Plant-wax D/H ratios in the southern European Alps record multiple aspects of climate variability

Stefanie B. Wirth^{a,b*} and Alex L. Sessions^a

^a Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena CA, United States

^b Present address: Centre for Hydrogeology and Geothermics, University of Neuchâtel, Neuchâtel, Switzerland

* Corresponding author:

Stefanie Wirth, University of Neuchâtel, CHYN, Rue Emile-Argand 11, 2000 Neuchâtel, Switzerland

e-mail: stefanie.wirth@alumni.ethz.ch

Abstract

We present a Younger Dryas–Holocene record of the hydrogen isotopic composition of sedimentary plant waxes (δD_{wax}) from the southern European Alps (Lake Ghirla, N-Italy) to investigate its sensitivity to climatic forcing variations in this mid-latitude region (45°N). A modern altitudinal transect of δD values of river water and leaf waxes in the Lake Ghirla catchment is used to test present-day climate sensitivity of δD_{wax} . While we find that altitudinal effects on δD_{wax} are minor at our study site, temperature, precipitation amount, and evapotranspiration all appear to influence δD_{wax} to varying extents. In the lake-sediment record, δD_{wax} values vary between -134 and -180 ‰ over the past 13 kyr. The long-term Holocene pattern of δD_{wax} parallels the trend of decreasing temperature and is thus likely forced by the decline of northern hemisphere summer insolation. Shorter-term fluctuations, in contrast, may reflect both temperature and moisture-source changes. During the cool Younger Dryas and Little Ice Age (LIA) periods we observe unexpectedly high δD_{wax} values relative to those before and after. We suggest that a change towards a more D-enriched moisture source is required during these intervals. In fact, a shift from northern N-Atlantic to southern N-Atlantic/western Mediterranean Sea sources would be consistent with a southward migration of the Westerlies with climate cooling. Prominent δD_{wax} fluctuations in the early and middle Holocene are negative and potentially associated with temperature declines. In the late Holocene (<4 kyr BP), excursions are partly positive (as for the LIA) suggesting a stronger influence of moisture-source changes on δD_{wax} variation. In addition to isotopic fractionations of the hydrological cycle, changes in vegetation composition, in the length of the growing season, and in snowfall amount provide additional potential sources of variability, although we cannot yet quantitatively assess these in the paleo-record. We conclude that while our δD_{wax} record from the Alps does contain climatic information, it is a complicated record that would require additional constraints to be robustly interpreted. This

also has important implications for other water-isotope-based proxy records of precipitation and hydro-climate from this region, such as cave speleothems.

Key words: Plant-wax D/H ratio; southern European Alps; Holocene; Younger Dryas; moisture source; temperature; vegetation; Westerlies; Mediterranean Sea; lake sediments

1 Introduction

The hydrogen isotopic composition (D/H ratio) of plant leaf waxes (δD_{wax}) has increasingly been used as proxy for reconstructing past hydro-climatic changes in many regions (e.g. Sauer et al., 2001; Schefuss et al., 2005; Nichols and Huang, 2012; Tierney and deMenocal, 2013; Feakins et al., 2014). Values of δD_{wax} from lacustrine and marine sedimentary sequences are considered to reflect past changes in the isotopic composition of the plants' source water, i.e. soil water recharged by infiltrating precipitation and river water (Sachse et al., 2012). However, a range of factors and processes controls the isotopic composition of the precipitation feeding the plants' source water, including the location and relative humidity of the initial moisture source, Rayleigh distillation by condensation processes during transport to the area of interest (i.e. temperature changes, rain- and snowfall events, and transport distance are important), and also re-evaporation during precipitation events (Craig, 1961; Dansgaard, 1964; Rozanski et al., 1997). A complete assessment of these isotopic fractionation processes is mostly unrealistic in the framework of paleo-climate studies, so simplifications have to be applied when interpreting δD_{wax} data. In addition, δD_{wax} is subject to physiological processes controlling isotope fractionation during plant-wax biosynthesis (e.g. Sessions et al. 1999; Smith and Freeman, 2006; Sachse et al., 2012; Kahmen et al. 2013). Despite these inevitable complications, δD_{wax} from sedimentary records has often been found to reliably record

aspects of past climate, but a key question is often *which* aspect of climate is being recorded. In the tropics, δD_{wax} primarily reflects changes in the amount of precipitation, whereas at high latitudes it seems to record mainly changes in temperature (Niedermeyer et al., 2010; Thomas et al., 2012). At mid-latitudes, greater variability in moisture sources and transport pathways often lead to a more complex forcing of δD_{wax} (Dansgaard, 1964; Aichner et al., 2015).

The southern European Alps (hereafter ‘southern Alps’) and the Mediterranean area (30–45°N) is a heavily populated and hydro-climatically sensitive area, for which a significant decrease in mean precipitation and an increased risk of drought with ongoing climate warming is expected (Giorgi and Lionello, 2008; Rajczak et al., 2013). Climatic reconstructions from this area are therefore crucial to understand past hydrologic variability as well as its potential impact on human civilization (e.g. Roberts et al., 2011; Magny et al., 2013).

Moisture arrives in the southern Alps by advection from the western Mediterranean and from the North Atlantic (N-Atlantic), as well as through land evapotranspiration in summer (Sodemann and Zubler, 2010; Winschall et al., 2014). The relative amount of southern N-Atlantic (~20–35°N) and western Mediterranean moisture vs. northern N-Atlantic (~35–60°N) moisture reaching the southern Alps is influenced by the meridional position of the westerly storm tracks (Westerlies) above the N-Atlantic, and thus potentially by the state of the North Atlantic Oscillation (NAO) (Hurrell et al. 2003; Trouet et al., 2012). During positive NAO (NAO+) conditions the Westerlies have a more northerly position causing wet and warm conditions in Scandinavia, and dry and cool conditions in southern Europe (including the southern Alps); almost opposite conditions occur during a negative NAO (NAO–) state (Wanner et al., 2001). The more southern position of the Westerlies during NAO– conditions thus entails an increased moisture advection from the southern N-Atlantic and from the western Mediterranean to the southern Alps.

Northern N-Atlantic (~35–60°N) surface waters are depleted in ^{18}O by ~1‰ (corresponding to a ~8‰ D-depletion; Craig, 1961) relative to southern N-Atlantic (~20–35°N) and western Mediterranean surface waters (Celle-Jeanton et al., 2001; LeGrande and Schmidt, 2006). On the one hand, this difference provides a potential opportunity to reconstruct past moisture-source changes using water isotope proxies such as $\delta\text{D}_{\text{wax}}$ or the oxygen isotopic composition of carbonate minerals ($\delta^{18}\text{O}_{\text{carb}}$) archived in sedimentary sequences. On the other hand, it is a potential complication for those proxies if one is primarily interested in interpreting them in terms of temperature or precipitation amount.

Previous studies from the south-alpine region investigating Holocene climate based on $\delta^{18}\text{O}_{\text{carb}}$ include a mollusc-based record from Lake Frassino (Baroni et al., 2006) and a stalagmite record from Grotta di Ernesto (McDermott et al., 1999; Scholz et al., 2012), both located in northeastern Italy (Fig. 1a). Whereas $\delta^{18}\text{O}_{\text{carb}}$ variations in the lake record are driven by lake-water evaporation and are therefore interpreted as wet-dry changes, the interpretation of $\delta^{18}\text{O}_{\text{carb}}$ variations at Grotta di Ernesto is complex and seems to reflect local effects as well as changing moisture sources (Scholz et al., 2012). In addition, the Holocene $\delta^{18}\text{O}_{\text{carb}}$ signal of stalagmites from the Spannagel Cave in the eastern Alps (Austria) (Fig. 1a) was intensively investigated over the last decade (e.g. Mangini et al., 2005; Fohlmeister et al., 2012). Spannagel Cave $\delta^{18}\text{O}_{\text{carb}}$ variations have primarily been interpreted as temperature changes, where lower $\delta^{18}\text{O}_{\text{carb}}$ corresponds to higher temperature. The mechanism proposed by the authors is that warm periods are characterized by relatively more winter precipitation compared to cool periods. As a result, speleothem $\delta^{18}\text{O}_{\text{carb}}$ integrating the annual, and not seasonal, precipitation $\delta^{18}\text{O}$ signal would decrease. Alternatively, it has also been proposed that the relative contribution of ^{18}O -enriched Mediterranean vs. ^{18}O -depleted N-Atlantic moisture is of importance (Mangini et al., 2005). In total, these $\delta^{18}\text{O}_{\text{carb}}$ studies indicate that

the interpretation of water isotope data from sedimentary proxies is not unambiguous in the alpine region.

The primary aim of our study is to evaluate the potential value of δD_{wax} as a (hydro-)climatic proxy in the mid-latitude region of the Alps. In doing so, we address the possible effects of changes in the relative importance of temperature and moisture source, of shifts in the growing season, and of changes in vegetation in determining δD_{wax} values over the course of the past 13 kyrs. In order to approach these goals, we established a Younger Dryas–Holocene δD_{wax} record based on *n*-C₂₈ alkanoic acids from the sediments of south-alpine Lake Ghirla (N-Italy). The sediments are well characterized and, importantly, hydro-climatic information is available in the form of a paleo-flood reconstruction (Wirth, 2013; Wirth et al., 2013). In addition, we conducted a catchment study, investigating the present-day factors controlling D/H of river water (δD_{river}), as well as δD_{wax} of modern tree leaves and of riverbed sediments. We compared our results to the water isotopic composition of precipitation and to meteorological data from the nearby Global Network of Isotopes in Precipitation (GNIP) and MeteoSwiss station Locarno (28 km north of Lake Ghirla).

2 Study area

2.1 Lake and catchment area

Lake Ghirla (45.916°N, 8.822°E) is a small lake with a surface area of 0.28 km² and a maximum water depth of 14 m situated at the foot of the southern Alps at an elevation of 442 m above sea level (asl) (Fig. 1). The maximum elevation in the catchment area is 1129 m asl. The lake is located in the N-S-oriented Valganna valley, which drains towards the north into Lake Maggiore (193 m asl). The southern limit of the Valganna coincides with a

morphological step of ~60 m, in the north of the city of Varese (382 m asl), that represents the southernmost physiographic limit of the Alps (Fig. 1b).

