

## **Point by point replies**

Dear Sébastien, dear reviewer,

All suggested minor revisions were incorporated. The reviewer had some valuable suggestions regarding word-use and terminology, and phrasing was adjusted where needed. We also updated some affiliations and sample storage details.

We would very much like to thank the reviewers and the editor again for improving this paper, we really appreciate their time.

Kind regards on behalf of all co-authors,

Tessa

# Dynamics of deep soil carbon – insights from <sup>14</sup>C time-series across a climatic gradient

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**Abstract.** Quantitative constraints on soil organic matter (SOM) dynamics are essential for comprehensive understanding of the terrestrial carbon cycle. Deep soil carbon is of particular interest, as it represents large stocks and its turnover ~~rates-times~~ remain highly uncertain. In this study, SOM dynamics in both the top and deep soil across a climatic (average temperature ~1-9 °C) gradient are determined using time-series (~20 years) <sup>14</sup>C data from bulk soil and water-extractable organic carbon (WEOC). Analytical measurements reveal enrichment of bomb-derived radiocarbon in the deep soil layers on the bulk level during the last two decades. The WEOC pool is strongly enriched in bomb-derived carbon, indicating that it is a dynamic pool. Turnover time estimates of both the bulk and WEOC pool show that the latter cycles up to a magnitude faster than the former. The presence of bomb-derived carbon in the deep soil, as well as the rapidly turning WEOC pool across the climatic gradient implies that there likely is a dynamic component of carbon in the deep soil. Precipitation and bedrock type appear to exert a stronger influence on soil C turnover time and stocks as compared to temperature.

## 1 Introduction

Within the broad societal challenges accompanying climate and land use change, a better understanding of the drivers of carbon turnover time of carbon in the largest terrestrial reservoir of organic carbon, as constituted by soil organic matter (SOM), is essential (Batjes, 1996; Davidson and Janssens, 2006; Doetterl et al., 2015; Prietzel et al., 2016). Terrestrial carbon turnover time remains one of the largest uncertainties in climate model predictions (Carvalhais et al., 2014; He et al., 2016). At present, there is no consensus on the net effect that climate and land use change will have on SOM stocks (Crowther et al., 2016; Gosheva et al., 2017; Melillo et al., 2002; Schimel et al., 2001; Trumbore and Czimczik, 2008). Deep soil carbon is of particular interest because of its large stocks (Jobbagy and Jackson, 2000; Balesdent et al., 2018; Rumpel and Kogel-Knabner, 2011) and perceived stability. The stability is indicated by low <sup>14</sup>C content (Rethemeyer et al., 2005; Schrumpf et al., 2013; van der Voort et al., 2016) and low microbial activity (Fierer et al., 2003). Despite its importance, deep soil carbon has not been ~~sparingly-frequently~~ studied and remains poorly understood (Angst et al., 2016; Mathieu et al., 2016; Rumpel and Kogel-Knabner, 2011). The inherent complexity of SOM and the multitude of drivers controlling its stability further impedes the understanding of this globally significant carbon pool (Schmidt et al., 2011). In this framework, there is a particular interest in the portion of soil carbon that could be most vulnerable to change, especially in colder climates (Crowther et al., 2016). Water-exactable organic carbon (WEOC) is seen as a dynamic and potentially vulnerable carbon pool in the soil (Hagedorn et al., 2004; Lechleitner et al., 2016).

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44 Radiocarbon ( $^{14}\text{C}$ ) can be a powerful tool to determine the dynamics of carbon turnover **time** over decadal to  
45 millennial timescales because of the incorporation of bomb-derived  $^{14}\text{C}$  introduced in the atmosphere in the 1950's  
46 as well as the radioactive decay of  $^{14}\text{C}$  naturally present in the atmosphere (Torn et al., 2009). Furthermore,  $^{14}\text{C}$   
47 can also be employed to identify petrogenic (or geogenic) carbon in the soil profile. Understanding the potential  
48 mobilization of stabilized petrogenic carbon is key because it could constitute an additional  $\text{CO}_2$  source to the  
49 atmosphere (Hemingway et al., 2018). Time-series  $^{14}\text{C}$  data is particularly insightful because it enables the  
50 tracking of recent decadal carbon. Furthermore, single time-point  $^{14}\text{C}$  data can yield two estimates for turnover  
51 time, whilst time-series data yields a single turnover **time** estimate (Torn et al., 2009). Given that the so-called  
52 “bomb radiocarbon spike” will continue to diminish in the coming decades, time-series measurements are  
53 increasingly a matter of urgency in order to take full advantage of this intrinsic tracer (Graven, 2015). Several  
54 case-studies have collected time-series  $^{14}\text{C}$  soil datasets and demonstrated the value of this approach (Baisden and  
55 Parfitt, 2007; Prior et al., 2007; Fröberg et al., 2010; Mills et al., 2013, Schrumpf and Kaiser, 2015). However,  
56 these studies are **sparse**, based on specific single sites and have been rarely linked to abiotic and biotic  
57 parameters. Much more is yet to be learned about the carbon cycling through time-series observations in top- and  
58 subsoils along environmental gradients. Furthermore, to our knowledge, there are no studies with pool-specific  
59  $^{14}\text{C}$  soil time-series focusing on labile carbon.

60  
61 This study assesses two-pool soil carbon dynamics as determined by time-series (~20 years) radiocarbon across a  
62 climatic gradient. The time-series data is analyzed by a numerically optimized model with a robust error reduction  
63 to yield carbon turnover **time** estimates for the bulk and dynamic WEOC pool. Model output is linked to potential  
64 drivers such as climate, forest productivity and physico-chemical soil properties. The overall objective of this  
65 study is to improve our understanding of shallow and deep soil carbon dynamics in a wide range of ecosystems.

## 67 **2 Materials and methods**

### 68 **2.1 Study sites, sampling strategy and WEOC extraction**

69 The five sites investigated in this study are located in Switzerland between 46-47° N and 6-10° E and encompass  
70 large climatic (mean annual temperature (MAT) 1.3-9.2°C, mean annual precipitation (MAP) 864-2126 mm  $\text{m}^{-2}\text{y}^{-1}$ ) and geological gradients (Table 1). The sites are part of the Long-term Forest Ecosystem Research program  
72 (LWF) at the Swiss Federal Institute for Forest, Snow and Landscape Research, WSL (Schaub et al., 2011; Etzold  
73 et al., 2014). The soils of these sites were sampled between 1995 and 1998 (Walthert et al., 2002, 2003) and were  
74 re-sampled following the same sampling strategy in 2014 with the aim to minimize noise caused by small-scale  
75 soil heterogeneity. In both instances sixteen samples were taken on a regular grid on the identical 43 by 43 meters  
76 (~1600  $\text{m}^2$ ) plot (Fig. 1; see Van der Voort et al., 2016 for further details). For the archived samples taken between  
77 1995 and 1998, mineral soil samples down to 40 cm depth (intervals of 0-5, 5-10, 10-20 and 20-40 cm) were taken  
78 on an area of 0.5 by 0.5 m (0.25  $\text{m}^2$ ). For samples >40 cm (intervals of 40-60, 60-80 and 80-100 cm), corers were  
79 used to acquire samples ( $n=5$  in every pit, area  $\sim 2.8 \times 10^{-3} \text{ m}^2$ ). The organic layer was sampled by use of a metal  
80 frame (30×30 cm). The samples were dried at 35-40°C, sieved to remove coarse material (2 mm), and stored in  
81 hard plastic containers under controlled climate conditions in the “Pedothek” at WSL (Walthert et al., 2002). For  
82 the samples acquired in 2014 the same sampling strategy was followed, and samples were taken on the exact same  
83 plot proximal (~10 m) to the legacy samples. For the sampling, a SHK Martin Burch AG HUMAX soil corer

84 (~2×10<sup>-3</sup> m<sup>2</sup>) was used for all depths (0-100 cm). For the organic layer, a metal frame of 20×20 cm was used to  
85 sample. Samples were sieved (2 mm), frozen and freeze-dried using an oil-free vacuum-pump powered freeze  
86 dryer (Christ, Alpha 1-4 LO *plus*). For the time-series radiocarbon measurements, all samples covering ~1600 m<sup>2</sup>  
87 were pooled to one composite sample per soil depth using the bulk-density. In order to determine bulk-density of  
88 the fine earth of the 2014 samples, stones > 2 mm were assumed to have a density of 2.65 g/cm<sup>3</sup>. For the Alptal  
89 site, sixteen cores were taken on a slightly smaller area (~1500 m<sup>2</sup>) which encompasses the control plot of a  
90 nitrogen addition experiment (NITREX project) (Schleppi et al., 1998). For this site, no archived samples are  
91 available and thus only the 2014 samples were analyzed. Soil carbon stocks were estimated by multiplying SOC  
92 concentrations with the mass of soil calculated from measured bulk densities and stone contents for each depth  
93 interval (Gosheva et al., 2017). For the Nationalpark site, the soil carbon stocks from 80-100 cm were estimated  
94 using data from a separately dug soil profile (Walthert et al., 2003) because the HUMAX corer could not penetrate  
95 the rock-dense soil below 80 cm depth. In order to understand very deep soil carbon dynamics (i.e. >100 cm), this  
96 study also includes single-time point <sup>14</sup>C analyses of soil profiles that were dug down to the bedrock between  
97 1995 and 1998 as part of the LWF programme on the same sites (Walthert et al., 2002). The sampling of the  
98 profiles has not yet been repeated.

