Variations in the aluminum fractions within soils associated with different tea varieties (Camellia sinensis L.): Insights at the aggregate scale

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Abstract

Background

Soil aggregates play an important role in the transformation of aluminum (Al) fractions, but little is known about the changes in Al fractions based on the aggregates in soils of different tea varieties.

Methods

The distribution of Al fractions in aggregates collected from soils planted with four tea varieties (*C. sinensis 'Fuding Dabaicha'* (FD), *Chuancha No. 3* (CC3), *Chuanmu No. 217* (CM217) and *Chuannong Huangyazao* (CN)) was investigated by continuous extraction method.

Results

Results showed significant differences in the distributions of Al fractions among the soils, which were related to the changes in soil aggregate compositions and the aggregate-associated Al fractions. The FD soil had the highest Or-Al concentration, which was related to its high proportion of > 5 mm aggregates and the high Or-Al concentration in the > 5 mm aggregates. The CC3 soil had the highest concentrations of total aluminum (T-Al), water-soluble aluminum (Ws-Al) and exchangeable aluminum (Ex-Al), and the accumulation of T-Al, Ws-Al and Ex-Al depended on the highest concentrations of T-Al, Ws-Al and Ex-Al in the > 5 mm and < 0.25 mm aggregates among the four soils. Therefore, the formation of aggregates and the high Al preservation capacity of the aggregates determined the high Al concentrations of the FD and CC3 soils.

Conclusions

This study helps elucidate the mechanisms of Al sequestration, and provides evidence for developing appropriate management plans to prevent an increase in Al concentrations in the soils of CC3 and FD by adjusting the soil aggregate composition and the Al concentration of aggregates.

Introduction

Tea (*Camellia sinensis* L.) is one of the most popular beverages in the world, and it contains higher concentrations of aluminum (Al) than many other plants (Ding et al. 2021; Fung et al. 2009; Zhang et al. 2016). The concentration of Al in tea leaves is generally as high as 20–30 g kg^{-1} (Xu et al. 2017). Once taken up by tea plants from soils, Al can enter the human body upon the ingestion of tea (Hu et al. 2017). Some investigations have shown that Alzheimer's disease is related to a high Al concentration in the human brain (Mirza et al. 2017). Therefore, the distribution of Al in soil-tea ecosystems has attracted attention.

The Al accumulation level of tea is affected by the distribution of Al fractions in soils (Ding et al. 2021; Pei et al. 2014; Xie et al. 2007). Studies on Al sequential extraction have attracted attention in recent decades (Martins et al. 2020), and Al is fractionated into water-soluble Al (Ws-Al), exchangeable Al (Ex-Al), Fe/Mn oxide-bound Al (Fe/Mn-Al), organic matter-bound Al (Or-Al), and residual Al (Res-Al) (Xie et al. 2007). Ws-Al and Ex-Al are the most chemically and biologically available forms of Al, and they are absorbed directly by tea pants (Martins et al. 2020). Although Fe/Mn-Al and Or-Al are nonbioavailable fractions, the transformations of Al fractions are controlled by the reactions of the Or-Al fraction. The residual fraction is considered the most stable, least reactive, and least bioavailable in comparison to the other fractions, as associated metals are occluded within crystal lattices of layered silicates and within well-crystallized oxide minerals (Ghanati et al. 2005; Palleir et al. 2018). The different Al fractions in soil have different physical-chemical reactivities and bioavailabilities and are in dynamic equilibrium with one another (Pei et al. 2014). Therefore, it is particularly important to study the distribution of Al fractions in the soils of tea plantations.

As essential building blocks of the soil structure (Alagöz et al. 2009), aggregates can determine the physical, chemical, and biological properties of soils (An et al. 2013; Ferro et al. 2012; Qiao et al. 2021), therefore, they are crucial in regulating the transformations of the Al fractions. The distribution of the Al fractions is influenced by the process of soil formation, and the types of cementing substances are different during each stage of soil aggregate formation (Artemyeva et al. 2021; Pihlap et al. 2021; Wang et al. 2013). In general, free primary particles are bound into microaggregates (< 0.25 mm) by highly disordered alum inosilicates, oxides, and persistent adhesives (Voltolini et al. 2017). On the other hand, macroaggregates (> 0.25 mm) are generated mainly through the association of particulate organic matter, minerals and microaggregates (Li et al. 2018). Thus, the aggregates of different sizes are distinct in terms of maintaining Al in the soil, and it has been reported that the bioavailability of the Al fractions are controlled by aggregates (Yang et al. 2019).

Tea is largely planted to achieve the goal of sustainable agricultural development in the hilly areas of western Sichuan (Wang et al. 2016; Yu et al. 2020), and the plants are frequently replaced by newly improved tea varieties to achieve higher economic returns. Many studies have reported variation in the soil Al fractions associated with different varieties of tea (Peng et al. 2018). The transformations of the Al fractions are controlled by aggregates. However, little information is available regarding the distribution of Al fractions within aggregates of different sizes in tea plantation soils. Therefore, the primary objective of the present study was to determine the differences in the Al fractions within soil aggregates present in the plantations of different tea varieties and to provide further support for the promotion of certain tea varieties. We hypothesized that (1) the soil aggregate composition and aggregate-associated Al fractions would differ among the soils planted with the four tea varieties, and (2) differences in the distributions of the Al fractions in the bulk soils planted with four tea varieties would be related to changes in the soil aggregate composition and the aggregate-associated Al fractions.

Materials And Methods

Experimental position

This study was performed in a long-term experimental tea garden located in the Zhongfeng ecological tea plantation in Ya'an, Sichuan, China (Fig. 1). This area has a subtropical monsoon climate with an average annual temperature of 15.4 °C and a maximum temperature of 35.2 °C. The average annual rainfall is approximately 1500 mm, and the relative humidity is 82% (Wang et al. 2018). The exposed layer is composed of sedimentary rock predominantly formed since the Mesozoic, and the soil is classified as a Luvisol with a clay loam lexture (IUSS Working Group 2014). To prevent further deterioration of natural ecosystems, the Mingshan District of Ya'an city has undertaken ecological preservation measures, returning farmlands to forests (tea) based on the regional reality since the 1950s (Wang et al. 2017).