Sediment cores used for this study were retrieved from the central, deepest (14 m) part of the lake (Fig. 1d). Complementary cores taken in the slightly shallower (13 m) area further south contain significantly less detrital material, indicating that the S-N-flowing stream entering the lake at its southern end provides only a minor portion of the sediment supply. Sediment is primarily delivered by tributaries draining the eastern and western slopes, leading to the accumulation of large delta structures extending into the lake (Fig. 1d). Detrital material found in the lake sediments reflects the mineralogy of the Permian granites and Triassic dolomites constituting the bedrock of the lateral slopes (Fig. 1c) (Atlas of Switzerland, 2010). Due to their steepness and thin soils they are unsuitable for agriculture and are therefore densely wooded. The most abundant tree species at present are beech (*Fagus*), hazel (*Corylus*) and chestnut (*Castanea*). A previous paleo-vegetation study reported an unusually low occurrence of agricultural activities in the Valganna until the Roman Period when pollen of *Castanea*, walnut (*Juglans*) and rye (*Secale*) appeared (Schneider and Tobolski, 1983).

2.2 Climatic setting

The study area is influenced by both N-Atlantic and western Mediterranean weather patterns. In winter, the N-Atlantic is the principal moisture source (Sodemann and Zubler, 2010), mirroring the influence of the Westerlies and of the NAO on European winter climate (Hurrell et al., 2003). NAO+ conditions are characterized by dry and warm conditions in the western Mediterranean, whereas the southern position of the Westerlies during NAO– conditions leads to an increased moisture transport to Mediterranean latitudes (Trigo et al., 2006; Mariotti and Dell'Aquila, 2012) and thus to rather wet conditions at Lake Ghirla. In spring and autumn, the western Mediterranean, and the Gulf of Genoa in particular, is the primary moisture source

(Fig. 1a); in summer, evapotranspiration from the Central European land surface constitutes the major moisture contribution (Sodemann and Zubler, 2010). However, modern event-to-event variability is large (Winschall et al., 2014).

Mean annual air temperature (MAAT) at the closest meteorological station (Locarno-Monti, 383 m asl, 28 km from Lake Ghirla; Fig. 1) is 11.9 °C; annual precipitation is 1893 mm with three quarters of the precipitation falling between April and October (Fig. 2b) (MeteoSwiss, 2015a). Compared to the continental climate in the north-alpine foreland (Bern; Fig. 2a) and at the Mediterranean coast (Genoa; Fig. 2c) our study area receives nearly twice as much precipitation. The reason is the relief and the shape of the alpine arc focusing the moisture flux from the south and acting as an orographic barrier (Rakovec et al., 2004; Isotta et al., 2014).

2.3 Water isotopes in precipitation

On an annual time scale, the isotopic composition of precipitation (δD_P , $\delta^{18}O_P$) at the GNIP station Locarno is best correlated with air temperature ($r^2_{\delta^{18}O_P-T}=0.76$; $+0.4 \text{ ‰/°C}$; monthly data used for calculations) (Fig. 2b; Table 1) while there is only a weak correlation with precipitation amount ($r^2_{\delta^{18}O_P-P}=0.19$). (We use $\delta^{18}O_P$ data ($n=370$) for calculating correlation factors since the δD_P time series ($n=280$) is incomplete.) The more marked rise of $\delta^{18}O_P$ in Mar-Apr-May as compared to the temperature increase (Fig. 2b) is possibly due to an enhanced contribution of Mediterranean moisture (Celle-Jeanton et al., 2001; Sodemann and Zubler, 2010). If considering only the growing season (Feb–Sep), relations among temperature, precipitation amount and $\delta^{18}O_P$ are similar to the annual time scale ($r^2_{\delta^{18}O_P-T}=0.64$; $r^2_{\delta^{18}O_P-P}=-0.05$).

The analysis of the instrumental climate data demonstrates that the annual temperature cycle dominates the isotopic composition of precipitation. In fact, ~70% of the variance in $\delta^{18}\text{O}_\text{P}$ at alpine GNIP stations is explained by the seasonal temperature cycle (Mariani et al., 2014). However, after removal of the seasonal temperature cycle, i.e. only taking into account interannual temperature variability, the fraction of $\delta^{18}\text{O}_\text{P}$ variation that is explained by temperature changes decreases to ~20% (Mariani et al., 2014). This indicates that the original water isotopic composition of the moisture source, atmospheric processes during the transport from the moisture source to the study area, as well as the irregular occurrence of precipitation in the course of a year all play important roles in determining the final $\delta^{18}\text{O}_\text{P}$ value.

3 Material and methods

3.1 Leaf and water sampling in the catchment

Leaves of the main tree species *Fagus*, *Corylus* and *Castanea*, as well as samples of river water and lake surface water were collected on 21 April 2013 and on 14 April 2014 about one week after leaf flush. Samples were collected along an altitude profile from 442 to 1030 m asl following one of the rivers draining the eastern catchment slope (Rio dei Pradisci; Fig. 1d). Leaf samples were taken from trees growing at a short distance (1–10 m) from the stream. In 2014, riverbed sediments at 442, 770, 1030 m asl were also sampled.

3.2 Lake-sediment coring

Lake sediments were collected in spring 2009 in 3 m-long sections using a UWITEC platform with a manual percussion piston coring system (for core location see Fig. 1d). Cores were retrieved as twin cores with a 1.5 m vertical offset. In addition, short gravity cores of ~80 cm length were taken to study undisturbed surface sediments.

208

209 *3.3 Lake-sediment characterization*

210 Sediment cores were scanned for density and magnetic susceptibility (MS) using a Geotek
211 multi-sensor core logger (MSCL) at a down-core resolution of 0.5 cm. Subsequently, cores
212 were split lengthwise into halves and the core surface was photographed with a line scan
213 camera system at a resolution of 72 pixel/inch (Avaatech core scanner). In order to obtain a
214 qualitative high-resolution density record, as well as x-ray images revealing internal sediment
215 structures, computed tomography (CT) scans (St-Onge and Long, 2009) with a resolution of
216 0.6 mm were undertaken on half cores at the Institute of Diagnostic and Interventional
217 Radiology of the University Hospital Zurich. The discrimination of sedimentary lithologies is
218 based on the visual observation of color and grain-size contrasts on core pictures and x-ray
219 images, as well as on rapid changes in density and MS data.

220

221 *3.4 Lipid extraction and quantification*

222 Samples from sediment cores were taken as 1 cm thick slices. Where organic matter content
223 was low, sample thickness was increased to 1.5–3 cm. 1.5 to 5 g of freeze-dried and
224 homogenized sample material was typically used for lipid extraction, though in a few cases of
225 a primarily clastic lithology (<0.5 wt% total organic carbon (TOC)) up to 23 g of sample were
226 extracted. In the case of catchment samples, 5–8 freeze-dried leaves yielding dry weights of
227 0.2–0.4 g were used.

228 Lipids were extracted with a microwave-assisted solvent extraction system (MARS 5, CEM
229 Corp) at 100 °C for 15 min using 20 ml dichloromethane (DCM)/methanol (MeOH) (9:1 v:v).
230 After filtering the total lipid extract (TLE) through a glass fiber filter, it was saponified
231 overnight with 10 ml aqueous 0.5 M NaOH at 70 °C in a dry bath and extracted with 3x 10 ml

methyl-t-butyl ether (MTBE). The saponified TLE was then separated into four fractions of increasing polarity by solid-phase extraction using 0.5 g Septra-NH₂ (Phenomenex) stationary phase. Eluted fractions contained hydrocarbons (7 ml hexane), esters and ketones (7 ml 4:1 hexane/DCM), alcohols (7 ml 9:1 DCM/acetone) and carboxylic acids (7 ml 3% formic acid in DCM). Alkanoic acids were esterified to fatty acid methyl esters (FAMES) in a mixture of 20:1 v:v anhydrous MeOH/acetyl chloride for 10 min at 100 °C. Equal volumes of hexane and deionized water were then added to quench the reaction and partition lipids into the solvent, which was removed and filtered through a Pasteur pipette packed with anhydrous NaSO₄ to eliminate residual water. Solvent volume was reduced by evaporation under an N₂ stream at 35 °C. For riverbed sediment and leaf samples, a further cleaning step using solid-phase extraction with Discovery Ag-Ion (Supelco) stationary phase was employed to separate the target saturated FAMES from other unsaturated lipids.

FAMES were identified via gas chromatography-mass spectrometry (GC-MS: Thermo Trace GC/DSQ II). A flame ionization detector (GC-FID) was used for quantification and concentrations were calculated relative to the peak area of an internal standard (palmitic acid isobutyl ester) added prior to analysis. Concentrations are reported in µg/g dry sediment.

3.5 Compound-specific δD analysis

Compound-specific hydrogen isotope analyses were obtained using a Thermo Trace GC_{ultra} coupled to a Thermo Delta_{plus}XP isotope ratio mass spectrometer via a GC/TC pyrolysis furnace operated at 1430 °C (GC-P-IRMS). The H₃⁺ correction factor was determined on a daily basis following the protocol of Sessions et al. (2001). The mean H₃⁺ factor over the duration of five measurement periods was 2.40 ppm/mV (1σ=0.08, n=41). An external standard consisting of 8 FAMES (A. Schimmelmann, Indiana University, Bloomington) with δD values ranging from -166.7 to -231.2 ‰ was run between samples to monitor precision

and accuracy of the system. δD values of samples were evaluated relative to 10 pulses of CH_4 reference gas ($\delta D_{CH_4} = -151.9$ ‰) added after the GC column as described by Wang and Sessions (2008). Precision was evaluated by measuring samples in triplicate, except for 5 samples where material was sufficient for only one injection. Triplicate measurements of n - C_{28} acids in sediment and leaf samples yielded a mean standard deviation (1σ) of 2.1 ‰ ($n=98$) and 2.3 ‰ ($n=25$), respectively. The δD value of MeOH used for derivatization was estimated by analyzing phthalic acid ($\delta D = -95.3$ ‰) as the dimethyl ester ($1\sigma = 1.2$ ‰, $n=21$). The contribution of the added methyl group was subtracted from all FAMES by isotopic mass balance. All δD values are reported as permil (‰) deviations from the V-SMOW standard.

