



- 1 Dynamics of soil aggregate-related stoichiometric characteristics with tea-planting age and
- 2 soil depth in the southern Guangxi of China
- 3 Ling Mao, Shaoming Ye, Shengqiang Wang*
- 4 Forestry College, Guangxi University, Nanning, 530004, China
- 5 *Corresponding author.
- 6 E-mail address: shengqiang@gxu.edu.cn
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8 ABSTRACT

Soil ecological stoichiometry offers a sort of effective way to explore the distribution, cycling, 9 limitation, and balance of chemical elements in tea plantation ecosystems. This study was aim to 10 explore how soil organic C (OC) and nutrient contents (total N (TN), total P (TP), Ca2+, Mg2+, 11 Fe²⁺, and Mn²⁺) as well as their stoichiometric ratios (C/N, C/P, N/P, Ca/Mg, and Fe/Mn) vary 12 with tea-planting age (8, 17, 25, and 43 years) and soil depth (0-10, 10-20, 20-40, and 40-60 cm) 13 at the aggregate scales in the southern Guangxi of China. Our results showed that tea-planting 14 age and soil depth significantly influenced soil stoichiometric characteristics in various sized 15 aggregates. In different aged tea plantations, soil OC, TN, and TP contents as well as C/N, C/P, 16 and N/P ratios significantly decreased as the soil depth increased. In addition, soil Ca²⁺ and Mg²⁺ 17 contents were significantly lower in the surface soil layer than the deeper soil layer, whereas soil 18 Fe²⁺ and Mn²⁺ contents showed totally opposite trends, and no significant differences were 19 detected among different soil depths in Ca/Mg and Fe/Mn ratios. Tea-planting age could 20 influence the variations in soil stoichiometric characteristics, but such effects were more obvious 21 22 at the 0-40 cm soil depth in contrast to the 40-60 cm soil depth. At the 0-40 cm soil depth, continuous planting of tea was beneficial for the cumulation of soil OC, total N (TN), Fe²⁺, and 23 Mn^{2+} , whereas soil Ca^{2+} and Mg^{2+} were susceptible to leaching losses over time. Compared with 24 other tea-planting regions in China, soil C/N ratio was higher in this tea-planting region, whereas 25 soil C/P and N/P ratios were much lower, indicating the lower contents of soil OC and TN, 26





27 especially the TN. Therefore, an appropriate increase in the amount of N fertilizer should be applied in this tea-planting region. During the process of tea planting, the losses of soil Ca^{2+} and 28 Mg^{2+} , especially the Ca^{2+} (as indicated by the decrease in soil Ca/Mg ratio), could lead to the soil 29 acidification. Soil acidification could reduce Fe^{2+} absorption and enhance Mn^{2+} uptake by tea 30 plant (as indicated by the increase in soil Fe/Mn ratio), thereby causing the aggravation of Fe^{2+} 31 insufficiency and the emergence of Mn²⁺ toxicity to tea plant. Overall, this study improved the 32 understanding of soil OC and nutrient dynamics in tea plantation ecosystems, and also provided 33 supplementary information for soil ecological stoichiometry in global terrestrial ecosystems. 34

35 KEYWORDS

36 Tea-planting age; Soil depth; Soil aggregate; Ecological stoichiometry

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38 1. Introduction

Ecological stoichiometry offers a sort of valid approach to explore the distribution, cycling, 39 restriction, and balance of nutrients in terrestrial ecosystems (Yu et al., 2019), and plays a critical 40 role in recognizing the influence factors and drive mechanisms in ecological processes (Su et al., 41 42 2019). On the one hand, carbon (C) is the most commonly seen element in plants (Prescott et al., 43 2020), and nitrogen (N) and phosphorus (P) are critical control factors for the growth of plants (Krouk and Kiba, 2020). The relationships amongst the three different elements are coupled 44 45 (Elser et al., 2003). Soil C/N, C/P, and N/P ratios represent not only the equilibrium features of 46 soil C, N, and P, but also the dynamics of fertility characteristics during the process of soil genesis (Bai et al., 2020). On the other hand, calcium (Ca), magnesium (Mg), iron (Fe), and 47 48 manganese (Mn) are pivotal metallic nutritive elements for the development of plants (Liu et al., 49 2021a). Soil total Ca, Mg, Fe, and Mn may exceed the demand of a single plant by more than a thousand-fold and cannot sensitively reflect the needs of plants (Miner et al., 2018), but the 50 available fractions of these nutrients may be insufficient or redundant, resulting in the 51 deficiencies or abundances of plant nutrients (Otero et al., 2013). Thus, soil exchangeable Ca and 52





Mg as well as available Fe and Mn generate significant effects on the development of plants. Soil Ca/Mg ratio reflects the relative effectiveness of these two ions and influences the buffering capacity and acidification process in soil (Yin et al., 2016). Moreover, maintaining a proper soil Fe/Mn ratio is pivotal for healthy soil because a lower ratio may indicate that plants have encountered Fe depletion and Mn poisoning (Wang et al., 2017a).

Over the past decade, soil stoichiometric characteristics (mainly C-N-P, rather than Ca-Mg 58 or Fe-Mn) has been broadly studied across the world (Tian et al., 2010; Yang et al., 2013; Zhang 59 et al., 2016; Yue et al., 2017; Yu et al., 2018; Qiao et al., 2020). A wide agreement exists 60 amongst these studies that soil depth is vital for the regulation of soil stoichiometric 61 characteristics. Substantial studies have identified the decreasing trend of soil organic C (OC), 62 total N (TN), and total P (TP) contents as the soil depth increased (Yue et al., 2017; Yu et al., 63 2018; Qiao et al., 2020), whereas conflicting vertical patterns were discovered for soil C/N, C/P, 64 and N/P ratios. For instance, decreasing trend of the C/P and N/P ratios was observed as the soil 65 depth increased in the data of the 2nd soil investigation in China (Tian et al., 2010). Nevertheless, 66 larger C/N ratio in the deeper soil layer, not the surface soil layer, was identified in a mollisol 67 plain in the northeast China (Zhang et al., 2016). Moreover, the C/N ratio displayed no 68 remarkable change throughout different soil depths in an investigation of alpine grassland on the 69 Qingzang Plateau (Yang et al., 2013). As shown above, inconsistent vertical patterns have been 70 71 reported for the C-N-P stoichiometric ratios in different soil ecosystems. Meanwhile, these 72 studies were mainly focused on the regional or global scales, rather than on the aggregate scales.

Soil aggregates constitute the fundamental parts of soil structure, and various sized aggregates exert different abilities in the supply and reserve of soil OC and nutrients (Six et al., 2004). Thus, to improve the comprehension about the structure and function of soil ecosystems, more efforts should be made to observe the soil stoichiometric characteristics at the aggregate scales (Xu et al., 2019; Cui et al., 2021). In recent period, lots of studies have reported the OC, TN, and TP distribution in various sized aggregates, but these studies are ended with different





results. To be specific, some studies revealed the significant increases in the OC, TN, and TP contents as the aggregate size decreased (Sarker et al., 2018; Piazza et al., 2020). Nevertheless, some other studies drew the totally opposite trends (Lu et al., 2019; Liu et al., 2021b). These show that the changes of soil OC, TN, and TP at the aggregate scales have received great attention, whereas soil exchangeable alkali cations (i.e., Ca^{2+} and Mg^{2+}) and available micronutrients (i.e., Fe^{2+} and Mn^{2+}) are rarely investigated.

In the past century, under the remarkable increase in population pressure, continuous tillage 85 and overmuch deforestation resulted in the dramatic decrease in soil fertility level in the southern 86 Guangxi of China (Jiang et al., 2018). For the purpose of tackling these challenges, the Chinese 87 government has rolled out the Grain for Green program in the hope of alleviating land 88 deterioration via converting farmlands to forest lands or grass lands (Zeng et al., 2020). Since the 89 initiation of such program, the south part of Guangxi has initiated the mode of transforming 90 farmlands into tea (Camellia sinensis L.) plantations as per the local geography and natural 91 92 resources (Zhang et al., 2017). Tea, as a pivotal cash crop, is commonly cultivated in the developing nations, particularly in China, India, Kenya, and Sri Lanka. China is the first nation 93 94 to plant tea across the globe, with the tea-planting area reaching 3.17 million hectares in 2020 95 and presenting an elevating trend in the future (Chinese Tea Committee, 2020). Guangxi has the subtropic monsoon climate and marks the key tea-planting region in China. According to the 96 97 statistics from Chinese Tea Committee (2020), more than 80% tea plantations of Guangxi are 98 situated at impoverished counties, and tea-planting industry turns to be the staple industry on 99 which poor counties depend to throw off poverty.

