

Dear Sébastien,

**Thank you** for going through the referee's comments and for your additional suggestions. You suggested to include soil type in the figure labeling and to clarify the counterintuitive turnover pattern in the Podzol. We have **incorporated** them:

- (1) **In the figures, we have now included both the soil type and location.** We have also taken the opportunity to improve the colour scheme of Figure 4.
- (2) **Thank you for highlighting the Podzol dynamics.** Indeed, you are right. Oxides and hydroxides are crucial for stabilization, so higher C dynamics in the deep soil versus the shallower soil is counterintuitive. We did not sufficiently explain the pedogenetic horizons in the Podzol in order to explain the pattern. The crux is the transition from the elluvial to the illuvial horizon. We have now corrected this.

**In detail:**

The elluvial horizon in Beatenberg extends from 4-5 cm downwards up to ~35 cm. The illuviation leeches the iron, aluminium out of the soil layer. Deeper, in the illuviation horizon (~35-60 cm), there is a redeposition of iron and aluminium which stabilizes the organic matter.

The counterintuitive pattern that you correctly noted is that in the shallower 20-40 cm layer - which encompasses the elluvial horizon – the turnover is slower than the deeper (40-60) layer - which encompasses the illuvial soil horizon. The reason the counter-intuitive pattern occurs is that elluvial horizon (mostly represented in the 20-40 layer) there are lower amounts of oxides and hydroxides as compared to the deeper layers. In the deeper layer (40-60), the organic matter can form organo-mineral complexes again and the carbon dynamics are faster.

Thank you very much again, we look forward to the feedback of the referees,

Tessa

# 1 Dynamics of deep soil carbon – insights from <sup>14</sup>C time-series across a 2 climatic gradient

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13  
14 **Abstract.** Quantitative constraints on soil organic matter (SOM) dynamics are essential for comprehensive  
15 understanding of the terrestrial carbon cycle. Deep soil carbon is of particular interest, as it represents large  
16 stocks and its turnover rates remain highly uncertain. In this study, SOM dynamics in both the top and deep soil  
17 across a climatic (average temperature ~1-9 °C) gradient are determined using time-series (~20 years) <sup>14</sup>C data  
18 from bulk soil and water-extractable organic carbon (WEOC). Analytical measurements reveal enrichment of  
19 bomb-derived radiocarbon in the deep soil layers on the bulk level during the last two decades. The WEOC pool  
20 is strongly enriched in bomb-derived carbon, indicating that it is a dynamic pool. Turnover time estimates of  
21 both the bulk and WEOC pool show that the latter cycles up to a magnitude faster than the former. The presence  
22 of bomb-derived carbon in the deep soil, as well as the rapidly turning WEOC pool across the climatic gradient  
23 implies that there likely is a dynamic component of carbon in the deep soil. Precipitation and bedrock type  
24 appear to exert a stronger influence on soil C turnover and stocks as compared to temperature.

## 25 26 1 Introduction

27 Within the broad societal challenges accompanying climate and land use change, a better understanding of the  
28 drivers of turnover of carbon in the largest terrestrial reservoir of organic carbon, as constituted by soil organic  
29 matter (SOM), is essential (Batjes, 1996; Davidson and Janssens, 2006; Doetterl et al., 2015; Prietzel et al.,  
30 2016). Terrestrial carbon turnover remains one of the largest uncertainties in climate model predictions  
31 (Carvalhais et al., 2014; He et al., 2016). At present, there is no consensus on the net effect that climate and land  
32 use change will have on SOM stocks (Crowther et al., 2016; Gosheva et al., 2017; Melillo et al., 2002; Schimel  
33 et al., 2001; Trumbore and Czimczik, 2008). Deep soil carbon is of particular interest because of its large stocks  
34 (Jobbagy and Jackson, 2000; Balesdent et al., 2018; Rumpel and Kogel-Knabner, 2011) and perceived stability.  
35 The stability is indicated by low <sup>14</sup>C content (Rethemeyer et al., 2005; Schrumpp et al., 2013; van der Voort et  
36 al., 2016) and low microbial activity (Fierer et al., 2003). Despite its importance, deep soil carbon has been  
37 sparsely studied and remains poorly understood (Angst et al., 2016; Mathieu et al., 2016; Rumpel and Kogel-  
38 Knabner, 2011). The inherent complexity of SOM and the multitude of drivers controlling its stability further  
39 impedes the understanding of this globally significant carbon pool (Schmidt et al., 2011). In this framework,  
40 there is a particular interest in the portion of soil carbon that could be most vulnerable to change, especially in  
41 colder climates (Crowther et al., 2016). Water-extractable organic carbon (WEOC) is seen as a dynamic and  
42 potentially vulnerable carbon pool in the soil (Hagedorn et al., 2004; Lechleitner et al., 2016). Radiocarbon  
43 (<sup>14</sup>C) can be a powerful tool to determine the dynamics of carbon turnover over decadal to millennial timescales

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

59

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

66

## 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  
70 encompass large climatic (mean annual temperature (MAT) 1.3-9.2°C, mean annual precipitation (MAP) 864-  
71 2126 mm  $\text{m}^{-2}\text{y}^{-1}$ ) and geological gradients (Table 1). The sites are part of the Long-term Forest Ecosystem  
72 Research program (LWF) at the Swiss Federal Institute for Forest, Snow and Landscape Research, WSL  
73 (Schaub et al., 2011; Etzold et al., 2014). The soils of these sites were sampled between 1995 and 1998  
74 (Walthert et al., 2002, 2003) and were re-sampled following the same sampling strategy in 2014 with the aim to  
75 minimize noise caused by small-scale soil heterogeneity. In both instances sixteen samples were taken on a  
76 regular grid on the identical 43 by 43 meters (~1600  $\text{m}^2$ ) plot (Fig. 1; see Van der Voort et al., 2016 for further  
77 details). For the archived samples taken between 1995 and 1998, mineral soil samples down to 40 cm depth  
78 (intervals of 0-5, 5-10, 10-20 and 20-40 cm) were taken on an area of 0.5 by 0.5 m (0.25  $\text{m}^2$ ). For samples >40  
79 cm (intervals of 40-60, 60-80 and 80-100 cm), corers were used to acquire samples ( $n=5$  in every pit, area  
80  $\sim 2.8 \times 10^{-3} \text{ m}^2$ ). The organic layer was sampled by use of a metal frame (30×30 cm). The samples were dried at  
81 35-40°C, sieved to remove coarse material (2 mm), and stored in hard plastic containers under controlled  
82 climate conditions in the "Pedothek" at WSL (Walthert et al., 2002). For the samples acquired in 2014 the same  
83 sampling strategy was followed, and samples were taken on the exact same plot proximal (~10 m) to the legacy

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

100

## 101 2.2 Climate and soil data

102 Temperature and precipitation data are derived from weather stations close to the study sites that have been  
103 measuring for over two decades, yielding representative estimates of both variables and over the time period  
104 concerned in this study (Etzold et al., 2014). The pH values for all sites and concerned depth intervals were  
105 acquired during the initial sampling campaign (Walthert et al. 2002). At Alptal, pH values were determined as  
106 described in Xu et al. (2009), values of 10-15 cm were extrapolated to the deeper horizons because of the  
107 uniform nature of the Gley horizon. The Beatenberg Podzol is marked by strong elluviation (~4-35 cm) and  
108 illuviation (~35-60 cm) (Walthert et al., 2003). Exchangeable cations were extracted (in triplicate) from the 2-  
109 mm-sieved soil in an unbuffered solution of 1 M  $\text{NH}_4\text{Cl}$  for 1 hour on an end-over-end shaker using a soil-to-  
110 extract ratio of 1:10. The element concentrations in the extracts were determined by inductively coupled plasma  
111 atomic emission spectroscopy (ICP-AES) (Optima 3000, Perkin-Elmer). Contents of exchangeable protons  
112 were calculated as the difference between the total and the Al-induced exchangeable acidity as determined (in  
113 duplicate) by the KCl method (Thomas, 1982). This method was applied only to soil samples with a pH ( $\text{CaCl}_2$ )  
114 < 6.5. In samples with a higher pH, we assumed the quantities of exchangeable protons were negligible. The  
115 effective cation-exchange capacity (CEC) was calculated by summing up the charge equivalents of  
116 exchangeable Na, K, Mg, Ca, Mn, Al, Fe and H. The base saturation (BS) was defined as the percental fraction  
117 of exchangeable Na, K, Mg, and Ca of the CEC (Walthert et al., 2002, 2013). Net primary production (NPP)  
118 was determined by Etzold et al. (2014) as the sum of carbon fluxes by woody tree growth, foliage, fruit  
119 production and fine root production. Soil texture (sand, silt and clay content) on plot-averaged samples taken in  
120 2014 have been determined using grain size classes for sand, silt and clay respectively of 0.05-2 mm, 0.002-0.05  
121 mm and <0.002 mm according to Klute (1986). The continuous distribution of grain sizes was also determined  
122 after removal of organic matter (350 °C for 12 h) using the Mastersizer 2000 (Malvern Instruments Ltd.). Soil  
123 water potential (SWP) was measured on the same sites as described in Von Arx et al., (2013). In accordance

Comment [TSvdV1]: Dear Sébastien, we included more detail regarding the Beatenberg Podzol soil horizons.

