Geogenic organic carbon in terrestrial sediments and its contribution to total soil carbon

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16 Abstract

17 Geogenic organic carbon (GOC) from sedimentary rocks is an overlooked fraction in soils that has not 18 yet been quantified, but influences the composition, age and stability of total organic carbon (OC) in 19 soils. In this context, GOC is the OC in bedrocks deposited during sedimentation. However, the 20 contribution of GOC to total soil OC may vary varies depending on the type of bedrock. However, As 21 yet, no far studies have been carried out to investigated the contribution of GOC derived from 22 different terrestrial sedimentary rocks to soil OC contents.

- 23 In order to fill this knowledge gap, 10-m long sediment cores from three sites recovered from Pleistocene Loess, Miocene Sand and Triassic Red Sandstone were analysed in 1 m depth intervals 24 and the amount of GOC calculated based on ¹⁴C measurements. The ¹⁴C ages of bulk sedimentary OC 25 revealed that OC is comprised of both biogenic and geogenic components. The biogenic component 26 27 relates to OC that recently entered the sediments from plant sources since soil development started. Assuming an average age for this biogenic component ranging from 1,000-4,000 years BP we 28 calculated average amounts of GOC in the sediments starting at 1.5 m depth based on measured ¹⁴C 29 ages. The median amount of GOC in the sediments was then taken and its proportion of soil mass (g 30 GOC per kg⁻¹ fine soil) calculated in the soil profile. All the sediments contained considerable 31 amounts of GOC (median amounts of 0.10 g kg⁻¹ in Miocene Sand, 0.27 g kg⁻¹ in Pleistocene Loess 32 and 0.17 in Red Sandstone) compared with subsoil OC contents (between 0.53-15.21 g kg⁻¹). Long-33 34 term incubation experiments revealed that the GOC appeared comparatively stable against biodegradation. Its possible contribution to subsoil OC stocks (0.3-1.5 m depth) ranged from 1 to 26 35 % in soil developed in the Miocene Sand, from 16 to 21 % in the Loess soil and from 6 to 36 % at the 36 Red Sandstone site. Thus GOC with no detectable ¹⁴C content influenced the ¹⁴C ages of subsoil OC. 37 and may partly explain the strong increase in ¹⁴C ages observed in many subsoils. This could be 38 particularly important in young soils on terrestrial sediments with comparatively low amounts of OC, 39 where GOC can make a large contribution to total OC stocks. 40
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49 experiment

⁴⁸ Keywords Geogenic organic carbon; sedimentary organic carbon; ¹⁴C; terrestrial sediments; incubation

51 1. Introduction

52 On average, the world's soils store more than 50 % of OC in the subsoil below 30 cm depth (Batjes, 2014). This type of carbon is considered a highly stable carbon pool due to its apparently high ¹⁴C ages 53 (Mathieu et al., 2015; Schrumpf et al., 2013). However, another explanation for this could be the 54 contribution of geogenic organic carbon (GOC), which is defined here as OC that originates from 55 56 deposition during sedimentation and rock formation, and may increasingly influence subsoil OC with depth (Graz et al., 2010; Kögel-Knabner et al., 2008; Schrumpf et al., 2013; Trumbore, 2009). GOC in 57 most cases is devoid of ¹⁴C and thus may lead to an overestimation of ancient OC sources although a 58 59 number of studies showed the importance of root derived, young OC inputs to subsoils (Angst et al., 2016; Crow et al., 2009). Therefore GOC may significantly influence and affect the overall ¹⁴C signal, 60 particularly in OC-poo subsoils. Vindušková et al. (2015) investigated the contribution of GOC to 61 soils in reclaimed mine soils, and found GOC contributions to total soil OC of between 26 and 99 %. 62 Furthermore OC-rich sediments with contents of 2-7 g kg⁻¹ (Hemingway et al., 2018) and 28-105 g kg⁻¹ 63 ¹ (Frouz et al., 2011) have been investigated with regard to the stability of OC in these sediments, but 64 65 no conclusion reached about GOC contributions in soils. However, the impact of GOC on soils 66 derived from sediments or sedimentary rocks with lower OC contentshas not yet been investigated. Considering that approximately 65 % of the continental earth's surface is covered with sediments and 67 68 sedimentary rocks (Amiotte Suchet et al., 2003), a potentially large fraction of soils could contain 69 GOC that contributes to soil OC stocks, even though a large portion might be derived from recent 70 sedimentation processes. There is not yet much literature about sediments containing only low amounts of OC. There are estimations that assume sandstones to be free of GOC(van der Voort et al., 71 2018) or, in contrast, a storage model that assumes generally high GOC amounts of 2.4 g kg⁻¹ for all 72 73 sandy deposits (Copard et al., 2007). Therefore, more information about the amounts of GOC in sediments is needed. 74

To estimate the possible contribution of GOC to subsoil OC stocks, it is important to establish the 75 76 amount of OC in sediments that comes from sedimentation (GOC) and to distinguish it from OC 77 derived from current vegetation (biogenic OC). There are many soil- and substrate-specific factors that might influence the OC contribution from current vegetation to sedimentary OC, such as potential 78 79 rooting depth or pore distribution. No method has yet been established to allow a direct quantification 80 of GOC in different soils or sediments, apart from promising methods to quantify the graphitic part of GOC in soils (Zethof et al., 2019). The only reliable approach to distinguish between both sources is 81 the use of ${}^{14}C$. Since deposition of sediments mostly took place > 50,000 years BP, they do not contain 82 14 C, which has a mean half-life time of 5,730 years (Libby, 1952). In addition, the δ^{13} C values of OC 83 in the sediments allow carbonaceous sources with δ^{13} C values around 0 ‰ to be distinguished from 84 organic sources with δ^{13} C values < -22 ‰. Thus, the use of both carbon isotopes could reveal wether 85 the OC is a mixture of GOC and OC from vegetation that is less than 50,000 years old. 86

87 One important question about the possible contribution of GOC in soils is whether the GOC is 88 mineralised when it becomes part of the soil. As GOC resists degradation once it has been deposited, it can be assumed that it already exhibits a strong inherent recalcitrance. Nevertheless, this could also be 89 due to a physical protection that prevents microbial accessibility. However, when it becomes part of 90 91 the subsoil during soil development, this OC pool could be degraded by the infiltration of water, 92 oxygen, fresh nutrients and microorganisms. Direct microbial coal degradation has already been observed in incubation experiments on mine soils (Rumpel and Kögel-Knabner, 2002; Waschkies and 93 94 Huttl, 1999) or shale bedrocks directly exposed to the surface (Soulet et al., 2017). There has been no 95 study to establish whether GOC is degradable in OC-poor sediments or sedimentary rocks has not 96 been investigated, but it could differ since the amount of available OC can also drive microbial respiration (Colman and Schimel, 2013). Therefore these sediments might contain fewer 97 microorganisms that can be spatially separated from the GOC, which may hamper its respiration. 98

99 To the best of the author's knowledge, there has only been one study by van der Voort et al. (2018) 100 that has investigated the amount of GOC in soils. They estimated that GOC makes up around 80 % of 101 soil OC in a moraine-derived soil, suggesting that GOC's contribution to soil OC is large. However, 102 apart from this study by van der Voort et al. (2018) on a very specific sediment, there have been no 103 direct calculations of the amount of GOC in soils.