Our past studies indicated that the landuse shift from farmlands to tea plantations could ameliorate soil fertility level (Zheng et al., 2011). Nevertheless, during the process of tea planting, the variations in soil stoichiometric characteristics are still unclear. Meanwhile, since tea plant serves as a deep root plant, it is vital to reveal how stoichiometric characteristics change with increasing soil depth in tea plantation ecosystems. Thus, the present study was carried out to





investigate how soil OC and nutrient contents as well as their stoichiometric ratios vary with tea-planting age (8, 17, 25, and 43 years) and soil depth (0-10, 10-20, 20-40, and 40-60 cm) at the aggregate scales (< 0.25, 0.25-1, 1-2, and > 2 mm). In addition, we assumed that the responses of soil OC and nutrient contents and their stoichiometric ratios to tea-planting age would be different amongst different soil depths.

110 2. Materials and methods

111 2.1. Experiment site

In January 2019, the present study was completed at the Hengxian Agriculture Experiment 112 Center of Guangxi University (altitude of 557-563 m and slope degree of 13-15°) (Figure S1). 113 Subtropic monsoon climate is predominant. Yearly average rainfall and temperature register 114 1304 mm and 21.6 °C, separately. Exposed soil horizon occurs early in the Mesozoic, which 115 gradually formed the Ultisols agrotype (IUSS Working Group, 2014). As early as in 1960s, due 116 to the high economic value of tea (especially "Baimao tea"), massive hectares of farmlands were 117 118 developed to tea plantations in such region. In the tea-planting course, tillage method is no tillage and tea-planting density is almost 6×10^4 plants ha⁻¹. Yearly fertilization regime has been 119 displayed in our past studies (Wang and Ye, 2020). In all tea plantations, herbicides were not 120 121 applied and yellow sticky boards were used to prohibit pests, because the color may attract pests and get them stuck on the boards. In addition, all tea plants were subjected to slight pruning in 122 123 September each year.

124 2.2. Experiment design

In general, examining the same location persistently has been considered a quite effective approach in the monitoring of the variations in soil with time (Sparling et al., 2003). Nevertheless, the challenges in long-period soil monitoring have made it urgent to develop substitutional approaches to research the changes of soil over time, amongst which the most common approach is the 'space-for-time' alternative (Zanella et al., 2018). To be specific, disperse sites ('space') in diverse developmental phases are identified simultaneously to obtain a





chronological sequence of ages ('time'). In accordance with the 'space-for-time' alternative,
disperse spots of increasing ages display alike initial status and synchro-sampling at these
disperse spots equals the re-sampling at the same spot in different ages.

- In this study, such approach was used to explore the variations in soil stoichiometric 134 135 characteristics in a chronological sequence of tea plantations. In general, certain underlying mixture effects exist in the spatial variations of soil, hence the present study manages to mitigate 136 such effects via choosing tea plantations, which were cultured with the same tea variety 137 ("Baimao tea") with different planting ages (8, 17, 25, and 43 years), and were located at the 138 same unit associated with geomorphological status. Every tea-planting age was duplicated in 139 quintuplicate, and afterwards generated 20 tea plantations. Separation amongst these tea 140 plantations was completed with distances of > 800 m between each other, hence decreasing the 141 space self-correlation and avoiding the pseudo-replication. For every tea plantation (S $\approx 1 \times 10^4$ 142 m²), a plot (S = 20 m \times 20 m) was randomly established with distance of > 50 m away from the 143 tea plantation margin. 144
- 145 2.3. Litterfall and soil sampling

146 For every plot, the 5 litterfall specimens had been acquired from the surface of soil in the 5 147 randomly chosen subplots (S = 1 m \times 1 m), and afterwards were integrated into a composite litterfall specimen. An overall the 20 (4 tea-planting ages \times 5 replicates) composite litterfall 148 149 specimens were desiccated at the 80 °C until steady weight. Then, the weights of these 150 desiccated litterfall specimens were measured, and the litterfall C (Nelson and Sommers, 1996) 151 and N (Bremner, 1996) contents were detected. Soil sampling was completed in the same sites of 152 the litterfall sampling. For every plot, the 5 soil specimens had been acquired by a spade from every soil layer (i.e., 0-10, 10-20, 20-40, and 40-60 cm) in the 5 subplots, and afterwards were 153 integrated into a composite soil specimen. An overall the 80 (4 tea-planting ages \times 4 soil layers \times 154 5 replicates) composite soil specimens were gently separated into naturally formed aggregates, 155 156 which were subjected to filtration by a 5 mm sifter to realize the removals of small stones, coarse





- roots, and macrofauna. After that, soil specimens were used for the aggregate separation. For every plot, moreover, extra 5 soil specimens were randomly chosen via cutting rings (V = 100 cm^{-3} , $\emptyset = 50.46$ mm, and depth = 50 mm) from every soil layer to assess the bulk density, clay (< 0.002 mm), pH, OC, and nutrients of bulk soil.
- 161 2.4. Soil aggregate separation

As per the process of wet screening, 250 g of every composite soil specimen was subjected 162 to filtration via the 2, 1, and 0.25 mm sieves in a successive way (Kemper and Chepil, 1965). To 163 be specific, the composite soil specimens were soaked by the aqua destillata for 15 min, and 164 afterwards were oscillated in the vertical direction for 15 min at the 1 s⁻¹ oscillating rate and 5 cm 165 amplitude. Consequently, we obtained 4 various sized aggregates, covering microaggregates (< 166 0.25 mm), fine (0.25-1 mm), medium (1-2 mm), and coarse (> 2 mm) macroaggregates. All of 167 the aggregates were desiccated and weighted, and then aggregate-related OC and nutrients were 168 detected. 169

170 2.5. Soil property analyses

Prior to the analyses of soil physical-chemical properties, soil specimens were subjected to 171 172 atmospheric drying under indoor temperature condition. According to the cutting ring method 173 (Lu, 2000), soil specimens were oven-dried at 105 °C to the stable weight in order to measure the bulk density. Soil clay was detected by the hydrometer (TM-85, Veichi, China) (Lu, 2000). Soil 174 175 pH was detected by the glassy electrode (MT-5000, Ehsy, China), with the ratio of soil : water 176 (mass : volume) as 1 : 2.5 (Lu, 2000). Soil OC and TN were identified via the acid dichromate wet oxidation method (Nelson and Sommers, 1996) and the micro-Kjeldahl method (Bremner, 177 178 1996), separately. Soil TP was identified via the molybdate blue colorimetry method (Bray and Kurtz, 1945). Soil exchangeable alkali cations (i.e., Ca^{2+} and Mg^{2+}) were abstracted by the 179 ammonium acetate (CH₃COONH₄) (Thomas, 1982). In short, 2.5 g of every aggregate fraction 180 was weighted into Erlenmeyer flask to blend with 50 mL 1 M CH_3COONH_4 (pH = 7.0). The 181 182 extract liquid was agitated for 30 min under 150 rpm, and afterwards subjected to filtration via





183 Whatman No. 2 V filtration paper (quantitative and ashfree). Soil available micronutrients (i.e., Fe^{2+} and Mn^{2+}) were abstracted by the diethylenetriamine pentaacetic acid (DTPA) (Lindsay and 184 Norvell, 1978). In short, 10 g of every aggregate fraction was weighted into Erlenmever flask to 185 blend with 20 mL 0.005 M DTPA + 0.01 M CaCl₂ + 0.1 M TEA (triethanolamine) (pH = 7.0). 186 187 The extract liquid was agitated for 2 h under 180 rpm, and afterwards subjected to filtration. Entire extractable metallic cations were detected by the atomic absorption spectrometer (AAS, 188 189 Shimadzu, Japan). In this study, 5 standard specimens (GBW-07401), 5 blank specimens, and 80 parallel specimens (accounted for 20% of the total soil specimens) were used to control quality, 190 191 and the error is controlled in 5%. 192 2.6. Calculations and statistics The mean weight diameter (MWD, mm) was utilized to indicate the stability of soil 193 aggregates (Kemper and Chepil, 1965): 194 $MWD = \sum_{i=1}^{n} (X_i \times M_i),$ 195 in the formula, X_i indicates the ith size aggregates' mean diameter (mm) and M_i indicates the 196 *i*th size aggregates' proportion (% in weight). 197 198 In the present study, since tea-planting age and soil depth serve as the two main factors, 199 statistic analysis was conducted separately by aggregate size. SPSS 22.0 was used for statistic analysis (Table S1). One-way analysis of variance (ANOVA) was taken for exploring the effect 200 201 of tea-planting age on the litterfall characteristics. Two-way ANOVA was taken for exploring 202 the effects of tea-planting age and soil depth on the soil characteristics. Besides that, Pearson 203 correlation analysis was utilized to test the relationships between pH and stoichiometric ratios 204 (i.e., Ca/Mg and Fe/Mn ratios) in bulk soil during the process of tea planting. 205 3. Results

206 3.1. Composition and stability of soil aggregates

At the 0-10 and 10-20 cm soil depths, continuous planting of tea resulted in remarkable variations in the proportions of various sized aggregates, apart from the medium and fine