124 with Mathieu et al., (2015), topsoil refers to the mineral soil up to 20 cm depth, and deep soil refers to mineral  
125 soil below 20 cm. Out of the five sites, two are hydromorphic (Gleysol and Podzol in Alptal and Beatenberg  
126 respectively), whilst the others are non-hydromorphic (Luvisol, Cambisol and Fluvisol in Othmarsingen,  
127 Lausanne and Nationalpark respectively).

128

### 129 **2.3 Isotopic ( $^{14}\text{C}$ , $^{13}\text{C}$ ) and compositional (C, N) analysis**

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

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

141 For the  $^{14}\text{C}$  measurements of the bulk soil samples were first graphitised using an EA-AGE (elemental analyser-  
142 automated graphitization equipment, Ionplus AG) system at the Laboratory of Ion Beam Physics at ETH Zürich  
143 (Wacker et al., 2009). Graphite samples were measured on a MICADAS (MInitirised radioCARbon DAting  
144 System, Ionplus AG) also at the Laboratory of Ion Beam Physics, ETH Zürich (Wacker et al., 2010). For three  
145 samples (Alptal depth intervals 40-60, 60-80 and 80-100 cm) the  $^{14}\text{C}$  signature was directly measured as  $\text{CO}_2$   
146 gas using the recently developed online elemental analyzer (EA) - stable isotope ratio mass spectrometers  
147 (IRMS)–AMS system et ETH Zürich (McIntyre et al., 2016). Oxalic acid (NIST SRM 4990C) was used as the  
148 normalising standard. Phthalic anhydride and in-house anthracite coal were used as blank. Two in-house soil  
149 standards (Alptal soil 0-5 cm, Othmarsingen soil 0-5 cm) were used as secondary standards. For the WEOC,  
150 samples were converted to  $\text{CO}_2$  by Wet Chemical Oxidation (WCO) (Lang et al., 2016) and run on the AMS  
151 using a Gas Ion Source (GIS) interface (Ionplus). To correct for contamination, a range of modern standards  
152 (sucrose, Sigma,  $\delta^{13}\text{C} = -12.4 \text{ ‰ VPDB}$ ,  $F^{14}\text{C} = 1.053 \pm 0.003$ ) and fossil standards (phthalic acid, Sigma,  
153  $\delta^{13}\text{C} = -33.6 \text{ ‰ VPDB}$ ,  $F^{14}\text{C} < 0.0025$ ) were used (Lechleitner et al., 2016).

154

155

156 **2.4 Numerical optimization to find carbon turnover and size of the dynamic pool**

157 **2.4.1 Turnover based on a single <sup>14</sup>C measurement**

158 The <sup>14</sup>C signature of a sample can be used to estimate turnover time of a carbon pool (Torn et al., 2009).

159 
$$R_{sample,t} = k \times R_{atm,t} + (1 - k - \lambda) \times R_{sample(t-1)} \quad (1)$$

160 
$$R_{sample,t} = \frac{\Delta^{14}C_{sample}}{1000} + 1 \quad (2)$$

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

166 For the turnover time estimation, we assumed the system to be in steady state over the modeled period ( $\sim 1 \times 10^4$   
167 years, indicating soil formation since the last glacial retreat (Ivy-Ochs et al., 2009)), hence accounting both for  
168 radioactive decay and incorporation of the bomb-testing derived material produced in the 1950's and 1960's  
169 (Eq. 1) (Herold et al., 2014; Torn et al., 2009). We assumed an initial fraction modern ( $F_m$ ) of <sup>14</sup>C value of 1 at  
170 10000 B.C.. For the period after 1900 atmospheric fraction modern ( $F_m$ ) values of the Northern Hemisphere  
171 were used (Hua et al., 2013). This equation could be solved in Excel with manual iterations (e.g. Herold et al.,  
172 2014), or alternatively a numerical optimization can be used to find the best fit automatically. In this paper, we  
173 used a numerical optimization constructed in MATLAB version 2015a (The MathWorks, Inc., Natick,  
174 Massachusetts, United States) to find the best fit. The numerical optimization is exhaustive, meaning that every  
175 single turnover value from 1 to 10.000 years with an interval of 0.1 year is tested. The error is defined as the  
176 difference between the fitted value of  $R$  and the measured value (Eq. 3). The turnover value with the lowest  
177 error is then automatically selected.

178 
$$Error_{single\ timepoint} = |R_{calculated} - R_{measured}| \quad (3)$$

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

182

183 **2.4.2 Turnover based on two <sup>14</sup>C measurements**

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

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

$$193 \quad Error_{two\ timepoints} = \sqrt{|R_{calculated} - R_{measured}|_{time\ point\ 1}^2 + |R_{calculated} - R_{measured}|_{time\ point\ 1}^2} \quad (4)$$

#### 194 2.4.3 Vegetation-induced lag

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

#### 200 2.4.4 Turnover and size vulnerable pool based on two-pool model

201 As SOM is complex and composed of a continuum of pools with various ages (Schrumpf and Kaiser, 2015) and  
 202 there is data available from two SOM pools, the  $^{14}C$  time-series data can be leveraged to create a two-pool  
 203 model. The following assumptions were made: First, both pools (slow & fast) make up the total carbon pool  
 204 (Eq. 5). Secondly, the total turnover of the bulk soil is made up out of the “dynamic” fraction turnover  
 205 multiplied by “dynamic” fraction pool size and the “slow” pool turnover multiplied by “slow” pool size (Eq. 6).  
 206 Furthermore, we assume that the signature of the sample (the time-series bulk data) is determined by the rate of  
 207 incorporation of the material (atmospheric signal) and the loss of carbon the two pools (Eq. 7). Lastly, we  
 208 assume that the radiocarbon signal of the WEOC pool is representative for a dynamic pool, as it could be  
 209 representative for a larger component of rapidly turning over carbon, even in the deep soil (Baisden and Parfitt,  
 210 2007; Koarashi et al., 2012). The turnover rate of the slow pool was set between 100 and 10.000 years, with a  
 211 time-step of 10 years. The size of the dynamic pool was set to be between 0 and 0.5, with a size-step of 0.01.

212

$$213 \quad 1 = F_1 + F_2 \quad (5)$$

$$214 \quad k_{total} = (F_1/k_1 + F_2/k_2)^{-1} \quad (6)$$

$$215 \quad R_{sample,t} = k_{total} \times R_{atm,t} + F_1[(1 - k_1 - \lambda) \times R_{sample(t-1)}] + F_2(1 - k_2 - \lambda) \times R_{sample(t-1)} \quad (7)$$

216 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  
 217 the inverse of the turnover time of the dynamic or WEOC as determined using the numerical optimisation of Eq.  
 218 1.4. The  $k_2$  is the inverse of the turnover time of the slow pool. The calculation of the error term becomes for  
 219 complex because it needs to be recalculated for each unique combination of pool-size distribution (Eq. 5) and  
 220 turnover time (inverse of  $k$ , Eq. 6). Therefore, the error space changes from column vector to a two-dimensional

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

223 
$$Error_{k_2, F_1} = \sqrt{|R_{calculated} - R_{measured}|_{time\ point\ 1}^2 + |R_{calculated} - R_{measured}|_{time\ point\ 2}^2} \quad (8)$$

224 
$$Error = Min(Error_{k_2, F_1}) \quad (9)$$

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

230

### 231 **3 Results**

#### 232 **3.1 Changes of radiocarbon signatures over time**

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

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

251

#### 252 **3.2 Carbon turnover patterns**

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



258 have relatively similar values with decadal turnover times for the 0-5 cm layer, increasing to an order of  
259 centuries down to 20 cm depth, and to millenia in deeper soil layers (~980 to ~3940 years at 0.6 to 1 m depth)  
260 (Fig. 5). In contrast, the hydromorphic soils are marked by turnover times that are up to an order of magnitude  
261 larger, from centennial in top soil to (multi-) millennial in deeper soils. At the Beatenberg Podzol, turnover time  
262 in the shallow layer which overlaps with the elluvial horizon is slower (20-40 cm, ~1900 y) than the deepest  
263 layer (40-60 cm, ~1300 y), which overlaps with the illuvial horizon (Figure 5, SI Table 5).

