

Interactive comment on “Dynamics of deep soil carbon – insights from ^{14}C time-series across a climatic gradient” by Tessa Sophia van der Voort et al.

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Response to Jerome Balesdent Understanding the dynamics of carbon in deep soil layers is an important issue, and this study uses an excellent sequence and provides a rare dataset: soil ^{14}C measurement at two dates using archived samples brings a precious information of C dynamics. One of the interesting results is the demonstration of the occurrence of rock-derived carbon. Another concerns the age of water extractable carbon. The analytical methods are high standard and highly relevant. I therefore consider it is worth publishing the data in Biogeochemistry. Unfortunately, there are major concerns that need revision. The most important is that the math-

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ematical and numerical interpretations look inappropriate, and this leads the authors to give conclusions that are in contrast with what the data show, whereas some unprecedented results could be derived. I finally suggest two alternative solutions: either the authors drop the modelling part and make a semi-quantitative interpretation of the data, either they use another model. I also noticed miscellaneous improvements to be done. The discussion should be updated according to these major points. The title and summary are nevertheless appropriate. Dear Prof. Balesdent,

→ Thank you very much for your positive feedback and thorough review. We very much appreciate that you value the importance of the data for the wider Biogeosciences community. Your comments about the turnover time modelling are also very insightful and the issues have subsequently been addressed. There was indeed a semantic issue which caused problems, so we incorporated all of your feedback. We realized that most of the modelling was explained in the code in the SI, and that therefore the text in the main text was absolutely inadequate in order to explain our calculations. Consequently, paper and especially the discussion was updated according to these major points. As you indicated, the title and summary remained appropriate.

We want to thank you again for your helpful review, which has further improved this paper. Please find detailed replies below. 1. The chosen model is unlikely to simulate observed data. Most of samples below 10 cm show an increase in $\Delta^{14}\text{C}$ between 1990's and 2010's, by several 10‰ (Figure 3), and even some above 10 cm do. As seen in the FIGURE below, which was built for this review, the ^{14}C content of well mixed compartments directly fed from atmospheric C has DECREASED with time since the 1990's (or increased by less than 4‰ for slow pools). The sum of two parallel pools cannot have a $\Delta^{14}\text{C}$ increased between 1995 and 2014. FIGURE: Simulated $\Delta^{14}\text{C}$ of a well-mixed compartment under steady state as a function of compartment turnover rate, for two dates of sampling. Thank you for these comments. Indeed, rapidly turnover compartments have decreased in ^{14}C in the last two decades, whilst the slower compartments have increased in ^{14}C signature (e.g. Figure 3a) (as you also indicate

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in your comments below). As you indicated in your supplemental, there was indeed a semantic issue with the turnover time definition when estimating the size of the two respective pools, which has now been adapted. Our apologies for the confusion.

I finally understood (from 14C data in Figure 3 and turnover time data in Table S5) that the the "mostly reliable" kWSOC value is more or less the arithmetic mean of two kWSOC values, one calculated in the 1990's and the other in the 2010's. The authors must invoke other processes to explain an increasing $\Delta^{14}\text{C}$. These processes may act together and interact: - Transit of carbon in another horizons or pool before entering the observed layer. This might be associated with either bioturbation or DOC production from an above layer, movement, and insolubilization. The data tend to indicate that carbon movement is a significant cause of the increase in $\Delta^{14}\text{C}$ across the sequence. - non-steady state, e.g. increased bioturbation due to warming, change in NPP and/or decay rates. To me, the fact that the $\Delta^{14}\text{C}$ of WSOC of all samples (except Othmarsingen 0-5 cm and Lausanne 0-5 cm) increased is a proof that WSOC is a by-product of SOM aged several 10th of years (usual age of OH horizons), and not directly fed by vegetation decomposition. This would be a bright finding and merit appropriate modelling. Thank you for these comments as well. We did not sufficiently explain how we estimate the turnover of the WEOC or 'labile' pool using the 14C time-series. We have addressed this now in the method section, by detailing the different steps and the error calculation (Equations 1-4). In short, we do not take the arithmetic mean, but rather use the standard equations (e.g. Herold et al., 2014; Torn et al., 2009) to find the likeliest turnover time considering both datapoints of the time series. Instead of the usual excel-based method, we do this in MatLab because it is automated and more repeatable. The solution which has the lowest calculated residual square root mean error (RSME) is automatically chosen, as opposed to a manual iteration.

