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INFLUENCE OF NATURAL REGENERATION ON FRACTAL FEATURES OF RESIDUE MICROAGGREGATES IN BAUXITE RESIDUE DISPOSAL AREAS

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ABSTRACT

Bauxite residue is often physically degraded, which limits vegetation establishment on the disposal areas. Microaggregate stability is an important physical property due to its significant effect on erosion and surface runoff, however this is rarely reported for bauxite residue. Native plant encroachment on a bauxite residue disposal area in Central China has revealed that natural regeneration may ameliorate the residue and help to support plant growth. Residue samples from five different disposal ages were collected to determine microaggregate stability and identify their fractal features. Following natural regeneration, the aggregate fraction 250-50 μm increased significantly from 27.4% to 40.3%, whilst the silt+clay size aggregate fraction decreased from 58.4% to 30.7%. With increasing disposal age, the residue clay dispersion ratio (CDR) ranged from 7.7% to 22.5%, whilst aggregated silt and clay (ASC) ranged from 15.3% to 19.0% indicating a stable microaggregate structure. The single-fractal dimension (D) of the residues for different disposal ages varied from 2.2 to 2.4. The high pH and salinity of bauxite residue indicated a high value of single-fractal dimension. The multi-fractal parameters of residue microaggregates, including capacity dimension (D_0), information dimension (D_1) and information dimension/capacity dimension (D_1/D_0) decreased which resulted in homogeneity following natural regeneration. Correlation analysis revealed that both single- and multi-fractal dimensions had significant correlations with residue microaggregate stability. Our results suggested that natural regeneration may improve microaggregate stability of bauxite residue, and fractal parameters of residue microaggregates may be used to describe residue microaggregate stability and the physical condition of bauxite residue.

KEY WORDS: Bauxite residue disposal area; bauxite residue; natural regeneration; microaggregate stability; fractal features

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INTRODUCTION

Residues from mineral ore processing are disposed on land in large residue disposal areas which may eventually create a series of ecological and environmental issues (Smart *et al*., 2016; Wu *et al*., 2016). In the aluminum industry, bauxite residue is an alkaline solid by-product generated when alumina is extracted from bauxite ore by the Bayer process (Goloran *et al*., 2016; Kong *et al*., 2017a). The global inventory has reached 3.4 billion tons, with an annual increase of 120 million tons (Xue *et al*., 2016a; Kong *et al*., 2017b). Large volumes of bauxite residue are deposited in bauxite residue disposal areas which cause potential environmental risks, as these bare areas are sensitive to erosion by wind and water, and can be regarded as a potential source of contamination due to their high alkalinity and salinity (Gelencsér *et al*. 2011; Ruyters *et al*. 2011). In situ rehabilitation and revegetation may however stabilize the residue surface and minimize wind erosion (Courtney *et al*., 2009; Kaur *et al*., 2016; Schmalenberger *et al*., 2013). Its poor physical structure is nevertheless a major limitation to support plant growth (Liu *et al.*, 2013; Zhu *et al.*, 2016a). Residue particle sizes range from 2 to 2000 μm and 60-80 % exist as <20 μm (Xue *et al*., 2016b). Jones *et al.* (2011) reported that addition of organic waste may influence aggregate size distribution and increase the proportion of water-stable aggregates. Zhu *et al*. (2016b) found that natural regeneration may improve the physical condition and aggregate stability of bauxite residue.

Soil aggregate stability is one of the most important properties in soils and affects water erosion, soil aeration, nutrient recycling and biological activity, as well as plant growth (Cerdà 2000; Le Guillou *et al.*, 2012; Moncada *et al.*, 2013). Physical forces, chemical bonds and biological agents may drive the aggregation processes of soil particles (Yagüe *et al*., 2016; Lehmann & Rillig, 2015). Microaggregate stability is an important soil property which is usually used to determine soil erosion resistance (Wang *et al*., 2016). A better understanding of microaggregate formation is essential to maintain structural stability in soils. Several major binding agents such as clay minerals, organic carbon and polyvalent ions have significant effects on colloid flocculation (Zhou *et al.*, 2005). Barbosa *et al.* (2015) observed a cementation effect by organic carbon from poultry manure applications and clay flocculation enhancing aggregation. Igwe *et al.* (2009) discovered that oxalate and pyrophosphate extractable iron-aluminum oxides may act as aggregation agents to colloidal stability; organic carbon had acted in association with the oxides as a linkage with clay particles and polyvalent cations to enhance aggregate stability. Virto *et al.* (2008) concluded that stable microaggregates were formed within the silt-size fraction and organic carbon was stored by adsorption and entrapment of fine organic residues.

