Land-use evolution in the catchment of Lake Murten, Switzerland

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\textbf{Highlights:}
\begin{itemize}
  \item Pollen and leaf waxes from Lake Murten record land-use evolution since prehistory
  \item Large-scale Roman deforestation and farming around the city of Aventicum
  \item Land-use and vegetation recovery times following Roman disturbance took centuries
\end{itemize}

\textbf{Abstract:}

Anthropogenic soil erosion is a problem of global concern and recently has become the focus of extensive research. In spite of this, our knowledge about the history of land-use and its long-term impact on soil erosion and the local environment remains limited. This study seeks to address this issue by investigating sediments of Lake Murten, Switzerland, using a multi-proxy approach to reconstruct the history of land-use and its impacts in the catchment. We analyzed pollen and charcoal to reconstruct past land-use and vegetation dynamics, and used the distributions of terrestrial leaf wax biomarkers, their $\delta^{13}$C isotopic composition and their soil retention time (compound-specific $^{14}$C) to evaluate long-term effects on past soil carbon dynamics.
Arboreal pollen abundances, charcoal influx and cultural indicators match the archaeological evidence and reveal an eventful past around the lake. The first signs of human presence were detected around 5000 BCE, when Neolithic pile dwellers occupied the lake's shores. However, human land-use had no significant effect on the pollen and the sedimentary organic matter (OM) composition during Neolithic times and the Bronze Age. This changed during the Late Iron Age and the Early Roman Period (ca. 70 BCE). Coincident with the rise of Aventicum, a Roman city, large-scale deforestation and agriculture began in the region. Severe soil degradation and outwash of soil organic carbon (SOC) at this time is documented by enhanced input of soil-derived and pre-aged leaf waxes, and resulted in cultural eutrophication ca. 2000 years ago. Soil erosion decreased after the fall of the Roman Empire and a short period of renaturation followed. Although the export of SOC returned to pre-Roman values after ca. 200 years, the forest never recovered to its past extent. The last two detected periods of land-use change correlate with the onset of Medieval agriculture (ca. 1000 CE) and the Industrial Period (ca. 1800 CE). Today, the mean transit time of leaf waxes is almost five times longer compared to the Roman Period, suggesting that substantial soil erosion has occurred and that an even longer time period would be necessary for the soil carbon dynamics to recover to their natural state.

Key words: Land-use Reconstruction, Pollen, Charcoal, Leaf Waxes, Compound-Specific δ¹³C, Compound-Specific Radiocarbon Dating

1. Introduction

Soil erosion as a consequence of human land-use is a pervasive problem of global concern (Lal, 2004; Quinton et al., 2010). Land-clearing, field cultivation and pastoralism are considered to be the land-use activities that are the most influential drivers of soil erosion. Such landscape modifying activities do not only have direct physical impacts on soils, but also have indirect impacts, including changes in local carbon dynamics (Foster et al., 2003). Soil organic carbon (SOC) is the largest terrestrial carbon reservoir on Earth (Quinton et al., 2010) and the magnitude of the changes in local carbon dynamics due to the mobilization, transport and redistribution of SOC has been much-debated in the context of land-use change (Lal, 2005; Van Oost et al., 2007). In particular, our knowledge of the long-term impact of soil degradation and its historical
evolution is rather limited. Past land-use and its legacy can influence terrestrial and aquatic ecosystems, including ecological functions, for centuries or even longer (Foster et al., 2003). Analyzing soil erosion rates in a historical to pre-historical context allows pre-anthropogenic conditions, the long-term influence of intensive land-use periods and the effects of renaturation periods to be evaluated.

Sedimentary repositories are promising archives that can be used to improve our understanding of the relationship between land-use history and SOC dynamics. Terrestrial archives integrating a catchment wide signal, such as lake or peat sediments, are natural recorders of environmental change and can be used to study soil erosion in a historical perspective (Huang and O’Connell, 2000). During phases of human land-use, i.e. activities that modify landscapes, the structure of top- and subsoil gets destroyed (Lal, 2002). As a consequence minerogenic and chemical erosional products from the impacted catchment soils, including SOC, are mobilized and often compiled in sedimentary repositories such as lakes (Edwards and Whittington, 2001). Previous studies have documented that organic matter (OM) and specific organic fractions deposited during land-use periods differ significantly in their bulk geochemical properties and in their isotopic composition compared to the material deposited as background sedimentation during undisturbed periods (Enters et al., 2006; Meyers, 1994). However, sedimentary OC is very heterogeneous and contains carbon from diverse allochthonous and autochthonous sources of both geological and biological origin (Gierga et al., 2016), which dilutes the initial SOC signal and can make it difficult to identify periods of land-use change from the properties of bulk soil alone.

In the last decades, new attempts to address the complications regarding the heterogeneity of sedimentary OC have been developed with a focus on source-specific organic compound classes of terrestrial origin (Douglas et al., 2018; Galy et al., 2011). In general, only a low percentage of eroded terrestrial OC is permanently buried in the sedimentary record due to early diagenesis and microbial consumption during transit and initial deposition (Vonk et al., 2010). This leads to the selective preservation of organic compounds with relatively recalcitrant molecular structures and affiliation to mineral matrices (Vonk et al., 2010). Among the compounds that are particularly well preserved in sediments, leaf wax lipids that are predominantly produced by higher terrestrial plants are among the most studied. These long-chain n-alkanes and n-carboxylic acids
have a persistent molecular structure and are well-represented in the refractory SOC pool (Douglas et al., 2018; Feng et al., 2013; Smittenberg et al., 2006).

By analyzing leaf wax molecular distributions, isotopic compositions and relative ages, it is possible to gain specific information not only about the paleoenvironment that prevailed during the time of synthesis but also about their past (and current) retention times in soils (Douglas et al., 2018; Dubois and Jacob, 2016). In recent studies, compound-specific radiocarbon analyses (CSRA) of leaf waxes from lake sediments has confirmed that periods of land-use have long-lasting effects on soil carbon dynamics (Douglas et al., 2018, 2014; Gierga et al., 2016). In these studies, the relative retention or transit time of leaf waxes provided an estimate of soil stability through time. However, in order to clearly identify anthropogenic effects and to exclude natural background erosion, additional independent proxies, such as the study of pollen and non-pollen palynomorphs, can provide important environmental context.

Pollen, spores and non-pollen palynomorphs retrieved from lake sediment are frequently used to estimate relative changes in vegetation cover within the catchment or to identify land-use periods. Palynological proxies have emerged as a robust tool to trace paleoecological changes (Bichet et al., 2013; Hillbrand et al., 2014; Tinner et al., 2003). Periods of human land-use are often associated with significant increases in the charcoal influx due to fire activity and with the occurrence of a variety of cultural indicator species, such as Cerealia type and Plantago lanceolata pollen or spores of coprophilous fungi (Behre, 1988; Cugny et al., 2010; Tinner et al., 2003; van Geel et al., 2003).

In a previous publication we detected drastic increases in soil erosion indicators correlating with the timing of Roman land-use 2000 years ago and the Industrial Period (Haas et al., 2019). Building on this framework, we applied in the current study a more source-specific approach based on pollen and leaf wax data in order to reconstruct the evolution of land-use and its impact on soil degradation in the catchment of Lake Murten, for the past 8000 years.

First, we analyzed the distribution of pollen in the sedimentary record to develop an overview of the regional to local vegetation history, to identify human activities, and to assess paleoecological changes triggered by
human land-use. Second, we estimated the influence of human land-use on local carbon dynamics in a pre-
historical to historical context, using the distribution, isotopic composition (δ¹³C values) and relative ages
(CSRA) of terrestrial leaf waxes. This allowed us to evaluate not only the long-term influence of intensive
land-use periods on soil degradation, but also to investigate pre-anthropogenic conditions and effects of
renaturation periods on the local carbon dynamics.