99

## 100 **2.2 Climate and soil data**

101 Temperature and precipitation data are derived from weather stations close to the study sites that have been  
102 measuring for over two decades, yielding representative estimates of both variables and over the time period  
103 concerned in this study (Etzold et al., 2014). The pH values for all sites and concerned depth intervals were  
104 acquired during the initial sampling campaign (Walthert et al. 2002). At Alptal, pH values were determined as  
105 described in Xu et al. (2009), values of 10-15 cm were extrapolated to the deeper horizons because of the uniform  
106 nature of the Gley horizon. The Beatenberg Podzol is marked by strong eluviation (~4-35 cm) and illuviation  
107 (~35-60 cm) (Walthert et al., 2003). Exchangeable cations were extracted (in triplicate) from the 2-mm-sieved  
108 soil in an unbuffered solution of 1 M NH<sub>4</sub>Cl for 1 hour on an end-over-end shaker using a soil-to-extract ratio of  
109 1:10. The element concentrations in the extracts were determined by inductively coupled plasma atomic emission  
110 spectroscopy (ICP-AES) (Optima 3000, Perkin–Elmer). Contents of exchangeable protons were calculated as the  
111 difference between the total and the Al-induced exchangeable acidity as determined (in duplicate) by the KCl  
112 method (Thomas, 1982). This method was applied only to soil samples with a pH (CaCl<sub>2</sub>) < 6.5. In samples with  
113 a higher pH, we assumed the quantities of exchangeable protons were negligible. The effective cation-exchange  
114 capacity (CEC) was calculated by summing up the charge equivalents of exchangeable Na, K, Mg, Ca, Mn, Al,  
115 Fe and H. The base saturation (BS) was defined as the percental fraction of exchangeable Na, K, Mg, and Ca of  
116 the CEC (Walthert et al., 2002, 2013). Net primary production (NPP) was determined by Etzold et al. (2014) as  
117 the sum of carbon fluxes by woody tree growth, foliage, fruit production and fine root production. Soil texture  
118 (sand, silt and clay content) on plot-averaged samples taken in 2014 have been determined using grain size classes  
119 for sand, silt and clay respectively of 0.05-2 mm, 0.002-0.05 mm and <0.002 mm according to Klute (1986). The  
120 continuous distribution of grain sizes was also determined after removal of organic matter (350 °C for 12 h) using  
121 the Mastersizer 2000 (Malvern Instruments Ltd.). Soil water potential (SWP) was measured on the same sites as  
122 described in Von Arx et al., (2013). In accordance with Mathieu et al., (2015), topsoil refers to the mineral soil  
123 up to 20 cm depth, and deep soil refers to mineral soil below 20 cm. Out of the five sites, two are hydromorphic

124 (Gleysol and Podzol in Alptal and Beatenberg respectively), whilst the others are non-hydromorphic (Luvisol,  
125 Cambisol and Fluvisol in Othmarsingen, Lausanne and Nationalpark respectively).

126

### 127 **2.3 Isotopic (<sup>14</sup>C, <sup>13</sup>C) and compositional (C, N) analysis**

128 Prior to the isotopic analyses, inorganic carbon in all samples was removed by vapour acidification for 72 hours  
129 (12M HCl) in desiccators at 60 °C (Komada et al., 2008). After fumigation, the acid was neutralised by substituting  
130 NaOH pellets for another 48 hours. All glassware used during sample preparation was cleaned and combusted at  
131 450°C for six hours prior to use. Water extractable organic carbon (WEOC) was procured by extracting dried soil  
132 with of 0.5 wt% pre-combusted NaCl in ultrapure Milli-Q (MQ) water in a 1:4 soil:water mass ratio (adapted from  
133 Hagedorn et al., (2004), details in Lechleitner et al., (2016)).

134 In order to determine absolute organic carbon and nitrogen content as well as <sup>13</sup>C values, an Elemental Analyser-  
135 Isotope Ratio Mass Spectrometer system was used (EA-IRMS, Elementar, vario MICRO cube – Isoprime, Vison).  
136 Atropine (Säntis) and an in-house standard peptone (Sigma) were used for the calibration of the EA-IRMS for  
137 respectively carbon concentration, nitrogen concentration and C:N ratios and <sup>13</sup>C. High <sup>13</sup>C values were used to  
138 flag if all inorganic carbon had been removed by acidification.

139 For the <sup>14</sup>C measurements of the bulk soil samples were first graphitised using an EA-AGE (elemental analyser-  
140 automated graphitization equipment, Ionplus AG) system at the Laboratory of Ion Beam Physics at ETH Zürich  
141 (Wacker et al., 2009). Graphite samples were measured on a MICADAS (MIniturised radioCARbon DAting  
142 System, Ionplus AG) also at the Laboratory of Ion Beam Physics, ETH Zürich (Wacker et al., 2010). For three  
143 samples (Alptal depth intervals 40-60, 60-80 and 80-100 cm) the <sup>14</sup>C signature was directly measured as CO<sub>2</sub> gas  
144 using the recently developed online elemental analyzer (EA) - stable isotope ratio mass spectrometers (IRMS)-  
145 AMS system et ETH Zürich (McIntyre et al., 2016). Oxalic acid (NIST SRM 4990C) was used as the normalising  
146 standard. Phthalic anhydride and in-house anthracite coal were used as blank. Two in-house soil standards (Alptal  
147 soil 0-5 cm, Othmarsingen soil 0-5 cm) were used as secondary standards. For the WEOC, samples were converted  
148 to CO<sub>2</sub> by Wet Chemical Oxidation (WCO) (Lang et al., 2016) and run on the AMS using a Gas Ion Source (GIS)  
149 interface (Ionplus). To correct for contamination, a range of modern standards (sucrose, Sigma, δ<sup>13</sup>C = -12.4 ‰  
150 VPDB, F<sup>14</sup>C = 1.053 ± 0.003) and fossil standards (phthalic acid, Sigma, δ<sup>13</sup>C = -33.6‰ VPDB, F<sup>14</sup>C <0.0025)  
151 were used (Lechleitner et al., 2016).

152

153

## 2.4 Numerical optimization to find carbon turnover **time** and size of the dynamic pool

### 2.4.1 Turnover **time** based on a single $^{14}\text{C}$ measurement

The  $^{14}\text{C}$  signature of a sample can be used to estimate turnover time of a carbon pool (Torn et al., 2009).

$$R_{\text{sample},t} = k \times R_{\text{atm},t} + (1 - k - \lambda) \times R_{\text{sample}(t-1)} \quad (1)$$

$$R_{\text{sample},t} = \frac{\Delta^{14}\text{C}_{\text{sample}}}{1000} + 1 \quad (2)$$

In Eq. 1-2, the constant for radioactive decay of  $^{14}\text{C}$  is indicated as  $\lambda$ , the decomposition rate  $k$  (inverse of turnover time) is the only unknown in this equation and is hence the variable for which the optimal value that fits the data is sought using the model. The  $R$  value of the sample is inferred from  $\Delta^{14}\text{C}$ , hence accounting for the sampling year, as shown in Eq. (2) (Herold et al., 2014; Solly et al., 2013). In order to avoid ambiguity, the term *turnover time* and not i.e. mean residence time is used solely in this manuscript (Sierra et al., 2016).

For the turnover time estimation, we assumed the system to be in steady state over the modeled period ( $\sim 1 \times 10^4$  years, indicating soil formation since the last glacial retreat (Ivy-Ochs et al., 2009)), hence accounting both for radioactive decay and incorporation of the bomb-testing derived material produced in the 1950's and 1960's (Eq. 1.) (Herold et al., 2014; Torn et al., 2009). We assumed an initial fraction modern ( $F_m$ ) of  $^{14}\text{C}$  value of 1 at 10000 B.C.. For the period after 1900 atmospheric fraction modern ( $F_m$ ) values of the Northern Hemisphere were used (Hua et al., 2013). This equation could be solved in Excel with manual iterations (e.g. Herold et al., 2014), or alternatively a numerical optimization can be used to find the best fit automatically. In this paper, we used a numerical optimization constructed in MATLAB version 2015a (The MathWorks, Inc., Natick, Massachusetts, United States) to find the best fit. The numerical optimization is exhaustive, meaning that every single turnover **time** value from 1 to 10,000 years with an interval of 0.1 year is tested. The error is defined as the difference between the fitted value of  $R$  and the measured value (Eq. 3). The turnover **time** value with the lowest error is then automatically selected.

$$\text{Error}_{\text{single timepoint}} = |R_{\text{calculated}} - R_{\text{measured}}| \quad (3)$$

The residual error of each fit are provided in the Supplemental Information (SI) Table 3. Turnover times determined with the numerical optimization match the manually optimized turnover **time** modeling published previously (Herold et al., 2014; Solly et al., 2013).