3.6 Total organic carbon (TOC) contents

10–20 mg of the sediment core samples were prepared for carbon analysis following standard procedures (e.g. Meyers and Teranes, 2001). Carbonate phases (mainly dolomite) were dissolved by treating the samples twice with hydrochloric acid (10%) over 20–24 h. Samples were washed with milliQ water until the pH was ≥ 6 after each acid treatment. The carbon content of the acid-treated samples was measured with a Costech CS4010 combustion Elemental Analyzer (EA). TOC content of the samples was calculated using the relationship:

$$TOC \text{ (wt\%)} = (TC \text{ of decarbonated sample (wt\%)} \times \text{Mass of decarbonated sample (mg)}) / \text{Mass of total sample (mg)}$$

TOC results are reported as wt% C of dry sediment. The standard deviation of replicate measurements (1σ , $n=18$) was 0.63 wt%.

3.7 Age-depth model

The most recent 60 years of the sediment sequence were dated by ^{137}Cs -activity

measurements defining depth horizons for 1986 (Chernobyl accident), 1963 (peak atmospheric weapon testing) and ~1951–53 (^{137}Cs zero-activity) (Appleby, 2001) (Table 2). ^{137}Cs -activity was measured at Eawag, Dübendorf, Switzerland. 16 radiocarbon (^{14}C) ages from terrestrial plant macrofossils were obtained by accelerator mass spectrometry (AMS) (^{14}C laboratories of ETH Zurich, Switzerland, and of Woods Hole Oceanographic Institution, United States) and used for dating the deeper sediment sequence (Table 2). The Laacher See Tephra (LST) (van den Bogaard, 1995) dated to 12,880 years BP (Brauer et al., 1999) provides an additional age horizon. Age-depth modeling was realized using the Clam R code (Blaauw, 2010) and applying a smooth spline interpolation between age horizons. All ages are reported as calendar years before present (cal yr BP), hereafter abbreviated as yr BP or kyr BP.

3.8 Water isotope analysis

The D/H and $^{18}\text{O}/^{16}\text{O}$ ratios of river and lake-surface water samples were analyzed with a DLT-100 spectroscopic liquid-water isotope analyzer (Los Gatos Research). Samples and two internal standards of known isotopic composition were analyzed as 6 replicate injections. The water isotope ratios of the samples were evaluated relative to the standards using a linear calibration. The mean precision of δD and $\delta^{18}\text{O}$ values of standards and samples is 0.54 ‰ and 0.24 ‰ (1σ , $n=51$), respectively.

3.9 Instrumental climate data

Data from the meteorological station Locarno-Monti (OTL; 46.172°N, 8.787°E; 383 m asl) located 28 km north of Lake Ghirla is available through GNIP, MeteoSwiss, and the Federal Office for the Environment of Switzerland (FOEN). We use monthly δD_p and $\delta^{18}\text{O}_\text{p}$ values of

precipitation (GNIP, 1976–2011; FOEN, 1992–2014); daily, monthly and annual data of temperature (T), precipitation (P), sunshine duration (SD), humidity (H) and evapotranspiration (ET) (MeteoSwiss, 2015a); as well as the record of days with snowfall (MeteoSwiss, 2015b).

4 Results

4.1 Catchment study 2013–2014

4.1.1 δD_{river} and δD_{wax} along altitude profile

δD_{river} values show a modest inverse altitude effect of +0.9 ‰/100 m ($r^2=0.81$) in 2013 (Fig. 3a) (for $\delta^{18}\text{O}_{\text{river}}$: +0.1 ‰/100 m; $r^2=0.62$). As a reference, the average altitude effect of $\delta^{18}\text{O}_\text{P}$ in the Alps is -0.2 to -0.3 ‰/100 m (Ambach et al. 1968; Schürch et al., 2003). No trend of δD_{river} with altitude is observed in 2014 ($r^2=-0.01$) (Fig. 3b).

δD_{wax} values of leaf samples collected in 2013 show an altitude effect of -3.7 ‰/100 m ($r^2=-0.60$) (Fig. 3a). In contrast, no significant altitude effect is observed for 2014 samples ($r^2=0.15$) (Fig. 3b). Average δD_{wax} values are identical at -131 ‰ in 2013 ($1\sigma=12$ ‰) and 2014 ($1\sigma=11$ ‰). Furthermore, no evidence was found for a systematic variation of δD_{wax} values between tree species (*Fagus*, *Corylus* and *Castanea*). The average apparent fractionation between leaf waxes and river water sampled from the same location and time [$\epsilon_{\text{wax/river}} = (\delta D_{\text{wax}} + 1)/(\delta D_{\text{river}} + 1) - 1$] is -71 ‰ ($1\sigma=14$ ‰) in 2013 and -78 ‰ ($1\sigma=11$ ‰) in 2014 (Table 3).

Riverbed sediment samples from 442, 805 and 1030 m asl exhibit δD_{wax} values of -149, -150 and -141 ‰, respectively, with little variation (Fig. 3b). They are D-depleted compared to the youngest sediment-core sample (-135 ‰) (asterisk in Fig. 3b) and to the leaf samples (on average -131 ‰).

329

330 *4.1.2 Feb-Mar-Apr climate conditions in 2013 and 2014*

331 We briefly outline the Feb-Mar-Apr climate conditions in 2013 and 2014, since they are the
332 most relevant to the δD values of leaves and water samples collected in the catchment in
333 April. Most importantly, March and April of 2014 were considerably warmer and drier than in
334 2013 (Fig. 4). In addition, February 2014 was the wettest of all months. These conditions are
335 reflected by higher average daily temperature (10.2 vs. 7.4 °C; 2014 vs. 2013),
336 evapotranspiration (1.5 vs. 1.0 mm/day), sunshine duration (5.9 vs. 5.2 h/day), and
337 precipitation amount (6.1 vs. 5.2 mm/day) in 2014 compared to 2013 (Fig. 4; Table 3). More
338 snow fell in 2014 (68 mm of precipitation in 5 days) than in 2013 (48 mm in 9 days). All
339 snowfall days occurred in February. Average Feb-Mar-Apr δD_P is 12 ‰ lower in 2014 than in
340 2013 (Table 3); however, inter-month variation of δD_P is high (Fig. 4).

341

342 *4.2 Lake-sediment record*

343 *4.2.1 Sediment lithologies*

344 The sediment sequence has a composite length of 12.6 m. Seven sediment lithologies are
345 distinguished (Fig. 5; and depth plot in supplementary material, Fig. S1):

346 *Clastic sediments – CL:* The lowermost 61 cm of the sediment sequence (1264–1203 cm) are
347 characterized by clay-to-silt-sized light-grey and sand-sized dark-grey layers with density
348 values of 1.4–1.8 g/cm³, magnetic susceptibility (MS) values of 19–40 10⁻⁵ SI, and a low
349 TOC content (<0.5 wt%). The interval with the highest MS values (38–40 10⁻⁵ SI) at 1237–
350 1234 cm depth has a yellowish color and contains the liquefied layer of the LST (van den
351 Bogaard, 1995). Liquefaction of this clay- and water-rich sediment section is probably due to
352 continuous hammering during coring.

353 From 1203 to 568 cm core depth, we distinguish three lithologies:

354 *Regular sediments – RS*: Dark-brown to black, organic-rich (6–15 wt% TOC) sediments with
355 a density of 1.0–1.3 g/cm³ and MS values of 0–10 10⁻⁵ SI represent the regular lacustrine
356 sedimentation.

357 *Flood layers – FLs*: Mineral-rich, often fining-upward deposits of dark-grey or red-brown
358 color with a beige-colored clay cap represent flood layers. Density and MS values lie in the
359 range of 1.3–1.7 g/cm³ and 5–25 10⁻⁵ SI, respectively. These layers consist of detrital material
360 (mineral grains from the catchment and terrestrial organic matter) flushed into the lake during
361 flood events and have thicknesses from <1 mm to 16.6 cm. Limits of flood layers were
362 primarily identified based on the color contrast with the regular sediments.

363 *Mass-movement deposit – MMD*: One thick mass-movement deposit (45 cm) consisting of
364 remobilized sediments occurs at a core depth of 1015 cm. This deposit clearly differs from
365 flood layers by its abnormally coarse grain size and its characteristic chaotic internal structure
366 (Schnellmann et al., 2005) imaged by CT scanning.

367 From 568 to 0 cm core depth, sediments alternate between dark-brown organic-rich and beige
368 to light-brown detrital-rich sections. The high TOC content (>6 wt%) and the near-absence of
369 minerals in the dark-brown intervals are similar to the regular sediments (RS) in the deeper
370 part of the core. The light-colored sections are rich in detrital material but poorer in TOC (<6
371 wt%). However, in contrast to the deeper part of the core, flood layers cannot be determined
372 since limits are smeared. Hence, we suggest that layering in this upper part was destroyed
373 after sediment deposition, most likely due to bioturbation. We categorize the interval 568–0
374 cm core depth into three lithologies: bioturbated regular sediments (*BT*) intercalated by
375 intervals rich in detrital material (*BT-high-detr*) and intervals with a slight enrichment in
376 detrital material (*BT-semi-detr*).

