

Crop residue management and oxalate-extractable iron and aluminium explain long-term soil organic carbon sequestration and dynamics

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1 **Crop residue management and oxalate-extractable iron and**
2 **aluminium explain long-term soil organic carbon sequestration**
3 **and dynamics**

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16 *Running title: Carbon sequestration in contrasting soil types*

17

18

19 *Keywords: carbon sequestration, crop residue, SOM fractionation, spatial analysis, iron*
20 *oxides, organo-metal complexes*

21

22 **Highlights**

23

- 24 • Different factors affect carbon stocks under two contrasting soils and four cropping systems
- 25 • Returning maize residues promotes substitution of native soil organic carbon by maize-derived
- 26 carbon
- 27 • Organo-mineral interactions in the silt and clay fraction enhance carbon sequestration
- 28 • The legacy of historic plaggen manuring has a strong effect on native soil carbon stocks

29

30

31 **Summary**

32

33 The management of crop residues affects carbon (C)-sequestration. This study aimed to
34 identify the interaction between residue management and soil properties on C-sequestration.
35 The hypothesis was that larger silt and clay contents and larger residue inputs enhance C-
36 sequestration. The soil was sampled in Belgium in long-term (≥ 15 years) cropping systems
37 with either grain maize, *Zea mays* L. (all stover (leaves and stalks) returned), silage maize (all
38 stover removed) or permanent grass. The fields sampled were distributed over two adjacent
39 regions; one with sandy soil (33% silt+clay) and one with silty loam soil (71% silt+clay). The
40 ^{13}C abundance of the soil organic carbon (SOC) revealed that topsoil (0–30 cm) under grain
41 maize contained more maize-derived SOC than that under silage maize (14 ± 1 and 9 ± 1 Mg
42 C ha^{-1} , respectively, $P < 0.001$, in sand, and 17 ± 1 and 14 ± 1 Mg C ha^{-1} , respectively,
43 $P < 0.001$, in silty loam). Total SOC stocks were unaffected by crop management, however,
44 which suggests substitution of native SOC by fresh SOC derived from residues, especially in
45 sandy soil where the silt and clay fraction is saturated with SOC. The maize-derived SOC
46 stocks of the silty loam soil are 3.5 to 5.5 Mg C ha^{-1} larger than those of the sandy soil, which
47 confirms the larger potential of the former to sequester C. Surprisingly, the native C_3 -SOC
48 stocks in the sandy soil were 25 to 30 Mg C ha^{-1} larger than those of the silty loam soil. The
49 concentration of SOC in the silt and clay fraction was on average three times larger for the
50 sandy than the silty loam soil. The concentration of oxalate-extractable Al and Fe in the same
51 fraction was on average 2.5 times larger for sandy than for silty loam soil. Silt and clay
52 associated SOC correlated with the Fe+Al concentration across all regions and treatments
53 ($R^2=0.62$). This suggests that organo-mineral associations and the formation of organo-metal
54 complexes promote C-sequestration in this temperate region.

55

56 **Introduction**

57

58 Crop residues are an important source of soil organic matter in agro-ecosystems, but the
59 increasing demand for biomass for renewable energy might reduce residue returns and
60 therefore soil organic carbon (SOC) stocks. The cultivation of maize (*Zea mays* L.) for biogas
61 can reduce fossil energy related CO₂ emissions considerably (Gerin *et al.*, 2008). However,
62 the sustainability of residue removal is unknown in relation to maintaining SOC content.
63 Larson *et al.* (1972) and Rasmussen *et al.* (1980) suggested that change in SOC is related
64 linearly to the amounts of residues returned to a soil irrespective of their type. In contrast,
65 Campbell *et al.* (1991) could not demonstrate that long-term straw removal reduced SOC
66 compared to straw retention and incorporation. The available studies on maize are
67 inconclusive. Reicosky *et al.* (2002) found no significant difference in SOC between the
68 removal of stover (leaves and stems) or its return to the soil after 30 years of maize
69 monoculture, whereas other studies (Robinson *et al.*, 1996; Dick *et al.*, 1998) found a
70 significant increase in SOC when stover was returned to the soil.

71

72 Soil properties and management affect C-sequestration and might obscure the effect of crop
73 residue management. Wilhelm *et al.* (2004) concluded that changes in SOC are proportional
74 to the amount of crop residue returned, but that tillage affects the actual retention of SOC.
75 Both silage and grain maize are often grown in long-term monoculture systems and will,
76 therefore, affect the soil's capacity to sequester carbon. The decline in SOC was reported to
77 be faster under continuous maize than under maize in rotation (Odell *et al.*, 1984; Huggins *et*
78 *al.*, 1998). Robinson *et al.* (1996) attributed this to the smaller quantities of residue returned to

79 the soil and a greater tillage intensity under maize monoculture. The silt and clay contents
80 might affect the role of crop residue input on C-sequestration because they affect the carbon
81 saturation of soil (Six *et al.*, 2002). Another soil property that might determine the retention of
82 C is the concentration of Al and Fe. Amorphous Al and Fe oxyhydroxides have been isolated
83 from a variety of soil types, in particular from podzolic soil (Adams & Kassim, 1984; Evans
84 & Wilson, 1985; Blaser *et al.*, 1997) for which positive correlations with SOC have been
85 shown.

86

87 In this study we aimed to elucidate the effect of crop residue management and soil properties
88 such as texture and metal concentration on C-sequestration; our hypothesis was that larger silt
89 and clay contents facilitate greater C-sequestration when the annual return of residues is
90 increased. We also hypothesized that a larger concentration of amorphous Al and Fe in the silt
91 and clay fraction increases the soil's capacity to sequester C irrespective of soil type.

92 Functional relations were sought, therefore, between soil organic carbon and either silt and
93 clay content or the concentration of oxalate-extractable Al and Fe. The soil was sampled in
94 fields that had been cropped for at least 15 years with either grain maize (all stover returned),
95 silage maize (all stover removed) or permanent grass from adjacent regions with sandy soil
96 and silty loam soil. We measured ¹³C natural abundance to identify the processes involved.

97 This technique has been used repeatedly to establish the relative proportion of C₃- and C₄-
98 crops to total SOC, and to gain insight into SOC turnover in long-term experimental fields
99 after the introduction of maize monoculture (Balesdent *et al.*, 1987; Clapp *et al.*, 2000; Wilts
100 *et al.*, 2004; Hooker *et al.*, 2005). Former studies were done on one or at best a few
101 experimental sites, whereas we adopted a regional scale perspective by sampling 70 farmers'
102 fields. Stable isotopes were measured both in the soil and in several SOC fractions.

103 Fractionation of soil samples enabled us to determine the extent to which both C₄- and C₃-
104 SOC were protected against mineralization.

105

106 **Materials and methods**

107

108 *Selection of the fields*

109 Soil samples were obtained from 70 fields in Flanders (the northern part of Belgium),
110 distributed over two adjacent regions: one with a sandy soil (40 fields) and the other with a
111 silty loam soil (30 fields). The soil of each field was classified according to World Reference
112 Base (WRB) by determining the reference soil groups (RSGs) for the geographic centroids of
113 the fields (Dondeyne *et al.*, 2014; IUSS Working Group WRB, 2014). Classification names
114 along with other major soil properties are provided in the supporting information (Table SI-2).
115 The fields were selected from a crop geodatabase developed to reconstruct the cultivation
116 history between 1997 and 2013 for about 500 000 agricultural fields in Flanders. The
117 development of the crop geodatabase and the selection criteria for the fields are described in
118 the supporting information, sections S1 and S2. The cropping systems were in place for at
119 least 15 (sandy region) or 17 years (silty loam region). All maize fields have a level
120 topography (confirmed by a digital elevation model) to minimize soil erosion. Four long-term
121 cropping systems were investigated: (i) silage maize monoculture (SM) where all stover is
122 harvested with short stubble only remaining, (ii) silage maize monoculture followed by grass,
123 mainly Italian ryegrass (*Lolium multiflorum* Lam.), sown directly between the maize stubble
124 by minimum or zero tillage and ploughed the next spring after a one-cut harvest (SMGRA),
125 (iii) grain maize monoculture (GM) where all stover is returned because the grain only is
126 harvested and (iv) permanent grass (pasture, PA) with a history of C₃-vegetation only.