### 2.4.2 Turnover **time** based on two $^{14}\text{C}$ measurements

A single  $^{14}\text{C}$  value could yield possible turnover **time** values (Torn et al., 2009; Graven et al., 2015). If there is a time-series  $^{14}\text{C}$  dataset, this problem can be eliminated. In this paper, we have time-series data of both the bulk soil, as well as the vulnerable fraction (WEOC). For all samples a time-series dataset is available, both data points are employed to give the best estimate of turnover time. The same numerical optimization (Eq. 1 and 2) as we did for a single time-point, except that we try to find the best fit for both time points whilst reducing the compounding residual mean square error (RSME, Eq. 4). As can be seen in Fig. 2a, single time points can yield two likely

188 turnover times but when two datapoints are available, a single value can be found. The input data for Figure 2 can  
 189 be found in SI Table 1. The results of the time-series turnover [time](#) modelling for both the bulk and WEOC pool  
 190 of the sub-alpine site Beatenberg are shown in Fig. 3.

$$191 \quad Error_{two\ timepoints} = \sqrt{|R_{calculated} - R_{measured}|_{time\ point\ 1}^2 + |R_{calculated} - R_{measured}|_{time\ point\ 1}^2} \quad (4)$$

### 192 2.4.3 Vegetation-induced lag

193 In order to account for vegetation-lag, two scenarios were run: firstly (1) with no assumed lag between the fixation  
 194 of carbon from the atmosphere and input into to the soil and (2) model run with a lag of fixation of the atmospheric  
 195 carbon as inferred from the dominant vegetation (Von Arx et al., 2013; Etzold et al., 2014). In the case of full  
 196 deciduous trees coverage a lag of two years was assumed, and for the case of 100% conifer-dominated coverage  
 197 a lag of 8 years was incorporated (Table 1).

### 198 2.4.4 Turnover [time](#) and size vulnerable pool based on two-pool model

199 As SOM is complex and composed of a continuum of pools with various ages (Schrumppf and Kaiser, 2015) and  
 200 there is data available from two SOM pools, the  $^{14}\text{C}$  time-series data can be leveraged to create a two-pool model.  
 201 The following assumptions were made: First, both pools (slow & fast) make up the total carbon pool (Eq. 5).  
 202 Secondly, the total turnover [time](#) of the bulk soil is made up out of the “dynamic” fraction turnover [time](#) multiplied  
 203 by “dynamic” fraction pool size and the “slow” pool turnover [time](#) multiplied by “slow” pool size (Eq. 6).  
 204 Furthermore, we assume that the signature of the sample (the time-series bulk data) is determined by the rate of  
 205 incorporation of the material (atmospheric signal) and the loss of carbon the two pools (Eq. 7). Lastly, we assume  
 206 that the radiocarbon signal of the WEOC pool is representative for a dynamic pool, as it could be representative  
 207 for a larger component of rapidly turning over carbon, even in the deep soil (Baisden and Parfitt, 2007; Koarashi  
 208 et al., 2012). The turnover [rate-time](#) of the slow pool was set between 100 and 10.000 years, with a time-step of  
 209 10 years. The size of the dynamic pool was set to be between 0 and 0.5, with a size-step of 0.01.

210

$$211 \quad 1 = F_1 + F_2 \quad (5)$$

$$212 \quad k_{total} = (F_1/k_1 + F_2/k_2)^{-1} \quad (6)$$

$$213 \quad R_{sample,t} = k_{total} \times R_{atm,t} + F_1[(1 - k_1 - \lambda) \times R_{sample(t-1)}] + F_2(1 - k_2 - \lambda) \times R_{sample(t-1)} \quad (7)$$

214 Where  $F_1$  is the relative size of the dynamic pool, and  $F_2$  is the relative size of the (more) stable pool. The  $k_1$  is  
 215 the inverse of the turnover time of the dynamic or WEOC as determined using the numerical optimisation of Eq.  
 216 1-4. The  $k_2$  is the inverse of the turnover time of the slow pool. The calculation of the error term becomes for  
 217 complex because it needs to be recalculated for each unique combination of pool-size distribution (Eq. 5) and  
 218 turnover time (inverse of  $k$ , Eq. 6). Therefore, the error space changes from column vector to a two-dimensional

219 matrix of length of the step size increments ( $F_i$ ) and width of the inverse of the turnover time of the slow pool  
220 ( $k_2$ ).

$$221 \quad Error_{k_2, F_1} = \sqrt{|R_{calculated} - R_{measured}|_{time\ point\ 1}^2 + |R_{calculated} - R_{measured}|_{time\ point\ 2}^2} \quad (8)$$

$$222 \quad Error = Min(Error_{k_2, F_1}) \quad (9)$$

223 The numerical optimization finds the likeliest solution for the given dataset. This model constitutes a best fit, and  
224 more data would better constrain the results. Additional details can be found in the Supplementary Information  
225 (SI) text and SI Fig. 1. **All Matlab-based numerical optimization codes can be found in the SI.** For correlations  
226 (packages HMISC, corrgram, method = pearson), statistical software R version 1.0.153 was used.

227

### 228 **3 Results**

#### 229 **3.1 Changes of radiocarbon signatures over time**

230 Overall, there is a pronounced decrease in radiocarbon signature with soil depth at all sites (Fig. 4). The time-  
231 series results show clear changes in radiocarbon signature over time from the initial sampling period (1995-1998)  
232 as compared to 2014, with the magnitude of change depending on site and soil depth. In the uppermost 5 cm of  
233 soils, the overarching trend in the bulk soil is a decrease in the  $^{14}\text{C}$  bomb-spike signature in the warmer climates  
234 (Othmarsingen, Lausanne), whilst at higher elevation (colder) sites (Beatenberg, Nationalpark) the bomb-derived  
235 carbon appears to enter the top soil between 1995-8 and 2014.

236 Water-extractable OC (WEOC) has an atmospheric  $^{14}\text{C}$  signature in the top soil at all sites in 2014. The  
237 deep soil in the 1990's still has a negative  $\Delta^{14}\text{C}$  signature of WEOC at multiple sites. There are two distinguishable  
238 types of depth trends for WEOC in the 2014 dataset: (1) WEOC has the same approximate  $^{14}\text{C}$  signature  
239 throughout depth (Othmarsingen, Beatenberg), (2) WEOC becomes increasingly  $^{14}\text{C}$  depleted with depth (Alptal,  
240 Nationalpark), or an intermediate form where WEOC  $^{14}\text{C}$  is modern throughout the top soil but becomes more  
241 depleted of  $^{14}\text{C}$  in the deep soil (Lausanne) (Fig. 4). The isotopic trends of WEOC co-vary with grain size as  
242 inherited from the bedrock type (Walther et al., 2003). Soils with a relatively modern WEOC  $^{14}\text{C}$  signature in 2014  
243 (down to 40 cm) are underlain by bedrock with large grained (SI Fig. 2, Table SI 3) components (the moraines  
244 and sandstone at Othmarsingen, Lausanne and Beatenberg respectively). Soils where WEOC  $^{14}\text{C}$  signature decreases  
245 with depth are underlain by bedrock containing fine-grained components. For instance, the Flysch in Alptal  
246 (Schleppi et al., 1998) and intercalating layers of silt and coarse grained alluvial fan in Nationalpark (Walther et  
247 al., 2003) respectively.

248

#### 249 **3.2 Carbon turnover time patterns**

250 Incorporation of a vegetation-induced time lag (Table 2, SI Table 2) has an effect on modelled carbon  
251 dynamics in the organic layer, but this effect is strongly attenuated in the 0-5 cm layer in the mineral soil and  
252 virtually absent for the deeper soil layers. The residual errors associated to the carbon turnover time estimates  
253 converge to a single point (Figure 2) and are low (i.e.  $< 0.06 R$ , SI Tables 3 and 4). Turnover times show two  
254 modes of behavior for well-drained soils and hydromorphic soils, respectively. The non-hydromorphic soils have  
255 relatively similar values with decadal turnover times for the 0-5 cm layer, increasing to an order of centuries down



256 to 20 cm depth, and to millenia in deeper soil layers (~980 to ~3940 years at 0.6 to 1 m depth) (Fig. 5). In contrast,  
257 the hydromorphic soils are marked by turnover times that are up to an order of magnitude larger, from centennial  
258 in top soil to (multi-) millennial in deeper soils. At the Beatenberg Podzol, turnover time in the shallow layer  
259 which overlaps with the elluvial horizon is slower (20-40 cm, ~1900 y) than the deepest layer (40-60 cm, ~1300  
260 y), which overlaps with the illuvial horizon (Figure 5, SI Table 5).

261 Carbon stocks also show distinct difference between drained and hydromorphic soils with greater stock  
262 in the hydromorphic soils (~15 kg C m<sup>-2</sup> at Beatenberg and Alptal vs. ~6 - ~7 kg C m<sup>-2</sup> at Othmarsingen, Lausanne  
263 and Nationalpark, Fig. 5, Table 3)).

264 The turnover times of the WEOC mimic the trends in the bulk soil but are up to an order of magnitude  
265 faster. Considering WEOC turnover time in the non-hydromorphic soils only, there is a slight increase in WEOC  
266 turnover time with decreasing site temperature, but the trend is not significant (SI Table 4). The modeled estimate  
267 for dynamic fraction is variable at the surface but decreases towards the lower top soil (from ~0.2 at 0-5 cm to  
268 ~0.01 at 10-20 cm in Othmarsingen). In the deep soil, the model indicates there could also be a non-negligible  
269 proportion of dynamic carbon (e.g. 0.10-0.23 at 20-40 cm). The residual errors associated to the error reduction  
270 of the two-pool model are also low (i.e. < 0.06 R), but do not converge as strongly as the single-pool model (SI  
271 Figure 1).

272

273

### 274 3.3 Pre-glacial carbon in deep soil profiles

275 The turnover times of deep soil carbon exceed 10,000 years in several profiles, indicating the presence of carbon  
276 that pre-dates the glacial retreat (Fig. 6). These profiles are located on carbon-containing bedrock and concern the  
277 deeper soil (80-100 cm) of the Gleysol (Alptal), as well as >100 cm in the Cambisol (Lausanne) (Fig. 6, SI Table  
278 6).

