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Effects of Natural Weathering on Organic Carbon Fractions in Red Mud Aggregates: Postprint

Authors: Zhu Feng, Li Meng, Xue Shengguo, Zou Qi, Wu Hao, Wang Qiongli

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Abstract

This study investigated the red mud disposal site of an alumina enterprise in Central China, collecting 0-20 cm samples from sites with different storage durations (1a, 10a, 20a) to examine the effects of natural weathering processes on organic carbon fractions in red mud aggregates using physical fractionation methods. The results showed that the proportion of light fraction organic carbon in red mud increased with prolonged storage time. Heavy fraction organic carbon accounted for approximately 97.24%-99.11% of total organic carbon in red mud, being dominated by coarse particulate organic carbon and mineral-associated particulate organic carbon. The particulate organic carbon content in red mud aggregates for the three storage durations (1a, 10a, 20a) ranged from 1.21-1.85, 2.62-2.95, and 3.52-4.15 g/kg, respectively. Particulate organic carbon content was highest in the 2-1 mm size fraction of red mud aggregates and decreased with decreasing particle size, with mineral-associated particulate organic carbon having the highest proportion and free particulate organic carbon the lowest. This indicates that natural weathering processes increased the content of organic carbon fractions in red mud and enhanced the stability of the organic carbon pool.

Full Text

Effects of Natural Weathering Processes on the Distribution Characteristics of Organic Carbon and Its Composition in Bauxite Residue Aggregates

ZHU Feng[1], **LI Meng**[1], **XUE Shengguo**[1,2,*], **ZOU Qi**[1], **WU Hao**[1], **WANG Qiongli**[1]

[1]School of Metallurgy and Environment, Central South University, Changsha 410083, China

[2]Chinese National Engineering Research Center for Control and Treatment of Heavy Metal Pollution, Central South University, Changsha 410083, China

Abstract

Revegetation is regarded as a promising approach for large-scale remediation of bauxite residue disposal areas. Formation of aggregate structure and the dynamic processes of the organic carbon pool are essential factors for achieving revegetation, given the high alkalinity, salinity, and poor physical structure of the residues. Physical fractionation of organic carbon can identify specific carbon pools responsible for carbon management and control. This study examined a bauxite residue disposal area in Central China, where spontaneous vegetation encroachment over a 20-year period has revealed that natural soil-forming processes may convert the residues into a soil-like medium. Residue samples from three different stacking ages (1 year, 10 years, and 20 years) were collected from the 0–20 cm layer to determine the effects of natural weathering processes on aggregate formation and organic carbon fractions. The contents and distribution ratios of light fraction organic carbon (LFOC), heavy fraction organic carbon (HFOC), particulate organic carbon (POC), and intra-aggregate POC were determined.

The results indicated that the content of organic carbon fractions in bauxite residue increased significantly under natural weathering processes. The proportion of LFOC relative to total organic carbon ranged from 0.36% to 2.06%, with its distribution ratio increasing with stacking age. Most organic carbon (97.24%–99.11%) was held in the HFOC pool, indicating that organic carbon dynamics in the residues were controlled by this fraction. In the HFOC, coarse POC and mineral-combined organic carbon predominated, whereas the distribution ratio of fine POC was relatively small. POC content was highest in 2–1 mm residue aggregates and lowest in the 0.25–0.05 mm fraction. Across the three stacking ages, POC content ranged from 1.21–1.85 g/kg (1 year), 2.62–2.95 g/kg (10 years), to 3.52–4.15 g/kg (20 years). Mean weight diameter was positively correlated with total organic carbon, LFOC, HFOC, and POC ($r = 0.908$, 0.908, 0.889, 0.793 respectively; $P < 0.01$). The content of free POC, occluded POC, and mineral-combined POC decreased with decreasing aggregate size. The distribution ratio of POC in intra-residue aggregates followed the order: 2–1 mm > 1–0.25 mm < 0.05 mm > 0.25–0.05 mm. Among these, mineral-combined POC was the major fraction, while the proportion of free POC was the lowest. Natural weathering processes increased total organic carbon and fraction contents, further enhancing the stability of organic carbon in bauxite residues, which was beneficial for organic carbon sequestration. These findings provide a theoretical basis for understanding carbon sequestration and contribute to improving the physical structure of bauxite residue.

Keywords: bauxite residue; natural weathering processes; light fraction organic carbon; heavy fraction organic carbon; particulate organic carbon; soil formation

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Introduction

Bauxite residue (red mud) is a highly alkaline solid waste generated during alumina production. Approximately 1.0–2.0 tons of red mud are produced per ton of alumina, with Chinese alumina enterprises generating about 70 million tons annually. The large-scale disposal of red mud has become an urgent problem constraining sustainable development of the alumina industry worldwide. Red mud is characterized by high alkalinity and salinity, poor physical structure, and severe limitations to plant growth. Current utilization rates remain below 5%, with most red mud being discharged via dry stacking into disposal areas, which not only occupies vast land areas but also causes serious atmospheric and water pollution.