127 Forty fields (ten fields each for GM, SM, SMGRA and PA) were selected from the crop
128 geodatabase in the northern part of Flanders. This region is characterized by a sandy to loamy
129 sandy soil texture and was dominated by heathland until the end of the 18th century (de
130 Ferraris, 2009). Because the region is dominated by intensive animal husbandry, the focus is
131 on fodder production. Another 30 fields (ten fields each for GM, SM and PA; no SMGRA)
132 were selected in the silty loam region in the southern part of Flanders. Agriculture in this
133 region is characterized by mixed farming that combines arable cultivation with livestock
134 farming. Within each region all fields were selected in a completely random way.

135

136 *Soil sampling*

137 All 40 fields in the sandy region were sampled between the end of November 2011 and mid-
138 March 2012. The 30 fields in the silty loam region were sampled in February and March
139 2014. Each field was sampled following a slightly modified version of the ‘area-frame
140 randomized soil sampling’ (AFRSS) protocol (Stolbovoy *et al.*, 2007). Briefly, the number of
141 sampling sites per field depended on the area of the field and their location was determined by
142 projecting a grid of random numbers over the field (detail is in the supporting information,
143 section S3). For each sampling site nine subsamples were taken with an auger of 3.5-cm
144 diameter that were mixed together to form a composite sample. The soil was sampled to a
145 depth of 30 cm. The upper 30 cm of soil under PA was sampled in intervals of 10 cm (i.e. 0–
146 10 cm, 10–20 cm and 20–30 cm). In contrast, for soil under maize a single sample was taken
147 over the 0–30-cm layer because it is considered homogenous from the annual tillage. For
148 every composite soil sample an undisturbed soil core was taken by a soil corer (with both a
149 diameter and height of five cm) and a Kopecky sampler, after pre-digging with an Edelman
150 auger. For the soil under maize the core was taken between 12.5 and 17.5 cm depth in the

151 profile. For the soil under permanent grass a separate core was taken for each of the three 10-
152 cm layers sampled.

153

154 *Measurements on bulk soil samples*

155 Details of soil sample preparation are provided in the supporting information, section S4. Soil
156 organic carbon and nitrogen contents were determined with an Elemental Analyzer (Thermo
157 Scientific Flash HT EA, Waltham, MA, USA) with a thermal conductivity detector (furnace
158 temperature of 1070 °C). The measurement error was determined from five repeated
159 measurements of ten different samples (with both small and large percentages of SOC); it was
160 0.01%. The SOC contents (g C 100 g⁻¹ soil, or %C) were then converted to SOC stocks (Mg
161 C ha⁻¹).

162

163 The ¹³C/¹²C ratio of SOC was determined with a Thermo Scientific, Delta V Advantage
164 Isotope Ratio MS coupled with the EA through ConFlo IV (Thermo Scientific, Waltham,
165 MA, USA). The carbon isotope ratio was converted to δ¹³C by

$$166 \quad \delta^{13}\text{C} = \left(\frac{R_{\text{sam}}}{R_{\text{std}}} - 1 \right) \times 10^3, \quad (1)$$

167 where R_{sam} is the ¹³C/¹²C ratio of the sample and R_{std} is the ¹³C/¹²C ratio of the Vienna Pee
168 Dee Belemnite standard. The fraction, f , of maize-derived carbon (C₄-carbon) in a soil sample,
169 expressed as a percentage of the total SOC, is given by

$$170 \quad \delta_s = f \cdot \delta_m + (1 - f) \cdot \delta_g, \quad (2)$$

171 where δ_s is the δ¹³C value determined on the soil sample, δ_m is the δ¹³C value of the maize
172 residue and δ_g is the initial δ¹³C value of the soil before the introduction of maize. The δ_m was

173 determined on 24 samples of maize residue which were taken in the field (more detail is given
174 in the supporting information, section S5). The δ_g was determined on the soil samples taken
175 from the 10 to 20-cm soil layer of the fields under permanent grass (PA fields) that served as
176 the ‘control’ treatment or isotopic reference. Data from a historical soil database suggest that
177 these fields had never had maize grown there and have always been under continuous grass
178 (see supporting information, section S2 for more detail). After calculating the fraction of SOC
179 derived from maize residues by Equation (2) for the maize fields, the total SOC of these fields
180 can be subdivided into C₄-SOC and C₃-SOC; the latter originated from the grassland
181 vegetation before the transition to maize.

182 The particle-size distribution of all soil samples was determined by laser diffraction
183 (Beckman Coulter LS13320 with Aqueous Liquid Module, Brea, CA, USA), see supporting
184 information, section S4 for detail. One soil sample was selected from each field to measure
185 pH (0.01 M CaCl₂; soil:water ratio 1:5).

186

187 *Fractionation of SOC*

188 For each of the 70 fields sampled, one composite topsoil sample was selected and fractionated
189 by the method of Zimmermann *et al.* (2007). The method combines several physical and
190 chemical fractionation techniques (Figure SI-3) to apportion the total SOC over five different
191 fractions. Each fraction is assumed to be protected from microbial degradation to different
192 extents. Two of the fractions are easily degraded (particulate organic matter (POM) and
193 dissolved organic carbon (DOC)), two are degraded more slowly (carbon associated with silt
194 and clay (s+c), and carbon stabilized in aggregates (S+A)), and one remains undegraded by
195 microbial activity (oxidation-resistant carbon (rSOC)). The oxidation-resistant rSOC is a sub-
196 fraction of the s+c fraction; the difference between s+c and rSOC fractions (to which we refer

197 hereafter as (s+c)-rSOC) is the oxidizable part of the s+c fraction. Details of the fractionation
198 procedure are given in the supporting information, section S6. All solid fractions (POM, S+A,
199 s+c and rSOC) were analysed for SOC, N and $\delta^{13}\text{C}$ in an analogous way to analysis of the
200 bulk soil. Prior to the analyses, the S+A, s+c and rSOC fractions were acidified with dilute
201 HCl to remove any inorganic carbon (IC). The DOC concentration was measured with a TOC
202 analyser (OI Analytical, Aurora 1030, College Station, TX, USA) with a nondispersive
203 infrared (NDIR) detector by wet oxidation with sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3$) after the
204 addition of phosphoric acid (H_3PO_4) to remove any IC. The $\delta^{13}\text{C}$ signature of the DOC was
205 determined with the same mass spectrometer that was used for the solid fractions and the bulk
206 soil samples. Maize-derived SOC in the fractions was calculated from the $\delta^{13}\text{C}$ values with
207 the fractionated PA soil, which served as the reference treatment (Equation (2)).

208

209 Oxalate-extractable Al and Fe were determined for the s+c fraction with a 0.2 M ammonium
210 oxalate extraction solution at pH 3 (solid to liquid ratio of 40 mg:2 ml, end-over-end shaking
211 for two hours in darkness; Schwertmann, 1964). Concentrations of Al and Fe were measured
212 by ICP-OES (inductively coupled plasma optical emission spectroscopy) with an Optima
213 3300 DV (Perkin Elmer, Waltham, MA, USA).