1.1. Study Site

Lake Murten is situated in the lowlands of western Switzerland, also known as the Swiss Plateau region
(Figure 1). The catchment has – geologically spoken - a relatively mixed bedrock composition, with various
conglomerates, sandstones, clays and marls (Pfiffner, 2014). The hillsides, such as Mont Vully, are glacially
imprinted side moraines. Towards the south of the lake lies the alluvial plain of the La Broye river,
representing the main inflow to the lake.

**Figure 1:** Location of Lake Murten (Switzerland). Locations of human settlements around the lake from the
Neolithic to the Medieval Period are indicated by the colored regions, and gray shading represents local
topography (Castella et al., 2015; Nast, 2006; UNESCO Palafittes, 2015). Data source: swisstopo (Art. 30
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The Swiss Plateau region is rich in archaeological sites from several periods of human activity. The shores of
Lake Murten have a remarkably dense history of settlement and cultural activities (Figure 1). Beginning 7000
- 6000 years ago, Neolithic pile dweller communities lived by Lake Murten (Menotti, 2004; UNESCO
Palafittes, 2015) and their sedentary lifestyle dominated the shores of the lake for millennia. In the Late Iron
Age (La Tène cultural period, 450 – 20 BCE), large-scale deforestation and farming first occurred in the
catchment area as the population became more dense and centralized. Architectural remnants of this Celtic era
include the remains of a fortress (lat. oppidum) on Mont Vully, overlooking the area of Lake Murten and Lake
Neuchâtel (Nast, 2006) (Figure 1).
The Romans made their entrance to the Swiss Plateau in the 1st century before the Common Era (BCE).

Numerous ruins and excavations in and around the village of Avenches provide evidence of an influential Roman city called Aventicum, capital of the Helvetians, within close proximity to the lake. At the peak of its influence (2nd century common era, CE) Aventicum contained approximately 20,000 inhabitants, which led to further large-scale deforestation and agriculture in the region (Castella et al., 2015). After the fall of the Roman Empire (5th century CE), the Great Migration Period led to socio-economic changes that contributed to a decentralization of the population. Archaeological findings from this period are rare in the vicinity of Lake Murten.

Starting in the Early Medieval Period (ca. 11th century CE) human activity in Switzerland intensified (Egli, 1988). The foundation and construction of the first Medieval fortress in Murten by the Burgundians began in the year 814 CE (Schöpfer, 2016). The city of Murten remains the largest conurbation in the catchment today. The Industrial Period was characterized by several water-corrections and drainage of the swamp area north of the lake, paving the way for intensive agriculture (Nast, 2006).

In a previous study we showed that drastic increases in soil erosion indicators such as relative X-ray fluorescence titanium (XRF Ti) concentrations, magnetic susceptibility (MS) and sediment mass accumulation rates (MAR) were correlated with the timing of Roman land-use 2000 years ago and the Industrial Period (Haas et al., 2019). The bulk sediment $^{14}$C time series that was previously established (Haas et al., 2019) offers an ideal base for our present study. Increasing age-offsets between bulk sedimentary organic carbon and the modeled deposition age revealed terrestrial runoff of pre-aged SOC due to human induced soil erosion. Interestingly, with the end of the Roman Period and relaxation of human pressure, soil erosion indicators including the bulk sediment age offsets decreased and reached steady conditions around 200 years after human activity ceased, which was interpreted as a period of renaturation and soil stabilization (Haas et al., 2019).

In the present study we worked on the same sedimentary record. Here, we add a new palynological data set that allows us to expand our knowledge about the vegetation dynamics and history of agriculture in the
catchment and to clearly identify land-use activity. Our present paleoecological overview compares well with former palynological work at Lake Murten (Hadorn, 1984). In addition, in the present study we focused on specific leaf waxes of terrestrial origin, which ensures better source control than the bulk SOC studied previously, and allows us to draw direct conclusions about the impact of land-use and abandonment on soil carbon dynamics.

2. Material and Methods

2.1. Chronology

The chronology of the Lake Murten sediment record (Latitude: 46.929220, Longitude: 7.067460, Haas et al., 2019) is based on 25 radiocarbon dates on terrestrial macrofossils, varve counting and $^{137}$Cs radionuclide dating. Compared to the age-depth model of Haas et al. (2019), 15 additional radiocarbon ages on terrestrial plant macrofossil were obtained (Figure 2, appendix A). We followed the procedure of Haas et al. (2019) to prepare the additional macrofossil assemblages for radiocarbon dating on an EA-AMS (MICADAS, ETH Zürich). Furthermore, the sampling error was estimated using the sample thickness and the average sedimentation rate of approximately 8 years/cm ($\text{y/cm}$) and added to the analytical error of the radiocarbon dates using error propagation (see appendix A). Instead of classic (linear) age-depth modeling as used in Haas et al. (2019) (clam by Blaauw, 2010), we used a new approach using Bayesian statistics to reconstruct sediment accumulation rates for sediment records, implemented in the program BACON (Blaauw and Christen, 2011). BACON subdivides a sediment record into numerous vertical sections and estimates the sedimentation rate (in $\text{y/cm}$) using millions of Markov Chain Monte Carlo (MCMC) iterations. The model was calculated with a mean sedimentation rate (acc.mean) of 5 $\text{y/cm}$ and an “accumulation shape” (acc.shape) of 1.5 on 101 sections of 10 cm thickness (Figure 2). Ages in this article are reported as Before Common Era (BCE) and Common Era (CE), respectively.

The application of this new age model resulted in larger confidence intervals, and some events described in our previous publication (Haas et al., 2019) became older by approximately a 100 years. All interpreted events and changes described in Haas et al. (2019), however, fall into the 95% confidence interval (CI) of the updated age-model and thus retain their significance.
Figure 2: A. Age-depth model of the Lake Murten sediment record based on varve counting, radionuclide $^{137}$Cs and radiocarbon dating on macrofossils. The lithological description follows Haas et al. (2019).

Abbreviations: Mesolithic Period (MP), Iron Age (IA), Bronze Age (BA), confidence interval (CI).

B. Enlarged zoom of the top 40 cm of the core. C. and D. prior (green) and posterior (grey) distributions of the sedimentation rate and memory, respectively, that were used in the program BACON (appendix A) for age-depth modelling (Blaauw and Christen, 2011).

2.2. Pollen, spore and microscopic charcoal analysis

The palynological results are presented in percentages of the terrestrial pollen sum (Fig. 3, appendix B). Laboratory treatment followed standard preparation procedures for glycerin samples after Moore et al. (1991). Lycopodium tablets were added to the samples prior to chemical and physical treatment in order to estimate pollen and charcoal concentrations (grains or particles/cm$^3$) and charcoal influx (particles/cm$^2$/year, Stockmarr, 1971). Pollen, spores and microscopic charcoal particles from 40 samples (1 cm$^3$) were analyzed. In each of these, a pollen sum of >500 terrestrial pollen grains (except sample at 317.5 cm) per sample were counted. The pollen results are presented in percentages calculated on the terrestrial pollen sum. Pollen grains, spores and other non-pollen palynomorphs (NPP) were identified using palynological keys and photo references (Beug, 2004; Moore et al., 1991; Reille, 1995). Coprophilous fungal spores were identified following van Geel et al. (2003). Microscopic charcoal particles were counted according to Finsinger and Tinner (2005) and Tinner and Hu (2003). In total, 146 palynological taxa were identified in the sedimentary record of Lake Murten. The results (counts) were normalized relative to the total pollen sum and displayed as percentages. The pollen record was subdivided into three statistically significant local pollen assemblage zones (PMUR-1-3) using constrained clustering, implemented in the program ZONE 1.2 (Juggins, 1991), and the broken stick model (BSTICK) to determine the number of statistically significant zones (Bennett, 1996). A diagram with a selection of the most important pollen, spore and non-pollen palynomorphs for the paleoecological overview and the discussion of land-use activity can be found in the appendix B.

