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**„Can lake sediments track past extreme weather events? – Evidence of event-related elemental, molecular and isotopic changes in sediments of subalpine Lake Lunz, Austria“**

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**Lisa – Maria Hollaus BSc**

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**Mag. Martin Kainz, Privatdoz. PhD**

## **Preface – Extreme weather events and their consequences**

### Climate change & extreme weather events

Global climate change affects aquatic ecosystems directly by altered precipitation and wind patterns (Edlund et al. 2017), and increasing temperature (specifically lake temperatures are predicted to rise up to 4°C (IPCC report 2013)). As a result, extreme weather events such as floods, droughts, tropical cyclones, heat waves, heavy rains and abrupt changes in temperatures (Jentsch et al. 2007) are predicted to increase in length, frequency and/or intensity (Field et al. 2012), which will eventually lead to extinction risks for several taxa (Isaac & Williams 2013; Blois et al. 2013) and is therefore a severe threat to biodiversity and ecosystem functions of aquatic and terrestrial habitats. Furthermore, Edlund et al. (2017) states that an extreme weather event can trigger a cascade of coupled physical, geochemical and biological processes in a clear – water lake with pervasive ecosystem effects that persist for an extended time period.

### Effects of extreme weather on lake ecology

The effects of extreme weather events on lakes influence both physical and ecological components of these ecosystems (Jankowski et al. 2006, Blenckner et al. 2002). The extreme weather event, which affected Lake Lunz in 1861 and is therefore most relevant for our study, was a strong precipitation event. Heavy rainfalls enhance runoff and the input of terrestrial material like suspended solids, nutrients and DOC, which will eventually lead to an increase in turbidity, eutrophication and brownification (Jennings et al. 2012). If accompanied by a severe storm they can shift the vertical thermal structure and mixing regime of a lake (Preusse et al. 2010), which are essential to lake biogeochemical processes and plankton community composition (Straile et al. 2010; Anneville et al. 2010; Gallina et al. 2011). Case studies in northern environments showed that climate change has increased the frequency of extreme precipitation events (Kundzewicz et al. 2006) along with increasing variation in terrestrial matter runoff into lakes and streams (Travnik & Jansson 2002).

### Effects on organisms

Phytoplankton has been used in the past to track past changes in temperature and nutrients (Winder and Schindler 2004; Smol et al. 2000). Weyhenmeyer et al. (2004) suggest that heavy rainfall events might favor larger phytoplankton species (e.g. Cryptophytes, Euglenophyceae), which are often associated with increased levels of dissolved organic matter. Droughts (which often cause a lack of dissolved inorganic nutrients) on the other hand side, may stimulate the growth of smaller phytoplankton, which have the option to feed on alternative nutrient rich food sources, i.e. bacteria (Findlay et al. 2001).

Sediment core studies in boreal regions (Jensen et al. 2007; Johnson et al. 2006) also show changes in phytoplankton community composition. In some lakes, a replacement of heavily silicified diatoms (e.g., *Aulacoseira* spp.) by smaller species (e.g., *Cyclotella* spp.) had appeared as a result of changing mixing regimes and increased stability of thermal stratification (Rühland et al. 2008).

### Importance of research on extreme weather events

Since climate change and its consequences are becoming an undeniable threat to ecosystems, scientists show increasing concern towards those climatic abnormalities (Jentsch et al. 2007). Extreme weather events influence aquatic and terrestrial ecosystems in various ways, of which a lot are still undefined and require further empirical testing. Ecologists have already proposed various theories about disturbance in ecosystems and how it affects biodiversity in the past (Lepori & Hjerdt, 2006). According to the intermediate disturbance hypothesis (Osman R.W. 2008), especially the stochastic appearance of extreme weather events in combination with increasing amplitude and frequency may cause a severe threat to biodiversity and calls for quantitative long-term research, particularly because of the importance and possible vulnerability of aquatic ecosystems (Hughes et al. 2003). In future, explicit research of climate change and its effects will eventually be critical to public understanding, management of the earth's natural resources, maintaining biodiversity and for devising possible solutions for changes that have already affected humanity.

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## Abstract

Lake sediments are used as indicators of organic matter (OM) sources and dynamics within lakes and their catchments. In this study, bulk carbon and nitrogen, stable carbon isotopes, and fatty acid biomarkers were determined in settling and sedimented OM to assess how recent and past hydrological extreme events affected the OM composition in settling particles and lake sediments in the subalpine, oligotrophic Lake Lunz, Austria. Heavier stable carbon isotope values ( $\delta^{13}\text{C}$ :  $\sim -22$  ‰) and higher C:N ratios ( $\sim 14:1$ ) were detected in a sediment layer that corresponded to a known extreme storm event that occurred 1861. Polyunsaturated fatty acids (PUFA;  $0.79 - 91.71 \mu\text{g}\cdot\text{g}^{-1}$ ), bacterial fatty acids (BAFA:  $0.18 - 122.46 \mu\text{g}\cdot\text{g}^{-1}$ ), and terrestrial fatty acids (terrFA;  $0.76 - 231.70 \mu\text{g}\cdot\text{g}^{-1}$ ), used biomarkers of mostly algae-derived, bacterial, and terrestrial OM in sediments, respectively, mostly decreased immediately below the sediment-water interface down core. Polyunsaturated FA showed a higher predictability for the presence of BAFA variability in sediments than terrestrial fatty acids (80% vs. 64%), suggesting that bacteria in these sediments depended more on algae-derived than terrestrial OM as energy-yielding substrates. The increase in BAFA coincided with lower  $\delta^{13}\text{C}$  values in the top sediment layers and a decrease of BAFA around the year 1885, when the major input of allochthonous matter into the lake occurred, accompanied by the highest measured  $\delta^{13}\text{C}$  values and C/N ratios. These high C/N ratios and heavy  $\delta^{13}\text{C}$  values at  $\sim 20$  cm depth co-occurred with the extreme storm event, suggesting that the combination of  $^{210}\text{Pb}$  as a tracer of lake sedimentation and these carbon indicators can fairly accurately back-trace historic carbon dynamics, which have been influenced by major storm events.



## Zusammenfassung

Seesedimente können als Indikatoren eingesetzt werden, die Veränderungen in der Zusammensetzung der organischen Materie und der Ökosystemdynamik in Seen und deren Einzugsgebiet widerspiegeln. Um in Erfahrung zu bringen, wie hydrologisch extreme Wetterereignisse in der Vergangenheit und Gegenwart die Zusammensetzung der organischen Materie in den Seesedimenten des oligotrophen subalpinen Lunzer Sees (Österreich) beeinflusst und verändert haben, analysierten wir die biochemische Beschaffenheit der Sedimente unter Verwendung von Sedimentprobekernen und Sedimentfallen. Durch das  $^{210}\text{Pb}$  Datierungsverfahren konnte eine durchschnittliche Sedimentationsrate von  $1.44 \text{ mm yr}^{-1}$  festgestellt werden, was es uns ermöglichte die einzelnen Sedimentschichten durchaus akkurat rück zu rechnen, bis zu einem bekannten extremen Sturmereignis im Jahr 1861. Die Sedimentschicht bei 18.5 cm lässt sich zu dem Jahr 1885 zurück rechnen, was der tatsächlichen Datierung des extremen Wetter Ereignisses (1861) sehr nahe kommt. Wir erhielten Ergebnisse zwischen -29.3‰ und -21.6‰ für die Analyse der stabilen Kohlenstoffisotope ( $\delta^{13}\text{C}$ ), welche signifikant mit den C/N Verhältnissen des Sedimentkerns in den oberen 20 cm korrelierten. Organischer Kohlenstoff (OC; 3.4 - 9.2 %) und Stickstoff (ON; 0.3 – 0.8 %) variierten ähnlich zueinander entlang der Schichten des gesamten Sedimentkerns. Ungesättigte Fettsäuren (polyunsaturated fatty acids, PUFA), welche als Maß für organische Materie, die hauptsächlich von Algen stammt, herangezogen werden ( $0.79 \mu\text{g}\cdot\text{g}^{-1}$  to  $91.71 \mu\text{g}\cdot\text{g}^{-1}$ ), bakterielle Fettsäuren (BAFA;  $0.18 \mu\text{g}\cdot\text{g}^{-1}$  to  $122.46 \mu\text{g}\cdot\text{g}^{-1}$ ), und terrestrische Fettsäuren (terrFA;  $0.76 \mu\text{g}\cdot\text{g}^{-1}$  -  $231.70 \mu\text{g}\cdot\text{g}^{-1}$ ) zeigten einen hauptsächlich abnehmenden Trend entlang des Sedimentkerns, beginnend unter der Sediment – Wasser Grenzschicht (etwa 5cm

unter der Sedimentoberfläche). Ungesättigte Fettsäuren konnten die Variabilität in bakteriellen Fettsäuren mit 80% Wahrscheinlichkeit besser vorhersagen, als terrestrische Fettsäuren mit 64%, was zu der Annahme leitet, dass die von Algen produzierte organische Materie mehr Energie für die bakterielle Fraktion bereit stellt, als terrestrische Fettsäuren. Die Zunahme von bakteriellen Fettsäuren überschnitt sich mit niedrigeren  $\delta^{13}\text{C}$  Ergebnissen in den oberen Sedimentkern Schichten und einem Abfall an BAFAs um das Jahr 1885, als der größte Eintrag von allochthoner Materie bei einem extremen Sturm stattgefunden hat. Gleichzeitig mit diesem Ereignis traten auch die höchsten gemessenen  $\delta^{13}\text{C}$  Werte und C/N Verhältnisse bei ~20 cm im Sedimentkern auf, was suggeriert, dass eine Kombination aus  $^{210}\text{Pb}$  als Tracer von Seesedimenten und den verschiedenen verwendeten Kohlenstoff Indikatoren, das durchaus akkurate Zurückverfolgen von extremen historischen Wetterereignissen ermöglicht.

## **CONTENT**

|                                                                       |                  |
|-----------------------------------------------------------------------|------------------|
| <b><u>PREFACE – EXTREME WEATHER EVENTS AND THEIR CONSEQUENCES</u></b> | <b><u>2</u></b>  |
| <b><u>ACKNOWLEDGEMENTS</u></b>                                        | <b><u>38</u></b> |
| <b><u>ABSTRACT</u></b>                                                | <b><u>8</u></b>  |
| ZUSAMMENFASSUNG .....                                                 | 9                |
| <b><u>CONTENT</u></b>                                                 | <b><u>11</u></b> |
| <b><u>FIGURES, TABLES &amp; ABBREVIATIONS</u></b>                     | <b><u>12</u></b> |
| <b><u>1. INTRODUCTION</u></b>                                         | <b><u>14</u></b> |
| <b><u>2. MATERIAL&amp;METHODS</u></b>                                 | <b><u>20</u></b> |
| 2.1 Study site.....                                                   | 20               |
| 2.2 Methods.....                                                      | 20               |
| 2.2.1. Sediment cores.....                                            | 20               |
| 2.2.2. Sediment traps.....                                            | 24               |
| 2.2.3. Data analysis.....                                             | 24               |
| <b><u>3. RESULTS</u></b>                                              | <b><u>24</u></b> |
| 3.1 Sediment cores.....                                               | 24               |
| 3.3.1 Biota in sediments .....                                        | 28               |
| 3.2 Sediment traps.....                                               | 29               |
| <b><u>4. DISCUSSION</u></b>                                           | <b><u>31</u></b> |
| 4.1 Sediment cores.....                                               | 31               |
| 4.2 Sediment traps.....                                               | 34               |
| <b><u>5. CLIMATE CHANGE &amp; FUTURE PROSPECTS.....</u></b>           | <b><u>36</u></b> |
| <b><u>6. APPENDIX</u></b>                                             | <b><u>37</u></b> |

## Figure caption

Fig. 1: Documentation on a house close to Lake Lunz of the water level after the flooding event in 1861.

Fig. 2: Location of subalpine Lake Lunz (608 m.a.s.l.) in Austria with the sampling site for settling particles and sediment cores at the deepest lake station (33 m depth).

Fig.3: Vertical distribution of C/N ratios and  $\delta^{13}\text{C}$  values in the sediment core of 2015.

Fig.4: Vertical distribution of organic nitrogen (%) and organic carbon (%) of sediment core 2015.

Fig.5: Correlation plot of  $\delta^{13}\text{C}$  values and C/N ratios in sediment layers (0-40 cm depth) taken at the deepest spot of Lake Lunz, Austria ( $R^2 = 0.84$ ,  $p < 0.0001$ ).

Fig.6: Vertical distribution of algal- (PUFA) and bacterial fatty acid (BAFA) biomarkers in the sediment core of 2015.

Fig.7: Linear regression analysis of PUFA and BAFA biomarkers in 'core2015';  $R^2 = 0.8$ ,  $p < 0.0001$

Fig. 8: Linear regression analysis of BAFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) and terrFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ );  $R^2 = 0.64$ ,  $p < 0.0001$

Fig.9: Vertical distribution of PUFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) and terrFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) of core 2015.

Fig.10: Vertical distribution of terrestrial fatty acids ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) and total phosphorus ( $\mu\text{g} \cdot \text{L}^{-1}$ ) of core 2015.

Fig.11: Distribution of total phosphorus ( $\mu\text{g} \cdot \text{L}^{-1}$ ) along the  $^{210}\text{Pb}$  dated sediment layers of core2015 (2016 – 1729).

Fig.12: Annual precipitation from 1913 until 2016; calculated mean precipitation of  $1620 \text{ mm} \cdot \text{yr}^{-1}$

Fig. 13: Accumulated carbon ( $\text{mg} \cdot \text{m}^{-2}$ ) over deployment period in the upper (15m) and lower (30m) sediment trap.