214

215 *Statistical analysis*

216 The statistical analysis was done in R, version 3.1.1 (R Core Team, 2015). The statistical
217 analysis of the bulk soil properties differed slightly from that of the SOC fractions. For the
218 latter, with only one fractionated composite soil sample per field we used a classical one-way
219 analysis of variance (ANOVA) in which the regions were analysed separately and cropping
220 history was the main effect, and a two-way ANOVA in which the regions were analysed

221 together with both region and cropping history as the main effects. However, to investigate
222 the effect of cropping history on several response variables for the bulk soil (with 3, 4 or 5
223 observations per field, i.e. a nested sampling scheme with within-field replicates) we averaged
224 the replicates before applying either of the ANOVAs. If the effect of cropping history on the
225 response variable was significant at $P = 0.05$, treatment means were compared with Fisher's
226 least significant difference (LSD) test (*LSD.test* in R-package *agricolae*). Means and standard
227 errors of the mean were reported. The normality of the residuals was verified by Q–Q plots. If
228 the residuals were not normally distributed, the data were transformed to natural logarithms.
229 The same was done if heteroscedasticity was detected by Levene's test. When two soil
230 variables were plotted against each other, we determined the functional relation between those
231 variables. Functional relations were estimated with Equations (18) and (19) in Webster
232 (1997).

233

234 **Results**

235

236 *Bulk soil*

237 Soil characteristics for the 70 fields sampled are summarized by region (Table SI-2). The
238 median of soil pH is about 1 unit lower in the sandy soil than in the silty loam soil. The
239 median of silt+clay content in the silty loam soil is twice that in the sandy soil. The sandy soil
240 under permanent grass (PA) has significantly ($P < 0.05$) larger SOC stocks in the 0–30-cm
241 topsoil layer than does the arable soil under maize (SM, SMGRA and GM; Table 1). Crop
242 residue management (removal or return of stover) or the sowing of a cover crop after harvest
243 had no significant effect on total SOC stocks. The $\delta^{13}\text{C}$ signatures show that sandy soil under
244 continuous cultivation of grain maize has a larger proportion of maize-derived SOC than that

245 under silage maize (18% and 11%, respectively; $P < 0.001$). There was no difference in C_3 -
246 SOC between sandy soil with continuous cultivation of silage maize whether or not this was
247 combined with a second annual C_3 -grass crop (Italian ryegrass), and there was no difference
248 in C_3 -SOC between grain maize and silage maize.

249

250 The effects of crop management for the silty loam soil are similar to those observed for sandy
251 soil. However, when the two soil types under maize are compared, total SOC and C_3 -SOC are
252 significantly smaller ($P < 0.001$) in the silty loam soil, whereas $\delta^{13}C$, C_4 -SOC and proportion
253 of C_4 -SOC are significantly larger. The soil under permanent grass, however, does not differ
254 significantly in total SOC or in $\delta^{13}C$. The total SOC stock for soil under maize is about 20 to
255 25 $Mg\ C\ ha^{-1}$ less for the silty loam than for the sandy soil, which is associated with a 25 to 30
256 $Mg\ C\ ha^{-1}$ smaller C_3 -SOC for the former. In contrast, the $\delta^{13}C$ values of soil under SM and
257 GM are about 2.5‰ larger in the silty loam soil than in the sandy soil, which results from the
258 greater proportion of maize-derived SOC in the silty loam soil of about 15%. In absolute
259 terms, the C_4 -SOC stocks of the silty loam soil are up to 5.5 $Mg\ C\ ha^{-1}$ larger than in the
260 sandy soil. The silty loam soil under GM contains ~3 $Mg\ C_4$ -SOC ha^{-1} more than that under
261 SM, and the sandy soil under GM contains ~5 $Mg\ C_4$ -SOC ha^{-1} more than that under SM, but
262 these differences are not significant. Some of the ANOVA-tables that are behind Table 1 are
263 reported in Table 2.

264

265 *The SOC fractions*

266 In both types of soil, about 60 to 70% of the SOC is present in the s+c fraction (Table 3). In
267 the sandy soil, the second largest SOC fraction is attributed to sand and stable aggregates
268 (S+A), followed by the POM fraction. In the silty loam soil, POM and S+A contribute equally

269 to SOC under maize, but under permanent grass S+A contributes significantly more ($P<0.01$)
270 to SOC than POM. For all SOC fractions in the sandy soil, no significant effects of cropping
271 history were detected, except that the S+A fraction contains significantly more SOC ($P<0.05$)
272 in soil under continuous grass than continuous maize. In contrast, the silty loam soil under PA
273 contains significantly more SOC ($P<0.05$) than the soil under maize in all fractions except for
274 POM; the largest difference is again for S+A. In spite of the larger SOC content of the s+c
275 fraction in the sandy soil under maize, both types of soil have the same relative contribution
276 of s+c to total SOC. Maize residue management has no significant effect either on the
277 absolute amount of SOC per fraction or on the contribution of the fraction to total SOC.

278

279 The differences in $\delta^{13}\text{C}$ between the cropping systems are consistent for all SOC fractions;
280 soil under PA has significantly ($P<0.001$) more negative $\delta^{13}\text{C}$ values than all soil under maize,
281 but soil under SM and SMGRA has significantly ($P<0.05$) more negative $\delta^{13}\text{C}$ than that under
282 GM (Figure 1). There are no significant differences between $\delta^{13}\text{C}$ of SM and that of SMGRA.
283 These effects are similar to those for the bulk soil. The $\delta^{13}\text{C}$ values under PA are similar for
284 all fractions of the sandy soil and there are small differences only in the silty loam soil. In
285 contrast, in maize-cropped soil the $\delta^{13}\text{C}$ value is less negative, and the maize-derived SOC is
286 larger in the labile (POM, DOC) than in the recalcitrant (rSOC) fractions. The $\delta^{13}\text{C}$ of the
287 fractions is less negative in the silty loam than in the sandy soil under maize, but not for POM
288 and DOC. The POM fraction in the sandy soil has on average $43 \pm 3\%$ of maize-derived SOC
289 under GM and $26 \pm 3\%$ under SM, whereas rSOC has only $10 \pm 1\%$ C₄-SOC under GM and 5
290 $\pm 1\%$ under SM. The GM-cropped soil has significantly more maize-derived SOC than does
291 the SM-cropped soil in each fraction ($P<0.001$), with on average 1.9 times more C₄-SOC. For
292 the silty loam soil the differences are smaller with on average only 40% more C₄-SOC
293 ($P<0.05$) under GM. The S+A fraction of the silty loam soil has the largest proportion of C₄-

294 SOC, and this is followed by the POM, (s+c)-rSOC, DOC and rSOC fractions. The latter is
295 again the most stable fraction and consists mainly of old C₃-SOC. When the two soil types are
296 compared, the largest difference is for the S+A and (s+c)-rSOC fractions, which both have
297 20% more C₄-SOC in the silty loam soil.

298

299 *The SOC and oxalate-extractable metals in the silt and clay fraction*

300 The amount of SOC associated with the s+c fraction is plotted against the silt+clay content of
301 the soil (Figure 2). The data are plotted separately for both types of soil, but all cropping
302 histories are combined because there is no difference in the functional relation between arable
303 cropping and permanent grass (data not shown). An increase in silt and clay content increases
304 the amount of SOC in the s+c fraction, especially in the sandy soil. The mean concentration of
305 oxalate-extractable Al and Fe in the s+c fraction is about 2.5-fold larger in the sandy than in
306 the silty loam soil, whereas the mean concentration of SOC in the s+c fraction differs by a
307 factor of three (Figure 3). The concentration of oxalate-extractable Al and Fe of the s+c
308 fraction explains the amount of SOC in this fraction ($P<0.001$).