Comment [TSvdV2]: As suggested we clarified the turnover pattern in the Podzol

264 Carbon stocks also show distinct difference between drained and hydromorphic soils with greater stock  
265 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,  
266 Lausanne and Nationalpark, Fig. 5, Table 3)).

267 The turnover times of the WEOC mimic the trends in the bulk soil but are up to an order of magnitude  
268 faster. Considering WEOC turnover in the non-hydromorphic soils only, there is a slight increase in WEOC  
269 turnover with decreasing site temperature, but the trend is not significant (SI Table 4). The modeled estimate for  
270 dynamic fraction is variable at the surface but decreases towards the lower top soil (from ~0.2 at 0-5 cm to  
271 ~0.01 at 10-20 cm in Othmarsingen). In the deep soil, the model indicates there could also be a non-negligible  
272 proportion of dynamic carbon (e.g. 0.10-0.23 at 20-40 cm). The residual errors associated to the error reduction  
273 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  
274 Figure 1).

275  
276

### 277 3.3 Pre-glacial carbon in deep soil profiles

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

282

### 283 3.4 Environmental drivers of carbon dynamics

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

291

## 292 4 Discussion

### 293 4.1 Dynamic deep soil carbon

#### 294 4.1.1 Rapid shifts in <sup>14</sup>C abundance reflect dynamic deep carbon

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

298 consistently more enriched in bomb-derived carbon than the bulk soil. Near-atmospheric signature WEO<sup>14</sup>C  
299 pervades up to 40 or even 60 cm depth. Hagedorn et al., (2004) also found WEOC to be a highly dynamic pool  
300 using <sup>13</sup>C tracer experiments in forest soils.

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

308

#### 309 4.1.2 Carbon dynamics reflect soil-specific characteristics at depth

310 Bulk carbon turnover for the top and deeper soil fall in the range of prior observations and models, although the  
311 data for the latter category is sparse (Scharpenseel and Becker-Heidelmann, 1989; Paul et al., 1997; Schmidt et  
312 al., 2011; Mills et al., 2013; Braakhekke et al., 2014). The carbon turnover is related to soil-specific  
313 characteristics. The slower turnover of hydromorphic as compared to non-hydromorphic soils is likely due to  
314 increased waterlogging and limited aerobicity (Hagedorn et al., 2001) which is conducive to slow turnover and  
315 enhanced carbon accumulation. The WEOC turns over up to an order of magnitude faster than the bulk and  
316 mirrors these trends, indicating that it indeed is a more dynamic pool (Hagedorn et al., 2004; Lechleitner et al.,  
317 2016). Results also reflect known horizon-specific dynamics for certain soil types, particularly in the deep soil.

318 [The hydromorphic Podzol at Beatenberg shows specific pedogenetic features such as an illuviation layer with an  
319 enrichment in humus and iron in the deeper soil (Walthert et al., 2003) where turnover of bulk and WEOC is  
320 faster and stocks are higher as compared to the elluvial layer above (Fig. 5). This is likely due to the input of  
321 younger carbon via leaching of dissolved organic carbon. The non-hydromorphic Luvisols are marked by an  
322 enrichment of clay in the deeper soil, which can enhance carbon stabilization (Lutzow et al., 2006). This also  
323 reflected in the turnover time of the 60-80 cm layer in the Othmarsingen Luvisol – in this clay-enriched depth  
324 interval (Walthert et al., 2003), turnover is relatively slow as compared to the other (colder) non-hydromorphic  
325 soils (Fig. 5). These patterns are consistent with findings by Mathieu et al., (2015) that the important role of soil  
326 pedology on deep soil carbon dynamics.

327

#### 328 4.1.3 Dynamic carbon at depth & implications for carbon transport

329 The analytical <sup>14</sup>C data as well as turnover time estimates indicate that there is likely a dynamic portion of  
330 carbon in the deep soil. The estimated size of the dynamic pool can be large, even at greater depth than it was  
331 observed by other <sup>14</sup>C time-series (Richter and Markewitz, 2001; Baisden and Parfitt, 2007; Koarashi et al.,  
332 2012). The two-pool modelling indicates that the size of dynamic pool in the deep soil can be upwards of ~10%.  
333 A deep dynamic pool is consistent with findings of a <sup>13</sup>C tracer experiment by Hagedorn et al., (2001) that  
334 shows with that relatively young (<4 years) carbon can be rapidly incorporated in the top soil (20% new C at 0-  
335 20 cm depth) but also in the deep soil (50 cm), and findings by Balesdent et al., (2018) which estimate that up to  
336 21% of the carbon between 30-100 cm is younger than 50 years. Rumpel and Kögel-Knabner (2011) have  
337 highlighted the importance of the poorly understood deep soil carbon stocks and a significant dynamic pool in

Comment [TSvdV3]: We have further clarified the Podzol turnover pattern here

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

347

#### 348 **4.2 Contribution of petrogenic carbon**

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

360 Other studies analyzing soils have observed the significant presence of petrogenic (geogenic in soil  
361 science terminology) in loess-based soils (Helfrich et al., 2007; Paul et al., 2001). Our results suggest that pre-  
362 glacial carbon may comprise a dominant component of deep soil organic matter in several cases, resulting in an  
363 apparent increase in the average age (and decrease in turnover) of carbon in these soils. Hemingway et al.,  
364 (2018) have highlighted that fossil carbon oxidized in soils can lead to significant additional  $\text{CO}_2$  emissions.  
365 Therefore, the potential of soils to ‘activate’ fossil petrogenic carbon should be considered when evaluating the  
366 soil carbon sequestration potential.

367

#### 368 **4.3 Controls on carbon dynamics and cycling**

369 In order to examine the effects of potential drivers on soil C turnover and stocks, we explore correlations  
370 between a number of available factors which have previously been proposed, such as texture, geology,  
371 precipitation, temperature and soil moisture (Doetterl et al., 2015; McFarlane et al., 2013; Nussbaum et al.,  
372 2014; Seneviratne et al., 2010; van der Voort et al., 2016).