→ Thank you for highlighting the importance of potentially DOC-driven transport of young(er) carbon through the deep soil, we have included this in our discussion. We have also now included your suggestion in Section 4.1.3 to highlight that WEOC is

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likely not fed by vegetation decomposition but rather is derived from several decades-old SOM.

2. Consistency in model implementation (to be confirmed). I tried to calculate by myself turnover time values, based on 14C data in Figure 3 and turnover time data in Table S5, and didn't find the author's results. This may arise from the fact that the basic differential equations of the model (equation 5 = SI.7) looks false, or at least do not correspond to authors' hypotheses. Equation SI.7 states: $F(t) = k \cdot F_{\text{atm}}(t) + m_1 \cdot F(t-1) \cdot (1 - \lambda - k_1) + m_2 \cdot F(t-1) \cdot (1 - \lambda - k_2)$ This equation indicates that the flux of 14C leaving the system (out of desintegration) is: $(m_1 \cdot k_1 + m_2 \cdot k_2) \cdot F(t-1)$, i.e., $k \cdot F(t)$ Since the corresponding flux of carbon is $k = m_1 \cdot k_1 + m_2 \cdot k_2$, this equation says that the 14C activity of carbon leaving the system is $F(t-1)$. So the equation would IMPLICITLY considers that the activity of the flux out is the same as that of the compartment itself. This is typically the assumption of a so-called 'well mixed' compartment, and is not the case of a system with two compartments. It would only accept the solution $k_1 = k_2$. Making this implicit assumption is a current mistake or at least a source of disagreement in isotope geochemistry. As a consequence, I guess that the authors have calculated a mean turnover time corresponding to a single compartment for bulk carbon, and an independent specific turnover time of WSOC. The error might be linked with my point 3 below. See a proposal for the correct equation as an appendix of this review. The authors are invited to check how eq SI.7 was implemented and how the couple (k_2, m_1) was inferred from bulk $F^{14}\text{C}$.

→ Thank you, there are two main things raised in this comment: A. Modelling Structure Indeed, we have calculated a mean turnover time corresponding to a single compartment for bulk carbon, and one independent specific turnover time of WSOC. We have clarified this in the text. B. Model consistency Thank you for your suggestions and example for Figure SI.7, we have implemented all of your suggestions (Eq. 6). More details can be found below. We would like to clarify that we merely transformed the usual excel file people use to find turnover time to MatLab-driven optimization, be-

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cause it saves time, is repeatable, unbiased and error can be quantified. We have now also quantified all our errors (See SI). Furthermore, the code can easily be used as well for longer time-series (i.e. > 2 timepoints). We benchmarked our results to the Excel-method, and the results agree.

3. Mathematical (and semantic) misuse of 'turnover time'. Let us call the turnover time of carbon in the compartment $T = 1/k$. Mathematically, the carbon input to the system is $m_1/T_1 + m_2/T_2$. The size of the compartment is $m_1 + m_2$. So, the turnover time, which is the ratio of pool size to the input, is: $T = (m_1 + m_2)/(m_1/T_1 + m_2/T_2)$. In Table SI.5, which presents the main result, i.e. the values of turnover time, the authors calculated the bulk turnover time as: $T = (m_1.T_1 + m_2.T_2)/(m_1 + m_2)$, which is wrong. What authors call "turnover time" is in fact the MEAN AGE of carbon, which is different of the mean turnover time in non-well mixed compartments. The error is not only semantic because it possibly have interfered in model and ^{14}C equation (point 2). Sierra et al. (2016), whom you cite lines 161-162, recommends the use of "age", not "turnover time" for this variable. See also Manzoni et al. (2009).

→ Indeed, there was a (semantic) inconsistency regarding turnover time between the Main text and the SI, which we have now addressed and corrected. We also implemented your equation. We have the ^{14}C -determined 'turnover time' for the bulk soil, whilst stating that we assume a steady state. We have also clarified our definition in the text, following Manzoni et al. (2009) as well.

4. Data availability. The authors must provide in SI a table including the primary data, i.e., $\Delta^{14}\text{C}$, C stock by horizon, WEOC stocks. Reference that were used to estimate atmospheric $\Delta^{14}\text{C}$ (post bomb and pre-bomb) should be indicated (e.g. Reimer, Hua etc.)

→ We have included an excel file with all the raw data regarding $\Delta^{14}\text{C}$ and stocks the WEOC material. The WEOC concentration is low (< 1 %) and can be found in SI Table 4. We had indicated the provenance of our pre- and post-bomb data already in the

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method section, but we have now further clarified it.