2.3. Leaf Wax Extraction, Purification and Quantification
Total lipid extracts (TLEs) were obtained from ~10g of freeze-dried, ground, and homogenized sediment from each selected depth using a microwave extraction system (Milestone, MLS 1200 mega) with 10 - 15 ml of 10% methanol in dichloromethane (DCM) (2 min with 300 W from room temperature to 70°C and 5 min with 500 W at 70°C). The TLE was eluted over an aminopropyl column (1 g, Sigma Aldrich, LC-NH2 SPE Bulk) with different solvents of increasing polarity. The first fraction (containing n-alkanes) was eluted with 10 ml of n-hexane, the second fraction with 4 ml of 20% DCM in n-hexane, the third fraction with 4 ml of 10% acetone in DCM and the fourth fraction (containing n-carboxylic acids) with 2% formic acid in DCM.

The fractions containing n-carboxylic acids were derivatized to form fatty acid methyl esters (FAMEs) using 10 ml of 5% hydrochloric acid (32%) in methanol and heated at 70°C for 12 h. The organic phase was recovered by liquid-liquid extraction with n-hexane. The resulting FAMEs were purified with a silver nitrate coated silica gel pipette column (0.4 g, Sigma Aldrich) using different solvents of increasing polarity. The first fraction was eluted with 2 ml of n-hexane and the second fraction (containing the saturated n-carboxylic acid FAMEs), with 4.5 ml of 20% DCM in n-hexane.

The fractions containing n-alkanes were purified using a zeolite pipette column (0.2 g, Geokleen, GHGeochemical Services) with n-hexane following the method of Haas et al. (2017). Aliquots of the purified n-alkane and n-carboxylic acid samples were analyzed by gas chromatography mass spectrometry (GC-MS, 5977B MSD, Agilent Technologies) and quantified for their concentrations, average chain lengths (ACL, after Poynter and Eglinton (1990)) and carbon preference indices (CPI, after Bray and Evans (1961)) on the GC-MS using the flame ionization detector (FID) attached to the same instrument. To minimize 14C contamination only external n-alkane (nC7-40) and n-carboxylic acid standards (nC16+22+24+28) were used for quantification. A summary of the leaf wax data is added in the appendix C.

2.4. Compound-specific radiocarbon dating

Nine samples with highest concentrations of leaf waxes were selected for compound specific radiocarbon dating. Specific n-alkane homologues n-C27, n-C29, n-C31, n-C33 and n-C35 were collected using a GC-FID (7890B, Agilent Technologies) coupled to a preparative fraction collector (PFC, Gerstel). The individual
Compound fractions were recovered from the PFC glass traps by elution with 1 ml n-hexane. Column bleed was removed by eluting the samples with n-hexane over a silica gel pipette column (0.1 g, Fluka Analytical). The transfer of the compound fraction into an EA tin capsule (Säntis AG) was performed by repetitive dissolution and transfer with DCM. A heat plate (37°C) was used to evaporate the DCM. To obtain sufficient quantities for radiocarbon dating, several compound fractions needed to be pooled (13 samples for CSRA) and in some cases the bulk n-alkane fraction, containing every chain length, was used for radiocarbon dating (19 bulk n-alkane samples). A sample list including the radiocarbon results is given in appendix D.

Radiocarbon analyses of the n-alkane fractions were performed on an accelerated mass spectrometer (AMS) equipped with an elemental analyser (EA) at the Laboratory for Ion Beam Physics of ETH Zürich, Switzerland (MICADAS, Wacker et al., 2013). To monitor recoveries and 14C-backgrounds, specific n-alkane standards (n-C28 and n-C32) with known 14C-concentration (Fm of 0 and Fm of 1.073, respectively) were processed. Samples that went through compound separation (GC-PFC) were corrected for the constant background contamination with GC-PFC processed n-C28 and n-C32. Bulk n-alkane fractions were corrected with unprocessed n-C28 and n-C32. A constant contamination correction was applied according to Hanke et al. (2017)(A summary of the compound-specific radiocarbon data is given in appendix E). Based on the blank assessment only data with errors (Fm) < 8% and sample masses larger than 10 µg of carbon were taken into account for the discussion.

All radiocarbon ages were calibrated with IntCal 13 (Reimer et al., 2013). Specific ages in this article are calibrated and represent the 95% confidence interval and median age. The results were compared with the macrofossil based age model and bulk sediment ages from Haas et al. (2019) and 15 additional measurements following the same procedure (Figure 2, appendix F). The mean transit time (MTT) represents the difference between the bulk sediment age (Median) and the n-alkane age (Median) to the age model (i.e. sedimentation age) and is reported in years. MTT errors were calculated using error propagation (appendix E).

2.5. Compound-specific δ13C isotope analysis
FAME and $n$-alkane $\delta^{13}C$ values were determined by gas chromatography – isotope ratio mass spectrometry (GC-IRMS) at Eawag’s Center for Ecology, Evolution and Biogeochemistry in Kastanienbaum, Switzerland. A GC-1310 (Thermo Scientific, Bremen, Germany) was interfaced to a Delta Advantage IRMS (Thermo Scientific) with a ConFlow IV (Thermo Scientific). Samples were injected with a TriPlusRSH autosampler to a split/splitless injector operated in splitless mode at 280 °C. The oven was equipped with an InertCap 5MS/NP column (0.25 mm x 30 m x 0.25 μm) (GL Sciences, Japan) and was heated from 80 to 215°C at 15°C min$^{-1}$, then to 320°C at 5°C min$^{-1}$, and then held at 320°C for 10 minutes. Column effluent was combusted at 1020 °C.

Raw isotope ratios were converted to the VPDB scale using Thermo Isodat 3.0 software and pulses of reference gas at the beginning and end of each analysis. The sample $\delta^{13}C$ values were further corrected using the linear regression of measured and known values for a standard mix of $n$-alkanes ($n$-C$^{17, 19, 21, 23, 25, 28, 34}$) of known isotopic composition (provided by Arndt Schimmelmann, Indiana University), which was run at varying concentrations at the beginning and end of each sequence, as well as after every 6-8 injections of samples. This standard was also used to monitor for instrumental drift and for any isotope effects associated with retention time or peak size. The average standard deviation for these standards during the analysis period was 0.6 ‰ ($n = 182$), and their average offset from known values was -0.1 ‰. An additional quality control standard of $n$-C$_{29}$-alkane was analyzed at least three times per sequence, and had a standard deviation of 0.18 ‰ ($n = 15$) during the analysis period. FAME $\delta^{13}C$ values were corrected for the carbon added during methylation by isotopic mass balance, and reported errors represent propagated errors from the replicate measurements and from the uncertainty associated with the added carbon, which was determined by methylating and analyzing phthalic acid of known isotopic composition (provided by Arndt Schimmelmann, Indiana University).

### 3. Interpretative Framework and Results

#### 3.1. Palynological indications for vegetation dynamics and land-use in the catchment

The relative abundances of the tree, shrub and herb pollen, microscopic charcoal influx (particles/cm$^2$/y), as well as cultural indicators such as *Cerealia* type and *Plantago lanceolata* (ribwort plantain) pollen, and spores
from coprophilous fungi have proven to be robust indicators for evaluating land-use dynamics and vegetation change (see Tinner et al. (2003) for further information). We focused on these proxies to reconstruct the history of agriculture and divided the pollen record into three local pollen zones (PMUR-1-3) in order to assess statistically significant vegetation changes (Fig. 3, appendix B).