The cultivation density of the tea (plant to plant distance = 0.30 ± 0.04 m, broad rows = 1.50 ± 0.04 m, and narrow rows = 0.30 ± 0.15 m) was set to approximately 8×10^4 plants ha^{-1}. The annual fertilization regime in the tea plantations is as follows: 15 t ha^{-1} organic fertilizer is applied yearly in November as the basal fertilizer in the region vertically below the tree crown. Subsequently, 300 kg ha^{-1}, 225 kg ha^{-1} and 225 kg ha^{-1} organic manure are applied in February, April and July of the following year in the same location as the basal fertilizer application. Furthermore, all of the tea varieties are pruned in April and October, and the pruned litter remains in situ (Wang et al. 2018).

In this study, we attempted to minimize external effects by selecting tea plantations that had been cultivated for the same period of time (15 years) with different varieties (*C. sinensis 'Fuding Dabaicha'* (FD), *Chuancha No.3* (CC3), *Chuanmu No.217* (CM217) and *Chuannong Huangyazao* (CN)) and the same management measures. The plantations for the four tea varieties were located in a close geomorphologic unit with the same soil parent material, comparable slope gradient and slope position, and similar climatic conditions (Table 1). Each tea variety was planted in 10 broad rows, and three broad rows of tea plants were stochastically selected as subplots for each tea variety. The complete details of the experimental design were previously described by Yu et al. (2020).

Table 1
Soil characteristics of different tea varieties.

Tea varieties	Geographical coordinates λ(E), Ψ(N)	Elevation (m)	Slope gradient (°)	Area (ha)
FD	103°11'31', 30°12'28'	678	20	5.18
CC3	103°12'14', 30°12'12'	678	20	5.20
CM217	103°12'03', 30°11'57'	679	21	5.18
CN	103°11'28', 30°12'26'	679	21	5.19

Litter and soil sampling

In each subplot, litter samples from 5 randomly selected points collected from the soil surface and then mixed to form a composite sample. Three composite samples were collected from three subplots for each tea variety. All of the collected litter samples were enclosed with a 30 cm×30 cm fixed frame and placed into plastic bags. All of the litter samples were oven-dried at 85 ℃ until the samples reached a constant weight. Next, the dry samples were weighed, and analyses were conducted to determine their total Al concentrations.

Soil samples were obtained from the same site as the litter samples. In each subplot, soil samples (0–20 cm) from 5 randomly selected points were collected from a narrow row and then mixed to form a composite sample. Three composite samples were collected from three subplots for each tea variety. All of the soil samples were carefully fragmented into natural aggregates and then sieved with a mesh size of 5 mm to remove large roots, stones and macrofauna. One portion of these samples was used to measure the properties of the bulk soil (Table 2), and the other part was used for aggregate separation.

Table 2
Soil properties of different tea varieties.

Tea varieties	Litter quantity ($g\ m^{-2}$)	w(Al/litter) ($mg\ kg^{-1}$)	pH	Bulk density ($g\ cm^{-3}$)	Organic C concentration ($g\ kg^{-1}$)	Exchangeable Ca^{2+} ($cmol\ kg^{-1}$)	Exchangeable Mg^{2+} ($cmol\ kg^{-1}$)
FD	1588.07 a	5215.00 bc	4.13 b	1.23 a	30.18 a	1.88 a	0.34 a
CC3	1525.37 ab	8830.00 a	4.22 b	1.09 b	20.83 c	1.80 a	0.33 a
CM217	1377.33 b	6397.50 b	4.36 a	1.22 a	24.68 b	1.65 b	0.24 b
CN	1540.17 a	4205.00 c	4.38 a	1.08 b	25.14 b	1.64 b	0.22 c

Note: Different letters indicate significant differences ($P < 0.05$) among the different tea varieties. (FD) *C. sinensis 'Fuding Dabaicha'*, (CC3) *Chuancha No.3*, (CM217) *Chuanmu No.217*, (CN) *Chuannong Huangyazao.*

Aggregate separation

The distribution of water-stable aggregates was established using a wet-sieving method described in detail by Amezketa (1996). The soil clods (100 g) were placed on the topmost section of a nest of sieves with diameters 5, 2, 1, 0.5, and 0.25 mm. Distilled water was added until the water level reached 2 cm above the uppermost clods, and the soil sample was presoaked in distilled water for 15 min. Sieving consisted of raising and lowering the sieves with an amplitude of 5 cm and an oscillation rate of 1 s^{-1} for 15 min. Finally, the residuum in the sieve was washed separately into a beaker, while the water-stable aggregates were air dried at indoor temperature. The following six aggregate sizes were acquired: >5, 5 − 2, 2 − 1, 1-0.5, 0.5 − 0.25, and < 0.25 mm.

Soil analyses

The soil samples (both the bulk soil and aggregates) were air dried at room temperature for one week before determining their chemical properties. Soil pH was measured using a glass electrode, and the soil-to-water ratio reached 1:2.5 m/v (Lu 2000). Soil organic matter was measured by the acid dichromate wet oxidation method (Nelson and Sommers 1996). Exchange cations were measured using an ammonium acetate replacement method (Thomas 1982).

Total Al was measured by microwave digestion-plasma emission spectrometry. The various forms of Al were extracted by a continuous extraction method and were divided into the Ws-Al, Ex-Al, Fe/Mn-Al, and Or-Al fractions. The forms of Al were established with a sequential extraction procedure as follows: deionized water, 1 mol L^{-1} $MgCl_2$, 0.04 mol L^{-1} $NH_2OH{\cdot}HCl$ and 0.02 mol L^{-1} $HNO_3 + 30\%H_2O_2$. All of these parameters were measured by ICP-OES (Xie et al. 2007).