377

378 4.2.2 Age-depth model

379 The age-depth relationship was established after excluding event deposits (flood layers and
380 mass-movement deposits) from the sediment record (upper curve in Fig. 5), since these
381 deposits represent near-instantaneous events deposited during only hours to days (Lambert
382 and Giovanoli, 1988). In the bioturbated section (above 568 cm core depth), this subtraction
383 was not possible (see section 4.2.1). Below the lowest ^{14}C age (at 1195 cm) (Table 2), we
384 linearly extrapolated to the boundary between regular and clastic sediments (RS-CL) yielding
385 an age of $12,814 \pm 126$ yr BP. For the clastic lithology, which is characterized by a
386 significantly higher sedimentation rate compared to the regular sediments (1 yr/cm vs. ~50
387 yr/cm) (Fig. 5), we used a linear sedimentation rate based on the RS-CL and LST time
388 horizons. As a result, the clastic section (dated to ~12.9–12.8 kyr BP) represents the end of
389 the Allerød climate oscillation, while the change to regular sediments at 1203 cm core depth
390 most likely represents the beginning of the Younger Dryas (YD) (~12.7 kyr BP; Brauer et al.,
391 1999). No lithological change is observed at the Younger Dryas–Holocene transition (11.6
392 kyr BP) that is expected at a depth of ~1163 cm. Modeled age ranges (minimum fit to
393 maximum fit) for each centimeter core depth varies from 6 to 317 years (average is 177 years)
394 (Fig. 5). The depth intervals of $\delta\text{D}_{\text{wax}}$ and TOC samples cover age ranges from 1 to 110 years.
395 In Figures 6 and 7, samples are plotted using the ‘best fit’ age of the Clam age-depth model.

396

397 4.2.3 Paleo-flood reconstruction

398 Information on the occurrence of past floods triggered by heavy precipitation events is
399 obtained from the temporal distribution of flood layers in the section with regular sediments
400 and of highly detrital and slightly detrital intervals within the bioturbated section (Fig. 5).

Variation of flood occurrence is best illustrated by the density and CT data (Figs. 6e and 6f). We observe high flood activity at ~11, 10.6–8.2, 6.7, 6–4.9, 2.8–2.7, 2.6–2.4, 1.2–1 and 0.4–0.1 kyr BP; and slightly elevated flood activity at 12.8–12.2, 6.2, 4.2–3.8, 3.3, ~2 and 1.8–1.4 kyr BP. The clastic lithology at the base of the record dated to the end of the Allerød (12.9–12.8 kyr BP) likely also reflects wet climatic conditions.

*4.2.4 Concentrations and δD values of sedimentary *n*-alkanoic acids*

n-alkanoic acids with 14–32 carbon atoms (C_{14-32}) and characterized by the common preference towards even-numbered chain lengths (Eglinton and Eglinton, 2008) were identified. Concentrations and δD values were determined for the even-numbered *n*-alkanoic acids. Over the entire sediment record ($n=104$), *n*- C_{28} is most abundant (average concentration: 23 $\mu g/g$), followed by *n*- C_{26} (19 $\mu g/g$) and *n*- C_{24} (17 $\mu g/g$) (suppl. Fig. S2). Long-chained *n*-alkanoic acids ($C_{\geq 24}$) are generally attributed to the cuticular waxes of terrestrial plants (Eglinton and Hamilton, 1967; Galy et al., 2011). In fact, δD values of *n*- C_{24-28} acids in Lake Ghirla correlate well with each other ($r^2=0.77-0.80$), whereas correlation of *n*- C_{24-28} with *n*- $C_{\leq 22}$ is poorer ($r^2 \leq 0.6$) (suppl. Fig. S2 and Table S1), supporting the plant-wax origin of *n*- C_{24-28} . The δD values of the longest-chain acids (*n*- C_{30-32}) do not correlate well with those of *n*- C_{24-28} (for *n*- C_{30} , $r^2=0.29-0.38$; for *n*- C_{32} , $r^2=0.58-0.79$; suppl. Table S1), a result we attribute to co-elution of minor other components with *n*- C_{30} and *n*- C_{32} . In conclusion, due to its high abundance and clean chromatographic separation, we chose the *n*- C_{28} acid as representative of δD_{wax} variations, rather than calculating an abundance-weighted average value.

4.2.5 Temporal evolution of plant-wax concentrations, δD_{wax} values and TOC content

425 Plant-wax concentrations vary considerably over time (Fig. 6b). They are very low ($<1 \mu\text{g/g}$)
 426 in the clastic lithology corresponding to the Allerød climate oscillation, but range between 8–
 427 $110 \mu\text{g/g}$ during the Younger Dryas, with $110 \mu\text{g/g}$ being the second-highest concentration
 428 found in the record. The period from 11.6 to ~ 5 kyr BP is characterized by generally low
 429 plant-wax abundance ($1\text{--}28 \mu\text{g/g}$), with particularly low concentrations at 11.6–8.2 kyr BP. At
 430 5.2 kyr BP concentrations increase to $\sim 50 \mu\text{g/g}$ and stay at this level until 4.2 kyr BP. After ~ 4
 431 kyr BP until today values remain at an intermediate level but with large fluctuations (average
 432 $38 \mu\text{g/g}$, $1\sigma=28 \mu\text{g/g}$). Here, low concentrations are often associated with an increased detrital
 433 content of the sediments diluting the organic material (*BT-high-detr* and *BT-semi-detr*
 434 lithologies), e.g. at 2.8, 2.6–2.4, and 0.4–0.1 (LIA) kyr BP.

435 δD_{wax} values vary between -134 and -180‰ over the course of the past 13 kyrs (Fig. 6c). The
 436 final phase of the Allerød is characterized by a decrease of δD_{wax} from -150 to -164‰ .
 437 During the Younger Dryas, δD_{wax} shows a slightly increasing trend ($+2\text{--}3 \text{‰}$) with values
 438 ranging between -160 and -153‰ . After the Younger Dryas, the increasing trend continues
 439 until ~ 9 kyr BP where δD_{wax} reaches -146‰ . The interval between ~ 9 and 6.8 kyr BP is
 440 characterized by elevated values including the highest value of the record at 6.8 kyr BP (-134
 441 ‰). At the same time, this 9–6.8 kyr BP period comprises short-term excursions to lower
 442 values, namely at 8.4–8.2 and ~ 7.6 kyr BP. After 6.8 kyr BP δD_{wax} values decrease to a low
 443 value of -169‰ at ~ 5.5 kyr BP, then rise gradually to a value of -152‰ at 2.9 kyr BP. Two
 444 negative shifts of -5‰ and -7‰ at 4.8 and 3.8 kyr BP are evident during this period. The
 445 flood-rich interval from 2.8–2.4 kyr BP is characterized by a strong δD_{wax} variability. At the
 446 onset of the highly detrital interval at 2.8–2.7 kyr BP δD_{wax} first increases from -152 to -149
 447 ‰ , and then decreases to -156‰ in the subsequent semi-detrital interval (Fig. 6; lithology
 448 plot in suppl. Fig. S1). The onset of the following highly detrital interval at 2.6–2.4 kyr BP is
 449 characterized by a sudden decrease of δD_{wax} from -150‰ to -163‰ . At 2.2 kyr BP, i.e. after

the end of the flood-rich period, δD_{wax} abruptly increases by 9 ‰ to -151 ‰. In the following, δD_{wax} shows a decreasing trend and reaches -180 ‰ at 700 yr BP, the lowest value of the record. During the LIA (~400–50 yr BP), δD_{wax} values increase and lie between -177 and -162 ‰. After the LIA, δD_{wax} values show a rapid and substantial increase by ~30 ‰ to -144 ‰ (sample dated to ~1963 AD) and -135 ‰ (~2004 AD).

TOC content varies between <0.5 and 15 wt% over the course of the 13 kyr record (Fig. 6a). While the clastic section at the base of the record (final ~200 years of the Allerød) is particularly poor in TOC, values range from 7–13 wt% during the Younger Dryas. The Holocene is characterized by TOC contents of 7 ± 3 wt%. As for plant-wax abundance, low TOC values occur where detrital material dilutes the organic matter (most striking at ~11, 10.5, 8.4–8.2, 2.8, 2.6–2.4, and 0.2–0.1 kyr BP). However, in contrast to plant waxes, TOC content does not show the same pattern of particularly low values between 11.6 and 8.2 kyr BP (Figs. 6a and 6b).

The correlation matrix among δD_{wax} , plant-wax concentration, TOC content and CT number (Table 4) provides some indication of the relationships among these sediment-derived proxy records. Importantly, there is no correlation between δD_{wax} and the CT number representing flood activity ($r^2_{\delta D_{\text{wax}}-\text{CT}}=0.02$). The negative correlations of the TOC content and of the plant-wax concentration with the CT number ($r^2_{\text{TOC}-\text{CT}}=-0.67$; $r^2_{\text{conc}-\text{CT}}=-0.56$) primarily reflect the sedimentary process of dilution of organic-rich lake sediments with detrital material.

5 Discussion

5.1 Modern δD_{river} and δD_{wax} values in the catchment

The main goal of our catchment study, realized with samples collected in April 2013 and 2014 (Fig. 3), was to assess the importance of the altitude effect on the D/H ratio of leaf

waxes and river water in support of the climatic interpretation of the paleo- δD_{wax} data. In addition, we sought to examine the relative influence of climate variables such as temperature, precipitation amount, snowfall, and evapotranspiration on δD_{river} and δD_{wax} values (Fig. 4; Table 3).

Climate conditions during late winter and spring (Feb-Mar-Apr) seem to be most decisive for δD_{wax} values of the leaves sampled in April, as these conditions largely determine the D/H ratio of the plants' source water at the time of leaf unfolding (Sachse et al., 2012). The isotopic composition of the source water is primarily influenced by (i) the water isotopic composition of precipitation; (ii) the amount of snow that fell during winter, i.e. the amount of D-depleted snowmelt contributing to source water; and (iii) isotopic enrichment due to evaporation. The particular reasons why we chose the Feb-Mar-Apr period for the comparison of δD_{river} and δD_{wax} values in the catchment of Lake Ghirla with instrumental data from Locarno-Monti (Fig. 4; Table 3) are the following: (i) late winter (Feb-Mar) snowfall may have a D- and ^{18}O -depleting effect on the source water; (ii) leaf unfolding at Lake Ghirla occurs in mid-April (MeteoSwiss, 2015c); and (iii) during the development from bud to leaf over a period of about 30 days important isotopic fractionation between leaf water and plant waxes occurs (Sachse et al. 2010; Kahmen et al. 2011; Tipple et al., 2013), i.e. from about mid-March to mid-April at Lake Ghirla.

