# Dynamics of carbon loss from an arenosol by a forest/vineyard land use change on a centennial scale

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Abstract. Few studies have focused on arenosols with regard to soil carbon dynamics despite the fact that they represent 7% of the world's soils and are present in key areas where food security is a major issue (e.g. in Sahelian regions). As for other soil types, land use changes (from forest or grassland to cropland) lead to a loss of substantial soil organic carbon (SOC) stocks

- 15 and have a lasting impact on the SOC turnover. Here we quantified long-term variations in carbon stocks and their dynamics in a 80 cm deep Mediterranean arenosol that had undergone a <u>forest to vineyard</u> land\_use <u>change</u> over <u>a</u> 100 years ago. Pairedsites of adjacent plots combined with carbon and nitrogen quantification and natural radiocarbon (<sup>14</sup>C) abundance analyses revealed a <u>C</u> stock of <u>53</u> t,\_ha<sup>-1</sup> in the 0-30 cm forest soil horizon, which was reduced to 3 t-\_ha<sup>-1</sup> after long-term grape cultivation. <u>Total organic carbon</u> in <u>the</u> vineyard was dramatically low, with around 1 g-C\_-kg<sup>-1</sup> and no vertical gradient as a
- 20 function of depth. <sup>14</sup>C showed that deep ploughing (50 cm) in <u>the</u> vineyard plot redistributed the remaining carbon both vertically and horizontally. This remaining carbon was old (compared to that of the forest), which had a C:N ratio characteristic of microbial <u>organic matter</u> and was probably stabilized within organomineral associations. Despite the drastic degradation of the OM pool in this <u>a</u>renosol, this soil would have a high carbon storage potential if agricultural practices, such as grassing or organic amendment applications, were to be implemented within the framework of the 4 per 1000 Initiative.

#### 25 1 Introduction

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Arenosol is one of the 30 soil groups in the FAO soil classification system. Arenosols account for about 7% of the world's soils and are found mostly under desert, tropical and Mediterranean climatic conditions. They are silty-sandy or sandy soils, with less than 35% by volume of coarse elements, exhibit no or partial diagnostic horizon and are generally 100 cm deep (FAO, 2014). Given their excessive permeability and low nutrient content, agricultural use of arenosols requires careful management.

The C concentration range in arenosols is wide, varying in topsoil (0-30 cm) from 100 g.kg<sup>-1</sup> (Andreetta et al., 2013) to 1 g.kg<sup>-1</sup> (Fourie et al., 2005; López-Piñeiro, 2013), with topsoil stocks ranging from 80 tC. ha<sup>-1</sup> (Marschner and Waldemar Wilczynski, 1991) down to 15 tC.-ha<sup>-1</sup> (Muñoz-Rojas et al., 2012). With the average for soils at the global scale being 80 tC.-ha<sup>-1</sup> (Mousset, 2014), arenosols belong to the soil groups with rather low organic matter content. In addition and as with other soil types, the

- 35 conversion from forest or grassland to cropland can lead to up to 50% native carbon loss in 10 years due to the acceleration of erosion, runoff and/or mineralization (Lal, 2004; Guillaume et al., 2021; Ramesh et al., 2019; Fourie et al., 2005; López-Piñeiro, 2013). Cropped arenosols that have lost a large percentage of their pre-cultivation SOC thereby represent a large potential sink for C uptake through the adoption of proper management strategies in the framework of the 4 per 1000 objectives (Zomer et al., 2017). Futhermore, arenosols are present in key areas for future food production to meet food security objectives
- 40 (FAO, 2018). Understanding <u>the</u> carbon dynamics in arenosols is therefore a <u>significant</u> societal challenge. <u>H</u>owever, few studies to date have focused on C dynamics for this type of soil (Kögel-Knabner and Amelung, 2021). Soils in Mediterranean climatic <u>condition</u> that have inherited a long history of viticulture <u>are</u> represent<u>ative of</u> situations where land use is likely to have affected C dynamics in a very significant way since vineyards <u>are among the most degraded</u> agricultural crop systems (Giagnoni et al., 2019; Panagos et al., 2015). Nevertheless, SOC studies in vineyards have received
- 45 less attention as compared to arable and pasture systems (Payen et al., 2021), while viticulture is now a major agricultural growth sector under Mediterranean climatic conditions worldwide (Eldon and Gershenson, 2015). High C losses in Mediterranean vineyards are due to accelerated mineralization, decreased nutrient content, topsoil compaction and reduced water infiltration capacity, enhanced soil erosion rates, accumulation of metals and organic pollutants, and associated loss of soil biodiversity (Bogunovic et al., 2019; Bordoni et al., 2019; Eldon and Gershenson, 2015; Ferreira et al., 2020, 2018;
- 50 Kratschmer et al., 2018). These degradations are a result of traditional wine-growing practices which involve frequent tillage to minimize weed cover and soil compaction, postharvest removal of crop residues, as well as high mineral fertilizer and phytopharmaceutical compound application rates (Ferreira et al., 2020).

In order to study the effects of long-term vineyard-use on the C dynamics of a soil, it is thus relevant to: (1) compare a long-term vine-cultivated soil with its undisturbed native vegetation-cover counterpart (paired-site strategy, as defined by Eldon

- 55 and Gershenson (2015)) and, (2) use a geochemical timescale proxy (C isotopes). A rigorous site pair is defined as two plots with different uses on the same soil before land-use, under the same climatic conditions, on the same bedrock, and on a flat landscape. However, as these conditions are hard to meet, few studies have been carried out on pairs of soils in strict compliance with the above criteria, let alone over a long period of time to assess significant differences in carbon content between cultivated and forest soils. In the metanalysis of Eldon and Gershenson (2015), for example, the study times did not exceed 50 years, i.e.
- a time scale that seems limited in the case of vines where replanting periods are about 70 years. Concerning the timescale proxy, <u>C isotopes have been</u> used in many studies to study <u>C dynamics at the profile scale</u>. For example, Balesdent et al. (2018) studied paired sites with a change in vegetation from  $C_3$  to  $C_4$ , or vice versa, to assess the age of deep carbon stocks. This is an efficient method but only applicable to specific conditions (difference in <sup>13</sup>C isotopic signature between two successive vegetation types). Otherwise <sup>14</sup>C may be applicable to any system to assess the impact of cultivation

- on carbon dynamics at the decadal (or longer) scale as it is a function of the carbon age (Trumbore, 2009). Studies that used <sup>14</sup>C in a paired soil context <u>showed that</u> cultivation mainly affects young (short turnover) carbon pools <u>in topsoil</u> by promoting their mineralisation. Yet more stable (long turnover) carbon pools may also be impacted via their transfer to carbon pools with faster turnover (Poeplau and Don, 2013), thus leading to overall ageing of soil organic matter (OM), at least <u>in topsoil</u> (Wang et al., 1999). However, likely due to the high cost of <sup>14</sup>C analysis which only allow for a single measurement, studies that also
- focus on the effect of agricultural practices at the scale of soil layers remain limited (Anon, 2020; Lawrence et al., 2020)(Chiti et al., 2016; van der Voort et al., 2016)—these samples are often pooled into a single composite sample to overcome heterogeneity issues (Jiang et al., 2020).

The present study was therefore carried out to highlight the impact of the long-term conversion (>100 yr) of a forest to a vineyard on the C dynamics at the profile scale, while focusing on an arenosol under a Mediterranean climate. We hypothesized

- 75 that the combination of arenosol, vineyard and conventional practices would, overall, have a major impact on C stocks and the dynamics of C remaining in the topsoil and subsoil. To test our hypothesis, we worked on paired soils, measuring carbon contents and stocks, vertical and intra-horizon heterogeneity of carbon, as measured by <sup>14</sup>C, and correlating the C:N ratio and radiocarbon (F<sup>14</sup>C). These parameters enabled us to: (1) determine how vineyard cultivation and deep ploughing impact carbon stocks and dynamics in a Mediterranean arenosol, at soil layer and entire soil profile scales, and (2) use this case study to
- 80 estimate, according to different calculation hypotheses, the time required for the vineyard soil to recover a C stock equivalent to that prevailing pre-cultivation.