Red mud resource utilization primarily includes building material preparation, valuable metal recovery, and environmental material applications. However, these approaches face challenges of high costs, potential secondary pollution, and difficulty in establishing long-term stable vegetation. Revegetation is considered the most promising and effective method for large-scale ecological disposal of red mud. Courtney and Harrington found that adding gypsum and organic compost could improve red mud physicochemical properties and fertility. Jones et al. reported that organic matter and coarse particle amendments enhanced red mud physical properties, facilitating vegetation reconstruction. However, amendment addition is costly and may cause secondary pollution.

Natural weathering processes, though requiring longer periods, can establish stable vegetation without secondary pollution. Santini et al. observed spontaneous vegetation encroachment on a Guyanese red mud disposal area, while Zhu et al. documented natural colonization by herbaceous plants at a Central China alumina enterprise site. These findings demonstrate that natural weathering can transform red mud into a soil-like growth medium, enabling natural succession of native plants. During this process, red mud physicochemical properties gradually improve, alkalinity decreases, and microbial community structure and abundance may increase significantly.

Soil organic carbon is a critical indicator of soil quality and fertility. Since the soil organic carbon pool comprises components with different turnover periods, it is not sensitive to short-term soil quality changes. Physical fractionation methods can separate soil organic carbon into light and heavy fractions, as well as particulate organic carbon (53–2000 m). Light fraction organic carbon and particulate organic carbon represent non-protected, labile carbon pools and serve as sensitive indicators for investigating soil carbon dynamics. Based on location within the soil matrix, particulate organic carbon can be divided into free, occluded, and mineral-combined forms. Physical fractionation minimally disrupts organic carbon structure, effectively reflecting carbon structure, function, and turnover characteristics.

This study investigated red mud samples from different stacking ages under natural weathering processes. Using physical density fractionation methods, we examined the effects of weathering on the distribution of organic carbon fractions in red mud aggregates, providing a theoretical basis for aggregate structure formation and organic carbon stabilization.

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1. Study Area and Methods

1.1 Study Area Description The study area was located at a red mud disposal site of an alumina enterprise in Central China (113°25 E, 35°24 N). The region has a warm temperate continental monsoon climate with four distinct seasons, an average annual temperature of 14 °C, and average annual precipitation of 560 mm. Red mud from alumina production is transported via elevated pipelines to the disposal site. Sampling was conducted in May, with systematic investigation of the site and collection from three areas with different stacking ages (1 year, 10 years, and 20 years). The 1-year and 10-year sites had no vegetation, while the 20-year site had sparse plant growth including bermudagrass (*Cynodon dactylon*) and black nightshade (*Solanum nigrum*).

1.2 Sample Collection From each of the three stacking age areas, five sampling plots were selected. At each plot, five random points were sampled from the 0–20 cm layer. Samples from the five points were mixed, and 1.5 kg was taken using the quartering method for laboratory analysis.

1.3 Light Fraction Organic Carbon Determination Air-dried samples were sieved to remove gravel and plant roots. Light fraction organic carbon (LFOC) was separated using the method of Roscoe and Buurman [22]. Briefly, 10 g of sample was placed in a centrifuge tube with 30 mL of sodium iodide solution (density 1.8 g/cm^3), shaken for 30 minutes, and centrifuged at 3000 rpm for 15 minutes. The floating light fraction was aspirated, washed with deionized water, dried at 60 °C, and weighed. Both light and heavy fractions were analyzed for organic carbon content using a VARIO MAX C/N elemental analyzer, and their distribution ratios were calculated.

1.4 Heavy Fraction Organic Carbon Determination Heavy fraction organic carbon (HFOC) was dispersed following the method of Geng et al. [23]. The heavy fraction was treated with sodium hexametaphosphate (0.5% mass fraction) and ultrasonic dispersion to obtain intra-aggregate coarse particulate, fine particulate, and mineral-combined fractions. Organic carbon content of each fraction was determined and distribution ratios calculated.

1.5 Particulate Organic Carbon Determination Particulate organic carbon (POC) was separated following the method of Cambardella and Elliott [24]. Samples were treated with sodium hexametaphosphate and heavy liquid (1.8 g/cm³) to separate free particulate organic matter with density < 1.6 g/cm³.

1.6 Intra-Aggregate Particulate Organic Carbon Determination Intra-aggregate POC fractions were separated using the method of Golchin et al. [25]. This yielded free POC (fPOM<1.6), occluded POC (oPOM<1.6 and oPOM1.6-2.0), and mineral-combined organic carbon (Mineral>0.05 mm and Mineral<0.05 mm). The content and distribution ratio of each fraction were determined.