Regarding the river water samples (δD_{river}), D-enrichment via evaporation at higher altitudes could be responsible for the weak inverse altitude effect observed in 2013 (+0.9 ‰/100 m) (Fig. 3a). However, the samples do not plot in the field characteristic for evaporated waters (i.e. ^{18}O -enriched compared to the Local Meteoric Water Line (LMWL)) in a δD_{river} vs. $\delta^{18}\text{O}_{\text{river}}$ graph (Fig. 3c)). Alternatively, exfiltration of non-evaporated groundwater into the river could play a role. In 2014, the lesser amount of precipitation and thus decreased groundwater recharge may have reduced this effect and led to the muted behavior of δD_{river}

with altitude (Fig. 3b). In any case, compared to typical altitude effects of precipitation along the northern and southern main slopes of the Alps of -1.6 to -2.4 ‰/100 m for δD_P (Ambach et al. 1968; Schürch et al., 2003) the observed values in the catchment of Lake Ghirla are minor. We speculate that the reason lies not only in the possible exchange of river water with groundwater but also in the setting and the small size of the catchment. The rivers responsible for most of the water and sediment input into the lake are oriented W-E, thus perpendicular to the N-S orientation of the main alpine slope and of the primary winds responsible for moisture advection. This slope orientation, combined with the location of the catchment at the foot of the Alps, probably minimizes the importance of orographic effects within the rather small area of the Lake Ghirla catchment. Nevertheless, decreasing temperature with elevation certainly entails an altitude effect on δD_P . The δD_{river} data presented here only represents one snapshot per year. For a better comparison of the different influences on the water isotopic composition with altitude both river water (δD_{river}) and precipitation (δD_P) should be regularly sampled along the same catchment slope.

δD_{wax} values of leaves show a normal altitude effect in 2013 (-3.7 ‰/100 m; Fig. 3a), which is presumably the result of decreasing temperature with higher altitude. By comparison, the absence of an altitude effect in 2014 is more difficult to explain. Possibly it is the result of the warm and dry Mar-Apr conditions (Fig. 4; Table 3) leading to increased evaporation of source and leaf water, and concomitant D-enrichment, at higher altitudes where the exposure to sunlight may be higher due to a less dense forest cover. However, in contrast to river water samples we cannot test this hypothesis by comparison with the LMWL. The higher amount of snowfall measured at the meteorological station Locarno-Monti in 2014 compared to 2013 (Table 3) has no obvious effect on δD_{wax} of leaves. Average leaf δD_{wax} values for 2013 and 2014 are both -131 ‰, thus identical within analytical uncertainties (Table 3). The lack of snowfall data from the catchment itself, however, poses a difficulty for a thorough assessment

of the influence of snowfall. The reason is that the catchment of Lake Ghirla is located at significantly higher altitudes (altitude range 442–1129 m asl) than the meteorological station at Locarno-Monti (383 m asl).

The observed difference between δD_{wax} of leaves and of riverbed sediments (Fig. 3b) could perhaps be due to continued turnover of leaf waxes throughout the growing season, with shifts in δD_{wax} towards more D-depleted values later in the season (Newberry et al., 2015). In contrast to the leaf samples collected early during the growing season, riverbed sediments would reflect the D/H ratio of leaf waxes at the time of leaf drop. However, it has also been proposed that δD_{wax} is set entirely during the 30 days of leaf development, with little influence from the rest of the growing season (Sachse et al. 2010; Kahmen et al. 2011; Tipple et al., 2013). No consensus currently exists on which pattern might apply to alpine vegetation. Our above proposed explanation for the offset between δD_{wax} of leaves and of riverbed sediments is thus only consistent with a scenario in which δD_{wax} changes continuously throughout the summer. Not too much emphasis is put on the offset between δD_{wax} of riverbed sediments and of the youngest lake-sediment sample due to differences in age. The lake-sediment sample reflects the δD_{wax} signal of the years ~2003–2005 AD and is thus ~10 years older than the sediment sample from the riverbed.

In summary, we suggest that shifts in the isotopic composition of leaf waxes with altitude are negligible in the catchment of Lake Ghirla, and thus in the interpretation of the paleo- δD_{wax} record. The fact that the average δD_{wax} values in 2013 and 2014 are identical (-131 ‰; Table 3) despite the different behavior with altitude supports this conclusion. Nonetheless, the influence of snow-rich winters potentially leading to D-depleted source waters cannot be entirely disregarded, even though it might become less important if synthesis of leaf waxes continues throughout the growing season (Feb–Sep), a point that is still being debated as mentioned above (Sachse et al., 2009; Sachse et al. 2010; Kahmen et al. 2011; Tipple et al.,

2013; Newberry et al., 2015). Regarding the evaluation and quantification of the relative importance of climate parameters such as temperature, precipitation amount, and evapotranspiration in defining δD_{wax} , our modern field data are not conclusive. A longer monitoring campaign over several years and with sampling at regular intervals in the course of the entire growing season could lead to a clearer understanding in this regard.

5.2 Discussion of the δD_{wax} paleo-climatic signal

Next we compare the Younger Dryas–Holocene paleo- δD_{wax} record with climate records from low- to high-latitudes of the northern hemisphere, with solar forcing (insolation, solar irradiance), and with paleo-flood evidence from Lake Ghirla (Fig. 7). Our primary goal in making these comparisons is to examine which climatic factors are the strongest determinants of δD_{wax} in the southern Alps over the course of the past 13 kyrs.

5.2.1 End of Allerød and Younger Dryas (12.9–11.6 kyr BP)

The clastic lithology at the base of the Lake Ghirla sediment core is dated to the final ~200 years (12.9–12.8 kyr BP) of the warm Allerød climate oscillation (von Grafenstein et al., 1999; Weaver et al., 2003; Vescovi et al., 2007). δD_{wax} decreases from -150 to -164 ‰ in this sediment section. This decrease could potentially be attributed to an early cooling before the actual onset of the Younger Dryas (Fig. 7g). An *n*-alkane δD study from Lake Meerfelder Maar (MFM) reported a similar cooling signal beginning 170 years before the onset of the Younger Dryas (Brauer et al., 1999; Brauer et al., 2008; Rach et al., 2014) — consistent with evidence from Greenland ice cores (Rasmussen et al., 2006).

The transition from the Allerød to the Younger Dryas (dated to $12,814 \pm 126$ yr BP in Lake Ghirla, see section 4.2.2) does not exhibit a remarkable change in δD_{wax} (Fig. 7g). The

lithology, however, shows a prominent change from clastic to organic-rich sediments within 1–2 centimeters (suppl. Fig. S1), indicating an important climatic and environmental change. Accordingly, δD_{wax} does not provide explicit evidence for climate cooling even though cool Younger Dryas climate conditions in the Alps as well as in the western Mediterranean are well documented (summer T anomaly in the southern Alps ~ -2 °C, Samartin et al., 2012; MAAT in the Alps -3 to -4 °C compared to today, Ivy-Ochs et al., 2009; Mediterranean sea-surface temperature (SST) anomaly ~ -3 to -7 °C; Fig. 7h; Combourieu-Nebout et al., 2013; Rodrigo-Gamiz et al., 2014). Circum-Mediterranean $\delta^{18}\text{O}_{\text{carb}}$ records from lake sediments also generally lack ^{18}O -depletion in connection with the Younger Dryas temperature decrease; on the contrary, they often even indicate ^{18}O -enrichment due to changes in atmospheric circulation as well as due to aridity and thus evaporation (Roberts et al., 2008).

For Lake Ghirla, it is possible that a moisture-source change due to a southward migration of the Westerlies coincident with climate cooling (Brauer et al., 2008; Baldini et al., 2015) overrode the effect of lower temperatures on δD_{wax} . The surface waters of the southern N-Atlantic are not only more D-enriched relative to more northern areas, they also experienced only minor cooling (-0 to -2 °C; Renssen, 1997), and thus minor D-depletion, during the Younger Dryas. Similarly, for the Last Glacial Maximum (~ 24 ka BP) an increased moisture advection from the south towards the Alps has been proposed due to a southern position of the storm tracks (Luetscher et al., 2015). One tangible effect of this shift towards a predominant southern as opposed to a northwestern moisture advection towards the Alps was that the most important alpine ice domes accumulated south of the alpine water divide (Florineth and Schlüchter, 2000).

A complete change from northern N-Atlantic to southern N-Atlantic/western Mediterranean moisture would imply an 8 ‰ (δD) heavier moisture source ($\delta^{18}\text{O}$: 1 ‰; see section 1). Applying the modern relationship of water isotopes in precipitation with temperature in the

study area (GNIP station Locarno; $\delta^{18}\text{O}_\text{P}$: 0.4 ‰/°C; δD_P : 3.2 ‰/°C; see section 2.3), a moisture-source change of this size could indeed compensate a Younger Dryas temperature decrease of 2–3 °C (corresponding to -6.4 to -9.6 ‰ δD_P). Due to the more southerly position of the storm tracks during the Younger Dryas a significantly increased contribution of the southern, i.e. D-enriched, source areas is reasonable. However, the D-enriched moisture signature would need to be conserved throughout all fractionation processes during transport to the study area. Also, a quantitative shift from northern to southern source areas should be considered as an end-member scenario. This is illustrated by a modern study (data from 1989–2009) of heavy precipitation events in the northwestern Mediterranean area documenting a high variability in the seasonal and event-to-event contribution of different moisture source regions (N-Atlantic, Mediterranean, tropics, land evapotranspiration) (Winschall et al., 2014).

An alternative, or complementary, interpretation of the Younger Dryas $\delta\text{D}_\text{wax}$ signal is that a significant change in the composition of the vegetation in the Lake Ghirla catchment from primarily trees to shrubs might have influenced $\delta\text{D}_\text{wax}$. In addition, a shortening of the growing season due to cold and long winters and thus a shift of leaf-wax synthesis towards the summer months could have led to an increase of $\delta\text{D}_\text{wax}$ that counterbalanced the effects of cooler temperature. The fact that Younger Dryas summer cooling was less pronounced than winter cooling (Denton et al., 2005) could have contributed to this seasonality effect. In turn, pronounced winter conditions with very low temperatures, as well as an increased fraction of solid precipitation (snow), could have led to important isotopic depletion of the source water of the vegetation and thus to a decrease of $\delta\text{D}_\text{wax}$.