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

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

398

#### 399 **4.4 Modular robust numerical optimization**

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

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

415

#### 416 **5 Conclusion**

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

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

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

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

434

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450

451 **References**

- 452 Angst, G., John, S., Mueller, C. W., Kögel-Knabner, I. and Rethemeyer, J.: Tracing the sources and spatial  
453 distribution of organic carbon in subsoils using a multi-biomarker approach, *Sci. Rep.*, 6(1), 29478,  
454 doi:10.1038/srep29478, 2016.
- 455 Von Arx, G., Graf Pannatier, E., Thimonier, A. and Rebetez, M.: Microclimate in forests with varying leaf area  
456 index and soil moisture: Potential implications for seedling establishment in a changing climate, *J. Ecol.*, 101(5),  
457 1201–1213, doi:10.1111/1365-2745.12121, 2013.
- 458 Baisden, W. T. and Parfitt, R. L.: Bomb <sup>14</sup>C enrichment indicates decadal C pool in deep soil?,  
459 *Biogeochemistry*, 85(1), 59–68, doi:10.1007/s10533-007-9101-7, 2007.
- 460 Baisden, W. T., Parfitt, R. L., Ross, C., Schipper, L. A. and Canessa, S.: Evaluating 50 years of time-series soil  
461 radiocarbon data : towards routine calculation of robust C residence times, *Biogeochemistry*, 112, 129–137,  
462 doi:10.1007/s10533-011-9675-y, 2013.
- 463 Batjes, N. H.: Total carbon and nitrogen in the soils of the world, *Eur. J. Soil Sci.*, 47(June), 151–163, 1996.
- 464 Balesdent, J., Basile-Doelsch, I., Chadoeuf, J., Cornu, S., Derrien, D., Fekiacova, Z. and Hatté, C.: Atmosphere–  
465 soil carbon transfer as a function of soil depth, *Nature*, 559(7715), 599–602, doi:10.1038/s41586-018-0328-3,  
466 2018.
- 467 Braakhekke, M. C., Beer, C., Schrupf, M., Ekici, A., Ahrens, B., Hoosbeek, M. R., Kruijt, B., Kabat, P. and  
468 Reichstein, M.: The use of radiocarbon to constrain current and future soil organic matter turnover and transport  
469 in a temperate forest, *J. Geophys. Res. Biogeosciences*, 119(3), 372–391, doi:10.1002/2013JG002420, 2014.
- 470 Carvalhais, N., Forkel, M., Khomik, M., Bellarby, J., Jung, M., Migliavacca, M., Mu, M., Saatchi, S., Santoro,  
471 M., Thurner, M., Weber, U., Ahrens, B., Beer, C., Cescatti, A., Randerson, J. T., Reichstein, M., Mu, M.,  
472 Saatchi, S., Santoro, M., Thurner, M., Weber, U., Ahrens, B., Beer, C., Cescatti, A., Randerson, J. T.,  
473 Reichstein, M., Mu, M., Saatchi, S., Santoro, M., Thurner, M., Weber, U., Ahrens, B., Beer, C., Cescatti, A.,  
474 Randerson, J. T. and Reichstein, M.: Global covariation of carbon turnover times with climate in terrestrial  
475 ecosystems, *Nature*, 514(7521), 213–217, doi:10.1038/nature13731, 2014.
- 476 Crowther, T., Todd-Brown, K., Rowe, C., Wieder, W., Carey, J., Machmuller, M., Snoek, L., Fang, S., Zhou,  
477 G., Allison, S., Blair, J., Bridgman, S., Burton, A., Carrillo, Y., Reich, P., Clark, J., Classen, A., Dijkstra, F.,  
478 Elberling, B., Emmett, B., Estiarte, M., Frey, S., Guo, J., Harte, J., Jiang, L., Johnson, B., Kröel-Dulay, G.,  
479 Larsen, K., Laudon, H., Lavalley, J., Luo, Y., Lupascu, M., Ma, L., Marhan, S., Michelsen, A., Mohan, J., Niu,  
480 S., Pendall, E., Penuelas, J., Pfeifer-Meister, L., Poll, C., Reinsch, S., Reynolds, L., Schmidh, I., Sistla, S.,  
481 Sokol, N., Templer, P., Treseder, K., Welker, J. and Bradford, M.: Quantifying global soil C losses in response  
482 to warming, *Nature*, 540(1), 104–108, doi:10.1038/nature20150, 2016.
- 483 Davidson, E. A. and Janssens, I. A.: Temperature sensitivity of soil carbon decomposition and feedbacks to  
484 climate change., *Nature*, 440(7081), 165–73 [online] Available from:  
485 <http://www.ncbi.nlm.nih.gov/pubmed/16525463> (Accessed 28 May 2014), 2006.

486 Davidson, E. A., Trumbore, S. E. and Amundson, R.: Soil warming and organic carbon content., *Nature*,  
487 408(December), 789–790, doi:10.1038/35048672, 2000.

488 Doetterl, S., Stevens, A., Six, J., Merckx, R., Oost, K. Van, Pinto, M. C., Casanova-katny, A., Muñoz, C.,  
489 Boudin, M., Venegas, E. Z. and Boeckx, P.: Soil carbon storage controlled by interactions between  
490 geochemistry and climate, *Nat. Geosci.*, 8(August), 1–4, doi:10.1038/NGEO2516, 2015.

491 Etzold, S., Waldner, P., Thimonier, A., Schmitt, M. and Dobbertin, M.: Tree growth in Swiss forests between  
492 1995 and 2010 in relation to climate and stand conditions: Recent disturbances matter, *For. Ecol. Manage.*, 311,  
493 41–55 [online] Available from: <http://linkinghub.elsevier.com/retrieve/pii/S0378112713003393> (Accessed 3  
494 June 2014), 2014.

495 Feng, X., Simpson, A. J., Wilson, K. P., Williams, D. D. and Simpson, M. J.: Increased cuticular carbon  
496 sequestration and lignin oxidation in response to soil warming, *Nat. Geosci.*, 1(December), 836–839, 2008.

497 Fierer, N., Schimel, J. P. and Holden, P. A.: Variations in microbial community composition through two soil  
498 depth profiles, *Soil Biol. Biochem.*, 35, 167–176, 2003.

499 Fontaine, S., Barot, S., Barré, P., Bdioui, N., Mary, B. and Rumpel, C.: Stability of organic carbon in deep soil  
500 layers controlled by fresh carbon supply., *Nature*, 450, 277–280, 2007.

501 Fröberg, M., Tipping, E., Stendahl, J., Clarke, N. and Bryant, C.: Mean residence time of O horizon carbon  
502 along a climatic gradient in Scandinavia estimated by <sup>14</sup>C measurements of archived soils, *Biogeochemistry*,  
503 104(1–3), 227–236 [online] Available from: <http://link.springer.com/10.1007/s10533-010-9497-3> (Accessed 2  
504 August 2013), 2010.

505 Galy, V., Beyssac, O., France-Lanord, C. and Eglinton, T. I.: Recycling of graphite during Himalayan erosion: a  
506 geological stabilization of carbon in the crust, *Science (80-. )*, 322(November), 943–945,  
507 doi:10.1126/science.1161408, 2008.

508 Giardina, C. P. and Ryan, M. G.: Evidence that decomposition rates of organic carbon in mineral soil do not  
509 vary with temperature, *Nature*, 404(6780), 858–861, doi:10.1038/35009076, 2000.

510 Gosheva, S., Walthert, L., Niklaus, P. A., Zimmermann, S., Gimmi, U. and Hagedorn, F.: Reconstruction of  
511 Historic Forest Cover Changes Indicates Minor Effects on Carbon Stocks in Swiss Forest Soils, *Ecosystems*,  
512 (C), doi:10.1007/s10021-017-0129-9, 2017.

513 Graven, H. D.: Impact of fossil fuel emissions on atmospheric radiocarbon and various applications of  
514 radiocarbon over this century, *Proc. Natl. Acad. Sci.*, (Early Edition), 1–4, doi:10.1073/pnas.1504467112,  
515 2015a.

516 Graven, H. D.: Impact of fossil fuel emissions on atmospheric radiocarbon and various applications of  
517 radiocarbon over this century, *Proc. Natl. Acad. Sci.*, (Early Edition), 1–4, doi:10.1073/pnas.1504467112,  
518 2015b.

519 Hagedorn, F., Bucher, J. B. and Schleppei, P.: Contrasting dynamics of dissolved inorganic and organic nitrogen  
520 in soil and surface waters of forested catchments with Gleysols, *Geoderma*, 100(1–2), 173–192,  
521 doi:10.1016/S0016-7061(00)00085-9, 2001a.

522 Hagedorn, F., Maurer, S., Egli, P., Blaser, P., Bucher, J. B. and Siegw: Carbon sequestration in forest soils :  
523 effects of soil type , atmospheric CO<sub>2</sub> enrichment , and N deposition, *Eur. J. Soil Sci.*, 52(December), 2001b.

524 Hagedorn, F., Saurer, M. and Blaser, P.: A <sup>13</sup>C tracer study to identify the origin of dissolved organic carbon in  
525 forested mineral soils, *Eur. J. Soil Sci.*, 55(1), 91–100 [online] Available from:  
526 <http://doi.wiley.com/10.1046/j.1365-2389.2003.00578.x> (Accessed 26 September 2013), 2004.

527 He, Y., Trumbore, S. E., Torn, M. S., Harden, J. W., Vaughn, L. J. S., Allison, S. D. and Randerson, J. T.:  
528 Radiocarbon constraints imply reduced carbon uptake by soils during the 21st century, *Science (80-. )*,  
529 353(6306), 1419–1424, 2016.