5. Hypothesis on WSOC as the labile pool. Line 180-182 and 190-191: A major (if not the major) assumption of the model is that the dynamic pools has the same decay rate as that of WEOC. The 'dynamic' pools contains as much as 88% of soil C (on the average 34%), whereas WEOC only a few %. Assigning the constant k of WEOC to the dynamic pool is therefore a surprising and very heavy hypothesis. (see also point 1.) Alternatively, the study may have targetted the study of WSOC dynamics for itself, e.g., considered that both WSOC and bulk C are heterogenous pools, each with a labile and a more stable component, but in varied proportion. Many other models use particulate organic matter (i.e. either sand-size primary organic particles or light OM, which has been described as having a good fit with labile carbon

→ Yes indeed, it was our assumption is we assumed the measured WEOC could be representative of the dynamic pool. There are studies that hypothesize WEOC could be indicative of a larger dynamic pool (Baisden and Parfitt, 2007; Koarashi et al., 2012). But indeed, this is a heavy assumption. We have therefore decreased the importance of the two-pool model in the paper, and highlighted this assumption. Indeed, both the WEOC and bulk themselves can heterogeneous pools, hence we also looked at biomarkers in another study (e.g. Van der Voort et al., 2017, Diverse Soil Carbon Dynamics Expressed at the Molecular Level, GRL). Looking at other fraction would be a worthwhile topic for future work.

6. Conclusions on correlation with MAP. Projecting conclusions on the effect of MAP on the basis of a "wet" sequence, i.e., where the water deficit is probably low if not nil, may look brash. The driest site is 800 mm, but with a MAT 1.3C and probably a small PET. Furthermore (Lines 360-361), authors state that 'The only climate-related driver which appears to be significant is precipitation' whereas the r^2 coefficient between MAP and turnover 0-20 cm is 0.04! I would recommend here to cite Carvalhais et al. (2013) and Mathieu et al. (2015), who highlighted the role of precipitation in SOM stabilization or ecosystem carbon turnover. I finally suggest to moderate the conclusions, but maybe

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discuss the role of precipitation on DOC movement (see point 1).

→ Thank you for these insights. As suggested, we have highlighted the role of precipitation as SOM stabilizer and interaction with DOC movement, and tempered our statements about precipitation. Indeed, Switzerland is a wet country! Your own 2018 paper also could show the important role of evapotranspiration but we unfortunately do not have this data. Also, we adapted the phrasing of line 360-61, the role of precipitation is pronounced for the deep soil. 7. Presentation of model and equations. The presentation of both the model and the optimization process is obscure throughout the text and should be more precise, in either text or SI. In the cases with four radiocarbon dates (2 sampling dates x two fractions), the optimization of three dynamic parameters is not a formal solution, but a best fit.

→ Indeed, we have now mentioned this specifically in the text.

The type of adjustment (least squares ?) and a criterion of the fit (e.g., RMSE) should be indicated.

→ This has been included in the main text instead of the SI, we use RSME. Harmonize the name of variables throughout the text and SI. For consistency with SI, please use m instead of F in eqn (3), (4) and (4); and possibly F instead of R . Also use the same character k in SI and main text. Harmonize M (Figure S2) and m , etc. Thank you, this has now been adjusted. How were single points managed ? (Line 194-195. " Due to limited availability of archived samples, there are only single time points available for some samples as indicated in Fig. 4.")

→ This has been clarified in the main text, we solve the standard radiocarbon decay equations (e.g. Torn et al., 2009,). This is done more traditionally in Excel, we did the same using a Matlab optimization.

8. Miscellaneous. lines 51-52 note the pioneer studies by Jenkinson et al (1992) on long-term experiments. The models by Braakheke et al. (2014) also simulates

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14C profiles in rather similar podzols, using WSOC as well, and may receive more attention in the discussion section. Also note (e.g. Line 34) the conclusions of Mathieu et al. (2015) concerning soil versus climate drivers of 14C, and (lines 39-40) the recent paper by Balesdent et al. (2018), which improved the understanding of the significance of deep soil C to the global C cycle.

→ Thank you, I have incorporated these literature suggestions. I had already cited Braakheke et al.

Move lines 126-128 (WEOC) to the end of 2.1. (WEOC extraction). Note that extraction with Na 0.86 M is not exactly Water extraction, since it moves some exchangeable calcium, disperses clays and therefore moves sorbed organic compounds that would not have been mobilized by water.