Based on current knowledge we cannot confidently distinguish which of the above scenarios (moisture-source change, changes in vegetation/growing season) is responsible for the

observed δD_{wax} signal during the Younger Dryas. Both topics require further research in order to achieve a more quantitative result.

5.2.2 Early to Mid-Holocene (11.6–5.2 kyr BP)

Similar to the onset, the end of the Younger Dryas shows no distinct δD_{wax} changes. Other climate reconstructions, however, document that air temperatures rose by $\sim 2\text{--}3^\circ\text{C}$ (southern Alps: Samartin et al., 2012; northern hemisphere: see Fig. 7a) and western Mediterranean SST by $\sim 3\text{--}7^\circ\text{C}$ (Fig. 7h) after the Younger Dryas. With this climate warming the Westerlies migrated northwards (towards the northern N-Atlantic) (Broccoli et al., 2006). We suggest that the same mechanism that was responsible for the lack of δD_{wax} decrease during the Younger Dryas, which one would expect if temperature were the solely forcing factor, was reversed at the end of the Younger Dryas. This effect may be a change of the primary moisture source due to the meridional migration of the Westerlies, or changes related to the composition of the vegetation and the timing of the growing season as discussed in section 5.2.1.

The period of low δD_{wax} variation during the earliest Holocene (11.6–9 kyr BP) is followed by the highest δD_{wax} values of our record, namely at ~ 8.9 , ~ 8 and 7.4–6.8 kyr BP (Fig. 7g). This period of high δD_{wax} values correlates with various lines of evidence for warm conditions, as well as for a northern position of the atmospheric circulation system (i.e. a northern position of the Westerlies and the ITCZ). Elevated air temperature is apparent in the Marcott et al. (2013) reconstruction from the northern hemisphere (Fig. 7a). In addition, warm western Mediterranean SST has been documented (Fig. 7h; Combourieu-Nebout et al., 2013). The marked northerly position of the atmospheric circulation system is illustrated by increased precipitation and strong monsoons at low-latitudes; e.g. Cariaco Basin (Fig. 7b; Haug et al., 2001); Dongge Cave, China (Dykoski et al., 2005); equatorial Africa (Tierney

and deMenocal, 2013); and formation of sapropel S1 in the eastern Mediterranean (Fig. 7; Hennekam et al., 2014). Furthermore, conditions with such a northerly position of the Westerlies resemble the positive state of the modern NAO circulation pattern. Under such conditions, moisture advection occurs towards northern Europe and perhaps to the northern Alps, while the southern Alps receive little northern N-Atlantic, and thus little D-depleted, moisture (Trigo et al., 2006; Wirth et al., 2013). If we apply this scenario to our study site, it suggests warm conditions with the Mediterranean Sea as the most important oceanic moisture source during the early Holocene.

The high δD_{wax} values between 9 and 6.8 kyr BP are interrupted by short negative shifts of δD_{wax} (at 8.4–8.2 and ~7.6 kyr BP). And they are followed by a longer interval of low values at 6.2–5.2 kyr BP. It seems likely that the negative shift of δD_{wax} at 8.4–8.2 kyr BP (Fig. 7g) is due to a decrease in temperature. Evidence is provided by cold conditions that appear in various climate reconstructions at the same point in time (Figs. 7d, 7e and 7h; Combourieu-Nebout et al., 2013). Increased concentrations of hematite-stained grains in the N-Atlantic indicating a southward advance of Greenland ice sheets, as well as phases of low solar irradiance (Fig. 7m; Bond et al., 2001), provide additional evidence for cooling. Likewise, cool conditions probably also apply to the period with low δD_{wax} values at 6.2–5.2 kyr BP, indicated by records from the Mediterranean Sea and from the N-Atlantic (Fig. 7h; Bond et al., 2001; Rohling et al., 2002), by worldwide glacier advances (Fig. 7f), by prominent solar lows around 5.6, 5.4 and 5.3 kyr BP (Fig. 7m), and by the northern hemisphere summer insolation reaching its turning point at ~5.5 kyr BP (Fig. 7c). As a consequence of the cool conditions, the fraction of solid precipitation (snow) could have increased, and would thus also have contributed to lower δD_{wax} values. The short excursion of δD_{wax} at ~7.6 kyr BP may also be driven by a temperature decrease. However, this interpretation remains uncertain, as comparisons with GISP $\delta^{18}\text{O}$ (Fig. 7d) or TSI (Fig. 7m) lack dating certainty.

At this point, a comparison with the $\delta^{18}\text{O}_{\text{carb}}$ record of the Spannagel Cave in the eastern Alps (Austria) seems useful to scrutinize our interpretation of $\delta\text{D}_{\text{wax}}$ forcing at Lake Ghirla. The Spannagel Cave stalagmite record shows — in contrast to high $\delta\text{D}_{\text{wax}}$ at Lake Ghirla — low $\delta^{18}\text{O}_{\text{carb}}$ values during the early Holocene (10–5.5 kyr BP) (Fig. 7i). Several scenarios could cause ^{18}O -depletion at the Spannagel Cave site: (i) increased contribution of ^{18}O -depleted winter precipitation as proposed by the authors (Mangini et al., 2005); (ii) cool temperatures; and (iii) an effect of [moisture source and transport](#), i.e. [a dominating contribution of \$^{18}\text{O}\$ -depleted moisture from northwest \(northern N-Atlantic as primary moisture source\)](#). As at Lake Ghirla, the Spannagel Cave site is sensitive to changes in the relative contribution of moisture from the N-Atlantic and from the western Mediterranean Sea. However, N-Atlantic moisture is normally more important than at Lake Ghirla and only strong cyclones forming in the northwestern Mediterranean [probably enable](#) significant moisture transport from the Mediterranean across the highest relief of the eastern Alps. Results from previous paleo-studies indicate that weather patterns including a south-to-north oriented moisture transport from the Mediterranean across the Alps, [or eastward around the Alps \(Vb cyclones; Nissen et al., 2013\)](#), probably occurred more frequently during cool climate conditions of the past (Czymzik et al., 2013; Glur et al., 2013). Hence, it seems reasonable that the contribution of ^{18}O -enriched Mediterranean moisture was probably low at the Spannagel Cave during the warm early Holocene. We therefore argue that different moisture sources, i.e. option (iii) above, may explain the discrepancy between the low $\delta^{18}\text{O}_{\text{carb}}$ at Spannagel Cave and the high $\delta\text{D}_{\text{wax}}$ values at Lake Ghirla. Without the aspiration to explain short-term variation in both records, this would imply that the Westerlies transported D-depleted moisture from the northern N-Atlantic mainly to north-alpine areas (including Spannagel Cave), and that Lake Ghirla probably received primarily Mediterranean moisture. This supports our above interpretation that temperature is primarily responsible for the early Holocene (until 5.2 kyr

BP) δD_{wax} variation at Lake Ghirla. Regarding Spannagel Cave, an increased contribution of ^{18}O -depleted winter precipitation remains a reasonable mechanism contributing to low $\delta^{18}\text{O}_{\text{carb}}$ values (option (i) above) during warm episodes. For further evaluation of the importance of this option (i), more detailed information about the seasonal distribution of precipitation in the past would be required.

5.2.3 Mid-Holocene (5.2–3 kyr BP)

At ~4.5 kyr BP, the onset of bioturbated sediments indicates a weakening of the water-column stratification. In combination with increasing plant-wax concentrations (Fig. 6b), this provides evidence for enhanced windiness and riverine water and sediment input, i.e. probably wetter conditions. The timing of this sedimentary transition matches the onset of late Holocene northern hemisphere cooling — also referred to as ‘Neoglaciation’ (Wanner et al., 2011) —, which is caused by the decrease of northern hemisphere summer insolation (Fig. 7c). Cooling, however, is not apparent in the δD_{wax} record. δD_{wax} exhibits a slight increase from 5.2–3 kyr BP, interrupted by two excursions to lower values at 4.8 and 3.8 kyr BP (Fig. 7g). The δD_{wax} increase perhaps reflects the increasing importance of the westerly storm tracks for the alpine climate at that time, with more D-enriched southern N-Atlantic and western Mediterranean moisture reaching the southern Alps. Moreover, the increase of $\delta^{18}\text{O}_{\text{carb}}$ at Spannagel Cave at this mid-Holocene climate transition is noteworthy (Fig. 7i). We suggest that the transport of ^{18}O -enriched moisture across the eastern Alps probably occurred more frequently with cooler conditions as discussed in section 5.2.2.

5.2.4 Late Holocene (3–0.1 kyr BP)

Between 2.8 and 2.2 kyr BP, rapid fluctuations of δD_{wax} are accompanied by prominent changes in flood activity (Figs. 7g and 7k). Concurrent temperature records from the Mediterranean, the Alps and the N-Atlantic, however, show no important changes (Figs. 7a, 7d and 7h; Geirsdóttir et al., 2013). Nonetheless, a major solar low, the Homeric Minimum (2.8–2.65 kyr BP), coincides with the shifts of δD_{wax} and the increased flood activity at 2.8–2.7 kyr BP. The Homeric Minimum has been associated with windier, cooler and wetter conditions at Lake Meerfelder Maar as well as in the southern Alps (Martin-Puertas et al., 2012; Wirth et al., 2013). Also, the flood reconstruction from Lake Ammersee located in the north-alpine foreland (close to Munich, Fig. 1a) indicates a high occurrence of floods (Fig. 7j; Czymzik et al., 2013). This agrees with evidence for a negative state of the NAO (Fig. 7l; Olsen et al., 2012), or a corresponding paleo-circulation pattern, and for a southern position at the ITCZ (Fig. 7b). Altogether, these records indicate a significant change in the climate system associated with the Homeric Minimum that also had an impact on δD_{wax} at Lake Ghirla. Similarly, the strong decrease of δD_{wax} at ~2.6 kyr BP coincides with a prominent increase in flood occurrence in the southern Alps (Fig. 7k; Wirth et al., 2013) and negative paleo-NAO conditions (Fig. 7l). However, the climatic forcing of this interval remains speculative. Possibly, a solar low at 2.4 kyr BP (Greek minimum) (Raspopov et al., 2013) and a recently reported major volcanic eruption at 2451 cal yr BP (Sigl et al., 2015) are of importance. In summary, δD_{wax} variability at 2.8–2.2 kyr BP is high. Currently, it is not possible to provide a coherent reasoning for the observed δD_{wax} fluctuations. We suspect that the frequent modification of the atmospheric circulation above the N-Atlantic, as well as processes at the catchment-scale such as vegetation and seasonality changes and the mobilization of lower sediment horizons due to flood erosion, were important.