1.7 Data Analysis Data processing and analysis were performed using Origin 8.0, Microsoft Excel 2003, and SPSS 13.0. One-way ANOVA was used to test for significant differences among treatments, with Duncan' s multiple comparison test at $P < 0.05$. Pearson correlation coefficients were calculated for bivariate relationships.

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2. Results

2.1 Light Fraction Organic Carbon Distribution in Red Mud Aggregates The LFOC content in red mud aggregates during natural weathering is shown in Table 1. Across the three stacking ages (1a, 10a, 20a), LFOC content ranged from 0.028–0.039, 0.029–0.065, and 0.056–0.241 g/kg, respectively. Significant differences were observed among aggregate sizes within each age group ($P < 0.05$), except between 2-1 mm and 0.25–0.05 mm fractions in the 1-year samples ($P > 0.05$). LFOC content in all aggregate sizes showed the trend A < B < C, indicating that LFOC increased with stacking age.

Table 1 Light fraction organic carbon (LFOC) content of residue aggregates in natural weathering processes

Aggregate size (mm)	LFOC content (g/kg)
2–1	0.032±0.01a
1-0.25	0.039±0.01a
0.25–0.05	0.029±0.01a
<0.05	0.034±0.01a

Note: Different lowercase letters indicate significant differences (P < 0.05).

The LFOC distribution ratio in different aggregate sizes is shown in Table 2. Across the three ages, LFOC proportions ranged from 0.52%–0.71%, 0.36%–0.87%, and 0.77%–2.06%, respectively. The 1-0.25 mm fraction had the highest proportion, while the 0.25-0.05 mm fraction had the lowest. Significant differences were observed among aggregate sizes ($P < 0.05$), except between 2-1 mm and 0.25-0.05 mm fractions ($P > 0.05$). The trend B < A < C in most fractions indicated that natural weathering and vegetation encroachment increased LFOC distribution ratios.

Table 2 LFOC proportion of residue aggregates in natural weathering processes

Aggregate size (mm)	LFOC proportion (%)
2-1	0.56±0.02b
1-0.25	0.71±0.02a
0.25-0.05	0.52±0.01a
<0.05	0.63±0.02a

2.2 Heavy Fraction Organic Carbon Distribution Heavy fraction organic carbon (HFOC) content is presented in Table 3. Across the three ages, HFOC ranged from 5.36-5.69, 7.46-8.49, and 11.06-12.11 g/kg, respectively. All aggregate sizes showed the trend A < B < C ($P < 0.05$), with significant differences among stacking ages.

Table 3 HFOC content of residue aggregates in natural weathering processes

Aggregate size (mm)	HFOC content (g/kg)
2-1	5.69±0.46a
1-0.25	5.41±0.51a
0.25-0.05	5.45±0.39a
<0.05	5.36±0.35a

The HFOC distribution ratio (Table 4) ranged from 99.29%-99.47%, 99.13%-99.64%, and 97.94%-99.23% across the three ages. The 2-1 mm fraction had the highest proportion, while the 0.25-0.05 mm fraction had the lowest. Significant differences were observed among aggregate sizes ($P < 0.05$).

Table 4 HFOC proportion of residue aggregates in natural weathering processes

Aggregate size (mm)	HFOC proportion (%)
2-1	99.02±1.67b
1-0.25	98.65±2.01b
0.25-0.05	98.83±1.22b
<0.05	98.76±2.57b

Analysis of HFOC fractions revealed that organic carbon was primarily distributed in coarse particulate and mineral-combined forms within aggregates

(Figure 1). Fine particulate organic carbon content was lowest. With increasing stacking age, coarse particulate organic carbon increased significantly, while fine and mineral-combined fractions showed minimal change. Coarse particulate organic carbon had the highest distribution ratio, followed by mineral-combined particulate organic carbon.

Figure 1 Content and proportion of organic carbon in the scattered aggregate of residue aggregates

2.3 Particulate Organic Carbon Distribution POC content in aggregates is shown in Table 5. Across the three ages, POC ranged from 1.21–1.85, 2.62–2.95, and 3.52–4.15 g/kg, respectively. POC content was highest in 2–1 mm aggregates and lowest in 0.25–0.05 mm fractions. All aggregate sizes showed the trend A < B < C ($P < 0.05$).

Table 5 POC content of residue aggregates in natural weathering processes

Aggregate size (mm)	POC content (g/kg)
2–1	1.85±0.25a
1–0.25	1.63±0.16a
0.25–0.05	1.21±0.12a
<0.05	1.57±0.18a

The POC distribution ratio (Table 6) ranged from 21.24%–28.65%, 20.46%–27.28%, and 25.64%–29.16% across the three ages. The 2–1 mm fraction had the highest proportion, while the 0.25–0.05 mm fraction had the lowest.