Notably, only a small amount of Ws-Al and Ex-Al was extracted by distilled water during the wet-sieving procedure, indicating that the wet-sieving process had little effect on the Ws-Al and Ex-Al concentrations in the soil aggregates of different sizes.

Calculation and statistical analyses

The formulas used to calculate the average weight diameter (*MWD*, mm) and geometric mean diameter (*GWD*, mm) of agglomerates are as follows (Li et al. 2013):

$$MWD=\frac{\sum_{i=1}^{n}\left(\overline{R}_i w_i\right)}{\sum_{i=1}^{n} w_i} \qquad GMD=Exp\left[\frac{\sum_{i=1}^{n} w_i \ln R_i}{\sum_{i=1}^{n} w_i}\right]$$

where R_i is the mean diameter of the ith size fraction (mm), and M_i is the mass percentage of the aggregate size (%).

The accumulation factor (AFx) was employed to estimate the enrichment of the Al fractions in each aggregate size fraction (Acosta et al. 2009). The formula used for the calculation is as follows:

$AFx = X_{fraction}/X_{bulk}$

where $X_{fraction}$ and X_{bulk} are the concentrations of Al (g kg^{-1}, mg kg^{-1}) in an individual aggregate size class and in bulk soil, respectively.

Another index to evaluate the contribution of aggregate sizes to the total accumulation of the Al fractions is the loading (Acosta et al. 2009). The loading was calculated as follows:

$GSF_{loading}=(X_i{\times}GS_i)/(\Sigma(X_i{\times}GS_i)){\times}100$

where X_i is the concentration of Al in an individual aggregate size (g kg^{-1}, mg kg^{-1}), and GS_i is the mass percentage of the individual aggregate size class (%).

Statistical analyses were conducted in SPSS software (Version 20.0). One-way analysis of variance (ANOVA) was used to determine the differences in the soil properties among the tea varieties. Two-way ANOVA was performed using tea varieties and aggregate sizes as factors to test the effects of tea varieties and aggregates on the concentration of each aggregate fraction and aggregate-associated Al fractions. The least significant difference (LSD) test was carried out to compare the different tea varieties and aggregate sizes, with $P < 0.05$ considered statistically significant.

Results

Distribution of the Al fraction in bulk soil

Res-Al was the predominant fraction, which varied from 54.92 ~ 70.42 g kg^{-1}, and Ws-Al was the fraction present in the lowest amounts, which varied from 1.86 ~ 2.71 mg kg^{-1} in the soils planted with the four tea varieties (Table 3). The distribution of Al factions clearly varied among the soils planted with the different tea varieties. The highest and lowest concentrations of T-Al, Ws-Al, Ex-Al, Fe/Mn and Res-Al were measured in the soils of CC3 and CN, respectively. The lowest concentration of Or-Al was measured in the soil of CC3.

Table 3
Distribution of aluminum fractions in soils of different tea varieties.

Tea varieties	Item						Effects
	T-Al (g kg^{-1})	Ws-Al (mg kg^{-1})	Ex-Al (mg kg^{-1})	Fe/Mn-Al (mg kg^{-1})	Or-Al (mg kg^{-1})	Res-Al (g kg^{-1})	tea plantation varieties (T)
FD	66.27 b	2.64 a	243.89 ab	544.08 b	131.71 a	65.35 b	**
CC3	71.31 a	2.71 a	254.05 a	562.29 a	71.48 d	70.42 a	
CM217	61.17 c	2.16 b	225.08 bc	531.15 b	82.84 c	60.33 c	
CN	55.74 d	1.86 c	206.78 c	502.40 c	106.51 b	54.92 d	

Note: Different letters indicate significant differences ($P < 0.05$) among the different tea varieties. T: tea varieties. **, * indicate significant at $P < 0.01$ and $P < 0.05$, respectively. (FD) *C. sinensis 'Fuding Dabaicha'*, (CC3) *Chuancha No.3*, (CM217) *Chuanmu No.217*, (CN) *Chuannong Huangyazao.*

Distribution of water-stable aggregates

The > 5 mm aggregates were the dominant fraction, varying from 28.71 ~ 33.89% in the soils planted with the four tea varieties, followed by the < 0.25 mm aggregates, which varied from 18.19 ~ 33.37% (Table 4). Specifically, the FD and CC3 soils were dominated by > 5 mm aggregates, and the CN soil was dominated by < 0.25 mm aggregates. The distribution of aggregate sizes differed significantly among the soils planted with the different tea varieties except for the 5 – 2 mm and 1-0.25 mm aggregates. The FD soil had the highest proportion of > 5 mm aggregates compared with the other soils, but an inverse trend was observed for the < 0.25 mm aggregates.

Table 4
Distribution of water-stable aggregates in soils of different tea varieties.

Tea varieties	**Concentration of soil aggregates (%)**						**Effects**		
	> 5 mm	**5 ~ 2 mm**	**2 ~ 1 mm**	**1 ~ 0.5 mm**	**0.5 ~ 0.25 mm**	**< 0.25 mm**	**tea plantation varieties (T)**	**Aggregate-size fraction(A)**	**T×A**
FD	33.89 a^{a}	11.94 b^{a}	13.75 c^{a}	12.99 c^{a}	9.25 d^{a}	18.19 b^{d}	**	**	**
CC3	30.83 a^{b}	9.69 d^{a}	14.96 c^{a}	11.60 d^{a}	8.97 d^{a}	21.89 b^{c}			
CM217	28.71 a^{b}	11.28 b^{a}	7.99 c^{b}	11.63 b^{a}	10.39 bc^{a}	29.36 a^{b}			
CN	29.87 b^{b}	9.18 c^{a}	8.65 c^{b}	11.00 c^{a}	9.80 c^{a}	33.37 a^{a}			

Note: Different lower-case letters indicate significant differences (*P*< 0.05) among the different aggregate sizes. Different superscript letters indicate significant difference (*P*< 0.05) among the different tea varieties. T: tea varieties; A: aggregate size. **, * indicate significant at *P*< 0.01 and *P*< 0.05, respectively. (FD) *C. sinensis ‘Fuding Dabaicha’*, (CC3) *Chuancha No.3*, (CM217) *Chuanmu No.217*, (CN) *Chuannong Huangyazao*.