The sediment record reaches back to approximately 5800 BCE, which falls into the Late Mesolithic Period (start of PMUR-1). There is no archaeological evidence of human settlements in the Swiss Plateau region at that time (UNESCO Palafittes, 2015). During PMUR-1, a dominance of pollen of temperate trees and shrubs such as Ulmus, Quercus, Tilia, Acer, Fraxinus, Corylus and Hedera helix and monolette fern spores were present, suggesting that mixed oak forests (Quercetum mixtum) dominated the region around Lake Murten (ca. 70% tree pollen during the Neolithic Period, Fig. 3). At the same time pollen of Abies and Fagus reached their empirical limit, suggesting that these trees expanded locally (Birks and Tinner, 2016). Fagus sylvatica (beech) and Abies alba (silver fir) are both shade-tolerant mesophilous late-successional trees species. The shift to mixed beech forests was found throughout the entire Swiss Plateau during the Neolithic Period. At Lake Murten, a representative site for the western part of the Swiss Plateau close to the Jura Mountains, Fagus sylvatica was co-dominant with Quercus (Ammann, 1989; Hadorn, 1992; Richoz and Haas, 1995).

During the Neolithic Period of the core, the charcoal influx increased to 6500 particles/cm²/y ($n = 14$), indicative of moderate fire activity in the region. Generally, the fire history is similar to the temporally high resolution charcoal results from nearby Lake Lobsigen (ca. 20 km away, Tinner et al., 2005). After ca. 4500 BCE, fire sensitive taxa such as Ulmus, Tilia, Fraxinus, Acer and Hedera helix decreased, while pollen of pioneer species such as Betula, Corylus avellana and Salix and heliophilous Juniperus shrubs increased.

These results are consistent with the overall slash and burn agricultural practices of the Neolithic Period. Plantago lanceolata pollen and Sporormiella spores were scattered throughout this sedimentary interval, but at concentrations that are suggestive of low levels of human activity during the Neolithic Period (Fig. 3).

Towards the end of the Neolithic Period (2540 BCE; Figure 3), the first pollen occurrence of Cerealia type (t.) was detected, revealing a probable human activity and field cultivation in particular. Around 2200 BCE, the
cultural period of the early Bronze Age started. However, no crucial vegetation change is visible in the overview pollen diagram from this time (Figure 3). The only indication of further human influence is a slight rise in charcoal influx coupled with an increase of pollen from more disruption-resistant *Fagus* and *Urtica*.

Starting towards the end of the Late Bronze Age period (ca. 1100 BCE, PMUR-2), crucial changes in the vegetation around the lake were observed, roughly coincident with the onset of the Iron Age cultural period (800 - 20 BCE). We observed a slow but significant decrease in tree pollen percentages, down to 30% (650 CE, Figure 3). In the course of this forest decline, *Fagus* and *Abies* pollen in particular decreased substantially. Furthermore, a notable peak in the charcoal influx of 14'360 particles/cm²/y occurred around 430 BCE (Figure 3). This so called Iron Age-peak in charcoal influx has also been observed at other sites in the Swiss Plateau and south of the Alps (Tinner et al., 2003). The timing of deforestation seems to correlate not only with the elevated charcoal influx, but also with the beginning of a continuous curve in the cultural indicator proxies, such as Cerealia t., *Plantago lanceolata* or Poaceae pollen (Figure 3). As Cerealia t. pollen are dispersed locally and do not reach large distances (Soepboer et al., 2007; Tinner et al., 2003), these results suggest cereal fields were in close proximity to Lake Murten. The agricultural intensification during the Late Iron Age to Roman Period was accompanied by a substantial expansion of meadow and ruderal species such as *Artemisia*, Cichorioideae, *Ranunculus acris* t. and *Rumex acetosa* t. A similar development has also been observed in pollen records from other sites in the Swiss Plateau, such as Lake Lobsigen (Ammann, 1989).

During the Iron Age, dung-spores (*Sporormiella*, Fig. 3) rose continuously and remained elevated throughout the Roman Period, where a temporal maximum of 1.4% at 350 CE was identified. Cerealia t. pollen percentages also increased steadily during the Roman Period (20 BCE to 5th century CE, Fig. 3). Furthermore, the Roman Period stands out clearly in the pollen record and is characterized by the introduction of new crops such as *Castanea sativa*, *Juglans regia* and *Secale cereale*.

The last local pollen zone PMUR-3 started ca. 380 CE and was characterized by further deforestation (until 650 CE, 30% tree pollen), mostly at the expense of *Abies* and *Fagus*. Following the deforestation peak (650 CE), tree pollen percentages increased again to 40-50%, mainly due to the increase of *Quercus* pollen percentages (Fig. 3). From the Medieval Period to the 19th century, oak stands provided an important
livelihood in Switzerland, consistent with our findings. Cereal cultivation (Cerealia t.; Secale cereale) was also prevalent during this time. Herb pollen percentages of cultural meadow and ruderal taxa such as Plantago lanceolata, Poaceae, Urtica, Artemisia, Trifolium pratense t., etc. were present at a continuously high level, indicating ongoing land-use in the nearby catchment area until today. The afforestation trend was accompanied by a simultaneous increase in the shrubs Juniperus and Salix but also slight increases in Calluna and Ericaceae pollen. The heathland components commonly grow on open patches at rather nutrient-poor and extensive meadows.

Figure 3: Palynological indications for past land-use around Lake Murten, Switzerland. A. Abundance of Cerealia type (including Secale cereale) and Plantago lanceolata pollen as well as Sporormiella spores. B. Charcoal influx. C. Normalized abundance of tree, shrub and herb pollen, as well as the subdivision into significant local pollen zones 1-3. The thick lines in A and B represent trends derived from a locally weighted scatterplot smoothing (LOESS, span = 0.3, n = 80). Abbreviations: Mesolithic Period (MP), Roman Period (R), Modern Times (MT).

3.2. Sedimentary leaf wax distributions

The range and relative molecular abundances of sedimentary leaf waxes have been widely used to trace environmental changes (Bush and McInerney, 2013). Leaf waxes are produced to coat the vascular leaf surface, and serve as an outer protection against the environment (Eglinton and Hamilton, 1967). Their molecular components, such as long-chained n-alkanes or n-carboxylic acids, have a refractory structure and are well preserved in the environment over geological time scales (Eglinton and Eglinton, 2008). A common ratio to describe leaf wax distribution patterns is the average chain-length (ACL) of n-alkanes or n-carboxylic acids (Bush and McInerney, 2013) (Formula in appendix C). For sediments it has been proposed that the ACL of n-alkanes is a broad indicator of shifts in the catchment’s plant community. A predominance of relatively long-chained n-alkanes (or n-carboxylic acids) is typical for leaf waxes originating from higher terrestrial plants (Eglinton and Hamilton, 1967). In contrast, n-alkanes from more aquatic sources, such as
microalgae or macrophytes, typically have relatively high abundances of middle and short-chained homologues (Ficken et al., 2000; Sachse et al., 2004).

In the sedimentary record of Lake Murten, ACL values of \( n \)-alkanes varied between 27.8 and 28.9 with a mean of 28.4 (\( n = 71 \), Figure 4). \( n \)-C\(_{29}\) alkane was the most abundant homologue. The predominance of relatively long-chained \( n \)-alkanes in the record indicates contributions of terrestrial leaf waxes sourced from higher plants. ACL trends from Lake Murten sediment (Figure 4) are correlated with arboreal pollen abundances (Figure 3), supporting the assumption that the leaf waxes are predominantly of terrestrial plant origin.