209 macroaggregates (Figure 1). To be specific, the proportions of coarse macroaggregates remarkably rose within the first 17 years and afterwards remarkably dropped, whereas the 210 211 proportions of microaggregates displayed an opposite trend over time. Meanwhile, the greatest value of soil MWD was identified in the tea plantations of 17 years (Figure 1). Notably, the role 212 213 of tea-planting age in the aggregate composition and stability is limiting at the 20-40 and 40-60 cm soil depths. Across the 4 tea-planting ages, the coarse macroaggregates were prevailing at the 214 0-10 cm soil depth, which accounted for 32.60%-53.18% of bulk soil. However, at the 10-20, 215 216 20-40, and 40-60 cm soil depths, the microaggregates were dominant, which accounted for 217 33.80%-49.51%, 42.12%-48.24%, and 44.80%-49.45%, respectively. These results showed that the coarse macroaggregate proportions reduced while the microaggregate proportions elevated 218 with increasing soil depth. 219

220 3.2. Contents of soil C, N, and P

At the aggregate scales, soil OC (Figure 2) and TN (Figure 3) contents increased with 221 increasing aggregate size, but the distribution of soil TP (Figure 4) was even in various sized 222 aggregates. In the tea-planting course (8-43 years), the aggregate-related OC and TN contents at 223 the 0-10, 10-20, and 20-40 cm soil depths were significantly elevated by 22%-35% and 224 225 14%-24%, 11%-22% and 9%-17%, and 8%-18% and 9%-13%, respectively. Nevertheless, no remarkable variation existed in the aggregate-related TP content. Furthermore, at the 40-60 cm 226 227 soil depth, the aggregate-related OC, TN, and TP contents did not show significant variations 228 over time. Regardless of the tea-planting age, decreasing trend of the aggregate-related OC, TN, 229 and TP contents was observed as the soil depth increased.

230 3.3. Stoichiometric ratios of soil C, N, and P

In this study, the increases in aggregate-related C/N (Table 2), C/P (Table 3), and N/P (Table 4) ratios were accompanied by the increasing aggregate size. At the 0-10, 10-20, and 20-40 cm soil depths, aggregate-related C/N ratio did not show remarkable variation while aggregate-related C/P and N/P ratios remarkably increased during the process of tea planting.





Moreover, there was little role of tea-planting age in the aggregate-related C/N, C/P, and N/P ratios at the 40-60 cm soil depth. In different aged tea plantations, aggregate-related C/N, C/P, and N/P ratios dropped as the soil depth increased. For example, at the 0-10 cm soil depth, aggregate-related C/N, C/P, and N/P ratios across the 4 tea-planting ages fluctuated in 20.81-23.04, 28.81-37.07, and 1.31-1.67, respectively. Meanwhile, at the 40-60 cm soil depth, aggregate-related C/N, C/P, and N/P ratios fluctuated in 16.41-20.74, 13.44-22.88, and 0.84-1.08, respectively.

242 3.4. Contents of soil alkali cations and micronutrients

At the aggregate scales, soil exchangeable alkali cations (i.e., Ca^{2+} and Mg^{2+}) were mainly 243 distributed in the microaggregates (Figures 5 and 6). However, soil available micronutrients (i.e., 244 Fe^{2+} and Mn^{2+}) were mainly existed in the coarse macroaggregates (Figures 7 and 8). In the 245 tea-planting course (8-43 years), the aggregate-related Ca²⁺ and Mg²⁺ contents at the 0-10, 10-20, 246 and 20-40 cm soil depths were significantly reduced by 31%-38% and 10%-24%, 23%-27% and 247 9%-18%, and 10%-16% and 5%-8%, respectively. However, the aggregate-related Fe²⁺ and 248 Mn^{2+} contents were significantly elevated by 16%-27% and 6%-9%, 11%-15% and 4%-7%, and 249 7%-12% and 3%-5%, respectively. In addition, at the 40-60 cm soil depth, the contents of 250 251 aggregate-related exchangeable alkali cations and available micronutrients did not show significant variations over time. Irrespective of the tea-planting age, increasing trend of the 252 aggregate-related Ca²⁺ and Mg²⁺ contents was observed with increasing soil depth, but the 253 aggregate-related Fe²⁺ and Mn²⁺ contents showed an opposite trend. 254

255 3.5. Stoichiometric ratios of soil alkali cations and micronutrients

In this study, soil Ca/Mg (Table 5) and Fe/Mn (Table 6) ratios were evenly distributed in various sized aggregates. At the 0-10, 10-20, and 20-40 cm soil depths, aggregate-related Ca/Mg ratio remarkably decreased while aggregate-related Fe/Mn ratio remarkably increased in the tea-planting course. Moreover, there was little role of tea-planting age in the aggregate-related Ca/Mg and Fe/Mn ratios at the 40-60 cm soil depth. In different aged tea plantations, no





variations were observed amongst different soil depths in aggregate-related Ca/Mg and Fe/Mn ratios. For example, at the 0-10 cm soil depth, aggregate-related Ca/Mg and Fe/Mn ratios across the 4 tea-planting ages ranged from 1.81 to 1.96 and 0.76 to 0.85, respectively. Meanwhile, at the 40-60 cm soil depth, aggregate-related Ca/Mg and Fe/Mn ratios ranged from 1.88 to 1.92 and 0.78 to 0.82, respectively.

266 4. Discussion

267 4.1. Composition and stability of soil aggregates

Tea-planting age significantly influenced the aggregate composition and stability at the 0-10 268 and 10-20 cm soil depths, whereas the effect at the 20-40 and 40-60 cm soil depths was quite 269 limited (Figure 1). In the early (8-17 years) period, tea planting was beneficial for the transition 270 from microaggregates to coarse macroaggregates at the 0-10 and 10-20 cm soil depths. By 271 comparison, in the middle (17-25 years) and late (25-43 years) periods, tea planting induced 272 coarse macroaggregate destruction and microaggregate release. According to the hierarchical 273 concept of soil aggregates (Six et al., 2004), the quality of plant litterfall returning to the soil 274 determines the distribution of decomposition products of litterfall in various sized aggregates, 275 276 which ultimately impacts the aggregate composition. In the early period of tea planting, tea 277 litterfall displayed greater availability (as indicated by the lower litterfall C/N ratio) (Table 1), revealing that the decomposition products of litterfall were easily combined into the coarse 278 279 macroaggregates, hence fostering the formation of coarse macroaggregates (Tisdall and Oades, 280 1982). Reversely, in the middle and late periods of tea planting, tea plants naturally encountered 281 aging processes and litterfall was progressively subjected to humification, which induced the 282 decomposition of coarse macroaggregates into microaggregates (Six and Paustian, 2014). 283 Moreover, the reduced litterfall amount and covering area after 17 years of tea planting (Table 1) enhanced the rainfall eluviation and artificial interferences (i.e., pruning of tea plants and 284 application of fertilizers), which also caused the destruction of coarse macroaggregates. In the 285 286 tea-planting course, variation in aggregate stability was indicated via the change of MWD value.





At the 0-10 and 10-20 cm soil depths, the MWD value was the greatest in the 17 years of tea planting (Figure 1), which was associated with the highest proportions of coarse macroaggregates in the 17-year tea plantations. These findings indicated that the 17-year tea plantations exhibited stronger aggregate stability in contrast to other plantations at the 0-10 and 10-20 cm soil depths.

292 Regardless of the tea-planting age, coarse macroaggregates were dominant in the topsoil (0-10 cm) while microaggregates were dominant in the subsoil (10-60 cm) (Figure 1), indicating 293 294 transformation of aggregate composition from coarse macroaggregate-prevailing to microaggregate-prevailing with the increase in soil depth. Also, alike outcomes were 295 corroborated by Li et al. (2015) and Zhu et al. (2017) from studies on tea plantations in the 296 southwest Sichuan of China. In the present study, coarse macroaggregates were the prevailing 297 fractions in the topsoil, not the subsoil, which was attributed to the surface cumulation of soil OC 298 (Figure 2). As an essential cementing agent, soil OC could foster the formation of coarse 299 macroaggregates (Al-Kaisi et al., 2014). Moreover, the reduced proportions of coarse 300 macroaggregates as the soil depth increased were also because of the elevated soil compactness 301 (as indicated by the bulk density) (Table 1). Soil densification could prevent the growth of plant 302 303 roots, hence causing the activities of soil microorganisms decreased, especially soil fungi (Kurmi et al., 2020). Reduced activities of soil fungi could diminish the production of polysaccharose 304 305 and glomalin-related soil protein (GRSP) from the fungal hyphae, hence inducing the 306 proportions of soil macroaggregates decreased (Ji et al., 2019). Likewise, as per our past studies (Wang et al., 2017b; Zhu et al., 2019), soil microbial activities and GRSP content served as the 307 308 vital effects in the formation and stabilisation of soil macroaggregates, and presented the higher 309 levels in the topsoil compared with the subsoil in tea plantation ecosystems. With increasing soil depth, the decrease in MWD value (Figure 1) was mainly related to the change of soil aggregate 310 composition, especially for the decomposition of coarse macroaggregates into microaggregates, 311