Table 5
Distribution of Al fractions in soil aggregates of different tea varieties.

Item	Tea varieties	Aggregate size						Effects		
		> 5 mm	5 ~ 2 mm	2 ~ 1 mm	1 ~ 0.5 mm	0.5 ~ 0.25 mm	< 0.25 mm	T	A	T×A
T-Al ($g\ kg^{-1}$)	FD	58.07 d^{a}	58.78 d^{ab}	61.62 cd^{ab}	68.09 bc^{b}	73.26 ab^{b}	78.67 a^{a}	**	**	**
	CC3	61.44 c^{a}	64.59 c^{a}	66.16 c^{a}	75.14 b^{a}	80.44 ab^{a}	84.72 a^{a}			
	CM217	55.94 b^{a}	55.91 b^{bc}	57.25 b^{bc}	58.33 b^{c}	60.14 ab^{c}	66.48 a^{b}			
	CN	48.43 c^{b}	49.26 bc^{c}	53.20 abc^{c}	55.68 ab^{c}	56.11 ab^{c}	58.17 a^{c}			
Ws-Al ($mg\ kg^{-1}$)	FD	2.15 d^{b}	2.50 c^{a}	2.53 c^{a}	2.82 b^{a}	3.08 ab^{a}	3.12 a^{a}	**	**	**
	CC3	2.37 d^{a}	2.55 cd^{a}	2.57 bc^{a}	2.76 b^{a}	3.02 a^{a}	3.19 a^{a}			
	CM217	1.90 d^{c}	2.03 cd^{b}	2.19 bc^{b}	2.25 b^{b}	2.32 ab^{b}	2.45 a^{b}			
	CN	1.68 b^{d}	1.79 b^{c}	2.05 a^{b}	2.23 a^{b}	2.22 a^{b}	2.23 a^{c}			
Ex-Al ($mg\ kg^{-1}$)	FD	221.42 d^{a}	231.50 cd^{a}	239.92 bc^{a}	248.50 bc^{a}	256.42 ab^{a}	267.00 a^{ab}	**	**	**
	CC3	229.25 b^{a}	236.25 b^{a}	238.63 b^{a}	260.00 a^{a}	263.87 a^{a}	277.63 a^{a}			
	CM217	185.00 d^{b}	219.50 c^{a}	228.92 bc^{a}	244.83 ab^{a}	254.13 a^{a}	257.50 a^{b}			
	CN	176.64 b^{b}	183.89 b^{b}	190.65 b^{b}	193.70 b^{b}	211.50 b^{b}	226.88 a^{c}			
Fe/Mn-Al ($mg\ kg^{-1}$)	FD	615.61 a^{a}	574.54 b^{a}	549.98 c^{a}	536.28 c^{a}	509.63 d^{ab}	501.18 d^{a}	**	**	ns
	CC3	615.12 a^{a}	583.46 b^{a}	555.00 c^{a}	553.80 c^{a}	520.17 d^{a}	506.82 d^{a}			

Note: Different lower case letters indicate significant differences ($P < 0.05$) among the different aggregate sizes. Different superscript letters indicate significant difference ($P < 0.05$) among the different tea varieties. T: tea varieties; A: aggregate size. **, * indicate significant at $P < 0.01$ and $P < 0.05$, respectively. (FD) *C. sinensis 'Fuding Dabaicha'*, (CC3) *Chuancha No.3*, (CM217) *Chuanmu No.217*, (CN) *Chuannong Huangyazao*.

	CM217	563.27 a^{b}	546.68 a^{b}	518.98 b^{b}	507.23 bc^{b}	496.28 cd^{b}	479.94 d^{b}			
	CN	542.09 a^{c}	520.53 b^{c}	517.06 b^{b}	495.41 c^{b}	476.38 cd^{c}	460.18 d^{c}			
Or-Al (mg kg^{-1})	FD	138.94 a^{a}	131.21 ab^{a}	123.32 bc^{a}	116.26 c^{a}	105.96 d^{a}	95.18 e^{a}	**	**	*
	CC3	81.78 a^{d}	75.13 ab^{d}	71.67 bc^{d}	67.13 bcd^{d}	65.50 cd^{c}	60.15 d^{c}			
	CM217	103.27 a^{c}	93.35 b^{c}	92.32 b^{c}	82.23 c^{c}	72.95 d^{c}	69.94 d^{b}			
	CN	128.76 a^{b}	113.72 b^{b}	107.19 bc^{b}	100.41 cd^{b}	94.72 de^{b}	90.18 e^{a}			
Res-Al (g kg^{-1})	FD	57.09 c^{a}	57.84 c^{ab}	60.70 c^{ab}	69.77 b^{a}	72.38 ab^{b}	77.81 a^{a}	**	**	**
	CC3	60.49 c^{a}	63.70 c^{a}	65.29 c^{a}	74.27 bc^{a}	79.59 ab^{a}	83.87 a^{a}			
	CM217	55.08 b^{a}	55.04 b^{bc}	56.40 b^{bc}	57.49 b^{b}	59.77 ab^{c}	65.67 a^{b}			
	CN	47.58 c^{b}	48.46 bc^{c}	52.38 abc^{c}	54.88 ab^{b}	55.32 a^{c}	57.39 a^{c}			

Note: Different lower case letters indicate significant differences ($P < 0.05$) among the different aggregate sizes. Different superscript letters indicate significant difference ($P < 0.05$) among the different tea varieties. T: tea varieties; A: aggregate size. **, * indicate significant at $P < 0.01$ and $P < 0.05$, respectively. (FD) *C. sinensis 'Fuding Dabaicha'*, (CC3) *Chuancha No.3*, (CM217) *Chuanmu No.217*, (CN) *Chuannong Huangyazao*.