309

310 **Discussion**

311

312 More than 15 years of stover incorporation in GM monoculture did not increase total SOC
313 significantly, but it increased the fraction of maize-derived SOC. The absence of significant
314 effects of residue management on total SOC could be attributed to the relatively short time
315 scale of the maize cropping systems studied. In separate experiments, we recorded that the
316 larger stover input in GM than for SM in this region is equivalent to an additional 3.54 Mg C
317 ha⁻¹ year⁻¹, or 53.05 Mg C ha⁻¹ over 15 years. With a retention coefficient of about 5 to 10%

318 of the fresh residue carbon (Allmaras *et al.*, 2004; Wilts *et al.*, 2004; Hooker *et al.*, 2005), this
319 contribution is small relative to the total SOC stocks of 55–80 Mg C ha⁻¹, and of the same
320 order of magnitude as the standard error of the stocks. Likewise, Reicosky *et al.* (2002) found
321 no effect of residue management on total SOC in the top 20 cm of the soil after 30 years of
322 continuous maize cultivation. They attributed this to intensive mouldboard plough tillage
323 (also used in Flanders), which stimulates decomposition and offsets any net gain in SOC from
324 the larger residue additions under GM. Soil carbon saturation (Six *et al.*, 2002) might also
325 explain why long-term stover return cannot increase total SOC stocks substantially more than
326 long-term stover harvest. In the sandy soil, in particular, the SOC dynamics have probably
327 reached a steady state, given the small silt and clay contents (Table SI-2), the relatively large
328 total SOC stocks (Table 1) and the same SOC content in the (s+c)-rSOC and rSOC fractions
329 for both permanent grass and maize monocropping (Table 3). The $\delta^{13}\text{C}$ analysis revealed a
330 clear effect of crop management on maize-derived SOC, but the additional C₄-SOC might
331 have been compensated for by an enhanced loss of old C₃-SOC. The latter might be explained
332 by a positive priming effect, which is defined as a strong short-term acceleration of the
333 decomposition of native SOM after the addition of an easily decomposable substrate
334 (Kuzyakov *et al.*, 2000).

335

336 The effects of maize residue management on the $\delta^{13}\text{C}$ in soil are consistent with several long-
337 term field trials. Balesdent *et al.* (1987) showed that $\delta^{13}\text{C}$ was 1‰ less negative after 23 years
338 of stover return under continuous maize cultivation than with stover removal. The maize-
339 derived SOC amounted to 21% in the top 0–30 cm of the profile with stover returned and 13%
340 with stover removed after 29 years of continuous maize cultivation (Wilts *et al.*, 2004). Clapp
341 *et al.* (2000) found that C₄-SOC content was 15% with stover returned, but only 5% with
342 stover removed after 13 consecutive years of maize under no tillage. With mouldboard or

343 chisel ploughing, however, both stover treatments led to a maize-derived SOC proportion of
344 about 10%.

345

346 The significantly larger C₄-SOC stocks in the silty loam than in the sandy soil is because finer
347 textured soil has a greater capacity to protect C₄-SOC physically against microbial
348 decomposition by the formation of soil aggregates, and to stabilize the C₄-SOC chemically
349 through association with silt and clay particles (Six *et al.*, 2002). The silty loam soil was also
350 sampled two years later than the sandy soil and therefore it had more time to incorporate C₄-
351 SOC. However, this explains only one third of the observed difference in SOC stocks at most,
352 when a retention coefficient of 10% is assumed for maize residue carbon and yearly stover
353 inputs for GM and SM of 6.06 and 2.53 Mg C ha⁻¹ year⁻¹, respectively (the latter were
354 determined in a separate experiment).

355

356 Native C₃-SOC and total SOC stocks are larger in the sandy than in the silty loam soil. This
357 might result from their past land use and different soil chemical properties. At the end of the
358 18th century, the sandy region was dominated by heathland (de Ferraris, 2009), which was
359 brought into cultivation during the following centuries. Peat sods from the heathland were
360 transferred to livestock stables for use as animal bedding, which was later returned to the field
361 as an organic amendment. This resulted in the development of Plaggic Anthrosols (IUSS
362 Working Group WRB, 2014) that contain large concentrations of refractory organic matter
363 (Springob & Kirchmann, 2002; Kalinina *et al.*, 2009). This recalcitrance can derive partially
364 from the large content of soluble phenolic compounds present in *Calluna vulgaris* L. litter.
365 The latter favours the formation of recalcitrant humic complexes (Kristensen & McCarty,
366 1999), i.e. native C₃-SOC is biochemically stable. The fact that the concentration of SOC in
367 the s+c fraction is about three times larger in the sandy than in the silty loam soil suggests

368 more chemical stabilization in the former. Other soil chemical factors might also be involved,
369 in particular the markedly different oxalate-extractable Al and Fe content of the s+c fraction
370 between both types of soil (Figure 3).

371

372 Six *et al.* (2002) showed that the regression lines between the silt and clay content of the soil
373 and SOC associated with the silt+clay fraction have different slopes for 2:1 clays (dominant in
374 temperate soil) and 1:1 clays (dominant in tropical soil), and for grassland and arable land.

375 The two regions we sampled are only 70 km apart, therefore, the clay mineralogy is likely to
376 be comparable and dominated by 2:1 clays because both soil types developed in a cool, humid

377 climate (Driessen & Dudal, 1991). The lack of effect of cropping system on the functional

378 relation for the sandy region (Figure 2) accords with the observations of Hassink (1997) that

379 the amounts of SOC associated with the silt+clay fraction are no different for arable and

380 grassland soil despite the smaller total SOC content of the former. Hassink (1997) concluded

381 that the silt+clay fraction was saturated with SOC, and that the larger total SOC content of

382 grassland soil could be explained by the greater accumulation of SOC than under arable

383 cropping in the coarser sand-sized fractions. In our study, the sandy soil with either

384 continuous grass or maize does not differ in the SOC content associated with the (s+c)-rSOC

385 and rSOC fractions, whereas the fraction that represents sand and stable aggregates (S + A)

386 contains significantly ($P < 0.01$) more SOC under permanent grass than under maize (Table 3).