The aim of this study was to quantify GOC in different terrestrial sediments and a sedimentary rock, and investigate its stability in incubation experiments in order to make assumptions about its possible contribution to soil OC stocks in soil profiles at the same site. The main research questions were: i) What is the relationship between sedimentary and subsoil OC contents? ii) Is OC in sediments ¹⁴C-free and how much is really geogenic? iii) Will sedimentary GOC be degraded? and iv) How much does GOC contribute to soil OC?

111 **2.** Material and methods

112 *2.1 Site description*

Three sites were selected with different sedimentary bedrocks derived from a single geologic substrate 113 114 that can be found close to the surface and is homogeneous down to 10 m depth. The sites represented 115 one sedimentary rock and two soft sediments. The sedimentary rock was a sandstone (Solling Formation, Triassic) under European beech forest (Fagus sylvatica) 11.5 km north-east of Göttingen 116 (51°35.012' N; 10°3.960' O), referred to below as "Red Sandstone". The soil is classified as a Folic 117 Brunic Arenosol according to the World Reference Base for Soil Resources (WRB, 2006). The 118 sediments were loessic deposits (Weichselian Glacial) that have been under agricultural landuse for 119 the past decades, 30 km north of Göttingen (51°48.101 N; 9°58.002' O), referred to as "Loess", and 120 terrestrial sandy deposits from the Miocene (Neogene formerly named Tertiary) in a European beech 121 122 forest 13 km south-west of Göttingen (51°28.673 N; 9°45.323' O) referred to as "Miocene Sand". The respective soils have been classified as a Haplic Luvisol and a Dystric Chromic Arenosol accordingly. 123 The sediments at the Loess site were deposited between the last glacial and interglacial periods 124 125 between 115,000 and 400,000 years BP, according to Jordan and Schwartau (1993). To the best of the 126 author's knowledge, these forest sites have never been under agricultural use. The associated soils are 127 classified as a Luvisol and a Cambisol respectively. Mean annual air temperature and precipitation 128 were 9.2 °C and 647 mm (1981-2010) according to the nearby weather station that covers all three 129 sites.

130 2.2 Sampling and sample preparation

Two 10-m long sediment cores with a diameter of 15 cm were drilled at each site in April 2017. For the soft sediments (Loess and Miocene Sand), drilling was conducted as percussion drilling, and for the hard sediment (Red Sandstone) as cable core drilling with water as the flushing solution. The replicates per site were drilled approximately 10 m apart. The sampled cores were subdivided into 1-m increments. For further chemical analysis, material was taken from a depth of 85-95 cm for each 1-m increment, and the outer 5 cm removed to avoid possible contamination. Thus the increment from 1-2 m for example, was represented by a sample from a 1.85-1.95 m depth.

One sample from each depth increment (1-2 m, 2-3 m, 3-4 m , 4-5 m, 5-6 m, 6-7 m, 7-8 m, 8-9 m, 9-10 m) for chemical analysis was oven dried at 60 °C and sieved to pass 2 mm. The Red Sandstone samples were crushed with a hammer before being dried and sieved. Additionally, approximately 1-m deep soil profiles were dug and soil samples taken from the different classified layers to obtain corresponding soil parameters (Fig. 1).



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Fig. 1: Respective soil profiles at the Loess (A), Miocene Sand (B) and Red Sandstone (C) sites. Soil
classification was conducted using the German classification system for soil horizons (Eckelmann et al., 2006)
and later transferred to the WRB (2006). Depth transitions to the starting sediment were 82 cm for Loess (Al +
Bt + elCv), 72 cm for Miocene Sand (Bv - ilCv) and 68 cm for Red Sandstone (ilCv₁).

Samples were oven dried at 60°C and stored and sieved to pass 2 mm. For the determination of OC
 and ¹³C, samples were ground in a planetary ball mill.

150 2.3 Chemical analysis and calculations

Three aliquots of each sieved sample were analysed by dry combustion for total C and total N content 151 (TruMac CN LECO, St. Joseph, MI, USA). Samples with a pH value of > 6 (measured in 0,01 mol L⁻¹ 152 CaCl) were analysed for carbonates after the sample was ignited for 16h in a muffle kiln at 450 °C 153 according to Nelson and Summers (1983), to remove the organic part of total C. The OC concentration 154 was calculated by subtracting carbonaceous C from total C, expressed as g OC kg⁻¹ dry matter. 155 Homogenised samples were further analysed for δ^{13} C values after carbonate removal in an isotope 156 ratio mass spectrometer (Delta Plus, Thermo Fisher, Waltham, MA, USA) coupled to an elemental 157 158 analyser (FLASH EA 1122 NA 1500; Wigan, United Kingdom). Resulting δ^{13} C values (‰) were 159 expressed relative to the international standard of Vienna Pee Dee Belemnite. The bulk densities for 160 the soil samples were obtained with 250 cm³ sampling rings from each layer of the soil profile. For the 161 sedimentary Loess and Miocene Sand samples, the bulk density of the deepest soil sample (94 and 100 cm respectively) was used. The bulk densities and densities without pore space of the intact Red 162 Sandstone cores were determined on four subsamples (from 1.6, 3.6, 7 and 9 m depth) with a Dryflow-163 pycnometer (GeoPyc 1360) and a gas pycnometer (AccuPyc 1330) respectively. Bulk densities for the 164 missing depth increments were linearly interpolated. For radiocarbon (¹⁴C) analysis, the sediment 165 samples were first treated with 1% HCl to remove inorganic C, and then transferred into pre-166

- 167 combusted quartz ampoules containing copper oxide and silver wool. The ampoules were evacuated,
- 168 flame-sealed, then combusted at 900 $^{\circ}$ C, and the CO₂ evolved was purified on a vacuum ring
- 169 (Rethemeyer et al., 2019). The 14 C contents were measured with the MICADAS accelerator mass
- 170 spectrometry (AMS) system at ETH Zürich, Switzerland. Where possible, one sedimentary sample per
- 171 depth increment and site and one sample per soil layer was analysed. Due to the very low OC contents
- in some sediment samples, ¹⁴C contents could only be determined for three samples from the Miocene
- 173 Sand (from 1.9, 4.9 and 7.9 m depth) and four from the Red Sandstone (1.9, 4.9, 7.9 and 9.9 m depth).
- 174 For the Loess, ¹⁴C of bulk OC was measured in all depth intervals (1.9-9.9 m).
- 175 Total OC stocks (Mg ha⁻¹) were calculated according to Eq. 1:

$$OC \ stock = \ OC \ \cdot BD \ \cdot (1 - stone \ content) \cdot depth \cdot 0.1$$
 Eq. 1

where *OC* is the weight-based OC content, either in the fine soil <2-mm fraction of the soil profiles or in the sediments (g kg⁻¹), *BD* is the bulk density of the fine soil (g cm⁻³), stone content is the volumebased proportion of stones (cm³ cm⁻³) and depth is the thickness of the depth increment (cm). To be able to compare OC stocks and contributions from GOC, it was decided to set the borders between the topsoils- and subsoils at 0.3 m and the transition from subsoils to the sediments at 1.5 m. According to Richter and Markewitz (1995) this represents a common border for the transition from soil to sediment. The sediments where further subdivided into an upper and a lower part at a_depth of 4 m.