Moreover, the trend of *MWD* and *GMD* in the soils planted with the different tea varieties were similar to those of the > 5 mm aggregates (Fig. 2). The *MWD* and *GMD* were 3.32 and 1.56, respectively, in the FD soil, which were significantly higher than those in the soils of CM217 (2.82, 1.00) and CN (2.87, 0.93).

Distribution of Al fractions in soil aggregates

In the soils planted with the four tea varieties, the concentrations of T-Al, Ws-Al, Ex-Al and Res-Al increased with decreasing aggregate sizes, reaching a maximum in the < 0.25 mm aggregates (Table 4). The concentrations of both Fe/Mn-Al and Or-Al decreased with decreasing aggregate sizes, with the highest concentrations in the > 5 mm aggregates.

The distributions of the Al fractions in the aggregates of the four tea varieties exhibited a trend similar to that of the bulk soils. Specifically, the highest and lowest concentrations of T-Al and Res-Al within all aggregate sizes were measured in the CC3 soil and CN soil, respectively. The Ws-Al and Ex-Al concentrations in the 5 – 2, 2 – 1, 1-0.5, 0.5 – 0.25 and < 0.25 mm aggregates of the FD and CC3 soils were significantly higher than those in the CN soil. The Fe/Mn-Al concentrations among all aggregate sizes of the FD soil and CC3 soil were significantly higher than those in the CM217 and CN soils. The concentrations of Or-Al were FD > CN > CM217 > CC3 in the > 5, 5 – 2, 2 – 1 and 1-0.5 mm aggregates.

Accumulation and loading of Al fractions

The accumulation of T-Al, Ws-Al, Ex-Al and Res-Al increased with decreasing aggregate sizes in the soils of the different tea varieties, reaching a maximum in the < 0.25 mm aggregates (Fig. 3). The accumulation factor (AF) values for T-Al, Ws-Al, Ex-Al and Res-Al exceeded 1.0 in the < 0.25 aggregates. Both the FD (1.18) soil and CC3 (1.18) soils exhibited a higher AF value for T-Al and Res-Al in the < 0.25 mm aggregates compared with the CN (1.04) soil. There were no significant differences among the soils planted with the four tea varieties with respect to the AF values for Ws-Al and Ex-Al in the < 0.25 aggregates. In contrast with the T-Al, Ws-Al, Ex-Al and Res-Al, the AF values for Fe/Mn-Al and Or-Al exceeded 1.0 in the > 5 mm aggregates. The FD soil exhibited a higher AF value for Fe/Mn-Al in the > 5 mm aggregates compared with CM217, and the inverse trend was observed for Or-Al.

The > 5 mm and < 0.25 mm aggregates were the predominant sinks for Al, ranging from 7.64–39.89% (Fig. 4). Moreover, the CN soil exhibited a higher loading of Al fractions within the < 0.25 mm aggregates compared with the FD and CC3 soils. In contrast, except for Ws-Al, the FD soil exhibited a higher loading of Al fraction within the > 5 mm aggregates compared with the CN soil. Nevertheless, no obvious differences were observed between the soils of the four tea varieties with respect to the loading of Al fractions within the 1-0.25 mm aggregates.

Discussion

Differences in Al fractions in soils of different tea varieties

T-Al in soil plays crucial roles in influencing tea quality (Kopittke et al. 2016), and the T-Al concentration in soils is positively related to the mass fraction of tea polyphenols (Tolrà et al. 2020). Peng et al. (2018) reported a variation in soil T-Al concentrations among tea varieties. Consistent with this study, our results showed that T-Al concentrations in the soils of four tea varieties were rank as CC3 > FD > CM217 > CN. This result is consistent with the findings of Lou (2011) and Huang (2015), who reported that the polyphenol concentrations in the leaves of CC3 (19.27%) was much higher than that in CN (15.83%). T-Al occurs in soils as Ws-Al, Ex-Al, Fe/Mn-Al, Or-Al and Res-Al; in this study, Res-Al was the main fraction and accounted for 98.53–98.75% of the T-Al. This result indicated that the high Res-Al concentration determined the high T-Al concentration in the soil of CC3 (Xie et al. 2007).

Although the Ws-Al, Ex-Al, Fe/Mn-Al and Or-Al fractions together accounted for 1.25–1.47% of the T-Al, these Al fractions have different bioavailabilities and are in dynamic equilibrium with one another, collectively affecting the activity of Al in soils (Pei et al. 2014). As the most labile forms of Al, Ws-Al and Ex-Al correspond to the bioavailability of Al (Frankowski et al. 2014). Many studies have confirmed that Ws-Al and Ex-Al concentrations are influenced by pH and exogenous Al in soils (Chen et al. 2018; Palleiro et al. 2018). A decrease in pH can cause Ex-Al to be released into the soil solution. The addition of exogenous Al can lead to an increase in the Ex-Al concentration in the soil (Chen et al. 2018). Therefore, the lower pH and higher Al concentrations in the litter resulted in the higher Ex-Al concentration found in the CC3 soil compared with the other tea varieties soils. This higher Ex-Al concentration also explained the enrichment of Ws-Al in the soil of CC3 because Ws-Al and Ex-Al exhibit similar behavior patterns in soils (de Campos et al. 2022). This finding is consistent with previous studies. Wang et al. (2016) also confirmed that soil Ws-Al showed a significant positive correlation with soil Ex-Al. Although the availabilities of Or-Al and Fe/Mn-Al are lower than those of Ws-Al and Ex-Al (Hobara et al. 2016), they are potential reservoirs for Al in soils (Pei et al. 2014). It has been recognized that Al tends to bind to organic matter, forming strong insoluble complexes. The FD soil had a higher Or-Al concentration due to its higher organic matter concentration in relation to the soils associated with the other tea varieties (Table 1). Taken together, these results indicated that the FD soil had a higher potential reservoir of Al, which poses a risk for the presence of excess Al in the soil. The CC3 soil had higher T-Al and bioavailable Al concentrations, which may affect the quality and accumulation of Al in the associated tea. It is necessary to develop and implement appropriate management strategies to prevent increases in T-Al, Ws-Al and Ex-Al concentrations in the CC3 soil and to prevent an increases in the Or-Al concentration in the FD soil.