279

### 280 3.4 Environmental drivers of carbon dynamics

281 Pearson correlation was used to assess potential relationships between carbon stocks and turnover time and their  
282 potential controlling factors (climate, NPP, soil texture, soil moisture and physicochemical properties (Table 4,  
283 SI Table 7, 8)). For the averaged top soil (0-20 cm, n=5), carbon stocks were significantly positive correlated to  
284 Mean Annual Precipitation (MAP). Turnover time in the bulk top soil negatively correlated with silt content and  
285 positively with average grain size. Turnover time in the WEOC of the top soil did not correlate significantly with  
286 any parameter except a weak positive correlation with grain size. Deeper soil bulk stock and turnover time  
287 positively correlated with MAP and iron content.

288

## 289 4 Discussion

### 290 4.1 Dynamic deep soil carbon

#### 291 4.1.1 Rapid shifts in <sup>14</sup>C abundance reflect dynamic deep carbon

292 The propagation of bomb-derived carbon into supposedly stable deep soil on the bulk level across the climatic  
293 gradient implies that SOM in deep soil contains a dynamic pool and could be less stable and potentially more  
294 vulnerable to change than previously thought. This possibility is further supported by the WEO<sup>14</sup>C which is  
295 consistently more enriched in bomb-derived carbon than the bulk soil. Near-atmospheric signature WEO<sup>14</sup>C

296 pervades up to 40 or even 60 cm depth. Hagedorn et al., (2004) also found WEOC to be a highly dynamic pool  
297 using <sup>13</sup>C tracer experiments in forest soils.  
298 We consider our <sup>14</sup>C comparison over time to be robust because the grid-based sampling and averaging was  
299 repeated on the same plots which excludes the effect plot-scale variability (Van der Voort et al., 2016). Our <sup>14</sup>C  
300 time-series data in the deep soil corroborate pronounced changes in <sup>14</sup>C (hence substantial SOM ~~turnover~~changes)  
301 ~~observed~~ in subsoils of an area with pine afforestation (Richter and Markewitz, 2001). The findings are also in  
302 agreement with results from an incubation study by Fontaine et al., (2007) which showed that the deep soil can  
303 have a significant dynamic component. Baisden et al., (2007) also found indications of a deep dynamic pool using  
304 modeling on <sup>14</sup>C time-series on the bulk level on a New Zealand soil under stable pastoral management.  
305

#### 306 4.1.2 Carbon dynamics reflect soil-specific characteristics at depth

307 Bulk carbon turnover ~~time~~ for the top and deeper soil fall in the range of prior observations and models, although  
308 the data for the latter category is ~~sparse-rare~~ (Scharpenseel and Becker-Heidelmann, 1989; Paul et al., 1997;  
309 Schmidt et al., 2011; Mills et al., 2013; Braakhekke et al., 2014). The carbon turnover ~~time~~ is related to soil-  
310 specific characteristics. The ~~slower-turnover~~higher turnover times of hydromorphic as compared to non-  
311 hydromorphic soils is likely due to increased waterlogging and limited aerobicity (Hagedorn et al., 2001) which  
312 is conducive to slow ~~turnover-decomposition rates~~ and enhanced carbon accumulation. The WEOC turns over up  
313 to an order of magnitude faster than the bulk and mirrors these trends, indicating that it indeed is a more dynamic  
314 pool (Hagedorn et al., 2004; Lechleitner et al., 2016). Results also reflect known horizon-specific dynamics for  
315 certain soil types, particularly in the deep soil. The hydromorphic Podzol at Beatenberg shows specific  
316 pedogenetic features such as an illuviation layer with an enrichment in humus and iron in the deeper soil (Walthert  
317 et al., 2003) where ~~the~~ turnover ~~time~~ of bulk and WEOC is ~~faster-more rapid~~ and stocks are higher as compared  
318 to the elluvial layer above (Fig. 5). This is likely due to the input of younger carbon via leaching of dissolved  
319 organic carbon. The non-hydromorphic Luvisols are marked by an enrichment of clay in the deeper soil, which  
320 can enhance carbon stabilization (Lutzow et al., 2006). This also reflected in the turnover time of the 60-80 cm  
321 layer in the Othmarsingen Luvisol – in this clay-enriched depth interval (Walthert et al., 2003), turnover ~~time~~ is  
322 relatively ~~slow-high~~ as compared to the other (colder) non-hydromorphic soils (Fig. 5). These patterns are  
323 consistent with findings by Mathieu et al., (2015) that the important role of soil pedology on deep soil carbon  
324 dynamics.  
325

#### 326 4.1.3 Dynamic carbon at depth & implications for carbon transport

327 The analytical <sup>14</sup>C data as well as turnover time estimates indicate that there is likely a dynamic portion of carbon  
328 in the deep soil. The estimated size of the dynamic pool can be large, even at greater depth than it was observed  
329 by other <sup>14</sup>C time-series (Richter and Markewitz, 2001; Baisden and Parfitt, 2007; Koarashi et al., 2012). The two-  
330 pool modelling indicates that the size of dynamic pool in the deep soil can be upwards of ~10%. A deep dynamic  
331 pool is consistent with findings of a <sup>13</sup>C tracer experiment by Hagedorn et al., (2001) that shows with that relatively  
332 young (<4 years) carbon can be rapidly incorporated in the top soil (20% new C at 0-20 cm depth) but also in the  
333 deep soil (50 cm), and findings by Balesdent et al., (2018) which estimate that up to 21% of the carbon between  
334 30-100 cm is younger than 50 years. Rumpel and Kögel-Knabner (2011) have highlighted the importance of the  
335 poorly understood deep soil carbon stocks and a significant dynamic pool in the deep soil could imply that carbon

336 is more vulnerable than initially suspected. One major input pathway of younger C into deeper soils is the leaching  
337 of DOC (Kaiser and Kalbitz, 2012; Sanderman and Amundson, 2009). Here, we have measured WEOC – likely  
338 primarily composed of microbial metabolites (Hagedorn et al., 2004) – carrying a younger <sup>14</sup>C signature than bulk  
339 SOM and thus, representing a translocator of fresh carbon to the deep soil. The WEOC turnover time is in the  
340 order of decades, implying that it is not directly derived from decaying vegetation, but rather composed of  
341 microbial material feeding on the labile portion of the bulk soil. In addition to WEOC, roots and associated  
342 mycorrhizal communities may also provide a substantial input of new C into soils in deeper soils (Rasse et al.,  
343 2005). Additional modelling such as in CENTURY and RotC could provide additional insights into the soil carbon  
344 dynamics and fluxes (Manzoni et al., 2009)

345

#### 346 4.2 Contribution of petrogenic carbon

347 Our results on deep soil carbon suggest the presence of pre-aged or <sup>14</sup>C-dead (fossil), pre-interglacial carbon in  
348 the Alptal (Gleysol) and Lausanne (Cambisol) profiles, implying that a component of soil carbon is not necessarily  
349 linked to recent (< millennial) terrestrial productivity and instead constitutes part of the long-term (geological)  
350 carbon cycle (> millions of years). In the case of the Gleysol in Alptal, the <sup>14</sup>C-depleted material could be derived  
351 from the poorly consolidated sedimentary rocks (Flysch) in the region (Hagedorn et al., 2001a; Schleppei et al.,  
352 1998; Smith et al., 2013), whereas carbon present in glacial deposits and molasse may contribute in deeper soils  
353 at the Lausanne (Cambisol) site. The potential contribution of fossil carbon was estimated using a mixing model  
354 using the signature of a soil without fossil carbon, the signature of fossil carbon and the measured values (SI Table  
355 4). Fossil carbon contribution in the Alptal profile between 80-100 cm (Fig. 6, SI Table 4) is estimated at ~40 %.  
356 Below one meter at Lausanne site the petrogenic percentage ranges from ~20% at 145 cm up to ~80 % at 310 cm  
357 depth (Fig. 6, SI Table 4).

358 Other studies analyzing soils have observed the significant presence of petrogenic (geogenic in soil  
359 science terminology) in loess-based soils (Helfrich et al., 2007; Paul et al., 2001). Our results suggest that pre-  
360 glacial carbon may comprise a dominant component of deep soil organic matter in several cases, resulting in an  
361 apparent increase in the average ~~age (and decrease in turnover)~~ turnover time of carbon in these soils. Hemingway  
362 et al., (2018) have highlighted that fossil carbon oxidized in soils can lead to significant additional CO<sub>2</sub> emissions.  
363 Therefore, the potential of soils to ‘activate’ fossil petrogenic carbon should be considered when evaluating the  
364 soil carbon sequestration potential.

365

#### 366 4.3 Controls on carbon dynamics and cycling

367 In order to examine the effects of potential ~~drivers-controls~~ on soil C turnover time and stocks, we explore  
368 correlations between a number of available factors which have previously been proposed, such as texture, geology,  
369 precipitation, temperature and soil moisture (Doetterl et al., 2015; McFarlane et al., 2013; Nussbaum et al., 2014;  
370 Seneviratne et al., 2010; van der Voort et al., 2016).