The carbon preference index (CPI) measures the abundance of odd-numbered relative to even-numbered \( n \)-alkane chain-lengths (formula in appendix C). High CPI values (>1) are characteristic for most modern plants (Bush and McInerney, 2013). Hence, in a sedimentary sequence CPI can provide an information on whether the \( n \)-alkanes are likely derived from a plant source, or whether they have been subject to significant microbial degradation or petrogenic inputs, both of which are associated with lower CPI values and greater relative contributions from even-numbered \( n \)-alkane homologues (Haas et al., 2017; Tao et al., 2015).

The \( n \)-alkane distributions from the sediments of Lake Murten had CPI values ranging from 4.3 to 9.4 with a mean of 7.2, demonstrating a strong dominance of odd-numbered \( n \)-alkanes (appendix C). We thus conclude that fossil leaf wax contribution from bedrock and microbial degradation seem to be minimal and that the leaf waxes are well preserved in the sediments.

The ACL and CPI values of sedimentary \( n \)-carboxylic acid distributions also support the conclusion that leaf waxes in Lake Murten are predominantly sourced from terrestrial leaf waxes (summarized leaf wax data is given in appendix C). However, \( n \)-carboxylic acid and \( n \)-alkane distributions are highly variable within different plant groups, species and even the plant itself and can be influenced by various environmental factors, such as temperature and relative humidity (Bush and McInerney, 2013). Furthermore, the original (terrestrial) leaf wax signal can be diluted during transport, deposition or degradation by addition of
compounds of other sources (Bush and McInerney, 2013). This problem of source control can be mitigated by using a compound-specific approach, focusing only on compound fractions of relatively known terrestrial origin.

3.3. $\delta^{13}C$ isotopic composition of sedimentary leaf waxes

The carbon isotopic composition of specific long-chained leaf waxes can be useful for inferring relative leaf wax sources (Diefendorf and Freimuth, 2017). Plant physiology, carbon fixation mechanisms (which differ for C$_3$ and C$_4$ plants) and environmental factors such as relative humidity and temperature are responsible for pronounced differences in the $\delta^{13}C$ signature of plant OM (Farquhar et al., 1982). Analyzing $\delta^{13}C$ values of specific leaf wax compounds in sediment allows relative inputs from various OM sources to be discriminated (Rieley et al., 1991).

In Lake Murten, the $\delta^{13}C$ values of sedimentary long-chained leaf waxes ($\delta^{13}C_{lw}$), including the n-C$_{28}$, n-C$_{30}$, n-C$_{32}$ carboxylic acids and n-C$_{29}$ and n-C$_{31}$ alkanes, ranged from -32.9 to -37.3‰ with a mean value of -34.6‰ (n = 41) (Figure 4). They were depleted by approximately 4‰ relative to the bulk sedimentary isotopic composition ($\delta^{13}C_{bulk}$) measured by Haas et al. (2019), which ranged from -28.2 to -32.0‰ with a mean value of -30.1‰ (n = 62). The $\delta^{13}C$ values of shorter homologues such as n-C$_{21}$ alkanes or n-C$_{20}$ carboxylic acids were in the same range as their longer chained counterparts (appendix D). The relatively depleted carbon isotopic composition is consistent with the $\delta^{13}C$ signature of higher terrestrial plants following the C$_3$ pathway for carbon fixation (Mays et al., 2017; Rieley et al., 1991). This assumption is supported by relatively high ACL and CPI values for both n-alkanes and n-carboxylic acids. Since the $\delta^{13}C$ values of long- and short-chained n-alkanes and n-carboxylic acids differed only slightly relative to each other, we assume a similar terrestrial leaf wax source for all these compounds. The offset of approximately 4‰ between $\delta^{13}C_{bulk}$ and $\delta^{13}C_{lw}$ values is attributed to the biosynthetic depletion of leaf waxes relative to the carbon isotopic composition of the bulk plant tissue (Collister et al., 1994). Differences between $\delta^{13}C_{bulk}$ and $\delta^{13}C_{lw}$ values can also occur if the sedimentary OM incorporates isotopically-enriched OM from autochthonous sources such as algae (Rieley et al., 1991). This possibility is unlikely in Lake Murten given the elevated ACL values and the relatively depleted $\delta^{13}C$ values of the short-chained n-carboxylic acids.
The evaluation of simultaneous changes among different compound-specific $\delta^{13}$C signatures is an additional approach to interpret changes in the sedimentary leaf wax sources (Diefendorf et al., 2010; Yamamoto et al., 2010). The $\Delta \delta^{13}$C offset (‰) represents the difference between specific long- and short-chained leaf wax $\delta^{13}$C values (Figure 4). In Lake Murten, the $\Delta \delta^{13}$C offset between $n$-C$_{32}$ and $n$-C$_{28}$ carboxylic acids ($\Delta \delta^{13}$C$_{32-28}$) showed the largest variations and seemed to react more sensitively to leaf wax source changes, compared to $\Delta \delta^{13}$C$_{32-30}$ and $\Delta \delta^{13}$C$_{30-28}$ (Figure 4). During most parts of the Neolithic Period and the Bronze Age, $\Delta \delta^{13}$C$_{32-28}$ values were positive, with the longer homologue typically ~2‰ enriched relative to the shorter one. Starting with the Iron Age, however, $\Delta \delta^{13}$C$_{32-28}$ decreased steadily and reached negative values during the Roman and the Medieval Period, indicating that $n$-C$_{28}$ carboxylic acid became relatively $^{13}$C-enriched relative to $n$-C$_{32}$ carboxylic acid. The overall decrease in $\Delta \delta^{13}$C from the Late Iron Age to the Medieval Period (Figure 4) is coeval with a decrease in the abundance of trees in the catchment (Figure 3).

One possible explanation is that during most parts of the Neolithic Period and the Bronze Age, a greater proportion of the $n$-C$_{28}$ carboxylic acids were sourced from trees, which on average have leaf waxes with lower $\delta^{13}$C values than do non-woody C$_3$ plants (Diefendorf and Freimuth, 2017; Magill et al., 2013). With ongoing deforestation, relatively more of the sedimentary $n$-C$_{28}$ carboxylic acid would have been sourced from non-woody C3 plants, making it more enriched than before and thus lowering the $\Delta \delta^{13}$C$_{32-28}$ during phases of land-clearing. This interpretation is supported by the overall variability in ACL and the arboreal pollen abundances.

Enhanced primary production in Lake Murten could also explain the negative shift in $\Delta \delta^{13}$C$_{32-28}$ values that began in the Late Iron Age and continued into the second half of the Medieval Period. $n$-Carboxylic acids produced by algae are relatively enriched in $\delta^{13}$C compared to terrestrial sources, and $n$-C$_{28}$ carboxylic acids can be produced by microalgae (van Bree et al., 2018; Volkman et al., 1980). Increased primary productivity within the lake could therefore also lower $\Delta \delta^{13}$C$_{32-28}$ values by increasing the $\delta^{13}$C values of $n$-C$_{28}$ carboxylic acids. However, clear evidence of enhanced primary production and eutrophic conditions that led to varve formation have only been observed during the Late Iron Age to Early Roman Period (200 BCE - 50 CE, with
the updated age-model, Haas et al., 2019). We assume that increased contributions of n-C$_{28}$ carboxylic acids from microalgae would only be relevant for these documented periods of hypoxia. Furthermore, if decreases in the $\Delta \delta^{13}C_{32-28}$ offset were caused by enhanced algal productivity, we would expect them to correspond to a decrease in the n-carboxylic acid ACL, which was not the case.