313 4.2. Contents of soil C, N, and P

In this study, more contents of soil OC and TN could be detected in coarse macroaggregates 314 (Figures 2 and 3), which conformed to the findings of Six et al. (2004) that macroaggregates 315 were comprised of microaggregates via temporary binding agents (i.e., microbe- and 316 317 plant-originated polysaccharides, fungal mycelium, and plant roots); meanwhile, macroaggregates could provide the protection for the organic matters (OMs), hence causing the 318 319 cumulation of OC and TN in macroaggregates. Unlike soil OC and TN, soil TP was evenly distributed in various sized aggregates (Figure 4). Moreover, Bhatnagar and Miller (1985) also 320 detected alike outcomes from soil specimens subjected to fresh poultry manure treatments, and 321 propelled the causal links influencing the TP distribution in soil aggregates. Specifically, (i) 322 introduced P was firstly adsorbed by clay particulates in soil and clay particulates were 323 discrepant in various sized aggregates, and (ii) introduced P had selective absorptive properties 324 for various sized aggregates. According to our findings, stochasticity seems to be one probable 325 mechanism that sheds light on the TP distribution in soil aggregates. 326

Continuous planting of tea could positively affect the cumulation of soil OC and TN, but 327 328 such positive effects were more obvious at the 0-40 cm soil depth in contrast to the 40-60 cm soil 329 depth (Figures 2 and 3). In this study, soil OC and TN contents exhibited a significant growing trend over time, which was possibly associated with the following mechanisms. First, many 330 331 long-period tests had demonstrated the proactive roles of manure and chemical fertilizer 332 applications in soil OM cumulation (Tong et al., 2009; Zhou et al., 2013). Similarly, in the tea-planting course, growing soil OC and TN contents were probably caused by the applications 333 of substantial swine manure every year (12 Mg ha⁻¹ year⁻¹) in this tea-planting region (Wang and 334 335 Ye, 2020). Second, plants serve as the prime OM sources in soil via root exudates and litterfall remains (Franklin et al., 2020). In the tea-planting course, soil OC and TN cumulation probably 336 occurred as a result of the growing root systems and the increasing amounts of aboveground 337 338 litterfall attained from trimmed branches and leaves. Third, no tillage could provide physical





339 protection for the OMs combined with soil aggregates, and then further improve soil OC and TN sequestration (Wulanningtyas et al., 2021). Notably, although the positive correlations of OC and 340 341 TN contents with clay content in soil have been reported, the present study revealed that significant increases in the OC and TN contents were accompanied by no significant variation in 342 343 the clay content during the process of tea planting (Table 1). Similarly, Li et al. (2015) and Wang et al. (2018) discovered as well that the changes of soil OC and TN contents were not influenced 344 by the clay content over time in tea plantation ecosystems, mainly because soil OC and TN 345 contents primarily depend on fertilization, tillage, root exudates, and litterfall remains, but soil 346 clay content is mainly controlled by its parent material (Rakhsh et al., 2020). Unlike soil OC and 347 TN, regardless of the soil depth, no remarkable difference existed in soil TP content amongst 348 different aged tea plantations (Figure 4), which implied the resistance of soil TP content to the 349 change of tea-planting age. Also, past studies verified that soil TP content was not related to the 350 tea-planting age (Wu et al., 2018; Yan et al., 2018), as soil P primarily derives from the 351 weathering release of soil minerals, instead of the short-period biology cycle (Cui et al., 2019). 352

In tea plantation ecosystems, the decreasing OC, TN, and TP contents with increasing soil depth (Figures 2, 3, and 4) coincided with some past findings in other ecosystems, such as tropic forests, bushlands, and grasslands (Stone and Plante, 2014; Yu et al., 2018; Qiao et al., 2020). In the present study, the higher contents of OC, TN, and TP in the topsoil were associated with the higher OM input, in which the soil OM contents in the topsoil were enriched by the input of surface tea litterfall, root debris and exudates, and swine manure.

359 4.3. Stoichiometric ratios of soil C, N, and P

Soil C/N, C/P, and N/P ratios act as vital indicators of soil health (Liu et al., 2018), which can be employed for exploring C circulation and guiding the equilibrium between N and P in soil ecosystems (Sardans et al., 2012). In this study, soil C/N ratio grew with growing aggregate size (Table 2), which indicated that the OMs in macroaggregates were younger and more unstable in contrast to microaggregates (Six et al., 2004). Meanwhile, the OMs associated with





365 microaggregates experienced more degradation, resulting in the lower C/N ratio in the microaggregates (Xu et al., 2019). In different aged tea plantations, soil OC and TN were 366 predominantly distributed in the coarse macroaggregates (Figures 2 and 3), but the TP was 367 evenly distributed in various sized aggregates (Figure 4). As a result, the associations of C/P and 368 N/P ratios to aggregate size primarily depended on the relationships of OC and TN contents with 369 aggregate size (Tables 3 and 4). As far as we know, the changes of soil C/P and N/P ratios at the 370 aggregate scales are rarely examined, although these kinds of knowledge are imperative because 371 of the biogeochemical cycles of N and P being influenced by the dynamics of soil aggregates 372 (Cui et al., 2021). Consequently, the impact generated by the aggregate size on the C/P and N/P 373 ratios ought to be studied more for the accurate forecast of soil N and P cycling under natural or 374 man-intervened ecosystems. 375

Irrespective of the soil depth, soil C/N ratio showed little significant variation in the 376 tea-planting course (Table 2). Meanwhile, tea-planting age significantly affected soil C/P and 377 N/P ratios at the 0-40 cm soil depth, not the 40-60 cm soil depth (Tables 3 and 4). Soil C/N ratio 378 is generally treated as the critical indicator which affects the formation and degradation of soil 379 380 OMs (Khan et al., 2016). Since response of soil TN content to soil environment change is almost 381 the same as soil OC content (Wang et al., 2018), soil C/N ratio did not show significant difference amongst different aged tea plantations (Table 2). Likewise, Zhou et al. (2018) proved 382 383 that no close correlation existed between soil C/N ratio and vegetation coverage, because C and 384 N are structure elements and their cumulation and consumption in soil remain relative consistency. Soil C/P ratio is the indicator suggesting P effectiveness, and higher C/P ratio often 385 386 denotes lower P effectiveness (Khan et al., 2016). In acidic soil (Table 1), available P was 387 adsorbed on the surfaces of Fe/Al oxides and clay minerals in a preferential way, because Fe/Al oxides and clay minerals with greater surface areas could afford enough sites to available P 388 adsorption (Wu et al., 2018). As the tea-planting age increased, therefore, soil acidification led to 389 390 the decrease in P effectiveness (evidenced by the significant increase in soil C/P ratio) (Table 3).





Soil N and P are the prohibiting factors mostly seen during the process of plant growth, and thus, N/P ratio can be utilized as one efficient indicator that shows nutrient restriction (Khan et al., 2016). In this study, soil N/P ratio remarkably increased in the tea-planting course (Table 4), mainly because soil TN content experienced significant increase while no such significant change was found in TP content over time.

Soil C/N ratio decreased with increasing soil depth, regardless of the tea-planting age 396 (Table 2), which coincided with the majority of studies (Cao et al., 2015; Feng and Bao, 2017, 397 Yu et al., 2019). Batjes (1996) suggested that the decrease in soil C/N ratio as the soil depth 398 increased was triggered by the stratification of humic substance in the soil profile. Moreover, in 399 this study, the lower soil C/P and N/P ratios in the subsoil (Tables 3 and 4) backed the outcomes 400 of past studies in terrestrial ecosystems of China, which were on the foundation of the data from 401 both the 2nd soil investigation in China (Tian et al., 2010) and the Chinese Ecosystem Research 402 Network (CERN) (Chai et al., 2015). 403

Across the 4 tea-planting ages, the mean contents of OC and TN in bulk soil (0-20 cm) were 404 16.70 and 0.77 g kg⁻¹, separately, which were below the mean contents of OC (21.30 g kg⁻¹) and 405 TN (2.17 g kg⁻¹) in Chinese tea plantations (Sun et al., 2020; Xie et al., 2020). Moreover, in this 406 407 tea-planting region, the mean content of TP in bulk soil (0-20 cm) was 0.57 g kg⁻¹, corresponding to the moderate level in Chinese tea plantations, where TP content varied in the range of 408 409 0.35-1.20 g kg⁻¹ (Wu et al., 2018; Sun et al., 2020). Herein, soil C/N ratio is higher compared 410 with other tea-planting regions in China, whereas soil C/P and N/P ratios are much lower (Sun et 411 al., 2020). These findings are primarily associated with the lower contents of soil OC and TN, 412 especially the TN. In general, N is the most limiting element in the net primary production of tea 413 plantation ecosystems (Miner et al., 2018), and this phenomenon also appeared in the southern 414 Guangxi of China.

