

1 **Aggregate stability and size distribution of red soils under different land uses**  
2 **integrally regulated by soil organic matter, and iron and aluminum oxides**

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16 **Abstract**

17 The stability and size distribution of soil aggregates is profoundly affected by land  
18 use, but the influencing mechanisms of land use are not clear. A study was carried  
19 out to investigate and attempted to interpret the effects of land use on soil  
20 aggregates from types of land use and soil properties in soil samples and size  
21 fractions of soil aggregates. Soil samples were taken from 9 sites under paddy,  
22 forest, and upland in southern China. The wet-sieving method was used to obtain  
23 6 size fractions of soil aggregates: >5, 5~2, 2~1, 1~0.5, 0.5~0.25, and <0.25 mm.  
24 The stability and size distribution of soil aggregates was measured as mean  
25 weight diameter (MWD), the percentage of water-stable aggregate (WSA) and the  
26 percentage of each size fraction (PSA). The quantities of soil organic carbon  
27 (SOC), humic substances, dithionite-citrate-bicarbonate (DCB) and oxalate  
28 extractable iron (Fe) and aluminum (Al) oxides were also measured. The results  
29 showed that types of land use solely explained 66.6% variation of soil  
30 aggregates; SOC, DCB-extractable Fe and Al oxides, and oxalate-extractable Al  
31 oxide caused 84.3% variation, in which SOC contributed 29.0%, Fe and Al oxides  
32 contributed 33.8%, and their interactions contributed 21.4%. The multiple linear  
33 regression and partial correlation analysis showed that soil organic matter and Fe  
34 and Al oxides had significant effects but played different roles on the stability and  
35 size distribution of soil aggregates. The study suggests that land use affects the  
36 stability and size distribution of soil aggregates through the integration of soil  
37 organic matter and Fe and Al oxides.

38

39 **Keywords:** soil aggregates; stability; size fraction; land use; soil organic matter;

40 iron and aluminum oxides

41

## 42 **1. Introduction**

43           Soil structure has important influences on edaphic conditions and the  
44 environment. The structure is often measured by the stability of soil  
45 aggregates(Six et al., 2000; Bronick and Lal, 2005). Soil aggregation sustains soil  
46 fertility because it reduces erosionand mediates soil aerationas well as water  
47 infiltration and retention. Furthermore, soil aggregation protects soil organic  
48 matter(SOM) from mineralizing because it physically reduces the accessibilityof  
49 organic compounds for microorganisms, extracellular enzymes,and oxygen  
50 (Oades, 1984; Six et al., 2002a; von Lützow et al., 2006; Spohn and Giani, 2010).  
51 The size distribution and stability ofsoil aggregatesareunder the control of  
52 various mechanisms. Generally, the size distribution and stability of soil  
53 aggregates positively correlates with the main cementing agents, such as SOM,  
54 clay minerals, multi-valent cationsand their complex in soil aggregates.According  
55 to the hierarchical theory of aggregate formation, these materials may be  
56 distributed unevenly in different size fractions of soil aggregates, andthey may  
57 have close relationships with the stability of soil aggregates(Tisdall and Oades,  
58 1982; Six et al., 2004).

59           Land use and associated management, such as crop sequencing,  
60 fertilization, soil conditioning, drainage and irrigation,arethe most important and  
61 direct ways to affect soil structure and properties,throughitsimpact on  
62 destruction forces and aggregate forming processes(Haynes et al., 1991; Besnard  
63 et al., 1996; Jastrow, 1996; John et al., 2005; Ashagrie et al., 2007; Lehrsch et al.,

64 [2012](#)). However, the extent of the impact and the associated mechanisms of land  
65 use on soil aggregates remains unclear. Studies on the stability of soil aggregates  
66 under different land use rarely focus on the effects of various soil organic  
67 components, Fe and Al oxides in different size fractions of soil aggregates, and  
68 their interactions.

69 In southern China, the red soils cover approximately 1.14 million km<sup>2</sup>.  
70 Because of intensive weathering and leaching, the soil is rich in Fe and Al oxides,  
71 and the dominant clay mineral is kaolinite ([He et al., 2004](#)). In recent years, soil  
72 structure has degraded rapidly, due to strong human intervention and high  
73 cultivating intensity ([Iqbal et al., 2009](#)), and the sustainability of the agricultural  
74 ecosystem is under serious threat.

75 Therefore, for soils that are rich in Fe and Al oxides, we hypothesize that  
76 the stability and size distribution of soil aggregates is controlled by SOM, which is  
77 heavily affected by the land use, while Fe and Al oxides and their interactions  
78 with SOM manifest another important regulating mechanism for the stability and  
79 size distribution of soil aggregates. In order to test this hypothesis, in this  
80 study, the stability and size distribution of soil aggregates of red soils in southern  
81 China were investigated from different types of land use, which were treated as a  
82 factor that represents the comprehensive effects brought about by the land use  
83 and its associated management, and the soil properties in soil samples and size  
84 fractions of soil aggregates.

85

## 86 2. Materials and Methods

### 87 2.1 Study sites and soil samplings

88 Nine sites located in Jinxian, Jiangxi Province, Changsha and Taoyuan,  
89 Hunan Province, and Xianning, Hubei Province, southern China, were chosen,  
90 taking into account the variability of soil type (Inceptisols and Ultisols), in order  
91 to compare how the land use (paddy, forest, and upland) and the associated soil  
92 properties affected the stability and size distribution of soil aggregates. All  
93 sampling sites are in the subtropics and share similar climate. Those regions are  
94 humid and have a monsoon climate, with an annual mean temperature above  
95 15°C and annual mean precipitation above 1300 mm. The paddy sites have been  
96 used for planting rice for more than ten years. The forest sites are secondary  
97 forests, with predominance of *Pinus massoniana* in Xianning (F<sub>1</sub>), *Cunninghamia*  
98 *lanceolata* in Changsha (F<sub>3</sub>), and a mixed forest in Changsha (F<sub>2</sub>). The uplands are  
99 used for grain or cash crops such as maize (*Zea mays*), rapeseed (*Brassica napus*),  
100 soybean (*Glycine max*), and sweet potato (*Ipomoea batatas*). Paddy and upland  
101 sites are managed with conventional tillage. The same type of land use shares  
102 similar agricultural practices.