The decline of δD_{wax} after 2.2 kyr BP to the lowest value of the record at 700 yr BP (-180 ‰) coincides with late Holocene cooling (Figs. 7a and 7f; Wanner et al., 2011) and is therefore

likely temperature-forced. The renewed increase of δD_{wax} during the LIA we interpret as an increase in D-enriched moisture (i.e. southern N-Atlantic and western Mediterranean) outweighing the temperature effect — similarly as for the Younger Dryas (section 5.2.1). Negative NAO conditions (Fig. 7l; Trouet et al., 2012), high $\delta^{18}\text{O}_{\text{carb}}$ values of the Spannagel Cave stalagmites (Fig. 7i), and high flood frequency at Lake Ammersee (Fig. 7j) support this interpretation. As for the Younger Dryas, the question if snow-rich winters and a shortened growing season also influence δD_{wax} values cannot presently be conclusively answered (section 5.2.1).

5.2.5 The past ~100 years

The prominent increase of δD_{wax} by ~30 ‰ during the past 50–100 years may be caused by both recent climate warming and land-use change. The long instrumental time series from the Locarno-Monti meteorological station (monthly resolution) shows a clear temperature increase towards the present (+1.2 °C for the period 1988–2014 relative to the 1864–2014 average) (MeteoSwiss, 2015a). In addition, recent land-use change may have modified the vegetation composition and increased evapotranspiration in the catchment, and thus impacted sedimentary δD_{wax} values. This hypothesis could be tested via complementary carbon isotope analysis ($\delta^{13}\text{C}_{\text{wax}}$), which is potentially capable of recording substantial vegetation changes.

5.2.6 Correlations between δD_{wax} and comparison data sets

For further evaluation of the climatic forcing of δD_{wax} , we calculated correlation factors between δD_{wax} and the comparison data sets illustrated in Figure 7 (Table 5). For the calculation, we used the period 0–11 kyr BP, thus focusing on the Holocene, and applied a Gaussian-kernel-based cross-correlation (Rehfeld and Kurths, 2014). We excluded the

Younger Dryas since δD_{wax} does obviously not correlate with any of the other data sets; and we eliminated the recent rapid increase of δD_{wax} because it significantly improved correlations, for instance with the temperature reconstruction of Marcott et al. (2013) (Fig. 7a).

The insolation-forced Holocene temperature decrease, well illustrated by Marcott et al. (2013) (Fig. 7a), plays a central role in the long-term behavior of δD_{wax} ($r^2_{\text{T-anomaly}-\delta D_{\text{wax}}}=0.78$). This corroborates our observation that the long-term decrease of δD_{wax} in the course of the Holocene is a response to temperature forcing. The good correlation with the Ti-record from the sediments of the Cariaco Basin ($r^2_{\text{Ti}-\delta D_{\text{wax}}}=0.59$) strengthens the link between changes in temperature and modifications of the atmospheric circulation system (meridional position of the Westerlies and of the ITCZ) during the Holocene. In this regard, the negative correlations of δD_{wax} with the Spannagel Cave $\delta^{18}\text{O}_{\text{carb}}$ ($r^2_{\delta^{18}\text{O}_{\text{carb}}-\delta D_{\text{wax}}}=-0.45$) and with the NAO reconstruction ($r^2_{\text{NAO}-\delta D_{\text{wax}}}=-0.40$) are also noteworthy. This result supports our hypothesis that the position of the Westerlies above the N-Atlantic is an important factor in defining δD_{wax} at Lake Ghirla, as well as $\delta^{18}\text{O}_{\text{carb}}$ at Spannagel Cave.

The correlation factors calculated with the detrended data sets are mostly insignificant. On the one hand, this underlines the observation that the short-term behavior of δD_{wax} is complex, as for instance during the period 2.8–2.2 kyr BP. On the other hand, this result may also emphasize that dating uncertainties that are common to paleo-climate reconstructions from natural archives pose challenges when comparing data sets with statistical methods.

6 Conclusions

We report a Younger Dryas–Holocene δD_{wax} record of Lake Ghirla located in the southern European Alps together with a modern catchment study, a paleo-flood reconstruction from the same study site, and proxy-record comparisons.

The modern catchment study indicates that the altitude effect on δD_{wax} is minor in the catchment of Lake Ghirla. For assessing the importance of various climate parameters in defining δD_{wax} our modern field study, however, yielded no conclusive data.

Decreasing temperature appears to control the δD_{wax} paleo-record on the long Holocene time scale. Declining northern hemisphere summer insolation seems therefore to be an important forcing factor of δD_{wax} . The shorter-term δD_{wax} fluctuations (decades to one millennium) are probably caused by both changes in temperature as well as in the main moisture source. Since the moisture from the northern N-Atlantic and from the southern N-Atlantic/western Mediterranean Sea are characterized by different isotopic signatures, changes in the relative contribution of these sources could be essential for shaping δD_{wax} at Lake Ghirla. In particular, during the cool periods of the Younger Dryas and the Little Ice Age, the southern position of the Westerlies appears to be – at least partly – responsible for unexpectedly high δD_{wax} values. Changes in the composition of the vegetation, as well as seasonality effects (length of growing season, snowfall amount), might also be potential sources of δD_{wax} variability. However, we cannot yet quantitatively assess these vegetation-related factors in the δD_{wax} paleo-record.

In conclusion, the interpretation of Younger Dryas–Holocene δD_{wax} variations in the southern Alps must consider a variety of possible forcing factors and processes, as well as a changing relative importance of these factors and processes over time. The information value of δD_{wax} as an alone-standing proxy is therefore limited. For improving our understanding of the δD_{wax} behavior in the Alps, as well as for the future use of δD_{wax} as paleo-climatic proxy in similar mid-latitude settings, a long-term monitoring and calibration campaign seems to be critical.

816

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1130 **Table captions**

1131 Table 1: Correlation matrix (1981–2014) of monthly (Jan–Dec) meteorological data from the
 1132 station Locarno-Monti (GNIP, 1976–2011; FOEN, 1992–2014; MeteoSwiss, 2015a) and of
 1133 the NAO index (NCAR, 2015). Length of used data set is limited by the time series of ET
 1134 beginning in 1981. T=temperature, P=precipitation amount, H=humidity, SD=sunshine
 1135 duration, ET=evapotranspiration, $\delta^{18}\text{O}_\text{P}$ =oxygen isotopic composition of precipitation.

1136

1137 Table 2: Radiocarbon ages (top) and age control from ^{137}Cs -activity measurements and the
 1138 LST (bottom) used for age-depth modeling. Radiocarbon ages were calibrated using the
 1139 IntCal13 calibration curve (Reimer et al., 2013).

1140

Table 3: Summary of results of catchment study realized in spring 2013 and 2014. Top: Average δD_{wax} of tree leaves and δD_{river} sampled along altitude profile (Fig. 3). Also provided are average apparent fractionation factors between leaf waxes and river water ($\epsilon_{\text{wax/river}}$). Bottom: Feb-Mar-Apr averages of daily instrumental data (MeteoSwiss, 2015a), and of monthly data of water isotopes in precipitation (FOEN, 1992–2014). FMA=Feb-Mar-Apr.

Table 4: Correlation matrix of sedimentary proxy data of Lake Ghirla emphasizing the absence of a simple relationship of δD_{wax} with other variables, in particular with the CT number representing flood activity.

Table 5: Pearson correlation factors and corresponding p-values (calculated with Gaussian-kernel-based cross-correlation; Rehfeld and Kurths, 2014) between Lake Ghirla δD_{wax} and comparison data sets presented in Figure 7. Correlations were calculated for the Holocene, 0–11'000 cal yr BP, thus excluding the recent climate change and the Younger Dryas. Bold font: correlations with p-value <0.05. 'Detrended': omitting long-term trends in data sets. Plots illustrating subtracted trends are presented in Figure S3.

Figure captions

Figure 1: Study area. a) Overview illustrating N-Atlantic and Mediterranean moisture-source areas relevant for Lake Ghirla. Blue arrows indicate primary wind directions (Digital Elevation Model over Europe (EU-DEM)). b) EU-DEM emphasizing the location of Lake Ghirla at the southernmost physiographic limit of the Alps. c) Greater catchment area with bedrock types (Atlas of Switzerland, 2010). d) Bathymetric map derived from reflection

seismic data with core location. Asterisks with altitude indication mark location of catchment samples taken in April 2013 and 2014. FRA (Lake Frassino), ERN (Grotta di Ernesto), SPA (Spannagel Cave) designate other study sites mentioned in the text, for which $\delta^{18}\text{O}_{\text{carb}}$ data, or paleo-flood data (AMM, Lake Ammersee), has been published.

Figure 2: Climate charts of (a) Bern, (b) Locarno, and (c) Genoa, illustrating the geographical (see Fig. 1a) as well as climatological position of Lake Ghirla (corresponding to (b) Locarno) in between (a) continental Europe and (c) the northwestern Mediterranean area. (Data: GNIP)

Figure 3: Results of catchment study. Presented are altitude profiles of $\delta\text{D}_{\text{wax}}$ (tree leaves and riverbed sediments) and of $\delta\text{D}_{\text{river}}$ for (a) April 2013 and (b) April 2014. In addition, $\delta\text{D}_{\text{river}}$ vs. $\delta^{18}\text{O}_{\text{river}}$ is compared to the Local (station Locarno-Monti) and Global Meteoric Water Lines (LMWL, GMWL) for (c) 2013 and (d) 2014. For sample locations see Figure 1d.