415 4.4. Contents of soil alkali cations and micronutrients

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





contents of exchangeable alkali cations (including Ca^{2+} and Mg^{2+}) were detected in both 2-4.76 417 and < 0.25 mm aggregates in the non-tillage soil. In the tillage course, however, the contents of 418 these two cations decreased in the 2-4.76 mm aggregates and increased in the < 0.25 mm 419 aggregates, revealing that the tillage practice could cause soil Ca^{2+} and Mg^{2+} to redistribute in 420 various sized aggregates. In comparison, the present study exhibited that the distribution of soil 421 Ca^{2+} and Mg^{2+} in aggregates was similar in different aged tea plantations (Figures 5 and 6), 422 implying that the distribution of these two cations in aggregates was seldom influenced by the 423 tea-planting age. To be specific, coarse macroaggregates had the lowest contents of Ca^{2+} and 424 Mg^{2+} , whereas microaggregates exhibited the highest contents. These findings could be ascribed 425 to the larger specific surface areas of microaggregates (Adesodun et al., 2007), which increased 426 microaggregates' adsorption to Ca²⁺ and Mg²⁺ derived from root exudates, litterfall remains, and 427 manure (Emadi et al., 2009). Unlike exchangeable alkali cations, the contents of soil available 428 micronutrients (including Fe^{2+} and Mn^{2+}) usually correspond to the contents of soil OMs (Wang 429 et al., 2017a), which are more abundant in macroaggregates (Six et al., 2004). Similarly, this 430 study also found that the Fe^{2+} and Mn^{2+} had a similar distribution pattern with OC at the 431 432 aggregate scales (Figures 7 and 8). Since the decomposition products of litterfall can be easily 433 integrated to the coarse macroaggregates (Six et al., 2004), the nutrient cycling of plant-soil systems might lead to the higher contents of soil Fe²⁺ and Mn²⁺ in the coarse macroaggregates 434 435 (Wang et al., 2017a).

At the 0-40 cm soil depth, the contents of soil Ca^{2+} and Mg^{2+} significantly decreased over time (Figures 5 and 6), which might be due to the applications of urea and NH_4^+ -N fertilizer in the tea-planting course for increasing tea leaf outputs. Urea hydrolysis can promote the production of ammonium ions which are readily nitrified into nitrate, and the excessive proton produced by the nitrification can compete for the adsorption sites with Ca^{2+} and Mg^{2+} (Wang et al., 2017a). As a result, these cations were easy to lose from soil in the manner of leaching. Except at the 40-60 cm soil depth, continuous planting of tea led to the remarkable increases in





soil Fe²⁺ and Mn²⁺ contents, which were elevated by 7%-27% and 3%-9% from 8 to 43 years of 443 tea planting, separately (Figures 7 and 8). This phenomenon was possibly caused by the soil 444 acidification (Table 1), which stimulates the release of soil Fe^{2+} and Mn^{2+} by mineralization and 445 desorption from soil OMs and minerals (Wang et al., 2017a). Tea, as an aluminium (Al) 446 cumulating crop, is able to cumulate Al in leaves (Li et al., 2016). Soil acidification in the 447 tea-planting course was due to the substantial tea litterfall into the soil annually via trimmed 448 branches and leaves (Li et al., 2016). At the same time, the rhizosphere deposition of massive 449 organic acids (i.e., malate, lemon acid, and oxalate acid) around the tea roots could provoke 450 localized acidification (Xue et al., 2006). In addition, for increasing the output of tea, tea 451 plantations needed to apply N fertilizers (i.e., urea and NH4⁺-N), thus leading to soil acidification 452 by the NH_4^+ nitration (Yang et al., 2018). 453

Across the 4 tea-planting ages, the contents of soil Fe^{2+} and Mn^{2+} were higher in the topsoil than the subsoil (Figures 7 and 8), primarily owing to the usage of swine manure and the inputs of tea litterfall and roots in the topsoil (Miner et al., 2018). Nevertheless, the contents of soil Ca²⁺ and Mg²⁺ showed an opposite trend as the soil depth increased (Figures 5 and 6), because soil Ca²⁺ and Mg²⁺ were easy to move from topsoil to subsoil in the manner of leaching (Hansen et al., 2017).

460 4.5. Stoichiometric ratios of soil alkali cations and micronutrients

461 Tea-planting age exerted a remarkable influence on the Ca/Mg and Fe/Mn ratios at the 0-40 462 cm soil depth, not the 40-60 cm soil depth (Tables 5 and 6). To be specific, a notable decline in the Ca/Mg ratio was found at the 0-40 cm soil depth over time (Table 5). From 8 to 43 years of 463 tea planting, the contents of Ca^{2+} and Mg^{2+} at the 0-40 cm soil depth decreased by 10%-38% and 464 5%-24%, separately (Figures 5 and 6), which revealed that the role of tea-planting age in the 465 content of soil Ca^{2+} was greater than that of soil Mg²⁺. Lu et al. (2014) suggested that the 466 selective losses of soil exchangeable alkali cations ($Ca^{2+} > Mg^{2+}$) could lead to the 467 468 disequilibrium of soil metal ions in forest ecosystems. Similarly, in this study, the preferential





loss of soil Ca^{2+} relative to Mg^{2+} was the prime cause of the notable decline in the soil Ca/Mg 469 ratio in the tea-planting course. The depletion of soil exchangeable alkali cations (especially the 470 Ca^{2+}) could lead to the decrease in soil buffering capacity and soil acidification (Hansen et al., 471 2017). Thus, the Ca/Mg ratio at the 0-40 cm soil depth was positively related (P < 0.05) to soil 472 473 pH across the 4 tea-planting ages (Figure 9). Soil acidification accelerated the mineralization and desorption of soil available micronutrients from soil OMs and minerals (Wang et al., 2017a), 474 conducive to the significant increases in Fe^{2+} and Mn^{2+} contents at the 0-40 cm soil depth, 475 especially the Fe²⁺ (Figures 7 and 8). In a chronological sequence of tea plantations, the negative 476 relationship (P < 0.05) of soil Fe/Mn ratio with soil pH in different soil depths (Figure 9) 477 indicated more cumulation of soil Fe^{2+} relative to Mn^{2+} over time (Table 6). Moreover, the 478 change of soil Fe/Mn ratio was also triggered by the antagonistic relationship between soil Fe²⁺ 479 and Mn^{2+} during the process of tea plant uptake (Wang et al., 2017a). Tian et al. (2016) 480 discovered that soil acidification could reduce Fe^{2+} absorption and enhance Mn^{2+} uptake by 481 various plant species, thereby causing the increase in soil Fe/Mn ratio and threatening plant 482 productivity. 483

484 **5. Conclusions**

485 Herein, soil OC, TN, and TP contents as well as C/N, C/P, and N/P ratios decreased as the soil depth increased. Moreover, soil Ca²⁺ and Mg²⁺ contents were lower in the topsoil than the 486 487 subsoil, whereas soil Fe^{2+} and Mn^{2+} contents showed an opposite trend, and no differences were 488 detected amongst different soil depths in soil Ca/Mg and Fe/Mn ratios. Tea-planting age could 489 influence the variations in soil OC and nutrient contents and their stoichiometric ratios, but such 490 effects were more obvious at the 0-40 cm soil depth in contrast to the 40-60 cm soil depth, thus supporting our hypothesis. At the 0-40 cm soil depth, continuous planting of tea was favorable to 491 the cumulation of soil OC, TN, Fe^{2+} , and Mn^{2+} , whereas soil Ca^{2+} and Mg^{2+} were susceptible to 492 leaching losses over time. Compared with other tea-planting regions in China, soil C/N ratio is 493 494 higher in this tea-planting region, whereas soil C/P and N/P ratios are much lower, indicating





- 495 that soil OC and TN contents in the present study were lower, especially the TN. Therefore, an appropriate increase in the amount of N fertilizer should be applied in this tea-planting region. In 496 the tea-planting course, the losses of soil Ca²⁺ and Mg²⁺, especially the Ca²⁺ (as indicated by the 497 decrease in soil Ca/Mg ratio), could lead to the soil acidification. Soil acidification could reduce 498 Fe²⁺ absorption and enhance Mn²⁺ uptake by tea plant (as indicated by the increase in soil Fe/Mn 499 ratio), thereby causing the aggravation of Fe^{2+} insufficiency and the emergence of Mn^{2+} toxicity 500 501 to tea plant. Overall, the present study improved the understanding of soil OC and nutrient dynamics in tea plantation ecosystems, and also provided supplementary information for soil 502 503 ecological stoichiometry in global terrestrial ecosystems.
- 504 Data availability
- 505 The data supporting the discovered information here can be presented by the relevant author
- 506 based on reasonable requests.