Differences in aggregate composition and aggregate-relevant Al fractions in soils of different tea varieties

Soil aggregate formation and its holding capacity for Al play vital roles in the Al fraction distribution of soils planted with different tea varieties. In this study, the > 5 mm and < 0.25 mm aggregates were the dominant fractions, consistent with a report by Li et al. 2015, indicating that the > 5 mm and < 0.25 mm aggregates had higher stabilities than the other aggregate sizes. The content of the > 5 mm and < 0.25 mm aggregates vary among tea varieties (Yang et al. 2019). Consistent with this study, our study also showed a significant difference in the soil aggregate composition among the different tea varieties, and the FD soil had the highest proportion of > 5 mm aggregates and the lowest proportion of < 0.25 mm aggregates. Many studies have confirmed that differences in aggregate formation are related to changes in divalent cations in soils and differences in the amount of tea litter (Jiang et al. 2010; Wang et al. 2018). As aggregate-cementing materials, divalent cations can form cationic bridges with clay particles and organic matter (Jiang et al. 2010; Wang et al. 2018), and litter decomposition can provide organic cement to flocculate silt and clay particles as well as free microaggregates (Rodrigues et al. 2019; Šimanský et al. 2014), ultimately promoting the formation of macroaggregates. In this study, higher divalent cation concentrations and litter quantities caused the formation of macroaggregates in the FD soil (Table 1) and resulted in the stabilization of soil structure.

The holding capacity of soil aggregates with respected to the Al fractions can explain the differences in the Al fraction distribution in the soils planted with the four tea varieties. The < 0.25 mm aggregates were key factors that determined the changes in the T-Al, Ws-Al and Ex-Al concentrations in the different soils, which was because the smaller aggregates had a stronger holding capacity for soil T-Al, Ws-Al and Ex-Al in this study. Consistent with a report by Yang et al. (2019), microaggregates favored the labile forms of Al due to their large surface areas and high clay mineral and organic matter contents. The CC3 soil had a greater T-Al, Ws-Al and Ex-Al holding capacity in the < 0.25 mm aggregates, which was the key factor influencing the high

T-Al, Ws-Al and Ex-Al concentrations in the bulk soil of CC3. In contrast, the > 5 mm aggregates were the key factors that determined the changes in the Fe/Mn-Al and Or-Al concentrations in the soils planted with the four tea varieties because Fe/Mn-Al and Or-Al were concentrated in the > 5 mm aggregates. The Or-Al fraction of soils is related to organic matter. With an increase in the organic matter concentration of soils, the amount of negative charge provided by organic acids relative to the total negative charge on the colloidal surface of soils increases correspondingly (Leelamanie et al. 2010). The functional groups of organic acids combine with Al^{3+} in soils to increase the concentration of Or-Al (Cory et al. 2015; Martins et al. 2013). Consistent with these studies, Or-Al tended to accumulate in the > 5 mm aggregates due to the higher organic matter concentration in these aggregates (Fig S1). In short, the high T-Al, Ws-Al and Ex-Al concentrations in the CC3 soil were due to the high holding capacity of the < 0.25 mm aggregates with respect to T-Al, Ws-Al and Ex-Al. The high Or-Al concentration in the FD soil was due to the formation of large aggregates (> 5 mm) and the high holding capacity of these aggregates for Or-Al. These finding support our first hypothesis.

Accumulation and loading of Al fractions in soil aggregates of different tea varieties

The metal loading of grain size fractions (GSF) is an important indicator for evaluating Al distribution in soil aggregates and is closely related to the mass proportion and Al concentration of each particle size (Sutherland et al. 2003). In this study, the FD soil exhibited a higher loading of Or-Al within the > 5 mm aggregates compared with the other aggregate sizes, indicating that the Or-Al concentrations in the FD soil were determined by the > 5 mm aggregates. The AF values of Or-Al in the > 5 mm aggregates exceeded 1.0, indicating that the > 5 mm aggregates had a stronger enrichment capacity with respect to Or-Al. The solubilization of Al is controlled by reactions of Or-Al, and the weak holding capacity of < 0.25 mm aggregates for Or-Al leads to Or-Al decomposition and transformation into labile forms, such as Ws-Al and Ex-Al (Eimil-Fraga et al. 2015). Therefore, the high Or-Al concentration in the FD soil could produce high quantities of labile Al, which depend on the high proportion of the > 5 mm aggregates in this soil and the greater holding capacity of the > 5 mm aggregates for Or-Al.

The CC3 soil exhibited a higher loading of T-Al, Ws-Al and Ex-Al within the > 5 mm and < 0.25 mm aggregates compared with the other aggregate sizes, indicating that the T-Al, Ws-Al and Ex-Al concentrations in this soil were determined by the > 5 mm and < 0.25 mm aggregates. The AF values for T-Al, Ws-Al and Ex-Al exceeded 1.0 in the < 0.25 aggregates. This indicates that the smaller size aggregates had stronger enrichment capacities for soil T-Al and bioavailable Al due to their greater specific surface areas (Huang et al. 2014; Jiang et al. 2015). Therefore, the stronger enrichment capacity of the < 0.25 mm aggregates with respect to T-Al and bioavailable Al was more conducive to the accumulation of T-Al and bioavailable Al in the CC3 soil. Although the > 5 mm aggregates had distinctly lower T-Al and bioavailable Al concentrations than the other aggregate sizes, the > 5 mm aggregates had a strong contribution to the accumulation of T-Al and bioavailable Al because of their high abundance in the CC3 soil. The results from this study validate the importance of the > 5 mm and < 0.25 mm aggregates for the accumulation of T-Al and bioavailable Al in the CC3 soil. Overall, the high T-Al and bioavailable Al concentrations in the CC3 soil were mainly due to the high

Al preservation capacity of the > 5 mm and < 0.25 mm aggregates. These results are consistent with our second hypothesis.