530 Helfrich, M., Flessa, H., Mikutta, R., Dreves, A. and Ludwig, B.: Comparison of chemical fractionation  
531 methods for isolating stable soil organic carbon pools, *Eur. J. Soil Sci.*, 58(6), 1316–1329, doi:10.1111/j.1365-  
532 2389.2007.00926.x, 2007.

533 Hemingway, J.: Microbial oxidation of lithospheric organic carbon in rapidly eroding tropical mountain soils,  
534 *Science (80-. )*, (April), doi:10.1126/science.aao6463, 2018.

535 Herold, N., Schöning, I., Michalzik, B., Trumbore, S. E. and Schrumpf, M.: Controls on soil carbon storage and  
536 turnover in German landscapes, *Biogeochemistry*, 119(1–3), 435–451, doi:x, 2014.

537 Hua, Q., Barbetti, M. and Rakowski, A. Z.: Atmospheric radiocarbon for the period 1950–2010, *Radiocarbon*,  
538 55(4), 2059–2072, 2013.

539 Ivy-Ochs, S., Kerschner, H., Maisch, M., Christl, M., Kubik, P. W. and Schluchter, C.: Latest Pleistocene and  
540 Holocene glacier variations in the European Alps, *Quat. Sci. Rev.*, 28(21–22), 2137–2149,  
541 doi:10.1016/j.quascirev.2009.03.009, 2009.

542 Jobbagy, E. G. and Jackson, R. .: Ther vertical distribution of soil organic carbon an its relation to climate and  
543 vegetation, *Ecol. Appl.*, 10(April), 423–436, 2000.

544 Kaiser, K. and Kalbitz, K.: Cycling downwards - dissolved organic matter in soils, *Soil Biol. Biochem.*, 52, 29–  
545 32, doi:10.1016/j.soilbio.2012.04.002, 2012.

546 Keiluweit, M., Bougoure, J. J., Nico, P. S., Pett-Ridge, J., Weber, P. K. and Kleber, M.: Mineral protection of  
547 soil carbon counteracted by root exudates, *Nat. Clim. Chang.*, 5(6), doi:10.1038/nclimate2580, 2015.

548 Klute, A.: *Methods of soil analysis, Part 1: Physical and Mineralogical Methods*, 2nd ed., Agronomy  
549 Monograph No 9, Madison WI., 1986.

550 Koarashi, J., Hockaday, W. C., Masiello, C. a. and Trumbore, S. E.: Dynamics of decadal cycling carbon in  
551 subsurface soils, *J. Geophys. Res. Biogeosciences*, 117(3), 1–13, doi:10.1029/2012JG002034, 2012.

552 Komada, T., Anderson, M. R. and Dorfmeier, C. L.: Carbonate removal from coastal sediments for the  
553 determination of organic carbon and its isotopic signatures ,  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  : comparison of fumigation and  
554 direct acidification by hydrochloric acid, *Limnol. Oceanogr. Methods*, (6), 254–262, 2008.

555 Lang, S. Q., McIntyre, C. P., Bernasconi, S. M., Früh-Green, G. L., Voss, B. M., Eglinton, T. I. and Wacker, L.:  
556 Rapid  $^{14}\text{C}$  Analysis of Dissolved Organic Carbon in Non-Saline Waters, *Radiocarbon*, 58(3), 1–11,  
557 doi:10.1017/RDC.2016.17, 2016.

558 Lechleitner, F. A., Baldini, J. U. L., Breitenbach, S. F. M., Fohlmeister, J., McIntyre, C., Goswami, B.,  
559 Jamieson, R. A., van der Voort, T. S., Pruber, K., Marwan, N., Culleton, B. J., Kennett, D. J., Asmerom, Y.,  
560 Polyak, V. and Eglinton, T. I.: Hydrological and climatological controls on radiocarbon concentrations in a  
561 tropical stalagmite, *Geochim. Cosmochim. Acta*, doi:10.1016/j.gca.2016.08.039, 2016.

562 Lutzow, M. V., Kogel-Knabner, I., Ekschmitt, K., Matzner, E., Guggenberger, G., Marschner, B. and Flessa, H.:  
563 Stabilization of organic matter in temperate soils: mechanisms and their relevance under different soil  
564 conditions - a review, *Eur. J. Soil Sci.*, 57, 426–445 [online] Available from:  
565 <http://doi.wiley.com/10.1111/j.1365-2389.2006.00809.x>, 2006.

566 McFarlane, K. J., Torn, M. S., Hanson, P. J., Porras, R. C., Swanston, C. W., Callahan, M. A. and Guilderson,  
567 T. P.: Comparison of soil organic matter dynamics at five temperate deciduous forests with physical  
568 fractionation and radiocarbon measurements, *Biogeochemistry*, 112(1–3), 457–476, doi:10.1007/s10533-012-  
569 9740-1, 2013.

570 Manzoni, S., Katul, G. G. and Porporato, A.: Analysis of soil carbon transit times and age distributions using  
571 network theories, *J. Geophys. Res. Biogeosciences*, 114(4), 1–14, doi:10.1029/2009JG001070, 2009.

572 Mathieu, J. A., Hatté, C., Balesdent, J. and Parent, É.: Deep soil carbon dynamics are driven more by soil type  
573 than by climate: A worldwide meta-analysis of radiocarbon profiles, *Glob. Chang. Biol.*, 21(11), 4278–4292,  
574 doi:10.1111/gcb.13012, 2015.

575 McIntyre, C. P., Wacker, L., Haghjipour, N., Blattmann, T. M., Fahrni, S., Usman, M., Eglinton, T. I. and Synal,  
576 H.-A.: Online  $^{13}\text{C}$  and  $^{14}\text{C}$  Gas Measurements by EA-IRMS-AMS at ETH Zürich, *Radiocarbon*,  
577 002(November 2015), 1–11, doi:10.1017/RDC.2016.68, 2016.

578 Melillo, J. M., Steudler, P. a, Aber, J. D., Newkirk, K., Lux, H., Bowles, F. P., Catricala, C., Magill, A., Ahrens,  
579 T. and Morrisseau, S.: Soil warming and carbon-cycle feedbacks to the climate system., *Science*, 298(5601),  
580 2173–6 [online] Available from: <http://www.ncbi.nlm.nih.gov/pubmed/12481133> (Accessed 21 January 2014),  
581 2002.

582 Mills, R. T. E., Tipping, E., Bryant, C. L. and Emmett, B. a.: Long-term organic carbon turnover rates in natural  
583 and semi-natural topsoils, *Biogeochemistry*, 118(1), 257–272 [online] Available from:  
584 <http://link.springer.com/10.1007/s10533-013-9928-z> (Accessed 18 December 2013a), 2013.

585 Mills, R. T. E., Tipping, E., Bryant, C. L. and Emmett, B. a.: Long-term organic carbon turnover rates in natural  
586 and semi-natural topsoils, *Biogeochemistry*, 118(1), 257–272 [online] Available from:  
587 <http://link.springer.com/10.1007/s10533-013-9928-z>, 2013b.

588 Nussbaum, M., Papritz, A., Baltensweiler, A. and Walthert, L.: Estimating soil organic carbon stocks of Swiss  
589 forest soils by robust external-drift kriging, *Geosci. Model Dev.*, 7(3), 1197–1210, doi:10.5194/gmd-7-1197-  
590 2014, 2014.

591 Paul, E. A., Collins, H. P. and Leavitt, S. W.: Dynamics of resistant soil carbon of midwestern agricultural soils  
592 measured by naturally occurring  $^{14}\text{C}$  abundance, *Geoderma*, 104(3–4), 239–256, doi:10.1016/S0016-  
593 7061(01)00083-0, 2001.

594 Paul, E. A., Follett, R. F., Leavitt, W. S., Halvorson, A., Petersen, G. A. and Lyon, D. J.: Radiocarbon Dating  
595 for Determination of Soil Organic Matter Pool Sizes and Dynamics, *Soil Sci. Soc. Am. J.*, 61(4), 1058–1067,  
596 1997.

597 Prietzel, J., Zimmermann, L., Schubert, A. and Christophel, D.: Organic matter losses in German Alps forest  
598 soils since the 1970s most likely caused by warming, *Nat. Geosci.*, 9(July), doi:10.1038/ngeo2732, 2016.