Soil structure is related to the size, shape and stability of soil aggregates (Aksakal *et al*., 2016; Ahmadi *et al*., 2011). Microaggregate stability depends on the size distribution of microaggregates and several procedures have been proposed for characterizing aggregate size distribution. Fractal theory is mainly used to analyze the relationship between local and overall irregular broken complex images and structural geometry under different scales. The concept of fractal dimension was proposed to provide a quantitative description for irregular shapes (Jing *et al*., 2016). Fractal theory has been widely applied in soil science to quantify and estimate aggregate size distribution of soils (Kolay & Kayabali, 2006). Fractal dimension reveals the difference between particle size distribution and related physical properties (Wei *et al.*, 2016; Wang *et al.*, 2015). Gao *et al.* (2014) suggested that fractal dimension could be regarded as a considerable and reliable parameter to reflect variations in soil properties. High values represent aggregates dominated by fine fragments, whilst low values represent large fragments. Many researchers have used fractal dimension to predict soil particle size distribution or the size distribution of water-stable aggregates (Peng *et al*., 2014).

With the development of soil fractal theory, the limitation of single-fractal dimension has been stressed to describe soil particle size distribution. In order to obtain more detailed information of soil structure, multi-fractal theory was introduced to soil science (Li *et al.*, 2016). Rodríguez-Lado & Lado (2016) found that particle size distribution behaved as multi-fractals, with scaling properties varying in different soil samples, whilst values of fractal dimension may be related to the degree of evolution of the soils. Peng *et al.* (2014) found that the single- and multi-fractal parameters could describe soil particle size distribution and the influences of soil structure effectively. There are, however, few studies focusing on multi-fractal parameters of microaggregate size distribution.

This work focuses on an alumina refinery in Central China. The inventory of bauxite residue is an estimated 35 million tons, which is currently increasing by approximately 2.2 million tons per annum (Zhu *et al*., 2016b). Bauxite ore is discharged in hot NaOH by the Bayer processes and the residues are pumped to the disposal areas using the dry stacking method. Spontaneous vegetation colonization over the past 20 years at the study site may reveal that natural weathering processes ameliorate the residue substrate and support plant growth. Natural regeneration also enhances the proportion of water-stable aggregates and resistance to erosion (Zhu *et al*., 2016c). As microaggregate stability is used to predict soil surface erosion (Wang *et al.*, 2016), this study focus on 1) the effect of natural weathering processes on microaggregate stability of bauxite residue; 2) to evaluate microaggregate size distribution in bauxite residue by fractal parameters; 3) to investigate whether fractal parameters may be used as an indicator to evaluate microaggregate stability of bauxite residue.

MATERIALS AND METHODS

Soil Sampling

Residue samples were collected from a disposal area in Central China. The climate is temperate continental monsoon, with a mean annual daily temperature of 12.8ºC-14.8ºC and average precipitation ranging from 600 mm to 1200 mm per year.

According to ecological field investigations, five different zones related to disposal age were selected during August to September 2014. These included (a) 1-year-old bauxite residue (R1), (b) 4-year-old bauxite residue (R2), (c) 6-year-old bauxite residue (R3), (d) 10-year-old bauxite residue (R4), and (e) 20-year-old bauxite residue (R5). Each zone was approximately 1500 m^2. Within the zones, natural colonization only occurred in R5. For each zone, five random points, taken within 100 m x 100 m, were designated as the replicates. For each sampling point, the residues were sampled with an auger to a depth of 20 cm. The samples were then stored in polyethylene bags, returned to the laboratory, air dried at room temperature for two weeks and then subsequently passed through a 2 mm sieve prior to analysis.

