

1 **Soil nutrient contents and stoichiometry within aggregate size classes varied with tea**  
2 **plantation age and soil depth in the southern Guangxi of China**

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8 **ABSTRACT**

9 Soil ecological stoichiometry offers a tool to explore the distribution, cycling, limitation, and  
10 balance of chemical elements in the tea plantation ecosystems. This study aimed to explore how  
11 soil organic C (OC) and nutrient contents (total N (TN), total P (TP),  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Fe}^{2+}$ , and  $\text{Mn}^{2+}$ )  
12 as well as their stoichiometric ratios (C/N, C/P, N/P, Ca/Mg, and Fe/Mn) varied with tea  
13 plantation age (8, 17, 25, and 43 years) and soil depth (0-10, 10-20, 20-40, and 40-60 cm) within  
14 aggregates in the southern Guangxi of China. Our results showed that tea plantation age and soil  
15 depth significantly affected soil nutrient stoichiometry in different sizes of aggregates. Among  
16 different ages of tea plantations, soil OC, TN, and TP contents as well as C/N, C/P, and N/P  
17 ratios significantly decreased when the soil depth increased. In addition, soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$   
18 contents were significantly lower in the surface soil layer than the deeper soil layer, whereas soil  
19  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  contents showed the totally opposite trends, and no significant differences were  
20 detected in Ca/Mg and Fe/Mn ratios among different soil depths. At the 0-40 cm soil depth,  
21 continuous planting of tea was beneficial for the significant increases in soil OC, TN,  $\text{Fe}^{2+}$ , and  
22  $\text{Mn}^{2+}$  contents, whereas soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents significantly decreased over time. During  
23 the process of tea growth, the losses of soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , especially the  $\text{Ca}^{2+}$  (as indicated by  
24 the decrease in soil Ca/Mg ratio), led to soil acidification. In the meanwhile, soil acidification  
25 reduced  $\text{Fe}^{2+}$  absorption and enhanced  $\text{Mn}^{2+}$  uptake by tea plants (as indicated by the increase in  
26 soil Fe/Mn ratio). In general, tea plantation age affected the variations of soil nutrient contents

27 and stoichiometry, and such effects were more obvious at the 0-40 cm soil depth in contrast to  
28 the 40-60 cm soil depth.

## 29 **KEYWORDS**

30 Tea plantation age; Soil depth; Soil aggregate; Ecological stoichiometry

31

## 32 **1. Introduction**

33 In the past century, under the remarkable increase in population pressure, continuous tillage  
34 and overmuch deforestation resulted in the dramatic decrease in soil fertility level in the southern  
35 Guangxi of China (Jiang et al., 2018). To overcome the existing challenges, the Chinese  
36 government has rolled out the Grain for Green program in the hope of alleviating land  
37 deterioration via converting farmlands to forest lands or grass lands (Zeng et al., 2020). Since the  
38 initiation of such program, the south part of Guangxi has initiated the mode of transforming  
39 farmlands into tea (*Camellia sinensis* L.) plantations as per the local geography and natural  
40 resources (Zhang et al., 2017). Tea, as a pivotal cash crop, is commonly cultivated in the  
41 developing nations, particularly in China, India, Kenya, and Sri Lanka. China is the world's  
42 largest producer of tea, with the tea-planting area reaching 3.17 million hectares in 2020, and it  
43 shows an elevating trend in the future (Chinese Tea Committee, 2020). Guangxi has the  
44 subtropic monsoon climate and marks the key tea-planting region in China. According to the  
45 statistics from Chinese Tea Committee (2020), over 80% tea plantations of Guangxi are situated  
46 at the impoverished counties, and tea-planting industry turns to be the staple industry on which  
47 poor counties depend to throw off poverty.

48 Ecological stoichiometry offers a tool to explore the distribution, cycling, restriction, and  
49 balance of nutrients in terrestrial ecosystems (Yu et al., 2019), and exerts a critical role in  
50 recognizing the influencing factors and driving mechanisms in ecological processes (Su et al.,  
51 2019). Carbon (C) is the most commonly seen element in plants (Prescott et al., 2020), and  
52 nitrogen (N) and phosphorus (P) are the critical control factors for the growth of plants (Krouk

53 and Kiba, 2020). The relationships amongst C, N, and P are coupled (Elser et al., 2003) and their  
54 stoichiometric ratios (C/N, C/P, and N/P) reflect the nutrient status during the process of soil  
55 genesis, making them being important indicators of soil quality (Bai et al., 2020). Additionally,  
56 calcium (Ca), magnesium (Mg), iron (Fe), and manganese (Mn) are the pivotal metallic nutritive  
57 elements for the development of plants (Liu et al., 2021a). The contents of soil total Ca, Mg, Fe,  
58 and Mn may exceed the demand of a single plant by more than a thousand-fold and cannot  
59 sensitively reflect the needs of plants (Miner et al., 2018), but the contents of these nutrients'  
60 available fractions may be insufficient or redundant, resulting in the deficiencies or abundances  
61 of plant nutrients (Otero et al., 2013). Thus, soil exchangeable Ca and Mg, together with the  
62 available Fe and Mn significantly affect the development of plants.

63 Over the past decade, soil nutrient stoichiometry (mainly C-N-P, rather than Ca-Mg or  
64 Fe-Mn) has been broadly investigated across the world (Tian et al., 2010; Yang et al., 2013;  
65 Zhang et al., 2016; Yue et al., 2017; Yu et al., 2018; Qiao et al., 2020). It has been widely  
66 acknowledged amongst these studies that soil depth is vital for the regulation of soil nutrient  
67 stoichiometry. Substantial studies have identified the decreasing trend of soil organic C (OC),  
68 total N (TN), and total P (TP) contents with the increase in soil depth (Yue et al., 2017; Yu et al.,  
69 2018; Qiao et al., 2020), whereas conflicting vertical patterns are discovered for soil C/N, C/P,  
70 and N/P ratios. For instance, a decreasing trend of the C/P and N/P ratios can be observed with  
71 the increase in soil depth, according to the data of the 2<sup>nd</sup> soil investigation in China (Tian et al.,  
72 2010). Nevertheless, a larger C/N ratio in the deeper soil layer, not the surface soil layer, is  
73 identified in a mollisol plain in the northeast China, because soil total C was measured and  
74 carbonates were observed in the deeper soil layer (Zhang et al., 2016). Moreover, the C/N ratio  
75 displays no remarkable change at different soil depths, as revealed by the investigation of alpine  
76 grassland on the Qingzang Plateau (Yang et al., 2013). As shown above, inconsistent vertical  
77 patterns have been reported for the C-N-P stoichiometric ratios in different soil ecosystems.

78 As the basic unite of soil structure, soil aggregates are complex ensembles composed of  
79 primary particles and organic matter (OM) (Tisdall and Oades, 1982). According to the  
80 differences in binding agents, soil aggregates can be classified into microaggregates (< 0.25 mm)  
81 and macroaggregates (> 0.25 mm) (Tisdall and Oades, 1982). In general, persistent binding  
82 agents (including humified OM and polyvalent metal cation complexes) contribute to the binding  
83 of primary particles into microaggregates (Six et al., 2004). On the contrary, temporary binding  
84 agents (including fungal hyphae, plant roots, and polysaccharides) aggregate with  
85 microaggregates, which facilitate to the formation of macroaggregates (Six et al., 2004). Based  
86 on the above discussion, soil aggregates with various sizes have different abilities in the supply  
87 and reserve of soil OC and nutrients. Thus, to improve the comprehension on the structure and  
88 function of soil ecosystems, more efforts should be made to observe the soil nutrient  
89 stoichiometry within aggregates (Xu et al., 2019; Cui et al., 2021). Recently, although lots of  
90 studies have reported the OC, TN, and TP distribution in different sizes of aggregates, these  
91 studies are ended with different results. To be specific, some studies reveal the significant  
92 increases in the OC, TN, and TP contents with the decrease in aggregate size (Sarker et al., 2018;  
93 Piazza et al., 2020). Nevertheless, some other studies draw the totally opposite trends (Lu et al.,  
94 2019; Liu et al., 2021b). These demonstrate that the changes in soil OC, TN, and TP contents  
95 within aggregates have received great attention, whereas soil exchangeable alkali cations (i.e.,  
96  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) and available micronutrients (i.e.,  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ ) are rarely investigated.

97 Our previous studies indicated that the landuse shift from farmlands to tea plantations  
98 ameliorated the soil fertility level (Zheng et al., 2011). However, during the process of tea  
99 growth, the variation of soil nutrient stoichiometry remains unclear. Meanwhile, since tea serves  
100 as a deep root plant, it is vital to reveal how nutrient stoichiometry changes with the increasing  
101 soil depth in the tea plantation ecosystems. Thus, this study was carried out to investigate how  
102 soil OC and nutrient contents as well as their stoichiometric ratios varied with tea plantation age  
103 (8, 17, 25, and 43 years) and soil depth (0-10, 10-20, 20-40, and 40-60 cm) within aggregates (<

104 0.25, 0.25-1, 1-2, and > 2 mm). We hypothesized that (i) soil OC and TN contents would  
105 increase with tea plantation age due to the annual fertilization, and (ii) decreases in soil Ca<sup>2+</sup> and  
106 Mg<sup>2+</sup> contents would be accompanied by increases in soil Fe<sup>2+</sup> and Mn<sup>2+</sup> contents because of soil  
107 acidification during the process of tea growth.

## 108 **2. Materials and methods**

### 109 2.1. Experiment site

110 In January 2019, this study was completed at the Hengxian Agriculture Experiment Center  
111 of Guangxi University (altitude of 557-563 m and slope degree of 13-15 °). The climate at the  
112 experiment site is dominated by subtropic monsoon climate, with the yearly average rainfall and  
113 temperature being 1304 mm and 21.6 °C, respectively. Exposed soil horizon occurs early in the  
114 Mesozoic, which gradually forms the Ultisols agrotypic (IUSS Working Group, 2014). As early  
115 as in 1960s, due to the high economic value of tea, massive hectares of farmlands were  
116 developed to tea plantations in this region.

117 The “*Baimao* tea” refers to a major cultivar in this area, and the ages of these tea plantations  
118 are distinct. Managed by different owners, tea plantations were both experimental trials  
119 (Guangxi University) and commercial plantings. In the tea-planting course, the tillage method  
120 was no tillage and tea-planting density was almost  $6 \times 10^4$  plants ha<sup>-1</sup>. Herbicides were not  
121 applied and yellow sticky boards were used to prohibit pests, because this color might attract  
122 pests and got them stuck on the boards. In addition, all the tea plants were subject to slight  
123 pruning in September each year.