#### 2 Materials and methods

#### 2.1 Study area

The study site was located at Plan de la Tour, in the Maures massif (France), under a Mediterranean climate: -3°C<T<sub>Winter</sub><18°C

- 85 and T<sub>Summer</sub>>22°C, P<sub>driest month</sub><40 mm (Csb of the (World Maps of Köppen-Geiger climate classification)). The site selection process was carried out in 7 successive stages. (1) In the French Mediterranean area, a granitic pluton outcrop was sought to ensure the presence of arenosols (Figure A7). (2) In the Plan-de-la-Tour granite area, places with adjacent vineyard and forest plots were identified on the basis of satellite images. (3) To ensure that the forest C dynamics were representative of a forest pedogenesis process and not the result of recent afforestation, we selected only sites that were already forested in the 1800s</p>
- 90 (Napoleonic land register 1808-1848, and Ordnance Survey map, 1820-1866, see Figure A8). (4) Among these sites, we only selected those with a comparable topographic situation for the two land uses, ideally with the flattest possible landscape (using topographic map at 1:25,000 scale) in order to minimize differences in C dynamics that could result from differential erosion between vineyards and forests. (5) On the basis of the field work on the 5 sites selected according to the above criteria, we selected a site ("Les Brugassières") according to its accessibility and sampling authorizations. (6) A structural analysis, as is
- 95 <u>conventionally done in pedology studies (e.g. Humbel, 1987)</u> was performed to identify areas within the plots where the soil had undergone an identical pedogenic evolution process prior to vine planting. The sampling zones were selected with (i)

relatively deep soil (about 80 cm), (ii) equivalent soil depths for the two land uses, and (iii) with a short distance (less than 20 m) between the forest and vineyard sampling zones. (7) Finally, we performed a screening (0-30 cm topsoil layer) to assess the homogeneity of the total organic carbon contents in the vineyard plots and adjacent forest. This selection process eliminated

100 <u>all soil variation factors other than the land use and agricultural practices at the soil profile scale (according to a paired-site strategy, as defined by Eldon and Gershenson, 2015).</u>

The soil was a poorly differentiated arenosol on granite. An analysis of aerial photographs and cadastral maps (from 1813 to present day) showed that these two plots had a history of continuous soil use for at least ~100 years in the case of the forest (with an age of 91 years, as measured by dendrochronology on a cork oak) and more than 150 years in the case of the vineyard

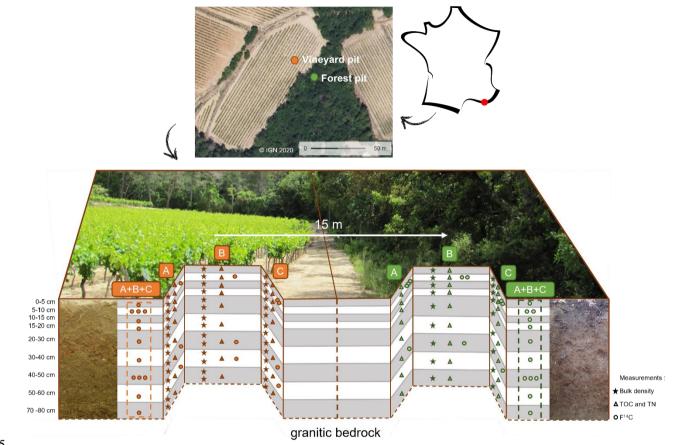
105 (Fig. A3). Additional field work ruled out the effects of terracing at the selected sampling sites. Concerning practices, the vineyard plot had undergone vine uprooting and deep ploughing (~50 cm) every 70 years on average. The last ploughing was carried out between 1998 and 2003. The soil was bare between rows (Figure 1 and Fig. A3).

#### 2.2 Sampling

Two pits were dug down to the underlying granite parent rock: the forest pit (43°19'37.35 "N, 6°32'12.89 "E) was 70 cm deep and the vineyard pit (43°19'37.74 "N, 6°32'11.90 "E) was 80 cm deep (Fig. 1). The pits were 15 m apart. The <u>soil particle-size</u> and mineralogy were similar at both sites (Fig. B<u>1</u> and Fig. B<u>2</u>). Three faces were sampled per pit (A, B and C). <u>9 layers were</u> <u>sampled on each face. 100 ml s</u>oil cylinders were taken from the three faces in the vineyard pit and from two faces in the forest

pit to determine the bulk density. Above 20cm, in the forest soil, the water measurement technique was preferred over the

- cylinder technique due to the high abundance of tree roots. Bulk density samples were oven dried at 105°C for 3 days before weighing. The profile samples were air-dried (25°C) for 1 week, sieved (2 mm) and weighed to determine the proportion of coarse elements (CE). Fine soil samples were ground in a planetary mill (50 g for 5 min, including 1 min direction reversal, at 400 rpm) down to <200 µm and the sample were then quartered. For the <sup>14</sup>C analysis, a 3 g composite sample (i.e. a mix of 1 g of A, B and C) was prepared for each depth range. To test the intra-horizon variability in topsoil and subsoil, 5-10 cm and 40-50 cm samples in the forest and vineyard, as well as 50-60 cm samples in the vineyard (below the ploughing sole), were
- 120 selected for further analyses (Figure 1). This variability was used to extrapolate the variability at all depths in the vineyard and forest soils.



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Figure 1: Scheme of the pits and sampling in the Brugassières arenosol under a Mediterranean climate: left, vineyard site (orange symbols); right, forest site (green symbols). A, B and C represent the three different sampled sides of each pit. Symbols indicate the sampling and analysis for each sampled layer: stars, sampling in cylinders for bulk density; triangles, sampling for total organic carbon and total nitrogen (TOC, TN), granulometry and mineralogy; circles, sampling for analysis in 14C; (A+B+C) represent composite samples resulting from the mixture of samples from the three faces at equal proportions.

#### 2.3 Methods

#### 2.3.1 Carbon and nitrogen measurement, and stocks calculation

Approximately 50 mg of fine soil samples were weighted in tin cups for total organic carbon and nitrogen measurements by

135 dry combustion with an elemental analyzer (NF ISO 10694 and 13878, respectively). Soil organic carbon stocks (SOC stock,  $\underline{tC.ha^{-1}}$ ) were calculated <u>as:</u>

$$SOC \, stock_i = BD_i \, (1 - CE_i) \times TOC_i \times e_i \div 10 \tag{1}$$

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$$SOC \ stock_n = \sum_{i=1}^n SOC \ stock_i$$
 (2)

Herer <u>i</u> is the considered as the layer and <u>n</u> is the number of layer increments, <u>TOC</u> is carbon concentration in fine soil (gC.kg<sup>-1</sup>). <u>BD</u> is the bulk density (g.cm<sup>-3</sup>), <u>CE</u> is the proportion of coarse elements (0 < CE < 1), and <u>e</u> the layer thickness (cm) (Poeplau et al., 2017).

A correction (equation <u>3</u>) was then applied to compare carbon stocks at equivalent mass and thus eliminate differences in bulk density between the two sites for the same depth (Ellert and Bettany, 1995; Poeplau and Don, 2013; Barré et al., 2020).

The reference soil mass was the layer with the heaviest density. A correction was applied for all cumulative increments from 0 to 60 cm (0-5, 0-10, 0-15, etc). The correction was performed as follows:

$$SOC \ stock_{corr} = SOC \ stock_i + \left(TOC_{i+1} * \frac{M_{heaviest} - M_i}{10}\right)$$
(3)

150 <u>Here *i* is the considered layer; SOC stock<sub>corr</sub> is the corrected cumulative SOC stock (tC.ha<sup>-1</sup>); SOC stock<sub>i</sub> is the uncorrected cumulative SOC stock (tC.ha<sup>-1</sup>);  $M_{heaviest}$  is the heaviest cumulative fine soil mass (g.cm<sup>-2</sup>) at both sites;  $M_i$  is the cumulative fine soil mass (g.cm<sup>-2</sup>) and TOC<sub>i</sub> is the fine soil carbon concentration of the underlying layer.</u>

#### 2.3.2 Radiocarbon dating

- 155 The <sup>14</sup>C contents were <u>measured in fine soil using the Mini Carbon Dating System ECHoMICADAS (Synal et al. 2007, Tisnérat-Laborde et al., 2015). Soil samples were weighed (20-200 mg) in tin capsules and converted into CO<sub>2</sub>. Two measurement modes (solid source or gas source) were used. (1) The solid source was used for C-rich samples (TOC>4 gC.kg<sup>-1</sup> to achieve a C mass of about 1,000 µg). CO<sub>2</sub> was reduced to C in the presence of H<sub>2</sub>, using automated graphitization equipment (AGE3) connected to an elemental analyzer (EA) (Wacker et al., 2010). Pure graphite was then pressed in the presence of the presence of the presence of the pressed in the presence of the pressed in the presence of the pressed in the pressence of the pressed in the pressence of t</u>
- 160 <u>ultrapure iron into a target to be introduced in the solid source. (2) The gas source required less C (30-140 µg) and could be used for both C-rich and C-poor samples. CO<sub>2</sub> was directly injected into the ECHoMICADAS gas source through the gas ion source interface (GIS) (Ruff et al. 2010) connected to an EA. The radiocarbon data are expressed in modern F<sup>14</sup>C fraction, as recommended by Reimer (2004). The range of variation of the analytical error, expressed as F<sup>14</sup>C, was between 0.002 and 0.014 and decreased with increasing carbon mass (Fig. <u>C</u>1). The difference between the highest and lowest F<sup>14</sup>C values for the</u>
- 165 same depth is expressed by  $\Delta F^{14}C$ . Many authors have used the  $\Delta^{14}C$  or conventional radiocarbon age to express <sup>14</sup>C (Lawrence et al., 2020) <u>;</u> the data expressed in  $\Delta^{14}C$  and conventional age are <u>thus also</u> shown in <u>the supplementary information</u> to facilitate comparison <u>with the literature</u>. All equations for the different units can be found in appendix <u>C</u>. <u>Due to the high analytical cost</u>, we opted to use composite samples for all depths: we thus obtained a mean <sup>14</sup>C value (mean of profils A, B and C). However, the composite samples did not enable us to determine the variability in <sup>14</sup>C at the scale of the