Physical and chemical analysis

Mechanical composition of residue samples were analyzed using a Malvern Mastersizer 2000 (Malvern Instruments Ltd., UK) (Santini and Fey, 2013). pH and electrical conductivity (EC) of residue samples were determined in 1:5 solid/solution extracts. Exchangeable Ca^{2+}, Mg^{2+}, K^+, and Na^+ were extracted with 1 M ammonium acetate and analyzed by ICP-AES (Jones *et al.*, 2011). Exchangeable sodium percentage (ESP) was calculated as the percentage of exchangeable Na^+ in the total exchangeable cations. The total contents of Ca, Mg, K and Na in bauxite residue were determined after microwave digestion using HF, HCl and HNO_3 and analysed by ICP-AES (Jones *et al.*, 2011). Total organic carbon was measured by the low-temperature external-heat potassium dichromate oxidation colorimetric method (Zhu *et al.*, 2016a). Chemical phases of residue samples were determined by X-ray powder diffraction (XRD) on a Bruker D8 discover 2500. XRD patterns were collected from 10° to 80° at a step size of 0.04° 2θ with a scan rate of 1° 2θ per minute and analysed using PANalytical analysis package (Zhu *et al*., 2017).

Microaggregate Stability Analysis

Laser sizing (for the <0.25 mm fraction) was used to determine particle size distribution of residue microaggregates (Santini & Fey, 2013). In this method, 10 g of air-dried residue samples were placed in a 0.25 mm sieve. The residue samples were then immersed in distilled water and oscillated for 24 h using an end-over-end shaker with a rate of 200 cycles per minute. Particle size distribution of the <0.25 mm aggregates was determined using a Malvern Mastersizer 2000. In order to observe residue microaggregate distribution characterization under natural regeneration, micro-morphological studies of the residue microaggregates from R1 and R5 were examined using a FET Quanta-200 scanning electron microscope (SEM), equipped with energy dispersive X-ray spectroscopy. The specimen was sputter coated with a layer of gold prior to examination (Zhu *et al.*, 2016b).

Water-dispersible clay (WDC) and water-dispersible silt (WDSI) were determined as the proportion of clay and silt in suspension in the distilled water. Clay dispersion ratio (CDR) and aggregated silt+clay indices (ASC) were selected as the two indicators to measure microaggregate stability of bauxite residue. Clay dispersion ratio (CDR) was determined as the following equation (Cammeraat & Imeson, 1998):

$$\mathrm{CDR}(\%) = \frac{\%\mathrm{clay} + \%\mathrm{silt}(\mathrm{water\ dispersed})}{\%\mathrm{clay} + \%\mathrm{silt}(\mathrm{calgon\ dispersed})} \times 100 \tag{1}$$

This is defined as the percentage ratio of clay+silt (<0.02 mm) obtained from both distilled water and sodium hexametaphosphate (calgon) dispersed residue samples. The value of ASC was negatively correlated to aggregate stability (Mbagwu & Auerswald, 1999).

Aggregated silt and clay (ASC) was calculated using the following equation:

$$\mathrm{ASC}(\%) = (\%\mathrm{clay} + \%\mathrm{silt})(\mathrm{calgon\ dispersed}) - (\%\mathrm{clay} + \%\mathrm{silt})(\mathrm{water\ dispersed}) \tag{2}$$

A higher ASC value indicates greater microaggregate stability (Monreal *et al*., 1995).

