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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Abstract

The stability and size distribution of soil aggregates is profoundly affected by land use, but the influencing mechanisms of land use are not clear. A study was carried out to investigate and attempted to interpret the effects of land use on soil aggregates from types of land use and soil properties in soil samples and size fractions of soil aggregates. Soil samples were taken from 9 sites under paddy, forest, and upland in southern China. The wet-sieving method was used to obtain 6 size fractions of soil aggregates: >5 , $5\sim2$, $2\sim1$, $1\sim0.5$, $0.5\sim0.25$, and <0.25 mm. The stability and size distribution of soil aggregates was measured as mean weight diameter (MWD), the percentage of water-stable aggregate (WSA) and the percentage of each size fraction (PSA). The quantities of soil organic carbon (SOC), humic substances, dithionite-citrate-bicarbonate (DCB) and oxalate extractable iron (Fe) and aluminum (Al) oxides were also measured. The results showed that types of land use solely explained 66.6% variation of soil aggregates; SOC, DCB-extractable Fe and Al oxides, and oxalate-extractable Al oxide caused 84.3% variation, in which SOC contributed 29.0%, Fe and Al oxides contributed 33.8%, and their interactions contributed 21.4%. The multiple linear regression and partial correlation analysis showed that soil organic matter and Fe and Al oxides had significant effects but played different roles on the stability and size distribution of soil aggregates. The study suggests that land use affects the stability and size distribution of soil aggregates through the integration of soil organic matter and Fe and Al oxides.

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

40 iron and aluminum oxides

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1. Introduction

Soil structure has important influences on edaphic conditions and the environment. The structure is often measured by the stability of soil aggregates (Six et al., 2000; Bronick and Lal, 2005). Soil aggregation sustains soil fertility because it reduces erosion and mediates soil aeration as well as water infiltration and retention. Furthermore, soil aggregation protects soil organic matter (SOM) from mineralizing because it physically reduces the accessibility of organic compounds for microorganisms, extracellular enzymes, and oxygen (Oades, 1984; Six et al., 2002a; von Lützow et al., 2006; Spohn and Giani, 2010). The size distribution and stability of soil aggregates are under the control of various mechanisms. Generally, the size distribution and stability of soil aggregates positively correlates with the main cementing agents, such as SOM, clay minerals, multi-valent cations and their complex in soil aggregates. According to the hierarchical theory of aggregate formation, these materials may be distributed unevenly in different size fractions of soil aggregates, and they may have close relationships with the stability of soil aggregates (Tisdall and Oades, 1982; Six et al., 2004).

Land use and associated management, such as crop sequencing, fertilization, soil conditioning, drainage and irrigation, are the most important and direct ways to affect soil structure and properties, through its impact on destruction forces and aggregate forming processes (Haynes et al., 1991; Besnard et al., 1996; Jastrow, 1996; John et al., 2005; Ashagrie et al., 2007; Lehrsch et al.,

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

In southern China, the red soils cover approximately 1.14 million km². Because of intensive weathering and leaching, the soil is rich in Fe and Al oxides, and the dominant clay mineral is kaolinite (He et al., 2004). In recent years, soil structure has degraded rapidly, due to strong human intervention and high cultivating intensity (Iqbal et al., 2009), and the sustainability of the agricultural ecosystem is under serious threat.

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

2. Materials and Methods

2.1 Study sites and soil samplings

Nine sites located in Jinxian, Jiangxi Province, Changsha and Taoyuan, Hunan Province, and Xianning, Hubei Province, southern China, were chosen, taking into account the variability of soil type (Inceptisols and Ultisols), in order to compare how the land use (paddy, forest, and upland) and the associated soil properties affected the stability and size distribution of soil aggregates. All sampling sites are in the subtropics and share similar climate. Those regions are humid and have a monsoon climate, with an annual mean temperature above 15°C and annual mean precipitation above 1300 mm. The paddy sites have been used for planting rice for more than ten years. The forest sites are secondary forests, with predominance of *Pinus massoniana* in Xianning (F₁), *Cunninghamia lanceolata* in Changsha (F₃), and a mixed forest in Changsha (F₂). The uplands are used for grain or cash crops such as maize (*Zea mays*), rapeseed (*Brassica napus*), soybean (*Glycine max*), and sweet potato (*Ipomoea batatas*). Paddy and upland sites are managed with conventional tillage. The same type of land use shares similar agricultural practices.