Conclusions

The FD soil had the highest concentration of Or-Al, and the CC3 soil had the highest concentrations of T-Al, Ws-Al and Ex-Al among the four tea varieties. These differences were related to differences in the soil aggregate composition and the aggregate-associated Al fractions of these soils. The high Or-Al concentration in the FD soil was related to the high proportion of > 5 mm aggregates and the high Or-Al concentration in the > 5 mm aggregates. Among the four tea varieties, the CC3 showed the highest T-Al, Ws-Al and Ex-Al concentrations, which were related to the high concentrations of T-Al, Ws-Al and Ex-Al in the > 5 mm and < 0.25 mm aggregates. Therefore, the formation of aggregates and the high Al preservation capacity of these aggregates determined the high Al concentrations of the FD soil and CC3 soil. This provides evidence for developing appropriate management plans to prevent an increase in Al concentrations in the soils of CC3 and FD by adjusting the soil aggregate composition and the Al concentration of the aggregates.

Declarations

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Author contributions

Jia Yang, Xizhou Zhang, Zicheng Zheng, and Tingxuan Li designed the paper. Jia Yang developed and implemented the research plan. All authors contributed to the writing of the manuscript, with writing led by Jia Yang.

Confict of interest

The authors declare that they have no conflicts of interest.

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Figures

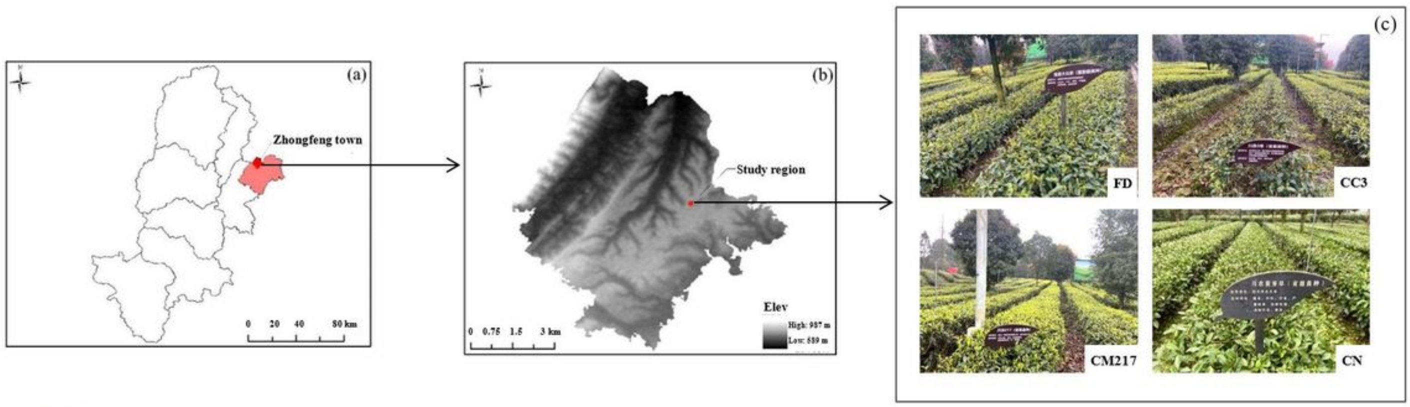

Fig.1

Figure 1

Location of study region. (a) is the location of Zhongfeng town in Ya'an; (b) is the location of Zhongfeng ecological tea garden (study region) in Zhongfeng town; (c) is the distribution map of four tea varieties. (FD) *C. sinensis 'Fuding Dabaicha'*; (CC3) *Chuancha No. 3*; (CM217) *Chuanmu No. 217*; (CN) *Chuannong Huangyazao*.