599 Prior, C. A., Baisden, W. T., Bruhn, F. and Neff, J. C.: Using a soil chronosequence to identify soil fractions for  
600 understanding and modeling soil carbon dynamics in New Zealand, *Radiocarbon*, 49(2), 1093–1102, 2007.

601 Rasse, D. P., Rumpel, C. and Dignac, M.-F.: Is soil carbon mostly root carbon? Mechanisms for a specific  
602 stabilisation, *Plant Soil*, 269(1–2), 341–356 [online] Available from: [http://link.springer.com/10.1007/s11104-  
603 004-0907-y](http://link.springer.com/10.1007/s11104-004-0907-y), 2005.

604 Rethemeyer, J., Kramer, C., Gleixner, G., John, B., Yamashita, T., Flessa, H., Andersen, N., Nadeau, M. and  
605 Grootes, P. M.: Transformation of organic matter in agricultural soils : radiocarbon concentration versus soil



606 depth, *Geoderma*, 128, 94–105, doi:10.1016/j.geoderma.2004.12.017, 2005.

607 Richter, D. D. and Markewitz, D.: *Understanding Soil Change*, Cambridge University Press, Cambridge., 2001.

608 Rumpel, C. and Kogel-Knabner, I.: Deep soil organic matter—a key but poorly understood component of

609 terrestrial C cycle, *Plant Soil*, 338, 143–158, 2011a.

610 Rumpel, C. and Kogel-Knabner, I.: Deep soil organic matter—a key but poorly understood component of

611 terrestrial C cycle, *Plant Soil*, 338, 143–158, 2011b.

612 Sanderman, J. and Amundson, R.: A comparative study of dissolved organic carbon transport and stabilization

613 in California forest and grassland soils, *Biogeochemistry*, 92(1–2), 41–59, doi:10.1007/s10533-008-9249-9,

614 2009.

615 Scharpenseel, H. W. and Becker-Heidelmann, P.: Shifts in <sup>14</sup>C patterns of soil profiles due to bomb carbon,

616 including effects of morphogenetic and turbation processes, *Radiocarbon*, 31(3), 627–636, 1989.

617 Schaub, M., Dobbertin, M., Kräuchi, N. and Dobbertin, M. K.: Preface-long-term ecosystem research:

618 Understanding the present to shape the future, *Environ. Monit. Assess.*, 174(1–4), 1–2, doi:10.1007/s10661-010-

619 1756-1, 2011.

620 Schimel, D. S., House, J. I., Hibbard, K. a, Bousquet, P., Ciais, P., Peylin, P., Braswell, B. H., Apps, M. J.,

621 Baker, D., Bondeau, A., Canadell, J., Churkina, G., Cramer, W., Denning, a S., Field, C. B., Friedlingstein, P.,

622 Goodale, C., Heimann, M., Houghton, R. a, Melillo, J. M., Moore, B., Murdiyarso, D., Noble, I., Pacala, S. W.,

623 Prentice, I. C., Raupach, M. R., Rayner, P. J., Scholes, R. J., Steffen, W. L. and Wirth, C.: Recent patterns and

624 mechanisms of carbon exchange by terrestrial ecosystems., *Nature*, 414(6860), 169–72 [online] Available from:

625 <http://www.ncbi.nlm.nih.gov/pubmed/11700548>, 2001.

626 Schleppei, P., Muller, N., Feyen, H., Papritz, A., Bucher, J. B. and Fluehler, H.: Nitrogen budgets of two small

627 experimental forested catchments at Alpental, Switzerland, *For. Ecol. Manage.*, 127(101), 177–185, 1998.

628 Schmidt, M. W. I., Torn, M. S., Abiven, S., Dittmar, T., Guggenberger, G., Janssens, I. a, Kleber, M., Kögel-

629 Knabner, I., Lehmann, J., Manning, D. a C., Nannipieri, P., Rasse, D. P., Weiner, S. and Trumbore, S. E.:

630 Persistence of soil organic matter as an ecosystem property., *Nature*, 478(7367), 49–56 [online] Available from:

631 <http://www.ncbi.nlm.nih.gov/pubmed/21979045> (Accessed 21 January 2014), 2011.

632 Schrumpf, M. and Kaiser, K.: Large differences in estimates of soil organic carbon turnover in density fractions

633 by using single and repeated radiocarbon inventories, *Geoderma*, 239–240, 168–178 [online] Available from:

634 <http://linkinghub.elsevier.com/retrieve/pii/S0016706114003577>, 2015.

635 Schrumpf, M., Kaiser, K., Guggenberger, G., Persson, T., Kogel-Knabner, I. and Schulze, E.-D.: Storage and

636 stability of organic carbon in soils as related to depth, occlusion within aggregates, and attachment to minerals,

637 *Biogeosciences*, 10, 1675–1691, 2013.

638 Seneviratne, S. I., Corti, T., Davin, E. L., Hirschi, M., Jaeger, E. B., Lehner, I., Orlowsky, B. and Teuling, A. J.:

639 Investigating soil moisture-climate interactions in a changing climate: A review, *Earth-Science Rev.*, 99(3–4),

640 125–161, doi:10.1016/j.earscirev.2010.02.004, 2010.

641 Sierra, C. A., Muller, M., Metzler, H., Manzoni, S. and Trumbore, S. E.: The muddle of ages, turnover, transit,

642 and residence times in the carbon cycle, *Glob. Chang. Biol.*, 1–11, doi:10.1111/gcb.13556, 2016.

643 Smith, J. C., Galy, A., Hovius, N., Tye, A. M., Turowski, J. M. and Schleppei, P.: Runoff-driven export of

644 particulate organic carbon from soil in temperate forested uplands, *Earth Planet. Sci. Lett.*, 365, 198–208,

645 doi:10.1016/j.epsl.2013.01.027, 2013.

646 Solly, E., Schöning, I., Boch, S., Müller, J., Socher, S. a., Trumbore, S. E. and Schrumpf, M.: Mean age of

647 carbon in fine roots from temperate forests and grasslands with different management, *Biogeosciences*, 10(7),

648 4833–4843, doi:10.5194/bg-10-4833-2013, 2013.

649 Torn, M. S., Swanston, C. W., Castanha, C. and Trumbore, S. E.: Storage and turnover of organic matter in soil,

650 in *Biophysico-Chemical Processes Involving Natural Nonliving Organic Matter in Environmental Systems*,

651 edited by N. Senesi, B. Xing, and P. M. Huang, p. 54, John Wiley & Sons, Inc., 2009.

652 Trumbore, S. E. and Czimeczik, C. I.: *Geology. An uncertain future for soil carbon.*, *Science*, 321, 1455–1456,

653 2008.

654 van der Voort, T. S., Hagedorn, F., McIntyre, C., Zell, C., Walthert, L. and Schleppei, P.: Variability in <sup>14</sup>C

655 contents of soil organic matter at the plot and regional scale across climatic and geologic gradients,

656 *Biogeosciences*, 13(January), 3427–3439, doi:10.5194/bg-2015-649, 2016.

657 Wacker, L., Bonani, G., Friedrich, M., Hajdas, I., Kromer, B., Němec, M., Ruff, M., Suter, M., Synal, H.-A. and

658 Vockenhuber, C.: MICADAS: Routine and high-precision radiocarbon dating, *Radiocarbon*, 52(2), 252–262,

659 2010.

660 Wacker, L., Němec, M. and Bourquin, J.: A revolutionary graphitisation system: Fully automated, compact and

661 simple, *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms*, 268(7–8), 931–934

662 [online] Available from: <http://linkinghub.elsevier.com/retrieve/pii/S0168583X09011161> (Accessed 2 August

663 2013), 2009.

664 Walthert, L., Blaser, P., Lüscher, P., Luster, J. and Zimmermann, S.: *Langfristige Waldökosystem-Forschung*

665 *LWF in der Schweiz. Kernprojekt Bodenmatrix. Ergebnisse der ersten Erhebung 1994–1999.*, 2003.