→ Indeed, we followed Hagedorn et al., 2014 when preparing the extraction, and have this stated this clearly in the method section.

Line 252 ' Deeper soil bulk stock and turnover positively...' and table S5: avoid "turnover " alone standing for "turnover time" in such sentences, because the common sense of turnover is turnover rate, i.e., the inverse of turnover time. This may lead to a reverse understanding of correlations.

→ Indeed! We adapted this now.

Line 262. Balesdent et al. (2018) reported that 21% of world subsoil C (30-100 cm) is less than 50 years old. We have included this The amount of WEOC (while not used in the modelling experiment) would be welcome.

→ We have included this in the SI Table 4. Concentrations are low (< 1%)

Surprisingly, the section of Material and methods indicates that NPP and its components were measured, which is a rare information in SOM studies. As a result, authors have an indicator of the true turnover time of soil C, i.e. the ratio of Soil C stock to C input is known, that they do not use.

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→ Indeed, there is NPP data, but we were recommended by the field experts that although it was representative for the tree vegetation, we had better not use it for estimating soil flux, as there would be too many assumptions to be considered. We did include the data, so others are free to use it.

Figure 4 contains the main primary result of the study. Policies Should be enlarged. The square signs for Aptal WEOC 1997 are misleading. Table S5 is the main final result and should take place in the main document.

→ We have adapted this figure slightly. Following your critique about the assumption of using WEOC as a dynamic pool we reduced the importance of the fraction modelling in the paper, so we opted to keep it in the SI.

→ Note that the bi-exponential age distribution is factually the age distribution of C in current "four pools" models such as RothC (or Century). All coupling of these models with radiocarbon more or less managed bi-exponential age distribution and 14C; e.g., Jenkinson et al. (1992).

→ Yes, we are familiar with Century (RothC), but feel applying them would be beyond the scope of this paper.

9. Appendix The differential equation should consider F1 and F2 the 14C fraction in pools 1 and 2, respectively, as illustrated in your Fig S1. Input flux to pool1 is $k_1.m_1$; input flux to pool2 is $k_2.m_2$ $F_1(t) = k_1.F_{atm}(t) + (1 - k_1 - \lambda).F_1(t - 1)$ $F_2(t) = k_2.F_{atm}(t) + (1 - k_2 - \lambda).F_2(t - 1)$ which give: $F(t) = m_1F_1(t) + m_2.F_2(t) = k.F_{atm}(t) + m_1.(1 - k_1 - \lambda).F_1(t - 1) + m_2.(1 - k_2 - \lambda).F_2(t - 1)$ And needs numerical resolution of F1 and F2. → Thank you, we implemented this. 10. Cited references Balesdent J., Basile-Doelsch I, Chadoeuf J., Cornu S., Derrien D. Fekiacova Z., Hatté C. Atmosphere-soil carbon transfer as a function of soil depth. *Nature*, 559, 599–602. (2018) doi.org/10.1038/s41586-018-0328-3 Jenkinson D.S., D.D. Harkness, E.D. Vance, D.E. Adams and A.F. Harrison. Calculating net primary production and annual input of organic matter to soil from the amount and radiocarbon content of

C9

soil organic matter. *Soil Biol. Biochem.* 24(4):295-308 (1992) Manzoni, S., Katul, G. G. & Porporato, A. Analysis of soil carbon transit times and age distributions using network theories. *J. Geophys. Res.* 114, G04025 (2009) Mathieu J., Hatté C., Parent E., Balesdent J. Deep soil carbon dynamics are driven more by soil type than by climate: a worldwide meta-analysis of radiocarbon profiles. *Global Change Biology* 21, 4278-4292. (2015) doi:10.1111/gcb.13012. → Thank you, we implemented these papers 11. Figure. Simulated $\Delta^{14}C$ of a well-mixed compartment under steady state as a function of compartment turnover rate, for two dates of sampling. Compartment has a single C7 exponential distribution of ages; system start 8050 BP; atmospheric $\Delta^{14}C$ after Reimer et al. (2009) and Hua et al. (2013); Northern hemisphere zone N2; May-August. Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2018-361>, 2018.

Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2018-361/bg-2018-361-AC1-supplement.zip>

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2018-361>, 2018.