103 At each site, 5 soil monoliths with diameter 10 cm were randomly taken  
104 from 0-15 cm surface between October and December 2010. All samples were  
105 transported to the laboratory in their intact form and broken into small pieces  
106 along the natural crack by hand during the air-drying process. The soils from  
107 paddies are Endoaquepts, and those from uplands and forests are Plinthudults,

108 according to Soil Survey Staff(2014). All soil developed out of middle  
109 Pleistocene(Q2) red clay. The basic soil properties, parent, land use, and soil type  
110 are shown in Table 1 and Table 2.

111

## 112 2.2 Water-stable aggregates

113 To obtain different size fractions of water-stable aggregates, for the 5  
114 samples from each site, 50 g of soil were placed in the top of a set of sieves with  
115 mesh sizes of 5 mm, 2 mm, 1 mm, 0.5 mm, and 0.25 mm from top to bottom. The  
116 sieve set was placed on the shock rack of a Yoder aggregates analyzer (Institute  
117 of Soil Science, Chinese Academy of Sciences, Nanjing, China), submerged in  
118 water, and shaken with an amplitude of 3 cm and a frequency of 30 min<sup>-1</sup> for a  
119 duration of 30 mins. The size fraction at each sieve was then washed into a beaker  
120 of known mass and then dried and weighed.

121 Mean weight diameter (MWD) was calculated as

$$\text{MWD} = \frac{\sum_{i=1}^6 d_i w_i}{w};$$

122 the percentage of water-stable aggregate (WSA) was calculated as

$$\text{WSA} = 100 \times \frac{\sum_{i=1}^5 w_i}{w};$$

123 and the percentage of each size fraction of soil aggregate (PSA) was given as

$$\text{PSA}_i = 100 \times \frac{w_i}{w};$$

124 where  $d_i$  is the average diameter of  $i^{\text{th}}$  size fraction of the aggregate,  $w_i$  is the mass  
125 of  $i^{\text{th}}$  size fraction of the aggregate, and  $w$  is the total mass of all size fractions of  
126 the aggregate.  $i=1,2,\dots,6$  represent the aggregate size >5 mm, 5~2 mm, 2~1 mm,

127 1~0.5 mm, 0.5~0.25 mm and <0.25 mm, respectively.

128

### 129 **2.3 Carbon content of soil organic components**

130 Humus (HF) in the soil sample or a size fraction of soil aggregates was  
131 extracted using the method proposed by the Institute of Soil Science, Chinese  
132 Academy of Sciences (1978). For each sample, 5g of air-dried soil were placed into  
133 a 250-mL Erlenmeyer flask. Then 100 mL 0.1 mol L<sup>-1</sup> sodium  
134 pyrophosphate-sodium hydroxide were added. The mixture was shaken for 30  
135 mins and then left to stand for 13~14h. Depending on the color depth, 2~15 mL  
136 supernatant were drawn into the digestive tract and neutralized with 0.5 mol  
137 L<sup>-1</sup>H<sub>2</sub>SO<sub>4</sub>. The supernatant was evaporated to dryness on a boiling water bath,  
138 after which the carbon content of HF was determined using the dichromate  
139 oxidization method (Bao, 2000).

140 Fifty milliliters of the previous supernatant were drawn into a 100-mL  
141 Erlenmeyer flask. The pH was adjusted to 3 or less with 0.5 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub> at  
142 60~70 °C. It was then kept at 80 °C for 30 mins with a water bath. After cooling  
143 for about 8h, it was filtered with a fine filter paper (Grade 44, Whatman). The  
144 precipitation on the filter paper was washed 3 times using 0.025 mol L<sup>-1</sup> H<sub>2</sub>SO<sub>4</sub>  
145 and then solubilized and washed into a 50-mL flask with 0.05 mol L<sup>-1</sup> NaOH at  
146 40~50 °C. Then, 10~25 mL solution were drawn into the digestive tract and  
147 neutralized with 0.5 mol L<sup>-1</sup>H<sub>2</sub>SO<sub>4</sub>. The solution was evaporated to dryness on a  
148 boiling water bath, after which the carbon content of humic acid (HA) was



149 determined using the dichromate oxidization method (Bao, 2000). The carbon  
150 content of fulvic acid (FA) was obtained by subtracting the carbon content of HA  
151 from HF (Institute of Soil Science, Chinese Academy of Sciences, 1978).

152

#### 153 **2.4 Organic carbon content in soil and size fractions**

154 The soil sample or a size fraction of soil aggregates was ground to pass  
155 through a sieve with a mesh size of 0.149 mm. For each sample, 0.3 g soil  
156 was placed in a small crucible to measure the total content of soil organic carbon  
157 (SOC) with an elemental analyzer (Vario MAX C/N, Elementar Analysensysteme  
158 GmbH, Hanau, Germany). A reference sample was added after each 15 soil  
159 samples.

160

#### 161 **2.5 Free-form Fe and Al oxides**

162 Free-form Fe and Al oxides were extracted using the  
163 dithionite-citrate-bicarbonate (DCB) method (Li, 1997). For each sample, 0.3 g  
164 air-dried soil was placed into a 50-mL centrifuge tube and then 20 mL 0.3 mol L<sup>-1</sup>  
165 sodium citrate and 2.5 mL 1 mol L<sup>-1</sup> sodium bicarbonate were added. When the  
166 temperature of the water bath reached 80 °C, 0.5 g sodium dithionite was added.  
167 The mixture was stirred constantly for 15 mins. After cooling and centrifugation,  
168 the supernatant was transferred into a 250 mL volumetric flask. The sediment in  
169 the centrifuge tube was treated with the above process again. Finally, all material  
170 in the tube was washed into the same flask with 1 mol L<sup>-1</sup> NaCl. The Fe and Al in

171 the solution were determined with an inductively coupled plasma (ICP) analyzer  
172 (Vista-MPX, Varian, Inc., Palo Alto, CA, USA).