Calculation of Single-fractal Dimension (D)

The power-law relationship between either number-diameter, mass-diameter or bulk density-diameter of soil aggregates are always used to determine the fractal dimension of soil aggregates. Here, according to Tyler & Wheatcarft (1989), mass-diameter of residue aggregates was selected to calculate the fractal dimension of microaggregates, designated as D, as follows:

$$D = 3 - \lg(W_i / W_o) / \lg(\bar{d}_i / \bar{d}_{max}) \tag{3}$$

where D is the mass fractal dimension; W_i is the cumulative mass of the <d_i residue aggregates; W_o is the total mass of the residue aggregates; $\bar{d}_i$ is the mean diameter of aggregates in adjacent particles and $\bar{d}_{max}$ is the mean diameter of the largest aggregates.

Calculation of Multi-fractal Parameters

In this study, the measurement interval of the laser particle size analyzer (I=[0.01 μm, 250 μm]) was considered as the residue microaggregate size volume percentages obtained from the previous results. The miacroaggregate size interval is divided into 74 subintervals I_i=[φ_i, φ_{i+1}], i=1, 2,…, 74. Based on standard microaggregate-size division methods, log(φ_{i+1}/φ_i) is the constant following the measurement interval of *I*=[0.01, 250]. In order to build a new measurement of the multi-fractal method, ψi=log(φi/φ_1) (i=1, 2, …, 74) was created to form a new dimensionless interval of J=[0, 4.40], which had 74 subintervals of equal length, Ji=[ψ_i,ψ_{i+1}] (i=1, 2, …, 74). In the interval J, ε was defined as 2^k same size subintervals, $\varepsilon=4.4\times2^{-k}$. The value of k ranged from 1 to 6 to make sure that every subinterval contained at least one measured value (Peng *et al.,* 2014). Thus, the multi-fractal parameters including capacity dimension (D_0), information dimension (D_1), correlation dimension (D_2) and information dimension/capacity dimension (D_1/D_0) were calculated as the following equations (Ahmadi *et al.*, 2011):

$$D(q) \approx \lim_{\varepsilon \to 0} \frac{1}{q-1} \times \frac{\log\left[\sum_{i=1}^{n(\varepsilon)} u_i(\varepsilon)^q\right]}{\log \varepsilon} \qquad q \neq 1 \qquad (4)$$

$$D_1 \approx \frac{\sum_{i=1}^{n(\varepsilon)} u_i(\varepsilon) \log u_i(\varepsilon)}{\log \varepsilon} \qquad q = 1 \qquad (5)$$

The value of q varied between −10 and 10 with a step size of 1. The multi-fractal spectrum of the residue microaggregate size distribution were determined by D(q). D_0 indicated the span of the residue microaggregate size distribution and the larger D_0 value representing a wider range; D_1 indicated the irregular degree of the residue microaggregate size distribution and the higher D_1 representing a higher level of dispersion in microaggregate size distribution; D_1/D_0 can measure the degree of heterogeneity of microaggregate size distribution (Peng *et al*., 2014).

Data Analysis

All analyses were performed in quintuplicate. The data were statistically treated with Microsoft Excel 2003, SPSS version 19.0 and Origin 8.0. A two-way ANOVA, followed by Tukey's post hoc test was used to determine the interaction between fragment size range and residue disposal ages. Chemical properties of bauxite residue samples with different chronosequences were individually determined using one-way ANOVA followed by Tukey's post hoc tests. In the case of no homogeneity, Dunnett's T3 test was performed. Bivariate correlation analyses were used to determine the relationships between fractal parameters and residue microaggregate size distribution. All figures were constructed using Origin 8.0.

RESULTS AND DISCUSSION

Particle Size Distribution of Residue Microaggregates

The particle size distribution of microaggregates is shown in Table Ⅰ. The main fraction, <0.02 mm aggregates, accounted for more than 55% of the total microaggregate weight in newly stacked residue (R1). Microaggregate fractions in the fresh residue (R1) decreased in the following order, 250-50 µm > 10-5 µm > 20-10 µm > 5-2 µm > 50-20 µm > <2 µm. With increasing disposal age, the aggregate fraction 250-50 µm, increased significantly from 27.4% to 40.3%, whilst the clay-size aggregate fraction decreased gradually. Microaggregate fractions which had been disposed for 20 years (R5) decreased in the following order, 250-50 µm > 50-20 µm > 20-10 µm > 10-5 µm > 5-2 µm > <2 µm. With increasing disposal age, the fine particles of bauxite residue became coarser and the silt+clay size (<0.02 mm) aggregate fraction effectively decreased.