371 From examination of data for all samples it emerges that C turnover time does not exhibit a consistent correlation  
372 with any specific climatological or physico-chemical factor. This implies that no single mechanism predominates  
373 and/or that there is a combined impact of geology and precipitation as these soil-forming factors affect grain size  
374 distribution, water regime and mass transport in soils. Exploring potential relationships in greater detail, we see  
375 that carbon stocks in the top soil and deep soil as well as turnover time is positively related to MAP, which could

376 be linked to waterlogging and anaerobic conditions even in upland soils leading to a lower decomposition and  
377 thus to a higher build-up of organic material (Keiluweit et al., 2015). Our results are supported by the findings  
378 based on >1000 forest sites that precipitation exerts a strong effect on soil C stocks across Switzerland (Gosheva  
379 et al., 2017; Nussbaum et al., 2014). Furthermore, Balesdent et al., (2008) also highlighted the role of precipitation  
380 and evapotranspiration on deep soil organic carbon stabilisation. Nonetheless, it has to be noted that for these  
381 sites, the precipitation range does not include very dry soils (MAP 864-2126 mm/y). ~~Topsoil carbon turnover~~  
382 ~~time in bulk and WEOC in both top and deep soil was most closely correlated with texture. The positively~~  
383 ~~correlation of top soil turnover with grain size which is likely caused by the large grain size of the waterlogged~~  
384 ~~Podzol Beatenberg, and negative correlation with the amount of silt-sized particles reflects lower stabilization in~~  
385 ~~larger-grained soils as opposed to clay-rich soils with a higher and more reactive surface area (Rumpel and Kogel-~~  
386 ~~Knabner, 2011). Mathieu et al., (2015) also stressed the decisive role of soil pedology on deep soil carbon storage.~~  
387 Overall, geology seems to impact the carbon cycling in three key ways. Firstly, when petrogenic carbon is present  
388 in the bedrock from shale or reworked shale (Schleppi et al., 1998; Walther et al., 2003), fossil carbon contributes  
389 to soil carbon. Secondly, porosity of underlying bedrock either prevents or induces waterlogging which in turn  
390 affects turnover time. Thirdly, the initial components of the bedrock (~~i.e. silt-sized layers in an alluvial fan~~)  
391 influence the final grain size distribution and mineralogy (SI Fig. 2, Table 3), which is also reflected in the carbon  
392 stock, -bulk and pool-specific turnover time. Within the limited geographic and temporal scope of this paper, we  
393 hypothesize that for soil carbon stocks and their turnover time, temperature is not the dominant driver, which has  
394 been concluded by some (Giardina and Ryan, 2000) but refuted by others (Davidson et al., 2000; Feng et al.,  
395 2008). The only climate-related driver which appears to be significant for the deep soil is precipitation. ▲

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#### 397 4.4 Modular robust numerical optimization

398 The numerical approach used here builds on previous work concerning turnover modeling of bomb-radiocarbon  
399 dominated samples (Herold et al., 2014; Solly et al., 2013; Torn et al., 2009) and the approach used in numerous  
400 time-series analysis with box modeling using Excel (Schrumpf and Kaiser, 2015) or Excel solver (Baisden et al.,  
401 2013; Prior et al., 2007). However certain modifications were made in order to (i) provide objective repeatable  
402 estimates, (ii) incorporate longer time-series data, and (iii) identify samples impacted by petrogenic (also called  
403 geogenic) carbon. Identifying petrogenic carbon in the deep soil is important considering the large carbon stocks  
404 in deep soils (Rumpel and Kogel-Knabner, 2011) and the wider relevance of petrogenically-derived carbon in the  
405 global carbon cycle (Galy et al., 2008). This approach is modular and could be adapted in the future to identify  
406 the correct turnover time for time-series <sup>14</sup>C data, which is becoming increasingly important with the falling bomb-  
407 peak (Graven, 2015). For the single and time-series data, the results from the numerical solution were  
408 benchmarked to the Excel-based model, and it was found that the results agree.

409 Other studies (e.g. Baisden and Canessa, 2012; Prior et al., 2007) also use time-series data to estimate the value  
410 for two unknowns simultaneously (size of the pool size and turnover time). The error does not always converge  
411 to single low point, but can have multiple minima (SI Fig. 1). This potential issue should be considered when  
412 interpreting the data. More time-series data is required to eliminate this problem.

413

#### 414 5 Conclusion

415 Time-series radiocarbon ( $^{14}\text{C}$ ) analyses of soil carbon across a climatic range reveals recent bomb-derived  
416 radiocarbon in both upper and deeper bulk soil, implying the presence of a rapidly turning over pool at depth.  
417 Pool-specific time-series measurements of the WEOC indicate this is a more dynamic pool which is consistently  
418 more enriched in radiocarbon than the bulk. Furthermore, the estimated modeled size of the dynamic fraction is  
419 non-negligible even in the deep soil (~0.1-0.2). This could imply that a component of the deep soil carbon could  
420 be more dynamic than previously thought.

421 The interaction between precipitation and geology appears to be the main control on carbon dynamics  
422 rather than site temperature. Carbon turnover time in non-hydromorphic soils is relatively similar (decades to  
423 centuries) despite dissimilar climatological conditions. Hydromorphic soils have turnover times which are up to  
424 an order of magnitude slower. These trends are mirrored in the dynamic WEOC pool, suggesting that in ~~sandy,~~  
425 non-waterlogged (aerobic) soils the transport of relatively modern (bomb-derived) carbon into the deep soil and/or  
426 the microbial processing is enhanced as compared to ~~fine-grained~~ waterlogged (anaerobic) soils.

427 Model results indicate certain soils contain significant quantities of pre-glacial or petrogenic (bedrock-  
428 derived) carbon in the deeper part of their profiles. This implies that soils not only sequester “modern” but can  
429 rather also mobilize and potentially metabolize “fossil” or geogenic carbon.

430 Overall, these time-series  $^{14}\text{C}$  bulk and pool-specific data provide novel constraints on soil carbon  
431 dynamics in surface and deeper soils for a range of ecosystems.

432

433 **Acknowledgements**

434 We would like to acknowledge the SNF NRP68 *Soil as a Resource* program for funding this project (SNF  
435 406840\_143023/11.1.13-31.12.15). We could also like to thank Jerome Balesdent, an anonymous reviewer and  
436 the journal editor Sébastien Fontaine for their helpful comments from which this paper greatly benefitted. We  
437 would like to thank various members of the Laboratory of Ion Beam Physics and Biogeoscience group for their  
438 help with the analyses, in particular Lukas Wacker. We thank Roger Köchli for his crucial help in the field which  
439 enabled an effective time-series comparison and for his help with subsequent analyses. We thank Emily Solly and  
440 Sia Gosheva for their valuable insights, Claudia Zell for her help on the project and in the field, Peter Waldner  
441 and Marco Waiser for facilitating the fieldwork, and Elisabeth Graf-Pannatier for her insights on soil moisture.  
442 The 2014 field campaign would not have been possible without the help of Thomas Blattmann, Lukas Oesch,  
443 Markus Vaas and Niko Westphal. Thanks to Stephane Beaussier for the insights into numerical modeling. Also  
444 thanks to Nadine Keller and Florian Neugebauer for their help in the lab. Last but not least, thanks to Thomas Bär  
445 for summarizing ancillary pH data. Data supporting this paper is provided in a separate data Table. [All soil samples  
446 used in this study are stored in the long-term soil storage facility of the Swiss Federal Institute of Forest, Snow  
447 and Landscape Research \(WSL\).](#)  
448

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676 **Author contributions**

677 T.S. van der Voort planned, coordinated and executed the sampling strategy and sample collection, performed the  
678 analyses, conceptualized and optimized the model and processed resulting data. U. Mannu led the model  
679 development. F. Hagedorn lent his expertise on soil carbon cycling and soil properties. C. McIntyre facilitated  
680 and coordinated the radiocarbon measurements and associated data corrections. L. Walthert and P. Schleppi lent  
681 their expertise on the legacy sampling and provided data for the compositional analysis. N. Haghypour performed  
682 in isotopic and compositional measurements. T. Eglinton provided the conceptual framework and aided in the  
683 paper structure set-up. T.S. van der Voort prepared the manuscript with help of all co-authors.

## Tables

**Table 1** Overview sampling locations and climatic and ecological parameters.

Location	Soil type	Geology	Latitude(N)/ Longitude (E)	Soil depth (m)	Depth waterlogging (m) <sup>1</sup>	Upper limit	Altitude Elevation (m a.s.l.)	MAT °C	MAP mm y <sup>-1</sup>	NPP g C m <sup>-2</sup> y <sup>-1</sup>
Othmarsingen <sup>1, 2, 3</sup>	Luvisol	Calcareous moraine	47°24'/8°14'	>1.9	2.5		467-500	9.2	1024	845
Lausanne <sup>1, 2, 3</sup>	Cambisol	Calcareous and shaly moraine	46°34'/6°39'	>3.2	2.5		800-814	7.6	1134	824
Alptal <sup>1, 2, 3, 4</sup>	Gleysol	Flysch (carbon-holding sedimentary rock)	47°02'/8°43'	>1.0	0.1		1200	5.3	2126	347
Beatenberg <sup>1, 2, 3</sup>	Podzol	Sandstone	46°42'/7°46'	0.65	0.5		1178-1191	4.7	1163	302
Nationalpark <sup>1, 2, 3</sup>	Fluvisol	Calcareous alluvial fan	46°40'/10°14'	>1.1	2.5		1890-1907	1.3	864	111

<sup>1</sup>Walthert et al. (2003) <sup>2</sup>Etzold et al., (2014) <sup>3</sup>Von Arx et al., (2013) <sup>4</sup>Krause et al., (2013) for Alptal data

**Table 2** Vegetation and soil data of the study sites. Soil water potential (hPa) are for 15 cm depth.

Location <sup>1</sup>	Deciduous tree species (%) <sup>3</sup>	Dominant tree species <sup>3</sup>	Inferred lag carbon fixation (y)	Organic layer Type <sup>1</sup>	Soil water potential (hPa) percentiles <sup>3</sup>		
					5%	50%	95%
Othmarsingen	100	<i>Fagus sylvatica</i>	2	Mull	-577	-39	-9
Lausanne	80	<i>Fagus sylvatica</i>	3	Mull	-547	-49	-8
Alptal <sup>4</sup>	15	<i>Picea abies</i>	7	Mor to anmoor	-38	-13	+1
Beatenberg	0	<i>Picea abies</i>	8	Mor	-50	-14	+1
Nationalpark	0	<i>Pinus montana</i>	8	Moder	-388	-65	-13

<sup>1</sup>Walthert et al. (2003) <sup>2</sup>Etzold et al., (2014), <sup>3</sup>Von Arx et al. (2013), <sup>4</sup>Krause et al., (2013)

**Table 3** Soil properties as well as carbon stocks and fluxes in 0-20, 20-60, and 60-100 cm depth of the study sites for the bulk and water-extractable organic carbon (WEOC).