507 Author contribution

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

510 Competing interests

511 The authors declare no conflict of interest.

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Item	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Litterfall		$821\pm21\;B$	$974\pm34\;A$	$786\pm28\;C$	$648\pm19 \; D$	
amount (g m ⁻²)						
Litterfall		$14.23\pm1.61\ C$	$12.68\pm1.26\ C$	$17.32\pm2.24\ B$	$21.37\pm3.11\;A$	
C/N ratio						
Soil bulk	0-10 cm	$1.28\pm0.02 \ Ab$	$1.20\pm0.02\;Bc$	$1.26\pm0.01\;Ad$	$1.31\pm0.04\ Ab$	
density (g cm ⁻³) 10-20 cm	$1.30\pm0.03\ Aab$	$1.22\pm0.03~Bc$	$1.30\pm0.03~Ac$	$1.29\pm0.02~Ab$	
	20-40 cm	$1.32\pm0.04\;Aab$	$1.31\pm0.01\;Ab$	$1.34\pm0.01 \ Ab$	$1.33\pm0.04 \ Ab$	
	40-60 cm	$1.36\pm0.01\;Aa$	$1.37\pm0.02\;Aa$	$1.39\pm0.02\;Aa$	$1.38\pm0.03~Aa$	
Soil clay (%)	0-10 cm	$34.69\pm3.21~Aa$	$35.91\pm2.77~\text{Aa}$	$33.12\pm2.46~\text{Aa}$	$35.08\pm2.41~\mathrm{Aa}$	
	10-20 cm	$34.88\pm2.08\;Aa$	$32.59\pm3.02\;\text{Aa}$	$34.92\pm3.67~Aa$	$32.35\pm2.68~Aa$	
	20-40 cm	$35.26\pm1.45~\text{Aa}$	$34.57\pm4.12\;\text{Aa}$	$34.51\pm3.21~\text{Aa}$	$34.29\pm3.54~\text{Aa}$	
	40-60 cm	$34.78\pm3.66~Aa$	$36.89\pm2.98\;Aa$	$33.68 \pm 1.91 \ Aa$	35.81 ± 3.69 Aa	
Soil pH	0-10 cm	$4.57\pm0.02\;Aa$	$4.49\pm0.01\;Ba$	$4.31\pm0.03\ Ca$	$4.15\pm0.02 \ Da$	
	10-20 cm	$4.55\pm0.03\;\mathrm{Aa}$	$4.50\pm0.01\;Aa$	$4.33\pm0.02\;Ba$	$4.17\pm0.02\ Ca$	
	20-40 cm	$4.60\pm0.04\;Aa$	$4.53\pm0.02\;Ba$	$4.34\pm0.04\ Ca$	$4.19\pm0.03 \; Da$	
	40-60 cm	$4.58\pm0.02\;Aa$	$4.54\pm0.03~Aa$	$4.32\pm0.01\;Ba$	$4.21\pm0.01~Ca$	

713 Table 1 Physical-chemical properties of litterfall and soil in different aged tea plantations.

714 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

715 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

716 (P < 0.05) among different soil depths.





Sample	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Bulk soil	0-10 cm	$22.21\pm0.12~\text{Aa}$	$21.72\pm0.11~\text{Aa}$	$22.06\pm0.06~Aa$	$21.99\pm0.07\;Aa$	
	10-20 cm	$21.73\pm0.08\;\text{Aa}$	$21.98\pm0.08\;Aa$	$21.10\pm0.13\;Aa$	$21.47\pm0.11\;Aa$	
	20-40 cm	$18.86\pm0.13\;ABb$	$18.07\pm0.09\;ABb$	$17.52\pm0.06\ Bc$	$19.38\pm0.08\;Ab$	
	40-60 cm	$18.48\pm0.04\;Ab$	$18.33\pm0.09\;Ab$	$19.40\pm0.04\;Ab$	$18.92\pm0.05\;Ab$	
	Mean	20.32	20.03	20.02	20.44	
> 2 mm	0-10 cm	$21.11\pm0.13\ Bab$	$22.19\pm0.08\;ABa$	$23.04\pm0.10\;\text{Aa}$	$22.24\pm0.11~\mathrm{ABa}$	
aggregates	10-20 cm	$22.39\pm0.04\;\text{Aa}$	$22.97\pm0.11~\text{Aa}$	$21.92\pm0.07\;Ab$	$21.83\pm0.03~Aab$	
	20-40 cm	$19.71\pm0.12 \ Ab$	$20.21\pm0.04\;Ab$	$19.51\pm0.14~Ac$	$20.40\pm0.04\;Ab$	
	40-60 cm	$20.74\pm0.07\;Aab$	$19.61\pm0.13\;Ab$	$19.67\pm0.08\;Ac$	$20.51\pm0.09\;Ab$	
	Mean	20.99	21.24	21.03	21.24	
1-2 mm	0-10 cm	$22.53\pm0.07~\text{Aa}$	$21.00\pm0.12\;\text{Aa}$	$21.31\pm0.08\;Aa$	$22.51\pm0.07~Aa$	
aggregates	10-20 cm	$22.09\pm0.04\;\text{Aa}$	$22.72\pm0.09\;Aa$	$22.69\pm0.07\;Aa$	$22.55\pm0.08~Aa$	
	20-40 cm	$19.05\pm0.05\;Ab$	$18.48\pm0.11\;Ab$	$19.29\pm0.04\;Ab$	$18.71\pm0.10\;Ab$	
	40-60 cm	$17.47\pm0.06\;Ac$	$17.69\pm0.10 \text{ Ab}$	$16.94\pm0.05\;Ac$	$17.27\pm0.06\;Ab$	
	Mean	20.29	19.97	20.06	20.26	
0.25-1 mm	0-10 cm	$22.05\pm0.08\;\text{Aa}$	$21.70\pm0.11~\text{Aa}$	$21.60\pm0.09~Aa$	$21.61\pm0.04\;Aa$	
aggregates	10-20 cm	$21.63\pm0.09~\text{Aa}$	$22.56\pm0.07\;\text{Aa}$	$21.90\pm0.04\;Aa$	$21.89\pm0.10\;\text{Aa}$	
	20-40 cm	$18.95\pm0.12 \ Ab$	$18.27\pm0.04\;Ab$	$19.01\pm0.07\;Ab$	$18.03\pm0.12\;Ab$	
	40-60 cm	$17.56\pm0.13~Ab$	$17.08\pm0.06\;Ab$	$17.48\pm0.06\;Ac$	$17.22\pm0.05\;Ab$	
	Mean	20.05	19.90	20.00	19.69	
< 0.25 mm	0-10 cm	$21.89\pm0.08\;Aa$	$21.30\pm0.07\;Aa$	$20.81\pm0.11~Aa$	$21.93\pm0.09~Aa$	
aggregates	10-20 cm	$21.51\pm0.08\;ABa$	$21.12\pm0.03\;ABa$	$20.27\pm0.07\ Ba$	$22.83\pm0.06~Aa$	
	20-40 cm	$17.78\pm0.02\;Ab$	$18.45\pm0.13\;Ab$	$17.46\pm0.06\;Ab$	$17.56\pm0.04\;Ab$	
	40-60 cm	$16.52\pm0.05\;Ab$	$16.92\pm0.07\;Ac$	$17.03\pm0.10\;Ab$	$16.41\pm0.08\;Ab$	
	Mean	19.42	19.45	18.89	19.68	

718 **Table 2** Effects of tea-planting age and soil depth on the soil C/N ratio in various sized aggregates.

719 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

720 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

721 (P < 0.05) among different soil depths.