3.4. Relative ages of sedimentary leaf wax compounds

In addition to the complications stemming from multiple sources for specific leaf wax compounds, their relative ages might also vary depending on the time needed for the cascade from synthesis to primary deposition in soils, to transport and finally to re-deposition in sediments (Douglas et al., 2014). Compound-specific radiocarbon dating (CSRA) allows the mean transit time of leaf waxes (MTT$_{lw}$) between plant synthesis and deposition in the sedimentary environment to be estimated (Douglas et al., 2018). Relative changes in MTT$_{lw}$ are closely associated with soil processes that can influence the recalcitrant “deep” SOC pool (Douglas et al., 2018; Feng et al., 2013; Smittenberg et al., 2006). It has been proposed that pre-aged leaf waxes, as part of the recalcitrant SOC pool, are introduced into the carbon cycle during phases of land-use, when the structure of top- and subsoil gets disturbed (Butman et al., 2015; Douglas et al., 2018; Lal, 2002). In a sedimentary archive, periods of human land-use may thus be recorded in terms of increasing age-offsets (i.e. MTT$_{lw}$) between individual long-chained leaf waxes and the corresponding deposition age (Douglas et al., 2014; Gierga et al., 2016). This has the advantage that the influence of a potential hard water effect can be limited substantially compared to analyses on the heterogenous bulk sedimentary OC due to better source control (Gierga et al., 2016). The predominance of relatively long-chained n-alkanes (or n-carboxylic acids) in the sediments of Lake Murten is characteristic for leaf waxes originating from higher terrestrial plants (increased ACL, Eglinton and Hamilton, 1967). The ACL even increased with ongoing land use during the Roman agricultural period, suggesting that the increased age-offsets are unlikely to be due to an increase of aquatic plant material which might have radiocarbon ages affected by hard water. Additional evidence that the leaf wax age-offsets are unlikely to be caused by aquatic plant inputs comes from the relatively depleted $\delta^{13}C$ values of these compounds ($\delta^{13}C$ from -32.9 to -37.3‰). These are consistent with a C3 terrestrial plant origin, as aquatic plants tend to be somewhat more enriched in $^{13}C$ (Fry and Sherr, 1984). In Lake Murten, we thus interpret increases in MTT$_{lw}$ as an indicator of enhanced input of soil-derived and pre-aged leaf waxes,
corresponding to periods of increased soil erosion. Decreasing MTT_{lw}, on the contrary, is interpreted as a sign of soil stabilization.

Compound-specific $^{14}$C dating of long-chained $n$-alkanes as well as bulk $n$-alkane fractions revealed that mean ages of leaf waxes in Lake Murten sediments are hundreds of years older than the estimated deposition age of the surrounding sediment. On average the MTT_{lw} ranged from -600 to 4850 years with a mean of 450 years (Figure 4). Due to low sample masses and the applied contamination corrections (appendix E) the errors of the $n$-alkane $^{14}$C ages were relatively large (approximately 700 y, $n = 32$). Compound-specific $n$-alkane samples with masses below 20 µg carbon had the highest errors. In three cases (50, 1110 and 1900 CE) we were able to date two different fractions from the same bulk $n$-alkane sample. In all cases the ages of the different chain-length fractions differed significantly by several hundred years. However, we assume that this effect was caused by the strong sample mass dependency of radiocarbon dating of small samples rather than a primary environmental signal (Hanke et al., 2017). Most of the reported ages that seem too young (e.g. all $n$-C$_{31-33}$ alkane fractions, Figure 4) were measured on samples with a relatively low carbon content (ca. 12 µg C, $n = 3$). The effect of mass dependency was also visible when comparing the ages and errors of compound-specific fractions (all < 20 µg C) and bulk $n$-alkane fractions (all > 20 µg C). Certain measurement artifacts could be countered by applying a locally weighted scatter-plot smoothing function (LOESS) including both compound-specific and bulk fractions. This smoothing function includes the assumption of a similar leaf wax source for short and long-chained homologues in the sediments of Lake Murten, which is supported by compound-specific $\delta^{13}$C measurements and the sedimentary leaf wax distributions (more details can be found in appendix C). Due to the relatively large errors in the CSRA data, not all changes in MTT_{lw} are significant. For this reason we only describe broad trends in the MTT_{lw} data and compare it to MTT_{OC}, which has much smaller errors.

Figure 4: A. Average chain length of $n$-alkanes $n$-C$_{23-33}$ (ACL) from sediments of Lake Murten, Switzerland. B. $\delta^{13}$C composition of leaf waxes. C. $\Delta\delta^{13}$C offset. D. Mean transit time of different $n$-alkane fractions
(MTT\textsubscript{lw}) and bulk sediment (MTT\textsubscript{OC}). The thick lines represent trends derived from LOESS (span \sim 0.3, n = 80). Abbreviations: Mesolithic Period (MP), Roman Period (R), Modern Times (MT).

4. Discussion: History of land use in the Lake Murten catchment

The first signs of human activity in the catchment area of Lake Murten appeared during the Neolithic Period, when several pile dweller communities occupied the lake shore (UNESCO Palafittes, 2015). In the Alpine region, early pastoral activity and slash and burn practices to gain arable land during this time are indicated by elevated charcoal influx, as well as the first occurrences of Sporormiella spores and Plantago lanceolata pollen (Dietre et al., 2017; Schwörer et al., 2014; Tinner et al., 2003; Wehrli et al., 2007). Although there is evidence of human activity around Lake Murten during this interval, no significant changes were observed in the sedimentary leaf wax distribution (ACL) and the overall $\delta^{13}$C\textsubscript{lw} isotopic composition (Figure 4). The MTT\textsubscript{lw} trend, on the other hand, indicates increased input of pre-aged leaf waxes from 5200 to 3400 BCE ($n = 3$). Also, MTT\textsubscript{OC} values suggest that input of pre-aged organic carbon increased during the same interval. Early human activity might be responsible for the excursions in MTT\textsubscript{lw} and MTT\textsubscript{OC} values. An alternative explanation comes from the lithological succession of the core. The early interval with elevated MTT\textsubscript{lw} and MTT\textsubscript{OC} values is mostly part of Unit 5 (5600 - 3700 BCE), which is composed of sediments with high detrital element concentrations (Haas et al., 2019). This suggests either a new source of sediments, which could be due to naturally occurring river diversions, or a climatically caused increase in soil erosion corresponding to wetter climate conditions as suggested by Magny (2004).

Unit 5 (957 - 766 cm) was deposited during a reported lake level high-stand for 26 lakes in Western Switzerland and France that occurred in the period of 4400 - 3950 BCE (Magny, 2004). In the Swiss lowlands phases of higher lake-level were triggered by enhanced annual precipitation and lower summer temperatures (Magny, 2004), leading to an outwash of detritus and probably soil material, including pre-aged leaf waxes. The age-depth model was updated substantially compared to the model in Haas et al. (2019). As a result, the progression of the age-depth model during the Neolithic Period is rather uncertain (Figure 2, Unit 5, 957 - 766 cm) and the variable MTT values are strongly dependent on the age-depth model. The largest confidence intervals (95%) for the age-depth model occurred during Unit 5 (ca. 500 years, Figure 2) due to two
radiocarbon dates that lie slightly outside the curve (Figure 2). If the model would be changed so that it would pass through these points more clearly, the corresponding deposition ages would have been older and thus the MTT_{lw} and MTT_{OC} values during Unit 5 much smaller. As a consequence, the errors in this part of the core would have been unrealistically small, which is why this version of the model was omitted (See appendix A for the alternative age-depth model).