387 In contrast, the silty loam soil contains significantly more SOC in the (s+c)-rSOC and rSOC

388 fractions under grass than under maize. This suggests that the s+c fraction is saturated with

389 SOC in the sandy soil, but not in the silty loam soil.

390

391 The difference in carbon sequestration per unit of silt and clay between both soil types might

392 be explained by the oxalate-extractable Al and Fe concentrations of the s+c fraction (Figure

393 3). The relation might be fortuitous because of the confounding effect of inputs from the
394 previous heath vegetation of the sandy soil and different Al and Fe concentrations for both
395 types of soil. The chemical interactions between organic matter and Al and Fe are well
396 established, however. Acid ammonium oxalate extracts amorphous Al and Fe (Schwertmann,
397 1964) as well as organically bound Al and Fe. The former is involved in stabilization of SOC
398 through organo-mineral associations, the latter through formation of organo-metal complexes.
399 Adams & Kassim (1984) extracted Fe from the clay fractions (<2 μm) of several Podzol
400 horizons and demonstrated a strong, positive linear relation between oxalate-extractable Fe in
401 the clay fraction and SOC associated with it. Several incubation studies have shown that
402 sorption on oxyhydroxides is a major mechanism by which Al (Schneider *et al.*, 2010) and Fe
403 (Wagai & Mayer, 2007; Eusterhues *et al.*, 2014) can stabilize SOC. However, Wagai &
404 Mayer (2007) observed that the OC:Fe ratio (g g^{-1}) was larger than the maximum sorptive
405 capacity of the iron oxyhydroxides for the spodic B-horizons of all Podzols investigated.
406 Therefore, other stabilization mechanisms for SOC are important for these types of soil, in
407 particular the complexation of Fe^{3+} ions with functional groups of SOC and subsequent
408 precipitation of these complexes. Both low pH and large SOC content favour the formation of
409 such complexes, not only with Fe (Karlsson *et al.*, 2008) but also with Al (Wagai & Mayer,
410 2007). This might explain why the sandy soil with its lower pH and often with podzolic
411 properties (Table SI-2) can stabilize more SOC than the silty loam soil.

412

413 **Conclusions**

414

415 The acid sandy soil studied is likely to stabilize more SOC in the silt+clay fraction than the
416 silty loam soil by sorption on Al and Fe oxyhydroxides and through the formation of organo-
417 metal complexes. The silt+clay fraction of the sandy soil is likely to be saturated with C,

418 unlike that of the silty loam soil which was suggested by the comparison of SOC fractions of
419 adjacent permanent grassland and arable soil. The concept of carbon saturation also explains
420 why recent C₄-SOC is larger in the unsaturated silty loam soil than in the saturated sandy soil.
421 Stover return under grain maize increases the C₄-SOC in the medium term (≥ 15 years) in both
422 soil types, but the lack of effect on total SOC suggests C-substitution, especially in the C-
423 saturated sandy soil. Increased stover return might have only small effects on soil carbon
424 sequestration in the long term, whereas other factors such as soil mineralogy are more
425 important.

426

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428

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436

437 **Supplementary material**

438

439 Supplementary material is available.

440

441 **References**

- 442 Adams, W.A. & Kassim, J.K. 1984. Iron oxyhydroxydes in soils developed from Lower
443 Palaeozoic sedimentary rocks in Mid-Wales and implications for some pedogenetic
444 processes. *Journal of Soil Science*, **35**, 117–126.
- 445 Allmaras, R.R., Linden, D.R. & Clapp, C.E. 2004. Corn-residue transformations into toot and
446 soil carbon as related to nitrogen, tillage, and stover management. *Soil Science Society of
447 America Journal*, **68**, 1366–1375.
- 448 Balesdent, J., Mariotti, A. & Guillet, B. 1987. Natural ¹³C abundance as a tracer for studies
449 of soil organic matter dynamics. *Soil Biology & Biochemistry*, **19**, 25–30.
- 450 Blaser, P., Kernebeek, P., Tebbens, L., Van Breemen, N., & Luster, J. 1997. Cryptopodzolic
451 Soils in Switzerland. *European Journal of Soil Science*, **48**, 411–423.
- 452 Campbell, C.A., Lafond, G.P., Zentner, R.P. & Biederbeck, V.O. 1991. Influence of fertilizer
453 and straw baling on soil organic matter in a thin black chernozem in western Canada.
454 *Soil Biology & Biochemistry*, **23**, 443–446.
- 455 Clapp, C.E., Allmaras, R.R., Layese, M.F., Linden, D.R. & Dowdy, R.H. 2000. Soil organic
456 carbon and ¹³C abundance as related to tillage, crop residue, and nitrogen fertilization
457 under continuous corn management in Minnesota. *Soil & Tillage Research*, **55**, 127–142.
- 458 Dick, W.A., Blevins, R.L., Frye, W.W., Peters, S.E., Christenson, D.R., Pierce, F.J. & Vitosh,
459 M.L. 1998. Impacts of agricultural management practices on C sequestration in forest-
460 derived soils of the eastern Corn Belt. *Soil & Tillage Research*, **47**, 235–244.

- 461 Dondeyne, S., Vanierschot, L., Langohr, R., Van Ranst, E. & Deckers, J. 2014. *The Soil Map*
462 *of the Flemish Region Converted to the 3rd Edition of the World Reference Base for Soil*
463 *Resources*. KU Leuven, Universiteit Gent & Departement Leefmilieu, Natuur & Energie
464 van de Vlaamse Overheid, Brussels.
- 465 Driessen, P.M. & Dudal, R. 1991. *The Major Soils of the World. Lecture Notes on Their*
466 *Geography, Formation, Properties and Use*. Wageningen University, The Netherlands
467 and Katholieke Universiteit Leuven, Belgium.
- 468 Eusterhues, K., Neidhardt, J., Hädrich, A., Küsel, K. & Totsche, K.U. 2014. Biodegradation
469 of ferrihydrite-associated organic matter. *Biogeochemistry*, **119**, 45–50.
- 470 Evans, L.J. & Wilson, W.G. 1985. Extractable Fe, Al, Si and C in B horizons of podzolic and
471 brunisolic soils from Ontario. *Canadian journal of soil science*, **65**, 489–496.
- 472 De Ferraris, J. 2009. *De Grote Atlas van Ferraris : De Eerste Atlas van België : 1777 :*
473 *Kabinetskaart van de Oostenrijkse Nederlanden en het Prinsbisdom Luik (The First*
474 *Atlas of Belgium: 1777: Cabinet Map of the Austrian Netherlands and the Prince-*
475 *Bishopric of Liège)*. Lannoo, Tielt.
- 476 Gerin, P.A., Vliegen, F. & Jossart, J.M. 2008. Energy and CO₂ balance of maize and grass as
477 energy crops for anaerobic digestion. *Bioresource Technology*, **99**, 2620–2627.
- 478 Gregorich, E.G., Drury, C.F. & Baldock, J.A. 2001. Changes in soil carbon under long-term
479 maize in monoculture and legume-based rotation. *Canadian Journal of Soil Science*, **81**,
480 21–31.
- 481 Hassink, J. 1997. The capacity of soils to preserve organic C and N by their association with
482 clay and silt particles. *Plant and Soil*, **191**, 77–87.

- 483 Hooker, B.A., Morris, T.F., Peters, R. & Cardon, Z.G. 2005. Long-term effects of tillage and
484 corn stalk return on soil carbon dynamics. *Soil Science Society of America Journal*, **69**,
485 188–196.
- 486 Huggins, D.R., Buyanovsky, G.A., Wagner, G.H., Brown, J.R., Darmody, R.G., Peck, T.R., *et*
487 *al.* 1998. Soil organic C in the tallgrass prairie-derived region of the corn belt: effects of
488 long-term crop management. *Soil and Tillage Research*, **47**, 219–234.
- 489 IUSS Working Group WRB 2014. *World Reference Base for Soil Resources 2014.*
490 *International Soil Classification System for Naming Soils and Creating Legends for Soil*
491 *Maps. World Soil Resources Reports No. 106.* FAO, Rome.
- 492 Kalinina, O., Chertov, O., Nadporozhskaya, M. & Giani, L. 2009. Properties of soil organic
493 matter of plaggic anthrosols from Northwest Germany, Northwest and North Russia.
494 *Archives of Agronomy and Soil Science*, **55**, 477–492.
- 495 Karlsson, T., Persson, P., Skyllberg, U., Mörth, C.-M. & Giesler, R. 2008. Characterization of
496 iron(III) in organic soils using extended X-ray absorption fine structure spectroscopy.
497 *Environmental Science & Technology*, **42**, 5449–5454.
- 498 Kristensen, H.L. & McCarty, G.W. 1999. Mineralization and immobilization of nitrogen in
499 heath soil under intact *Calluna*, after heather beetle infestation and nitrogen fertilization.
500 *Applied Soil Ecology*, **13**, 187–198.
- 501 Kuzyakov, Y., Friedel, J.K. & Stahr, K. 2000. Review of mechanisms and quantification of
502 priming effects. *Soil Biology & Biochemistry*, **32**, 1485–1498.