Fig. 14: Mean PUFA and terrestrial fatty acid concentrations ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) with standard deviations of the 15 m and 30 m placed sediment traps.

Fig. 15: Vertical distribution of biological fossil remains (cladocera – blue tones, copepods - red diatoms – green, tones, zygnemophyceae – yellow/orange tones) of the sediment core retrieved in 2015.

## Tables

Table 1: Different types of lipids and fatty acids used as source indicators for OM of bacterial communities, bulk algae, macroalgae, vascular plants and diatoms.

Table 2: Comparison of different alpine and subalpine lakes in respect of their depth, lake area and sedimentation rates.

Table 3: Accumulated carbon in the 15m (up) and 30m (low) positioned sediment traps over the time of deployment period and per day.

Table 4: Different fatty acid types and their maximum and minimum values, of the analysed sediment cores

## **Abbreviations**

DOC – dissolved organic carbon

POC – particulate organic carbon

FA – fatty acids

PUFA – Polyunsaturated fatty acids

BAFA – bacterial fatty acids

TerrFA – terrestrial fatty acids

TP – total phosphorus

OC – organic carbon

ON – organic nitrogen

DW – dry weight

## 1. Introduction

Lake sediments are composed of allochthonous and autochthonous organic matter (OM) and thus represent a mixture of various OM sources over time (Meyers & Ishiwatari, 1995). Allochthonous OM in sediments consists of mostly decomposed terrestrial plant and soil material (Lozovik et al, 2005), whereas autochthonous OM is based on lake primary producer and consumer material (Dean et al, 2006). Since the deposition of these OM sources has varied in the past, their quantity and biochemical quality had also been subject to change that may be tracked in sediments (Meyers & Ishiwatari, 1995).

Biological remains preserved in lake sediments can be used as indicators for various changes, such as temperature (Smol et al., 1988), variations in nutrient supply and OM sources to lakes (i.e., paleolimnological approach: Vreca et al., 2010; Perga et al., 2015). Thus, high-resolution paleolimnological investigations strongly rely on a variety of biochemical indicators, including stable isotopes (Leng et al, 2003), C/N ratios (Kaushal S. & Binford M.W. 1999), lipids and their fatty acids (Kainz et al, 2004 & 2009), total phosphorus (Wilson et al, 2010), and sediment core dating using the decay of radioactive isotopes, such as  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  (O'Reilly et al, 2010).

Carbon and nitrogen, as elemental C/N ratios and their stable isotopes, are commonly applied to examine the OM composition of lake sediments. Stable carbon and nitrogen isotope values ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) in combination with C/N ratios are used as indicators of OM sources (Talbot et al., 2001), in particular to differentiate between autochthonous and allochthonous OM. For example, algae-derived OM contains C/N ratios between 5 to 8, whereas C/N ratios in vascular plants are typically higher (Meyers et al., 1994). Phytoplankton OM as well as C3

plants show  $\delta^{13}\text{C}$  values ranging between -33‰ and -22‰, whereas C4 plants contain much heavier  $\delta^{13}\text{C}$  values (e.g., from -15 to -10‰; Vreca & Muri 2010). Thus, these biomarkers can often provide sufficient differentiation of OM sources in sediments, however, if pooled together, can also challenge source-specific OM assessments.

Lipids and their fatty acids in sediments can provide additional information on OM sources in sediments (Dunn et al, 2008). An important difference between stable isotopes and fatty acids in OM source tracking in sediments is that fatty acids can be reported as mass fractions (e.g., mg fatty acid per unit sediment mass) and thus be used for quantifying loads of different OM sources. Such source assessments gain particular value when related to sedimentation rates, as a particular time can be associated to the retention of specific OM source deposition. Furthermore, some lipids can be fairly resistant compounds to OM degradation (Muri & Wakeham, 2006) and are therefore useful for paleolimnological research. Using lipids and fatty acids as OM source indicators has been successfully applied for a number of aquatic ecosystems to identify different OM sources, including bacterial communities, algae, vascular plants, and diatoms (Kainz et al., 2009; Currie and Johns, 1988; Volkman et al., 1980; Rajendran et al., 1993; Nichols et al., 1982; Canuel et al., 1995; Kharlamenko et al., 1995; Meziane et al., 1997) .

The OM content in lake sediments depends on the allochthonous OM load, autochthonous production, and subsequent OM degradation, which also cause alteration of the OM composition (Meyers & Ishiwatari, 1995). Organic matter degradation already starts in the water column, thus OM deposited on lake sediments may not entirely reflect the biogeochemical composition of its origin (Meyers & Eadie, 1992). For example, sediment trap studies in Lake Michigan show

that nearly 85% of the carbon is oxidized before leaving the epilimnion (Eadie et al., 1984). The OM sinking time is strongly influenced by lake depth and wind-induced lake water movements, and bioturbation at the upper sediment layers can alter the OM composition and microbial OM degradation (Meyers & Ishiwatari, 1995). Depending on the biochemical OM reactivity with autochthonous OM tending to be more reactive than allochthonous, settled OM will either remain in the sediments or be utilized by microbes and benthic feeders (Meyers & Ishiwatari, 1995).

Seasonal OM sedimentation patterns in a lake can be reflected by plankton succession stages (Matthews & Mazumder, 2005), but may also be altered by allochthonous OM input from the lake catchment (Fuentes et al 2012). Extreme weather events, such as heavy rainfalls, supply considerable loads of terrestrial material into lakes (Raymond & Saiers, 2010). Storm events have the most impact right after spring mixing when they influence seasonal metabolic balance and reduce primary productivity for a larger period of the growing season (Sadro & Melack 2015). Mountainous catchments might be especially affected, since steep slopes and thin soils cause accelerated water runoff into streams and lakes (Sadro & Melack, 2015), but may deliver less OM content to lake sediments than OM supply from lowland lake catchments with thicker soil layers.

Previous studies documented how OM compounds within the water column and in sediments change over time in large lakes with rather large and flat catchments, such as the Great Lakes in North America (Meyers & Eadie, 1992; Kruger et al, 2015; Meyers P. A., 2006; Neilson & Stevens, 2011; Cory et al, 2016; Bouchard V., 2007). Within the European Alps, several lakes, including subalpine Lake Lunz, Austria, are well documented for over 100 years in terms of their water temperature and lake ice conditions (Kainz et al, 2017). However, little is known about



sedimentation rates, OM composition in sediments and recently settled OM particle composition in subalpine lakes within high relief–energy. To fill such gaps, the objectives for this study were to;

- a) compare the sedimentation rate of the 34 m deep, oligotrophic, subalpine Lake Lunz with other subalpine, alpine, but also flatland lakes,
- b) assess the particulate organic carbon (POC) content, the biochemical OM composition, and OM sources in lake sediments as tracers of previous extreme weather events (descriptive and predictive biogeochemical approach), and,
- c) explore how the presence of zooplankton remains (counts/measures of carapace) can be related to total carbon, stable carbon isotope values (as bulk source indicators), and fatty acid source biomarkers (algal, bacterial, and terrestrial sources).

We hypothesized that,

- a) OM quantity decreases over time (down-core) because of selective OM degradation, thus,
- b) the most labile OM, as characterized by PUFA, degrade faster than the more recalcitrant terrestrial OM, and,
- c) past extreme weather events can be traced back by concurrent increased accumulation of terrestrial OM in sediments.

## **2. Materials & Methods**

### **2.1 Study site**

Lake Lunz (Lunzer Untersee), an oligotrophic subalpine lake (608 m.a.s.l.), is located in Lower Austria (47.85° N, 15.05° E) in the eastern Austrian Alps. The glacially formed lacustrine basin covers an area of 68 ha with a maximum depth of ~33 m. The region typically remains snow covered during winter months with long-term average annual air temperature of 6.7°C (Fasching et al., 2015) and a precipitation average of 1608 mm per year.

### **2.2 Methods**

Lake sediment cores (historical OM record) and settling particles in sediment traps (recent OM deposition) were collected to investigate temporal dynamics of settling and sedimented OM in Lake Lunz.

#### **2.2.1 Sediment Cores**

Three sediment cores were collected using a Kajak – Brinkhurst gravity corer at the deepest point of Lake Lunz (33 m) in September and November 2015. Each core had a maximum length of 60 cm, in which about 40 cm of sediments were retrieved. The cores were sub-selected into 0.5 cm slices, which resulted in 80 samples per core. Immediately after collection, all sediment samples were stored either in 50 mL Eppendorf tubes or 20 mL plastic scintillation vials, kept at -80°C and subsequently freeze-dried.

One of the sediment cores was used solely for biotic community counts of diatoms, cladocera and crustaceae, using a binocular compound microscope (100 – 400x magnification). The core samples assigned for the organism count were

stored at cryogenic temperature ( $-80^{\circ}\text{C}$ ) to preserve biological remains. Subsequently, samples were thawed and filtered through a  $40\ \mu\text{m}$  sieve using untreated water. The other sediment samples were used for total phosphorus, carbon, nitrogen analysis, and fatty acid analysis, and for OM dating using  $^{210}\text{Pb}$ . In all cores, the redox-potential (Eh) and pH analyses were taken at 0.5 cm intervals downcore, before the cores were sub-selected.

To measure the total phosphorus content of the sediment samples, we decided on an approach that included an acidic digestion with nitric acid, according to the CEM Microwave Instruction Manual (2006) combined with a molybdate - ascorbic acid colour reaction for eighty samples with 100 – 110 mg sediment each. The treated samples were measured using a photometer (HITACHI™ U2800).

Total nitrogen and carbon were measured using an elemental analyser (THERMO Elemental Analyzer Flash 2000 with MAS200R). Forty sediment samples were acidified using hydrochloric acid (35%) in a fumigation glass chamber for twelve hours to remove inorganic carbon. Thereafter, the absence of carbonates before and after fumigation was verified by adding hydrochloric acid (10%) to the samples, as there was no carbonate degassing visible after applying the acid to the samples after fumigation. Additionally, we analysed 10 sediment samples, which were not treated with acid beforehand, to achieve an estimation of the inorganic carbon content of the sediment samples. Additionally, stable carbon ( $^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) isotopes were measured with the elemental analyzer alongside this process to track changes of carbon sources and trophic levels.

Sediment dating was performed (GÉOTOP, Université du Québec à Montréal, Canada) from one reference core using radiometric methods ( $^{210}\text{Pb}$ ), which provided data for assessing annual sedimentation rates of the lake. Further

calculations, to determine chronological age of each sediment layer were performed according to Ghaleb et al. (2009).

Fatty acids in lake sediments were used to assess algal, bacterial, and terrestrial OM sources. Total lipids and their fatty acids were extracted on freeze-dried, homogenized, pre-weighed (100 – 120 mg dry weight) sediment samples. The process of obtaining lipids out of sediments involved three steps: extraction, gravimetric analysis, and esterification. By running all steps under nitrogen aeration and keeping chemicals and samples on ice or in cold environment (-20°C, -80°C), the preservation of fatty acids in the lake sediments was assured.

To achieve a separation of the organic layers, a mixture of chloroform - methanol (2:1), ice – cold methanol, and a 0.9% NaCl - water solution was added to small centrifuge vials (10 mL) containing the sediments. Afterwards the samples were placed in an Elma S15 Ultrasonic bath for 10 minutes. During this time, the bath was covered with crushed ice to assure low temperatures. After interfusing the samples for 1 minute each, we centrifuged them for 5 minutes at 4°C with 3000 rpm. The extraction process was repeated three times, trying to get the greatest possible amount of lipids out of the samples. The bottom layer inside the centrifugation vials was transferred into 8 mL glass vials after each repetition. Using a gas evaporator (N-EVAP 111) we reduced the extracted solution to 1.5 mL under nitrogen flow. At this point of the process, samples can be optionally stored at -20°C until further analysis.

We applied a gravimetric analysis to determine the total amount of lipids in the sampled sediments. One hundred µL of the extracted solution were transferred into previously weighed tin capsules (duplicates) and left under the fume hood for

about 2 hours, until they were completely dried off. Accordingly, the tin capsules were weighed again with a micro scale (SARTORIUS™ CPA2P). Further calculations, to predict the total lipid concentration and obtain the required volume for methylation and gas chromatography, were done in Microsoft Excel<sup>®</sup>. Due to low total lipid concentrations in the sediment samples, all extracted lipids were used for the derivatization of fatty acids to methyl esters (FAME).

Prior to the derivatization, we added C19:0 as internal standard to the total lipid extract that was subsequently evaporated under a gentle flow of nitrogen. Thereafter, ice-cold toluene (1 mL) was applied as a solvent along to a sulfuric acid (1%, H<sub>2</sub>SO<sub>4</sub>) – methanol (CH<sub>3</sub>OH) solution and sealed under nitrogen. The samples were then placed in a water bath at 50°C for 16 hours. After the samples had cooled down to room temperature again, 2 mL of KHCO<sub>3</sub> (2%) and 5 mL of hexane (C<sub>6</sub>H<sub>14</sub>) were added. To release produced CO<sub>2</sub>, the centrifuge vials have to be shaken, opened up again to release the gaseous components and sealed under nitrogen. The samples were vortexed, followed up by 2 minutes of centrifuging at 4°C with 1500 rpm. The upper layers containing FAME were transferred into centrifuge vials (14 mL). To extract the highest possible amount of FAME the methylation process was repeated 3 times. The vials containing the transferred samples were then placed in a warm water (35°C) and evaporated under a gentle flow of nitrogen. Once the samples were evaporated, the vials were rinsed with ice – cold hexane (1.5 mL) to wash all the fatty acids off the inside of the vial. This solution was then transferred into labeled GC vials and, using the nitrogen gas flow once more, was reduced to the calculated volume (in the case of the sediments 0.5 mL for the entire sampling series). Finalized samples can be stored in the GC – vials at -80°C for gas chromatography.