666 Walthert, L., Graf Pannatier, E. and Meier, E. S.: Shortage of nutrients and excess of toxic elements in soils  
667 limit the distribution of soil-sensitive tree species in temperate forests, *For. Ecol. Manage.*, 297, 94–107,  
668 doi:10.1016/j.foreco.2013.02.008, 2013.  
669 Walthert, L., Lüscher, P., Luster, J. and Peter, B.: Langfristige Waldökosystem- Forschung LWF. Kernprojekt  
670 Bodenmatrix. Aufnahmeanleitung zur ersten Erhebung 1994–1999, Swiss Federal Institute for Forest, Snow and  
671 Landscape Research WSL, Birmensdorf., 2002a.  
672 Walthert, L., Lüscher, P., Luster, J. and Peter, B.: Langfristige Waldökosystem-Forschung LWF in der Schweiz.  
673 Kernprojekt Bodenmatrix. Aufnahmeanleitung zur ersten Erhebung 1994–1999, Birmensdorf. [online]  
674 Available from: <http://e-collection.ethbib.ethz.ch/show?type=bericht&nr=269,2002b>.  
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678 **Author contributions**

679 T.S. van der Voort planned, coordinated and executed the sampling strategy and sample collection, performed  
680 the analyses, conceptualized and optimized the model and processed resulting data. U. Mannu led the model  
681 development. F. Hagedorn lent his expertise on soil carbon cycling and soil properties. C. McIntyre facilitated  
682 and coordinated the radiocarbon measurements and associated data corrections. L. Walthert and P. Schleppi lent  
683 their expertise on the legacy sampling and provided data for the compositional analysis. N. Haghypour  
684 performed in isotopic and compositional measurements. T. Eglinton provided the conceptual framework and  
685 aided in the 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 (m a.s.l.)	Elevation	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>Walther 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 bulk (y)	Average turnover 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>Walther et al., 2002, Walther 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.

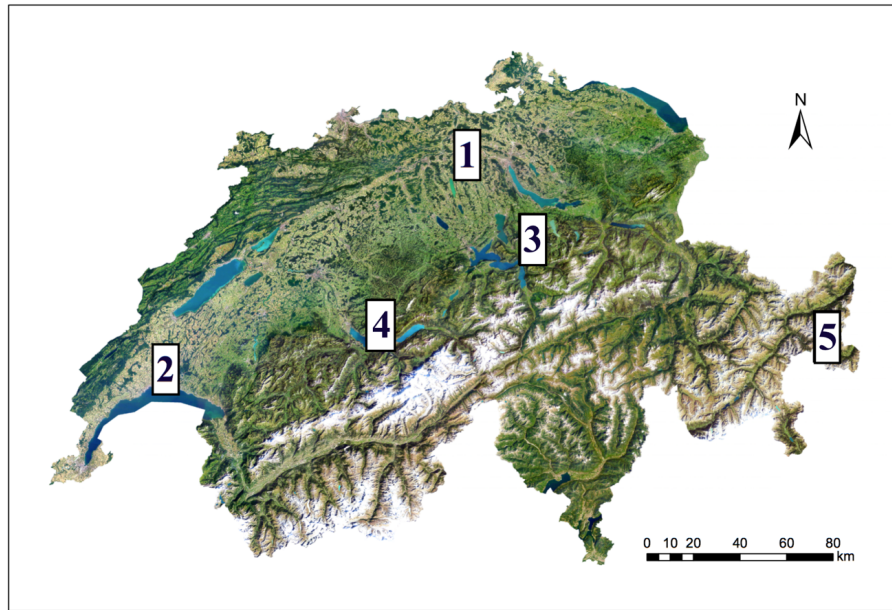
<sup>2</sup>Krause et al., 2013

<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

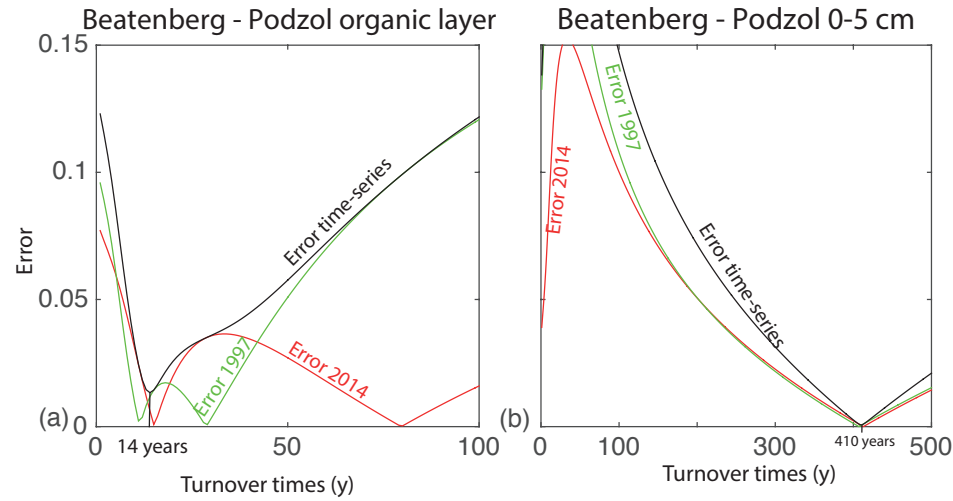
**Table 4** Pearson correlations for averaged depth intervals for the top soil (0-20 cm, n=5) and deep soil (20-60 cm, n=5). Significance denoted with ; \*, \*\* or \*\*\* for respectively p-values smaller than 0.1 (marginally significant) 0.05, 0.005 and 0.0005 (significant). Non-significant correlations 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



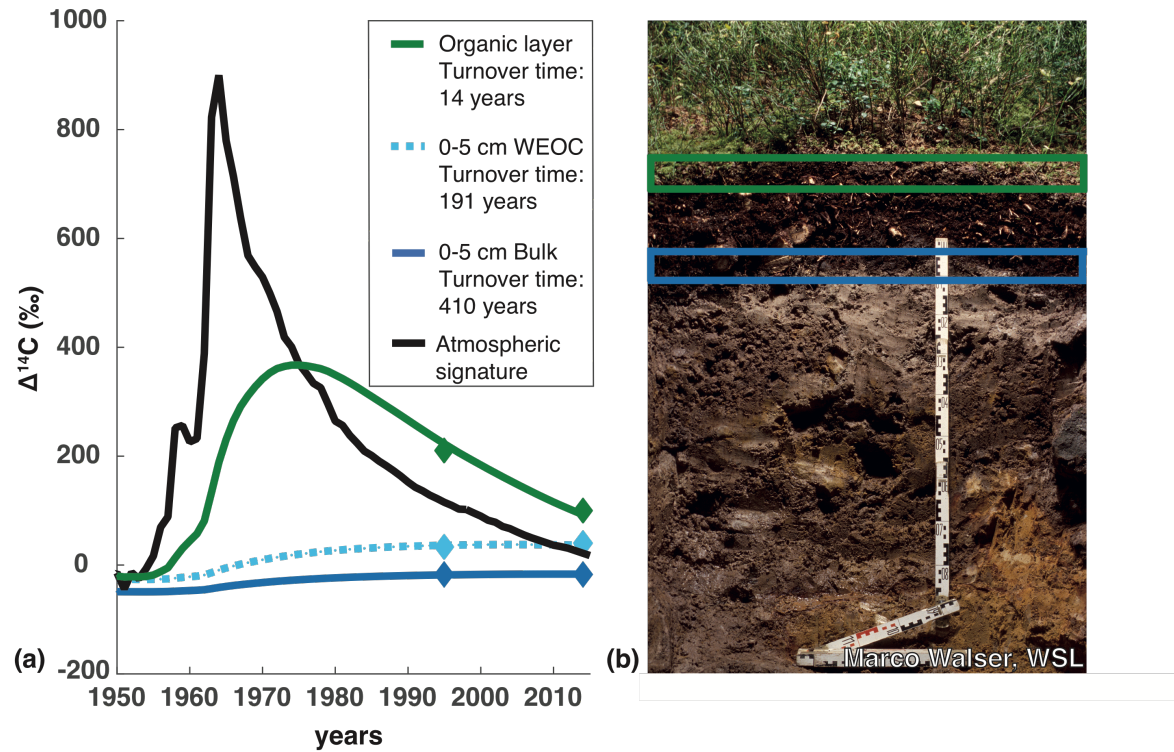
**Figure 1** Sample locations, all of which are part of the Long-term ecosystem research program (LWF) of the Swiss Federal Institute WSL, 1) Othmarsingen, 2) Lausanne, 3) Alptal, 4) Beatenberg and 5) Nationalpark Image made using 2016 swisstopo (JD100042).