SEM images of residue microaggregates from R1 and R5 are shown in Fig. 1. The residue microaggregate in R1 contained numerous amorphous substances and fine particles. Compared to R1, the microaggregate in R5 consisted of a great number of larger aggregates. With increasing disposal age, the residue microaggregates had a denser structure and the particles were distributed uniformly. Total sodium content decreased from 9.27% to 1.07%, whilst total calcium content increased from 13.89% to 27.88% (Fig. 1), which suggested that natural weathering processes decreased sodium but increased calcium content. The result from Fig. 1 was consistent with the variation trend of calcium and sodium content in Table Ⅱ.

Natural weathering processes may have a positive effect on particle aggregation; the finer particles aggregating to form larger particles (Santini & Fey, 2013). Climate affects soil aggregation through alterations in temperature and moisture regimes and wet-dry and freeze-thaw cycles, which can re-orientate particles and improve aggregation (Singer *et al*., 1992). Weathering alters materials, which are translocated within the soil through leaching, eluviation, and illuviation resulting in horizonation (Garcia-Franco *et al*., 2015; Zhou *et al.*, 2017). Plant roots and their rhizospheres have positive effects on soil aggregation. Roots realign soil particles and release exudates, which result in physical, chemical and biological alterations that influence aggregation (Rillig *et al*., 2001). Courtney *et al.* (2009) revealed that gypsum and spent mushroom compost application may decrease microaggregate breakdown and hence the dominance of less erodible aggregates. Courtney *et al.* (2013) found that addition of gypsum and compost produced a significant decrease in clay- and silt-size particles, whilst mean particle size (<53 μm) was lowest in the unamended residues. Natural weathering processes may ameliorate physical and chemical properties of bauxite residue, which lead to clay-size particles flocculating and the formation of more stable aggregates.

Microaggregate Stability of the Residues

Colloidal stability indices of the residues are presented in Fig. 2. Water-dispersible clay (WDC) which was used to estimate microaggregate instability, ranged from 0.64% to 2.04%, whilst water-dispersible silt (WDSI), also used to estimate instability, ranged from 30.72% to 58.39%. A combination of WDC and WDSI gave values of between 31.4% and 58.4%. Clay dispersion ratio (CDR) of the residues ranged from 7.7% to 22.5%, and aggregated silt and clay (ASC) ranged from 15.3% to 19%. Following natural disposal processes, WDC, WDSI, CDR and ASC of the residues stacked for 20 years (R5) increased by 218.75%, 90.07%, 192.21% and 24.18%, respectively.

The WDC, WDSI, CDR and ASC may be used to estimate the rate of soil dispersibility. A high WDC and dispersion indices have negative implications for the soil environment in terms of water and wind erosion (Virto *et al.*, 2008). With increasing disposal age, WDC, WDSI and CDR significantly decreased, whilst ASC increased indicating that natural stacking processes may improve microaggregate stability; the finer particles may combine together to form larger particles due to binding agents related to physical and chemical properties of the residues (Zhu *et al.*, 2016c). Plant growth and root penetration may also have positive effects on particle aggregation and stability as the residues stacked for 20 years (R5) had improved colloidal stability compared to the other locations.

Stability of microaggregates, as opposed to its dispersion, is a very important soil property that regulates soil degradation. Several major binding agents including clay content, organic carbon and electrolytes had significant effects on microaggregate stability. Clay content, organic carbon, pH and exchangeable cations were selected to identify correlational relationships with colloidal stability indices. The selected physical and chemical properties were determined in a previous study (Zhu *et al*., 2016c). Silt and clay contents ranged from 48.8%-23.8% and 5.9%-1.5% respectively. With increasing disposal age, pH and EC were significantly reduced. Total organic carbon (TOC) content ranged from 5.7-10.8 g/kg. Exchangeable Ca and Na varied regularly but with opposite trends (Table Ⅱ).