All data in the lower part of the core until ca. 3700 BCE should be interpreted with caution. There are indications that the last two data points in the age-depth model could also be classified as uncertain due to a biostratigraphic event, namely the rise of *Abies alba* and *Fagus sylvatica* around 6000 BCE, that was observed in other palynological publications near the study area (Tinner et al., 2018). In the sediments of Lake Murten the first rise of *Abies alba* (above 2%) occurred slightly earlier approximately 5000 – 5500 BCE (see appendix B). This could imply that the deposition ages of the lowermost sediments could also be older by ca. 500 – 1000 years, and that the age-model rather shows a minimum age (see also appendix A for an alternative age-depth model). However, the resolution of the palynological analysis would need to be increased substantially in order to verify this assumption. After 3700 BCE, the age-depth model is relatively stable until today (Figure 2) and more accurate statements about environmental changes in the catchment can be made.

Towards the transition into the Bronze Age, around 2500 BCE, increasing *Sporormiella, Plantago lanceolata* percentages and also the first occurrence of Cerealia t. pollen were documented (Figure 3), demonstrating that human activity including cattle raising and arable farming in close proximity to the lake shore were increasing. This result is also in close agreement with archaeological evidence. Numerous pile dwelling remains from the Neolithic Period were found in and around Lake Murten and dated using dendrochronology (Schibler and Jacomet, 2010). The interval of increased human activity (ca. 3000 - 2000 BCE) in the Lake Murten sedimentary record corresponds well with the reported ages of pile dwellings excavated in Faug, Vallamand and Muntelier (Figure 1, UNESCO Palafittes, 2015).

Despite new innovations in agricultural field cultivation (as indicated by Cerealia t. pollen), no significant changes in the leaf wax distributions and $\delta^{13}C_{lw}$ signature could be detected at this time (Figure 4). Only the MTT_{lw} gradually decreased, whereas no changes in the bulk sediment age offset were visible during this
period. Based on the long-lasting decrease in $MTT_{lw}$ and the disagreement with land-use indicators, we assume climatic controls were more important than anthropogenic controls on the leaf wax transit time during this period. Magny (2004) also suggested that warmer and dryer climatic conditions prevailed during this period (ca. 2500 BCE), favoring wheat production. Decreased precipitation might naturally lower the outwash of SOC and pre-aged leaf waxes without changing their distributions or $\delta^{13}C$ values.

During the Bronze Age, from 2200 - 800 BCE, land-use indicators remained low, even though nine archaeological sites of pile dwellings are known from this time (Figure 1, UNESCO Palafittes, 2015). The only signs of land-clearing were a minor increase in the charcoal influx and decrease in tree pollen percentages in the Early Bronze Age (1500 BCE). This timing also roughly correlates with one elevated $MTT_{lw}$ value, suggesting a short period of soil erosion. However, a higher sample resolution would be needed to confirm that land-clearing and shorter periods of soil erosion existed during this interval.

During the Iron Age, 800 - 15 BCE, tree percentages started to decrease steadily and the charcoal influx rose to a distinct peak, ca. 430 BCE, indicating severe land clearing (Figure 3). The interval of deforestation during the La Tène cultural period (Late Iron Age) to the Early Roman Period was also accompanied by sharp increases in Sporormiella, Plantago lanceolata and Cerealia t. percentages, indicating the beginning of large-scale farming with cattle raising and field cultivation in the region (Figure 3). The earlier increase in Sporormiella spores might suggest that pastoralism was dominant during the Late Iron Age. This past land-use period corresponds nicely with the increases in sediment mass accumulation rate and detrital input documented in our previous study (Haas et al., 2019).

Evidently, land-use and deforestation also had an influence on the leaf wax composition. With the onset of the Late Iron Age, ACL increased significantly to a temporal maximum of 28.8 at 140 BCE (Figure 4). At the same time, $\Delta \delta^{13}C$ offsets started to decrease steadily, suggesting a change in the leaf wax source towards open grasslands. $MTT_{lw}$ and $MTT_{OC}$ trends increased simultaneously during the Iron Age, consistent with the growing influence of agriculture on soil erosion.
The deforestation trend continued with the onset of the Roman Period, reflecting the high demand for wood and arable land that was needed for the newly founded Roman city Aventicum and its 20,000 inhabitants, at the southern shore of Lake Murten (Castella et al., 2015) (Figure 1). Increased land-use indicators such as Sporormiella, Plantago lanceolata and Cerealia point towards large-scale farming and the extension of human pressure on the environment.

In the leaf wax record, the Roman Period was accompanied by relatively high but decreasing ACL (from 28.4 to 28.0, 70 BCE – 280 CE) and a slight decrease towards more $^{13}$C-depleted values for the leaf waxes (Figure 4). The $\Delta^{13}$C offset increased slightly despite the overall downward trend, due to a pulse of $^{13}$C-depleted $n$-C$_{28}$ carboxylic acids. This effect might originate from the affiliation of leaf waxes with soil aggregates. Mineral matrix protection of leaf waxes could have served as an effective protection from microbial degradation or re-synthesis, which leads to a relative enrichment in $\delta^{13}$C (Douglas et al., 2018; Matsumoto et al., 2007; Vonk et al., 2010). The elevated CPI of about 9 at 110 CE (appendix D) would support the assumption that less microbially influenced leaf wax material affiliated with soil aggregates was washed into the lake during this time.

Correlating with the onset of Late Iron Age to Early Roman land-use, soil erosion indicators drastically increased, consistent with our previous study (Haas et al., 2019). The mean transit time of bulk sedimentary OC (MTT$_{OC}$ of 1500 years) peaked at 170 BCE with the updated age-model, revealing terrestrial runoff of pre-aged SOC due to human induced soil erosion. In the leaf wax record, a similar peak was detected in the transit time (MTT$_{lw}$ of around 700 years from 70 BCE to 150 CE), supporting the hypothesis of Early Roman soil degradation in the catchment (Figure 4). There was minimal time-lag of around 100 years between the MTT$_{lw}$ and MTT$_{OC}$ maxima. This might be a source effect of the two different organic fractions. Sedimentary OM has a very heterogeneous composition, containing OC of various allochthonous and autochthonous sources of different origin and ages (Gierga et al., 2016). Additionally, bulk sediment $^{14}$C data is prone to so-called “hardwater effects”, depending on the bedrock composition of the catchment. Due to the dissolution of limestone, algae and aquatic plants incorporate dissolved inorganic carbon (DIC) that is considered radiocarbon dead, which may lead to a dilution of the bulk sediment $^{14}$C-concentration (Fowler et al., 1986).
The sedimentary leaf waxes, on the contrary, are predominantly of terrestrial instead of aquatic origin and offer a better source control. We thus posit that the MTT$_{lw}$ maximum during the Early Roman Period was mainly the result of anthropogenic soil erosion.

The input of pre-aged leaf waxes might have an effect on common ratios such as ACL, CPI and $\delta^{13}$C$_{lw}$ records (Douglas et al., 2014). Due to the time lag between synthesis and sedimentary deposition, the environmental signals might be delayed by the transit time of the individual compounds. In the Lake Murten setting, however, this effect appears to be restricted to the periods of land-use. During times of land-use and enhanced soil input we detected rising ACL, $\Delta\delta^{13}$C offsets and MTT$_{lw}$ but decreasing $\delta^{13}$C$_{lw}$ values (Figure 4). The described excursions correspond well with the palynologically derived land-use periods and do not show significant time lags, suggesting that only a smaller portion of the long-chained leaf waxes were pre-aged.

During undisturbed times, the sedimentary leaf wax distribution (ACL and $\Delta\delta^{13}$C offsets) seems to have been controlled by vegetation and the ecological changes inferred from the independent pollen record (Figure 3).