Figure 4: Comparison of climatic conditions during Feb-Mar-Apr 2013 and 2014 for investigating the climatic forcing of $\delta\text{D}_{\text{wax}}$ and $\delta\text{D}_{\text{river}}$ of catchment samples (see also Table 3). (Data: MeteoSwiss, 2015a; FOEN, 1992–2015)

Figure 5: Age-depth model with lithological profile, as well as with graphs illustrating accumulation rates and the uncertainty of modeled ages.

Figure 6: Temporal evolution of $\delta\text{D}_{\text{wax}}$ and of sedimentary proxy records from Lake Ghirla. a) TOC content; (b) concentration of plant waxes represented by $n\text{-C}_{28}$ acids; (c) $\delta\text{D}_{\text{wax}}$ (represented by $n\text{-C}_{28}$ acids); (d) magnetic susceptibility (MS); (e) density; and (f) CT number used as high-resolution density record in Figure 7. Elevated values in (d), (e) and (f) indicate

increased occurrence of floods. The grey area marks the Younger Dryas (YD), blue areas emphasize periods with important variation of δD_{wax} and of flood activity discussed in the text. LIA=Little Ice Age.

Figure 7: Comparison of δD_{wax} from Lake Ghirla with temperature, precipitation, and atmospheric circulation reconstructions from low- to high-latitudes of the northern hemisphere, as well as with solar forcing. a) 30–90° N temperature anomaly (Marcott et al., 2013); (b) precipitation record from the Cariaco Basin indicating changes in the meridional position of the ITCZ (Haug et al., 2001); (c) 30° N summer insolation (Berger and Loutre, 1991); (d) $\delta^{18}\text{O}$ of the Greenland ice core GISP2 (Grootes et al., 1993); (e) Holocene cold events (Wanner et al., 2011); (f) worldwide glacier advances (Denton and Karlén, 1973); (g) δD_{wax} Lake Ghirla; (h) Alboran Sea SST (western Mediterranean) (Cacho et al., 2001); (i) $\delta^{18}\text{O}_{\text{carb}}$ record from the Spannagel Cave (eastern Alps; Fig. 1a) (Fohlmeister et al., 2012); (j) flood reconstruction from Lake Ammersee (north-alpine foreland; Fig. 1a) (Czymzik et al., 2013); (k) high-resolution sediment density record from Lake Ghirla indicating flood activity; (l) paleo-NAO reconstruction (Olsen et al., 2012); and (m) variation of total solar irradiance (Steinhilber et al. 2009).

Table 1: Correlation matrix (1981-2014) of monthly (Jan-Dec) meteorological data from the station Locarno-Monti (GNIP, 1976-2011; FOEN, 1992-2014; MeteoSwiss, 2015a) and of the NAO index (NCAR, 2015). Length of used data set is limited by the time series of ET beginning in 1981. T=temperature, P=precipitation amount, H=humidity, SD=sunshine duration, ET=evapotranspiration, $\delta^{18}\text{O}_\text{P}$ =oxygen isotopic composition of precipitation.

	T	P	H	SD	ET	$\delta^{18}\text{O}_\text{P}$	NAO
T	1	0.28	-0.01	0.77	0.88	0.76	0.00
P	0.28	1	0.50	0.15	0.09	0.19	-0.20
H	-0.01	0.50	1	-0.54	-0.36	-0.08	-0.29
SD	-0.01	-0.15	-0.54	1	0.89	0.62	0.16
ET	0.88	0.09	-0.36	0.89	1	0.69	0.01
$\delta^{18}\text{O}_\text{P}$	0.76	0.19	-0.08	0.62	0.69	1	0.07
NAO	0.00	-0.20	-0.29	0.16	0.01	0.07	1

Table 2: Radiocarbon ages (top) and age control from ^{137}Cs -activity measurements and the LST (bottom) used for age-depth modeling. Radiocarbon ages were calibrated using the IntCal13 calibration curve (Reimer et al., 2013).

Lab code	Core Section	Section depth (cm)	Sample material	Depth (cm)	Depth without event layers (cm)	^{14}C age (yr BP)	1 σ	Calibrated age (cal yr BP), 2 σ
OS-117514	GHI09-1-A2	7.5-11	macrofossils	128.5	-	355	20	317-491
ETH-39227	GHI09-2-A1	12-13	leaf fragments	188.7	-	1055	35	923-1055
ETH-43301	GHI09-1-A3	31.5-32.5	macrofossils, cone	252.1	-	1495	35	1306-1514
OS-117551	GHI09-2-A2	64.5-65.5	leaf	328.2	-	2010	20	1899-1999
ETH-38339	GHI09-2-A2	71.5-72.5	macrofossils	335.2	-	2120	35	1994-2299
ETH-39228	GHI09-2-A3	86.5-87.5	leaf	458.2	-	2680	35	2748-2850
ETH-43302	GHI09-2-B1	81.5-82	wood	546.3	-	3460	35	3640-3831
ETH-39229	GHI09-1-B3	76.5-77.5	bud leaves	589.8	586.1	3840	35	4150-4409
OS-117553	GHI09-2-B3	20.5-21.5	branch fragment	683.9	669.3	4490	30	5039-5297
ETH-38340	GHI09-2-B3	44-45	wood	707.5	686.9	4770	35	5332-5590
ETH-39230	GHI09-2-C1	74-75	fruit	808.7	759.8	5645	35	6318-6496
OS-117552	GHI09-1-C3	30-31	needles	847.2	788.8	6610	30	7440-7566
ETH-40409	GHI09-2-C2	43-43.5	branch fragment	873.7	812.5	7160	35	7883-8029
ETH-38341	GHI09-2-C3	53.5-56	wood	981.6	871.3	8380	45	9290-9491
ETH-39231	GHI09-2-D1	92.5-93	fruit	1164.7	927.7	10060	45	11338-11821
ETH-43303	GHI09-1-D3	25-26	bark	1194.7	948.2	10580	45	12420-12630

Type of age control	Core Section	Section depth (cm)	Depth (cm)	Depth without event layers (cm)	Age (cal yr BP)
^{137}Cs	GHI09-SC2	7.5	7.5	-	-36
^{137}Cs	GHI09-SC2	22	22	-	-13
^{137}Cs	GHI09-SC2	34-37	35.5	-	-1
LST	GHI09-2-D2	65.3-68.3	1236.2	988.1	12880*

* Brauer et al. (1999)

Table 3: Summary of results of catchment study realized in spring 2013 and 2014. Top: Average δD_{wax} of tree leaves and δD_{river} sampled along altitude profile (Fig. 3). Also provided are average apparent fractionation factors between leaf waxes and river water ($\epsilon_{\text{wax/river}}$). Bottom: Feb-Mar-Apr averages of daily instrumental data (MeteoSwiss, 2015a), and of monthly data of water isotopes in precipitation (FOEN, 1992-2014). FMA=Feb-Mar-Apr.

	2013	1σ	2014	1σ
$\delta D_{\text{river}} (\text{‰})$	-64	2	-62	1
$\delta D_{\text{wax}} (\text{‰})$	-131	11	-131	12
$\epsilon_{\text{wax/river}} (\text{‰})$	-71	14	-78	11
$\epsilon_{\text{wax/FMA-P}} (\text{‰})$	-30		-48	

	2013	2014	2014-2013
T (°C)	7.4	10.2	+37 %
P (mm)	5.2	6.1	+17 %
S (h)	5.2	5.9	+13 %
H (%)	65	63	-3 %
ET (mm)	1	1.5	+42 %
Days with snow	9	5	
P on days with snow (mm)	48	68	
$\delta D_{\text{FMA-P}} (\text{‰})$	-104	-92	
$\delta^{18}\text{O}_{\text{FMA-P}} (\text{‰})$	-14.3	-12.5	

Table 4: Correlation matrix of sedimentary proxy data of Lake Ghirla emphasizing the absence of a simple relationship of δD_{wax} with other variables, in particular with the CT number representing flood activity.

	Plant- wax δD	Plant-wax conc.	TOC content	CT number
Plant-wax δD	1	-0.16	0.03	0.02
Plant-wax conc.	-0.16	1	0.54	-0.56
TOC content	0.03	0.54	1	-0.67
CT number	0.02	-0.56	-0.67	1

Table 5: Pearson correlation factors and corresponding p-values (calculated with Gaussian-kernel-based cross-correlation; Rehfeld and Kurths, 2014) between Lake Ghirla δD_{wax} and comparison data sets presented in Figure 7. Correlations were calculated for the Holocene, 0–11'000 cal yr BP, thus excluding the recent climate change and the Younger Dryas. Bold font: correlations with p-value <0.05. ‘Detrended’: omitting long-term trends in data sets. Plots illustrating subtracted trends are presented in Figure S3.

	δD_{wax}	p-value	Detrended		Reference	Plot in Fig. 7
			δD_{wax}	p-value		
T-anomaly N-hemisphere	0.78	0.000	0.31	0.002	Marcott et al., 2013	a
%Ti, Cariaco Basin	0.59	0.000	0.10	0.168	Haug et al., 2001	b
SST, Alboran Sea	0.26	0.036	0.07	0.284	Cacho et al., 2001	h
$\delta^{18}\text{O}$, GISP	0.23	0.004	0.14	0.027	Grootes et al., 1993	d
$\delta^{18}\text{O}$, Spannagel Cave	-0.45	0.007	-0.10	0.169	Fohlmeister et al., 2012	i
NAO reconstruction, Greenland	-0.40	0.000	-0.17	0.078	Olsen et al., 2012	l
Flood activity, Lake Ammersee	-0.17	0.150	-0.04	0.346	Czymzik et al., 2013	j
Δ Total Solar Irradiance	-0.13	0.075	0.04	0.293	Steinhilber et al., 2009	m
Flood activity, Lake Ghirla	0.07	0.249	-0.06	0.301	Wirth, 2013	k

Figure1

* meteorological station Locarno-Monti

Figure3

Figure4

Legend to lithological profile

0-568 cm core depth:

- Bioturbated background sediments (BT)
- BT rich in detrital material (BT-high-detr)
- BT slightly enriched in detrital material (BT-semi-detr)

568-1265 cm core depth:

- Regular background sediments (RS)
- Flood layer (FL)
- Mass-movement deposit (MMD)
- Clastic sediments (CL)

Figure6

Figure 7