As natural weathering processes had a significant effect on bauxite residue mineral chemistry, the residue samples, including R1 and R5, were selected to investigate the variation in chemical phases (Figure 3). Slaked lime addition by the Bayer process resulted in the formation of calcium minerals including calcite ($CaCO_3$), hydrogarnet ($Ca_3Al_2(SiO_4)_x(OH)_{12-4x}$), and tri-calcium aluminate ($Ca_3Al_2(OH)_{12}$). Other alkaline minerals in the residue included sodalite ($Na_6Al_6Si_6O_{24}$ • [2NaOH or Na_2CO_3]) and cancrinite ($Na_6Al_6Si_6O_{24}$ • $2CaCO_3$). Following natural processes, sodalite, hydrogarnet and calcite decreased, as did pH. Reduction of pH resulted in the precipitation of $Ca(OH)_2$ but also leaching of NaOH by exchange reactions as the charged colloid (such as $Al(OH)_6^{3-}$) was a regulator for cation exchange (Kong *et al.*, 2017). As a result, exchangeable sodium percentage decreased. In addition, sodium ions could not be coordinated with negatively charged surfaces which led to the formation of alkaline dust, reduction on erosion resistance and a poor physical structure (Zhu *et al*., 2016d).

Linear regression analysis showed that the value of clay dispersion ratio (CDR) was positively correlated to clay content, pH and exchangeable Na^+ content (r=0.898, 0.943, and 0.826 respectively; P<0.01), but negatively correlated to exchangeable Ca^{2+} and total organic carbon content (r = -0.972 and -0.936, P<0.01) (Fig. 4). The value of aggregated silt and clay (ASC) was negatively correlated to clay content, pH and exchangeable Na^+ content (r = -0.903, -0.927, and -0.865, respectively; P<0.01), and positively correlated to exchangeable Ca^{2+} and total organic carbon content (r = 0.948 and 0.932 respectively, P<0.01) (Fig. 5). This indicated that high exchangeable Ca^{2+} content and low exchangeable Na^+ stimulated microaggregate flocculation, whilst the decrease in pH and the accumulation of organic carbon may have improved microaggregate stability. Courtney *et al.* (2009) established a field scale investigation to promote vegetation cover on bauxite residue, and found that spent mushroom compost and gypsum amendments decreased pH and ESP which positively impacted on microaggregate stability. Addition of Ca had a positive effect on flocculating clay particles, reducing mechanical dispersion and lowering exchangeable Na^+ content, thereby stabilizing microaggregates (Harris & Rengasamy, 2004). Pojasok & Kay (1990) reported that increasing organic carbon content stimulated particle aggregation.

Single-fractal Features of Residue Microaggregates

The fractal dimension of soil microaggregates may reflect the geometry parameters of soil aggregate structure, with higher clay content indicating the higher value of the fractal dimension (D). Under natural weathering processes, residue fractal dimension (D) was significantly affected (Fig. 6). The single-fractal dimension ranged from 2.2 to 2.4 and with increasing disposal age, microaggregate fractal dimension (D) decreased. R1 had a low proportion of 250-20 µm size aggregates and a high proportion of 10-2 μm aggregates, which resulted in a high fractal dimension value. Under natural soil forming processes, fine particle aggregation led to the decrease in single-fractal dimensions. Certainly, single-fractal dimension (D) was positively correlated with the proportion of 10-5 μm, 5-2 μm, and <2 μm sized microaggregates (r=0.859, 0.977, 0.991 respectively, $P<0.01$), but negatively correlated with the proportion of 250-50 μm and 50-20 μm sized microaggregates (r=0.876 and 0.761 respectively, $P<0.01$). There was no significant correlation between single-fractal dimension and the proportion of 20-10 μm sized microaggregates. Single-fractal dimension may be regarded as an important indicator to reflect aggregate structure of bauxite residue.