124 An annual fertilizer regime in tea plantations was shown below. Both 0.65 Mg ha<sup>-1</sup> complex  
125 fertilizer (granule, N-P<sub>2</sub>O<sub>5</sub>-K<sub>2</sub>O: 18%-6%-6%) and 12 Mg ha<sup>-1</sup> swine manure (slurry,  
126 N-P<sub>2</sub>O<sub>5</sub>-K<sub>2</sub>O: 0.54%-0.48%-0.36%) were applied vertically below tree crown yearly in  
127 mid-November as the basal fertilizer at the surrounding region. Subsequently, the top-dressing,  
128 applied to the site treated with replenished basal fertilizer, was replenished thrice per year. Both  
129 1.2 Mg ha<sup>-1</sup> complex fertilizer and 0.5 Mg ha<sup>-1</sup> urea were applied onto soil surface in mid-March,

130 while 0.65 Mg ha<sup>-1</sup> complex fertilizer and 0.3 Mg ha<sup>-1</sup> urea were applied in late-June and in  
131 early-September.

## 132 2.2. Experiment design

133 In general, examining the same location persistently has been considered as an effective  
134 approach to monitor the variations of soil with time (Sparling et al., 2003). Nevertheless, the  
135 challenges in long-time soil monitoring have made it urgent to develop the substitutional  
136 approaches to investigate the changes in soil over time, amongst which the most common  
137 approach is the ‘space-for-time’ alternative (Zanella et al., 2018).

138 In this study, this approach was used to explore the variation of soil nutrient stoichiometry  
139 in a chronological sequence of tea plantations. In general, confounding factors existed in the  
140 spatial variations of soil, hence the present study managed to mitigate such effects by choosing  
141 tea plantations, which were cultured with the same tea variety (“*Baimao* tea”) with different  
142 planting ages (8, 17, 25, and 43 years), and were located at the same unit associated with  
143 geomorphological status.

144 Each of the 4 tea plantation age groups was replicated in 5 locations for totally 20  
145 experimental units (Figure 1). Separation amongst these units was completed with distances of >  
146 800 m between each other, thereby decreasing the spatial autocorrelation and avoiding the  
147 pseudo-replication. For every unit ( $S \approx 1 \times 10^4 \text{ m}^2$ ), a plot ( $S = 20 \text{ m} \times 20 \text{ m}$ ) was randomly  
148 established with distance of > 50 m away from the unit margin.

## 149 2.3. Litter and soil sampling

150 For every plot, 5 surface litter (a stock) specimens were acquired from the surface of soil in  
151 the 5 randomly chosen subplots ( $S = 1 \text{ m} \times 1 \text{ m}$ ), which were afterwards integrated into a  
152 composite litter specimen. In total, 20 (4 tea plantation ages  $\times$  5 replicates) composite litter  
153 specimens were desiccated at 80 °C until the weight became constant. Then, the weights of these  
154 desiccated litter specimens were measured, and the litter C (Nelson and Sommers, 1996) and N  
155 (Bremner, 1996) contents were detected. The amounts of litter were 821, 974, 786, and 648 g m<sup>-2</sup>

156 in the 8, 17, 25, and 43 years of tea plantations, respectively, and the C/N ratios of litter were  
157 14.23, 12.68, 17.32, and 21.37, respectively.

158 Soil sampling was completed at the same sites of litter sampling. For every plot, 5 soil  
159 specimens were acquired by a spade from every soil layer (i.e., 0-10, 10-20, 20-40, and 40-60  
160 cm) in the 5 subplots ( $S = 1 \text{ m} \times 1 \text{ m}$ ), which were afterward integrated into a composite soil  
161 specimen. In total, 80 (4 tea plantation ages  $\times$  4 soil layers  $\times$  5 replicates) composite soil  
162 specimens were gently separated into the naturally formed aggregates, and were then sieved by a  
163 5-mm sifter to remove small stones, coarse roots, and macrofauna. Afterwards, soil specimens  
164 were used for the aggregate separation. For every plot, another 5 soil specimens were randomly  
165 chosen via cutting rings (volume =  $100 \text{ cm}^3$ , diameter = 50.46 mm, and depth = 50 mm) from  
166 every soil layer to assess the bulk density, clay ( $< 0.002 \text{ mm}$ ), pH, OC, and nutrients of bulk soil.

#### 167 2.4. Soil aggregate separation

168 According to the wet screening process, 250 g of every composite soil specimen was sieved  
169 via the 2, 1, and 0.25 mm sieves successively (Kemper and Chepil, 1965). To be specific, the  
170 composite soil specimens were soaked by the aqua destillata for 15 min, and were then oscillated  
171 in the vertical direction for 15 min at the  $1 \text{ s}^{-1}$  oscillating rate and 5 cm amplitude. Consequently,  
172 we obtained 4 different sizes of aggregates, covering microaggregates ( $< 0.25 \text{ mm}$ ), fine (0.25-1  
173 mm), medium (1-2 mm), and coarse ( $> 2 \text{ mm}$ ) macroaggregates. All the aggregates were  
174 desiccated and weighted, and later the aggregate-related OC and nutrients were detected.

#### 175 2.5. Soil property analyses

176 Prior to the analyses of soil physical-chemical properties, soil specimens were subject to  
177 atmospheric drying under indoor temperature condition. According to the cutting ring method  
178 (Lu, 2000), soil specimens were oven-dried at  $105 \text{ }^\circ\text{C}$  until constant weight, so as to measure the  
179 bulk density. Soil clay was detected by the hydrometer (TM-85, Veichi, China) (Lu, 2000). Soil  
180 pH was detected by the glassy electrode (MT-5000, Ehsy, China), with the ratio of soil : water  
181 (mass : volume) being 1 : 2.5 (Lu, 2000). Soil OC and TN were identified via the acid dichromate

182 wet oxidation method (Nelson and Sommers, 1996) and the micro-Kjeldahl method (Bremner,  
183 1996), respectively. Soil TP was identified via the molybdate blue colorimetry method (Bray and  
184 Kurtz, 1945). Soil exchangeable alkali cations (i.e.,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) were determined by the  
185 ammonium acetate ( $\text{CH}_3\text{COONH}_4$ ) (Thomas, 1982). Briefly, 2.5 g of every aggregate fraction  
186 was weighed into the Erlenmeyer flask to blend with 50 mL 1 M  $\text{CH}_3\text{COONH}_4$  (pH = 7.0). The  
187 extract liquid was agitated for 30 min under 150 rpm, and then sieved via Whatman No. 2 V  
188 filtration paper (quantitative and ashfree). Soil available micronutrients (i.e.,  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ )  
189 were determined by the diethylenetriamine pentaacetic acid (DTPA) (Lindsay and Norvell,  
190 1978). Briefly, 10 g of every aggregate fraction was weighed into the Erlenmeyer flask to blend  
191 with 20 mL 0.005 M DTPA + 0.01 M  $\text{CaCl}_2$  + 0.1 M TEA (triethanolamine) (pH = 7.0). The  
192 extract liquid was agitated for 2 h under 180 rpm, and then sieved. The entire extractable  
193 metallic cations were detected by the atomic absorption spectrometer (AAS, Shimadzu, Japan).  
194 In this study, 5 standard specimens (GBW-07401), 5 blank specimens, and 80 analytical  
195 replicates (accounted for 20% of the total soil specimens) were used to control quality. Besides,  
196 the error between parallel specimen and experimental specimen was controlled in 5%.

## 197 2.6. Calculations and statistics

198 The mean weight diameter (MWD, mm) was utilized to indicate the stability of soil  
199 aggregates. To be specific, the higher MWD value indicated the stronger aggregate stability  
200 (Kemper and Chepil, 1965):

$$201 \text{MWD} = \sum_{i=1}^4 (X_i \times M_i),$$

202 in the formula,  $X_i$  indicates the  $i^{\text{th}}$  size aggregates' mean diameter (mm) and  $M_i$  indicates the  
203  $i^{\text{th}}$  size aggregates' proportion (% in weight).

204 SPSS 22.0 software (SPSS, Inc., Chicago, IL, USA) was used for statistic analysis (Table  
205 1). Means were tested by the Tukey's HSD and the significant level was set at  $p \leq 0.05$ .  
206 Two-way analysis of variance (ANOVA) was used for exploring the effects of soil depth, tea  
207 plantation age, and their interactions on the physico-chemical properties of bulk soil. Three-way



208 ANOVA was used for exploring the effects of soil depth, tea plantation age, aggregate size, and  
209 their interactions on the physico-chemical properties of soil aggregates. Besides that, Pearson  
210 correlation analysis was utilized to test the relationships between pH and stoichiometric ratios  
211 (i.e., Ca/Mg and Fe/Mn) in bulk soil during the process of tea growth.

### 212 **3. Results**

#### 213 3.1. Soil bulk density, clay content, and pH

214 At the 0-10 and 10-20 cm soil depths, bulk density significantly decreased within the first  
215 17 years and afterwards significantly increased, whereas the effect of tea plantation age on the  
216 bulk density is limiting at the 20-40 and 40-60 cm soil depths (Tables 1 and 2). Regardless of the  
217 tea plantation age, a significant increase in bulk density was observed when the soil depth  
218 increased. A two-way ANOVA analysis showed that the effects of soil depth, tea plantation age,  
219 and their interactions on the clay content were not significant (Tables 1 and 2). Soil pH  
220 significantly decreased during the process of tea growth (Tables 1 and 2). Moreover, no  
221 significant variation in soil pH was observed with the increasing soil depth.

#### 222 3.2. Composition and stability of soil aggregates

223 At the 0-10 and 10-20 cm soil depths, continuous planting of tea resulted in significant  
224 variations in the proportions of different sizes of aggregates, apart from the medium and fine  
225 macroaggregates (Table 3). To be specific, the proportions of coarse macroaggregates  
226 significantly rose within the first 17 years and then significantly dropped, whereas the  
227 proportions of microaggregates displayed an opposite trend over time. At the same time, the  
228 greatest value of soil MWD was identified in the tea plantations of 17 years (Table 3). Notably,  
229 the role of tea plantation age in the aggregate composition and stability is limited at the 20-40  
230 and 40-60 cm soil depths. Across the 4 tea plantation ages, the coarse macroaggregates were  
231 dominant at the 0-10 cm soil depth, accounting for 32.60%-53.18% of bulk soil. However, at the  
232 10-20, 20-40, and 40-60 cm soil depths, the microaggregates were dominant, accounting for  
233 33.80%-49.51%, 42.12%-48.24%, and 44.80%-49.45%, respectively. According to the obtained

234 results, the coarse macroaggregate proportions significantly reduced while the microaggregate  
235 proportions significantly elevated with the increasing soil depth.

### 236 3.3. Contents of soil C, N, and P

237 Within aggregate size classes, soil OC (Figure 2) and TN (Figure 3) contents significantly  
238 increased with the increasing aggregate size, while the distribution of soil TP (Figure 4) was  
239 even in different sizes of aggregates. From 8 to 43 years of tea plantations, the OC and TN  
240 contents in soil aggregates were significantly elevated by 22%-35% and 14%-24%, 11%-22%  
241 and 9%-17%, and 8%-18% and 9%-13% at the 0-10, 10-20, and 20-40 cm soil depths,  
242 respectively. Nevertheless, there existed no significant variation in the aggregate-related TP  
243 content. Furthermore, at the 40-60 cm soil depth, the aggregate-related OC, TN, and TP contents  
244 did not exhibit significant variations over time. Regardless of the tea plantation age, significant  
245 decreases in the aggregate-related OC, TN, and TP contents were observed when the soil depth  
246 increased.