170 soil layer. We estimated this variability in 3 layers: a C-rich topsoil layer (5-10 cm) in the forest and its equivalent depth in the vineyard, a C-poor subsoil layer in the ploughed horizons (40-50 cm), and a layer below the ploughing horizon for which only the soil in the vineyard was measured (50-60 cm) (in view of the 5-10 and 40-50 cm results in the forest, we did not expected that there would be any variability in the forest 50-60 cm F<sup>14</sup>C).

## 2.3.2 Statistical analyses

- 175 <u>Statistical analyses were performed using the R statistical environment. A Student's-t test was used to compare, depth by depth, the TOC between vineyard and forest soils. This test is applicable if the variances are in the same order of magnitude. We therefore performed the test on log<sub>10</sub>(TOC) to have similar orders of magnitude in the variances between vineyard and forest soils. A Student's t-test was also used to compare the C:N levels between the vine and forest soils. We used Spearman's test (no data normality) for the linear regression between F<sup>14</sup>C and C:N.</u>
- 180 We tested the intra-layer variability with a limited number of data, by applying a permutation test on the RMS<sub>vinevard</sub>/RMS<sub>forest</sub> ratio (RMS being the residual mean squares), calculated on the F<sup>14</sup>C data. We repeated 1,000 times a permutation test of the RMS ratios between forest and vineyard (simulation). The RMS ratio allowed us to compare the degrees of variance between the forest and vineyard findings, which we then compared to the observed ratio value. The permutation test allowed us to test whether the ratio result was significant or not (Manly, 2006).

#### 185 **3 Results**

## 3.1 Carbon content, C:N ratio and stocks

The results of the carbon content profiles are presented in <u>Fig. 2a and Table D1</u>. Under the forest, <u>the</u> carbon content and variability were high <u>in topsoil</u>, with <u>32-51</u> gC.kg<sup>-1</sup> in the 0-5 cm layer (<u>mean 42.4 ± 9</u> gC.kg<sup>-1</sup>), but it decreased with depth down to 1.89-2.70 gC.kg<sup>-1</sup> in the <u>50-60</u> cm layer (<u>mean 2.3 ± 0.4 gC.kg<sup>-1</sup></u>). Under vines, the carbon content was comparatively very <u>low</u> and equivalent in the <u>A</u>. <u>B</u> and <u>C</u> profiles throughout the depth. <u>In topsoil</u> (0-5 cm), the TOC ranged from 0.9 to 2.4 gC.kg<sup>-1</sup> (<u>mean 1.8 ± 0.8 gC.kg<sup>-1</sup></u>), and at depth (<u>50-60 cm</u>) it ranged from 0.8 to 0.9 gC.kg<sup>-1</sup> (<u>mean 0.9 ± 0.05 gC.kg<sup>-1</sup></u>). The TOC values under vines were <u>thus</u> extremely low compared to those under the forest (<u>p<0.01</u>, <u>Table D2</u>) and this depletion was even observed <u>in subsoil</u>. Under the forest, the average C:N ratio was high, *i.e.* around 16 in the 0-5 cm layer, and decreased with depth to 10 in the 60-70 cm layer (Fig. <u>2b</u>). The C:N ratio under vines was <u>significantly different from C:N</u> in the forest

horizon (p<0.1, Table D3) in the 0-50 cm layer. Beyond this depth, the vineyard profile became similar to that under the forest. Finally, stocks in the 0-30 cm layer in the forest soil contained 53.3 tC\_ha<sup>-1</sup> while the vineyard soil contained only 3.3 tC\_ha<sup>-1</sup> (Fig. 2c).

## 3.2 Radiocarbon dating

The radiocarbon profile results are presented in Fig. 2d (Table D4, Fig. D1 and Fig. D2). Young carbon, *i.e.* younger than the

- 1960 bomb peak (F<sup>14</sup>C>1), was detected in the forest topsoil profile. The carbon age then increased with depth (F<sup>14</sup>C<1 around 40 cm). The forest soil profile indicated a conventional undisturbed soil (Jreich, 2018; Mathieu et al., 2015; van der Voort et al., 2016). It showed a 'belly' shape curve between 5 and 20 cm depth, which corresponded to penetration of the <sup>14</sup>C signal of the bomb peak in the profile. Concerning the variability in a single soil layer, F<sup>14</sup>C ranged from 1.095 to 1.124 (ΔF<sup>14</sup>C=0.029) at 5-10 cm. Meanwhile, at 40-50 cm. depth, F<sup>14</sup>C ranged from 0.974 to 1.005 (ΔF<sup>14</sup>C=0.031).
- 205 Conversely, the vineyard profile revealed the presence of old carbon from the <u>top</u> to the bottom of the pit ( $F^{14}C=0.893$  at the <u>top</u> and 0.990 at depth), despite the heterogeneity within the horizons (one point with an  $F^{14}C>1$ , at 40-50 cm). The variation pattern in the profile was not progressive from the <u>topsoil</u> to the depth, contrary to the pattern noted in the forest profile. Under vines, the intra-horizon variability was much more marked than under the forest. In the 0-10 cm layer,  $F^{14}C$  ranged from 0.880 to 0.969 ( $\Delta F^{14}C=0.089$ ), and from 0.909 to 1.081 ( $\Delta F^{14}C=0.172$ ) at 40-50 cm depth.

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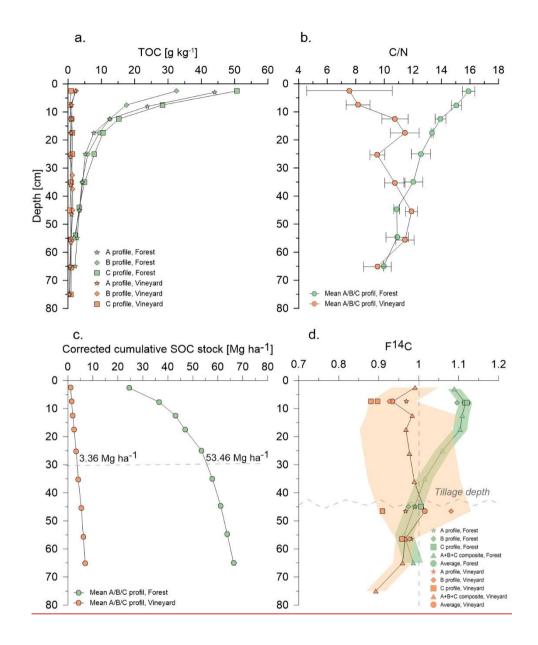


Figure 2: TOC variations (a), C:N ratio (b), cumulative stocks corrected for equivalent mass (c) and F<sup>14</sup>C profiles (d) as a function of depth, under vines (orange) and under forest (green), for an arenosol under a Mediterranean climate. The F<sup>14</sup>C measurement variability (d) is represented by green (forest) and orange (vine) bands.

#### **4** Discussion

#### 4.1 Comparison with Mediterranean arenosols

Under the forest, the TOC profiles (22 ± 5 gC.kg<sup>-1</sup> in the 0-20 cm layer and 3.94 ± 0.25 gC.kg<sup>-1</sup> in the 30-50 cm layer) obtained
for topsoil and subsoil were comparable to those obtained for other arenosols under Mediterranean climatic conditions (Figure 3a) (Andreetta et al., 2013; Caravaca et al., 2002; Fierro et al., 2007; Pinzari et al., 1999; Vittori Antisari et al., 2016). However, to our knowledge, very few data are available beyond 30 cm soil depth. For the vineyard, we only identified 5 references of studies concerning arenosols in Mediterranean climatic conditions (Conradie, 2001; Fourie et al., 2005; López-Piñeiro, 2013; Nogales et al., 2019; Okur et al., 2009). The Brugassières arenosol was found to be among the soils with the lowest organic carbon contents that were comparable to those of the soil studied here, both in the toposoil and at depth (Fourie et al., 2005; López-Piñeiro, 2013). The arenosol in this study, although very depleted in C, does not seem to represent a unique case of organic matter (OM) depletion after arenosol vineyard cultivation.