High alkalinity and salinity resulted in poor aggregate structure of the residue and clearly affected revegetation on disposal areas (Jones *et al.*, 2011). Zhu *et al.* (2016d) found that natural vegetation encroachment ameliorated residue physicochemical properties and stimulated aggregate stability. Courtney *et al.* (2013) investigated the physical condition of revegetated residue and found that gypsum and organic carbon decreased pH and ESP, which enhanced the proportion of water-stable aggregates, which supported plant growth. The related relationships between single-fractal dimension and pH, EC, and ESP are displayed in Fig. 7. The single-fractal dimension was positively correlated with pH, ESP, exchangeable Na^+ content and EC (r=0.935, 0.984, 0.859 and 0.912 respectively, $P<0.01$), but negatively correlated with exchangeable Ca^{2+} content (r=-0.968, $P<0.01$). It indicated that the single-fractal dimension of residue microaggregates may reflect the related physical and chemical properties of bauxite residue.

Multi-fractal Dimension of Residue Microaggregates

Multi-fractal spectrums of residue microaggregate size distributions between -10 and 10 at 1.0 lag increments for different disposal ages are presented in Fig. 8. The multi-fractal spectrums show a typical anti-S-decreasing function. The information entropy ($D_{(q)}$) of residue microaggregates decreased with increasing disposal age. Furthermore, in each residue sample, $D_0>D_1$ always existed, meaning that microaggregate size distribution with different disposal ages were not homogeneous or monofractal. Therefore, multi-fractal dimension analysis was essential.

The value of D_0, D_1 and D_1/D_0 decreased with increasing disposal age (Table Ⅲ). In residues which had been stacked for 20 years (R5), these values were nearly the lowest (D_0, D_1 and D_1/D_0 values of 0.942, 0.853 and 0.906, respectively), whilst for R1, these values were the highest (D_0, D_1 and D_1/D_0 values of 0.968, 0.891 and 0.920, respectively). Analysis of variance in the different residues showed that the multi-fractal parameters of R1 and R2 were significantly different ($P<0.05$).

The larger D_0 means a wider range of microaggregate size distributions. Nevertheless, the calculation of D_0 is based on the assumption that particle size distribution was homogeneous. The value of D_1/D_0 may make a quantitative description of the heterogeneous degree of soil particle size distribution. Miranda *et al.* (2006) pointed out that if the value of D_1/D_0 was closer to 1 this specified that the particle size distribution was more concentrated. The value of D_1/D_0 in the residue microaggregates ranged from 0.896 to 0.920. With increasing disposal age, the value of D_1/D_0 decreased indicating that natural weathering processes decreased the concentration of microaggregate distribution. Natural processes accumulate organic carbon over time and this may have ameliorated the high alkalinity and salinity in the residue, stimulating fine particle aggregation. A significant increase in the proportion of 250-50 μm residue microaggregates resulted in homogeneity with increasing disposal age.

Bivariate correlation analysis between multi-fractal parameters and residue microaggregate distribution showed that the value of D_0, D_1, D_1/D_0 was significantly correlated with the proportion of <2 μm microaggregates (r=0.915, 0.786 and 0.523 respectively, P<0.05). In addition, the value of D_1 was positively correlated with the proportion of 10-5 μm and 5-2 μm microaggregates (r=0.912 and 0.671 respectively, P<0.05). According to correlation analysis between multi-fractal parameters, D_1 was positively correlated with D_0 and D_1/D_0 (r=0.933 and 0.917 respectively, P<0.05). Multi-fractal parameters of residue microaggregate size distribution were mainly affected by the proportion of <10 μm microaggregates, especially the silt-sized (<2 μm) microaggregates.