### 247 3.4. Stoichiometric ratios of soil C, N, and P

248 A three-way ANOVA analysis revealed that the lone and interactive effects of soil depth,  
249 tea plantation age, and aggregate size on the C/P and N/P ratios were significant, and the effects  
250 of soil depth, aggregate size, and their interactions on the C/N ratio were significant (Table 1). In  
251 this study, significant increases in aggregate-related C/N (Table S1), C/P (Table S2), and N/P  
252 (Table S3) ratios were accompanied by the increasing aggregate size. At the 0-10, 10-20, and  
253 20-40 cm soil depths, aggregate-related C/N ratio did not exhibit significant variation while  
254 aggregate-related C/P and N/P ratios significantly increased with the increase in tea plantation  
255 age. Moreover, there was little role of tea plantation age in the aggregate-related C/N, C/P, and  
256 N/P ratios at the 40-60 cm soil depth. Among different ages of tea plantations, aggregate-related  
257 C/N, C/P, and N/P ratios significantly dropped when the soil depth increased. For example, at the  
258 0-10 cm soil depth, aggregate-related C/N, C/P, and N/P ratios across the 4 tea plantation ages  
259 fluctuated in 20.81-23.04, 28.81-37.07, and 1.31-1.67, respectively. In the meanwhile, at the

260 40-60 cm soil depth, aggregate-related C/N, C/P, and N/P ratios fluctuated in 16.41-20.74,  
261 13.44-22.88, and 0.84-1.08, respectively.

### 262 3.5. Contents of soil alkali cations and micronutrients

263 Within aggregate size classes, soil exchangeable alkali cations (i.e.,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) were  
264 more concentrated in the microaggregates (Figures 5 and 6). However, soil available  
265 micronutrients (i.e.,  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ ) were mainly found in the coarse macroaggregates (Figures 7  
266 and 8). From 8 to 43 years of tea plantations, the  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents in soil aggregates were  
267 significantly reduced by 31%-38% and 10%-24%, 23%-27% and 9%-18%, and 10%-16% and  
268 5%-8% at the 0-10, 10-20, and 20-40 cm soil depths, respectively. From 8 to 43 years of tea  
269 plantations, however, the  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  contents in soil aggregates were significantly elevated  
270 by 16%-27% and 6%-9%, 11%-15% and 4%-7%, and 7%-12% and 3%-5%, respectively. In  
271 addition, at the 40-60 cm soil depth, the contents of aggregate-related exchangeable alkali  
272 cations and available micronutrients did not present significant variations over time. Irrespective  
273 of the tea plantation age, significant increases in the aggregate-related  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents  
274 were observed with the increasing soil depth, whereas the aggregate-related  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$   
275 contents showed an opposite trend.

### 276 3.6. Stoichiometric ratios of soil alkali cations and micronutrients

277 A three-way ANOVA analysis demonstrated that the effect of tea plantation age on the  
278 Ca/Mg and Fe/Mn ratios in soil aggregates was significant (Table 1). In this study, soil Ca/Mg  
279 (Table S4) and Fe/Mn (Table S5) ratios did not vary among different sizes of aggregates. At the  
280 0-10, 10-20, and 20-40 cm soil depths, aggregate-related Ca/Mg ratio significantly decreased  
281 while aggregate-related Fe/Mn ratio significantly increased in the tea-planting course. Moreover,  
282 there was little role of tea plantation age in the aggregate-related Ca/Mg and Fe/Mn ratios at the  
283 40-60 cm soil depth. In tea plantations, no significant variations were observed amongst different  
284 soil depths in aggregate-related Ca/Mg and Fe/Mn ratios. For example, at the 0-10 cm soil depth,  
285 aggregate-related Ca/Mg and Fe/Mn ratios across the 4 tea plantation ages ranged from 1.81 to

286 1.96 and 0.76 to 0.85, respectively. Meanwhile, at the 40-60 cm soil depth, aggregate-related  
287 Ca/Mg and Fe/Mn ratios ranged from 1.88 to 1.92 and 0.78 to 0.82, respectively.

## 288 **4. Discussion**

### 289 4.1. Composition and stability of soil aggregates

290 Tea plantation age significantly influenced the aggregate composition and stability at the  
291 0-10 and 10-20 cm soil depths, whereas the effect at the 20-40 and 40-60 cm soil depths was  
292 extremely limited. In the early (8-17 years) period, tea planting was conducive to the transition  
293 from microaggregates to coarse macroaggregates at the 0-10 and 10-20 cm soil depths (Table 3).  
294 Comparatively, in the middle (17-25 years) and late (25-43 years) periods, tea planting induced  
295 coarse macroaggregate destruction and microaggregate release (Table 3). According to the  
296 hierarchical concept of soil aggregates (Six et al., 2004), the quality of plant litter returning to the  
297 soil determines the distribution of decomposition products of litter in different sizes of  
298 aggregates, ultimately impacting the aggregate composition. In the early period of tea planting,  
299 tea litter displayed greater availability (as indicated by the lower litter C/N ratio), revealing that  
300 the decomposition products of litter were easily combined into the coarse macroaggregates,  
301 thereby fostering the formation of coarse macroaggregates (Tisdall and Oades, 1982). Reversely,  
302 in the middle and late periods of tea planting, tea plants naturally encountered aging processes  
303 and litter was progressively subjected to humification, inducing the decomposition of coarse  
304 macroaggregates into microaggregates (Six and Paustian, 2014). Moreover, the reduced litter  
305 amount and covering area after 17 years of tea planting enhanced the rainfall eluviation and  
306 artificial interferences (i.e., pruning of tea plants and application of fertilizers), which could also  
307 cause the destruction of coarse macroaggregates. In the tea-planting course, variation in  
308 aggregate stability was indicated via the change of MWD value (Table 3). At the 0-10 and 10-20  
309 cm soil depths, the MWD value was the greatest in the 17 years of tea planting, which was  
310 associated with the highest proportions of coarse macroaggregates in the 17-year tea plantations.

311 The above findings indicated that the 17-year tea plantations exhibited stronger aggregate  
312 stability in contrast to other plantations at the 0-10 and 10-20 cm soil depths.

313       Regardless of the tea plantation age, coarse macroaggregates were dominant in the topsoil  
314 (0-10 cm) while microaggregates were dominant in the subsoil (10-60 cm), suggesting  
315 transformation of aggregate composition from coarse macroaggregate-prevailing to  
316 microaggregate-prevailing with the increase in soil depth (Table 3). In addition, similar  
317 outcomes were corroborated by Li et al. (2015) and Zhu et al. (2017) from studies on tea  
318 plantations in the southwest Sichuan of China. In this study, coarse macroaggregates were the  
319 prevailing fractions in the topsoil, not the subsoil, which was caused by the surface cumulation  
320 of soil OC (Figure 2). As an essential cementing agent, soil OC could foster the formation of  
321 coarse macroaggregates (Al-Kaisi et al., 2014). Moreover, the reduced proportions of coarse  
322 macroaggregates as the soil depth increased were also resulted from the elevated soil  
323 compactness (as indicated by the bulk density) (Table 2). Soil densification could prevent the  
324 growth of plant roots, hence causing the activities of soil microorganisms decreased, especially  
325 soil fungi (Kurmi et al., 2020). Reduced activities of soil fungi could diminish the production of  
326 polysaccharose and glomalin-related soil protein (GRSP) from the fungal hyphae, thereby  
327 inducing the proportions of soil macroaggregates decreased (Ji et al., 2019). Similarly, as per our  
328 past studies (Wang et al., 2017b; Zhu et al., 2019), soil microbial activities and GRSP content  
329 served as the vital effects in the formation and stabilisation of soil macroaggregates, and also  
330 presented the higher levels in the topsoil compared with the subsoil in the tea plantation  
331 ecosystems. With the increasing soil depth, the decrease in MWD value was mainly associated  
332 with the change of soil aggregate composition (Table 3), especially for the decomposition of  
333 coarse macroaggregates into microaggregates, implying that the topsoil exhibited stronger  
334 aggregate stability in contrast to the subsoil.

## 335 4.2. Contents of soil C, N, and P

336 In this study, more contents of soil OC and TN could be detected in coarse macroaggregates  
337 (Figures 2 and 3), which conformed to the findings of Six et al. (2004) that macroaggregates  
338 were comprised of microaggregates via temporary binding agents. Meanwhile, macroaggregates  
339 could provide the protection for the OM, causing the cumulation of OC and TN in  
340 macroaggregates. Different from soil OC and TN, soil TP was evenly distributed in different  
341 sizes of aggregates (Figure 4). Moreover, Bhatnagar and Miller (1985) also detected similar  
342 outcomes from soil specimens subjected to fresh poultry manure treatments, and promoted the  
343 mechanisms influencing the distribution of TP in soil aggregates. Specifically, (i) introduced P  
344 was firstly adsorbed by clay particulates in soil and clay particulates were discrepant in different  
345 sizes of aggregates, and (ii) introduced P had selective absorptive properties for the different  
346 sizes of aggregates. According to our findings, stochasticity seems to be one probable  
347 mechanism that sheds light on the TP distribution in soil aggregates.