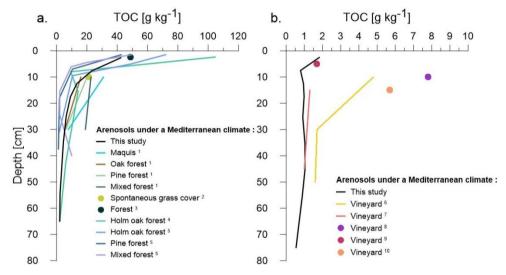


Figure 3: TOC comparison between the Brugassières arenosol and other forest arenosols (a) and vineyard arenosols (b) under a Mediterranean climate (1) Pinzari et al., (1999); (2) Caravaca et al., (2002) ; (3) Fierro et al., (2007) ; (4) Andreetta et al., (2013) ; (5) Vittori Antisari et al., (2016) ; (6) Conradie, (2001) ; (7) Fourie et al., (2005) ; (8) Okur et al., (2009) ; (9) López-Piñeiro, (2013) ; (10) Nogales et al., (2019). Data available in Table E1 (details about Mediterranean climate Table E2).

4.2 Drastic carbon stock loss: a combination of land use change / agricultural practices / unfavourable soil texture

These very low carbon contents in the vineyard resulted in a 12-fold lower carbon stock in the vineyard than in the forest throughout the profile (*e.g.* in the 0-30 cm layer, the SOC stock was  $3.3 \text{ tC.ha}^{-1}$  in the vineyard compared to  $53.5 \text{ tC.ha}^{-1}$  in the forest) (Figure 2c). Arenosol carbon stocks under the forest, in the 0-30 cm layer, were lower than stocks under the forest irrespective of the soil type (80 tC.ha<sup>-1</sup>, Mousset, 2014). The difference between the national forest average and that of the studied forest was:  $80-50.8 = 29.2 \text{ tC.ha}^{-1}$ . The difference between the national mean for French vineyards (30 tC.ha<sup>-1</sup>, Mousset, 2014).

240 <u>2014</u>) and that of the studied vineyard was:  $30_{3_2} = 26.8$  tC.ha<sup>-1</sup>. Consequently, arenosols had about 30 tC\_ha<sup>-1</sup> less than the French average under both forests and vines.

Cultivation in the vineyard plot resulted in a very high carbon stock loss throughout the entire depth: 94% in the 0-30 cm layer and 76% in the 30-60 cm layer. Although this carbon stock loss phenomenon has already been widely reported (Guillaume et al., 2021; Ramesh et al., 2019), it has generally been found to be around 50% in topsoil during a forest (or grassland) to vineyard transition under all climatic conditions (Carlisle et al., 2006; Eldon and Gershenson, 2015). Moreover, contrary to our findings here, the loss is usually much greater in topsoil than in subsoil layers, ranging from 30 to 63% on average in the 30-100 cm horizon (Batjes, 2014; Poeplau and Don, 2013). However, if we focus the comparison on arenosols under a Mediterranean climate, losses (in TOC) during a natural vegetation to vine transition can reach 85% in the 0-20 cm layer over a 1-year period (Caravaca et al., 2002). The soil carbon loss noted in this study thus resulted in extremely high carbon loss

after more than 150 years of grapevine cultivation, which does not seem to be out of line with observations described in the

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arenosol literature.

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This extreme carbon loss throughout the cultivated soil profile could be explained by a combination of four aggravating factors at the Brugassières site: (1) The initial disturbance of the arenosol, due to the forest to vineyard land\_use change in the 19<sup>th</sup> century (Caravaca et al., 2002; Tsozué et al., 2020); (2) the absence of vegetation cover (apart from vines) for more than 150 years was probably also an important factor. Carbon inputs were almost nil in topsoil (soil kept bare, Figure 1). Deep inputs were limited to the depth of the grapevine root system, while the vine plants were uprooted every 70 years. However, the age of the carbon distribution as a function of depth proposed by Balesdent et al. (2018) shows that almost half of the carbon in a soil is on average younger than 150 years at the soil profile scale. Although this distribution concerns soils under tropical climates, the drastic long-term reduction of carbon inputs to the soil could likely largely explain the carbon stocks observed in the vineyard throughout the soil profile; (3) deep ploughing (50 cm), carried out every 70 years at the same time as the grapevine plant uprooting, was probably a third factor favouring carbon loss via accelerated SOC mineralisation; and (4) the arenosol texture, characterised by a low proportion of fine particles (<20 µm fraction, Fig. <u>B1</u>) is also an unfavorable factor for C storage within the mineral-associated OM pool.

#### 4.3 Intra-layer radiocarbon variability

Carbon spatial heterogeneity is generally not taken into account in soil studies on carbon dynamics using the <sup>14</sup>C proxy (van der Voort et al., 2016). Chiti et al. (2016) and van der Voort et al. (2016) showed that the intra-layer radiocarbon signature under forests is relatively homogeneous in topsoil and at depth. This finding is in line with our forest soil results (Figure 2d, Figure 4), where the low intra-layer F<sup>14</sup>C variability (represented by the standard deviation SD) in the forest soil was noted both in the 5-10 cm layer with a high carbon concentration (SD<sub>F14C</sub> = 0.008 ; TOC<sub>avemage</sub>=42.4 gC<sub>2</sub>kg<sup>-1</sup> and SD<sub>TOC</sub>= 9.1 gC<sub>2</sub>kg<sup>-1</sup>
and in the 40-50 cm layer with a low carbon concentration (SD<sub>F14C</sub> = 0.011 ; TOC<sub>avemage</sub>= 3.4 gC<sub>2</sub>kg<sup>-1</sup> and SD<sub>TOC</sub>= 0.1 gC<sub>2</sub>kg<sup>-1</sup>
Below the ploughing depth, low intra-layer variability was also observed in the vineyard soil (Figure 2d, Figure 4), (50-60 cm layer, SD<sub>F14C</sub> = 0.012\_; TOC<sub>avemage</sub>= 0.9 gC<sub>2</sub>kg<sup>-1</sup> and SD<sub>TOC</sub>=0.1 gC<sub>2</sub>kg<sup>-1</sup>). Thus, in a horizon undisturbed by agricultural

cultivation, <sup>14</sup>C showed little intra-layer variability on a metric scale, even when measurements were carried out on samples with a very low TOC (Fig. <u>1</u>a and Table D1).

- In cultivated systems, to our knowledge, no studies have reported measurements of intra-layer <sup>14</sup>C variability. Our findings therefore cannot be compared with those of previous studies. In comparison to undisturbed horizons, much higher intra-layer variability was observed at 5-10 cm depth (SD<sub>F14C</sub>=0.029) and within the 40-50 cm ploughing depth (SD<sub>F14C</sub>=0.058), with both layers being characterized by low total carbon (over 5-10 cm, TOC=0.79 gC.kg<sup>-1</sup> with SD=0.26 gC.kg<sup>-1</sup>; over 40-50 cm, TOC=1.03 gC.kg<sup>-1</sup> with SD=0.42 gC.kg<sup>-1</sup>). The variance under vines was significantly different from that under forest, both in the topsoil (p<sub>permutation test</sub>=0.02) and subsoil (p<sub>permutation test</sub>=0.01) (Fig. D3, Fig. D4). Furthermore, the F<sup>14</sup>C measurements at 40-50 cm depth in the B profile and in vineyard pit composite soils had a post-bomb value (F<sup>14</sup>C<sub>mean</sub>=1.001), which was higher
- than that obtained in the forest soil (F<sup>14</sup>C<sub>mean</sub>=0.990). At 40-50 cm depth, and only for this horizon, OM in the vineyard was younger than that in the forest soil<u>for some samples</u>. This highly suggests that the variability in F<sup>14</sup>C measured between samples on sides A, B and C was a consequence of multiple ploughing whereby the soil is mixed vertically but also horizontally on a metric scale.

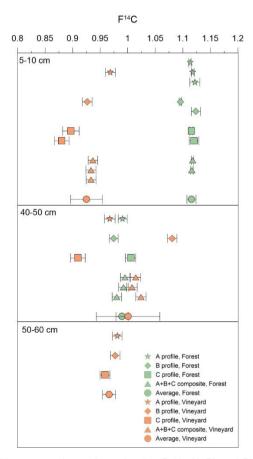


Figure 4: Comparison of intra-layer  $F^{14}C$  heterogeneity at three depths (5-10, 40-50 and 50-60 cm) in forest and vineyard soils.  $F^{14}C$  data were obtained for profiles A (star), B (diamond), C (square), composites A+B+C (triangle) and the average of these data (round),

in forest (green) and vinevard (orange) soils. Error bars represent the analytical error for the profiles A. B and C and the standard 290 deviation for the mean.