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

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

2.2 Water-stable aggregates

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

Mean weight diameter (MWD) was calculated as

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

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

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

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

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

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

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

2.3 Carbon content of soil organic components

Humus (HF) in the soil sample or a size fraction of soil aggregates was extracted using the method proposed by the Institute of Soil Science, Chinese Academy of Sciences (1978). For each sample, 5 g of air-dried soil were placed into a 250-mL Erlenmeyer flask. Then 100 mL 0.1 mol L⁻¹ sodium pyrophosphate-sodium hydroxide were added. The mixture was shaken for 30 mins and then left to stand for 13~14 h. Depending on the color depth, 2~15 mL supernatant were drawn into the digestive tract and neutralized with 0.5 mol L⁻¹ H₂SO₄. The supernatant was evaporated to dryness on a boiling water bath, after which the carbon content of HF was determined using the dichromate oxidization method (Bao, 2000).

Fifty milliliters of the previous supernatant were drawn into a 100-mL Erlenmeyer flask. The pH was adjusted to 3 or less with 0.5 mol L⁻¹ H₂SO₄ at 60~70 °C. It was then kept at 80 °C for 30 mins with a water bath. After cooling for about 8 h, it was filtered with a fine filter paper (Grade 44, Whatman). The precipitation on the filter paper was washed 3 times using 0.025 mol L⁻¹ H₂SO₄ and then solubilized and washed into a 50-mL flask with 0.05 mol L⁻¹ NaOH at 40~50 °C. Then, 10~25 mL solution were drawn into the digestive tract and neutralized with 0.5 mol L⁻¹ H₂SO₄. The solution was evaporated to dryness on a boiling water bath, after which the carbon content of humic acid (HA) was

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

2.4 Organic carbon content in soil and size fractions

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

2.5 Free-form Fe and Al oxides

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

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

2.6 Amorphous Fe and Al oxide

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

2.7 Statistical analysis

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

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

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

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

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

3. Results

3.1 Soil properties and land use

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

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

3.2 Stability and size distribution of soil aggregates

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

order as paddy>forest >upland (Fig. 1). Paddy has the highest MWD, followed by forest. Upland has the lowest MWD, with half of this region being under forest land use. PSA₁ in paddy was highest, while the rest of the size fractions were smaller. PSA₆ was highest in upland. The differences among MWD, WSA, PSA₁, PSA₄, and PSA₆ under different types of land use were significant (Fig. 1). MWD, WSA, and PSA₆ showed similar patterns and no significant differences existed between paddy and forest, but significant differences were found among upland and the other 2 types. While PSA₁ and PSA₄ showed similar patterns of significant differences between paddy and other types of land use, there was no significant difference between forest and upland. PSA₂, PSA₃, and PSA₅ were not significantly different among the 3 types of land use.

The effects of land use type on the stability and size distribution of soil aggregates (MWD, WSA, and PSA) were explored by the RDA. The effects of land use accounted for 66.6% of variation of the stability and size distribution of soil aggregates. The first axis of RDA accounted for 45.3%, mainly contributed by upland and paddy, while the second axis of RDA accounted for 21.3%, which was mainly contributed by forest (Fig. 2). MWD, WSA and PSA presented a clear pattern, where MWD, WSA and PSA₁ were grouped and closely related with each other. Similarly, the middle-sized fractions (PSA₂, PSA₃, and PSA₄), and small-sized fractions of soil aggregates (PSA₅, and PSA₆) were grouped separately (Fig. 2). Paddy, forest and upland had a positive contribution to group 3, respectively. Permutation tests showed that the land use types had significant