Location	Depth interval (m)	pH <sup>1</sup>	CEC <sup>1</sup> (mmolc/kg)	Fe <sub>exchangeable</sub> (mmolc/kg)	Al <sub>exchangeable</sub> (mmolc/kg)	Sand content (%)	Silt content (%)	Clay content (%)	Carbon stock kgC/m <sup>2</sup>	Average turnover <u>time</u> bulk (y)	Average turnover <u>time</u> WEOC (y)
Othmarsingen <sup>1</sup>	0.0-0.2	4.4	62.2	0.15	42	46.8	35.5	17.6	4.84	173	35
	0.2-0.6	4.4	62.8	0.10	49	44.3	33.3	22.4	1.69	868	518
	0.6-0.8	4.9	99.5	0.06	41	46.7	28.4	25.0	0.28	3938	-
Lausanne <sup>1</sup>	0.0-0.2	4.5	60.8	0.13	43	49.2	32.6	18.2	3.24	353	77
	0.2-0.6	4.6	43.9	0	34	50.2	32.0	17.8	2.12	1239	588
	0.6-1.0	4.8	49.7	0	35	50.5	31.5	18.1	0.69	2246	1502 <sup>5</sup>
Alptal <sup>2,3,4</sup>	0.0-0.2	4.5	417	-	19	19.3	39.4	41.3	7.73	437	162
	0.2-0.6	4.7	340	-	14	4.90	47.0	48.1	7.24	3314	893 <sup>6</sup>
	0.6-1.0	4.7	340	-	-	-	-	-	6.54	5165	-
Beatenberg <sup>1</sup>	Organic layer	3.1	260.2	2.8	33	-	-	-	7.05	53	-
	0.0-0.2	4.0	35.6	1.7	18	84.9	12.4	2.7	3.65	1224	293
	0.2-0.6	4.1	23.1	0.40	17	83.2	12.3	4.6	4.10	1607	677
Nationalpark <sup>1</sup>	0.0-0.2	8.3	171.8	0.1	0.0	47.5	34.8	17.7	3.23	180	92
	0.2-0.6	8.8	106.3	0.0	0.0	61.9	32.5	5.7	0.36	612	214
	0.6-0.8	-	-	0.0	0.0	60.6	33.6	5.9	0.08	983	-

<sup>1</sup>Walthert et al., 2002, Walthert et al., 2003., Fe and Al content (mmolc/kg) determined by NH<sub>4</sub>Cl extraction.

For the 0.2-0.6 depth interval the CEC determined for 0.2-0.4 m was taken, and similarly for the depth interval 0.6-1.0 m the values for 0.6-0.8 m were taken in the case of Othmarsingen, Lausanne Beatenberg and Nationalpark.

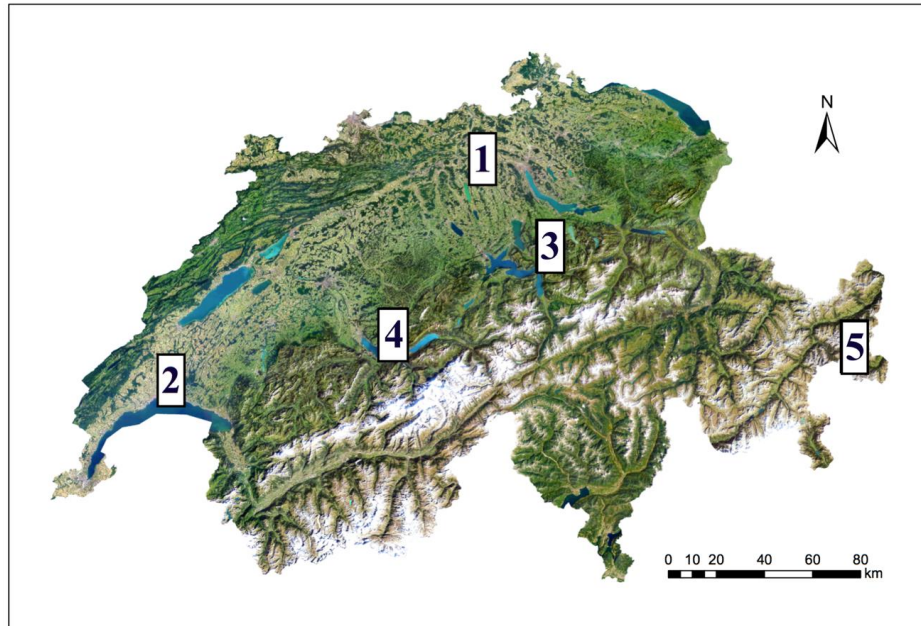
<sup>2</sup>Krause et al., 2013

<sup>3</sup>Diserens et al,1992, CEC determined (mmeq/kg), hydrogen lead and zinc ions were not included, Aluminium content determined by Lakanen method. CEC values for 0.2-0.4 m were extrapolated to 1 m. <sup>4</sup>Xu et al., 2009 <sup>5</sup>Depth to 0.8 m <sup>6</sup>Depth to 0.4 m

**Table 4** Pearson correlations for averaged depth intervals for the top soil (0-20 cm, n=5) and deep soil (20-60 cm, n=5). Significance denoted with ; \*, \*\* or \*\*\* for respectively p-values smaller than 0.1 (marginally significant) 0.05, 0.005 and 0.0005 (significant). Non-significant correlations are indicated by the superscript **ns**. SWP or soil water potential used are the median values at 15 cm for each of these 5 sites (Von Arx et al., 2013). Water-extractable carbon is abbreviated to WEOC. Results indicate that no single climatic or textural factor consistently co-varies with carbon stocks, or turnover time.

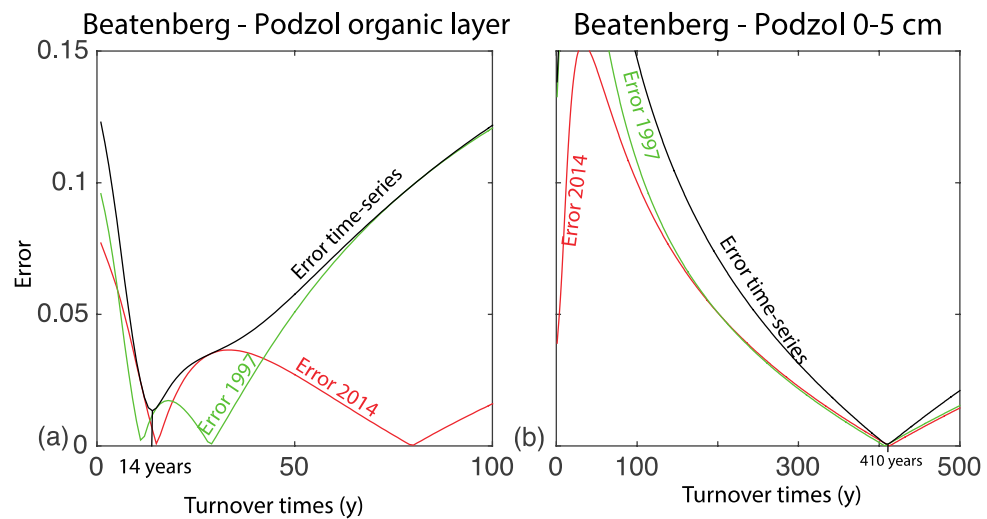
Explaining variable	Stock <sub>0-20 cm</sub>	Turnover time bulk <sub>0-20 cm</sub>	Turnover time WEOC <sub>0-20 cm</sub>	Stock <sub>20-60 cm</sub>	Turnover time <sub>20-60 cm</sub>
MAT	0.17 <sup>ns</sup>	-0.14 <sup>ns</sup>	-0.36 <sup>ns</sup>	0.02 <sup>ns</sup>	0.02 <sup>ns</sup>
MAP	<b>0.96*</b>	0.11 <sup>ns</sup>	0.28 <sup>ns</sup>	<b>0.93*</b>	<b>0.98**</b>
NPP	0.2 <sup>ns</sup>	0.65 <sup>ns</sup>	0.39 <sup>ns</sup>	0.03 <sup>ns</sup>	-0.10 <sup>ns</sup>
Sand	-0.66 <sup>ns</sup>	0.72 <sup>ns</sup>	0.55 <sup>ns</sup>	-0.56 <sup>ns</sup>	-0.70 <sup>ns</sup>
Silt	0.38 <sup>ns</sup>	<b>-0.91*</b>	-0.79 <sup>ns</sup>	0.29 <sup>ns</sup>	-0.47 <sup>ns</sup>
Clay	<b>0.81*</b>	-0.51 <sup>ns</sup>	-0.31 <sup>ns</sup>	0.71 <sup>ns</sup>	0.80 <sup>ns</sup>
CEC	-0.67 <sup>ns</sup>	-0.24 <sup>ns</sup>	0.03 <sup>ns</sup>	0.74 <sup>ns</sup>	<b>0.82*</b>
pH	-0.74 <sup>ns</sup>	-0.47 <sup>ns</sup>	-0.31 <sup>ns</sup>	-0.51 <sup>ns</sup>	-0.46 <sup>ns</sup>
Fe	0.24 <sup>ns</sup>	0.98*	0.97*	0.98*	-0.78 <sup>ns</sup>
Al	0.18 <sup>ns</sup>	-0.16 <sup>ns</sup>	-0.41 <sup>ns</sup>	-0.17 <sup>ns</sup>	-0.17 <sup>ns</sup>
SWP	0.70 <sup>ns</sup>	0.68 <sup>ns</sup>	0.70 <sup>ns</sup>	-	-
Average Grain size	-0.25 <sup>ns</sup>	<b>0.97*</b>	<b>0.88*</b>	0.05 <sup>ns</sup>	-0.16 <sup>ns</sup>