## 4.4 OM in arenosols : younger C than in other soil types

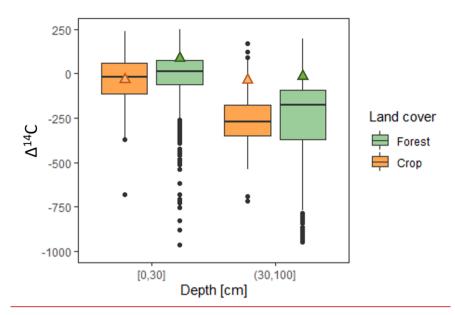
The  $\Delta^{14}$ C of soil profiles reported in 185 papers culled from version 1.7.8.2021-01-04 of the ISRaD database (Anon, 2020; Lawrence et al., 2020), under forest and cultivation, in topsoil (0-30 cm) and at depth (30-100 cm), are compared in Figure 5. The arenosol studied here had a higher  $\Delta^{14}$ C than the median  $\Delta^{14}$ C (all soil types combined), in topsoil and even more marked in subsoil layers, e.g. in topsoil, the  $\Delta^{14}C_{\text{forest}} = 84.88 \pm 22.5\%$  relative to a median around 7% from the literature review; and 295  $\Delta^{14}C_{crop} = -32.7 \pm 21.8\%$ , relative to a median of -20%. This was probably due to the lower fine particle (<2 µm) content than the overall average in the meta-analysis. Indeed, arenosols have few reactive mineral phases that stabilize OM in the long term, which is in line with the above discussion on stocks. The fact that the OM fractions were systematically younger than those generally described in the literature could thus be explained by the soil type (i.e. the fine fraction was minimal in the arenosol) and by the long cropland history (>150 years).



#### 4.5 Land-use impact on OM borne <sup>14</sup>C

In the ploughed horizon, with the exception of the 40-50 cm layer,  $\Delta^{14}$ C was always more negative in cultivated soils than in forest soils. Cultivation therefore led to carbon aging (by loss of the most recent carbon pool) to 40 cm depth. This impact of cultivation had already been highlighted in a ploughed horizon by Wang et al. (1999), where the carbon of a cultivated soil in

- the 0-30 cm layer was older than its equivalent in forest soils. This trend was also revealed in a meta-analysis (Figure 5, 305 cropland-soil n=34, forest-soil, n=151 papers). The median values confirmed that the carbon age of SOM was older in cultivated soils in both top and deep horizons. Cultivation affects the mean carbon turnover mainly by removing carbon from fast-turnover pools and mostly retaining slow-turnover carbon pools (Poeplau and Don, 2013). It is likely that these slowturnover OM pools are organic compounds associated with the mineral-associated OM (MAOMs) pool (Cotrufo et al., 2019).
- 310 Furthermore, the findings in the 40-50 cm horizon, with a younger post-bomb OM than all other horizons in the vineyard profile, showed that the full inversion tillage practiced effectively dragged surface OM down to 50 cm. Cultivating the deep ploughed arenosol under vines therefore led to: (1) loss of the young and poorly stabilised OM pools, and (2) redistribution of the remaining MAOMs throughout the ploughed horizon and, as shown in section 4.3, in a horizontally heterogeneous way.



315 Figure 5: Comparison of the Δ<sup>14</sup>C average at 0-30 cm and 30-100 cm depth between this study forest (green triangle), this study crop (orange triangle) and the Δ<sup>14</sup>C average from <u>185 papers</u>, <u>extracted using R software from the ISRaD database</u>, version <u>1.7.8.2021-01-04</u> (Anon, 2020; Lawrence et al., 2020). Black dots represent outliers. Crop-soil n=34, forest-soil n=151 papers. <u>The central value of the boxplot is the median</u>, the edges are the quartiles and the ends of the whiskers represent the maximum and minimum values. <u>50% of the observations are inside the boxplot</u>. Values outside the whiskers are represented by dots.

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## 4.6 Possible microbial origin of OM in the vineyard

The C:N profile of the forest soil was a classic profile, with a C:N ratio that tended to decrease with depth. This decrease reflects an enrichment in N of the SOM in connection with an increased proportion of the contribution of molecules of microbial origin (Cotrufo et al., 2013). The vineyard profile did not follow a similar trend but was the result, as for F<sup>14</sup>C, of disturbances linked to tillage. However, there was a positive linear correlation (R<sup>2</sup><sub>Speamnan</sub>= 0.78) between C:N and F<sup>14</sup>C: the microbial signature was higher in older SOM (Figure 6). This suggests that the ancient carbon in the soil was mainly borne by molecules originating from N-rich microbial metabolism and presumably stabilised within MAOM (Cotrufo et al., 2019; Kleber et al., 2015; Rumpel and Kögel-Knabner, 2011). The change in vineyard use associated with conventional practices (absence of inter-row cover crops and deep ploughing) thus only seemed allow maintenance of this small MAOM pool, to the

330 detriment of other less stable OM pools lost through erosion, leaching or mineralisation.

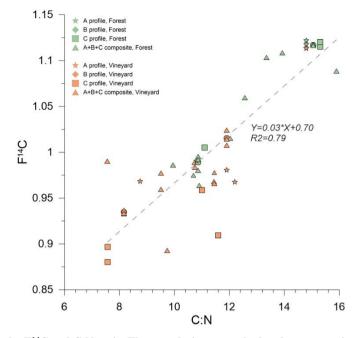


Figure 6: Correlation between the F<sup>14</sup>C and C:N ratio. The correlation was calculated on composite samples (F<sup>14</sup>C) and the average for the 3 profiles A, B and C (C:N), as well as on samples of profiles A, B and C only, from the forest (green) and vineyard (orange).

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#### 4.7 Is arenosol a good target for the 4 per 1000 Initiative?

To restore OM stocks in soils and meet the 4p1000 objectives, the land-use may be changed (cropland returned to grassland or forest), or cropland may be maintained by adopting practices that foster C storage, e.g. establishment of permanent grasslands, application of organic amendments, grassing of vinevards, etc. (Pellerin et al, 2019). Arenosols, whose carbon 340 stocks are very low in cultivated systems (Figure 2c), thus seem to be good candidates for the 4p1000 Initiative because they have a high C storage potential. Storage experiments conducted on arenosols measured an increase of 40.2 to 45.6 tC.ha<sup>-1</sup> and 39.4 to 49.0 tC.ha<sup>-1</sup> of carbon stocks in the 0-30 cm layer in 20 years following, respectively, cropland abandonment (+ 0.27 tC.ha<sup>-1</sup>.yr<sup>-1</sup>) and a change of grassland management (+ 0.48 tC.ha<sup>-1</sup>.yr<sup>-1</sup>) (Kazlauskaite-Jadzevice et al., 2019). In these experiments the annual increase in carbon stock was +5.9  $\frac{50}{20}$  and +9.8, respectively, *i.e.* more than 2-fold higher than the  $4\frac{1000}{1000}$  annual increase targeted by the  $4\underline{p}1000$  Initiative.

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In the case of the studied arenosols, the potentially achievable reference stock could be considered equal to the forest soil stock. In the 0-30 cm range, the C storage potential was therefore  $50 \text{ t.ha}^{-1}$  (Figure 2c). If we consider an annual C stock increase rate equivalent to that obtained by Kazlauskaite-Jadzevice et al. (2019) (about 8%), it could be calculated that an arenosol could recover this stock in <u>117</u> years under appropriate practices. If calculated differently, considering not the same storage proportion as Kazlauskaite-Jadzevice et al. (2019) but the same storage rate (a mean of +0.37 tC.ha<sup>-1</sup>.yr<sup>-1</sup>), an arenosol 350

could recover its C stock in 131 years of storage. Even if the system would probably not respond linearly in terms of C storage

rate, these simple calculations show that the additional C storage potential in cultivated arenosols is thus high, but the timing to recover a stock level equivalent to that of the forest is around a century. Although this timing is long in comparison to policy-defined C-neutrality urgency timetables, low-C cultivated arenosols are likely to represent a sustainable annual C sink upon the adoption of C storage practices—a sink that could exceed the 4p1000 targets.