173

## 174 **2.6 Amorphous Fe and Al oxide**

175 Amorphous Fe and Al oxide was extracted using the ammonium oxalate  
176 method(Li, 1997). For each sample, 0.5 g air-dried soil was placed into a 50-mL  
177 centrifuge tube and then 25 mL 0.2 mol L<sup>-1</sup> ammonium oxalate was added  
178 (soil:solution = 1:50). The mixture was shaken for 2h in the dark and then  
179 centrifuged immediately with rotation speed 4000 rmin<sup>-1</sup> to obtain supernatant.  
180 The supernatant was diluted 5 times and the Fe and Al content in the solution  
181 were determined with an ICP analyzer (Vista-MPX, Varian, Inc., Palo Alto, CA,  
182 USA).

183

## 184 **2.7 Statistical analysis**

185 All experimentally determined data are expressed as mean±SE (standard  
186 error of mean). For the soil properties in each site, the mean and SE that were  
187 calculated from the measurements of 5 samples ( $n = 5$ ), while for the soil  
188 properties in a specific type of land use, they were calculated from 3 averages ( $n$   
189 = 3). The differences of MWD, WSA, and PSA of soil aggregates under different  
190 types of land use were tested by one-way analysis of variance (ANOVA). If the  
191 results were statistically significant, then the post hoc multiple comparisons were  
192 performed with least significant difference (LSD).

193 Redundancy analysis (RDA) was used to explore the relationship between  
194 response variables (i.e., MWD, WSA, and PSA) and explanatory factors such  
195 as types of land use, and the contents of Fe and Al oxides, FA, HA, humus, HA/FA  
196 ratio, and SOC in soil and in various size fractions of soil aggregates. The RDA  
197 model with up to 6 explanatory variables was built through best subset searching.  
198 The multicollinearity between explanatory variables was checked with variance  
199 inflation factors (VIF), and any model containing a variable with VIF greater than  
200 10 was discarded. The Akaike information criterion (AIC) was used to select an  
201 optimal model. The validation of the RDA model and the significance of each  
202 variable on MWD and WSA were checked by permutation tests. The permutation  
203 was repeated 1999 times. The significant level was set to 0.05. The variation of  
204 response variables with respect to SOC and Fe and Al oxides were also partitioned,  
205 using the coefficient of determination of the RDA model ([Borcard et al., 1992](#)).

206 The relationship between individual response variables and soil properties  
207 in size fractions of soil aggregates was explored with multiple linear  
208 regression, based on best subset searching. The same procedures as in RDA were  
209 used to check the multicollinearity and to choose an optimal model. Partial  
210 correlation analysis was used to explore the correlation between the  
211 individual response variable and the corresponding soil properties in the optimal  
212 model.

213 All statistical analysis and plotting were carried out using the statistical  
214 and computing environment R version 3.3.0 ([R Core Team, 2016](#)). RDA, variance

215 partition, and permutation tests were provided by vegan version 2.3 (Oksanen et  
216 al., 2016), an add-on package for R.

217

## 218 **3. Results**

### 219 **3.1 Soil properties and land use**

220 The basic soil properties, such as pH, bulk density, SOC, soil total nitrogen  
221 (STN), C/N ratio and texture of the 9 sites, are presented in Table 2. Although the  
222 soils were developed from the same parent material, the properties varied  
223 significantly among the different types of land use. Paddy sites had the lowest  
224 bulk density ( $1.09 \pm 0.07 \text{ g cm}^{-3}$ ), highest pH ( $5.0 \pm 0.14$ ), and highest STN  
225 ( $1.56 \pm 0.04 \text{ gN kg}^{-1}$ ), which were significantly different from those in upland or  
226 forest sites. The contents of silt, sand, and clay were not significantly different  
227 among the types of land use.

228 Other soil properties, such as SOC and its components, revealed significant  
229 differences among different types of land use (Table 3). The differences between  
230 paddy and forest were not significant, but in both types of land use, SOC, HF, and  
231 FA were significantly higher than those in uplands ( $p < 0.05$ ), while HA and HA/FA  
232 ratios were not significantly different. The Fe and Al oxides did not vary  
233 significantly between any types of land use.

234

### 235 **3.2 Stability and size distribution of soil aggregates**

236 Under different types of land use, the average MWD and WSA appeared in

237 order as paddy>forest >upland (Fig. 1). Paddy has the highest MWD, followed by  
238 forest. Upland has the lowest MWD, with half of this region being under forest  
239 land use. PSA<sub>1</sub> in paddy was highest, while the rest of the size fractions  
240 were smaller. PSA<sub>6</sub> was highest in upland. The differences among MWD, WSA, PSA<sub>1</sub>,  
241 PSA<sub>4</sub>, and PSA<sub>6</sub> under different types of land use were significant (Fig. 1). MWD,  
242 WSA, and PSA<sub>6</sub> showed similar patterns and no significant  
243 differences existed between paddy and forest, but significant differences were  
244 found among upland and the other 2 types. While PSA<sub>1</sub> and PSA<sub>4</sub> showed  
245 similar patterns of significant differences between paddy and other types of land  
246 use, there was no significant difference between forest and upland. PSA<sub>2</sub>, PSA<sub>3</sub>,  
247 and PSA<sub>5</sub> were not significantly different among the 3 types of land use.

248         The effects of land use type on the stability and size distribution of soil  
249 aggregates (MWD, WSA, and PSA) were explored by the RDA. The effects of land  
250 use accounted for 66.6% of variation of the stability and size distribution of soil  
251 aggregates. The first axis of RDA accounted for 45.3%, mainly contributed by  
252 upland and paddy, while the second axis of RDA accounted for 21.3%, which  
253 was mainly contributed by forest (Fig. 2). MWD, WSA and PSA presented a clear  
254 pattern, where MWD, WSA and PSA<sub>1</sub> were grouped and closely related with each  
255 other. Similarly, the middle-sized fractions (PSA<sub>2</sub>, PSA<sub>3</sub>, and PSA<sub>4</sub>), and  
256 small-sized fractions of soil aggregates (PSA<sub>5</sub>, and PSA<sub>6</sub>) were grouped separately  
257 (Fig. 2). Paddy, forest and upland had a positive contribution to group 3,  
258 respectively. Permutation tests showed that the land use types had significant

259 effects( $p<0.05$ ) on the stability and size distribution of soil aggregates.