**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.

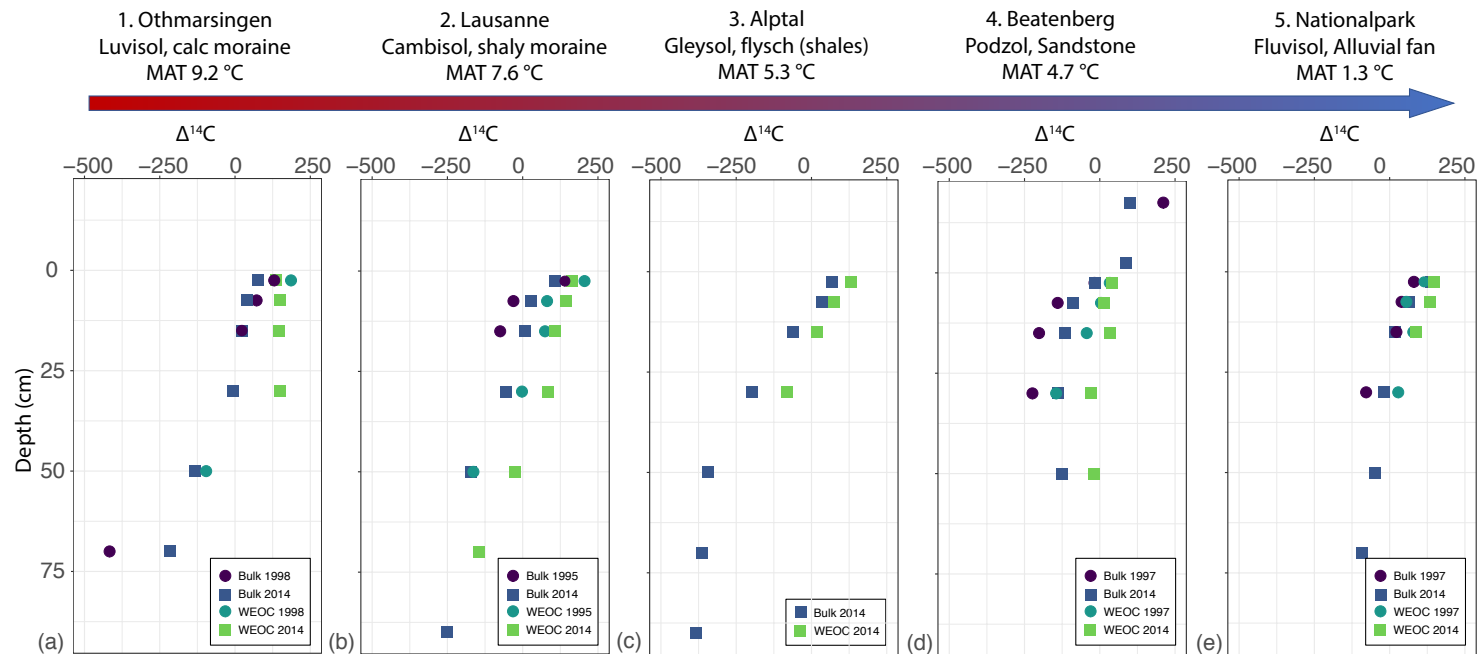
**Comment [TSvdV4]:** Figure also augmented with soil type as suggested





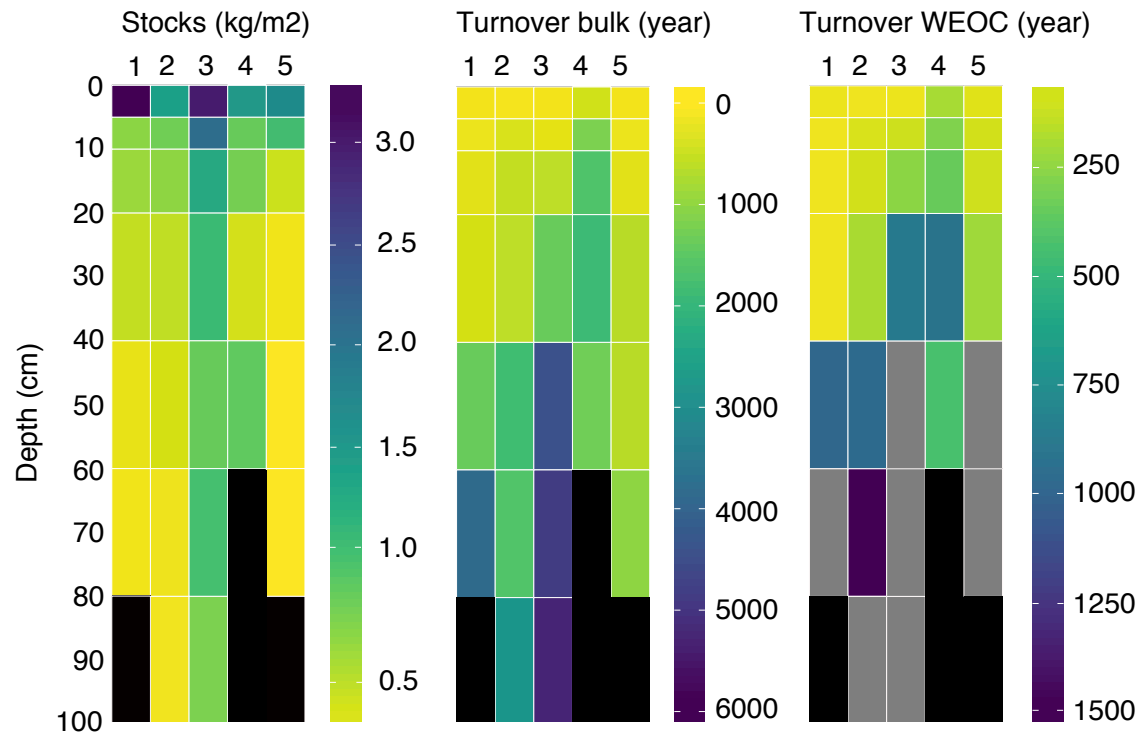
**Figure 3** (a) Time-series soil carbon turnover time in years (y) as determined by numerical modelling for (b) sub-alpine site Podzol Beatenberg. The bulk turnover in the organic layer is rapid (14 years), followed by the turnover of the water-extractable organic carbon (WEOC) (191 years) and the bulk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

**Comment [TSvdV5]:** Figure also augmented with soil type as suggested



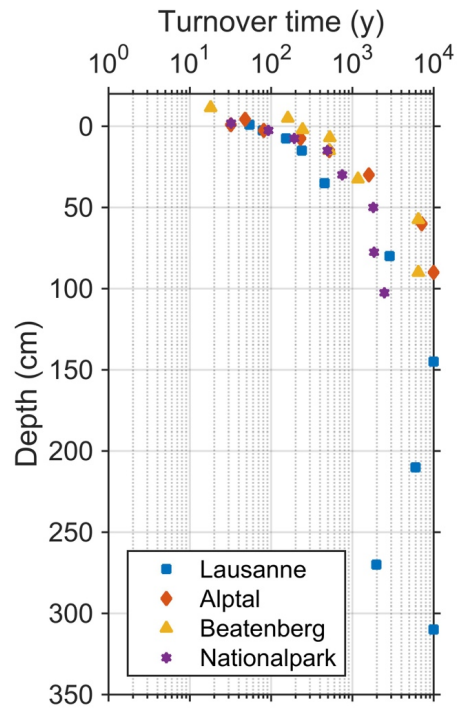
**Figure 4 (a-e)** Changes in radiocarbon signature of both bulk soil and WEOC over two decades at four sites on a climatic gradient. For Alptal (Gleysol) (c) only the 2014 time-point was available. For the warmer locations Othmarsingen and Lausanne (Luvisol, Cambisol MAT 9.2-7.6 °C), depletion in bomb-derived 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.

**Comment [TSvdV6]:** Dear Sébastien, as suggested we have now included the soil types both in the figure heading as well as the text. We have improved the text following this suggestion elsewhere as well. We have also augmented the colours in this figure as compared to the previous version.



**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).

**Comment [TSvdV7]:** Figure also augmented with soil type as suggested



**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.