**Figures**

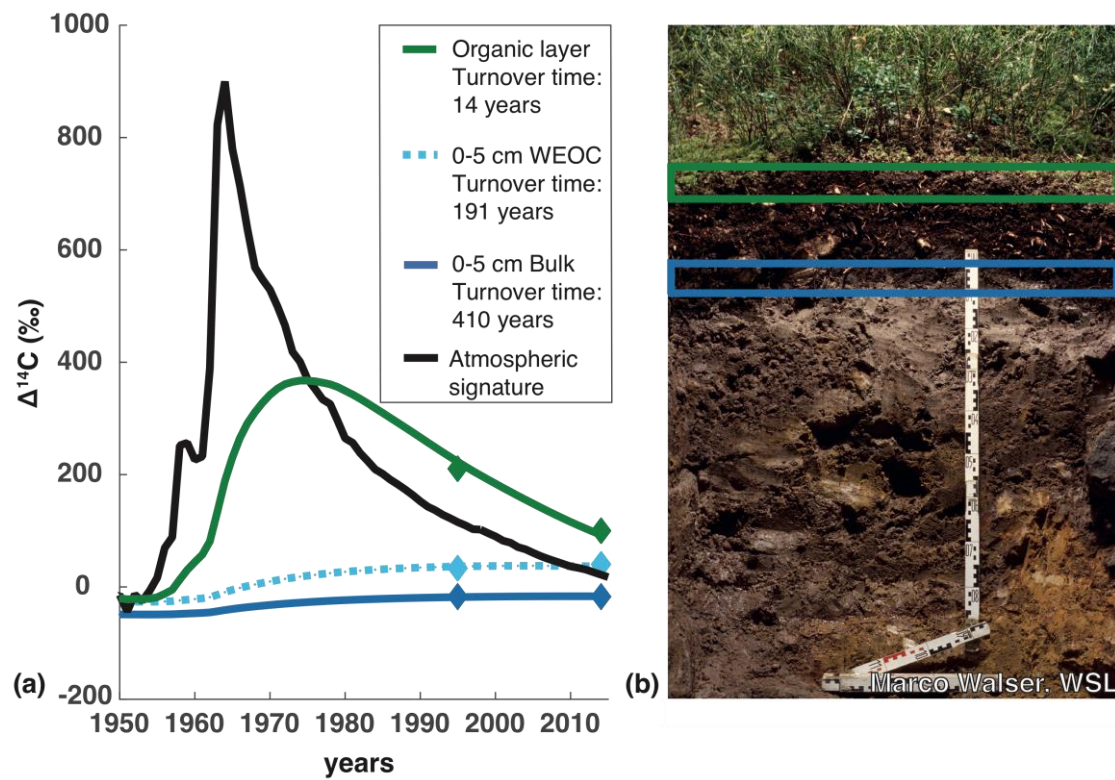


**Figure 1** Sample locations, all of which are part of the Long-term ecosystem research program (LWF) of the Swiss Federal Institute WSL, 1) Othmarsingen, 2) Lausanne, 3) Alptal, 4) Beatenberg and 5) Nationalpark Image made using 2016 swisstopo (JD100042).