348 Tea plantation age could positively affect the cumulation of soil OC and TN, while such  
349 positive effects were more obvious at the 0-40 cm soil depth in contrast to the 40-60 cm soil  
350 depth. In this study, soil OC and TN contents exhibited a significant growing trend over time  
351 (Figures 2 and 3), which was possibly associated with the following mechanisms. At first,  
352 numerous long-period tests had demonstrated the proactive roles of manure and chemical  
353 fertilizer applications in soil OM cumulation (Tong et al., 2009; Zhou et al., 2013). Similarly, in  
354 the tea-planting course, growing soil OC and TN contents were probably caused by the  
355 application of substantial swine manure every year ( $12 \text{ Mg ha}^{-1} \text{ year}^{-1}$ ) in this tea-planting region  
356 (Wang and Ye, 2020). Second, plants serve as the prime OM sources in soil via root exudates  
357 and litter remains (Franklin et al., 2020). In the tea-planting course, soil OC and TN cumulation  
358 probably occurred as a result of the growing root systems and the increasing amounts of  
359 aboveground litter attained from trimmed branches and leaves. Third, no tillage could provide  
360 physical protection for the OM combined with soil aggregates, then further improving soil OC

361 and TN sequestration (Wulanningtyas et al., 2021). Notably, although the positive correlations of  
362 OC and TN contents with clay content in soil have been reported, this study revealed that  
363 significant increases in the OC and TN contents were accompanied by no significant variation in  
364 the clay content during the process of tea growth (Table 2). Similarly, Li et al. (2015) and Wang  
365 et al. (2018) also discovered as well that the changes of soil OC and TN contents were not  
366 influenced by the clay content over time in the tea plantation ecosystems, mainly because soil  
367 OC and TN contents primarily depend on fertilization, tillage, root exudates, and litter remains,  
368 whereas soil clay content is mainly controlled by its parent material (Rakhsh et al., 2020).  
369 Different from soil OC and TN, regardless of the soil depth, there existed no significant  
370 difference in soil TP content amongst different aged tea plantations (Figure 4), implying the  
371 resistance of soil TP content to the change of tea plantation age. Moreover, previous studies  
372 verified that soil TP content was not associated with the tea plantation age (Wu et al., 2018; Yan  
373 et al., 2018), as soil P primarily derives from the weathering release of soil minerals, instead of  
374 the short-period biology cycle (Cui et al., 2019). In the tea plantation ecosystems, the decreasing  
375 OC, TN, and TP contents with the increasing soil depth (Figures 2, 3, and 4) coincided with  
376 some previous findings in other ecosystems, including tropic forests, bushlands, and grasslands  
377 (Stone and Plante, 2014; Yu et al., 2018; Qiao et al., 2020). In this study, the higher contents of  
378 OC, TN, and TP in the topsoil were associated with the higher OM input, where the soil OM  
379 content in the topsoil was enriched by the input of surface tea litter, root debris and exudates, and  
380 swine manure.

#### 381 4.3. Stoichiometric ratios of soil C, N, and P

382 Soil C/N, C/P, and N/P ratios serve as vital indicators of soil health (Liu et al., 2018), which  
383 can be employed to explore C circulation and guiding the equilibrium between N and P in soil  
384 ecosystems (Sardans et al., 2012). In this study, soil C/N ratio grew with increasing aggregate  
385 size (Table S1), indicating that the OM in macroaggregates was younger and more unstable in  
386 contrast to microaggregates (Six et al., 2004). Meanwhile, the OM associated with

387 microaggregates experienced more degradation, resulting in the lower C/N ratio in the  
388 microaggregates (Xu et al., 2019). Among different ages of tea plantations, soil OC and TN were  
389 predominantly distributed in the coarse macroaggregates, whereas the TP was evenly distributed  
390 in different sizes of aggregates. As a result, the associations of C/P and N/P ratios to aggregate  
391 size primarily depended on the relationships of OC and TN contents with aggregate size (Tables  
392 S2 and S3). As far as we know, the changes of soil C/P and N/P ratios within aggregates are  
393 rarely examined, even though these kinds of knowledge are imperative due to the  
394 biogeochemical cycles of N and P being influenced by the dynamics of soil aggregates (Cui et al.,  
395 2021). Consequently, the impact generated by the aggregate size on the C/P and N/P ratios is  
396 required to be studied more for the accurate forecast of soil N and P cycling under natural or  
397 man-intervened ecosystems.

398 Irrespective of the soil depth, soil C/N ratio showed little significant variation in the  
399 tea-planting course (Table S1). Meanwhile, tea plantation age significantly affected soil C/P and  
400 N/P ratios at the 0-40 cm soil depth, rather than the 40-60 cm soil depth (Tables S2 and S3). Soil  
401 C/N ratio is generally treated as the critical indicator which can affect the formation and  
402 degradation of soil OM (Khan et al., 2016). Since response of soil TN content to soil  
403 environment change is almost the same as soil OC content (Wang et al., 2018), soil C/N ratio did  
404 not present significant difference amongst different aged tea plantations (Table S1). Similarly,  
405 Zhou et al. (2018) proved that no close correlation existed between soil C/N ratio and vegetation  
406 coverage, because C and N are structure elements and their cumulation and consumption in soil  
407 remain relatively consistent. Soil C/P ratio is the indicator suggesting P effectiveness, and higher  
408 C/P ratio often denotes lower P effectiveness (Khan et al., 2016). In acidic soil (Table 2),  
409 available P was adsorbed on the surfaces of Fe/Al oxides and clay minerals in a preferential way,  
410 because Fe/Al oxides and clay minerals with greater surface areas could afford enough sites to  
411 available P adsorption (Wu et al., 2018). Therefore, when the tea plantation age increased, soil  
412 acidification generated the decrease in P effectiveness (evidenced by the significant increase in



413 soil C/P ratio) (Table S2). Soil N and P are the prohibiting factors mostly observed during the  
414 process of plant growth, and thus, N/P ratio can be utilized as an efficient indicator that shows  
415 nutrient restriction (Khan et al., 2016). In this study, soil N/P ratio significantly increased in the  
416 tea-planting course (Table S3), mainly because soil TN content experienced significant increase  
417 while no such significant change was observed in TP content over time.

418       Regardless of the tea plantation age, soil C/N ratio decreased with the increasing soil depth  
419 (Table S1), which coincided with the results from Cao et al. (2015), Feng and Bao (2017), and  
420 Yu et al. (2019). They suggested that the decrease in soil C/N ratio as the soil depth increased  
421 was triggered by the older and more processed OM in the deeper soil layer. Moreover, in this  
422 study, the lower soil C/P and N/P ratios in the subsoil (Tables S2 and S3) backed the outcomes  
423 of past studies in terrestrial ecosystems of China, which were on the foundation of the data  
424 obtained from both the 2<sup>nd</sup> soil investigation in China (Tian et al., 2010) and the Chinese  
425 Ecosystem Research Network (CERN) (Chai et al., 2015).

426       Across the 4 tea plantation ages, the mean contents of OC and TN in bulk soil (0-20 cm)  
427 were 16.70 and 0.77 g kg<sup>-1</sup>, respectively, which were below the mean contents of OC (21.30 g  
428 kg<sup>-1</sup>) and TN (2.17 g kg<sup>-1</sup>) in Chinese tea plantations (Sun et al., 2020; Xie et al., 2020).  
429 Moreover, in this tea-planting region, the mean content of TP in bulk soil (0-20 cm) was 0.57 g  
430 kg<sup>-1</sup>, corresponding to the moderate level in Chinese tea plantations, where TP content varied in  
431 the range of 0.35-1.20 g kg<sup>-1</sup> (Wu et al., 2018; Sun et al., 2020). Herein, soil C/N ratio is higher  
432 compared with other tea-planting regions in China, whereas soil C/P and N/P ratios are much  
433 lower (Sun et al., 2020). The above findings are primarily associated with the lower contents of  
434 soil OC and TN, especially the TN. In general, N is the most limiting element in the net primary  
435 production of the tea plantation ecosystems (Miner et al., 2018). Besides, this phenomenon also  
436 appeared in the southern Guangxi of China.

#### 437 4.4. Contents of soil alkali cations and micronutrients

438       According to the findings from Adesodun et al. (2007) and Emadi et al. (2009), the higher

439 contents of exchangeable alkali cations (including  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) were detected in both 2-4.76  
440 and  $< 0.25$  mm aggregates in the non-tillage soil. However, in the tillage course, the contents of  
441 these two cations decreased in the 2-4.76 mm aggregates and increased in the  $< 0.25$  mm  
442 aggregates, revealing that the tillage practice could lead soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  to redistribute in  
443 different sizes of aggregates. Comparatively, this study exhibited that the distribution of soil  $\text{Ca}^{2+}$   
444 and  $\text{Mg}^{2+}$  in aggregates was similar among different ages of tea plantations (Figures 5 and 6),  
445 suggesting that the distribution of these two cations in aggregates was seldom influenced by the  
446 tea plantation age. Specifically, coarse macroaggregates had the lowest contents of  $\text{Ca}^{2+}$  and  
447  $\text{Mg}^{2+}$ , whereas microaggregates exhibited the highest contents. These findings could be ascribed  
448 to the larger specific surface areas of microaggregates (Adesodun et al., 2007), which increased  
449 microaggregates' adsorption to  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  derived from root exudates, litter remains, and  
450 manure (Emadi et al., 2009). Different from exchangeable alkali cations, the contents of soil  
451 available micronutrients (including  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ ) usually correspond to the content of soil OM  
452 (Wang et al., 2017a), which are more abundant in macroaggregates (Six et al., 2004). Moreover,  
453 this study also found that the  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  had a similar distribution pattern with OC within  
454 aggregates (Figures 7 and 8). Since the decomposition products of litter can be easily integrated  
455 to the coarse macroaggregates (Six et al., 2004), the nutrient cycling of plant-soil systems might  
456 contribute to the higher contents of soil  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  in the coarse macroaggregates (Wang et  
457 al., 2017a).

458 At the 0-40 cm soil depth, the contents of soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  significantly decreased over  
459 time (Figures 5 and 6), which might be caused by the applications of urea and  $\text{NH}_4^+$ -N fertilizer  
460 in the tea-planting course for increasing tea leaf outputs. Urea hydrolysis can promote the  
461 production of ammonium ions which are readily nitrified into nitrate, and the excessive proton  
462 produced by the nitrification can compete for the adsorption sites with  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  (Wang et  
463 al., 2017a). As a result, these cations were easy to lose from soil in the manner of leaching.  
464 Except at the 40-60 cm soil depth, continuous planting of tea generated the significant increases

465 in soil  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  contents (Figures 7 and 8), which were elevated by 7%-27% and 3%-9%  
466 from 8 to 43 years of tea planting, respectively. This phenomenon was possibly caused by the  
467 soil acidification (Table 2), stimulating the release of soil  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  by mineralization and  
468 desorption from soil OM and minerals (Wang et al., 2017a). Tea, as an aluminium (Al)  
469 cumulating crop, is capable of cumulating Al in leaves (Li et al., 2016). Soil acidification in the  
470 tea-planting course was due to the substantial tea litter into the soil annually through trimmed  
471 branches and leaves (Li et al., 2016). At the same time, the rhizosphere deposition of massive  
472 organic acids (i.e., malate, lemon acid, and oxalate acid) around the tea roots could provoke  
473 localized acidification (Xue et al., 2006). Apart from that, to increase the output of tea, tea  
474 plantations needed to apply N fertilizers (i.e., urea and  $\text{NH}_4^+\text{-N}$ ), thus leading to soil acidification  
475 by the  $\text{NH}_4^+$  nitrification (Yang et al., 2018). Across the 4 tea plantation ages, the contents of soil  
476  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  were higher in the topsoil than the subsoil (Figures 7 and 8), primarily due to the  
477 usage of swine manure and the inputs of tea litter and roots in the topsoil (Miner et al., 2018).  
478 Nevertheless, the contents of soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  showed an opposite trend when the soil depth  
479 increased (Figures 5 and 6), because soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  were easy to move from topsoil to  
480 subsoil in the manner of leaching (Hansen et al., 2017).