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

The relationships between soil properties and the stability and size distribution of soil aggregates were also explored by RDA, and showed that the DCB-extractable Fe and Al oxides, oxalate-extractable Al oxide, and SOC could explain 84.3% of the variation in the stability and size distribution of soil aggregates under different types of land use. The first RDA axis accounted for 57.4%, mainly contributed by SOC, while the second RDA axis accounted for 25.0%, mainly contributed by the oxalate-extractable Al oxide (Fig. 3). The DCB-extractable Fe and Al oxides contributed equally to both ordination axes. All soil properties in the RDA model had significant positive correlations with MWD, WSA, and PSA_1 , but negative with PSA_5 and PSA_6 . For PSA_2 , PSA_3 and PSA_4 , Fe and Al oxides were positive contributors, while SOC presented a negative contribution (Fig. 3). Permutation tests showed that all soil properties significantly affected soil aggregates ($p<0.05$).

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

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

indicated the vegetation increasing to top or anthropogenic disturbance
increasing to bottom.

3.3 Soil properties of different aggregatesize fractions

Different types of land use had a significant impact on soil properties of
different size fractions of soil aggregates ([Table 4](#)). HA in 0.5~0.25mm aggregate
(HA₅), organic carbon in <5 mm aggregate (SOC₂₋₆), and FA in >5mm aggregate
(FA₁)were significantly different under different types of land use. In the
<0.25 mm aggregate size fraction,the oxalate-extractable Al oxide(Al₆^{0xa}) and
HA/FA ratio (HA₆/FA₆) werealso significantly different under different types of
land use.

The stability of soil aggregates (MWD and WSA)wereclosely related to the
corresponding soilproperties in various size fractions of soil aggregates([Table 5](#)).
The oxalate-extractable Al oxide in <0.25 mm aggregates (Al₆^{0xa}) and the organic
matter in >2mm aggregate(HA₁ and FA₂) positively contributed to MWD and
WSA, while the oxalate-extractable Fe (Fe₄^{0xa})and Al oxides(Al₂^{0xa})and
humus(HF₃) in mid-sized aggregates negatively contributed to MWD and WSA.

Except for PSA₂, percentages ofall size fractions of soil aggregatesresulted
insignificantly linear relationships with their corresponding soilproperties.The
organic matter(including SOC, HA, and FA) wasnegatively related to the
percentage of smalleraggregates (<1 mm) but positively correlated with the
percentage of bigger aggregates (>1 mm). However, the correlations of Fe and Al

oxides werenot consistent within different size fractions of soil aggregates.

4. Discussion

4.1 Effects of land use on soil aggregates

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

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

Additional,66.6% variations of soil aggregateswereexplained by land use

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

4.2 Effects of soil properties on soil aggregates

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

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

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

4.3 Role of soil properties in different size fractions of soil aggregates

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

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

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

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

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

5. Conclusions

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

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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

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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 ⁻³)	pH	SOC (gC kg ⁻¹)	STN (gN kg ⁻¹)	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.

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Table 3 Soil properties under different types of land use

	Fe^{Oxa}	Fe^{DCB}	Al^{Oxa}	Al^{DCB}	HF	FA	HA	SOC	HA/FA
	g kg ⁻¹				g C kg ⁻¹				
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^{Oxa} and Al^{Oxa} are the oxalate extractable Fe and Al oxides; Fe^{DCB} and Al^{DCB} are the
13 DCB extractable Fe and Al oxides; HF, FA, HA, SOC are the carbon content of humus, fulvic acid, humic aid and soil organic matter,
14 respectively.

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

	Al_6^{Oxa}	HA ₅	FA ₁	SOC ₂	SOC ₃	SOC ₄	SOC ₅	SOC ₆	(HA/FA) ₆
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 aid 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.