FAME were analyzed using a gas chromatograph (TRACE GC THERMO) equipped with flame-ionization detection, a temperature- programmable injector and an auto-sampler. To perform the FAME separation a Supelco<sup>TM</sup> SP-2560 column (100 m, 25 $\mu$ m i.d., 0.2 mm film thickness) was used (McMeans et al., 2015). The fatty acid concentrations were calculated using calibration curves based on known standard concentrations (FAME C19:0 standard added to the samples during the procedure).

#### 2.2.2. Sediment traps

Sediment traps were deployed to assess the current sedimentation rate of settling particles. Sediment traps were placed at the center of the pelagic zone at a depth of 15 m (beneath the photic zone) and 30 m (just above the sediments) and retrieved every three weeks, in triplicates, for a period of one year. Due to ice-cover during winter months, no sediment trap sampling was possible from January to March 2015. Samples were filtered onto pre-combusted and pre-weighed GF/C filters, stored at -80°C and freeze-dried before being weighed to assess sedimentation rates per time units (e.g., days). For lipid and fatty acid analysis, freeze-dried and weighed samples were transferred into glass tubes (15 mL) and, before being stored at -80°C, chloroform and methanol was added and kept under nitrogen. The fatty acid analysis was performed as described above.

#### 2.2.3. Data analysis

Data were analyzed using SPSS and the statistical software R version 3.3.3 (R Core Team 2017). Linear regression analysis was applied to identify factors that could explain variation in the biogeochemical parameters in the water

column (15 and 30 m) and the lacustrine sediments of Lake Lunz.

### 3. Results

#### 3.1. Sediment cores

The average annual sedimentation rate of Lake Lunz, based on the  $^{210}\text{Pb}$  decay measurements in the upper sediment layers, was  $1.44 \text{ mm yr}^{-1}$ . Stable carbon isotope values ( $\delta^{13}\text{C}$ ) ranged between  $-29.35\text{‰}$  and  $-21.56\text{‰}$ . The  $\delta^{13}\text{C}$  values significantly correlated with C/N ratios in the top 20 cm of the sediment core; however, two  $\delta^{13}\text{C}$  peaks were recorded at 1.5 cm ( $-26.59\text{‰}$ ) and 27.5 cm ( $-25.61\text{‰}$ ).  $\delta^{13}\text{C}$  values decreased steadily from the detected peak ( $\sim 1885$ ) to the upper sediment layers (3.5 cm). The most noticeable  $\delta^{13}\text{C}$  peak occurred in 1885, which coincided with an event of high terrestrial organic matter input caused by a massive flood event in 1861 (historic record; Fig. 1). In addition, concurrent increases in C/N ratios at this sediment layer indicate higher terrestrial OM sources (Fig. 3).  $\delta^{15}\text{N}$  peaked at 4.5 cm (coincides with the years around 1986) with  $4.76\text{‰}$ , followed by its minimum value  $0.66\text{‰}$  at 2.5 cm ( $\sim 1999$ ).

Organic carbon (OC) and organic nitrogen (ON) showed similar fluctuations between a maximum of 9.16% and a minimum of 3.43% for OC and a maximum and minimum of 0.77% and 0.28% for ON (Fig. 4). The OC and ON content was highest at 1.5 cm below the sediment-water interface, followed by a drop to the minimum value of OC at 6.5 cm. The lowest recorded value of ON was located at 19.5 cm.  $\delta^{13}\text{C}$  values and C/N ratios correlated significantly in these sediment layers ( $R=0.83$ ,  $p<0.0001$ ; Fig. 5).

Polyunsaturated fatty acids (PUFA;  $0.79 - 91.71 \mu\text{g g}^{-1}$ ), bacterial fatty acids (BAFA:  $0.18 - 122.46 \mu\text{g g}^{-1}$ ), and terrestrial fatty acids (terrFA;  $0.76 - 231.70 \mu\text{g g}^{-1}$ ), used biomarkers of mostly algae-derived, bacterial, and terrestrial OM in



sediments, respectively, mostly decreased immediately below the sediment-water interface down core (Fig. 6 and 9).

The OM source indicators PUFA and BAFA revealed co-occurring decreasing algal and bacterial OM down-core, suggesting a positively correlated relationship between bacterial and algal (i.e. labile) OM (Fig. 6). Linear regression analysis showed that PUFA significantly predict 80% of the BAFA variability in sediments (Fig. 7;  $R^2 = 0.80$ ,  $p < 0.0001$ ). PUFA and terrestrial fatty acids (terrFA) did not decrease steadily downcore (Fig. 9).

Terrestrial FA were not significantly associated with total phosphorus (TP;  $p=0.5$ ; regression analysis; Fig. 10). Furthermore, linear regression analyses with terrFA and BAFA showed that terrFA significantly predicted 64% of the BAFA variability (Fig. 8).

The highest total P content was  $0.371 \mu\text{g}\cdot\text{L}^{-1}$  at the surface sediment layer (0 – 0.5 cm), whereas the downcore total P content fluctuated between a minimum of  $0.221 \mu\text{g}\cdot\text{L}^{-1}$  at 22 cm,  $0.327 \mu\text{g}\cdot\text{L}^{-1}$  at 6.5 cm, 0.321 at 28.5 cm and  $0.312 \mu\text{g}\cdot\text{L}^{-1}$  at 12 and 25 cm (Fig. 11). Total P content in sediments did not vary significantly with annual precipitation ( $R^2 = 0.01$ ,  $F = 0.264$ ,  $p = 0.611$ ), or terrestrial OM deposition (using terrFA as proxy;  $R^2 = 0.00$ ,  $F = 0.002$ ,  $p = 0.964$ ).

BAFA decrease around the year 1885, when the major input of allochthonous matter into the lake co-occurred with the highest  $\delta^{13}\text{C}$  values and C/N ratios (Fig. 5).

### 3.1.1 Biota in sediments

The phytoplankton assembly resulted in 26 genera over 17 analysed sediment core

layers (Fig. 15). The community composition of the sediment core layers indicates that phytoplankton abundance decreased downcore and disappeared in the bottom sediment layers (20 – 40 cm depth). The following diatom genera were abundant in the top core layers (0 – 5 cm): *Navicula*, *Cymbella*, *Gyrosigma*, *Fragilaria*, *Stauroneis*, *Tabellaria*, *Surirella* and *Pinnularia*.

The zooplankton community contained 4 genera in over 17 sediment layers, including *Daphnia*, *Bosmina*, *Alonella* and *Alona*, with *Bosmina* being the most abundant genera. Densities of the large bodied *Daphnia* remained low throughout the sediment core, despite their high abundance in the upper water column of Lake Lunz. The total number of zooplankton in the sediment layers was not significantly correlated with organic carbon content, stable isotope values, or terrFA, PUFA, BAFA, or total P (see Appendix).

### 3.2. Sediment traps

Particulate OC accumulation in the upper sediment trap (15 m below the lake-air interface) ranged from 8 to 154 mg C m<sup>2</sup> d<sup>-1</sup> over the time of the deployment period, with highest accumulation in June 2016. A second accumulation peak was recorded in December 2015 (94 mg C m<sup>2</sup> d<sup>-1</sup>). The OC accumulation in the profundal sediment trap (30 m) ranged from 10 mg (December 2015) to 68 mg C m<sup>2</sup> d<sup>-1</sup> (December 2015, Fig. 12). The average OC settling rate in the water column was lower at 30 m (36 mg C m<sup>-2</sup> d<sup>-1</sup>) than at 15 m lake depth (49 mg C m<sup>-2</sup> d<sup>-1</sup>).

The observed OC differences between the upper and lower sediment traps were larger during the summer and smaller during spring and fall. The lower OC load at 30 m compared to 15 m suggests carbon being more degraded during summer than during winter months. On average, 12.5 mg C m<sup>-2</sup> got lost between 15 m and

30 m per day. The lowest carbon production between 15 and 30 m occurred during the winter months, with an absolute minimum average in December 2016 of  $1.14 \text{ mg} \cdot \text{m}^{-2}$  per day. Differences in carbon content between the upper (15 m) and lower (30 m) sediment traps were highest during the summer time (e.g., June 2016:  $80 \text{ mg C m}^{-2} \text{ d}^{-1}$ ; and August 2016:  $105 \text{ mg C m}^{-2} \text{ d}^{-1}$ ; Table 3) and markedly lower in winter (December 2015:  $33 \text{ mg C m}^{-2} \text{ d}^{-1}$ ) and spring (May 2015:  $30 \text{ mg C m}^{-2} \text{ d}^{-1}$ ). An annual OC load of 4.47 t was assessed for the lake sediment accumulation area, defined as the lake bottom area in which continuous particle sedimentation takes place (Hakanson and Jansson, 1983); this area was assumed to be 50% of the total lake area (34 ha), estimated based on the lake trough-shaped morphometry, and calculated using results of the lower sediment trap and an annual load of 6.02 t OC (per 34 ha) for the upper sediment trap. This adds up to a total difference of 1.55 t OC per 34 ha between the two sediment traps.

PUFA in the upper trap ranged from  $0.45$  to  $6.36 \text{ } \mu\text{g g}^{-1}$ , in the lower trap PUFA varied from a minimum of  $0.32 \text{ } \mu\text{g g}^{-1}$  to a maximum of  $2.95 \text{ } \mu\text{g g}^{-1}$ . Terrestrial fatty acids were much lower in both traps ( $0.03 \text{ } \mu\text{g g}^{-1}$  to  $0.22 \text{ } \mu\text{g g}^{-1}$  in the upper and  $0.02 \text{ } \mu\text{g g}^{-1}$  to  $0.11 \text{ } \mu\text{g g}^{-1}$  in the lower trap).

A difference of  $2.264 \text{ } \mu\text{g g}^{-1}$  between PUFA and terrFA measurements was detected in the upper sediment trap in November 2016 with average PUFA values of  $2.29 \text{ } \mu\text{g g}^{-1}$  and terrFA values of  $0.026 \text{ } \mu\text{g g}^{-1}$ . Results for the lower sediment traps showed similar values for PUFA and terrFA. Differences of  $2.915 \text{ } \mu\text{g g}^{-1}$  were mainly visible in December, where PUFA peaked at  $2.95 \text{ } \mu\text{g g}^{-1}$ , but terrFA decreased to  $0.035 \text{ } \mu\text{g g}^{-1}$  in the upper trap. Both types of fatty acids show peaks in the late summer season ending of September.

## 4. Discussion

### 4.1. Sediment cores

The annual sedimentation rate ( $\sim 1.44 \text{ mm yr}^{-1}$ ) of oligotrophic, subalpine Lake Lunz is similar to many oligotrophic lakes and seems to be independent of the high relief-energy of its mountain lake catchment as it coincides with lowland oligotrophic lakes (i.e.,  $\sim 1 \text{ mm yr}^{-1}$  in eastern Canada; Lucotte et al. 1995). It is likely that the trophic lake status is more important than the lake catchment relief for the annual lake sedimentation rates. For example, Lake Mondsee, also a subalpine lake in the Austrian Alps with a similar mountainous catchment, had a much higher annual sedimentation rate ( $6.6 \text{ mm yr}^{-1}$ ) during times of eutrophication (Klee and Schmidt 1987). Although difficult to record, irregularities of sedimentation rates in Lake Lunz are likely due to extreme weather events with high precipitation and consequently high OM loading from the lake catchment. According to historical records, a disastrous storm event happened in 1861 in the Lake Lunz catchment. A conservative back-calculation of the current  $1.44 \text{ mm yr}^{-1}$  lake sedimentation rate to this historic storm event yields the year 1885, which almost matches the actual extreme weather event in 1861. However, if the extreme event of 1861 had yielded higher deposition of OM on the lake sediments, an offset of about 20 years may have been compensated for it. Concurrently, the high C/N ratios and heavy  $\delta^{13}\text{C}$  values at  $\sim 20 \text{ cm}$  sediment depth co-occur with this storm event, suggesting that the combination of  $^{210}\text{Pb}$  (tracer of lake sedimentation rates) and the C/N ratios (OM source bioindicator) can fairly accurately track historic carbon dynamics. The hypothesis that extreme weather events, in a lake located at higher altitudes, can be biochemically tracked in lake sediments could thus be confirmed.

Atomic C/N ratios from 6 to 10 in lake sediments typically indicate that the

sedimentary matter is mostly of autochthonous origin (Teranes & Berasconi, 2000). The C/N ratios in sediment layers of Lake Lunz are generally >10, and even reach ~15 during the extreme weather event of 1861, which implies higher terrestrial OM input to the sediments (Teranes & Berasconi, 2000). Concurrent to the significant correlation between C/N and  $\delta^{13}\text{C}$  values ( $R^2 = 0.83$ ), the heavier  $\delta^{13}\text{C}$  values at the sediment layer referring to the extreme weather event also suggest higher terrestrial OM input. The significant relationship between  $\delta^{13}\text{C}$  values and C/N ratios in the sediment layers also signifies that these two OM source indicators have very similar predictive values. Changes in  $\delta^{13}\text{C}$  values downcore in these sediments are likely due to changes in deposited carbon sources and/or isotopic carbon fractionation (Gälman et al., 2009). Lighter carbon isotope values just below the sediment water interface may be the result of methanogenic bacteria, which are isotopically very light and possibly provide as an important food source for pelagic zooplankton (Bastviken et al., 2003). The lack of significant relationships between bacterial fatty acids and  $\delta^{13}\text{C}$  values in these sediment layers suggest that the used bacterial FA biomarkers cannot predict the  $\delta^{13}\text{C}$  variability and that other processes, such as changes in OM sources, determined the stable carbon isotope composition in these sediment layers. However, C/N and  $\delta^{13}\text{C}$  values do not provide detailed or quantitative OM source information. Thus, we applied fatty acids as quantifiable molecular biomarkers and distinguished between bacterial, algal, and terrestrial sources.