#### 481 4.5. Stoichiometric ratios of soil alkali cations and micronutrients

482 Tea plantation age exerted a significant influence on the Ca/Mg and Fe/Mn ratios at the  
483 0-40 cm soil depth, rather than the 40-60 cm soil depth (Tables S4 and S5). To be specific, a  
484 significant decline in the Ca/Mg ratio was found at the 0-40 cm soil depth over time. From 8 to  
485 43 years of tea planting, the contents of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  at the 0-40 cm soil depth decreased by  
486 10%-38% and 5%-24%, respectively, revealing that the role of tea plantation age in the content  
487 of soil  $\text{Ca}^{2+}$  was greater than that of soil  $\text{Mg}^{2+}$  (Figures 5 and 6). Lu et al. (2014) suggested that  
488 the selective losses of soil exchangeable alkali cations ( $\text{Ca}^{2+} > \text{Mg}^{2+}$ ) could lead to the  
489 disequilibrium of soil metal ions in forest ecosystems. Similarly, in this study, the preferential  
490 loss of soil  $\text{Ca}^{2+}$  relative to  $\text{Mg}^{2+}$  was the prime reason for the significant decline in the soil

491 Ca/Mg ratio in the tea-planting course. The depletion of soil exchangeable alkali cations  
492 (especially the  $\text{Ca}^{2+}$ ) could generate the decrease in soil buffering capacity and soil acidification  
493 (Hansen et al., 2017). Thus, the Ca/Mg ratio at the 0-40 cm soil depth was positively related ( $p \leq$   
494 0.05) to soil pH across the 4 tea plantation ages (Figure S1). Soil acidification accelerated the  
495 mineralization and desorption of soil available micronutrients from soil OM and minerals (Wang  
496 et al., 2017a), which was conducive to the significant increases in  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  contents at the  
497 0-40 cm soil depth, especially the  $\text{Fe}^{2+}$  (Figures 7 and 8). In a chronological sequence of tea  
498 plantations, the negative relationship ( $p \leq 0.05$ ) of soil Fe/Mn ratio with soil pH in different soil  
499 depths indicated more cumulation of soil  $\text{Fe}^{2+}$  relative to  $\text{Mn}^{2+}$  over time (Figure S1).  
500 Furthermore, during the process of tea plant uptake, the change of soil Fe/Mn ratio was also  
501 triggered by the antagonistic relationship between soil  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  (Wang et al., 2017a). Tian  
502 et al. (2016) discovered that soil acidification could reduce  $\text{Fe}^{2+}$  absorption and enhance  $\text{Mn}^{2+}$   
503 uptake by various plant species, causing the increase in soil Fe/Mn ratio and threatening plant  
504 productivity.

## 505 **5. Conclusions**

506 To conclude, soil OC, TN, and TP contents as well as C/N, C/P, and N/P ratios decreased  
507 when the soil depth increased. Moreover, soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents were lower in the topsoil  
508 than the subsoil, whereas soil  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  contents showed an opposite trend, and no  
509 differences were detected amongst different soil depths in soil Ca/Mg and Fe/Mn ratios. At the  
510 0-40 cm soil depth, continuous planting of tea was favorable to the increases in soil OC, TN,  
511  $\text{Fe}^{2+}$ , and  $\text{Mn}^{2+}$  contents, whereas soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents decreased over time, thus  
512 supporting our hypotheses. Compared with other tea-planting regions in China, soil C/N ratio is  
513 higher in this tea-planting region, whereas soil C/P and N/P ratios are much lower, suggesting  
514 that soil OC and TN contents in this study were lower, especially the TN. In the tea-planting  
515 course, the losses of soil  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , especially the  $\text{Ca}^{2+}$  (as indicated by the decrease in soil  
516 Ca/Mg ratio), could lead to the soil acidification. Meanwhile, soil acidification could reduce  $\text{Fe}^{2+}$

517 absorption and enhance  $Mn^{2+}$  uptake by tea plants (as indicated by the increase in soil Fe/Mn  
518 ratio). In general, tea plantation age could influence the variations in soil nutrient contents and  
519 stoichiometry, whereas such effects were more obvious at the 0-40 cm soil depth in contrast to  
520 the 40-60 cm soil depth.

#### 521 **Data availability**

522 The data supporting the discovered information here can be presented by the relevant author  
523 based on reasonable requests.

#### 524 **Author contribution**

525 S.W. and S.Y. designed the experiments; L.M. carried out the experiments; S.W. and L.M.  
526 analyzed the experimental results; L.M., S.W. and S.Y. wrote and edited the manuscript.

#### 527 **Competing interests**

528 The authors declare no conflict of interest.

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**Table 1** Three-way ANOVA regarding the effects of soil depth, tea plantation age, aggregate size, and their interactions on the physico-chemical properties of soil aggregates, and Two-way ANOVA regarding the effects of soil depth, tea plantation age, and their interactions on the physico-chemical properties of bulk soil.

Soil properties	Three-way ANOVA							Two-way ANOVA		
	S	T	A	S × T	S × A	T × A	S × T × A	S	T	S × T
Bulk density								*	*	*
Clay								NS	NS	NS
pH								NS	*	NS
MWD								*	*	*
Aggregate proportion	*	*	**	*	*	*	*			
Organic C	**	**	**	**	**	**	**	**	**	**
Total N	**	**	**	**	**	**	**	**	**	**
Total P	*	NS	NS	NS	NS	NS	NS	*	NS	NS
Exchangeable Ca <sup>2+</sup>	*	**	*	*	*	*	*	*	**	*
Exchangeable Mg <sup>2+</sup>	*	*	*	*	*	*	*	*	*	*
Available Fe <sup>2+</sup>	*	**	*	*	*	*	*	*	**	*
Available Mn <sup>2+</sup>	*	*	*	*	*	*	*	*	*	*
C/N ratio	*	NS	*	NS	*	NS	NS	*	NS	NS
C/P ratio	*	*	*	*	*	*	*	*	*	*
N/P ratio	*	*	*	*	*	*	*	*	*	*
Ca/Mg ratio	NS	*	NS	NS	NS	NS	NS	NS	*	NS
Fe/Mn ratio	NS	*	NS	NS	NS	NS	NS	NS	*	NS

S: soil depth; T: tea plantation age; A: aggregate size. \*\*, \*, and NS indicate significant differences at  $p < 0.01$ ,  $p \leq 0.05$ , and  $p > 0.05$ , respectively.



**Table 2** Effects of soil depth and tea plantation age on the bulk density, clay, and pH in bulk soil.

Soil depth	Tea plantation age	Bulk density (g cm <sup>-3</sup> )	Clay (%)	pH
0-10 cm	8 years	1.28 ± 0.02 b	34.69 ± 3.21 a	4.57 ± 0.02 a
	17 years	1.20 ± 0.02 c	35.91 ± 2.77 a	4.49 ± 0.01 ab
	25 years	1.26 ± 0.01 bc	33.12 ± 2.46 a	4.31 ± 0.03 b
	43 years	1.31 ± 0.04 b	35.08 ± 2.41 a	4.15 ± 0.02 c
10-20 cm	8 years	1.30 ± 0.03 b	34.88 ± 2.08 a	4.55 ± 0.03 a
	17 years	1.22 ± 0.03 c	32.59 ± 3.02 a	4.50 ± 0.01 a
	25 years	1.30 ± 0.03 b	34.92 ± 3.67 a	4.33 ± 0.02 b
	43 years	1.29 ± 0.02 b	32.35 ± 2.68 a	4.17 ± 0.02 c
20-40 cm	8 years	1.32 ± 0.04 ab	35.26 ± 1.45 a	4.60 ± 0.04 a
	17 years	1.31 ± 0.01 b	34.57 ± 4.12 a	4.53 ± 0.02 a
	25 years	1.34 ± 0.01 ab	34.51 ± 3.21 a	4.34 ± 0.04 b
	43 years	1.33 ± 0.04 ab	34.29 ± 3.54 a	4.19 ± 0.03 c
40-60 cm	8 years	1.36 ± 0.01 a	34.78 ± 3.66 a	4.58 ± 0.02 a
	17 years	1.37 ± 0.02 a	36.89 ± 2.98 a	4.54 ± 0.03 a
	25 years	1.39 ± 0.02 a	33.68 ± 1.91 a	4.32 ± 0.01 b
	43 years	1.38 ± 0.03 a	35.81 ± 3.69 a	4.21 ± 0.01 bc

Data represent the mean of 5 replicates ± standard deviations. Means in the same column with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages.

**Table 3** Effects of soil depth and tea plantation age on the aggregate stability and composition.

Soil depth	Tea plantation age	MWD (mm)	Aggregate composition (%)			
			> 2 mm	1-2 mm	0.25-1 mm	< 0.25 mm
0-10 cm	8 years	1.88 ± 0.03 b	44.26 ± 3.24 bA	16.23 ± 2.45 abC	8.46 ± 1.37 abD	31.05 ± 5.78 bcB
	17 years	2.20 ± 0.04 a	53.18 ± 2.78 aA	18.02 ± 1.63 aB	6.69 ± 0.98 bC	22.11 ± 4.01 cB
	25 years	1.78 ± 0.01 b	40.29 ± 4.01 bA	17.97 ± 2.03 aC	8.81 ± 0.88 abD	32.93 ± 3.58 bcB
	43 years	1.53 ± 0.03 c	32.60 ± 3.61 cB	19.61 ± 2.04 aC	7.64 ± 1.57 bD	40.15 ± 4.27 abA
10-20 cm	8 years	1.62 ± 0.02 c	37.31 ± 2.47 cA	13.58 ± 1.56 bB	9.24 ± 2.04 abC	39.87 ± 2.69 abA
	17 years	1.82 ± 0.04 b	43.02 ± 2.69 bA	14.31 ± 1.38 abC	8.87 ± 1.14 abD	33.80 ± 4.58 bB
	25 years	1.56 ± 0.03 c	34.87 ± 1.45 cB	15.03 ± 2.47 abC	9.36 ± 1.09 abD	40.74 ± 3.94 abA
	43 years	1.34 ± 0.02 d	29.24 ± 3.28 dB	13.97 ± 1.65 bC	7.28 ± 0.82 bD	49.51 ± 2.56 aA
20-40 cm	8 years	1.43 ± 0.01 cd	31.25 ± 1.68 cdB	15.47 ± 2.49 abC	7.62 ± 0.47 bD	45.66 ± 4.77 aA
	17 years	1.48 ± 0.03 cd	32.08 ± 3.60 cdB	16.89 ± 2.51 abC	8.91 ± 2.14 abD	42.12 ± 2.05 abA
	25 years	1.39 ± 0.02 d	30.72 ± 3.25 dB	14.23 ± 0.58 abC	6.81 ± 1.36 bD	48.24 ± 3.59 aA
	43 years	1.48 ± 0.03 cd	32.49 ± 2.98 cdB	15.40 ± 2.11 abC	9.05 ± 0.91 abD	43.06 ± 4.32 aA
40-60 cm	8 years	1.30 ± 0.01 d	28.48 ± 2.57 dB	12.02 ± 3.08 bC	10.05 ± 0.58 aC	49.45 ± 3.68 aA
	17 years	1.36 ± 0.02 d	29.68 ± 2.61 dB	13.78 ± 1.14 bC	9.47 ± 1.03 abC	47.07 ± 3.47 aA
	25 years	1.36 ± 0.01 d	30.09 ± 1.47 dB	11.98 ± 0.98 bC	10.64 ± 0.45 aC	47.29 ± 4.01 aA
	43 years	1.34 ± 0.03 d	28.42 ± 3.02 dB	14.33 ± 1.57 abC	12.45 ± 2.13 aC	44.80 ± 2.99 aA

Data represent the mean of 5 replicates ± standard deviations. Means in the same column with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means in the same row with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 1** Location of the experiment site. Tea plantations were cultured with the same tea variety with different planting ages (8, 17, 25, and 43 years), and were located at the same unit associated with geomorphological status. Each of the 4 tea plantation age groups was replicated in 5 locations for a total of 20 experimental units.