Multi-fractal dimension of residue microaggregates was closely linked with related physicochemical properties. Residues which had been stacked for 20 years had a low pH and EC which led to a low multi-fractal dimension, whilst the newly stacked residue had a high pH and EC and a high multi-fractal dimension. This suggested that the multi-fractal parameters of microaggregates may reflect physical and chemical properties and may be used as an effective indicator to characterize alkalinity and salinity of bauxite residue. According to multiple linear models between multi-fractal parameters and the related properties of bauxite residue, the following equations were obtained:

$$D_0=1.01926+0.0018x_1-0.0079x_2-0.00376x_3+2.06\times10^{-4}x_4 \quad (6)$$

$$D_1=0.91468+0.00325x_1-0.00476x_2+1.9669\times10^{-4}x_3-3.91826\times10^{-4}x_4 \quad (7)$$

$$D_1/D_0=0.88054+0.00498x_1-0.0019x_2+0.00231x_3+6.58484\times10^{-5}x_4 \quad (8)$$

where x_1 is the content of TOC, x_2 is the value of pH, x_3 is the value of EC, and x_4 is the value of ESP. This demonstrated that organic carbon content and pH were the main properties influencing the values of multi-fractal dimension of residue microaggregates.

Relationship between Microaggregate Stability and Fractal Parameters

Microaggregate stability is usually used to estimate or predict soil erosion and surface runoff (Wang *et al.*, 2016). Bauxite residue has poor physical structure to resist water erosion and support revegetation. Zhu *et al.* (2016d) discovered that following natural weathering processes, the erodibility factor of the residue decreased indicating improved resistance to erosion. Correlation analysis showed that single- and multi-fractal parameters were significantly correlated to microaggregate stability indicating that fractal parameters of residue microaggregate distribution may reflect microaggregate stability. The fractal dimension of microaggregate size distribution may exhibit variation in microaggregate size distribution. The high value fractal parameters indicated a dense physical structure and poor erosion resistance.

Tang *et al.* (2013) revealed a significant negative relationship between fractal dimension and soil microaggregate content (<0.25 mm) in karst rocky desertification areas and suggested that fractal dimension could be used as a reliable indicator of soil quality. A small fractal dimension value for granular structure indicated a stable soil structure. Ahmadi *et al.* (2011) found that both number- and mass-based fragmentation fractal dimension may describe the aggregate size distribution and estimate splash and inter-rill soil erosion. In our study, the single-fractal dimension (D) of microaggregate distribution was negatively correlated with ASC (r=-0.977, P<0.01), whilst positively correlated with CDR (r=0.995, P<0.01). The value of D_0 and D_1 showed a significant correlation with ASC (r=-0.823 and -0.739 respectively, P<0.01) and CDR (r=0.822 and 0.709 respectively, P<0.01), whilst D_0/D_1 had little significant difference with ASC and CDR (Table Ⅳ). With increasing disposal age, aggregation of fine particles resulted in a lower value of fractal dimension and a more stable aggregated structure. This suggests that fractal dimension may be useful to characterize microaggregate stability. Compared to multi-fractal parameters, single-fractal dimension (D) was more significantly correlated with microaggregate stability in bauxite residue.

CONCLUSIONS

Microaggregate stability, an important physical indicator, is required to sustain a stable physical structure. This study has clearly demonstrated that natural weathering processes significantly affect particle size distribution of residue microaggregates. With increasing disposal age, the proportion of silt- and clay-sized microaggregates significantly decreased. Clay dispersion ratio (CDR) decreased from 22.5% to 7.7%, and aggregated silt and clay (ASC) increased from 15.3% to 19% indicating that natural weathering processes enhanced microaggregate stability. Clay content, organic carbon, exchangeable bases and pH were significantly correlated with ASC and CDR which indicated that organic carbon and exchangeable cations had significant effects on microaggregate stability. The value for single-fractal dimension (D) varied from 2.2 to 2.4. With increasing disposal age, both single-fractal dimension (D) and multi-fractal parameters (D_0, D_1 and D_1/D_0) decreased, revealing that natural weathering process promoted aggregation of microaggregates. Correlation analyses demonstrated that fractal parameters were significantly correlated with microaggregate stability and physicochemical properties, indicating that fractal parameters may be used to characterize residue physical structure and related properties. This study may help to provide an improved understanding of physical microstructures, appropriate indicators to use when evaluating microaggregate stability, and a scientific basis for the revegetation of bauxite residue disposal areas.

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