The significant relationship between PUFA and BAFA suggest that autochthonous OM (PUFA) provided energy-yielding substrates for bacteria throughout the sediment core. The bacterial fatty acids of the analyzed sediments represent a sum of known BAFA, but do not allow specific differentiation between

anaerobic or aerobic bacteria. BAFA values decrease downcore, which suggests simultaneous decrease of available OM as energy-yielding substrate and/or a change in bacteria taxa, which are probably not traced with the used BAFA in this study. Meier (1993) states that autochthonous produced matter is generally more sensitive to bacterial degradation than materials of terrestrial origin, therefore lacustrine sediments often contain large relatively unreactive fraction of land-derived organic residues.

The lower correlation coefficient between terrFA and BAFA suggests that terrFA supply less readily available energy for bacteria than PUFA. Additionally, terrFA were not significantly associated with total phosphorus (TP;  $p=0.5$ ; regression analysis), suggesting that TP input to the sediments is not directly linked to deposited terrFA and was probably derived from external sources like atmospheric deposition (Tammatamah et al., 2005), which show no relation to terrFA.

The high predictive power of PUFA and also terrFA for BAFA suggests that both, autochthonous and allochthonous OM supports bacteria in lake sediments, although algae-derived PUFA more strongly than terrestrial OM. The lack of correlations between  $\delta^{13}\text{C}$  and BAFA indicates that there is no linear relation between the stable carbon isotope values and BAFA evident in sediments.

Moreover, we hypothesized that autochthonous OM, as characterized by PUFA, degrades faster than the more recalcitrant terrestrial OM. PUFA and terrFA did not decrease steadily downcore and thus show that both source indicators can be used as a quantitative measure for the determination of OM sources. Furthermore, PUFA and terrFA values did not indicate degradation preferences of

one or the other fatty acid type. Thus, the hypothesis, which described PUFA in the ecosystem Lake Lunz as the faster degrading fatty acid type, can be rejected.

Decreasing OC and ON below the sediment-water interface generally indicate diagenetic degradation of freshly sedimented OM (Teranes & Bernasconi, 2000). Fluctuations of OC values along the sediment core suggest that varying amounts of allochthonous or autochthonous matter influenced the ecosystem at different points in time (e.g., strong indication for large terrestrial matter input in 1886). Further investigations on how exactly the differently abundant OM sources shaped the lake over time may be subject for further research.

The range of  $^{15}\text{N}$  values with terrestrial or aquatic origin (Fry, 2006) mainly results from selective metabolic processing of nitrogen compounds (Saino & Hattori, 1987). Other possible influences on  $^{15}\text{N}$  budget are external nitrate loading from agricultural runoff, sewage influx, shifts in phytoplankton species composition, or the addition of heterotrophs (Teranes & Bernasconi, 2000). Lake Lunz as a remotely situated sub – alpine lake is not exposed to extensive anthropogenic influences like fertilizers or sewage runoff. Rather, the maximum peak of  $\delta^{15}\text{N}$  in the top sediment layers might be related to the recent introduction of Northern pike (*Esox lucius*) in Lake Lunz. With the addition of heterotrophic matter to the OM pool,  $^{15}\text{N}$  levels can increase even up to 3 – 4‰ with each trophic transfer (Teranes & Bernasconi, 2000). A theoretical explanation for the increases  $^{15}\text{N}$  could be that, profiting from increased lake temperatures during their spawning time, the pike population size enlarged strongly from a few rarely sighted individuals up to hundreds of individuals by 2016. Additionally, the pike as a predator at the top of the food chain reduces other fish populations like the Brown trout (*Salmo trutta fario*) or the Arctic charr (*Salvelinus alpinus*). However, this remains highly

hypothetical and is beyond the scope of this study, thus it may be examined in future studies.

#### 4. 2. Sediment traps

The decrease of terrestrial fatty acids in the 30 m sediment trap of the December sampling might indicate an increased use of terrestrial matter in the water column during the winter season. Since photosynthesis and primary production eventually reach a low during winter months, terrestrial resources probably are of greater importance for the ecosystem lake during this time of the year.

High standard deviation values for the fatty acid markers in the sediment traps of September and December samples indicate a strong spatial variability of OM abundance in the water column, since the standard deviation was calculated from three sediment trap cylinders situated close to each other (within 1 m). The spatial distribution of OM in the water column can be influenced by the onset of circulation due to wind events during lake mixing. As comparison Eadie (1984) states that nearly 85% of the carbon in Lake Michigan is oxidized before leaving its epilimnion (above the thermocline). Compared to Lake Michigan, Lake Lunz with 33 m maximum depth, should consequently show sediment records richer in OM, due to reduced sinking times, which can be confirmed by considering results of the labile organic matter indicators PUFA in the 15 m and 30 m sediment traps. Both traps show similar values for PUFA as well as terrFA. Differences in peaks of PUFA in September between the high and low sediment trap can probably be explained by enhanced mixing in the water body. Also the high standard deviation values for the measurements of September 29<sup>th</sup> 2016 in upper and lower sediment traps might



be an indicator for this event.

To understand the full spectrum of POC influx to lake sediments, we also have to consider DOC as an important player in this circuit, since the proportion of POC, which is in part transformed out of DOC in the water column via flocculation (Helms et al. 2013), also contributes to the sediment organic carbon pool (Travnik et al. 2009). The importance of potential DOC input fluctuates with the parameters snow cover and permafrost (modification of carbon flow due to alteration of runoff and vegetation), which especially affects high altitude lakes (Zimov et al. 2006). As a consequence, DOC levels can be estimated by taking the length of the main growing and thawing season into account (Karlsson et al. 2009). The role of climate change might be a more influencing one in this scenario than expected, since changes in the trend of seasonality can raise DOC levels of high – altitude lakes and therefore alter their organic carbon cycle. With climate change, the frequency of extreme weather events (i.e. heavy rainfalls) will eventually increase as well, which affects sub – alpine lakes even more than lowland lakes.

## 5. Climate change and future prospects

Lake Lunz is a lake ecosystem that provides important ecosystem services, including tourism and fisheries, and eventually may become more strongly affected by climate change than lowland lakes. According to paleolimnological records, especially high altitude lakes are already affected by prevailing climatic conditions (Battarbee, 2010). The difficulty that arises from working with sediment records is that although the cores can be accurately dated up until 200 years and more in the past with  $^{210}\text{Pb}$ , the time resolution in between the sediment layers (which often covers decades) doesn't allow precise conclusions for the effects of climate change on lake ecosystems. Moreover, even typically measured lake parameters like surface water temperature, stratification length or duration of ice cover tend to vary more with situational influences like altitude, magnitude of shading or surface area (Livingstone & Padisak, 2007; Livingstone et al., 2010), which impedes understanding the effects of climatic forcing.

It is equally difficult to determine how climate change has affected the biotic community in Lake Lunz (Kainz et al. 2017), since the composition of lake biota is a specific variable for each lake and often even incomparable in two neighboring lakes (Dokulil & Teubner, 2002). Generally, biological parameters and their responsive extent to climate change tend to be more unpredictable than chemical or hydrological variables (Hering et al., 2010). Even in the same water column within the same lake, chemical parameters might show differences in response to climate change between deep water and surface water layers (Danis et al., 2004).

In conclusion, it is clear that tracking past and current carbon loadings, in particular from extreme weather events, to lake sediments is highly a highly complex scientific endeavor. It involves detailed understanding of interrelated

chemical, biological and physical parameters, which have to be observed individually with respect to their catchments, anthropogenic influence, nutrient levels, and primary production. Future studies should therefore include extended long-term observations, accurate time resolutions (climate change leaves more pronounced traces on monthly or seasonal scales than yearly or decadal time lines) and the analysis of multiple sediment cores, since full understanding of the complexity of a lake ecosystem and its response to changing climatic conditions can probably only be achieved through broad and long-term coverage of such variables.

## 6. Appendix

The zooplankton abundance (Fig. 14) could occur due to the diverging body sizes of *Bosmina* and *Daphnia*. Larger organisms might be a more favourable prey for planktivorous fish, in comparison to the small-bodied *Bosmina*, because of lower energy expenditure while hunting and higher nutrient intake in case of success.

Hence, more small – bodied zooplankton will survive, sink below the thermocline and eventually add to the top sediment layers. Insignificant results of the linear regression analysis between the sum of *Bosmina* carapace and total phosphorus, fatty acids, stable isotopes and organic carbon, might indicate that during its life cycle and after subsidence to the surface sediment layer of the lake, other factors probably explain the number of deposited carapace better. Since there is no trend in the number of zooplankton carapace discernible, which can be related to measured biochemical sediment composition, the situation of small-bodied zooplankton might just be a mirror for the biochemical constitution of the water column during their life cycle in the lake, although also this is yet to be tested.

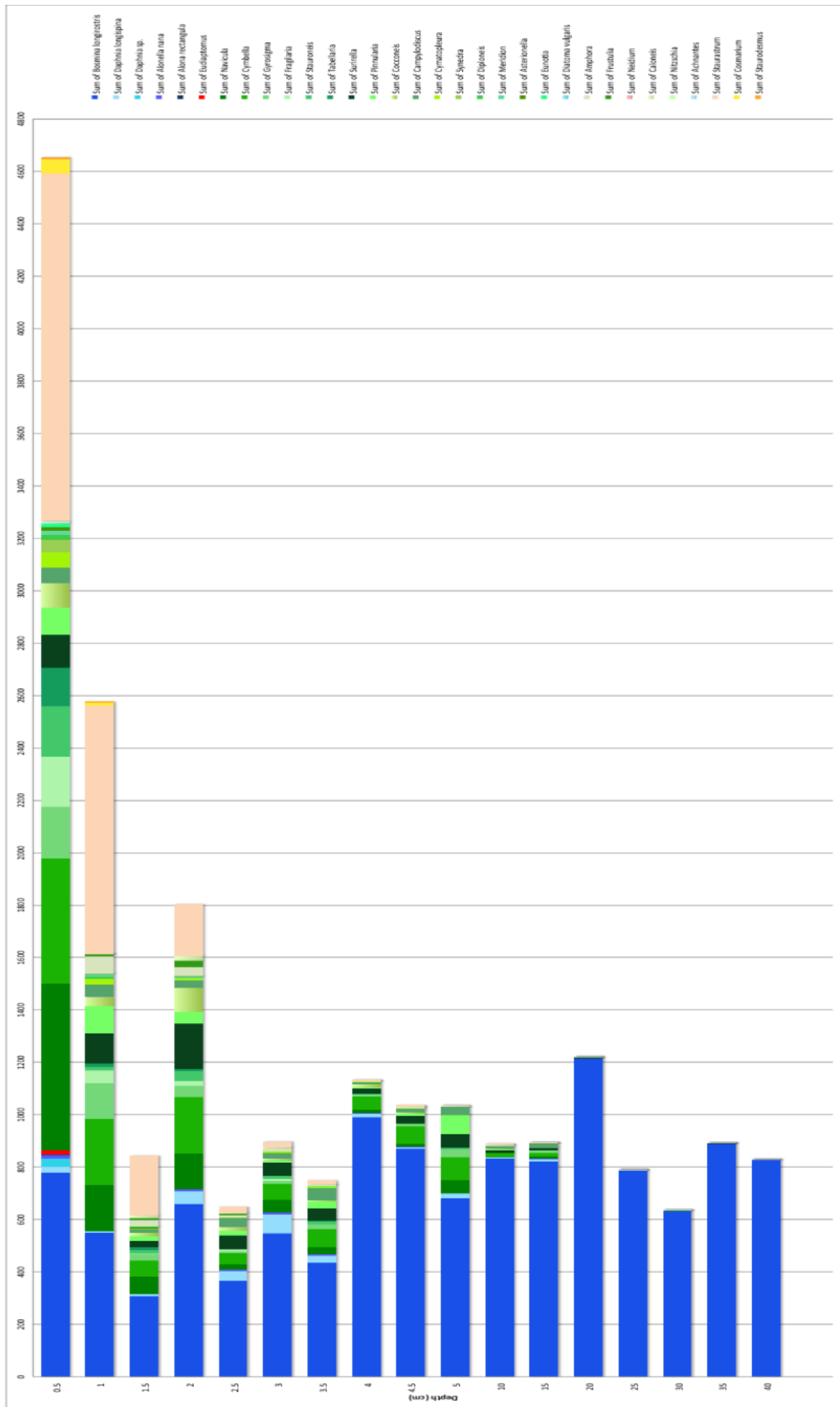


Fig. 15: Vertical distribution of biological fossil remains (cladocera – blue tones, copepods - red tones, diatoms – green, tones, zygnerophyceae – yellow/orange tones) of the sediment core retrieved in 2015. 37