**Figure 2** Effects of soil depth and tea plantation age on the organic C content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 3** Effects of soil depth and tea plantation age on the total N content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 4** Effects of soil depth and tea plantation age on the total P content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 5** Effects of soil depth and tea plantation age on the exchangeable  $\text{Ca}^{2+}$  content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 6** Effects of soil depth and tea plantation age on the exchangeable  $\text{Mg}^{2+}$  content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 7** Effects of soil depth and tea plantation age on the available  $\text{Fe}^{2+}$  content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

**Figure 8** Effects of soil depth and tea plantation age on the available  $\text{Mn}^{2+}$  content in bulk soil and different sized aggregates. Data represent the mean of 5 replicates and error bars represent the standard deviations. Means with the same lower case letter are not significantly different ( $p > 0.05$ ) among different soil depths and tea plantation ages. Means with the same capital letter are not significantly different ( $p > 0.05$ ) among different sized aggregates.

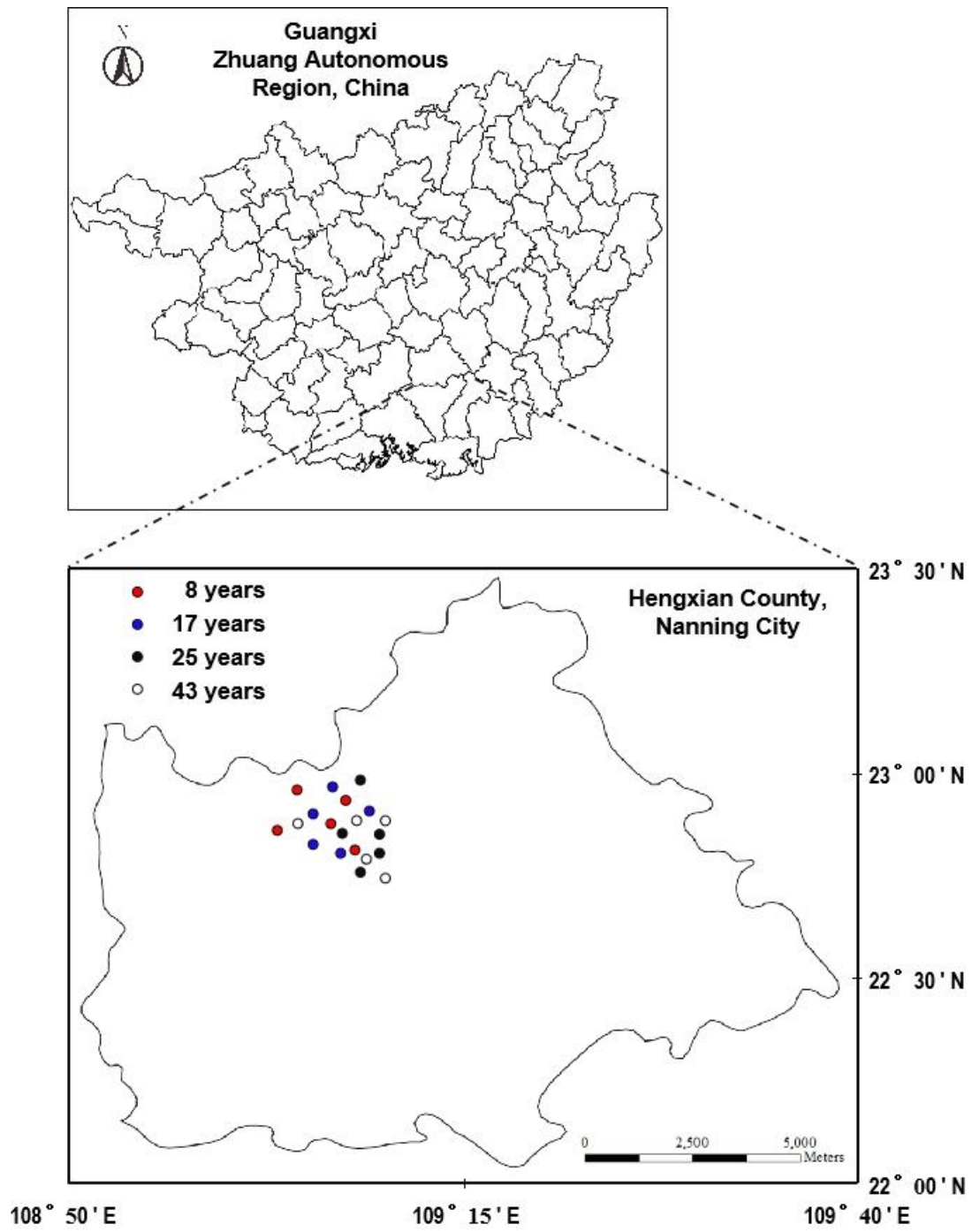


Figure 1

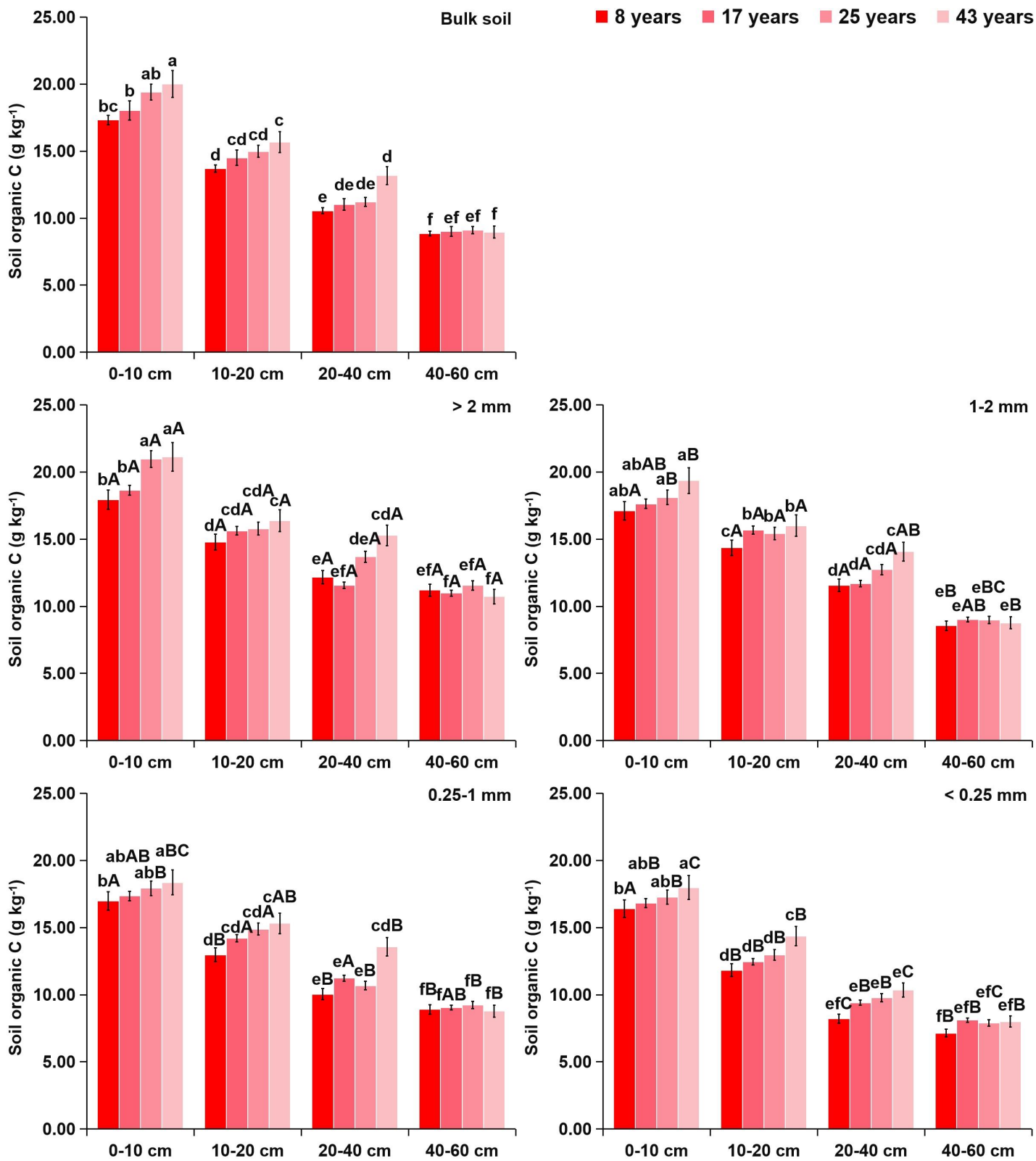


Figure 2

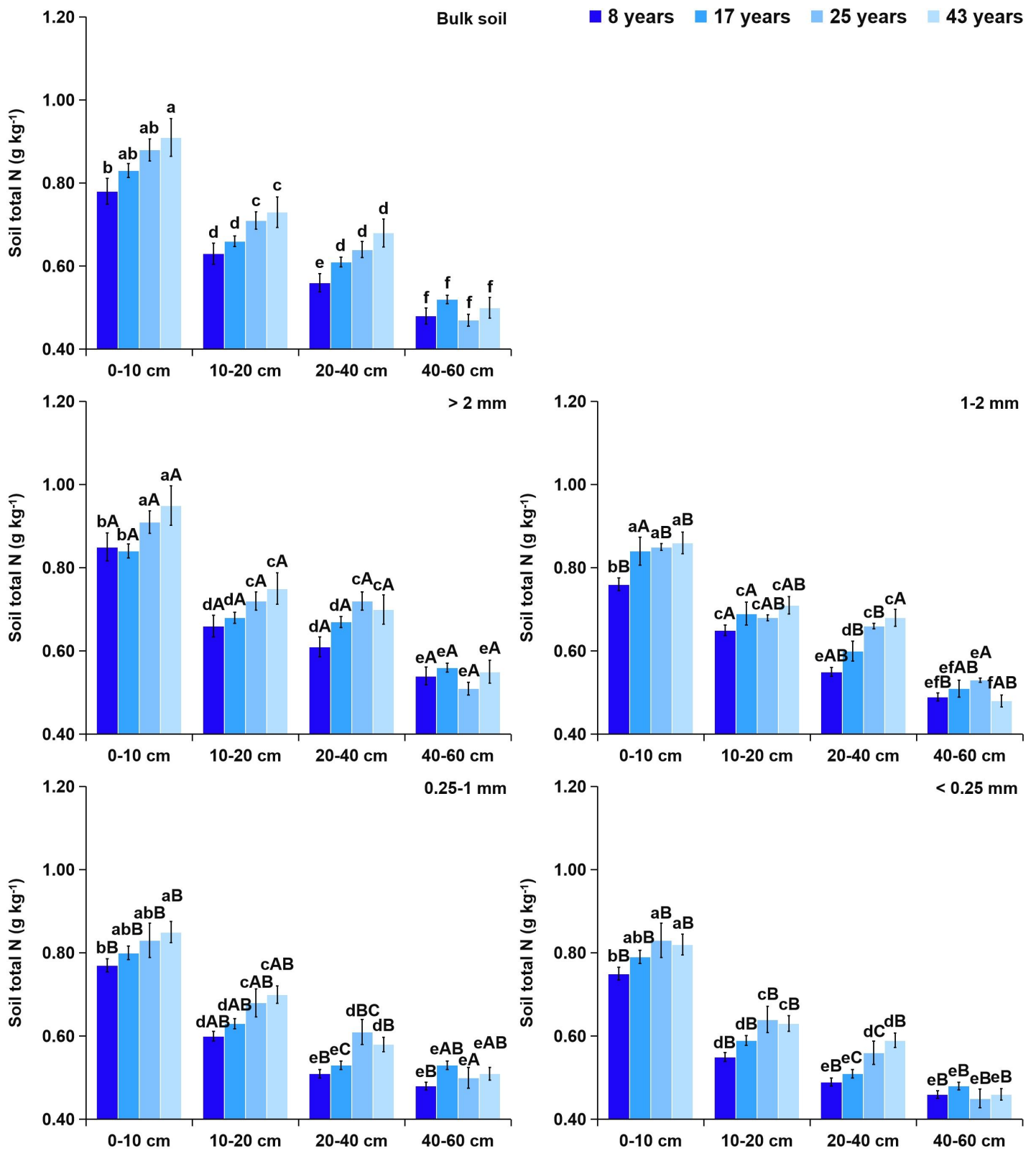


Figure 3

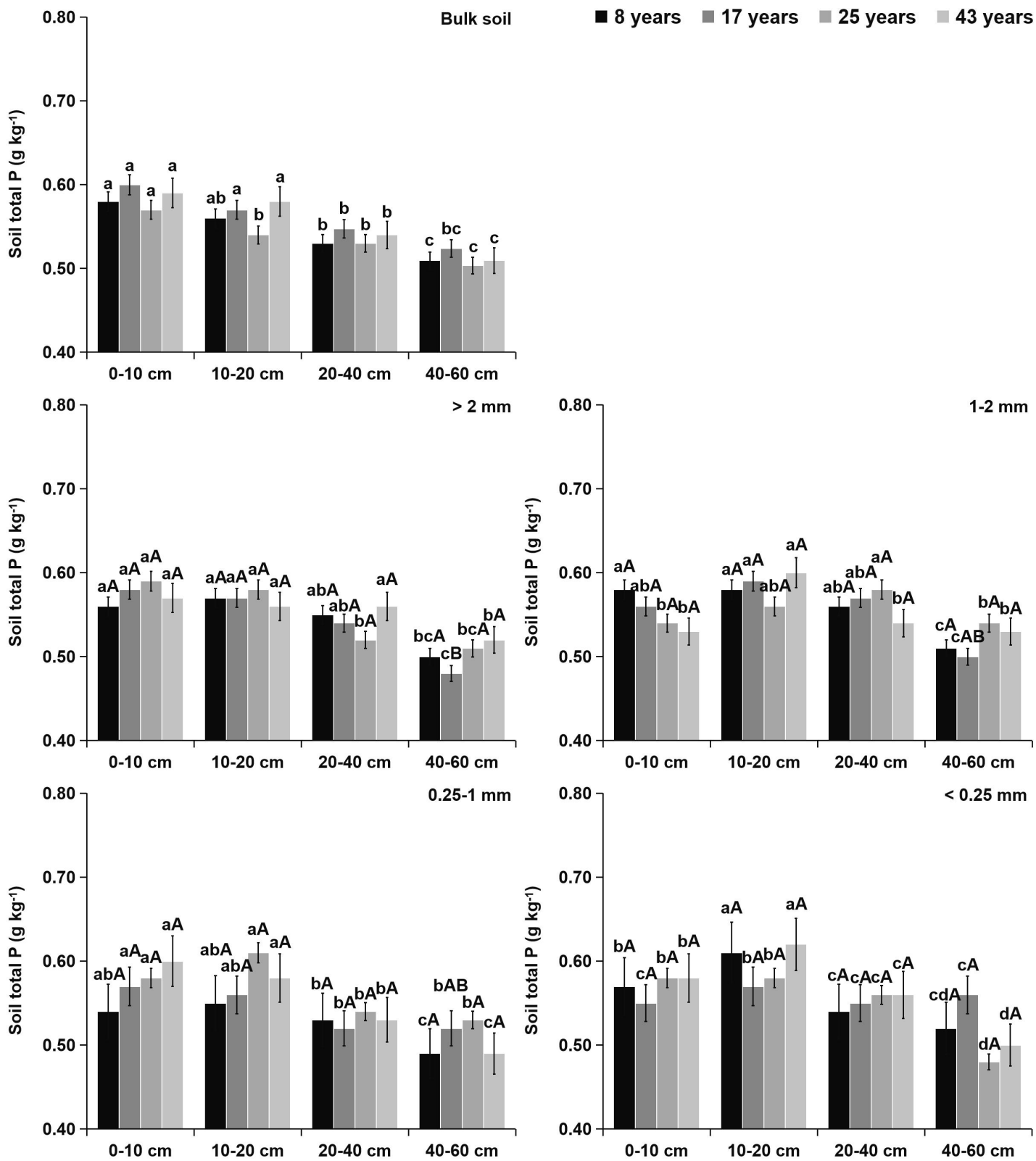


Figure 4

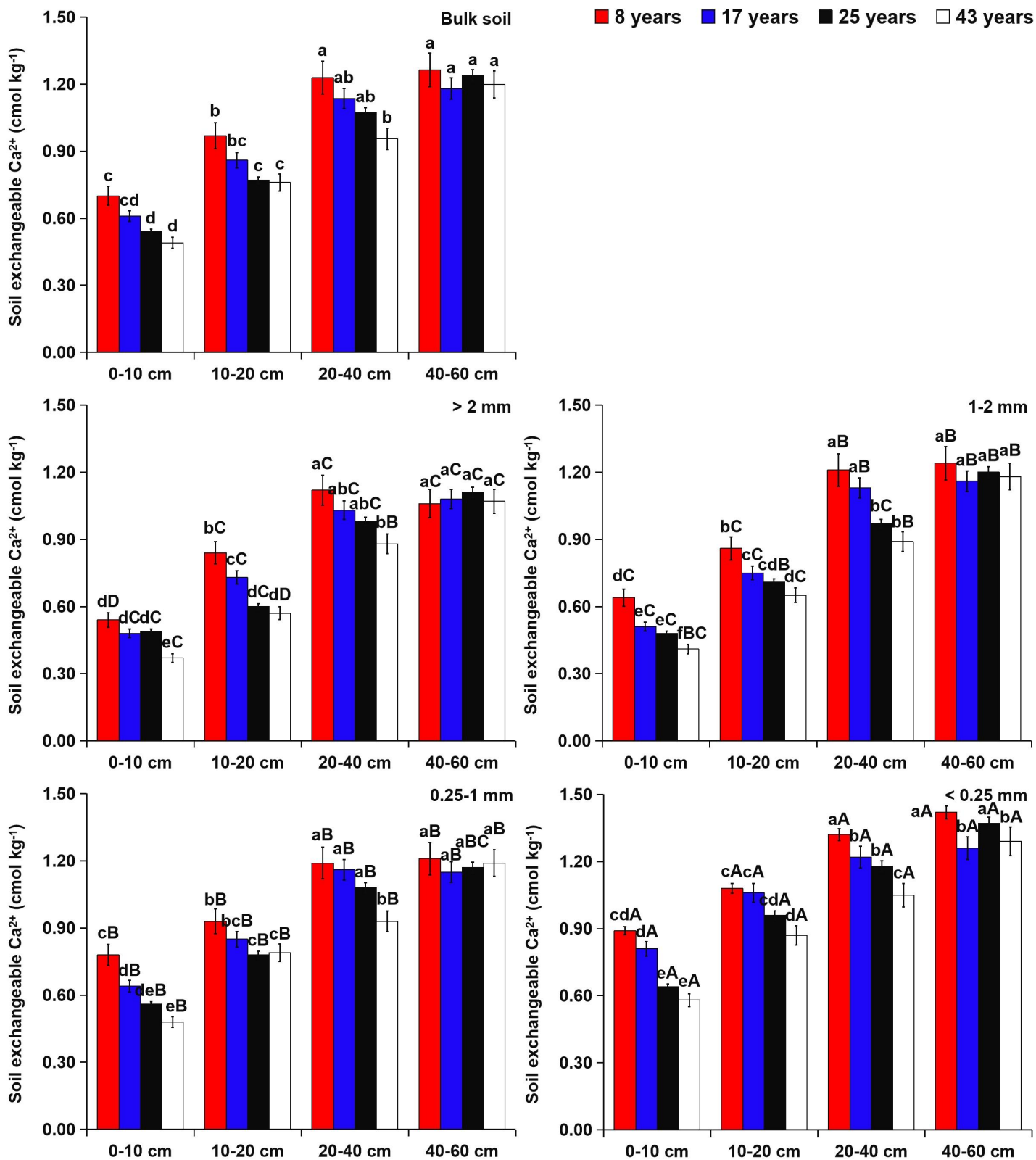


Figure 5



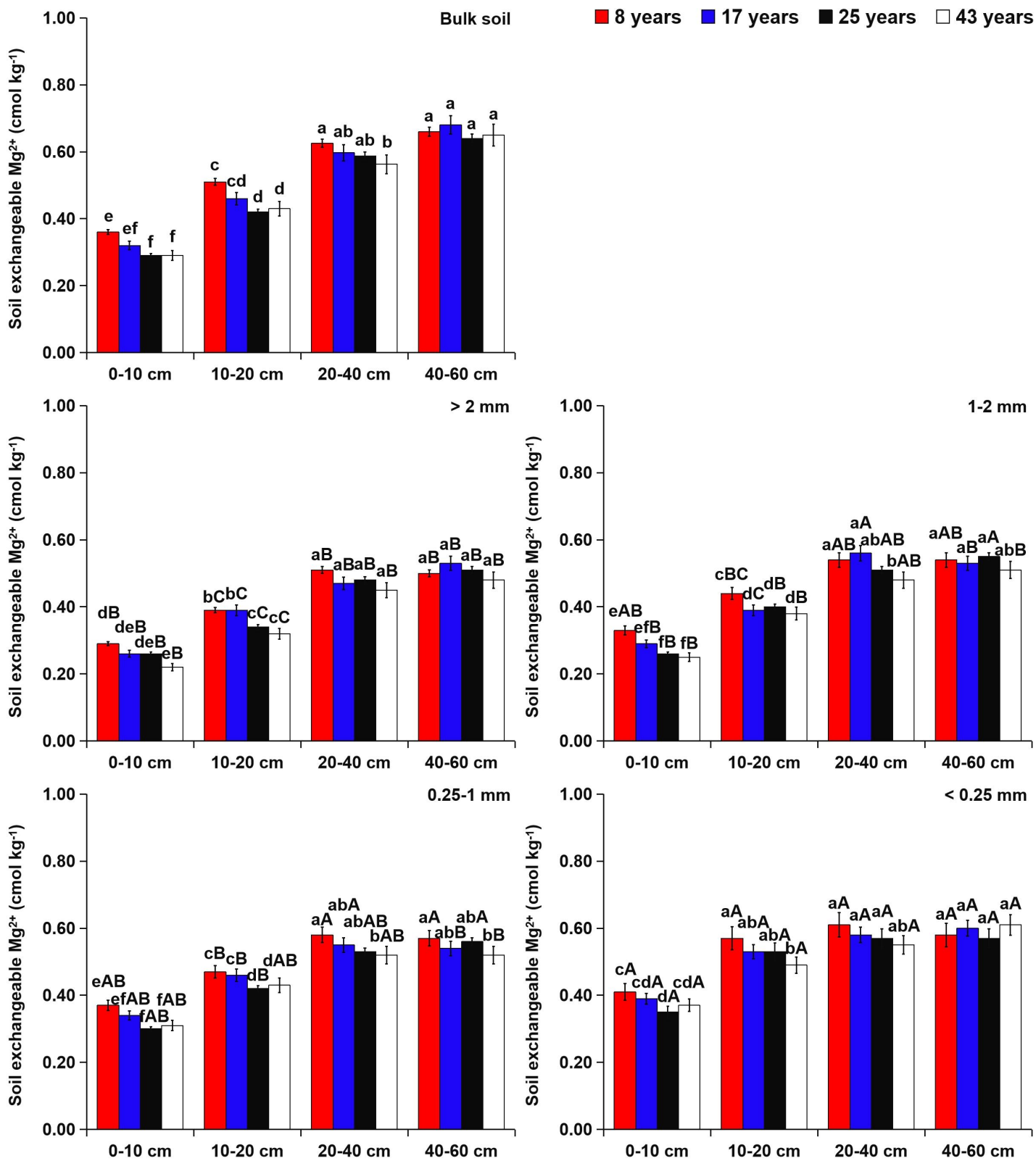


Figure 6

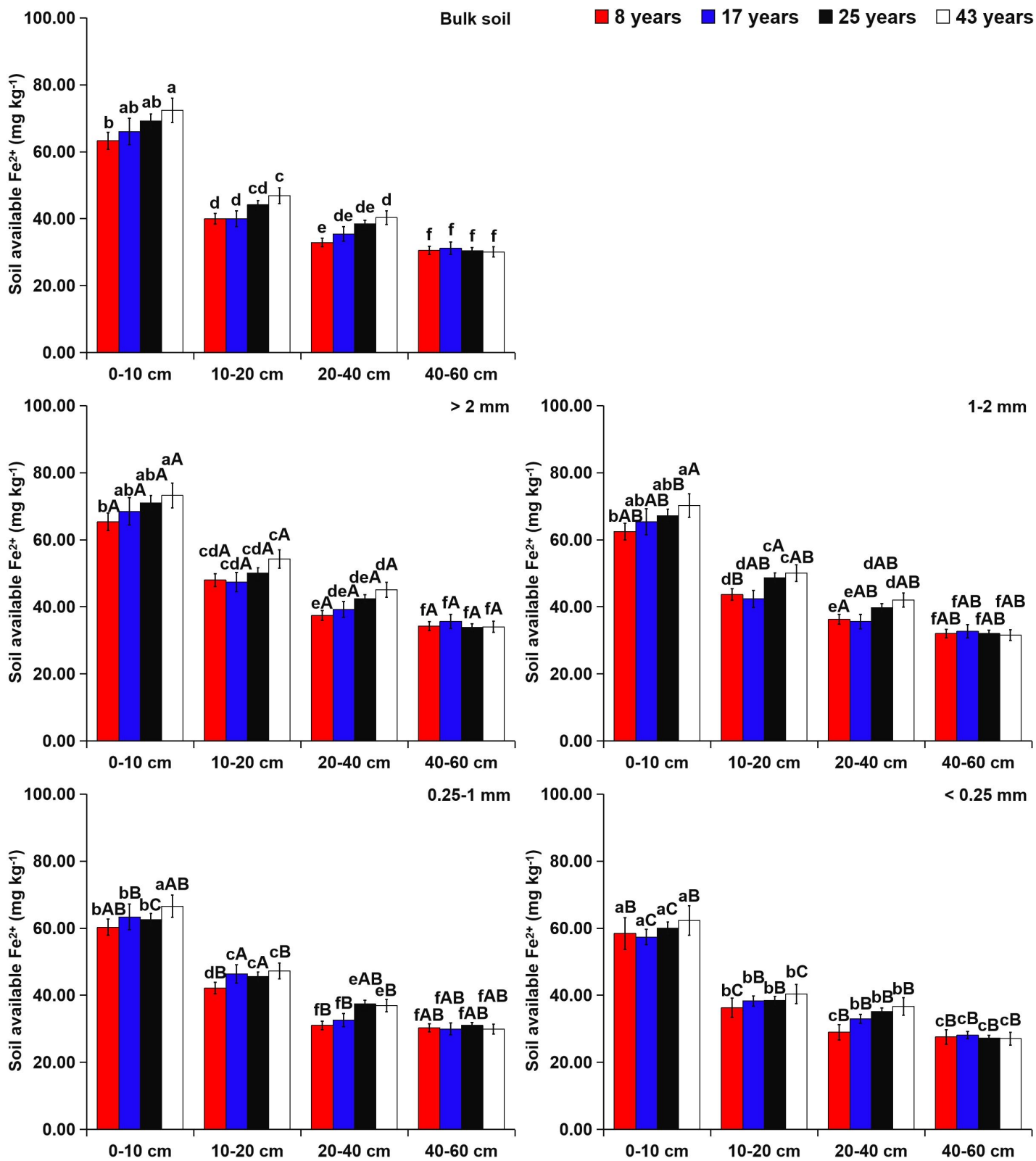


Figure 7

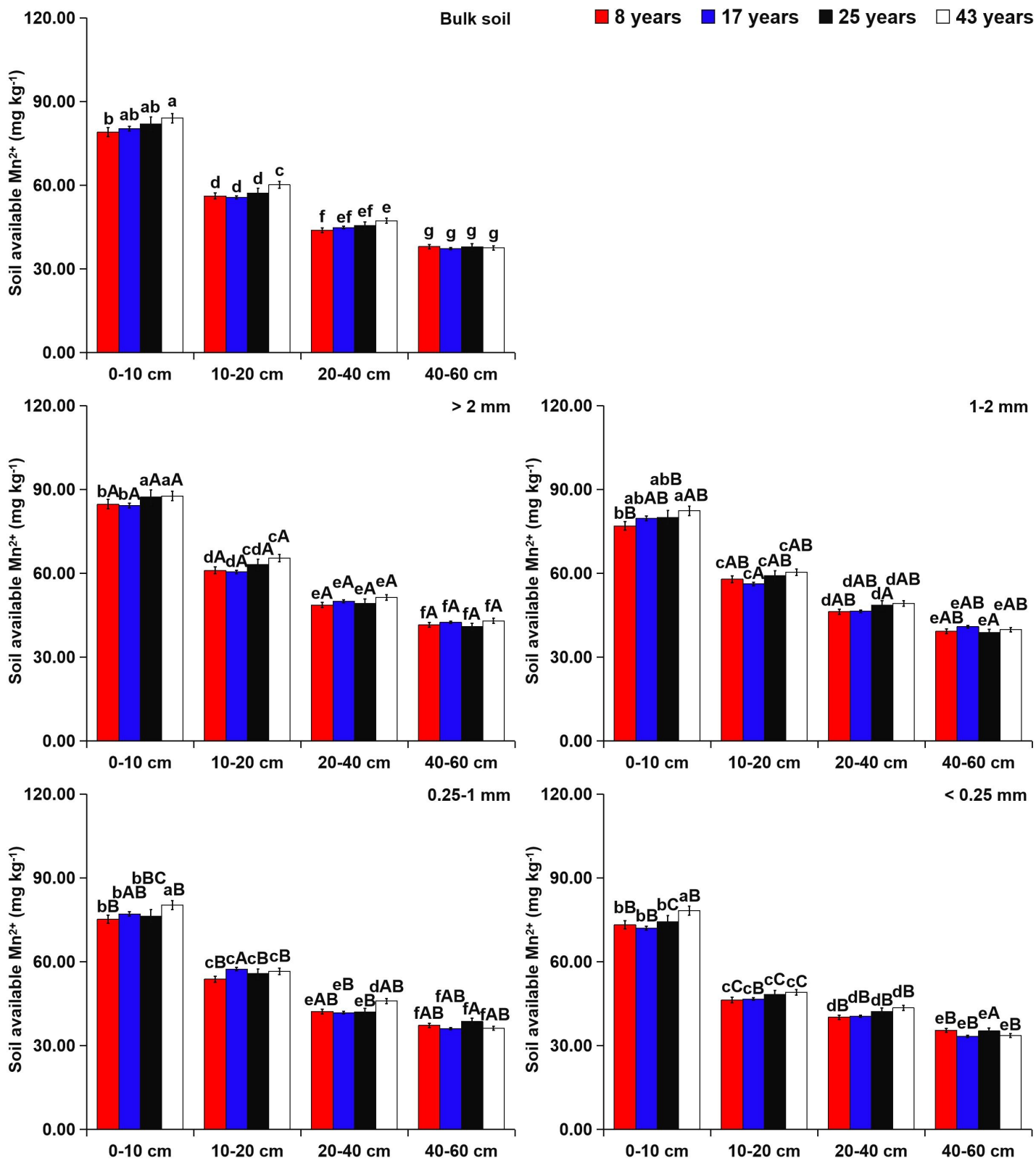


Figure 8