503 Larson, W.E., Clapp, C.E., Pierre, W.H. & Morachan, Y.B. 1972. Effects of increasing
504 amounts of organic residues on continuous corn: II. Organic carbon, nitrogen,
505 phosphorus, and sulfur. *Agronomy Journal*, **64**, 204–208.

506 Odell, R.T., Melsted, S.W. & Walker, W.M. 1984. Changes in organic carbon and nitrogen of
507 Morrow plot soils under different treatments, 1904–1973. *Soil Science*, **137**, 160–171.

508 The R Foundation. 2015. R: The R Project for Statistical Computing (At: [http://www.R-](http://www.R-project.org/)
509 [project.org/](http://www.R-project.org/) . Accessed: 27/02/2016). R Foundation for Statistical Computing, Vienna,
510 Austria.

511 Rasmussen, P.E., Allmaras, R.R., Rhode, C.R. & Roager, N.C. 1980. Crop residue influences
512 on soil carbon and nitrogen in a wheat-fallow system. *Soil Science Society of America*
513 *Journal*, **44**, 596–600.

514 Reicosky, D.C., Evans, S.D., Cambardella, C.A., Allmaras, R.R., Wilts, A.R. & Huggins,
515 D.R. 2002. Continuous corn with moldboard tillage: Residue and fertility effects on soil
516 carbon. *Journal of Soil and Water Conservation*, **57**, 277–284.

517 Robinson, C.A., Cruse, R.M. & Ghaffarzadeh, M. 1996. Cropping system and nitrogen effects
518 on Mollisol organic carbon. *Soil Science Society of America Journal*, **60**, 264–269.

519 Schneider, M.P.W., Scheel, T., Mikutta, R., van Hees, P., Kaiser, K. & Kalbitz, K. 2010.
520 Sorptive stabilization of organic matter by amorphous Al hydroxide. *Geochimica et*
521 *Cosmochimica Acta*, **74**, 1606–1619.

522 Schwertmann, U. 1964. Differenzierung der Eisenoxide des Bodens durch Extraktion mit
523 Ammoniumoxalat Lösung (Differentiation of iron oxide in soils by extraction with an

524 ammonium oxalate solution). *Zeitschrift für Pflanzenernährung, Düngung, Bodenkunde*,
525 **105**, 194–202.

526 Six, J., Conant, R.T., Paul, E.A. & Paustian, K. 2002. Stabilization mechanisms of soil organic
527 matter: implications for C-saturation of soils. *Plant and Soil*, **241**, 155–176.

528 Springob, G. & Kirchmann, H. 2002. C-rich sandy A_p horizons of specific historical land-use
529 contain large fractions of refractory organic matter. *Soil Biology & Biochemistry*, **34**,
530 1571–1581.

531 Stolbovoy, V., Montanarella, L., Filippi, N., Jones, A., Gallego, J. & Grassi, G. 2007. *Soil*
532 *Sampling Protocol to Certify the Changes of Organic Carbon Stock in Mineral Soil of*
533 *the European Union, Version 2*. Office for Official Publications of the European
534 Communities, Luxembourg.

535 Wagai, R. & Mayer, L.M. 2007. Sorptive stabilization of organic matter in soils by hydrous
536 iron oxides. *Geochimica et Cosmochimica Acta*, **71**, 25–35.

537 Webster, R. 1997. Regression and functional relations. *European Journal of Soil Science*, **48**,
538 557-556.

539 Wilhelm, W.W., Johnson, J.M.F, Hatfield, J.L, Voorhees, W.B. & Linden, D.R. 2004. Crop
540 and soil productivity response to corn residue removal. *Agronomy Journal*, **96**, 1–17.

541 Wilts, A.R., Reicosky, D.C., Allmaras, R.R. & Clapp, C.E. 2004. Long-term corn residue
542 effects: Harvest alternatives, soil carbon turnover and root-derived carbon. *Soil Science*
543 *Society of America Journal*, **68**, 1342–1351.

544 Zimmermann, M., Leifeld, J., Schmidt, M.W.I., Smith, P. & Fuhrer, J. 2007. Measured soil
545 organic matter fractions can be related to pools in the RothC model. *European Journal of*
546 *Soil Science*, **58**, 658–667.

547

548

549 Table 1 Mean SOC and $\delta^{13}\text{C}$ in the top 30 cm of soil with selected cropping histories in the sandy and silty loam regions. The fraction of maize-
 550 derived SOC (Equation (2)), $\text{C}_4\text{-SOC}$ and $\text{C}_3\text{-SOC}$ were determined for cropping histories SM, SMGRA and GM. The standard error of the mean
 551 is also given.

Cropping History	SOC / g C 100 g ⁻¹ soil	SOC / Mg C ha ⁻¹	$\delta^{13}\text{C}$ / ‰	Maize-derived SOC fraction / %	Maize-derived C ₄ -SOC / g C 100 g ⁻¹ soil	Maize-derived C ₄ -SOC / Mg C ha ⁻¹	Native C ₃ -SOC / g C 100 g ⁻¹ soil	Native C ₃ -SOC / Mg C ha ⁻¹
Sand region								
PA†	2.24 ± 0.12	91.19 ± 4.18	-28.40 ± 0.13					
SM	1.85 ± 0.10	78.76 ± 3.47	-26.63 ± 0.13	11.3 ± 0.8	0.21 ± 0.02	8.86 ± 0.74	1.64 ± 0.09	69.90 ± 3.17
SMGRA	1.65 ± 0.06	73.21 ± 2.41	-26.41 ± 0.18	12.7 ± 1.1	0.20 ± 0.01	9.07 ± 0.66	1.44 ± 0.06	64.15 ± 2.60
GM	1.76 ± 0.10	75.34 ± 3.45	-25.53 ± 0.18	18.3 ± 1.2	0.32 ± 0.02	13.74 ± 1.02	1.44 ± 0.09	61.60 ± 3.08
Silt Loam Region								
PA	2.44 ± 0.28	94.23 ± 8.99	-28.20 ± 0.22					
SM	1.21 ± 0.08	54.16 ± 2.96	-24.04 ± 0.20	26.9 ± 1.3	0.32 ± 0.02	14.32 ± 0.68	0.89 ± 0.08	39.84 ± 2.70
GM	1.20 ± 0.12	54.64 ± 4.89	-23.02 ± 0.38	33.5 ± 2.5	0.38 ± 0.01	17.30 ± 0.56	0.83 ± 0.11	37.33 ± 4.81

552 † PA = permanent grass (pasture), SM = monoculture of silage maize, SMGRA = monoculture of silage maize where the maize is followed by a second annual grass crop, GM = monoculture of
 553 grain maize

554 Table 2 (a) One-way ANOVA table for the dependent variable ‘maize-derived C₄-SOC (Mg C
555 ha⁻¹)’ for the sandy region with averaged within-field replicates , (b) two-way ANOVA table
556 for the dependent variable ‘maize-derived C₄-SOC (Mg C ha⁻¹)’ for the two regions combined
557 with averaged within-field replicates, (c) one-way ANOVA table for the dependent variable
558 ‘native C₃-SOC (Mg C ha⁻¹)’ for the sandy region with averaged within-field replicates and
559 (d) two-way ANOVA table for the dependent variable ‘native C₃-SOC (Mg C ha⁻¹)’ for the two
560 regions combined with averaged within-field replicates .