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**Table 1: Different source indicators for macroalgae, vascular plants, and types of OM of lipids and bacterial fatty acids used as communities, bulk algae, diatoms.**

| <b>Fatty acids</b>                                                                     | <b>Used as tracer for</b>                                                                    | <b>References</b>                                                           |
|----------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------|
| <b>18:1<math>\omega</math>7, odd, linear &amp; branched fatty acids</b>                | Bacterial communities                                                                        | Currie & Johns 1988                                                         |
| <b>10-methylpalmitic acid; cyclopropyl 17:0, 19:0 <math>\nabla</math> 19:0 (11,12)</b> | Bacteria                                                                                     | Perry et al., 1979                                                          |
| <b>Polyunsaturated fatty acids (PUFA)</b>                                              | Algae<br><br>Synthesized by vascular plants, used as tracers in sediments and animal tissues | Kharlamenko et al. 1995<br><br>Rajendran et al. 1992<br>Meziane et al. 1997 |
| <b>eicosapentaenoic acid (EPA, 20:5n-3)</b>                                            | Algae – derived PUFA, highly retained in zooplankton                                         | Kainz et al, 2004                                                           |
| <b>C<sub>18</sub> and C<sub>20</sub> PUFA</b>                                          | Macroalgae                                                                                   | Kharlamenko et al. 1995                                                     |
| <b>&gt;C22:0</b>                                                                       | Terrestrial OM                                                                               | Sun and Wakeham 1994                                                        |

**Table 2: Comparison of different alpine and subalpine lakes in respect of their depth, lake area and sedimentation rates.**

| Lake name                      | Sedimentation rate                                                                   | location                                                        | max. depth                                               | Lake area              | author              |
|--------------------------------|--------------------------------------------------------------------------------------|-----------------------------------------------------------------|----------------------------------------------------------|------------------------|---------------------|
| <b>Zgornje Krisko jezero</b>   | 7.0 mm*yr <sup>-1</sup>                                                              | Slovenian mountain lakes                                        | 3 m                                                      | 0.0061 km <sup>2</sup> | Vreca et al., 2010  |
| <b>Jezero v Ledvicah</b>       | 1.4 mm*yr <sup>-1</sup>                                                              | Slovenian mountain lakes, Valley of the seven lakes             | 15 m                                                     | 0.0219 km <sup>2</sup> | Vreca et al., 2010  |
| <b>Lake Geneva (Lac Léman)</b> | 1.2 mm*y <sup>-1</sup>                                                               | North side of the Alps, between Switzerland & France            | 310 m                                                    | 582 km <sup>2</sup>    | Vernet et al., 1983 |
| <b>Upper Lake Raduńskie</b>    |                                                                                      | Northern Poland, central part of Kashubian Lakeland, hilly area | 43 m<br>Samples taken at:<br>41 m<br>8 m<br>20 m<br>13 m | 3.872 km <sup>2</sup>  | Tylmann 2004        |
| <b>Lake Mansar</b>             | 1.4 & 3.7 mm*yr <sup>-1</sup><br><br>mean rate<br><br>2.3 ± 0.02 mm*yr <sup>-1</sup> | Himalayan foothills, 666m a.b.s.l., Siwalik Terrain             | 38.25 m                                                  | 0.59 km <sup>2</sup>   | Rai et al., 2007    |



|                       |                                                                |                                                            |        |                          |                                                |
|-----------------------|----------------------------------------------------------------|------------------------------------------------------------|--------|--------------------------|------------------------------------------------|
| <b>Lake Bourget</b>   | 1.3 mm yr <sup>-1</sup>                                        | Northwest edge of French Alps, glacially formed,           | 65 m   | 27 km <sup>2</sup>       | Perga et al., 2015<br><br>Chapron et al., 2001 |
| <b>Lake Annecy</b>    | 1 mm yr <sup>-1</sup>                                          | Northwest edge of French Alps, glacially formed            | 147 m  | 45 km <sup>2</sup>       | Perga et al., 2015<br><br>Vernet et al., 1982  |
| <b>Lake Mondsee</b>   | 6.6 mm yr <sup>-1</sup>                                        | Austria, Salzburg, Salzkammergut                           | 68.3 m | 14.2 km <sup>2</sup>     | Klee & Schmidt 1987                            |
| <b>Lake Iseo</b>      | 8.1 - 10 mm yr <sup>-1</sup> (past 50 years)                   | Italy, southern Alps                                       | 251 m  | 61 km <sup>2</sup>       | Bettinetti et al., 2011                        |
| <b>Canadian Lakes</b> | 1-3 mm yr <sup>-1</sup>                                        | North-south gradient; Quebec                               | 6-13 m | 0.04 – 6 km <sup>2</sup> | Lucotte et al. 1995                            |
| <b>Lake Traunsee</b>  | 20 – 30 mm yr <sup>-1</sup> (past 50 years, measured at basin) | Austria, Salzburg, Salzkammergut, northern rim of the Alps | 189 m  | 24.4 km <sup>2</sup>     | Schneider et al. 1987                          |

**Table 3: Accumulated carbon in the upper (15 m lake depth) and lower (30 m lake depth) sediment traps over the time of deployment period and per day.**

| deployment date | days deployed | mgC m <sup>-2</sup> d <sup>-1</sup><br>upper | mgC m <sup>-2</sup> d <sup>-1</sup><br>lower | $\Delta_{u-l}$ * |
|-----------------|---------------|----------------------------------------------|----------------------------------------------|------------------|
| 08/10/15        | 21            | 24.20                                        | 19.18                                        | 5.02             |
| 27/10/15        | 19            | 10.66                                        | 15.40                                        | 4.74             |
| 17/11/15        | 21            | 8.17                                         | 24.93                                        | 16.76            |
| 15/12/15        | 28            | 93.96                                        | 60.84                                        | 33.12            |
| 21/04/16        | 23            | 15.70                                        | 12.09                                        | 3.61             |
| 17/05/16        | 26            | 28.54                                        | 58.12                                        | 29.58            |
| 07/06/16        | 21            | 153.94                                       | 64.05                                        | 89.90            |
| 23/06/16        | 16            | 42.83                                        | 30.02                                        | 12.81            |
| 15/07/16        | 22            | 71.84                                        | 68.03                                        | 3.82             |
| 04/08/16        | 20            | 116.26                                       | 11.76                                        | 104.50           |
| 18/08/16        | 14            | 42.61                                        | 61.97                                        | 19.37            |
| 01/09/16        | 14            | 32.07                                        | 42.95                                        | 10.88            |
| 14/09/16        | 13            | 43.90                                        | 55.42                                        | 11.52            |
| 29/9/16         | 15            | 29.70                                        | 18.64                                        | 11.07            |
| 18/10/16        | 19            | 25.24                                        | 29.18                                        | 3.94             |
| 10/11/16        | 23            | 12.23                                        | 13.88                                        | 1.65             |
| 01/12/16        | 21            | 11.41                                        | 10.27                                        | 1.14             |

$\Delta_{u-l}$  = Difference in carbon between upper and lower sediment trap per indicated time period.

annual t C per 34 ha: 4.47

average load per day: 36.05

**Table 4: Different fatty acid types and their maximum and minimum values, of the analysed sediment cores.**

| <b>Fatty acid type</b>                     | <b>Minimum (<math>\mu\text{g g}^{-1}</math>)</b> | <b>Maximum (<math>\mu\text{g g}^{-1}</math>)</b> |
|--------------------------------------------|--------------------------------------------------|--------------------------------------------------|
| <b>Polyunsaturated fatty acids (PUFAs)</b> | 0.79                                             | 91.71                                            |
| <b>Bacterial fatty acids (BAFAs)</b>       | 0.18                                             | 122.46                                           |
| <b>Terrestrial fatty acids (terrFA)</b>    | 0.76                                             | 231.70                                           |

## Figures



Fig. 1: Documentation on a house close to Lake Lunz of the water level after the flooding event in 1861.

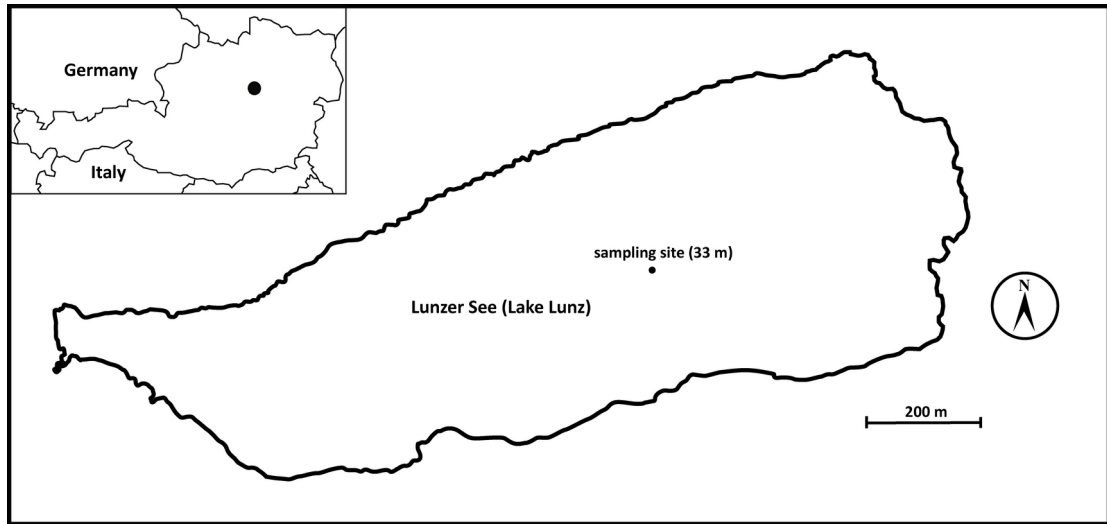


Fig. 2: Location of subalpine Lake Lunz (608 m.a.s.l.) in Austria with the sampling site for settling particles and sediment cores at the deepest lake station (33 m depth).

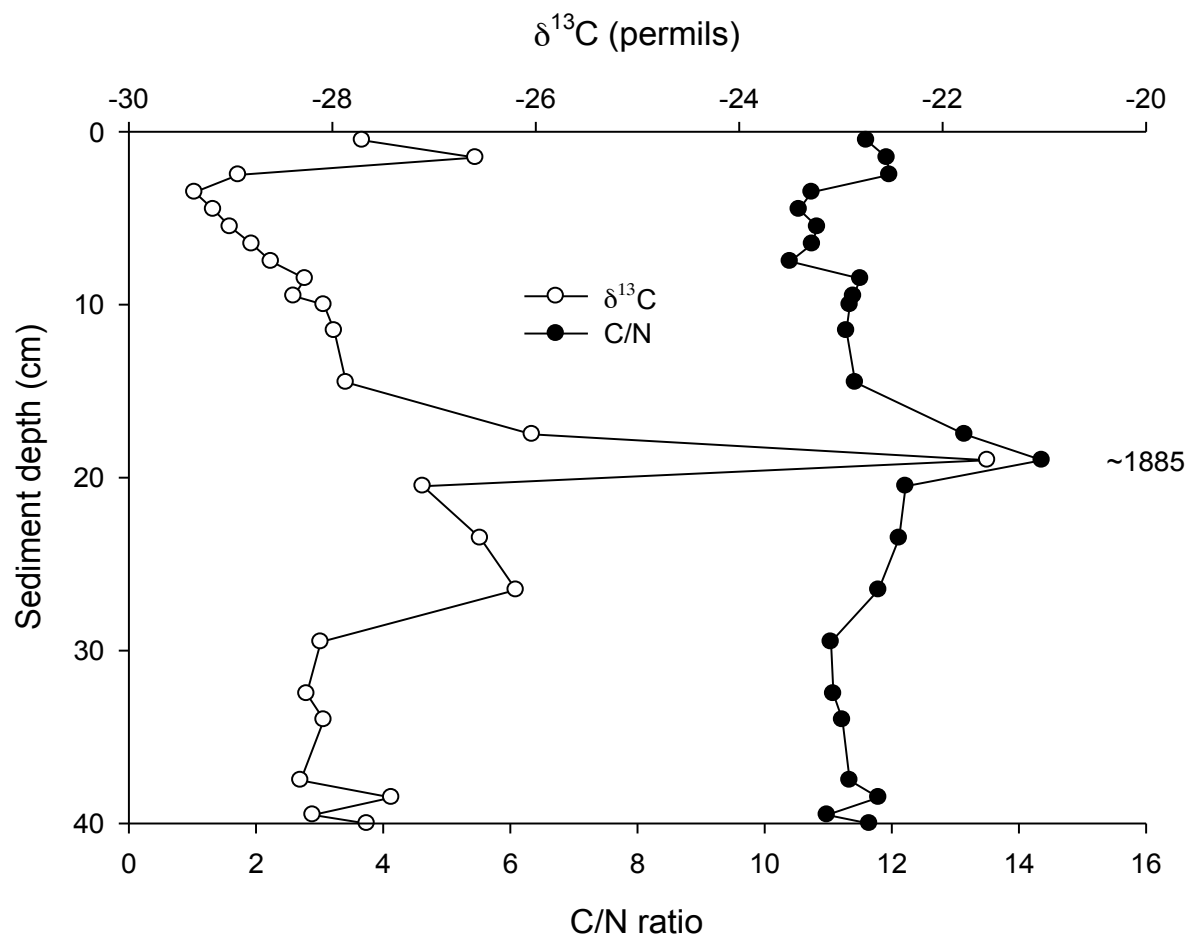


Fig.3: Vertical distribution of C/N ratios and  $\delta^{13}\text{C}$  values in the sediment core of 2015.