183 In a second step, the amount of GOC and biogenic OC in the sediments was calculated, -considering 184 GOC as one carbon pool free of ¹⁴C. For the sediments, the proportion of biogenic OC ($f_{biogenic}$) of the 185 total amount of OC was calculated in a two pool model (Eq. 2) used by Cerri et al. (1985):

$$f_{biogenic} (\%) = \frac{F_{biogenic \, OC} - F_{GOC}}{F_{sample} - F_{GOC}} \cdot 100$$
 Eq. 2

where *F* represents the ¹⁴C content in the fraction modern carbon ($F^{14}C$) from a source compared with the ¹⁴C content of an oxalic standard (Stuiver and Polach, 1977; Torn et al., 2009). The sources were the GOC fraction (F_{GOC}), the sample (F_{sample}) and the biogenic OC fraction ($F_{biogenic OC}$). Since the ¹⁴C content of the GOC fraction can be set to zero, this equation could be simplified to:

$$f_{biogenic} (\%) = \frac{F_{biogenic \ OC}}{F_{sample}} \cdot 100$$
 Eq. 3

For the biogenic OC in the sediments, an average ¹⁴C age ranging from 1,000-4,000 years BP was assumed, based on published ¹⁴C results of dissolved OC reaching greater depths (Artinger et al., 1996; Schiff et al., 1997). The ¹⁴C contents in the sediment from 2 to 4 m depth of the loess led to ages
< 3,000 years BP, and were therefore even younger than at 74 cm depth (4,413 years BP). Thus, they

were treated like the soil part for the calculation below of a GOC fraction. Respective times were converted into ¹⁴C contents ($F_{biogenic OC}$) according to Torn et al. (2009):

$$F_{biogenic \, OC} = e^{\left(\frac{t}{-8033}\right)}$$
 Eq. 4

where *t* represents the ¹⁴C age (1,000 or 4,000 years BP respectively) and 8033 represents the mean life of radiocarbon in years. The proportion of GOC in the sediments (f_{GOC}) is therefore the portion left (Eq. 5):

$$f_{GOC} = 100 \% - f_{biogenic}$$
Eq. 5

For the depth increments without measured ¹⁴C ages (Tab. S1), the calculated amounts were linearly 199 interpolated with measured ¹⁴C ages from samples above and below. This was done by assuming a 200 201 depth-dependent correlation and using the adjacent values. To calculate the amount of GOC in the soil 202 profiles, the weight-based amount of GOC in the sediments was first calculated by multiplying its fraction (f_{GOC}) by the respective OC content (in g OC per kg⁻¹ dry mass). The median amount of GOC 203 (g GOC per kg⁻¹ dry mass) of these sedimentary values was then taken and its proportion of soil OC 204 content calculated (g OC per kg^{-1} fine soil) in the soil profile. This was done for the proportion of 205 GOC in the sediments calculated with a 1,000 and a 4,000-year-old biogenic OC fraction ($F_{biogenic OC}$ in 206 207 Eq. 3) to obtain a range of GOC contributions. It was assumed that the GOC fraction resisted degradation during soil formation. Therefore, this proportion represents the largest possible amount of 208 GOC that could contribute to soil OC stocks. On this assumption, it was also possible to define the 209 influence of GOC in the soil profile on the resulting ¹⁴C ages. Since the calculated ¹⁴C ages represent a 210 mixture of the ¹⁴C content from GOC and the biogenic fraction (Eq. 5), the GOC fraction has the same 211 influence on the soil ¹⁴C age as on bulk OC (according to Eq. 3). Reducing the age by this fraction 212 213 would therefore represent an "unbiased" age of soil-derived-OC.

214 2.4 Incubation experiment

To assess the potential stability of OC in the sediments against microbial decay, two laboratory 215 216 incubation experiments were conducted at 20 °C for 50 and 63 days respectively. This was done to reveal the potential degradation of OC from the sediments. A temperature of 20 °C was chosen 217 because only very low degradation rates were expected at lower temperatures. The first, 50-day 218 experiment was conducted with intact Red Sandstone core samples, while the second experiment was 219 performed after the Red Sandstone was crushed to pieces < 2 mm to simulate the process of 220 221 weathering when the intact sediment or sedimentary rock becomes part of the (sub)soil. For the 222 incubations, four subsamples of 1,340-6,890 g per sample from different depth intervals were used. 223 The sample material was stored at room temperature until the start of the incubation experiment. Four 224 samples were taken from each sediment and from each of four depth ranges for Miocene Sand (1.2-225 2.8, 3.2-4.8, 6.1-7.8 and 8.2-9.9 m), Red Sandstone (2-3, 4-5.8, 7-8 and 8.4-9.8 m) and Loess (1.4-2,

2.7-3, 4.8-6 and 9.1-10 m). Water content was adjusted to correspond to 40 % of water-holding 226 227 capacity based on the poured bulk density, determined by filling the loose material into a defined volume and measuring its weight. The material for the four repetitions was mixed in large plastic vats 228 229 and water added before the respective four subsamples were transferred to incubation vessels. There 230 were also four blank samples with no material. Based on preliminary tests and their calculated bulk density and porosity, the intact Red Sandstone samples were kept in a barrel with pure water for 14 231 hours to reach a water content of nearly 40 %. Samples were placed in polycarbonate vessels with a 232 233 volume of 7069 cm³ and made airtight. The lids contained two tube connectors so that the samples 234 could be flushed with ambient air. After flushing, samples were set to a starting pressure of about 235 1,300 mbar and kept closed until the end of the incubation. Nine gas samples were taken in evacuated 236 glass vials (20 mL), 0, 3, 7, 13, 21, 30, 59 and 63 days after the start of incubation. Samples were 237 analysed for CO₂ concentrations by gas chromatography (Agilent 7890A, GC, Agilent Technologies, Santa Clara, USA) to account for the amount of accumulated CO₂. Three additional gas samples were 238 239 taken 0, 30 and 63 days after the incubation started and analysed with an isotope ratio mass spectrometer (Delta Plus XP, Thermo Fisher Scientific, Bremen, Germany) to account for the 240 development of δ^{13} C of CO₂ during the respiration. The corresponding pressure was measured on each 241 sampling date. When the over-pressure of a vessel was lost due to leakages, it was removed from the 242 243 sampling because contamination with ambient air could no longer be ruled out. This happened for a third of all the samples. 244

The amount of respired CO_2 -C (mg CO_2 -C d⁻¹) was calculated using Eq 6.

$$CO_2 - C = \frac{0.1 \cdot p \cdot x_i \cdot M \cdot V}{R \cdot T \cdot t}$$
 Eq. 6

where *p* is the pressure (mbar), x_i is the difference in CO₂ concentration between the samplings (ppm), *M* is the molar mass of C (g mol⁻¹), *V* is the air volume of the sample (m³), *R* is the molar gas constant (J kmol⁻¹ K⁻¹), *T* is the incubation temperature (K) and *t* is the elapsed time (d) between the samplings. Based on the δ^{13} C values of CO₂ the proportion of CO₂ derived from carbonates was subtracted where necessary according to Bertrand et al. (2007). This respiration rate was related to the OC content of the samples (called "OC-normalised respiration") by being divided by the total amount of OC in g in the sample.

Since we observed an almost linear respiration behaviour was observed in both incubation experiments, a simple linear regression model was fitted to describe the mineralisation rate per time (mineralised OC (%) = $k \cdot t$).