260         The relationships between soil properties and the stability and size  
261 distribution of soil aggregates were also explored by RDA, and showed that the  
262 DCB-extractable Fe and Al oxides, oxalate-extractable Al oxide, and SOC  
263 could explain 84.3% of the variation in the stability and size distribution of soil  
264 aggregates under different types of land use. The first RDA axis accounted for  
265 57.4%, mainly contributed by SOC, while the second RDA axis accounted for  
266 25.0%, mainly contributed by the oxalate-extractable Al oxide (Fig. 3). The  
267 DCB-extractable Fe and Al oxides contributed equally to both ordination axes. All  
268 soil properties in the RDA model had significant positive correlations with MWD,  
269 WSA, and PSA<sub>1</sub>, but negative with PSA<sub>5</sub> and PSA<sub>6</sub>. For PSA<sub>2</sub>, PSA<sub>3</sub> and PSA<sub>4</sub>, Fe and  
270 Al oxides were positive contributors, while SOC presented a negative contribution  
271 (Fig. 3). Permutation tests showed that all soil properties significantly affected soil  
272 aggregates ( $p<0.05$ ).

273         The variation of the stability and size distribution of soil aggregates with  
274 respect to SOC and Fe and Al oxides was partitioned (Fig. 4). The contribution of  
275 SOC was 29.0%, while the contribution of Al and Fe oxides was 33.8%. The  
276 interactions between SOC and Fe and Al oxides contributed 21.4% to the  
277 variation of the stability and size distribution of soil aggregates.

278         The pattern of site scores presented in the ordination plot (Fig. 2 and Fig.  
279 3) was same for both RDA models. It clearly showed that the first RDA axis  
280 indicated SOC in different sites increasing to left, while the second RDA axis

281 indicated the vegetation increasing to top or anthropogenic disturbance

282 increasing to bottom.

283

### 284 **3.3 Soil properties of different aggregatesize fractions**

285 Different types of land use had a significant impact on soil properties of  
286 different size fractions of soil aggregates (Table 4). HA in 0.5~0.25mm aggregate  
287 (HA<sub>5</sub>), organic carbon in <5 mm aggregate (SOC<sub>2-6</sub>), and FA in >5mm aggregate  
288 (FA<sub>1</sub>) were significantly different under different types of land use. In the  
289 <0.25 mm aggregate size fraction, the oxalate-extractable Al oxide (Al<sub>6</sub><sup>0xa</sup>) and  
290 HA/FA ratio (HA<sub>6</sub>/FA<sub>6</sub>) were also significantly different under different types of  
291 land use.

292 The stability of soil aggregates (MWD and WSA) were closely related to the  
293 corresponding soil properties in various size fractions of soil aggregates (Table 5).  
294 The oxalate-extractable Al oxide in <0.25 mm aggregates (Al<sub>6</sub><sup>0xa</sup>) and the organic  
295 matter in >2mm aggregate (HA<sub>1</sub> and FA<sub>2</sub>) positively contributed to MWD and  
296 WSA, while the oxalate-extractable Fe (Fe<sub>4</sub><sup>0xa</sup>) and Al oxides (Al<sub>2</sub><sup>0xa</sup>) and  
297 humus (HF<sub>3</sub>) in mid-sized aggregates negatively contributed to MWD and WSA.

298 Except for PSA<sub>2</sub>, percentages of all size fractions of soil aggregates resulted  
299 insignificantly linear relationships with their corresponding soil properties. The  
300 organic matter (including SOC, HA, and FA) was negatively related to the  
301 percentage of smaller aggregates (<1 mm) but positively correlated with the  
302 percentage of bigger aggregates (>1 mm). However, the correlations of Fe and Al

303 oxides werenot consistent within different size fractions of soil aggregates.

304

## 305 **4. Discussion**

### 306 **4.1 Effects of land use on soil aggregates**

307 The effects of land use on soil were profound and comprehensive. The  
308 stability and size distribution of soil aggregatesweresignificantly different in  
309 terms of types of land use ([Fig. 1](#)). The macro-aggregates, which generally had  
310 anorder offorest>rangeland/grassland>arable land, have been observed  
311 todecrease or disappear with sustained planting([Haynes et al., 1991](#); [Jastrow,](#)  
312 [1996](#); [John et al., 2005](#); [Ashagrie et al., 2007](#)). However, the percentage of  
313 macro-aggregates in the paddy washighest among the 3 types of land use.  
314 Although the paddy is one kind of arable land that involves long-term  
315 anthropogenic disturbances,its special water management systems  
316 ([Kögel-Knabner et al., 2010](#))resulted in highest SOC, HF,and HA([Table 3](#)).

317 The soil properties, especially SOM related properties, were significantly  
318 different in terms of land use type ([Table 3](#)). On the one hand,SOM is significantly  
319 affected by land use([John et al., 2005](#); [Don et al., 2011](#); [Li et al., 2015](#)); on the  
320 other hand, SOM playsprofoundly important roles in the stability and size  
321 distribution of soil aggregates([Tisdall and Oades, 1982](#); [Barral et al., 1998](#);  
322 [Abiven et al., 2009](#); [Alagöz and Yilmaz, 2009](#)), it is suspected that the effects of  
323 land use on soil aggregates are driven by the SOM or its components.

324 Additional,66.6% variations of soil aggregateswereexplained by land use



325 (Fig. 2), implying that the driving mechanism for the stability and size  
326 distribution of soil aggregates should include other factors, such as Fe and Al  
327 oxides, although they were not influenced by land use.

328

#### 329 **4.2 Effects of soil properties on soil aggregates**

330 The 84.3% variations of soil aggregates interpreted by organic matter and  
331 Fe and Al oxides (Fig. 3) mean that SOC and Fe and Al oxides and their  
332 interactions may be an important mechanism for the stability and size  
333 distribution of soil aggregates. It is also explicitly demonstrated by the partition  
334 of the variation of soil aggregates (Fig. 4).