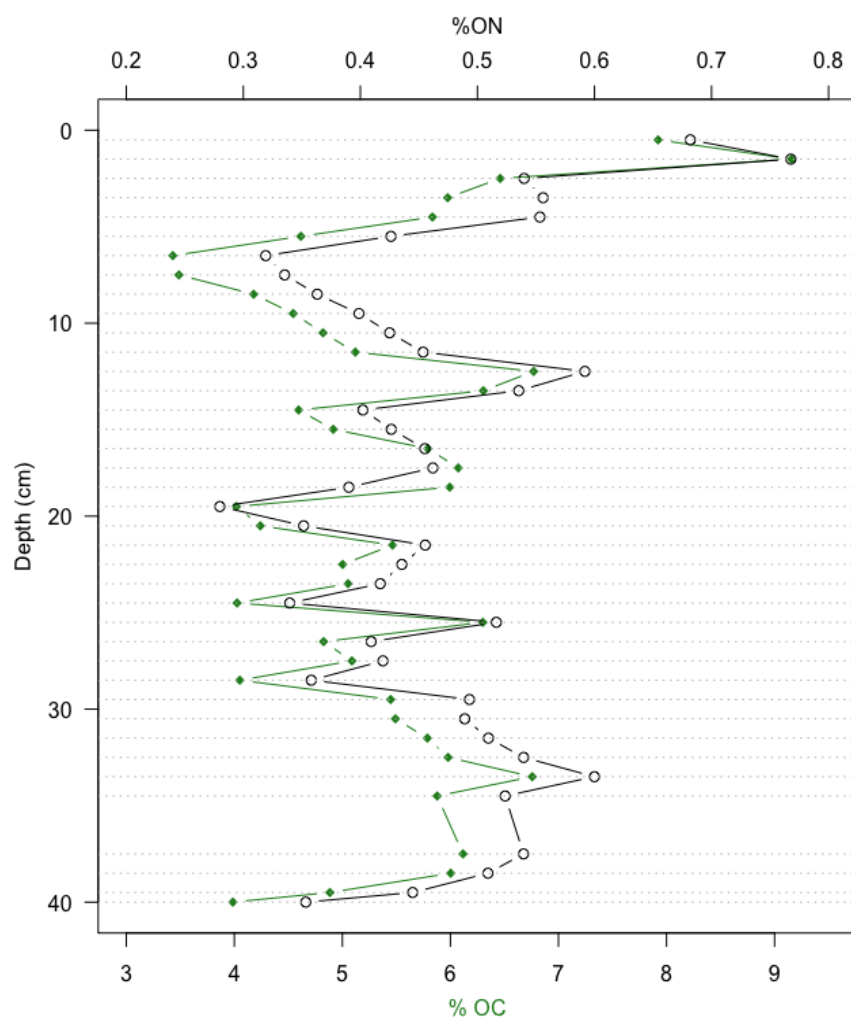


Fig. 4: Vertical distribution of organic nitrogen (%) and organic carbon (%) of sediment core 2015.

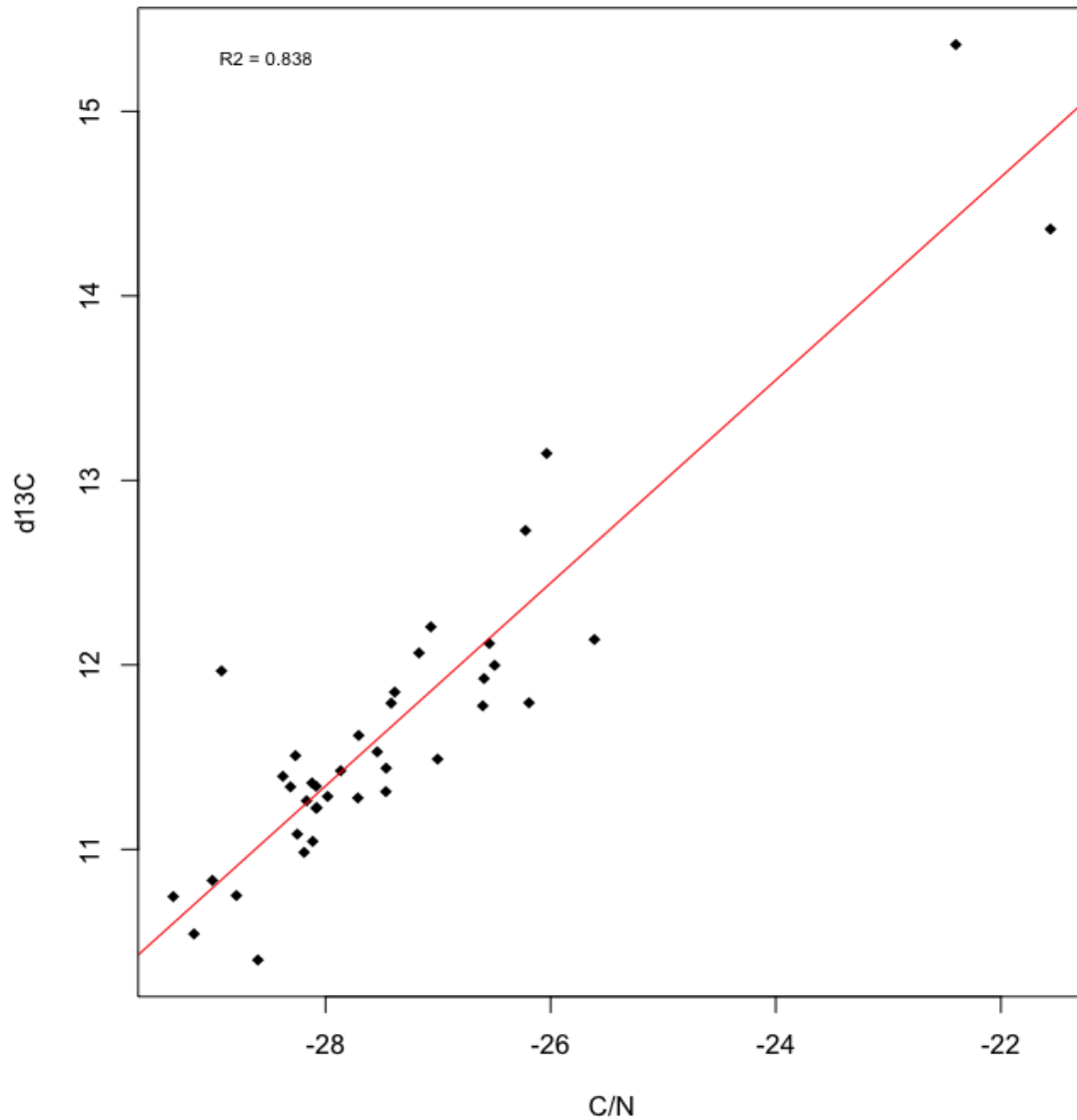


Fig.5: Correlation of  $\delta^{13}\text{C}$  values and C/N ratios in sediment layers (0-40 cm depth) taken at the deepest spot of Lake Lunz, Austria ( $R^2 = 0.84$ ,  $p < 0.0001$ ). N.b.: the two extreme data points (C/N ~15 and  $\delta^{13}\text{C}$  ~-22 permils) are assigned to the extreme weather event of 1861.



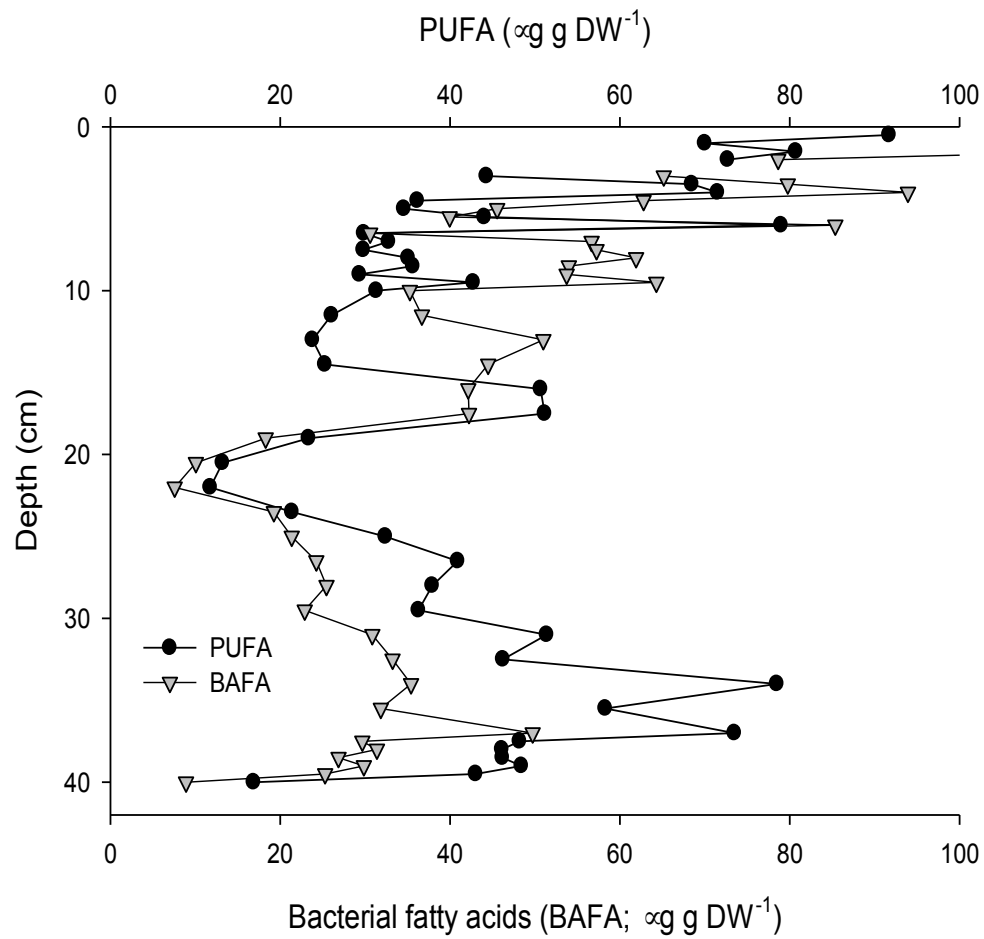


Fig. 6: Vertical distribution of algal- (PUFA) and bacterial fatty acid (BAFA) biomarkers in the sediment core of 2015.

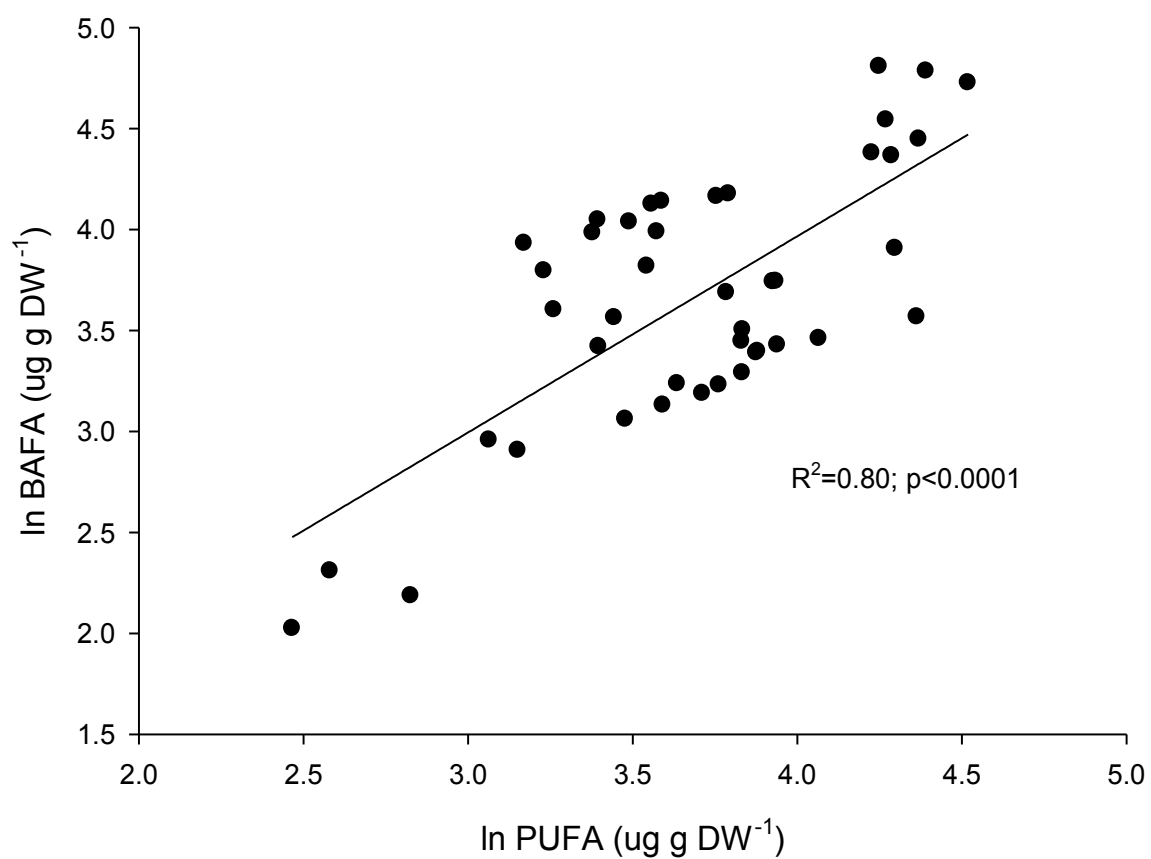


Fig.7: Linear regression analysis of PUFA and BAFA biomarkers in 'core2015';  $R^2 = 0.8$ ,  $p < 0.0001$