258 Statistical analyses were conducted using the statistical environment R (R Core Team (2018)) including the function "lm" to fit linear models and the package ggplot2 (Wickham, 2016) for 259 graphical presentation. The models were tested for deviations from homoscedasticity, normality of 260 261 residuals and absence of collinearity. The tests revealed heteroscedasticity of the residuals, which can be explained by an increasing standard deviation with time. It subsequently became apparent that the 262 residuals were not normally distributed since the dependent variable, representing the proportion of 263 264 OC being mineralised, only allows values between 0 and 1. Bearing this in mind, the results should be 265 treated as an indicator of differences between the samples and a scale for the mineralisation of the OC 266 pool. Therefore calculations of standard errors and significance of the parameters were omitted since 267 this would not produce reasonable results with the model used.

268 **3. Results**

269 3.1 Relationship between sedimentary and subsoil organic carbon

270 Using three laboratory replicates per sample and comparing them with the muffled samples, 271 measurable OC contents were detected in all the sediments analysed. The mean relative standard deviation of the laboratory replicates was 9.5 %. In terms of the detection limit, the sample with the 272 lowest total C content (mean of 0.04 g C kg⁻¹ soil) showed values between 0.00 and 0.01 g C kg⁻¹ soil 273 after removal of OC at 450 °C. Thus the range of 0.00 to 0.01 g C kg⁻¹ soil was assumed to be the 274 mean standard error from the measurement. Despite having the same material down to 10 m depth at 275 276 each site, there were still some inhomogeneities that were visible and measurable. This was especially 277 true for Loess and Red Sandstone. The amount of OC in the sediments from 1 to 10 m depth was comparatively low in Miocene Sand and Red Sandstone (0.04-0.71 g kg⁻¹ and 0.01-0.53 g kg⁻¹, 278 respectively), while much higher OC contents of 0.21-9.71 g C kg⁻¹ were found in Loess (Fig. 2 a). 279 The median OC content of the sediments was within a range comparable to that of the respective 280 deepest subsoil horizon. This deepest horizon was a Cv horizon at 94 cm depth for Loess, 100 cm 281 depth for Miocene Sand and 74 cm depth for Red Sandstone. In detail, the median OC content in the 282 sediments, compared with the respective Cv horizons, corresponded to 27 % for Loess, 29 % for 283 284 Miocene Sand and 39 % for Red Sandstone. The Loess OC contents were highly variable, highlighting 285 the changing sedimentary conditions during the past glacial and interglacial periods (Jordan and Schwartau, 1993). At 4-5 m depth, the OC contents of Loess were even higher (9.7 g kg⁻¹) than in the 286 subsoil (3 g kg⁻¹). This was not an outlier because the high OC content could be visually confirmed by 287 the very dark colour of the sample. In Miocene Sand and Red Sandstone, no clear depth gradient of 288 289 OC was found at 2-10 m depth (Fig. 2 a). Even though the OC content in the sediments were low, the 290 OC stocks could be very large. A comparison of OC stocks in topsoils (0-0.3 m), subsoils (0.3-1.5 m) 291 and the sediments down to 10 m depth revealed quite high OC stocks in the sediments. For Loess, OC in the sediment contributed up to 71 % of the total OC amount while it was 51 % for Red Sandstone 292 293 and 21 % for Miocene Sand (Table 1).

The distribution of the $\delta^{13}C$ values of OC in the soil and sediment profiles showed an increase in $\delta^{13}C$ 294 with depth in the soil down to 1 m (Fig. 2 b). In contrast, the δ^{13} C values of OC in the sediments 295 showed no clear trend with increasing depth, but they all were within the range of C₃ plant material. A 296 value above -25 ‰ for Red Sandstone at 4 m depth could be explained by the high values of inorganic 297 298 carbon (IC) at this depth. It can be assumed that decarbonisation of this sample was not completely 299 successful. Unexpectedly high amounts of IC were found in parts of Red Sandstone, indicating the 300 presence of calcareous deposits in this terrestrial material (Fig. 2 c). The Loess also included some 301 distinct calcareous layers at 5 and 10 m depth, while there were only small amounts of around 0.1 mg IC g⁻¹ soil at the other depths. This could be due to the fact that the investigated loess deposits belong 302 to the "Leine Ilme Basin", a region with aoelian loamy loess that has been decalcified during 303 304 weathering and soil genesis (Wagner, 2011), while the contents in the soil profile can be explained by 305 liming. No IC was present in Miocene Sand.



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Fig. 2: Depth distribution of different bulk properties of the soil profiles and deep drilling cores. Presented parameters include the log scale organic carbon (OC) and inorganic carbon (IC) content (a and c) and the δ^{13} C values of the organic carbon (b) related to the amount of fine soil or dry mass respectively. Filled and unfilled symbols represent the two different cores. For the Loess, only one core could be analysed.

311 Table 1: OC stocks and proportions for the three sites down to 10 m depth. Proportions of biogenic and 312 geogenic OC were calculated based on ¹⁴C results and assumptions described in the material and methods 313 section. Represented ranges are calculated based on the assumption of a 1,000 or 4,000-year-old biogenic OC 314 fraction reaching the sediments.

| ubstrate | Layer | Depth (m) | TOC | OC stocks (Mg ha ⁻¹) | Proportion of OC (%) ^b |
|----------|-------|-----------|-----|----------------------------------|-----------------------------------|
|----------|-------|-----------|-----|----------------------------------|-----------------------------------|

| | | | . 1 | | geogenic | biogenic | geogenic | biogenic |
|-----------------|----------------|------------|-----------|------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| | | | (Mg·ha⁻¹) | (%) ^a | 4,000 1,000 yrs. yrs. | 1,000 4,000 yrs. yrs. | 4,000 1,000 yrs. yrs. | 1,000 4,000 yrs. yrs. |
| Loess | Topsoil | 0.0 - 0.3 | 40.6 | 11 | 1.1 - 1.2 | 40 - 39 | 3 - 3 | 97 - 97 |
| | Subsoil | 0.3 - 1.5 | 66.0 | 17 | 5.1 - 5.3 | 61 - 61 | 8 - 8 | 92 - 92 |
| | Upper Sediment | 1.5 - 4.0 | 218.6 | 57 | 10.6 - 11.0 | 208 - 208 | 5 - 5 | 95 - 95 |
| | Lower Sediment | 4.0 - 10.0 | 55.1 | 15 | 42.9 - 46.7 | 8 - 12 | 78 - 85 | 15 - 22 |
| Red andstone | Topsoil | 0.0 - 0.3 | 22.2 | 30 | 0.6 - 0.7 | 22 - 22 | 3 - 3 | 97 - 97 |
| | Subsoil | 0.3 - 1.5 | 13.7 | 19 | 1.6 - 1.7 | 12 - 12 | 12 - 12 | 88 - 88 |
| | Upper Sediment | 1.5 - 4.0 | 18.7 | 25 | 13.1 - 14.9 | 4 - 6 | 70 - 80 | 20 - 30 |
| | Lower Sediment | 4.0 - 10.0 | 19.3 | 26 | 14.1 - 15.7 | 4 - 5 | 73 - 82 | 18 - 27 |
| Miocene | Topsoil | 0.0 - 0.3 | 39.1 | 31 | 0.3 - 0.3 | 39 - 39 | 1 - 1 | 99 - 99 |
| | Subsoil | 0.3 - 1.5 | 60.8 | 48 | 1.3 - 1.6 | 59 - 60 | 2 - 3 | 97 - 98 |
| Sand | Upper Sediment | 1.5 - 4.0 | 10.6 | 8 | 3.7 - 5.8 | 5 - 7 | 34 - 55 | 45 - 66 |
| | Lower Sediment | 4.0 - 10.0 | 16.3 | 13 | 8.3 - 10.8 | 6 - 8 | 51 - 66 | 34 - 49 |