Sample	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Bulk soil	0-10 cm	$30.93 \pm 1.02 \text{ Ba}$	$31.63\pm1.45\ Ba$	$35.94 \pm 1.41 \text{ Aa}$	$34.50\pm2.89~Aa$	
	10-20 cm	$23.60\pm0.85\;Bb$	$24.18\pm0.84\ Bb$	$26.28\pm1.21 \ Ab$	$26.56\pm1.47\;Ab$	
	20-40 cm	$19.92\pm0.48\;Cc$	$20.13\pm0.71\ BCc$	$21.15\pm0.89\ Bc$	$24.41\pm0.98\;Ab$	
	40-60 cm	$17.41\pm0.69\;Ac$	$17.21\pm0.58\;Ad$	$18.12\pm0.24\;Ad$	$17.59\pm1.22\;Ac$	
	Mean	22.97	23.29	25.37	25.76	
> 2 mm	0-10 cm	$32.04\pm1.04\ Ca$	$32.14\pm0.98\ Ca$	$35.54\pm1.07\ Ba$	$37.07\pm0.38\;Aa$	
aggregates	10-20 cm	$25.93\pm0.84\ Cb$	$24.41\pm1.07\ Cb$	$27.21\pm0.37\ Bb$	$29.23\pm0.98\;Ab$	
	20-40 cm	$22.13\pm0.97~Bc$	$21.43\pm1.12\ Bc$	$26.33\pm0.86\;Ab$	$27.29\pm1.24\;Ab$	
	40-60 cm	$22.40\pm2.02~Ac$	$22.88\pm0.87~Abc$	$22.67\pm1.24~Ac$	$21.63\pm1.56~Ac$	
	Mean	25.62	25.21	27.94	28.81	
1-2 mm	0-10 cm	$29.52\pm1.01~\text{Da}$	$31.48\pm0.47\ Ca$	$33.54\pm0.97\ Ba$	$36.53\pm0.81~\text{Aa}$	
aggregates	10-20 cm	$24.76\pm0.38\ Bb$	$26.58\pm0.58\;Ab$	$27.55\pm0.47\;Ab$	$26.68\pm0.97\;Ab$	
	20-40 cm	$20.68\pm1.14\ Bc$	$20.51\pm1.48\ Bc$	$21.95\pm1.05\ Bc$	$26.07\pm0.78\;Ab$	
	40-60 cm	$16.78\pm0.87\;Bd$	$18.04\pm0.98\;Ac$	$16.63\pm1.24\ Bd$	$17.55\pm1.05~\text{ABc}$	
	Mean	22.93	24.15	24.92	26.71	
0.25-1 mm	0-10 cm	31.44 ± 1.27 Aa	$30.46\pm0.78~Aa$	30.91 ± 1.08 Aa	$30.62\pm0.98\;Aa$	
aggregates	10-20 cm	$23.60\pm0.27\;Bb$	$25.38\pm0.38~ABb$	$24.41 \pm 1.14 \; ABb$	$26.41\pm0.57\;Ab$	
	20-40 cm	$18.96\pm1.47~\mathrm{Cc}$	$21.62\pm0.45\ Bc$	$19.78\pm0.87\ Cc$	$25.60\pm1.02\;Ab$	
	40-60 cm	$18.18\pm0.87~Ac$	$17.40\pm0.38\;Ad$	$17.43\pm0.91\;Ac$	$17.92\pm1.34~Ac$	
	Mean	23.05	23.71	23.13	25.14	
< 0.25 mm	0-10 cm	$28.81 \pm 1.01 \text{ Ba}$	$30.60\pm1.07~Aa$	$29.78\pm0.87~ABa$	$31.00\pm0.38~Aa$	
aggregates	10-20 cm	$19.39\pm1.17\ Cb$	$21.86\pm0.68\ Bb$	$22.36\pm0.78\;ABb$	$23.19\pm0.98\;Ab$	
	20-40 cm	$15.22\pm0.87~Bc$	17.11 ± 1.14 Ac	$17.46\pm0.94\;Ac$	$18.50\pm0.75\;Ac$	
	40-60 cm	$13.73\pm0.74~Ac$	$14.50\pm0.74\;Ad$	$13.44\pm1.00\;Ad$	$14.02\pm0.91\;Ad$	
	Mean	19.29	21.02	20.76	21.68	

723 Table 3 Effects of tea-planting age and soil depth on the soil C/P ratio in various sized aggregates.

724 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

725 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

726 (P < 0.05) among different soil depths.





Sample	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Bulk soil	0-10 cm	$1.39\pm0.04\ Ba$	$1.46\pm0.02\;Ba$	$1.63\pm0.04~Aa$	1.57 ± 0.02 ABa	
	10-20 cm	$1.09\pm0.01\;Bb$	$1.10\pm0.04\;Bb$	$1.25\pm0.08\;Ab$	$1.24\pm0.03~Ab$	
	20-40 cm	$1.06\pm0.02\;Bb$	$1.11\pm0.07\;ABb$	$1.21\pm0.02\;Ab$	$1.26\pm0.02~Ab$	
	40-60 cm	$0.94\pm0.06\;Ab$	$0.99\pm0.06\;Ab$	$0.93\pm0.01\;Ac$	$0.98\pm0.04\ Ac$	
	Mean	1.12	1.17	1.25	1.26	
> 2 mm	0-10 cm	$1.52\pm0.05\;ABa$	$1.45\pm0.01\;Ba$	$1.54\pm0.04\;ABa$	$1.67\pm0.02~Aa$	
aggregates	10-20 cm	$1.16\pm0.02\;ABb$	$1.06\pm0.02~Bc$	$1.24\pm0.03\ Ab$	$1.34\pm0.01~Ab$	
	20-40 cm	$1.11\pm0.03\;Bb$	$1.24\pm0.02\ ABb$	$1.25\pm0.05\;ABb$	$1.38\pm0.04~Ab$	
	40-60 cm	$1.08\pm0.04\;Ab$	$1.07\pm0.03~Ac$	$1.00\pm0.04\;Ac$	$1.06\pm0.03~Ac$	
	Mean	1.22	1.20	1.26	1.36	
1-2 mm	0-10 cm	$1.31\pm0.03\;Ba$	$1.50\pm0.01\;Aa$	$1.57\pm0.02\;Aa$	$1.62\pm0.04~Aa$	
aggregates	10-20 cm	$1.12\pm0.04\;Ab$	$1.17\pm0.02~Ab$	$1.21\pm0.03\ Ab$	$1.18\pm0.05~Abc$	
	20-40 cm	$0.98\pm0.01\;Bc$	$1.05\pm0.01\;Bb$	$1.14\pm0.04\;ABbc$	$1.26\pm0.02~Ab$	
	40-60 cm	$0.96\pm0.06\;Ac$	$1.02\pm0.03\ Ab$	$0.98\pm0.06\;Ac$	$1.03\pm0.03~Ac$	
	Mean	1.09	1.19	1.23	1.27	
0.25-1 mm	0-10 cm	$1.43\pm0.02\;Aa$	$1.40\pm0.04\;Aa$	$1.43\pm0.02\;\text{Aa}$	$1.42\pm0.01~Aa$	
aggregates	10-20 cm	$1.09\pm0.02\;Bb$	$1.13\pm0.01\;ABb$	$1.11\pm0.01\;ABb$	$1.21\pm0.01~Ab$	
	20-40 cm	$0.96\pm0.06\ Bb$	$1.02\pm0.02\;Bb$	$1.13\pm0.04\;ABb$	$1.29\pm0.03~Ab$	
	40-60 cm	$0.98\pm0.04\;Ab$	$1.02\pm0.03\ Ab$	$0.94\pm0.02\;Ac$	$1.04\pm0.02~Ac$	
	Mean	1.11	1.14	1.15	1.24	
< 0.25 mm	0-10 cm	$1.32\pm0.04\ Ba$	$1.44\pm0.03~Aa$	$1.43\pm0.02\;\text{Aa}$	$1.41\pm0.04~Aa$	
aggregates	10-20 cm	$0.90\pm0.01\;Bb$	$1.04\pm0.02~ABb$	$1.10\pm0.01 \ Ab$	$1.02\pm0.03~ABb$	
	20-40 cm	$0.91\pm0.04\ Bb$	$0.93\pm0.02 \; Bbc$	$1.00\pm0.03\ Ab$	$1.05\pm0.02~Ab$	
	40-60 cm	$0.88\pm0.03\ Ab$	$0.86\pm0.04\;Ac$	$0.84\pm0.02\;Ac$	$0.85\pm0.04~Ac$	
	Mean	1.00	1.06	1.09	1.08	

728 Table 4 Effects of tea-planting age and soil depth on the soil N/P ratio in various sized aggregates.

729 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

730 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

731 (P < 0.05) among different soil depths.