335 SOM participates in various processes of the formation and stability of soil  
336 aggregates (Tisdall and Oades, 1982; Six et al., 2004). Furthermore, Fe and Al  
337 oxides and SOM can form stable organo-mineral complexes that can enhance the  
338 tensile strength and stability of the aggregate (Barral et al., 1998). Edwards and  
339 Bremner (1967) suggested that the formation of soil aggregates is a specific  
340 process that involves the absorption of organic matter into clay through the  
341 bridge of the multivalent metal cation; this is also known as a composite process  
342 of soil organic-inorganic complexes (Bronick and Lal, 2005).

343 Amézketa (1999) argued that Fe and Al oxides, which bond with organic and  
344 inorganic compounds, or aggregates through a cation bridge to improve soil  
345 structure, are the most active components in soil.

346

### 347 **4.3 Role of soil properties in different size fractions of soil aggregates**

348 The soil properties do not differ significantly in terms of fraction size,  
349 which is inconsistent with the hierarchical theory of soil aggregates (Tisdall and  
350 Oades, 1982; Puget et al., 2000; Six et al., 2000; John et al., 2005). It may result  
351 from kaolinite being the dominant clay, which has a 1:1 structure. Six et al. (2000;  
352 2002b) suggested that soil aggregates were not hierarchical in this kind of soil,  
353 and the bonding material between aggregates included not only organic materials  
354 but also Fe and Al oxides and other cementing materials..

355 The soil properties in each size fraction played different roles in the  
356 stability of soil aggregates. The oxalate-extractable Al oxide in the  
357 smaller aggregate ( $Al_6^{Oxa}$ ) and the soil organic components in the bigger  
358 aggregates ( $HA_1$ ,  $FA_2$ ) contributed positively to the MWD and WSA (Table 5).  
359 However, the oxalate-extractable Al oxides in 5~2 mm aggregate ( $Al_2^{Oxa}$ ), the  
360 humus in 2~1 mm aggregate ( $HF_3$ ), and the oxalate-extractable Fe oxides in  
361 1~0.5 mm aggregate ( $Fe_4^{Oxa}$ ) had negative contributions to the MWD and WSA.

362 In the smaller aggregates (<0.5 mm), the role of organic components ( $SOC_5$ ,  
363  $HF_6$ , and  $HA_6$ ) to the size distribution of soil aggregates were consistent. When  
364 organic components increased, the percentage of the  
365 smaller aggregates was significantly reduced, which indicates that the  
366 smaller aggregates were combining to form larger aggregates. This is also  
367 consistent with the theory that macro-aggregates are formed from  
368 micro-aggregates.

369           Meanwhile, the roles of different forms of Fe and Al oxides in those  
370 processes were inconsistent. In the smaller aggregates (0.5~0.25 mm and  
371 <0.25mm), the oxalate-extractable Al oxide ( $Al_5^{Oxa}$  and  $Al_6^{Oxa}$ ) was negatively  
372 correlated with the corresponding PSA, indicating that it was an important  
373 cementing substance beneficial to the formation of large aggregates, while the Fe  
374 oxide ( $Fe_5^{Oxa}$ ,  $Fe_5^{DCB}$ , and  $Fe_6^{Oxa}$ ) was a basic component of the aggregates due to  
375 a positive correlation with the corresponding PSA. In the >0.5mm aggregates, the  
376 oxalate-extractable Fe oxide ( $Fe_3^{Oxa}$  and  $Fe_4^{Oxa}$ ) acted as a cementing substance.

377           The roles of soil properties in the middle size fractions on the stability and  
378 size distribution of soil aggregates are worthy of further study. On the one hand,  
379 these middle-sized aggregates can be combined through the cementing agents,  
380 such as Fe and Al oxides, and organic matter, to form larger aggregates. In this case,  
381 the soil properties were positively related to the MWD and WSA and negatively  
382 related to the corresponding PSA. On the other hand, middle-sized aggregates  
383 can breakdown into smaller components, after which the corresponding variables  
384 should have the completely opposite pattern. Both processes of soil aggregates are  
385 homeostatic; they are indistinguishable unless dynamically monitored. In addition,  
386 because there was no conversion among the three types of land use, the  
387 relationships of transformations among the various size fractions of soil  
388 aggregates could not be determined.

389

## 390 **5. Conclusions**

391           The effects of land use on the stability and size distribution of soil  
392 aggregates were investigated based on soil properties and size fractions of soil  
393 aggregates in red soils in southern China with three types of land use, i.e., paddy,  
394 forest, and upland. The results suggested that land use significantly affected the  
395 stability and size distribution of soil aggregates through changing the SOM in soil  
396 and size fractions by high cultivating intensity or management related  
397 hydro-regime. Additionally, the Fe and Al oxides, which were not affected by land  
398 use, also had remarkable influence. The interaction of Fe and Al oxides and  
399 SOM, and the differential roles of Fe and Al oxides in formation or break-down of  
400 soil aggregates with middle size, were key to the stability and size distribution of  
401 soil aggregates in red soil in southern China.

402

### 403 **Acknowledgements**

404           This study is funded by the National Sciences Foundation of China  
405 (41171212), the National Key Basic Research Program of China (2012CB417106),  
406 and the China Scholarship Council (201308420455). The authors are grateful to  
407 several anonymous reviewers for their valuable comments on the early  
408 manuscript. The authors also thank Mr. Wei Yang and Dr. Junguang Wang for their  
409 helps in sampling and chemical analysis.