After the fall of the Roman Empire, during the Early Medieval Period, land-use indicators such as the charcoal influx as well as Sporormiella or Plantago lanceolata percentages decreased (ca. 650 CE). At this point the forest around Lake Murten had been decimated to ca. 20% of its initial extent. Historically, these sediments were deposited during the Great Migration Period (ca. 4th to 6th century CE), which is poorly represented in archaeological findings (Nast, 2006). Tinner et al. (2003) documented a reduction of agricultural activities around the 6th century CE accompanied by a short period of renaturation in three Swiss lakes. In the Lake Murten pollen record, afforestation was also identified, but several hundred years later, ca. 700 CE (Figure 3). The timing discrepancy can either be assigned to the low sample resolution, an inaccurate age-depth relation, and/or to regional differences in population discontinuities in Switzerland.

In the leaf wax record a minimum in the ACL trend occurred (ACL of ca. 28.2, 350 CE), which might be indicative of a slight renaturation and forest regrowth during this period (Figure 4). This agrees with the MTT$_{lw}$ values, which steadily decreased and reached a temporal minimum of -340 years at 390 CE, suggesting soil stabilization due to vegetation regrowth. A similar trend in the MTT$_{OC}$ was observed. Since Aventicum
most likely had its largest population in the first two centuries CE, approximately 200 - 300 years of soil stabilization was needed to reach MTT_{lw} values similar to those before the great land-clearing episode.

After 1000 CE, the tree pollen percentages decreased slightly, probably marking the growing influence of Medieval agriculture in the region (Figure 3). Charcoal influxes, however, remained at a relatively low level (around 7100 particles/cm²/ y), which might indicate that less effort was needed to re-open the shrub-dominated lands compared to the Late Iron Age to Early Roman Period. Palynological indicators, such as Cerealia t. and Plantago lanceolata pollen and Sporormiella spores, were present in approximately the same range as in the Roman Period, demonstrating active Medieval land-use in the catchment. The observed changes also agree with historical developments. The Burgundians built the Early Medieval fortress of Murten in 814 CE on a pre-existing village back from Celtic to Roman times (documented as Muratum, 515 CE) (Schöpfer, 2016).

In the leaf wax record, Medieval land-use was only partly recorded. ACL rose to relatively high values (>28.5, 1000 CE) starting in the second half of the Medieval Period, probably indicating elevated input of leaf waxes of terrestrial origin (Figure 4). Around the same time, MTT_{lw} rose to 1900 years (900 CE), reaching a temporal maximum. In the MTT_{OC} no temporal maximum was detected but rather a steady increase (Haas et al., 2019). This agrees with the results of our previous study, where we documented elevated detrital input during the Medieval Period (Haas et al., 2019). Building on previous argumentation we thus assume that an increase of human-induced soil erosion was responsible for this aging-effect in the sedimentary leaf-wax record.

Around 1830 CE, the charcoal influx increased again and tree pollen percentages reached another minimum, revealing a short period of deforestation during the course of the industrialization (Figure 3). Cultural indicators such as Plantago lanceolata, and new indicators, e.g. Platanus, Zea mays, and Carum carvi, increased significantly, suggesting high human activity in the catchment. This last development agrees with the mechanization of agriculture, drainage of swamps and several water level corrections that accompanied the intensification of agriculture in the region around the lake (Nast, 2006).
The effect of land-use on the leaf wax composition is more pronounced during the Industrial Period compared to the Roman or Medieval land-use periods (Figure 4). Simultaneous changes in the $\delta^{13}$C values of leaf wax compounds, towards more depleted values, and the steep increase in MTT$_{lw}$ (Maximum of 4850 years, 1950 CE) show that soil erosion increased substantially. New agricultural innovations, including mechanization, allowed deeper plowing and most likely led to a more efficient mobilization of the deep SOC pool. Furthermore, the large-scale water corrections that were applied during this time contributed to declining lake surface levels and to the draining of the large swamp areas north of the lake to make new land for agriculture available. Therefore, destabilization of the peatlands, leading to soil subsidence and erosion, might also be responsible for the recent increase in MTT$_{lw}$.

5. Conclusions

A detailed reconstruction of the land-use history and its long-term impact on the environment in the Swiss Plateau was produced through a combination of lake sediment pollen, spore, non-pollen palynomorphs and microscopic charcoal analysis, as well as the evaluation of the distribution, carbon isotopic composition and the relative ages of sedimentary leaf waxes in Lake Murten. Although pile dweller communities of the Neolithic Period and the Bronze Age left their traces in the sedimentary record (low level slash and burn agriculture, first cultural indicators for human activity), their influence on the local environment seems to have been minimal. Based on a comparison of the relative input of soil-derived leaf waxes and independent lake level reconstructions, we conclude that soil erosion during the Neolithic Period and the Bronze Age was mostly driven by precipitation.

With the onset of the Late Iron Age and the rise of the La Tène culture, human activities had a greater impact on soil erosion in Lake Murten’s catchment. At the beginning of the Iron Age (ca. 800 BCE), continuous deforestation was initiated, including severe slash and burn practices around 400 BCE (Figure 3). Cultural indicators increased distinctly. Land-use activities had an impact on the local carbon dynamics, triggering soil erosion and the outwash of SOC (Figure 4). With ongoing deforestation during the following Roman Period, the effect of human pressure on the environment grew further.
Following the rise of the Roman city Aventicum south of the lake, large-scale agriculture with field cultivation made its entrance (Figure 2). Continuous soil degradation altered the sedimentary leaf wax distribution and δ¹³C values, indicating enhanced input of terrestrial OM (Figure 4). Moreover, leaf wax radiocarbon dating revealed an increase in the mean transit time (MTTₜₖₑ) of the soil-derived compounds, suggesting that soil erosion was able to deplete the recalcitrant SOC pool. This supports the main findings from our previous study where we documented that outwash of soil OM due to soil erosion during the Early Roman Period was responsible for eutrophication of the lake, resulting in deposition of varved sediments.

After the fall of the Roman Empire human pressure decreased and a period of renaturation occurred. During the Great Migration Period (4th to 6th century CE), land abandonment and a partial regrowth of the natural vegetation in the catchment helped stabilize the soils and remediated the SOC export back to pre-Roman values in approximately 200 years, as suggested by decreasing transit times of leaf waxes (Figure 4). A similar recovery rate was described in previous studies (Haas et al., 2019; Hillbrand et al., 2014). We assume that land-use was able to deplete the SOC pool, which might have resulted in long-lasting changes of the soil properties. For instance, we detected a significant shift in the plant community after the episode of Roman land-use, with mainly open, intensive to extensive cultivated area compared to the spacious woodlands (beech forests with silver fir and mixed oak elements) that existed during Neolithic times (Figure 3).

With the onset of Medieval agriculture (ca. 1000 CE) and the foundation of the city of Murten, a new wave of soil erosion and SOC outwash started. The following intensification of agriculture is visible not only in the pollen but also in the leaf wax record (Figure 3, 4). In particular, during the Industrial Period and the mechanization of agriculture a final but most prominent period of soil degradation was detected. During the modern land-use period, we assume soil erosion was also jointly responsible for recent eutrophication in Lake Murten that started around 1950.

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7. Data Availability:
Appendices A to F containing raw data tables are made available in the Mendeley data repository.

8. References:

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Archaeological Overview

- Pile dwelling site
- Roman roads
- Lake shore reconstruction (1 CE)

**Medieval Period**, 5th - 15th century CE
**Roman Period**, 20 BCE - 5th century CE
**Late Iron Age** (La Tène), 450 - 20 BCE
**Early Iron Age** (Hallstatt), 800 - 450 BCE
**Bronze Age**, 2200 - 800 BCE
**Neolithic Period**, 5500 - 2200 BCE