Sample	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Bulk soil	0-10 cm	$1.94\pm0.12\;\text{Aa}$	$1.91\pm0.05~Aa$	$1.86\pm0.12\;ABa$	$1.82\pm0.07\;Bab$	
	10-20 cm	$1.93\pm0.08\;Aa$	$1.87\pm0.07\;ABa$	$1.87\pm0.08\;ABa$	$1.84\pm0.12\;Bab$	
	20-40 cm	$1.96\pm0.14~\mathrm{Aa}$	$1.90\pm0.14\ Ba$	$1.88\pm0.04\;Ba$	$1.80\pm0.14\;Cb$	
	40-60 cm	$1.91\pm0.11~Aa$	$1.88\pm0.09\;Aa$	$1.90\pm0.07\;Aa$	$1.89\pm0.06~Aa$	
	Mean	1.94	1.89	1.88	1.84	
> 2 mm	0-10 cm	$1.96\pm0.17~Aa$	$1.89\pm0.08\;ABa$	$1.88\pm0.16~ABa$	$1.83\pm0.04\;Bab$	
aggregates	10-20 cm	$1.92\pm0.14\;Aa$	$1.87\pm0.18\ Ba$	$1.86\pm0.06\ Ba$	$1.88\pm0.07\;Bab$	
	20-40 cm	$1.95\pm0.08\;Aa$	$1.88\pm0.06\;ABa$	$1.89\pm0.07\;ABa$	$1.81\pm0.12\;Bb$	
	40-60 cm	$1.90\pm0.11~\mathrm{Aa}$	$1.91\pm0.09\;Aa$	$1.89\pm0.11~Aa$	$1.90\pm0.13~Aa$	
	Mean	1.93	1.89	1.88	1.86	
1-2 mm	0-10 cm	$1.94\pm0.20\;Aa$	$1.86\pm0.17\ Ba$	$1.85\pm0.08\;Ba$	$1.84\pm0.14\ Ba$	
aggregates	10-20 cm	$1.95\pm0.15\;\text{Aa}$	$1.90\pm0.16\ Ba$	$1.88\pm0.08\;Ba$	$1.87\pm0.10\;Ba$	
	20-40 cm	$1.92\pm0.07\;Aa$	$1.84\pm0.05\;Ba$	$1.86\pm0.12\;Ba$	$1.85\pm0.08\;Ba$	
	40-60 cm	$1.90\pm0.06\;Aa$	$1.89\pm0.06\;Aa$	$1.88\pm0.03~Aa$	$1.89\pm0.09~Aa$	
	Mean	1.93	1.87	1.87	1.86	
0.25-1 mm	0-10 cm	$1.95\pm0.14\;\mathrm{Aa}$	$1.88\pm0.17\ Ba$	$1.87\pm0.06\ Ba$	$1.81\pm0.07\ Cb$	
aggregates	10-20 cm	$1.93\pm0.11~\text{Aa}$	$1.90\pm0.08\;ABa$	$1.86\pm0.07\;ABa$	$1.84\pm0.04\;Bb$	
	20-40 cm	$1.94\pm0.12\;\mathrm{Aa}$	$1.86\pm0.10\;ABa$	$1.87\pm0.03~ABa$	$1.82\pm0.06\;Bb$	
	40-60 cm	$1.92\pm0.07\;Aa$	$1.90\pm0.06\;Aa$	$1.91\pm0.05\;Aa$	$1.89\pm0.09~Aa$	
	Mean	1.94	1.89	1.88	1.84	
< 0.25 mm	0-10 cm	$1.92\pm0.06\;Aa$	$1.85\pm0.04\ Ba$	$1.83\pm0.08\;Ba$	$1.82\pm0.12\;Bb$	
aggregates	10-20 cm	$1.94\pm0.17~Aa$	$1.86\pm0.12\ Ba$	$1.87\pm0.08\;Ba$	$1.85\pm0.07\;Bab$	
	20-40 cm	$1.91\pm0.08\;Aa$	$1.85\pm0.07\ Ba$	$1.84\pm0.07\;Ba$	$1.86\pm0.04\;Bab$	
	40-60 cm	$1.92\pm0.11~\mathrm{Aa}$	$1.90\pm0.03~Aa$	$1.89\pm0.06\;Aa$	$1.91\pm0.13~Aa$	
	Mean	1.92	1.87	1.86	1.86	

733 Table 5 Effects of tea-planting age and soil depth on the soil Ca/Mg ratio in various sized aggregates.

734 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

735 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

736 (P < 0.05) among different soil depths.





Sample	Soil depth	Tea-planting age				
		8 years	17 years	25 years	43 years	
Bulk soil	0-10 cm	$0.78\pm0.04\ Ba$	$0.82\pm0.01\;ABa$	$0.81\pm0.04\;ABa$	$0.84\pm0.02~Aab$	
	10-20 cm	$0.76\pm0.02\;Ba$	$0.81\pm0.04\;Aa$	$0.82\pm0.01\;Aa$	$0.83\pm0.04 \ Aab$	
	20-40 cm	$0.75\pm0.03\ Ca$	$0.80\pm0.03\;Ba$	$0.81\pm0.05\ Ba$	$0.85\pm0.02~Aa$	
	40-60 cm	$0.78\pm0.05\;Aa$	$0.80\pm0.05\;Aa$	$0.79\pm0.03\;Aa$	$0.81\pm0.03\ Ab$	
	Mean	0.77	0.81	0.81	0.83	
> 2 mm	0-10 cm	$0.77\pm0.02\;Ba$	$0.81\pm0.01\;ABa$	$0.81\pm0.03\;ABa$	$0.83\pm0.02 \ Aab$	
aggregates	10-20 cm	$0.75\pm0.04\ Ca$	$0.80\pm0.04\ Ba$	$0.79\pm0.01\;Ba$	$0.84\pm0.02~Aa$	
	20-40 cm	$0.77\pm0.01\;Ba$	$0.78\pm0.01\;Ba$	$0.82\pm0.02\;Aa$	$0.82\pm0.03~Aab$	
	40-60 cm	$0.78\pm0.03\;Aa$	$0.81\pm0.02\;Aa$	$0.79\pm0.04\;Aa$	$0.80\pm0.01~Ab$	
	Mean	0.77	0.80	0.80	0.82	
1-2 mm	0-10 cm	$0.76\pm0.01\;Cab$	$0.82\pm0.02\;ABa$	$0.80\pm0.01\;Ba$	$0.85\pm0.03~Aa$	
aggregates	10-20 cm	$0.75\pm0.02\;Bb$	$0.81\pm0.01\;Aa$	$0.82\pm0.02\;Aa$	$0.84\pm0.01~Aa$	
	20-40 cm	$0.78\pm0.02\;Bab$	$0.80\pm0.03\;ABa$	$0.82\pm0.01\;ABa$	$0.85\pm0.04\;\text{Aa}$	
	40-60 cm	$0.79\pm0.01\;Aa$	$0.82\pm0.01\;Aa$	$0.81\pm0.03\;Aa$	$0.82\pm0.02~Aa$	
	Mean	0.77	0.81	0.81	0.84	
0.25-1 mm	0-10 cm	$0.78\pm0.03\;Bab$	$0.82\pm0.02\;ABa$	$0.82\pm0.03\;ABa$	$0.84\pm0.01~Aab$	
aggregates	10-20 cm	$0.77\pm0.01\;Bab$	$0.81\pm0.01\;ABab$	$0.82\pm0.02\;ABa$	$0.85\pm0.02~Aa$	
	20-40 cm	$0.75\pm0.02\;Bb$	$0.78\pm0.04\;ABb$	$0.81\pm0.05\;ABa$	$0.84\pm0.01~Aab$	
	40-60 cm	$0.80\pm0.04\;Aa$	$0.79\pm0.02 \ Aab$	$0.80\pm0.02\;Aa$	$0.81\pm0.03\ Ab$	
	Mean	0.78	0.80	0.81	0.84	
< 0.25 mm	0-10 cm	$0.79\pm0.02\;Ba$	$0.81\pm0.03\;Ba$	$0.81\pm0.03\;Ba$	$0.85\pm0.01~\text{Aa}$	
aggregates	10-20 cm	$0.77\pm0.01\;Ba$	$0.82\pm0.02\;Aa$	$0.80\pm0.01\;ABa$	$0.83\pm0.03~\text{Aa}$	
	20-40 cm	$0.78\pm0.03\;Ba$	$0.80\pm0.02\;ABa$	$0.82\pm0.02\;ABa$	$0.83\pm0.04~\mathrm{Aa}$	
	40-60 cm	$0.80\pm0.01\;Aa$	$0.81\pm0.03~Aa$	$0.80\pm0.04\;Aa$	$0.82\pm0.02~Aa$	
	Mean	0.79	0.81	0.81	0.83	

738 Table 6 Effects of tea-planting age and soil depth on the soil Fe/Mn ratio in various sized aggregates.

739 Data represent the average of 5 replicates ± standard deviations. Different capital letters indicate significant

740 differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences

741 (P < 0.05) among different soil depths.





Figure 1 Effects of tea-planting age and soil depth on the composition and stability of soil aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 2 Effects of tea-planting age and soil depth on the soil organic C content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 3 Effects of tea-planting age and soil depth on the soil total N content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 4 Effects of tea-planting age and soil depth on the soil total P content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 5 Effects of tea-planting age and soil depth on the soil exchangeable Ca^{2+} content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 6 Effects of tea-planting age and soil depth on the soil exchangeable Mg^{2+} content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 7 Effects of tea-planting age and soil depth on the soil available Fe^{2+} content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 8 Effects of tea-planting age and soil depth on the soil available Mn^{2+} content in various sized aggregates. Data represent the average of 5 replicates and error bars represent the standard deviations. Different capital letters indicate significant differences (P < 0.05) among different tea-planting ages. Different lower case letters indicate significant differences (P < 0.05) among different soil depths.

Figure 9 Relationships of soil Ca/Mg and Fe/Mn ratios with soil pH in different soil layers. ** indicates significant differences at P < 0.01.





















