410

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502

1 **Table 1 Location, land use, annual mean precipitation, parent matherial and soil type of study sites**

Location	Code	Coordinates	Land use	Site description	AMP (mm)	Parent material	Soil type
Jinxian	P1	N28°21'31.9" E116°10'26.8"	Paddy	>50 yrs paddy field	1580	Redeposited Q2 red clay	Endoaquepts
Jinxian	P2	N28°21'02.4" E116°10'30.9"	Paddy	>20 yrs paddy field	1580	Redeposited Q2 red clay	Endoaquepts
Changsha	P3	N28°13'31.1" E113°09'94.2"	Paddy	>10 yrs paddy field	1410	Q2 red clay	Endoaquepts
Xianning	F1	N30°00'03.6" E114°22'38.7"	Forest	>40 yrs horsetail pine forest	1547	Q2 red clay	Plinthudults
Changsha	F2	N28°13'31.9" E113°09'45.7"	Forest	>10 yrs mixed forest	1319	Q2 red clay	Plinthudults
Changsha	F3	N28°15'40.3" E113°11'10.6"	Forest	>30 yrs <i>Cunninghamia lanceolata</i> forest	1319	Q2 red clay	Plinthudults
Taoyuan	U1	N29°13'43.4" E111°31'17.8"	Upland	>3 yrs corn with straw return	1510	Q2 red clay	Plinthudults
Xianning	U2	N30°01'18.9" E114°21'44.4"	Upland	>3 yrs rapeseed and soybean	1547	Q2 red clay	Plinthudults
Changsha	U3	N28°16'06.4" E113°14'25.9"	Upland	>3 yrs soybean and sweet potato	1410	Q2 red clay	Plinthudults

2 Note: Q2 means Middle Pleistocene, the second part of Quaternary.

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**Table 2 Basic soil properties, texture and structure of study sites**

Code	BD (g cm <sup>-3</sup> )	pH	SOC (gC kg <sup>-1</sup> )	STN (gN kg <sup>-1</sup> )	C/N	Silt (%)	Clay (%)	Sand (%)	Texture	Structure
P1	0.96±0.01	4.9±0.04	18.6±0.35	1.71±0.02	10.8±0.17	66.6±0.12	30.5±0.29	2.9±0.17	silty clay loam	Subangular blocky
P2	1.14±0.03	4.8±0.05	14.9±0.39	1.51±0.06	9.9±0.15	67.9±2.67	25.8±1.06	6.3±3.73	silty loam	Subangular blocky
P3	1.18±0.06	5.2±0.06	13.9±0.12	1.47±0.01	9.5±0.09	46.9±3.56	41.9±3.50	11.2±0.06	silty clay	Subangular blocky
F1	1.31±0.12	4.5±0.06	15.5±0.51	1.34±0.04	11.6±0.43	52.2±0.08	41.8±0.85	6.1±0.77	silty clay	Subangular blocky
F2	1.45±0.08	4.3±0.05	11.5±0.15	1.15±0.02	10.0±0.08	46.7±0.71	37.7±1.71	15.6±1.00	silty clay loam	Crumb
F3	1.39±0.13	4.2±0.06	11.9±0.22	1.28±0.01	9.3±0.14	42.9±0.47	52.7±0.71	4.4±0.24	silty clay	Crumb
U1	1.25±0.12	4.9±0.05	8.2±0.32	1.30±0.05	6.4±0.24	53.4±1.50	24.4±0.88	22.2±0.62	silty loam	Subangular blocky
U2	1.35±0.05	4.6±0.05	10.0±0.60	1.42±0.04	7.1±0.45	57.9±0.62	34.0±0.54	8.2±1.16	silty clay loam	Subangular blocky
U3	1.32±0.15	4.7±0.02	6.0±0.23	0.99±0.03	6.1±0.18	45.5±1.12	48.2±0.87	6.3±1.99	silty clay	Crumb

Note: All data are expressed in mean±SE, which are calculated based on 5 samples. BD is bulk density; SOC is soil organic carbon; and STN is soil total nitrogen; C/N is the ratio of SOC to STN. pH was measured in a 2.5:1 mixture of water:soil.

9

**Table 3 Soil properties under different types of land use**

	<b>Fe<sup>Oxa</sup></b>	<b>Fe<sup>DCB</sup></b>	<b>Al<sup>Oxa</sup></b>	<b>Al<sup>DCB</sup></b>	<b>HF</b>	<b>FA</b>	<b>HA</b>	<b>SOC</b>	<b>HA/FA</b>
	g kg <sup>-1</sup>				g C kg <sup>-1</sup>				
Paddy	5.68±1.59	16.29±2.08	1.54±0.25	1.43±0.25	5.07±0.66a	3.87±0.42a	1.20±0.50	15.8±1.41a	0.32±0.12
Forest	1.93±0.58	17.49±2.55	2.34±0.40	1.52±0.17	4.53±0.31ab	3.39±0.08a	1.15±0.23	13.0±1.28a	0.33±0.06
Upland	3.73±0.82	14.20±1.01	1.41±0.33	1.10±0.20	2.93±0.42b	2.06±0.27 b	0.87±0.15	8.1±1.16b	0.42±0.02

10 Note: All data is expressed as mean±SE, where sample size is 3, i.e., the number of sites for a specific land use. Different letters in a column  
11 indicates the significant difference of the specific property under the different types of land use based on the post hoc multiple comparison  
12 by least significant difference with significant level 0.05; Fe<sup>Oxa</sup> and Al<sup>Oxa</sup> are the oxalate extractable Fe and Al oxides; Fe<sup>DCB</sup> and Al<sup>DCB</sup> are the  
13 DCB extractable Fe and Al oxides; HF, FA, HA, SOC are the carbon content of humus, fulvic acid, humic acid and soil organic matter,  
14 respectively.

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16

**Table 4 Soil properties in size fractions of soil aggregates under different types of land use**

	$Al_6^{Oxa}$	HA <sub>5</sub>	FA <sub>1</sub>	SOC <sub>2</sub>	SOC <sub>3</sub>	SOC <sub>4</sub>	SOC <sub>5</sub>	SOC <sub>6</sub>	(HA/FA) <sub>6</sub>
Paddy	1.77±0.16a	1.22±0.17a	3.47±0.27a	17.2±1.55a	17.8±1.70a	16.5±0.94a	15.6±1.35a	14.6±1.94a	0.32±0.06b
Forest	1.66±0.08a	0.52±0.02b	3.37±0.30a	11.4±0.50ab	10.0±0.36b	9.0±0.26b	8.4±0.32b	8.6±0.79b	0.24±0.04b
Upland	1.20±0.11b	0.86±0.14ab	2.10±0.41b	9.3±2.60b	9.9±2.82b	8.7±2.18b	7.8±1.68b	7.1±1.50b	0.61±0.08a

17 Note: Only the properties that significantly affected by land use are show; All data is expressed as mean±SE, where sample size is 3, i.e., the  
18 number of sites for a specific land use. Different letters in a column indicates the significant difference of the specific property in terms of types of  
19 land use based on the post hoc multiple comparison by least significant difference with significant level 0.05;  $Al^{Oxa}$  is the oxalate extractable Al  
20 oxides; FA, HA, and SOC are the carbon content of humus, fulvic acid, humic acid and soil organic matter, respectively; The subscript of each  
21 properties indicates the fraction size of the soil aggregate, i.e., 1, 2, ..., 6 represents the size of >5 mm, 5~2 mm, 2~1 mm, 1~0.5 mm, 0.5~0.25mm  
22 and <0.25mm.