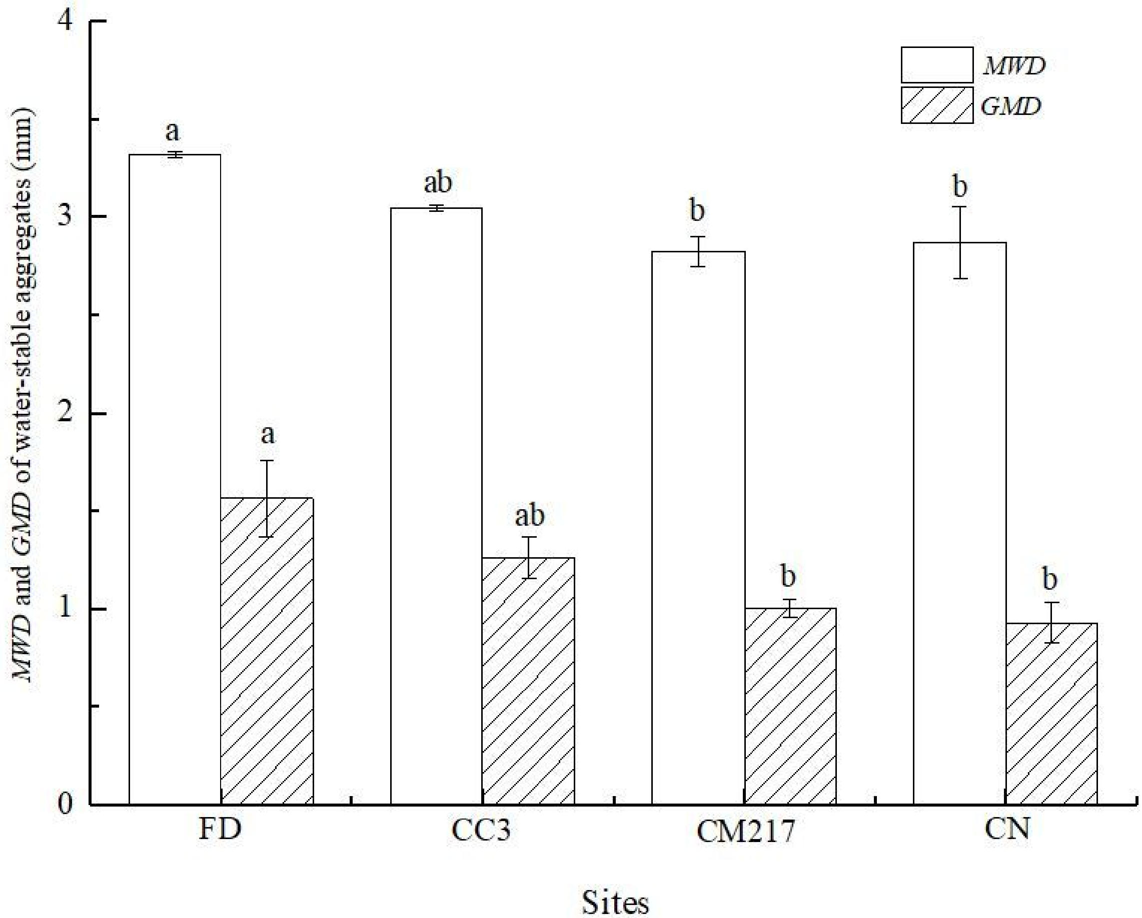

Figure 2

Mean weight diameter (*MWD*) and geometric mean diameter (*GMD*) in the soil water-stable aggregates of different tea varieties. Means of three replicates are reported. Error bars represent standard deviations. Different letters indicate significantly different (P<0.05) among different tea varieties. (FD) *C. sinensis 'Fuding Dabaicha'*; (CC3) *Chuancha No. 3*; (CM217) *Chuanmu No. 217*; (CN) *Chuannong Huangyazao*.

Figure 3

Accumulation factor of Al fractions in water-stable aggregates of different tea varieties. Means of three replicates are reported. Error bars represent standard deviations. Different letters indicate significantly different (P<0.05) among different tea varieties. (FD) *C. sinensis 'Fuding Dabaicha'*; (CC3) *Chuancha No. 3*; (CM217) *Chuanmu No. 217*; (CN) *Chuannong Huangyazao*.

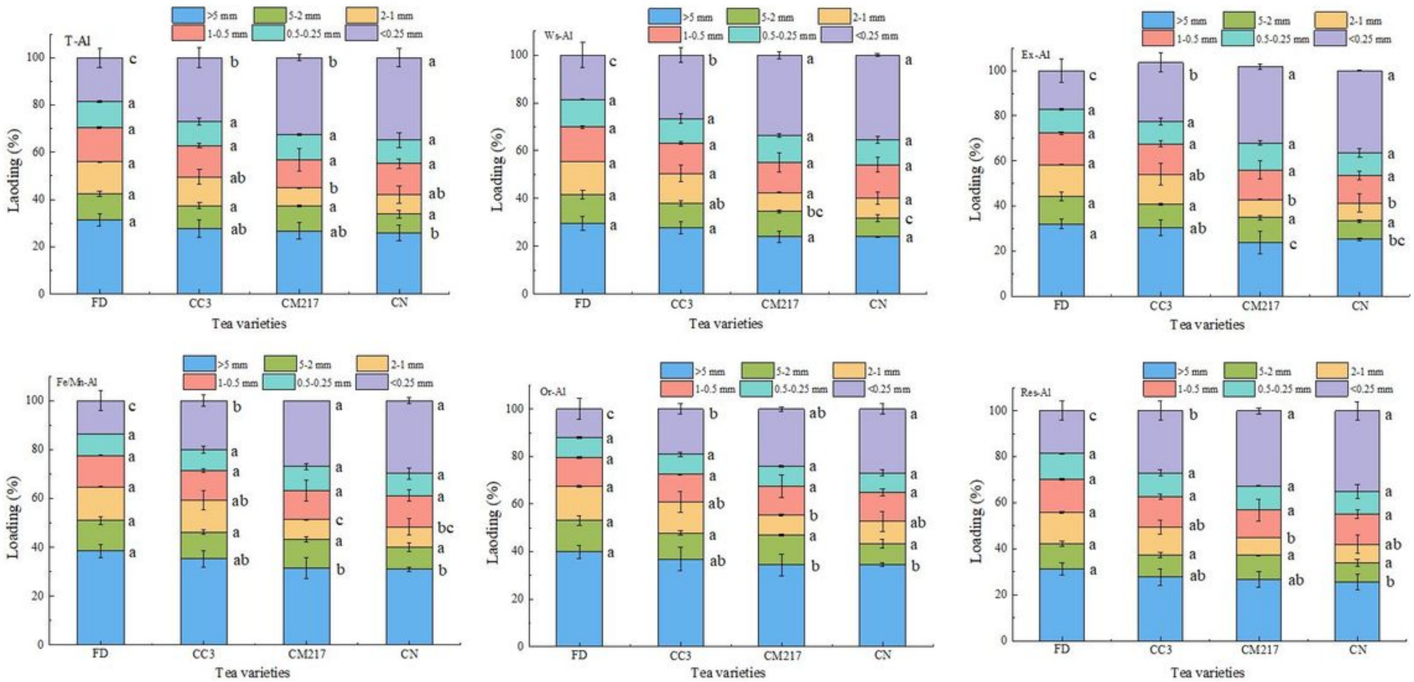

Figure 4

Loading (%) of Al fractions in water-stable aggregates of different tea varieties. Means of three replicates are reported. Error bars represent standard deviations. Different letters indicate significantly different (*P*<0.05) among different tea varieties. (FD) *C. sinensis 'Fuding Dabaicha'*; (CC3) *Chuancha No. 3*; (CM217) *Chuanmu No. 217*; (CN) *Chuannong Huangyazao*.

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