315 ^a % of total OC stock from 0-10 m

316 ^b % of OC stock in the respective depth increment

317 *3.2 Ages of organic carbon in soils and sediments and contributions from geogenic organic carbon*

318 The ages of OC in the Loess soil profiles revealed a modern carbon signature (0 years BP) at 0.3 m depth, with a sharp increase up to $4,413 \pm 51$ years BP at 0.7 m depth (Fig. 3 a). For the Red 319 320 Sandstone soil profile there was only an increase in the ages from a modern signature at 0.04 m to 532 \pm 41 years BP at 0.3 m depth. The Miocene Sand soil profile at 0.4 m depth showed an increase from 321 $1,277 \pm 41$ to $1,771 \pm 44$ years BP at 0.6 m depth. Thus, OC of the subsoil (around 0.6 m depth) in 322 Loess was more than twice as old as in Miocene Sand. In contrast, Loess had a modern signature at 0.3 323 324 m, while the soil developed in Red Sandstone showed an average age of 532 ± 41 years BP at 0.3 m 325 depth.

326 The ages of OC in the sediments ranged from 2,200-30,730 years BP, with respective mean ages of $9,077 \pm 3,234$ years BP for Miocene Sand, $13,674 \pm 9,632$ years BP for Loess, and $14,463 \pm 1,992$ 327 years BP for Red Sandstone. For all sediments, 11 out of 16 samples had a ¹⁴C content that led to an 328 ¹⁴C age older than 11,600 years BP, which we assumed to be the time after the latest glacial period 329 when soil development started (Litt et al., 2007). Therefore the sediments contained a mixture of 330 geogenic (¹⁴C-free) and biogenic (with ¹⁴C) OC. Despite being the youngest sediment, Loess partly 331 revealed the highest apparent ${}^{14}C$ ages up to $30,730 \pm 631$ years BP (Fig. 3 a). The ages of OC in the 332 sediment of Red Sandstone and Miocene Sand ranged from 12,940 ± 132 to 17,390 ± 206 years BP 333 334 and from $6,750 \pm 86$ to $12,770 \pm 151$ years BP respectively, revealing no depth trend with age in the deeper sediment. The calculated GOC fraction in the sediments was highest for Red Sandstone, 335 ranging from 67 to 87 %, with a mean of 77 % (Fig. 3 b). For the three Miocene Sand samples, the 336 GOC fraction ranged from 29 to 77 % with a mean of 53 %. Loess showed a sharp increase at a depth 337

of five metres where GOC contribution rising to between 71 to 98 % while it was only 5 to 19 % at the

339 2 to 4 m depth.

- 340 The calculated weight-based content of GOC in the sediment revealed a comparatively uniform
- 341 distribution for all the sediments with depth, except the extremely high contents of Loess at 5 m depth
- 342 (Fig. 3 c). The investigated sediment depths revealed quite a narrow range of GOC contents with 0.10
- ± 0.03 g kg⁻¹ for Miocene Sand, 0.17 ± 0.12 g kg⁻¹ for Red Sandstone and 0.27 ± 0.08 g kg⁻¹ for Loess.



344

Fig. 3: Depth distribution of apparent ¹⁴C ages (in years BP) (a), GOC contribution to OC contents in the sediments (b) and
 resulting weight based amounts of GOC (c) in the sediments. The range in the contribution from GOC is due to the
 assumption of an average biogenic OC age of 4,000 (empty shapes) and 1,000 years (filled shapes).

348 3.3 Biodegradability of sedimentary derived organic carbon

The incubation experiment revealed a potential, but low biodegradability of OC for all samples, but 349 without a clear depth gradient from 1 to 10 m (Fig. 4). To compare the effect of crushing on the 350 351 respiration of Red Sandstone samples, the mineralisation rates from the first incubation experiment were compared with those of the second incubation experiment (Table 2). While Red Sandstone 352 showed very low mineralisation when the samples were incubated as intact cores (0.3-1.0 mg CO₂-C 353 g^{-1} OC y^{-1}), the mineralisation rate constants were up to five times higher when the samples were 354 crushed (1.0-2.1 mg CO₂-C g⁻¹ OC y⁻¹). In a comparison of Loess and Miocene Sand, both revealed 355 large differences between samples with the lowest (1.2 and 1.8 mg CO₂-C g⁻¹ OC y⁻¹) and highest (34.2 356 and 12 mg CO_2 -C g⁻¹ OC y⁻¹) respiration rate constants. Interestingly, there was no depth gradient for 357

the different substrates, but samples from 7 and 9 m depth of Miocene Sand tended to have up to 6.8 358 times lower respiration rate constants than samples from the 2 and 4 m depth. There was also a very 359 wide variation between samples from the same depth (see Fig. A1) as revealed by the high standard 360 deviations. Assuming a constant mineralisation rate over time, the results of the incubation experiment 361 362 would result in mean residence times of between 29 and 135 years for the Loess, 83 and 556 years for the Miocene Sand and between 476 and 1,000 years for the crushed Red Sandstone. Since the CH₄ 363 levels of the samples remained at a low level (Fig. A3) there were no indications of oxygen-limited 364 365 conditions during the incubation.

366

367 Table 2: Comparison of mineralisation rate constants between the first (intact Red Sandstone samples) and second incubation368 experiments (crushed Red Sandstone samples) calculated using linear models.

| Substrate | Depth (m) | Mineralisation rate constant (mg CO ₂ -C g ⁻¹ OC y ⁻¹) ^a |
|---------------|--------------|---|
| | 2 | 12.0 |
| Maria San 1 | 4 | 10.5 |
| Miocene Sand | 7 | 1.8 |
| | 9 | 2.9 |
| | 1.7 | 7.4 |
| I | 2.9 | 34.2 |
| Loess | 5.4 | 10.2 |
| | 9.6 | 1.3 |
| | 2.5 | 0.3 |
| Uncrushed | 5 | 1.0 |
| Red Sandstone | 7.5 | 0.4 |
| | 9 | 0.4 |
| | 2.5 | 1.3 |
| Crushed | 5 | 2.1 |
| Red Sandstone | 7.5 | 2.0 |
| | 9 | 1.0 |

^a represents the slope of the fitted linear model for respiration during the incubation experiment



371

Fig. 4: Potential degradability of sedimentary OC from three sites. Results represent cumulative respiration from Loess and Miocene Sand samples and both Red Sandstone incubation experiments with uncrushed and crushed samples, with respective standard deviations (n = 4). Dashed lines represent a fitted linear model to the respiration data.