Response to Jerome Balesdent

Understanding the dynamics of carbon in deep soil layers is an important issue, and this study uses an excellent sequence and provides a rare dataset: soil ^{14}C measurement at two dates using archived samples brings a precious information of C dynamics. One of the interesting results is the demonstration of the occurrence of rock-derived carbon. Another concerns the age of water extractable carbon. The analytical methods are high standard and highly relevant. I therefore consider it is worth publishing the data in Biogeochemistry. Unfortunately, there are major concerns that need revision. The most important is that the mathematical and numerical interpretations look inappropriate, and this leads the authors to give conclusions that are in contrast with what the data show, whereas some unprecedented results could be derived. I finally suggest two alternative solutions: either the authors drop the modelling part and make a semi-quantitative interpretation of the data, either they use another model. I also noticed miscellaneous improvements to be done. The discussion should be updated according to these major points. The title and summary are nevertheless appropriate.

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1. The chosen model is unlikely to simulate observed data.

Most of samples below 10 cm show an increase in $\delta^{14}\text{C}$ between 1990's and 2010's, by several 10‰ (Figure 3), and even some above 10 cm do. As seen in the FIGURE below, which was built for this review, the $\delta^{14}\text{C}$ content of well-mixed compartments directly fed from atmospheric C has DECREASED with time since the 1990's (or increased by less than 4‰ for slow pools). The sum of two parallel pools cannot have a $\delta^{14}\text{C}$ increased between 1995 and 2014.

FIGURE: Simulated $\delta^{14}\text{C}$ of a well-mixed compartment under steady state as a function of compartment turnover rate, for two dates of sampling.

Thank you for these comments. Indeed, rapidly turning-over compartments have decreased in ^{14}C in the last two decades, while the slower compartments have increased in ^{14}C signature (e.g. Figure 3a) (as you also indicate in your comments below). As you indicated in your supplemental, there was indeed a semantic issue with the turnover time definition when estimating the size of the two respective pools, which has now been adapted. Our apologies for the confusion.

Fig. 1. Point by point replies

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1 Dynamics of deep soil carbon – insights from ^{14}C time-series across a 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 $-1.9\text{ }^{\circ}\text{C}$) gradient are determined using time-series (~ 20 years) ^{14}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; Goshiva 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 ^{14}C content (Rehemeyer 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 (^{14}C) can be a powerful tool to determine the dynamics of carbon turnover over decadal to millennial timescales

1

Fig. 2. Paper included changes

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Supporting Information for

2 Dynamics of deep soil carbon – insights from ¹⁴C time-series across a climatic gradient

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12
13 **Contents of this file:**
14 Tables S1-5
15 Figures S1-3
16 **Introduction**
17 This supporting information provides details on the modeling approach used in this paper, contains ancillary
18 data and details on the estimation of petrogenic carbon.
19
20 **S1 Numerical model in Matlab Environment**
21 The purpose of this section is to explain the necessity of a robust numerical modelling approach for ¹⁴C time-
22 series which can be applied ubiquitously in radiocarbon turnover estimates in oceanic and terrestrial reservoirs.
23 **The code that can be employed to do this is freely available with this paper.** Tere et al. (2009) explains that
24 a single measured radiocarbon value collected on the falling arm of the bomb-spike yields two estimates of the
25 turnover time. In the case of two time-points, this uncertainty is avoided and a single estimate can be produced.
26 For this reason, time-series radiocarbon can be crucial. Craven et al. (2015) highlighted that owing to continued
27 burning of fossil fuels, the importance of time-series measurements can only increase. In this section, we
28 elaborate on a sensitivity analysis and error propagation analysis. The Matlab numerical optimization rou-
29 tinizations until the lowest mean-squared error for both time points is reached. [There are separate scripts to](#)
30 [download software for a single time-point, a time-series and multiple points.](#)
31
32 **S1.1 Necessity of numerical approach**
33 **As the incorporation of atmospheric ¹⁴C into the atmosphere reservoir is inherently time-dependent, and**
34 **therefore as to be solved numerically, as can be proven in the following manner:** For the isotopic signature of any
35 reservoir the value of a variable q_i can be formulated as the following:
36
$$\frac{dq_i}{dt} = -\lambda q_i + \dots$$
 Eq. S1.1
37 Here q_i refers to the new value q_i relative to the previous point, λ is the derivative (i.e. slope) of the
38 previous point and Δt refers to the time-step between t and $t + 1$. For any case of uptake of atmospheric ¹⁴C,
39 the derivative can be determined.

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Fig. 3. Paper tracked changes