## Conclusion

Land use change from a Mediterranean forest to a vineyard on an arenosol resulted in loss of <u>a very high proportion of</u> the soil's carbon throughout the entire depth of the soil profile: 93.7% less SOC <u>in topsoil</u> and 76.2% at depth. <u>Few papers in the literature showed</u> comparable levels of carbon under forests as well as in vineyards. The radiocarbon study highlighted the very high vertical homogeneity (as a function of depth) together with horizontal heterogeneity (intra-layer) of the carbon distribution, induced by deep ploughing. The carbon remaining in the <u>0-50 cm of the vineyard</u> soil layer was old stabilized microbial carbon that was, <u>for some samples</u>, mixed with younger carbon at depth. The study of <sup>14</sup>C data and the C:N ratio revealed a link between the degree of OM biotransformation by the microbial compartment and its age, *i.e.* F<sup>14</sup>C (old and stabilized carbon) decreased with <u>N enrichment</u>. Finally, arenosols are soils for which the adoption of C stocking practices can meet ambitious annual soil carbon storage objectives. The findings of this study thus generated fresh knowledge on the carbon dynamics of arenosols following a land-use change, with a view to application of the 4p1000 Initiative\_In the case of vineyards cover cropping is an effective carbon storage practice and economically interesting for farmers (Payen et al., 2021; Pellerin, 2019). The large area of land devoted to viticulture worldwide (7.45 Mha) means that the widespread use of this practice would be a significant step towards the adoption of carbon-storage practices on a global scale.

## Appendix A: Site identification

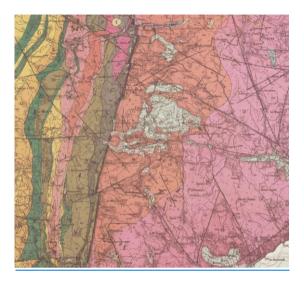
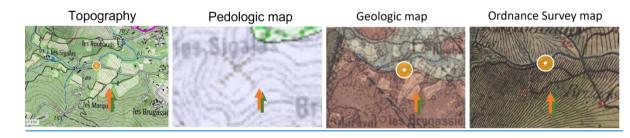
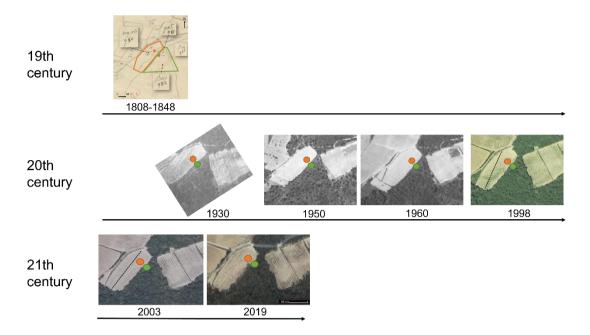


Figure A7: Geological map (source https://www.geoportail.gouv.fr/carte) of the granite of Plan de la Tour (Maures, South of
 France), represented by the north-south elongated red zone, in the center of the geological map (source https://www.geoportail.gouv.fr/carte)



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Figure A8: topographic, pedologic (Source: Soil reference system of the VAR), geological and Ordnance Survey map, 1820-1866 (source: https://www.geoportail.gouv.fr). Brown arrow=crop; green arrow=forest



385 Figure <u>A3</u>: History of land use at the "Les Brugassières" site from the 19<sup>th</sup> century to present day, through the study of old maps and aerial photos. The boundaries of the forest (green) and vineyard (orange) plots were shown on the Napoleonic <u>land</u> register (1808-1848, https://archives.var.fr). All aerial photos from the 20<sup>th</sup> century to present day were from the IGN *Remonter le temps* website (https://remonterletemps.ign.fr/). The pit locations are indicated by orange and green circles.

#### Appendix B: textural and mineralogical comparison of vineyard and forest soil profiles

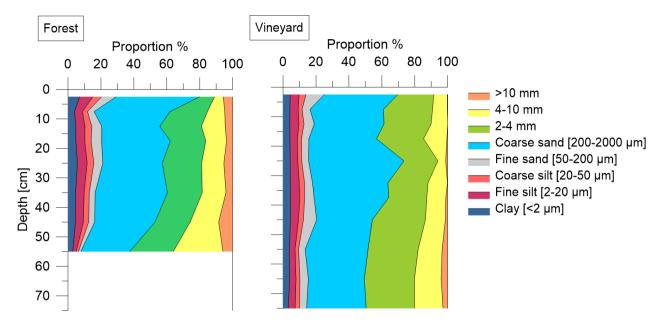
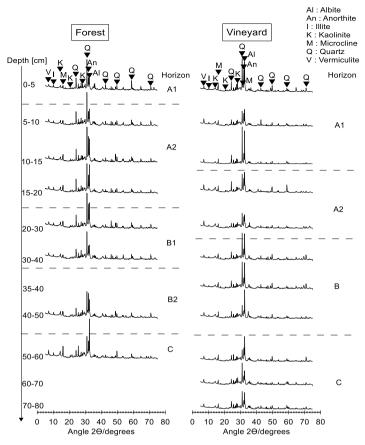


Figure B1: Soil particle-size distribution of the pit samples under the forest and under the vineyard, as a function of depth.
The eight particle-size fractions, clay <2 μm (dark blue), fine silt 2-20 μm (burgundy), coarse silt 20-50 μm (pink), fine sand 50-200 μm (grey), coarse sand 200-2000 μm (sky blue), 2-4 mm (green), 4-10 mm (yellow) and >10 mm (orange). The different fractions are expressed in % of the mineral phase. The particle size profiles in the forest and vineyard soils showed about 50% coarse sand and did not vary significantly with depth. There was no significant difference in soil particle-size distribution between the forest and vineyard plots.



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Figure B2: Forest/vineyard comparison of X-ray diffractograms as a function of soil depth. The rhombs indicate the peak considered and the letters above are the corresponding minerals. The mineralogy was determined by X-ray diffraction on powder samples, deposited on a silicon plate, and measured using a PANalytical X'pert PRO diffractometer, with a cobalt radiation source. The range of 20 was between  $5_{2}^{\circ}$  and  $75^{\circ}$ , with a step size of 0.033° and a measurement time of 5 h 10 min per sample. The forest and cultivated soil mineralogy is characteristic of a granitic bedrock with quartz, feldspar and secondary 410 minerals (illite and vermiculite), throughout the entire profile. The mineralogy was equivalent in both soils.

Appendix C: radiocarbon equations and analytical errors according the type of source used in ECHoMICADAS

415 The data expressed in  $\Delta^{14}$ C is shown in Fig. D1 to facilitate comparison with the literature findings, Eq.(A1):

$$\Delta^{14}C = (F^{14}C/\exp\left(((years - 1950)/(5730/ln2)) - 1) * 1000$$
(A1)

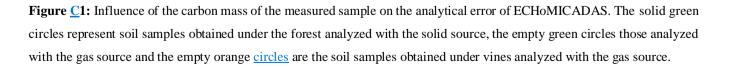
From the radiocarbon data, it is possible to access a relative age, expressed in years before present (BP). The starting date of the age scale is 1 January 1950, which was before the bombs peak, and corresponds to the first publications with radiocarbon dates. The BP age takes the radiocarbon decay equation into account and was calculated according to the Libby half-life of

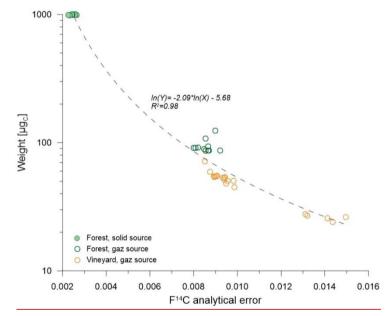
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5,568 years (Libby et al., 1949), Eq.(A2):

 $Age BP = -5568 * \frac{\ln(F^{14}C)}{\ln 2}$ (A2)





## 430 Appendix D: data

**Table D1:** Table of bulk density  $[g \text{ cm}^{-3}]$ , <u>coarse elements [%]</u>, <u>TOC</u>  $[gC_2 - kg^{-1}]$ , C: N ratio as a function of plant cover and depth. A,B,C are the pit profiles, BD is the bulk density, CE is the coarse elements, ± is the analytical error, X is the mean of the 3 A, B and C profiles and SD is the standard deviation.