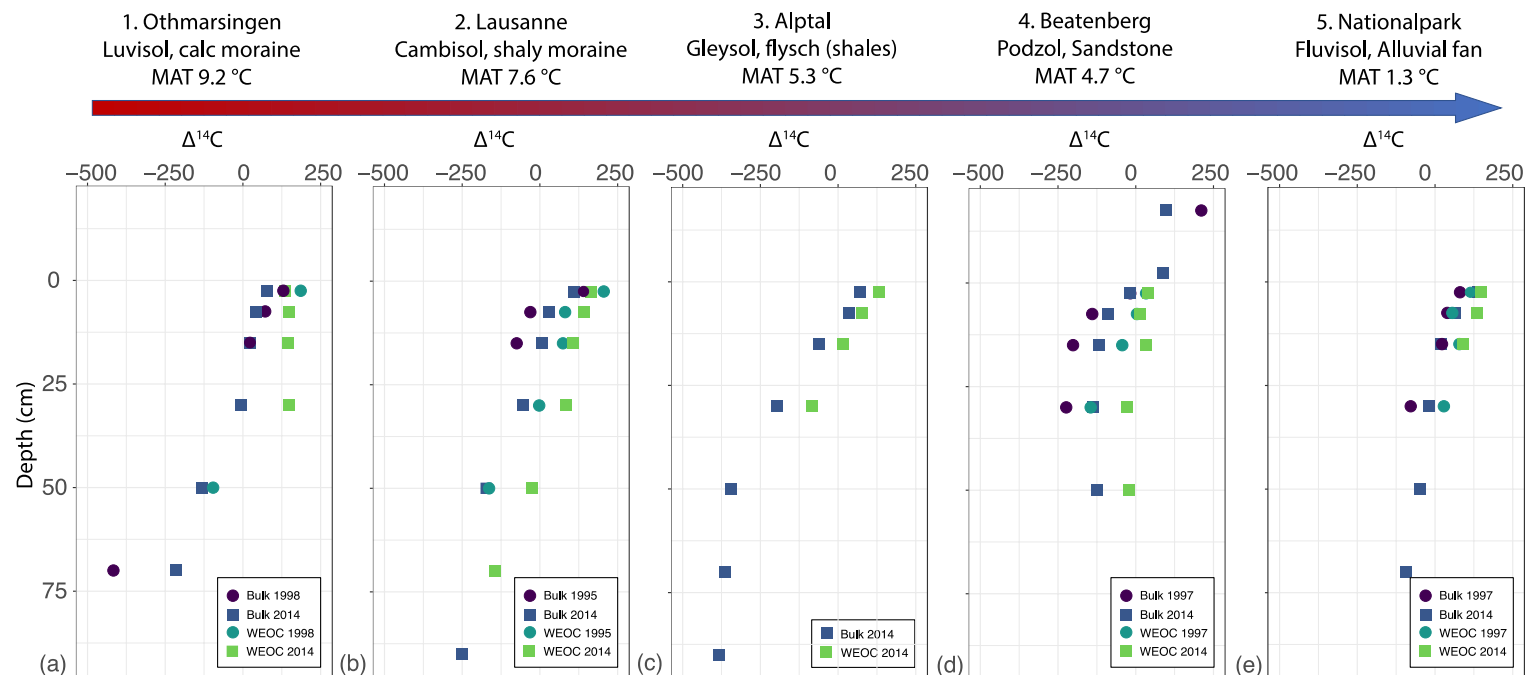




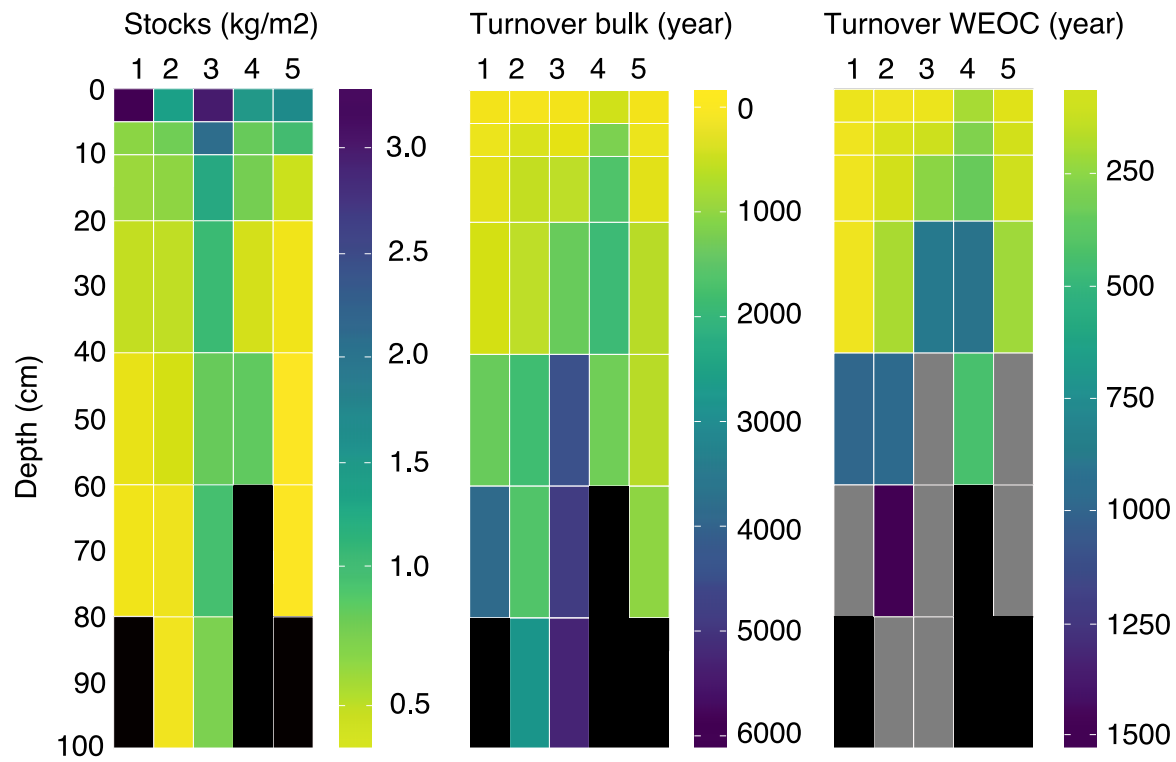
**Figure 2** Numerical optimization of least mean-square error reduction, showing and the reduction of error spread for two soil depths. For the Beatenberg Podzol organic layer (a) the individual  $^{14}\text{C}$  time-points for both 1997 and 2014 both yield two solutions are almost equally likely (i.e. the error nears zero). The combined optimization using both the time-points reveal the likeliest option. For the (b) 0-5 cm layer the single time points only have a single likely solution.

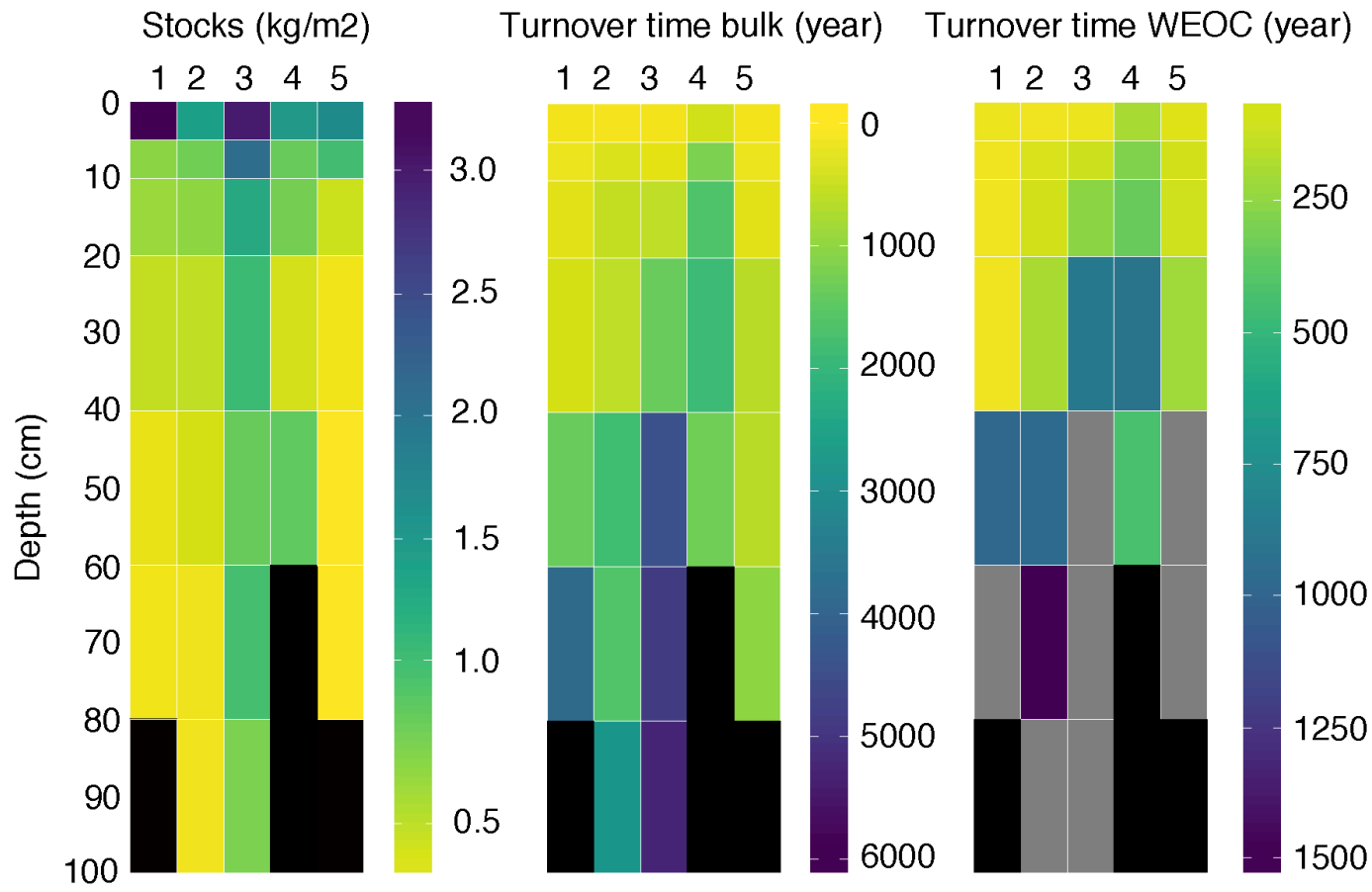


**Figure 3** (a) Time-series soil carbon turnover time in years (y) as determined by numerical modelling for (b) sub-alpine site Podzol Beatenberg. The bulk turnover time in the organic layer is rapid (14 years), followed by the turnover time of the water-extractable organic carbon (WEOC) (191 years) and the bulk turnover time of the soil (410 years) at 0-5 cm depth. Photo soil profile courtesy of Marco Walser, WSL.

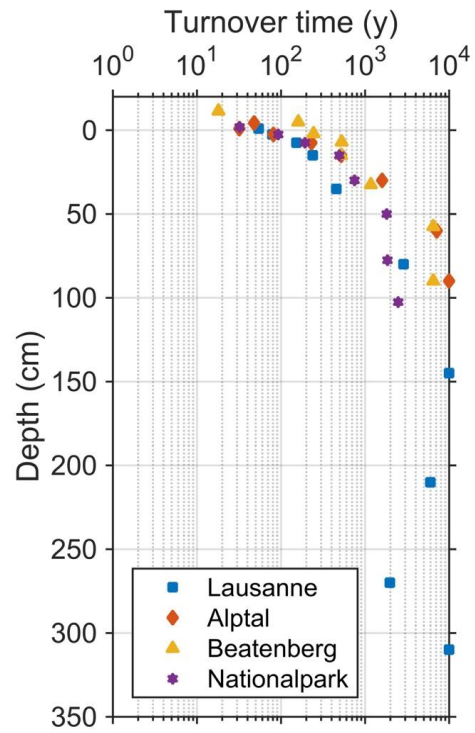


**Figure 4** (a-e) Changes in radiocarbon signature of both bulk soil and WEOC over two decades at four sites on a climatic gradient. For Alptal (Gleysol) (c) only the 2014 time-point was available. For the warmer locations Othmarsingen and Lausanne (Luvisol, Cambisol MAT 9.2-7.6 °C), depletion in bomb-derived radiocarbon occurs in the first five centimeters soil in 2014 as compared to 1995-8. The colder Beatenberg site (Podzol, MAT 4.7 °C) is marked by a clear enrichment of  $^{14}\text{C}$  in the mineral soil in 2014 w.r.t. 1997. At the coldest site Nationalpark (Fluvisol, MAT 1.3 °C) almost all samples taken two decades after the initial sampling show an enrichment in radiocarbon signature. WEOC contains bomb-derived carbon in the topsoil in 2014 at all sites.





**Figure 5** Carbon (a) stocks in the mineral soil kgC/m<sup>2</sup>, (b) turnover time bulk soil in years and (c) turnover time water extractable organic carbon soil in years. Locations are ordered from the warmest to coldest sites i.e. (1) Othmarsingen (Luvisol), (2) Lausanne (Cambisol), (3) Alptal (Gleysol), (4) Beatenberg (Podzol) and (5) Nationalpark (Fluvisol). Grey boxes indicate absence of material, black boxes indicate the occurrence of the C-horizon (poorly consolidated bedrock-derived stony material or bedrock itself).



**Figure 6** Modeled turnover times (y) of single profiles sampled down to the bedrock between 1995 and 1998.  $\Delta^{14}\text{C}$  published in Van der Voort et al. (2016). Results indicate presence of petrogenic (bedrock-derived) carbon as modeled turnover time exceeds soil formation since the end of last ice age (10,000 years) in Lausanne (>100 cm, Cambisol) and Alptal (80-100 cm, Gleysol). For Beatenberg (Podzol) and Nationalpark (Fluvisol), no petrogenic carbon was found.