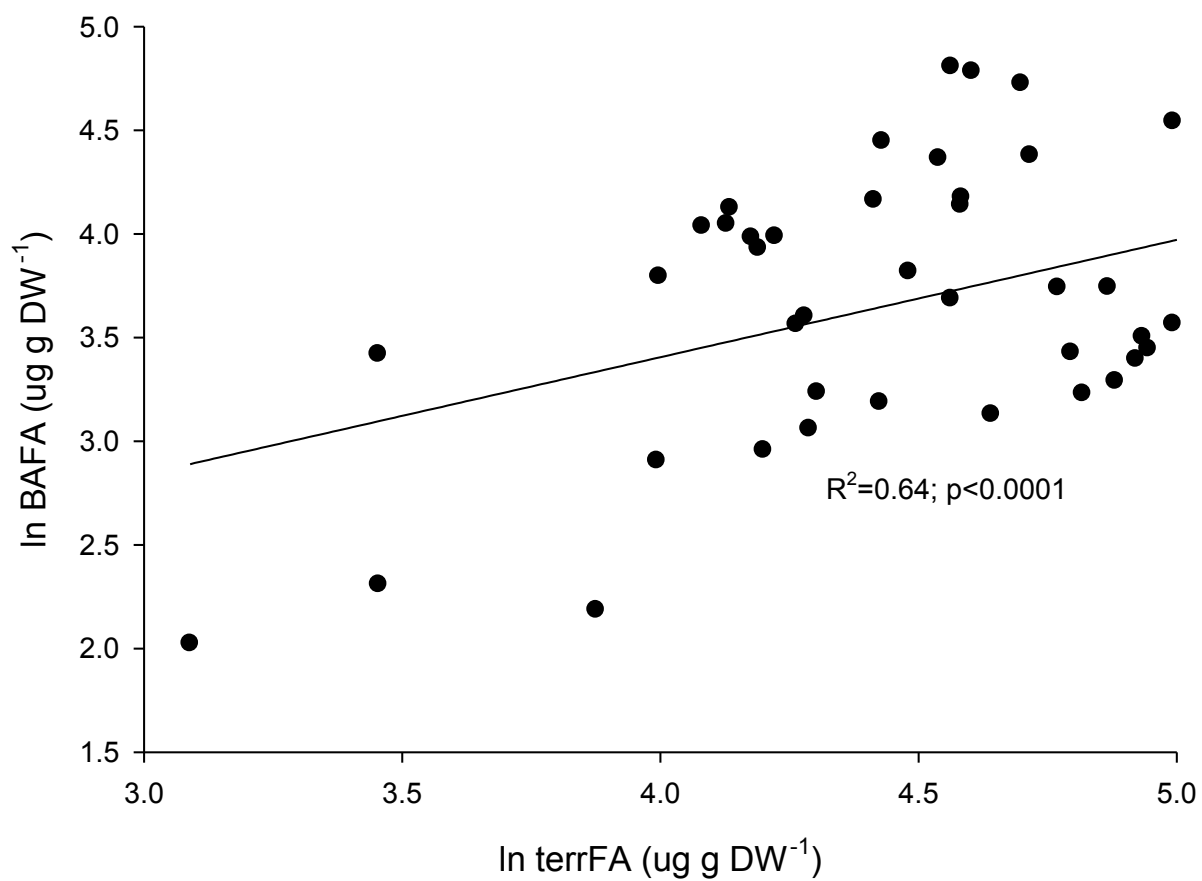


Fig. 8: Linear regression analysis of BAFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ ) and terrFA ( $\mu\text{g} \cdot \text{g DW}^{-1}$ );  $R^2 = 0.64$ ,  $p < 0.0001$

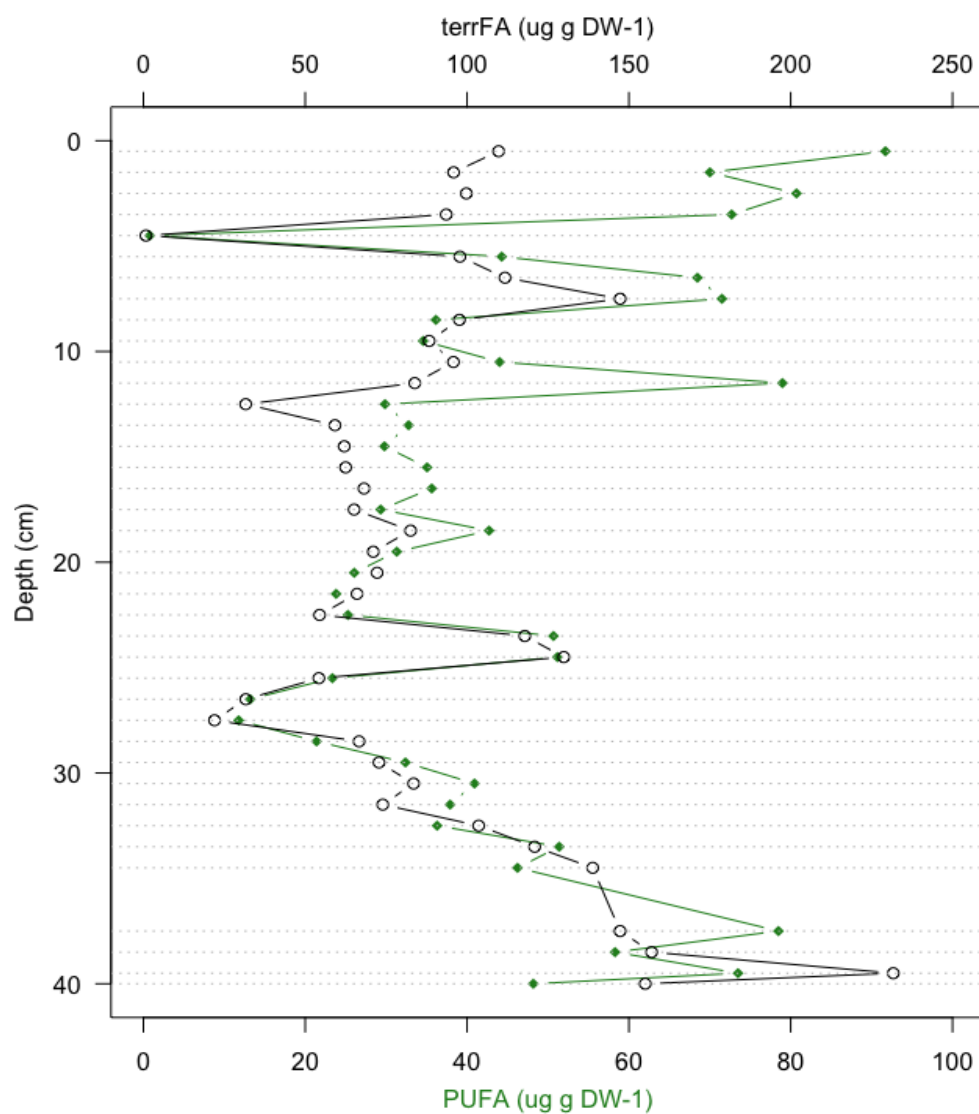


Fig.9: Vertical distribution of PUFA ( $\mu\text{g} \cdot \text{g}^{-1} \cdot \text{DW}^{-1}$ ) and terrFA ( $\mu\text{g} \cdot \text{g}^{-1} \cdot \text{DW}^{-1}$ ) of core 2015.

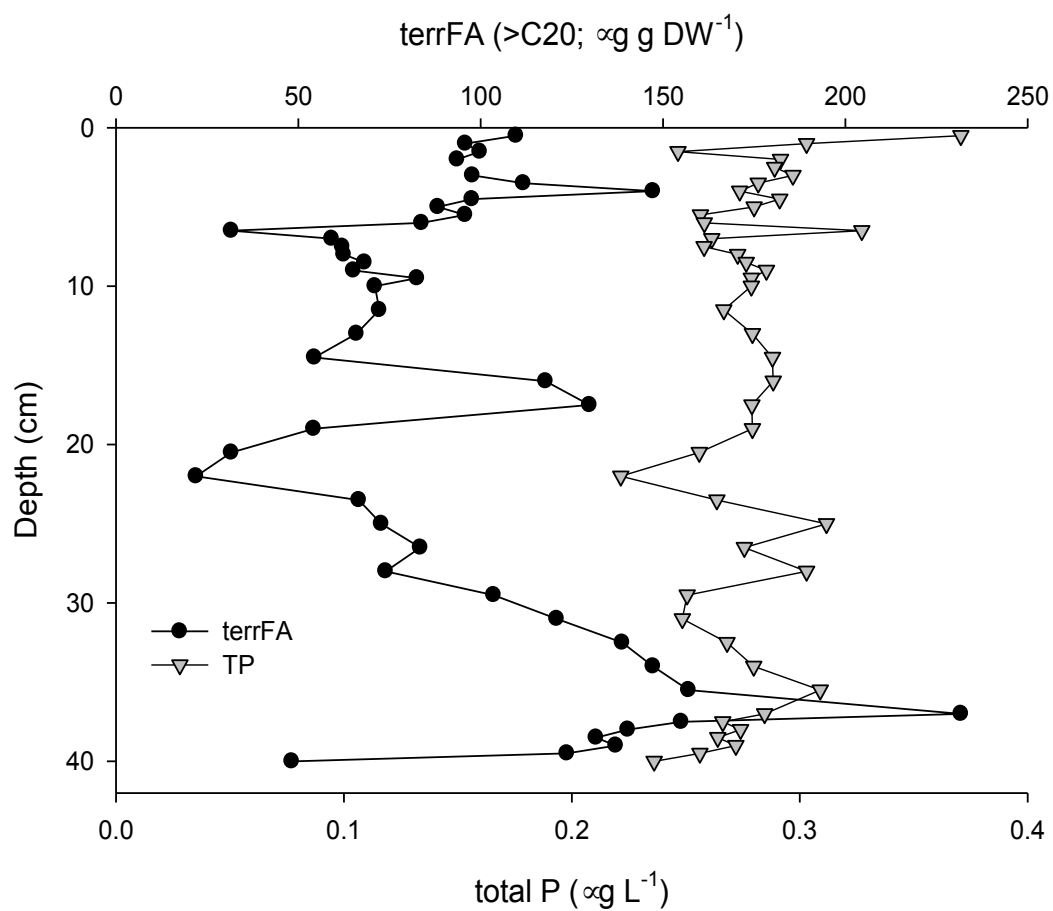


Fig.10: Vertical distribution of terrestrial fatty acids ( $\mu$ g\*g\*DW<sup>-1</sup>) and total phosphorus ( $\mu$ g\*L<sup>-1</sup>) of core 2015.

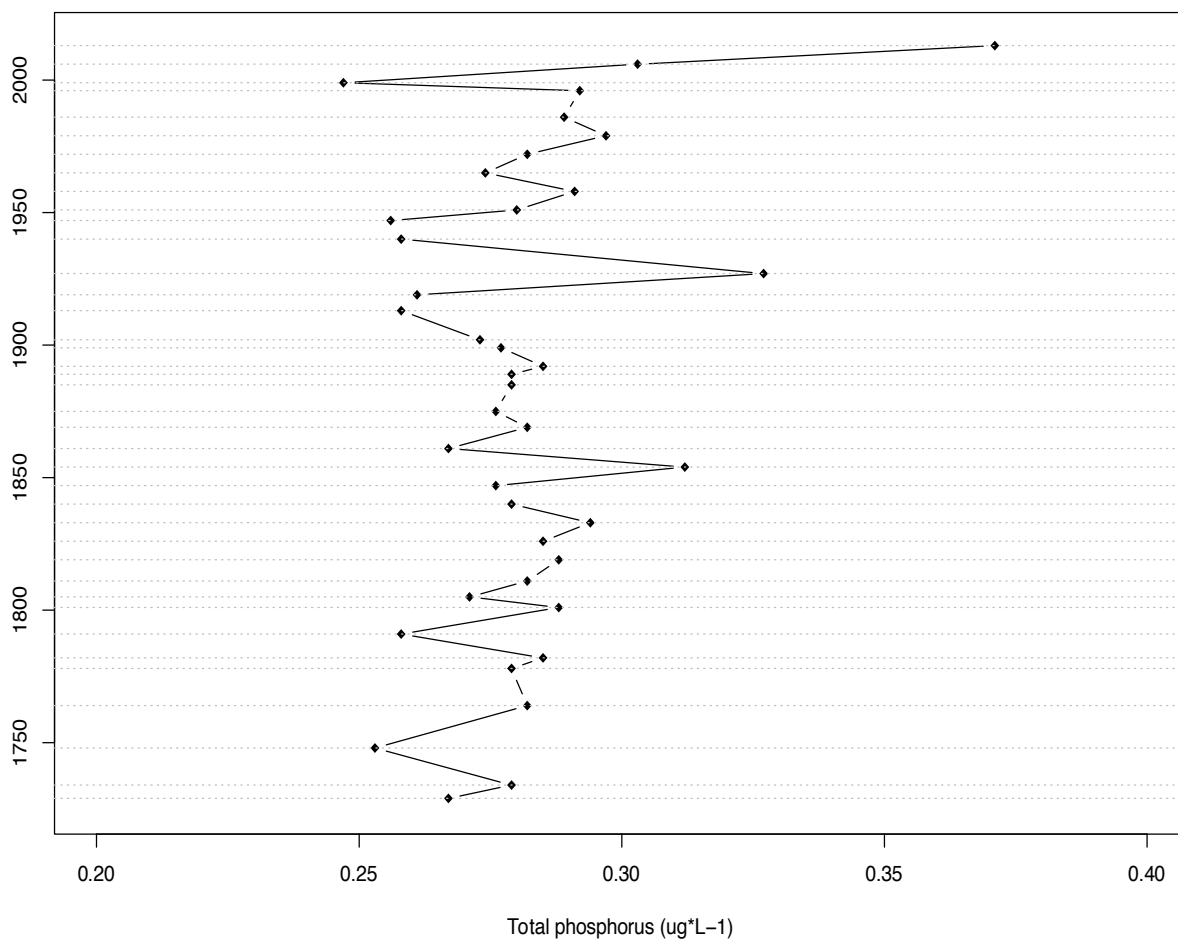


Fig.11: Distribution of total phosphorus ( $\mu\text{g}\cdot\text{L}^{-1}$ ) along the  $^{210}\text{Pb}$  dated sediment layers of core2015 (2016 – 1729).