376 *3.4 Possible contribution from geogenic organic carbon to soil organic carbon*

377 As bedrock weathers, it becomes part of the soil and GOC also becomes soil OC. The potential amount of GOC in the subsoil was dependent on the sedimentary bedrock (Fig. 5). In the subsoil of 378 Miocene Sand the proportion of GOC amounted to 2-3 %, at the Loess site it was 8 %, and at the Red 379 380 Sandstone site it was 12 % (Table 1). For the defined topsoils (0-30 cm depth), contributions of GOC to soil OC were smaller with 0.7-0.9 % for Miocene Sand, 2.8-2.9 % for Loess and 2.8-3.0 % for Red 381 Sandstone The possible contribution of GOC to soil OC was also calculated under the assumption that 382 383 the biogenic fraction had an unrealistically high average age of 10,000 years (Fig. A5). This resulted in the GOC fraction falling to 0 for Miocene Sand. For Loess the geogenic fraction fell by ~ 25 %, and 384 for Red Sandstone by ~ 16 %. 385

The presence of ¹⁴C-free GOC in soil OC reduced the mean bulk soil OC ¹⁴C ages, depending on its proportion in soil OC content. Topsoils that developed in Loess and Red Sandstone had a modern ¹⁴C content of 1.029 and 1.035 F¹⁴C, similar to the atmospheric ¹⁴C content in 1950. Due to the large proportion of biogenic OC, no influence of a geogenic fraction was detected in the topsoil of these sites. No ¹⁴C data were available for the topsoil of Miocene Sand. For all subsoils, the influence of

GOC on bulk soil OC ¹⁴C contents depended greatly on the depth and corresponding OC contents. For 391 Loess, the possible influence on ¹⁴C ages in the subsoils was quite high, with an average of 10 % 392 reduction in mean apparent ¹⁴C ages in the subsoil. Thus it would reduce the measured age of 4,413 393 years BP in 74 cm depth by 532-555 years BP. Geogenic OC potentially reduced the mean apparent 394 395 radiocarbon age of 1,277 years BP at 0.39 m depth in Miocene Sand by about 7-9 years, and the radiocarbon age of 1,771 years BP at 0.61 m depth by 20-24 years. These reductions are below the 396 respective standard deviations of the measurement. Nevertheless, at 1 m depth a given possible 397 proportion of 13.1-16.3 % would reduce an interpolated ¹⁴C age of 3,053 years BP by 399-497 years 398 For Red Sandstone the influence of GOC on ¹⁴C ages would be highest in the subsoil. At 74 cm depth 399 400 it would increase an age of 1,453 years BP by 451-490 years BP. Due to the low amounts of soil OC at 401 90 cm depth at the Red Sandstone site, the weight-based median amount of GOC in the sediments was 402 four times higher than the biogenic amount of soil OC.



404 Fig. 5: Largest possible contribution of GOC to OC (red area) in relation to bulk OC content, taking into account the median
 405 GOC contents of the sediments for the respective horizontal weight based OC contents. The contribution of GOC is the
 406 mean amount based on the assumption of an average biogenic OC age of between 4,000 and 1,000 years

407 4. Discussion

408 *4.1 Geogenic OC in the sediments*

409 It should be noted that these GOC calculations in sediments were based on the assumption of biogenic 410 OC in the sediments not being older than 4,000 years BP on average. The influence of a biogenic OC fraction derived from soils that developed before the latest glacial period was also excluded. Thus it is 411 412 possible that the biogenic OC fraction in the sediments is even older. Nevertheless, even with an 413 assumed age of 10,000 years for the biogenic OC fraction, the greatest possible contribution was 15.4 % for Loss (94 cm depth) and 21.5 % for Red Sandstone (74 cm depth) (Fig. A5). Considering 414 that modern OC entering the sediment has a quantitatively greater influence on measured ¹⁴C ages than 415 416 very old OC, such high ages for the biogenic OC fraction seem unrealistic and would mean that almost no young OC enters the sediment. Given the high ¹⁴C ages, it can therefore be assumed that some of 417 418 the OC in the sediments comes from sedimentation as geogenic OC.

419

420 4.1.1 Site dependent contents of GOCRegarding the calculated contribution of GOC to OC in the sediments, the assumed range of biogenic ¹⁴C ages from 1,000-4,000 years BP was within the typical 421 range for ages of dissolved OC leaching from soils (Artinger et al., 1996; Jia et al., 2019). 422 423 Nevertheless, the range of 1,000-4,000 years BP did not greatly influence the range of calculated 424 sedimentary contribution from GOC, especially for Loess (~ 6 % difference) and Red Sandstone (~ 9 425 % difference). The calculated GOC contribution for Miocene Sand was comparatively low (29-77 %) 426 compared with the contribution for Red Sandstone and Loess, especially at 5 m depth. Compared with 427 the Red Sandstone, this could be due to deep biogenic carbon inputs, such as roots and root exudates from the trees (Angst et al., 2016; John et al., 2016; Kirfel et al., 2017; Tückmantel et al., 2017), since 428 429 the loosely bedded Miocene Sand allows for deep infiltration compared with the Red Sandstone site 430 with its shallow bedrock as a root-restricting layer (Schneider and Don, 2019). Although it was not 431 possible to define the exact rooting depth for Miocene Sand, this depth probably does not exceed 4 m, according to Schenk and Jackson (2005). Therefore the increase in sedimentary contribution from 5 to 432 8 m depth of Miocene Sand could be due to the decreasing influence of roots and root exudates at 8 m 433 434 depth.

In Loess, the low ¹⁴C ages at 2-4 m depth (2,200-2,770 years BP) in contrast to a ¹⁴C age of 4,413 435 years BP at 0.74 m depth were surprising. This might indicate past anthropogenic activities or erosion-436 437 driven material movement that might have led to a mixing of the upper part of the profile. Furthermore, the modern ¹⁴C signature at 21 cm depth could be due to the plough layer at the Loess 438 site mixing the upper 30 cm. Nevertheless, at depths below 4 m the high ¹⁴C ages of OC in the 439 sediments indicated a large proportion of GOC. This could be due to different sedimentation periods, 440 soil forming and also soil burial processes (Chaopricha and Marin-Spiotta, 2014) that took place 441 during the Pleistocene. These processes can lead to the presence of buried layers in Loess with varying 442 443 amounts of rather recalcitrant OC, as shown by Marin-Spiotta et al. (2014). For the investigated Loess 444 site, different sedimentation and soil forming processes can be expected due to the presence of 445 completely different material in the cores in terms of colour and measured OC contents. For example, 446 the very dark Loess at 4 m depth with its high OC content support the assumption of sedimentation 447 circumstances that favoured the accumulation and preservation of OC. This is in accordance with 448 Jordan and Schwartau (1993) who investigated the same site and assigned the different layers to 449 specific Pleistocene sedimentation periods.

In summary, the contribution of GOC to sedimentary OC was substrate dependent. A loosely bedded sediment like Miocene Sand with extremely low concentrations of OC could be more prone to infiltration of biogenic OC and dilution of GOC. This resulted in contributions of biogenic OC to the sediments of about 50 %. In contrast, the Loess site with comparatively low infiltration rates or the Red Sandstone site with reduced possibilities for deep rooting seemed to contain relative constant contributions from GOC of around 80 %.