561 (a)

Source	Degrees of freedom	Sum of squares	Mean square	<i>F</i> ratio	<i>P</i>
Crop	2	154.9	77.44	11.21	< 0.001
Residuals	28	193.4	6.91		
Total	30	348.3	11.61		

567 (b)

Source	Degrees of freedom	Sum of squares	Mean square	<i>F</i> ratio	<i>P</i>
Region	1	342.7	342.7	59.67	< 0.001
Crop	2	190.2	95.1	16.56	< 0.001
Region × Crop	1	9.2	9.2	1.60	0.21
Residuals	46	264.2	5.7		
Total	50	806.3	16.1		

575 (c)

Source	Degrees of freedom	Sum of squares	Mean square	<i>F</i> ratio	<i>P</i>
Crop	2	382.3	191.17	2.08	0.14
Residuals	28	2570.8	91.81		
Total	30	2953.1	98.44		

581 (d)

Source	Degrees of freedom	Sum of squares	Mean square	<i>F</i> ratio	<i>P</i>
Region	1	8718	8718	75.55	< 0.001
Crop	2	328	164	1.42	0.25
Region × Crop	1	86	86	0.74	0.39
Residuals	46	5309	115		
Total	50	14 441	289		

584 Table 3 Mean concentrations of SOC in the different SOC fractions of soil with selected
 585 cropping histories in the sandy and silty loam regions. The standard error of the mean is also
 586 reported. The contribution of the SOC associated with a fraction relative to the total SOC of
 587 the bulk soil (percentage of total SOC) is reported in brackets.

Cropping History	POM*	DOC	S+A	(s+c)-rSOC	rSOC
/ g C kg ⁻¹ soil					
Sand region					
PA ^{\$}	1.51 ± 0.39 (8)	0.45 ± 0.03 (2)	6.64 ± 0.55 (32)	10.20 ± 0.82 (49)	1.83 ± 0.35 (9)
SM	1.24 ± 0.18 (7)	0.40 ± 0.03 (2)	3.82 ± 0.45 (21)	12.05 ± 1.17 (63)	1.51 ± 0.13 (8)
SMGRA	2.04 ± 0.41 (11)	0.35 ± 0.02 (2)	4.22 ± 0.61 (24)	9.00 ± 0.50 (52)	1.79 ± 0.25 (11)
GM	1.30 ± 0.15 (7)	0.39 ± 0.02 (2)	3.88 ± 0.45 (22)	9.84 ± 0.82 (57)	2.10 ± 0.56 (11)
Silt Loam Region					
PA	1.97 ± 0.30 (10)	0.69 ± 0.07 (3)	6.14 ± 1.65 (27)	10.28 ± 1.43 (49)	2.37 ± 0.43 (11)
SM	2.35 ± 0.34 (19)	0.29 ± 0.02 (2)	1.44 ± 0.25 (13)	6.54 ± 0.71 (53)	1.53 ± 0.11 (13)
GM	1.83 ± 0.26 (16)	0.30 ± 0.02 (3)	1.42 ± 0.46 (12)	6.87 ± 0.99 (58)	1.38 ± 0.14 (12)

588 \$ PA, SM, SMGRA and GM: see Table 1

589 * POM = particulate organic matter, DOC = dissolved organic carbon, S+A = SOC associated with the fraction containing
 590 sand and stable aggregates, (s+c)-rSOC = oxidizable part of the SOC associated with the silt and clay fraction, rSOC = non-
 591 oxidizable, chemically resistant part of the SOC associated with the silt and clay fraction

592

593 **Figure 1** The $\delta^{13}\text{C}$ in the bulk soil (BS) and in the different SOC fractions of soil with
594 selected cropping histories in (a) the sandy region and (b) the silty loam region, and the
595 fraction of SOC derived from maize (%) determined by Equation (2) for the bulk soil and
596 different SOC fractions in soils with selected cropping histories in (c) the sandy region and (d)
597 the silty loam region. Abbreviations of cropping histories and SOC fractions are given in
598 Tables 1 and 3.

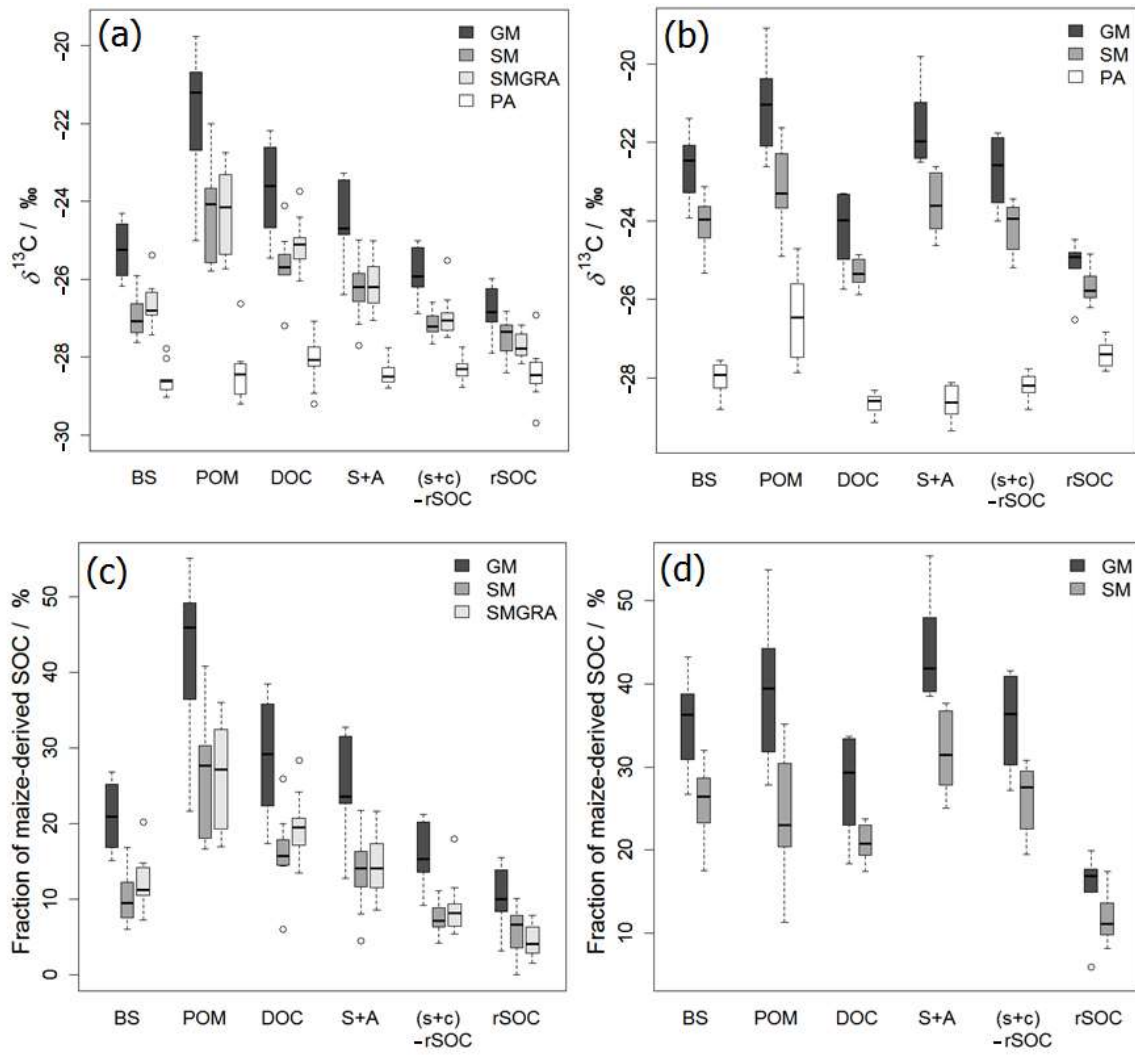
599

600 **Figure 2** Functional relation between silt + clay content and the amount of SOC associated
601 with the silt and clay fraction. All cropping histories are combined. The line for the sandy soil
602 is the estimated functional relation (based on Equations (18) and (19) in Webster, 1997) of
603 SOC associated with silt and clay as the dependent variable on silt + clay content. No such
604 meaningful relation could be demonstrated for the silty loam soil.