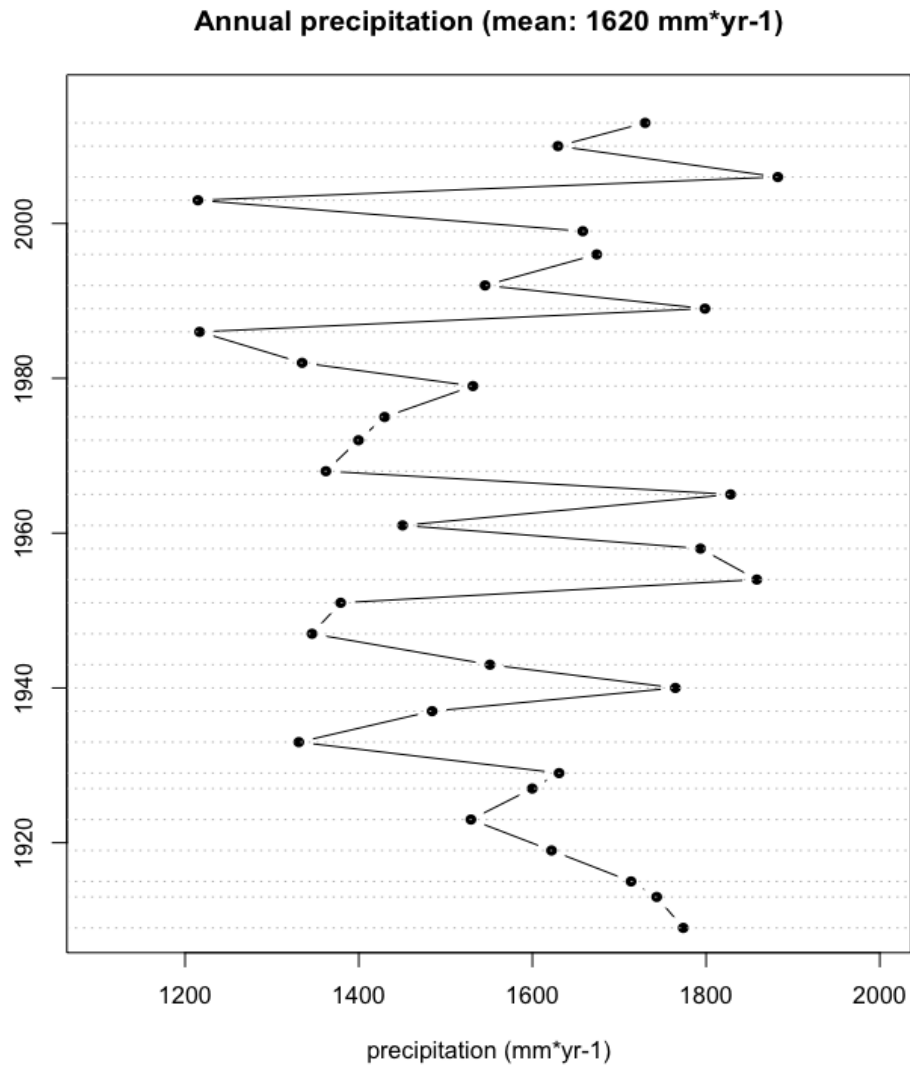


Fig.12: Annual precipitation from 1913 until 2016; calculated mean precipitation of 1620 mm\*yr<sup>-1</sup>

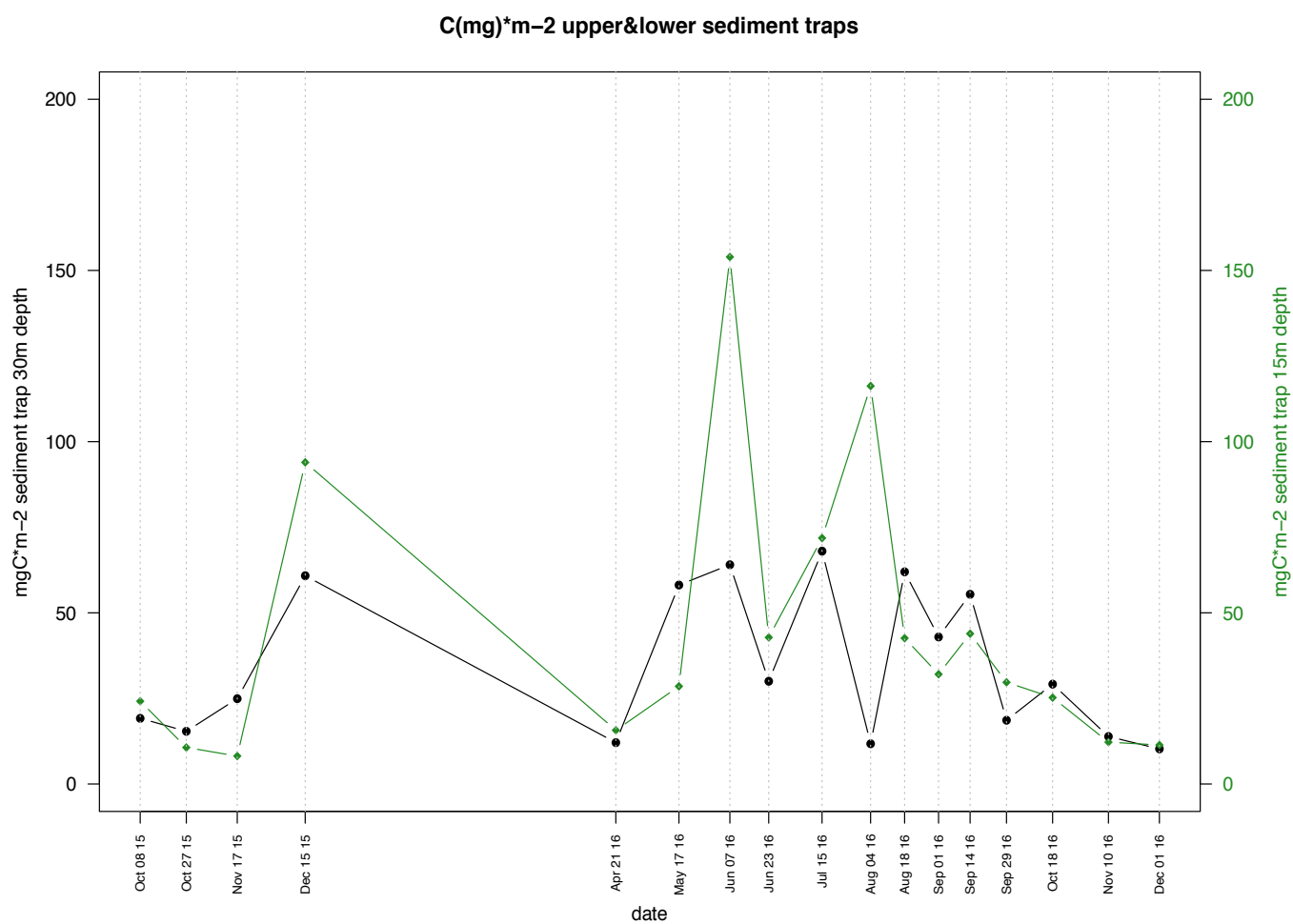


Fig. 13: Accumulated carbon (mg\*m<sup>-2</sup>) per day in the upper (15 m) and lower (30 m) sediment traps.



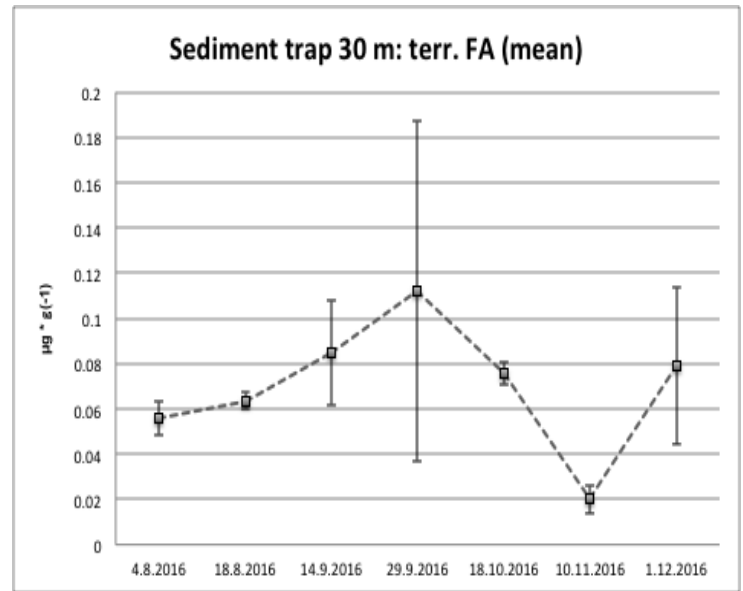
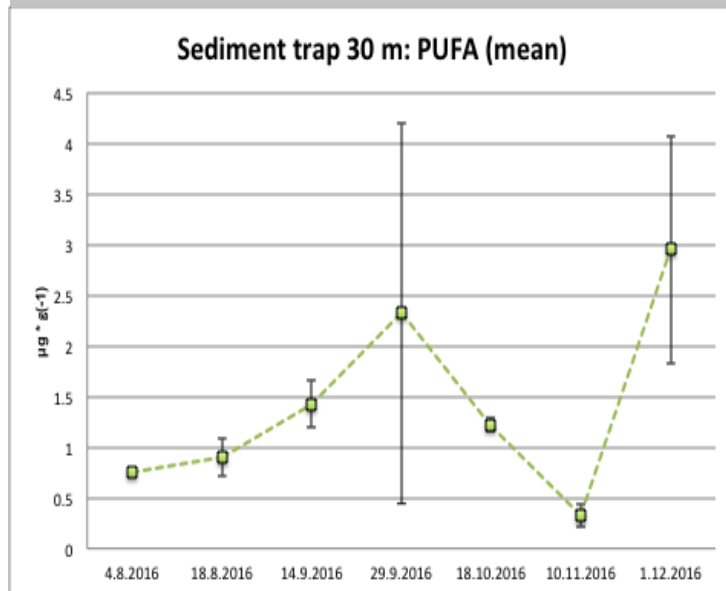
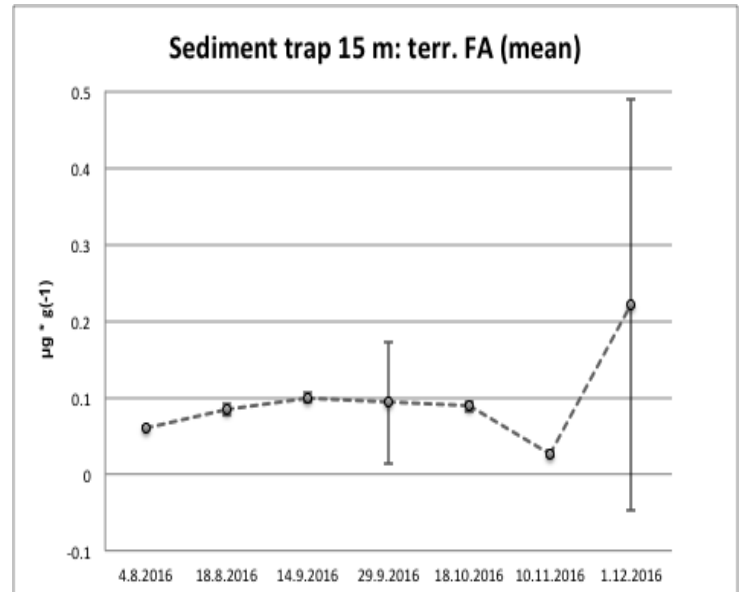
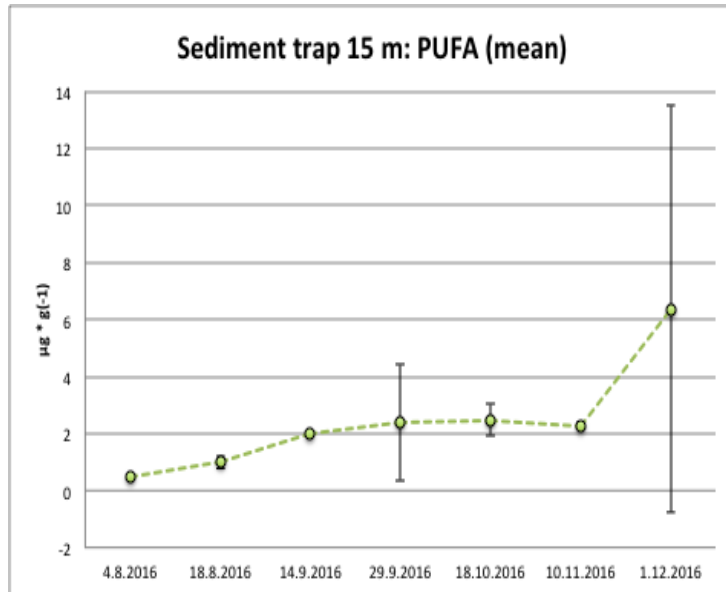


Fig. 14: Mean PUFA and terrestrial fatty acid concentrations ( $\mu\text{g} * \text{g DW}^{-1}$ ) with standard deviations of the 15 m and 30 m placed sediment traps.