The GOC contribution within the sediments was not found to increase with soil depth. This is in 456 contrast to the results of Frouz et al. (2011) which showed that different sediment types from a 457 458 Miocene clay sediment had higher weight-based carbon contents at 150 m compared with 30 m depth. However, a comparison with the present study is difficult since Frouz et al. (2011) did not distinguish 459 between the geogenic and biogenic OC fractions, and OC contents were much higher (28-112 g kg⁻¹ 460 dry mass compared to 0.008-10 g kg⁻¹ dry mass in our results). However, it also underlines the 461 importance of different sedimentation processes for the amount and depth distribution of OC in 462 463 sediments and sedimentary rocks. 4.1.2 Geogenic OC in Miocene Sand and Red Sandstone compared 464 with other studies

465 It is hard to compare the weight-based amounts of GOC in terrestrial sediments since most studies in 466 this field rarely determine the amounts of OC in terrestrial sediments, or presume that sandy 467 sediments, for example, do not contain large amounts of OC (Artinger et al., 1996). Quite high 468 amounts of OC have been found in the skeleton part of different soils on sandstones with 0.61-1.97 t ha⁻¹ (Corti et al.,2002). Nevertheless, they mentioned the possible strong influence of organic 469 substances from the soil solution without quantifying it, and did not directly investigate OC in the 470 sediments. Additionally Copard et al. (2007) assumed an OC amount of 2.4 g kg⁻¹ from an unknown 471 source for all sandy sediments in a global storage modelling approach for the first metre of sediments. 472 This would fit with the amounts of around 1 and 5 mg OC g^{-1} found by Krummholz et al. (1997) for 473 474 Dakota sandstone layers at > 180 m depth, but is much greater than the median GOC amount in Red Sandstone (0.2 g kg^{-1}) and Miocene Sand (0.1 g kg^{-1}) in the present study. 475

476 *4.1.3 Geogenic OC in Loess compared with other studies*

Loess deposits are relatively well investigated because they provide a record of paleoenvironmental
conditions (Hatté et al., 1998; Head et al., 1989; Murton et al., 2015; Wang et al., 1996). The median
amount of 0.27 g kg⁻¹ from the present study was low compared with studies by Hatté et al. (1998),

Wang et al. (1996) and Strauss et al. (2012). Hatté et al. (1998) investigated 20-m deep loess deposits 480 in the Rhine valley and found OC contents between 1.0-8.6 g kg⁻¹, Wang et al. (1996) investigated 12-481 m deep loess deposits in China and found OC contents of 31.2 ± 30.5 g kg⁻¹ and Strauss et al. (2012) 482 found OC contents of 15 ± 14 g kg⁻¹ in Yedoma loess deposits in Siberia. This shows that the deposits 483 from the site in the present study stored comparatively low GOC contents, although most of the above-484 485 mentioned studies do not distinguish between a biogenic and geogenic OC pool. Nevertheless, as also shown in the present study, Loess had a high OC content compared with other sediments.. This was in 486 487 line with highest OC contents in subsoils at the Loess site and may indicate the importance and 488 contribution of bedrock OC to subsoil OC.

489 4.2 Is sedimentary derived organic carbon biodegradable?

490 The incubation experiment revealed a mineralisation of OC within the sediments with values between 0.1 and 3.4 % of total OC being mineralised after one year, assuming a constant mineralisation rate. 491 492 The incubation temperature of 20 °C should therefore be considered to be above the typical mean 493 temperatures in the subsoils. For subsoils with comparable climatic conditions, Wordell-Dietrich et al. (2019) found temperatures at 150 cm depth ranging from 4 to 14.4°C over a two-year period. 494 Assuming a Q_{10} value of around two for the assumable difference (Hamdi et al., 2013), the respiration 495 rate at typical subsoil temperatures would be roughly half that. A direct mineralisation of OC from 496 497 sediments is in agreement with several studies investigating the direct mineralisation from outcrops 498 (Copard et al., 2007; Horan et al., 2017; Petsch et al., 2000; Soulet et al., 2017; Seifert et al., 2001). 499 The difference to the present study is, that those studies observed this mineralisation when the 500 sediments were directly exposed to the surface or/and were part of a very rapidly eroding area. Thus 501 GOC from the sediments is already in touch with the atmosphere and inputs of recent vegetation. 502 However, Frouz et al. (2011) conducted an incubation experiment with sedimentary samples from OCrich Miocene clay sediments and found quite high respiration rate constants, with values between 3.5-503 12.3 mg CO₂-C g⁻¹ OC y⁻¹ during a 91 day incubation experiment. They attribute this to the prevailing 504 presence of aliphatic compounds in their samples being decomposed. Kieft and Rosacker (1991) also 505 found high respiration rates of sedimentary samples, with values between 0.9-9.5 mg CO₂-C g⁻¹ OC y⁻ 506 507 ¹, which they primarily attribute to the physiological status of the soil microbial community expressed as adenylate energy charge. Those results are in fairly good agreement with the respiration rate 508 constants observed in the present study (1 - 34.2 mg CO₂-C g⁻¹ OC y⁻¹). Meanwhile, compared with 509 the subsoil incubation experiments, the mineralisation found in the present incubation experiment was 510 511 quite low. For example, in subsoil incubation experiments at 20 °C, Wordell-Dietrich et al. (2017) found that between 5-9.5 mg CO₂-C g⁻¹ OC of OC are mineralised after incubation for 63 days, Wang 512 et al. (2013) report values between 5-15 mg CO₂-C g⁻¹ OC after 28 days, and Soucemarianadin et al. 513 (2018) report values between 10- and 12.5 mg CO₂-C g⁻¹ OC after 70 days. The difference between 514 515 the respiration rates observed in the present incubation experiment and the results from sedimentary and subsoil incubation experiments could be due to different microbial communities, OC quality and the physical connection between OC and potential decomposers. As shown in a meta-analysis by Colman and Schimel (2013), different microbial compositions, their abundance and the quality of OC, strongly affects respiration rates. This might also be indicative of extreme differences in respiration rates even within the same substrate and sample.

The results of the present study indicated that there was a considerable portion of biogenic OC in the sediments. The observed mineralisation could therefore be due primarily to the consumption of this biogenic OC part. Furthermore, the low mineralisation rate of Red Sandstone during the incubation as intact cores (Fig. A1) promoted the stability of GOC when it is part of the sediments. This might be due tothe low accessibility of OC in the sediments for microorganisms and the low availability of water due to a preferential flow through the sandstone (Swanson et al., 2006).

527 Altogether, the low mineralisation rates of the OC in the sediments might be caused by a lack of fresh 528 substrates and/or microorganisms that could enhance the degradation of OC (Fontaine et al., 2007). 529 Seifert et al. (2011) have shown that microorganisms are able to degrade sedimentary OC after the addition of glucose in black slate outcrops. Nevertheless, mineralisation of OC could only be observed 530 531 with the addition of water, which indicates the widely recognised assumption of the presence of an 532 active microbial community in the sediments (Bomberg et al., 2017; Joergensen and Wichern, 2018; 533 Magnabosco et al., 2018). Since fresh substrates were not added during the incubation, respiration 534 rates could be even higher if a fresh substrate-induced priming effect occurs. Furthermore the inherent and active microbial communities in the sediments might have assimilated ¹⁴C-free GOC into their 535 536 biomass, as shown by Schwab et al. (2019). Thus part of the labile OC pool in the sediments might also be derived from metabolised ¹⁴C-free microbial biomass. 537

538 The incubation experiment was unable to answer the question of whether GOC is mineralised when it 539 becomes part of the (sub-)soil. Assuming that a large part of the biogenic OC in the sediments was 540 mineralised during the incubation experiment, geogenic OC could still be preserved during soil 541 formation especially in the subsoils. This is in accordance with the indirect approach taken by Graz et 542 al. (2010) to determine the mineralisation of sedimentary OC when it becomes part of the subsoil. 543 They stated that 30 % of GOC resists degradation when it becomes part of the soil due to the results of 544 a quantitative palynofacies analysis of bedrock and soil samples. Hemingway et al. (2018) found that sedimentary OC directly exposed to the surface in a rapidly eroding tropical mountain area exhibits 545 considerable mineralisation down to 1 m below the surface, also leading to around 30 % of GOC 546 remaining in the soil. Based on ¹⁴C measurements they found out that on average 67 ± 11 % of the OC 547 fraction in the sediments could be lost during soil formation, but no distinction was made between 548 biogenic and geogenic OC fractions. This indicates that a microbial mineralisation of bedrock OC 549 550 takes place but may be partly restricted to biogenic OC.