	-			BD g.cm <sup>-3</sup>		CE %						g	-				
	Depth [cm]	Α	в	C	X SD	А	в	C	х	SD	А	В	C	х	SD	Α	В
Vineyard	0-5	1.55	1.63	1.61	$1.60 \pm 0.0$	24	30	29	28	± 4	2.17 ± 0.3	$2.38 \pm 0.3$	$0.93 \pm 0.2$	1.83 ±	± 0.8	7 ± 2	11 ± 3
	5-10	1.59	1.38	1.59	$1.52 \pm 0.1$	31	39	32	34	± 4	$1.05 \pm 0.2$	$0.79 \pm 0.2$	$0.53 \pm 0.2$	0.79 ±	± 0.3	9 ± 4	
	10-15	1.60	1.45	1.58	$1.54 \pm 0.1$	31	39	41	37	± 5	1.21 ± 0.2	$0.61 \pm 0.2$	$1.03 \pm 0.2$	0.95 ±	± 0.3	10 ± 4	
	15-20	1.60	1.39	1.56	$1.52 \pm 0.1$	33	43	39	39	± 5	$0.93 \pm 0.2$	$0.73 \pm 0.2$	$1.30 \pm 0.2$	0.99 ±	± 0.3	10 ± 5	12 ± 8
	20-32	1.57	1.48	1.51	$1.52 \pm 0.0$	48	27	30	35	± 11	$0.79 \pm 0.2$	$0.67 \pm 0.2$	$1.28 \pm 0.2$	0.91 ±	± 0.3	10 ± 6	
	32-40	1.51	1.38	1.49	$1.46 \pm 0.1$	34	36	42	37	± 4	$0.74 \pm 0.2$	$1.35 \pm 0.2$	$1.01 \pm 0.2$	1.03 ±	± 0.3	11 ± 7	12 ± 4
	40-53	1.52	1.45	1.41	$1.46 \pm 0.1$	34	46	38	39	± 6	$1.10 \pm 0.2$	$1.42 \pm 0.2$	$0.58 \pm 0.2$	1.03 ±	± 0.4	12 ± 6	
	53-60	1.70	1.39	1.46	$1.51 \pm 0.2$	38	48	29	38	± 10	$0.83 \pm 0.2$	$0.94 \pm 0.2$	$0.88 \pm 0.2$	0.88 ±	£ 0.1	12 ± 7	
	60-70	1.72	1.59	1.62	$1.64 \pm 0.1$	46	51	39	45	± 6	$0.53 \pm 0.2$	$0.69 \pm 0.2$	$0.92 \pm 0.2$	0.71 ±	£ 0.2	9 ± 7	
	70-80					40	49	76	55	± 19	$0.42 \pm 0.2$	$0.34 \pm 0.2$	$0.87 \pm 0.2$	0.54 ±	± 0.3	7 ± 6	11 ± 15
Forest	0-5	1.21	1.42		$1.32 \pm 0.2$	24	19	27	23	± 4	$44.00 \pm 1.6$	$32.60 \pm 1.2$	50.7 ± 1.8	42.43 ±	9.2	16 ± 1	15 ± 1
	5-10	1.46	1.65		$1.56 \pm 0.1$	24	37	34	32	± 7	$23.80\pm0.9$	$17.50 \pm 0.7$	$28.4 \pm 1.1$	23.23 ±	5.5	15 ± 1	
	10-15	1.49	1.73		$1.61 \pm 0.2$	36	43	38	39	± 4	$12.50 \pm 0.6$	$12.50 \pm 0.6$	$15.2 \pm 0.7$	13.40 ±	± 1.6	16 ± 1	14 ± 1
	15-20	1.48	1.40		$1.44 \pm 0.1$	38	37	35	37	± 1	$7.72 \pm 0.4$	$9.29 \pm 0.5$	$10.5 \pm 0.5$	9.17 ±	± 1.4	13 ± 2	
	20-30	1.65	1.62		$1.63 \pm 0.0$	33	42	35	37	± 5	$5.18 \pm 0.4$	$5.94 \pm 0.4$	$7.87 \pm 0.4$	6.33 ±	± 1.4	12 ± 2	12 ± 2
	30-40	1.74	1.67		$1.70 \pm 0.0$	41	39	33	38	± 4	$4.34 \pm 0.3$	$4.16 \pm 0.3$	$4.98 \pm 0.4$	4.49 ±	± 0.4	12 ± 2	12 ± 2
	40-50	1.72	1.55		$1.64 \pm 0.1$	36	47	30	38	± 9	$3.44 \pm 0.3$	$3.41 \pm 0.3$	$3.32 \pm 0.3$	3.39 ±	£ 0.1	11 ± 2	11 ± 2
	50-60	1.70	1.58		$1.64 \pm 0.1$	35	62	50	49	± 13	$2.70 \pm 0.3$	$1.89 \pm 0.3$	$2.25 \pm 0.3$	2.28 ±	± 0.4	10 ± 2	11 ± 3
	60-70	1.74	1.68		1.71 ±0.0	27			27		$2.09 \pm 0.3$			2.09	9.2	10 ± 3	

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Table D2: p values the Student's t-test to compare, depth by depth, the TOC between vineyard and forest soils. This test is applicable if the variances are in the same order of magnitude. We therefore performed the test on log<sub>10</sub>(TOC) to have similar orders of magnitude of variances between vineyard and forest soils. The p-value results are:

Depth [cm]	<u>t-test p-value</u>
0-5	0.00059
5-10	0.00015
10-15	0.00024
15-20	0.00028
20-30	0.00104
30-40	0.00118
40-50	0.00928

<u>50-60</u> 0.00100 <u>60-70</u> 0.07454

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The p-values showed a significant difference (<0.01) in TOC between forest and vineyard soils to 60 cm depth.

Table D3: p-values the Student's t-test to compare the C:N ratio between vine and forest soils. Up to 50 cm depth, the p-values were under 0.05 except for the 15-20 cm and 30-40 cm horizons, where they were less than 0.1. This result shows that there

445 <u>was a significant difference in C:N, with lower values in the vineyard than in the forest soils. This result tended to confirm</u> that, at equivalent depth, the C pool remaining in the vineyard had a more marked microbial signature than the C pool in the forest soil.

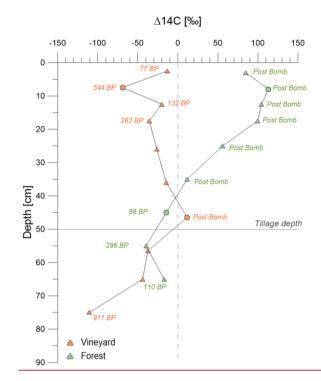
Depth [cm]	<u>t-test p-value</u>
0-5	0.0255
5-10	<u>0.0143</u>
10-15	<u>0.0122</u>
15-20	<u>0.0990</u>
20-30	<u>0.0098</u>
30-40	<u>0.0778</u>
40-50	<u>0.0310</u>
50-60	0.4627
60-70	<u>0.7696</u>

#### 450

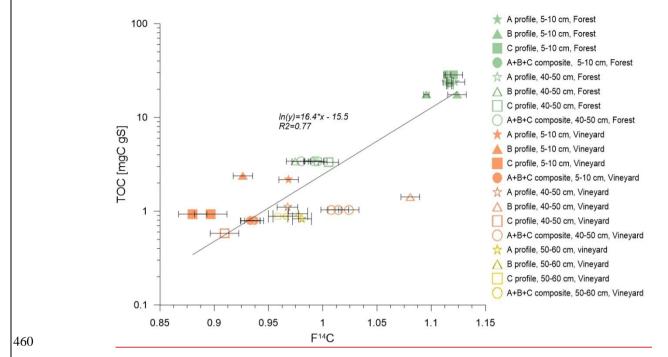
**Table <u>D4</u>:** Amount of <u>C measured (micrograms) and  $F^{14}C$  per composite sample (A+B+C) or per profile (A, B and C) according to soil depth, the numbers 1, 2 or 3 indicate the repetitions, ± is the analytical error.</u>

					A+B+C						А				E	3				;	
			1		2		3		1		2		3		1		2		1		2
	Depth	С	F <sup>14</sup> C	С	F <sup>14</sup> C	С	F <sup>14</sup> C	С	F <sup>¹⁴</sup> C	С	F <sup>¹⁴</sup> C	С	F <sup>14</sup> C	С	F <sup>¹⁴</sup> C	С	F <sup>14</sup> C	С	F <sup>14</sup> C	С	F <sup>14</sup> C
	cm	μg		μg		μg		μg		μg		μg		μg		μg		μg		μg	
Vineyard	0-5	71	0.990 ±0.009																		
	5-10	53	0.937 ±0.009	54	0.933 ± 0.009	54	$0.933 \pm 0.009$	54	$0.969 \pm 0.009$					54	0.926 ± 0.009			26	0.897±0.01	5 27	$0.880 \pm 0.013$
	10-15	54	0.984 ±0.009																		
	15-20	45	0.968 ±0.010																		
	20-30	50	0.977 ±0.010																		
	30-40	55	$0.989 \pm 0.009$																		
	40-50	50	1.014 ±0.010	53	1.008 ± 0.009	53	$1.024 \pm 0.009$	48	0.967±0.009					86	1.081 ± 0.009			27	$0.909 \pm 0.01$	3	
	50-60		0.966 ±0.009					40	$0.981 \pm 0.009$					36	0.977 ± 0.009			34	$0.959 \pm 0.00$	9	
	60-70		0.959 ±0.014																		
	70-80		0.893 ±0.014																		
Forest	0-5	998	1.089 ±0.003																		
	5-10		1.118 ±0.002	988	$1.116 \pm 0.002$			1 000	$1.113 \pm 0.003$	987	$1.118 \pm 0.003$	123	$1.122 \pm 0.009$	998	1.095 ± 0.002	108	$1.124 \pm 0.009$	997	$1.115 \pm 0.00$	3 93	$1.120 \pm 0.009$
	10-15		1.108 ±0.002																		
	15-20		1.104 ±0.002																		
	20-30	983	1.059 ±0.002																		





**Figure** <u>D1:</u> Comparison of the BP age patterns, via  $\Delta^{14}$ C [‰], as a function of the soil depth and vegetation cover.