23

24 **Table 5 Multiple linear regression and partial correlation analysis between stability and size distribution and soil**  
 25 **properties in the different size fraction of soil aggregates under different types of land use**

Aggregate properties	Linear regression model with partial correlation coefficient	R <sup>2</sup>
<b>MWD</b>	-2.785 + 2.792 Al <sub>6</sub> <sup>Oxa</sup> (0.999) - 0.400 Al <sub>2</sub> <sup>Oxa</sup> (-0.974) - 0.044 Fe <sub>4</sub> <sup>Oxa</sup> (-0.985) + 1.144 HA <sub>1</sub> (0.999)	0.999 <u>7</u>
<b>WSA</b>	-34.344 + 50.763 Al <sub>6</sub> <sup>Oxa</sup> (0.999 <u>8</u> ) - 10.233 HF <sub>3</sub> (-0.998) + 32.434 HA <sub>1</sub> (0.999 <u>7</u> ) + 7.513 FA <sub>2</sub> (0.995)	0.999 <u>6</u>
<b>PSA<sub>1</sub></b>	-19.104 - 3.150 Fe <sub>1</sub> <sup>DCB</sup> (-0.825) - 17.367 Al <sub>1</sub> <sup>Oxa</sup> (-0.919) + 56.646 Al <sub>1</sub> <sup>DCB</sup> (0.948) + 15.731 FA <sub>1</sub> (0.956)	0.952
<b>PSA<sub>3</sub></b>	18.556 - 0.613 Fe <sub>3</sub> <sup>Oxa</sup> (-0.560) - 1.212 Fe <sub>3</sub> <sup>DCB</sup> (-0.657) + 9.730 Al <sub>3</sub> <sup>Oxa</sup> (0.762)	0.797
<b>PSA<sub>4</sub></b>	13.508-0.286 Fe <sub>4</sub> <sup>Oxa</sup> (-0.890)+0.311 Fe <sub>4</sub> <sup>DCB</sup> (0.897) - 2.233 Al <sub>4</sub> <sup>DCB</sup> (-0.874) - 1.523 HF <sub>4</sub> (-0.979)	0.975
<b>PSA<sub>5</sub></b>	9.995 + 0.651 Fe <sub>5</sub> <sup>Oxa</sup> (0.985) + 0.151 Fe <sub>5</sub> <sup>DCB</sup> (0.913) -1.624 Al <sub>5</sub> <sup>Oxa</sup> (-0.937) - 0.443 SOC <sub>5</sub> (-0.992)	0.987
<b>PSA<sub>6</sub></b>	126.066 + 7.673 Fe <sub>6</sub> <sup>Oxa</sup> (0.966) - 40.242 Al <sub>6</sub> <sup>Oxa</sup> (-0.978) - 6.175 HF <sub>6</sub> (-0.937) - 32.253 HA <sub>6</sub> (-0.892)	0.977

26 Note: All variables are partially correlated with MWD, WSA and PSA with  $p < 0.05$ , except for the ones underlined; The partial correlation  
 27 coefficient is shown in parentheses. The significance test of the partial correlation is based on a  $t$  distribution with degree of freedom:  $n-2-nv$ ,  
 28 where  $n$  is sample size, i.e., 9,  $nv$  is the number of the controlled variables. Fe<sup>Oxa</sup> and Al<sup>Oxa</sup> are the oxalate extractable Fe and Al oxides; Fe<sup>DCB</sup> and  
 29 Al<sup>DCB</sup> are the DCB extractable Fe and Al oxides; HF, FA, HA, SOC are the carbon content of humus, fulvic acid, humic acid and soil organic matter,  
 30 respectively; The subscript of each properties indicates the fraction size of the soil aggregate, i.e., 1, 2, ..., 6 represents the size of >5 mm, 5~2 mm,  
 31 2~1 mm, 1~0.5 mm, 0.5~0.25mm and <0.25mm.

32

1 **Figure captions:**

2

3 **Fig. 1.** Mean weight diameter (MWD), water-stable aggregate (WSA), and size  
4 distribution of soil aggregates under 3 types of land use.  $PSA_i$  is the percentage of  
5 the  $i^{\text{th}}$  size fraction of soil aggregates, with  $i = 1, 2, \dots, 6$  representing the size  
6 of  $> 5$  mm,  $5\sim 2$  mm,  $2\sim 1$  mm,  $1\sim 0.5$  mm,  $0.5\sim 0.25$ mm and  $<0.25$ mm. Different  
7 letters on the top of the error bars indicate the significant differences in terms of  
8 types of land use. The error bar indicates the standard error ( $n = 3$ ) of a  
9 measurement.

10

11 **Fig. 2.** Ordination plot of redundancy analysis (RDA) for the stability and size  
12 distribution of soil aggregates with types of land use as the constraining variable.  
13 The solid points indicate scores of sampling sites (P for paddy, F for forest, and U for  
14 upland). The squares are centroids of types of land use. The ellipses indicate the  
15 standard errors ( $n = 3$ ) of types of land use with a 95% confidence levels. MWD is  
16 mean weight diameter; WSA is water-stable aggregate (WSA),  $PSA_i$  is the percentage  
17 of the  $i^{\text{th}}$  size fraction of soil aggregates, with  $i = 1, 2, \dots, 6$  representing the size  
18 of  $> 5$  mm,  $5\sim 2$  mm,  $2\sim 1$  mm,  $1\sim 0.5$  mm,  $0.5\sim 0.25$ mm and  $<0.25$ mm.