Regarding the depth distribution of GOC in the sediments, the amount of GOC (in g kg⁻¹) did not 551 increase with depth, but there were clear differences. This reflects the sedimentation history with 552 553 different initial amounts of OC and degradation during sedimentation, which is particularly evident in the high amounts of GOC at 5 m depth in Loess. However, the contents of GOC, especially in Red 554 Sandstone and Miocene Sand, were within the same range for the whole depth. This might indicate 555 that degradation of GOC is not depth-dependent within the sediments. If there were a stronger 556 degradation of GOC with decreasing depth, a decreasing amount of GOC could be expected due to the 557 558 input of water, microorganisms and fresh nutrients from above. Furthermore, there was a relatively 559 constant contribution of biogenic OC within the sediments, meaning that if biogenic OC enters the 560 sediments with possibly degrading microorganisms, this biogenic OC might also be largely 561 mineralised. A study by Heitkötter et al. (2018) demonstrate effectively that degradation of OC in 562 subsoils is primarily limited to small hotspot areas. Also in sediments, Krummholz et al. (1997) show 563 that microbial communities can particularly be found in spatially discrete areas. Nevertheless, 564 Heitkötter et al. (2018) also show that microorganisms outside the hotspots can be activated when 565 substrate is supplied. Thus the bioavailability of GOC might be very site-dependent since root channels as microbial hotspots, for example, are less abundant and stable in sandy soils (Schneider and 566 567 Don, 2019). With regard to the sites investigated here, the solid Red Sandstone might only obtain 568 water in preferential flow paths (Swanson, 2006) leading to a comparatively stable OC pool in the sediment. Meanwhile for weathered, poorly structured sandy soils, prevalent matrix flow conditions 569 can be assumed (Flury et al., 1994). Thus the bulk GOC might be supplied with fresh substrates and 570 571 water from above more often than for the well-structured Loess soil with more frequent and stable preferential flow paths (Schneider and Don, 2019). This might lead to a lower accessibility and 572 573 therefore slower turnover rates of OC (Dungait et al., 2012).

- 574 The conclusion can be drawn that GOC can and will be degraded when it becomes part of the subsoil575 but probably to a comparatively limited extent.
- 576 *4.3 How much does GOC contributes to soil organic carbon?*

577 The contribution of GOC to soil OC stocks in this study was driven by the amount of OC in the soil, the amount of GOC in the respective sediment and also the turnover of GOC when it becomes part of 578 579 the soil. Our results revealed that despite differing between sediments, GOC content varied in a quite narrow range between 0.1 and 0.3 g kg⁻¹. The contribution of GOC to topsoil OC was negligible. 580 581 Assuming no degradation of GOC, the greatest possible contributions to total subsoil OC were found for Red Sandstone (~30 %) and the lowest for Miocene Sand (0.6 %). This was due to the range of 582 OC contents in the subsoils $(0.53 \text{ g kg}^{-1}-15.21 \text{ g kg}^{-1})$. When soil OC contents were low, the possible 583 contributions from GOC were high and vice versa For the investigated soils, the OC contents of 3 g 584 kg⁻¹ soil allowed for possible GOC contributions of between 5- and 10 %. For OC contents around 1 g 585 kg⁻¹ soil, a GOC contribution of between 10- and 20 % seemed possible. Thus, greater contributions 586

587 were made by GOC rich sediments such as Loess, while smaller contributions from sandy sediments. 588 In comparison, van der Voort et al. (2018) estimated the contribution of GOC in a soil derived from glacial deposits (flysch) at between 80-100 cm depth to be around 40 %. For a soil developed from a 589 590 poorly consolidated sedimentary rock (calcareous and shaly moraine), they calculate the contribution 591 of GOC to range from 20 % at 145 cm depth to 80 % at 310 cm depth. An attempt has also been made to fractionate subsoils to extract the most stable OC that may be derived from GOC. Paul et al. (2001) 592 investigated a soil developed on loess over till with 30 % of subsoil OC as a non-hydrolysable fraction 593 showing a ¹⁴C age of 13,000 years BP. They also concluded that this high ¹⁴C age can partly be 594 explained by a GOC fraction. These results indicate that deposits from the past glacial periods, such as 595 596 flysch or till in particular, have much greater potential for OC contributions from GOC, possibly due to the higher amounts of GOC in their sediments. Since only terrestrial sediments were investigated in 597 598 the present study, it should be noted that marine sediments or shales also contain much higher amounts of OC, up to 250 g kg⁻¹ (Hemingway et al., 2018; Petsch et al., 2000). Thus, the amount of GOC they 599 conatin and their possible contribution to subsoil OC stocks might therefore be much higher. 600

Nevertheless, the ¹⁴C ages of OC in the subsoil can be high in soils derived from igneous parent 601 602 materials without GOC (Rumpel et al., 2002), although even crystalline bedrocks contain microbial 603 communities (Purkamo et al., 2020). Furthermore, on a global scale, the ¹⁴C ages of soil OC are primarily driven by climatic conditions, clay content and age of the soil (Mathieu et al., 2015). 604 605 However, for terrestrial sediments with comparatively low amounts of GOC that started their soil development after the latest glacial period, a scale of possible contributions could be obtained when 606 the amount of OC is known. Thus, on a global scale, the high 14 C age of subsoils is not driven just by 607 the GOC fraction but the presence of GOC may greatly influence subsoil ¹⁴C. 608

609 5. Conclusions

In this study the amount of GOC in sediments and in the soil was analysed by radiocarbon dating. The 610 611 aim was to find out if GOC from different terrestrial sediments can have an influence on soil OC 612 stocks. This approach of estimating the GOC contribution to soil OC showed that common and abundant terrestrial sediments with low amounts of sedimentary OC can make a considerable 613 contribution to subsoil OC stocks. One fraction of OC in the sediments is of geogenic origin and could 614 therefore influence measured ¹⁴C ages in soil, particularly in subsoils. Subsoils are known for their 615 high ¹⁴C ages and slow turnover rates and slow reaction to changing environmental condition. These 616 subsoil OC properties may partly be derived from the GOC in the subsoil. The sediments at the 617 investigated sites contained OC in a range of 0.1-0.3 g kg⁻¹,... These amounts alloweding for 618 contributions from GOC of between 10-30 % in subsoils, defined here as soil horizons ranging from 619 0.3 to 1.5 m depth. Incubation of sediments indicated that this geogenic contribution presents a quite 620 stable OC pool, especially for subsoils. Thus, even sediments with comparatively low amounts of OC 621 were also able to demonstrate the large contribution of GOC. 622

623 Data availability

624 The data will be made available on request

625 Author contribution

AD conceived of and designed the study, FK performed the sampling and analysis, and wrote the first
draft. All the authors contributed to generating and reviewing the subsequent versions of the
manuscript.

629 Competing interest

630 The authors declare that they have no conflict of interest

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