**Figure D2:** Variations in carbon content as a function of  $F^{14}C$ . Profiles A, B and C are represented by stars, triangles and squares, respectively. These symbols are solid when they represent topsoil samples (5-10 cm) and empty when they represent deep samples (40-50 cm). Soil samples obtained under the forest are green and those under vines are orange. The error bars represent the analytical error. The TOC values were higher with younger  $F^{14}C$  (usually topsoil samples):  $R^2$ =0.77. Under the vineyard, ploughing had eliminated the young carbon pools.

## Figure D3 and D4: Permutation tests

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At 5-10 cm depth, the observed ratio was 9.16 ( $\neq$ 1). We repeated 1,000 times a permutation test of the RMS ratios between forest and vines (simulation), which we then compared to the observed ratio value (Fig. D3). The observed value was outside

470 the simulated critical values with p=0.02 (<0.05). This showed that the variance under vines was significantly different from the variance under the forest.

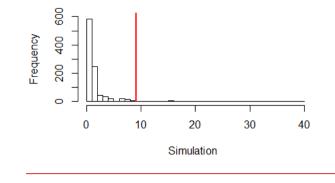


Figure D3: RMS' simulation ratios in relation to the observed ratio (red) at 5-10 cm depth

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At 40-50 cm depth, the observed ratio is 27.53 ( $\neq$ 1). Similarly, we repeated a permutation test 1,000 times. The observed value was within the simulated critical values (Fig. D4), with a p=0.01 (<<0.05). This showed that the variance under vines was significantly different from the variance under the forest.

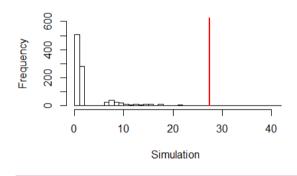




Figure D4: RMS' simulation ratios in relation to the observed ratio (red) at 40-50 cm depth.

## **Appendix E: comparison with literature data**

Table E1: listing papers used in this study, with land use type for each soil and associated TOC as a function of depth. These papers all deal with arenosols, or at least sandy soils, in Mediterranean climates according to the Köppen-Geiger criteria (csa 485 means temperate climat, dry and hot summer). They were found by accessing the ISRaD database or the Web of Science with the keywords "14C arenosol heterogeneity".

Paper

Publication Country DOI

	year						years	cm	g <u>.</u> -kg <sup>-1</sup>	gkg <sup>-1</sup>
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	0-5	104.7	na
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	5-11	9.3	na
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	11-30	14.2	na
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	30-55	6.1	na
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	55-75	2.1	na
Andreetta et al.	2013	Italy	10.1007/s10533-011-9693-9	Haplic arenosol	csa	Holm oak forest	50	75-120	1.4	na
Caravaca et al.	2002	Spain	10.1016/S0167-1987(02)00080-6	Calcaric arenosol	csa	Spontaneous grass cover	na	0-20	21.3	na
Caravaca et al.	2002	Spain	10.1016/S0167-1987(02)00080-6	Calcaric arenosol	csa	vineyard	na	0-20	3.2	na
Conradie	2001	South Africa	https://doi.org/10.21548/22-2-2192	Sandy soil	scb	vineyard	na	0-20	4.8	na
Conradie	2001	South Africa	https://doi.org/10.21548/22-2-2192	Sandy soil	scb	vineyard	na	20-40	1.7	na
Conradie	2001	South Africa	https://doi.org/10.21548/22-2-2192	Sandy soil	scb	vineyard	na	40-60	1.6	na
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	47	7
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	48	8
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	45	15
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	50	21
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	54	21
Fierro et al.	2007	Italy	10.1071/WF06114	Calcaric arenosol	csa	forest	na	0-5	48.8	14.4
Fourie et al.	2005	South Africa	https://doi.org/10.21548/26-2-2129	Sandy soil	csb	vineyard	na	0-30	1.3	na
Fourie et al.	2005	South Africa	https://doi.org/10.21548/26-2-2129	Sandy soil	csb	vineyard	na	30-60	1.0	na
López-Piñeiro et al.	2013	Spain	http://dx.doi.org/10.1016/j.still.2012.09.007	Loamy sand soil	csa	vineyard	na	0-10	1.73	na
López-Piñeiro et al.	2013	Spain	http://dx.doi.org/10.1016/j.still.2012.09.007	Loamy sand soil	csa	vineyard	na	0-10	1.68	na
Nogales et al.	2018	Portugal	10.3389/fpls.2018.01906	Arenosol	csa	vineyard	na	0-30	5.7	0.213
Okur et al.	2009	Turkey	10.3906/tar-0806-23	Sandy loamy soil	csa	vineyard	na	0-20	7.8	na
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	Natural oak foret	100	0-20	16	1.62
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	Natural oak foret	100	20-40	5.6	0.33
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	maquis	na	0-20	31	2.45
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	maquis	na	20-40	7.7	0.45
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	pine forest plantation	60	0-20	20.1	2.56
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	pine forest plantation	60	20-40	6.1	0.25
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	natural mixed forest	na	0-20	22.7	2.06
Pinzari et al.	1999	Italy	10.1016/S0167-7012(99)00007-X	Sandy soil	csa	natural mixed forest	na	20-40	19	0.41
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Holm forest	na	0-3	72.0	5.6
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Holm forest	na	3-7	49.3	2.4
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Holm forest	na	7-12	10.5	0.5
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Holm forest	na	12-50	1.8	0.3
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Pine forest	na	0-3	42.7	1.5
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Pine forest	na	3-11	10.5	1.3
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Pine forest	na	11-25	1.9	0.4
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Haplic arenosol	csa	Pine forest	na	25-50	1.2	0.3
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest (oak)	245	0-3	49.7	3.1
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest	245	3-6	19.6	1.2
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest	245	6-12	9.6	1.8
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest	245	12-19	2.1	0.3
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest	245	19-30	2.1	0.6
Vittori Antisari et al.	2016	Italy	10.1007/s12665-016-5581-x	Brunic arenosol	csa	Hygro forest	245	30-50	10.0	0.2

Table E2: Köppen-Geiger criteria. The Köppen-Geiger Mediterranean climate classes including the defining criteria, adapted from (Beck et al., 2018): MAT=mean annual air temperature (°C); T<sub>cold</sub>=air temperature of the coldest month (°C); T<sub>hot</sub>=air temperature of the warmest month (°C); T<sub>mon10</sub>=the number of months with air temperature >10-°C (unitless); MAP=mean annual precipitation (mm y<sup>-1</sup>); P<sub>sdry</sub>=precipitation in the driest month in summer (mm month<sup>-1</sup>); P<sub>wwet</sub>=precipitation in the wettest month in winter (mm\_month<sup>-1</sup>); P<sub>threshold</sub>=2×MAT if >70% of precipitation falls in winter, P<sub>threshold</sub>=2×MAT+14. Summer (winter) is the 6-month period that is warmer (colder) between April-September and October-March.

1st	2nd	3rd	Description	Criterion
в			Arid	MAP<10×P <sub>threshold</sub>
	W		desert	MAP<5×P <sub>threshold</sub>
	S		steppe	MAP≥5×P <sub>threshold</sub>
		h	hot	MAT≥18
		k	cold	MAT<18
С			Temperate	Not (B) & T <sub>hot</sub> >10 & 0 <t<sub>cold&lt;18</t<sub>
	s		Dry summer	$P_{sdry}$ <40 & $P_{sdry}$ < $P_{wwet}$ /3
	f		without dry season	Not (Cs)
		а	hot summer	T <sub>hot</sub> ≥22
		b	warm summer	Not (a) & T <sub>mon10</sub> ≥4
		с	cold summer	Not (a or b) & 1≤T <sub>mon10</sub> <4

#### Author contributions

The conceptualization of the study for this paper was done by SQ, SC and IBD with input from NC, FJ, DB, AD and CH. All

<sup>510</sup> authors participated in the <u>resource</u> collection. The data curation and formal analysis and methodology were done by SQ, IBD and CH. The visualization for the paper was performed by SQ, with substantial input from IBD and CH as well as feedback from all authors. SO and IBD wrote the initial draft and all authors were involved in the review and editing of the paper.

#### **Competing interests**

The authors declare that they have no conflict of interest.

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