19

20 **Fig. 3.** Ordination plot of redundancy analysis (RDA) for the stability and size  
21 distribution with soil properties as constraining variables. The solid points indicate

22 scores of sampling sites (P for paddy, F for forest, and U for upland). MWD is mean  
23 weight diameter; WSA is water-stable aggregate (WSA),  $PSA_i$  is the percentage of the  
24  $i^{\text{th}}$  size fraction of soil aggregates, with  $i = 1, 2, \dots, 6$  representing the size of  $> 5$  mm,  
25  $5 \sim 2$  mm,  $2 \sim 1$  mm,  $1 \sim 0.5$  mm,  $0.5 \sim 0.25$  mm and  $< 0.25$  mm.  $Al^{0\text{xa}}$  is the oxalate  
26 extractable Al oxides;  $Al^{\text{DCB}}$  and  $Fe^{\text{DCB}}$  are the DCB extractable Al and Fe oxides; SOC  
27 is soil organic carbon.

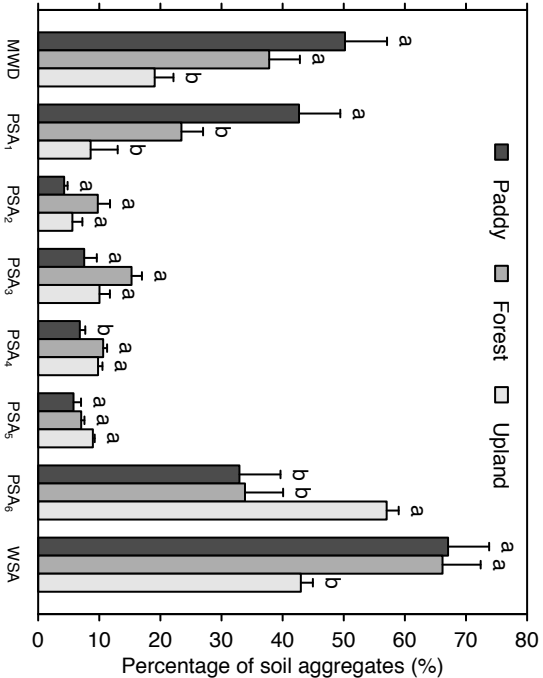
28

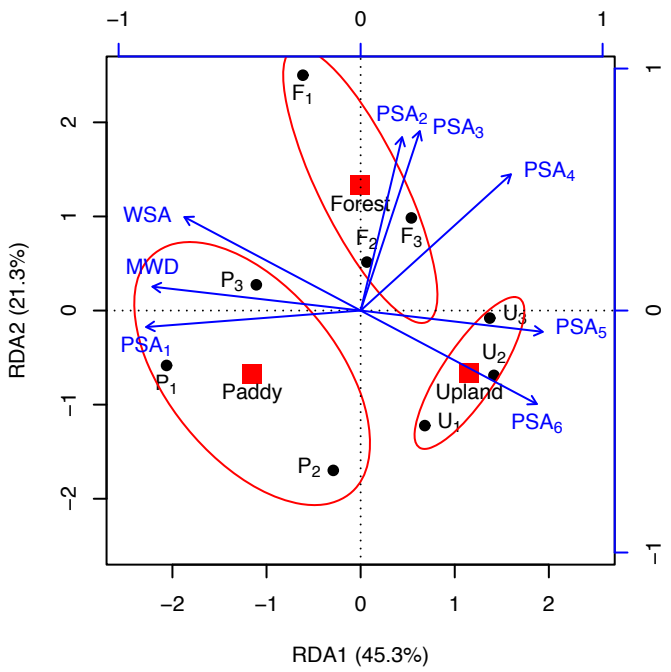
29 **Fig. 4.** Diagrams describing the partitions of variation of the stability and size  
30 distribution of soil aggregates by soil organic carbon (SOC), and Fe and Al oxides.

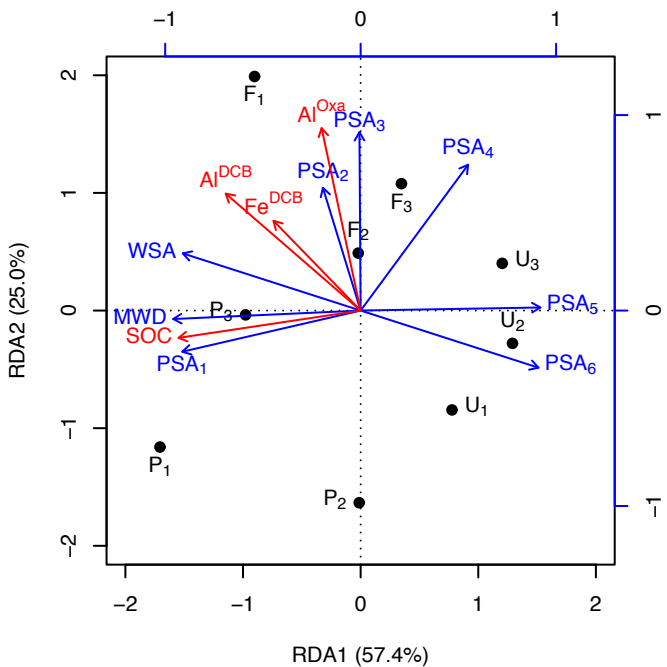


Mean weight diameter (mm)

0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0

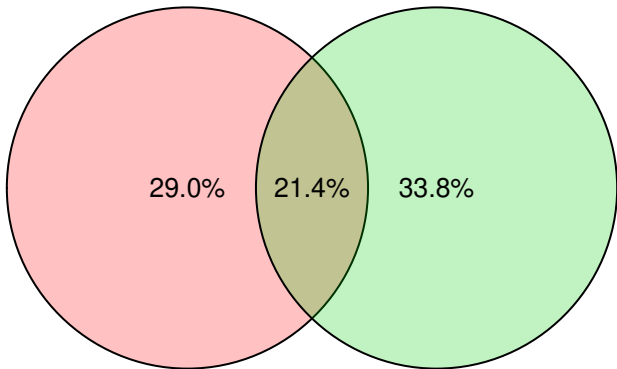






SOC

Fe & Al oxides



Residuals = 15.7%