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Table 5 Multiple linear regression and partial correlation analysis between stability and size distribution and soil 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^2
MWD	$-2.785 + 2.792 \text{ Al}_6^{\text{Oxa}} (0.999) - 0.400 \text{ Al}_2^{\text{Oxa}} (-0.974) - 0.044 \text{ Fe}_4^{\text{Oxa}} (-0.985) + 1.144 \text{ HA}_1 (0.999)$	0.999 <u>7</u>
WSA	$-34.344 + 50.763 \text{ Al}_6^{\text{Oxa}} (0.9998) - 10.233 \text{ HF}_3 (-0.998) + 32.434 \text{ HA}_1 (0.9997) + 7.513 \text{ FA}_2 (0.995)$	0.999 <u>6</u>
PSA₁	$-19.104 - 3.150 \text{ Fe}_1^{\text{DCB}} (-0.825) - 17.367 \text{ Al}_1^{\text{Oxa}} (-0.919) + 56.646 \text{ Al}_1^{\text{DCB}} (0.948) + 15.731 \text{ FA}_1 (0.956)$	0.952
PSA₃	$18.556 - 0.613 \text{ Fe}_3^{\text{Oxa}} (-0.560) - 1.212 \text{ Fe}_3^{\text{DCB}} (-0.657) + 9.730 \text{ Al}_3^{\text{Oxa}} (0.762)$	0.797
PSA₄	$13.508 - 0.286 \text{ Fe}_4^{\text{Oxa}} (-0.890) + 0.311 \text{ Fe}_4^{\text{DCB}} (0.897) - 2.233 \text{ Al}_4^{\text{DCB}} (-0.874) - 1.523 \text{ HF}_4 (-0.979)$	0.975
PSA₅	$9.995 + 0.651 \text{ Fe}_5^{\text{Oxa}} (0.985) + 0.151 \text{ Fe}_5^{\text{DCB}} (0.913) - 1.624 \text{ Al}_5^{\text{Oxa}} (-0.937) - 0.443 \text{ SOC}_5 (-0.992)$	0.987
PSA₆	$126.066 + 7.673 \text{ Fe}_6^{\text{Oxa}} (0.966) - 40.242 \text{ Al}_6^{\text{Oxa}} (-0.978) - 6.175 \text{ HF}_6 (-0.937) - 32.253 \text{ HA}_6 (-0.892)$	0.977

Note: All variables are partially correlated with MWD, WSA and PSA with $p < 0.05$, except for the ones underlined; The partial correlation coefficient is shown in parentheses. The significance test of the partial correlation is based on a t distribution with degree of freedom: $n-2-nv$, where n is sample size, i.e., 9, nv is the number of the controlled variables. Fe^{Oxa} and Al^{Oxa} are the oxalate extractable Fe and Al oxides; Fe^{DCB} and Al^{DCB} are the DCB extractable Fe and Al oxides; HF, FA, HA, SOC are the carbon content of humus, fulvic acid, humic aid and soil organic matter, 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, 2~1 mm, 1~0.5 mm, 0.5~0.25mm and <0.25mm.

Figure captions:

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

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

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

scores of sampling sites (P for paddy, F for forest, and U for upland). MWD is mean weight diameter; WSA is water-stable aggregate (WSA), PSA_i is the percentage of the i^{th} size fraction of soil aggregates, with $i = 1, 2, \dots, 6$ representing the size 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. $Al^{0_{oxa}}$ is the oxalate extractable Al oxides; Al^{DCB} and Fe^{DCB} are the DCB extractable Al and Fe oxides; SOC is soil organic carbon.

Fig. 4. Diagrams describing the partitions of variation of the stability and size 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

MWD

PSA₁

PSA₂

PSA₃

PSA₄

PSA₅

PSA₆

WSA

Paddy Forest Upland

Percentage of soil aggregates (%)

SOC

Fe & Al oxides

29.0%

21.4%

33.8%

Residuals = 15.7%