605

606 **Figure 3** Functional relation between the concentrations of oxalate-extractable Al and Fe and
607 the amount of SOC associated with the silt and clay fraction for the sandy and the silty loam
608 soil. All cropping histories are combined. The line is the estimated functional relation (based
609 on Equations (18) and (19) in Webster, 1997) of SOC amount associated with silt and clay as
610 the dependent variable on oxalate-extractable Al and Fe for the sandy and silty loam soil
611 combined.

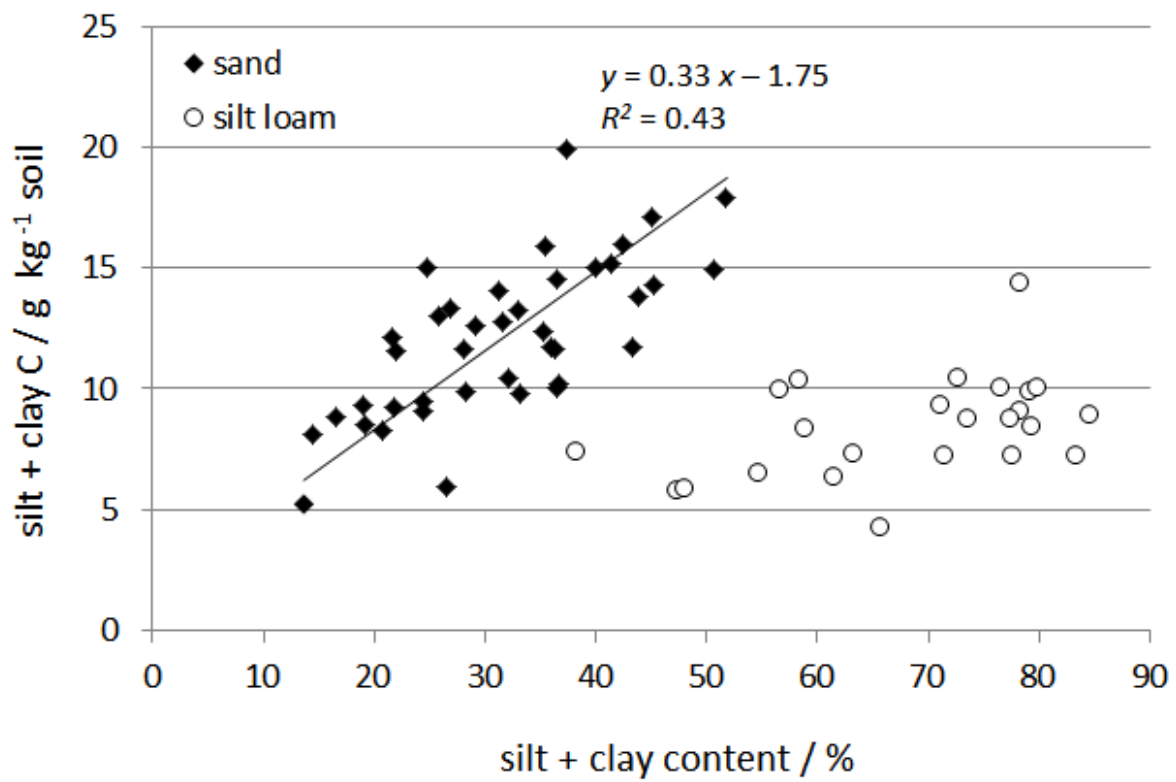
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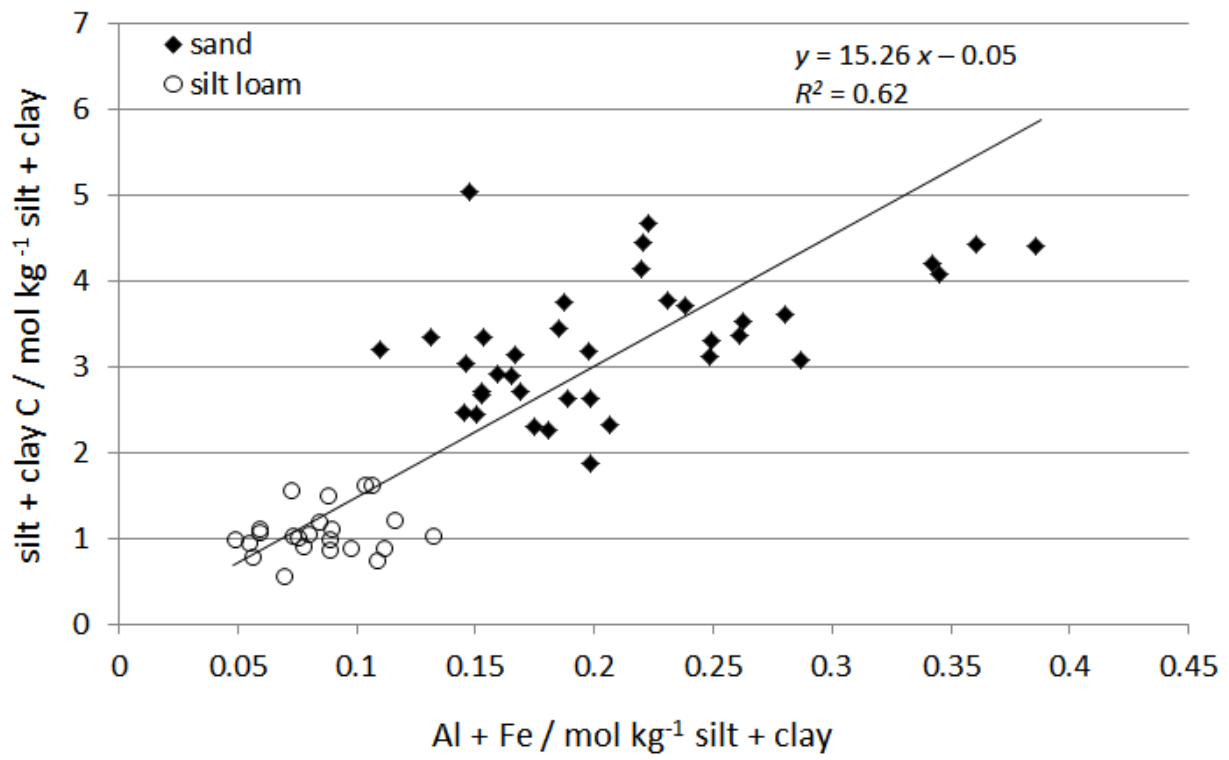
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614 Figure 1

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617 Figure 2
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 620 Figure 3
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