Table 6 POC proportion of residue aggregates in natural weathering processes

Aggregate size (mm)	POC proportion (%)
2–1	28.65±1.32b
1–0.25	25.78±1.08b
0.25–0.05	21.24±0.64b
<0.05	28.46±1.48c

2.4 Intra-Aggregate Particulate Organic Carbon Distribution Intra-aggregate POC content increased with stacking age across all aggregate sizes. The order of POC content from highest to lowest was: 2–1 mm, 1–0.25 mm, <0.05 mm, 0.25–0.05 mm. Among the physical components, mineral-combined organic carbon (Mineral<0.05 mm) had the highest content, followed by occluded POC (oPOM1.6–2.0), free POC (fPOM<1.6), and mineral-combined organic carbon (Mineral>0.05 mm) (Figure 2).

Figure 2 Content and proportion of particulate organic carbon in residue aggregates

Mineral-combined organic carbon (Mineral>0.05 mm) had the highest distribution ratio (40%-50%), followed by mineral-combined organic carbon (Mineral<0.05 mm) (40%-45%). These results indicate that mineral-combined organic carbon is the dominant intra-aggregate POC fraction in red mud.

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3. Discussion

3.1 Effects of Natural Weathering on Physical Fractions of Organic Carbon Light fraction organic carbon consists of plant residues and microbial debris at various decomposition stages, with chemical components including carbohydrates, polyphenols, and alkyl compounds. It plays an important role in soil structure formation and stability, particularly for macroaggregates (>0.25 mm). Aboveground litter and underground roots are the primary sources of light fraction organic carbon. In red mud disposal sites, vegetation establishment is extremely difficult. Although sparse herbaceous plants were observed on the 20-year site, LFOC content and distribution ratios remained very low, with HFOC dominating the organic carbon pool.

The slight increase in LFOC at the 20-year site may be attributed to minor plant growth and root inputs, though the effect was limited. With increasing stacking age, the overall LFOC distribution ratio did not change significantly, possibly because microbial debris also contributes to LFOC as physicochemical properties improve and microbial communities develop.

Heavy fraction organic carbon comprises thoroughly decomposed or newly synthesized aromatic substances that are more stable. HFOC responds more slowly to system changes but better reflects carbon retention capacity. During natural weathering, total organic carbon content increased significantly, primarily from microbial metabolism and plant residues. The sum of heavy and light fractions was slightly lower than total organic carbon, likely due to loss of salt-soluble organic carbon during fractionation.

Analysis of HFOC fractions showed that organic carbon was mainly distributed in mineral-combined and coarse particulate forms. Coarse particulate formation requires more organic fragments for cementation, resulting in higher carbon content. Mineral-combined particles interact with clay, which protects organic carbon from decomposition and adsorbs hydrophobic substances, enhancing stability.

Particulate organic carbon reflects the amount of non-protected organic carbon, with higher proportions indicating greater lability. The POC distribution ratio in red mud aggregates (20%-30%) was lower than typical soil values (55%-68%), indicating lower lability. However, the increasing trend with stacking age suggests gradual enhancement of aggregate stability. POC primarily originates from fresh litter inputs and decomposition rates. Despite limited vegetation, POC content increased significantly over time, indicating that microbial activity and physicochemical improvements contributed to carbon accumulation.

3.2 Effects on Intra-Aggregate Particulate Organic Carbon Intra-aggregate POC is crucial for microaggregate formation and stability. With increasing stacking age, free POC content increased significantly, particularly in 2–1 mm aggregates, consistent with findings that free POC accumulation depends on aggregate size and litter quantity. The presence of roots and leaf litter at the 20-year site influenced free POC content and distribution.

Occluded POC represents a relatively labile carbon pool and is an important indicator of total carbon changes. Its content was higher than free POC and increased significantly with stacking age, decreasing with aggregate size. This suggests that macroaggregate POC is more stable than microaggregate POC, likely due to protection within microaggregates.

Mineral-combined organic carbon showed the highest distribution ratio among intra-aggregate fractions and increased gradually with stacking age. This fraction is more stable than occluded POC and less susceptible to microbial decomposition. The increasing contribution of mineral-combined organic carbon in microaggregates (<0.25 mm) aligns with findings from low organic carbon agricultural soils. Natural weathering improved red mud physicochemical properties, enhancing microbial decomposition of labile carbon and promoting mineral-combined organic carbon formation.

Overall, natural weathering increased total organic carbon and altered its composition, with stable carbon pools dominating over labile fractions. This transformation is crucial for improving red mud physical structure and enabling natural vegetation establishment.

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4. Conclusion

Natural weathering processes significantly increased both light and heavy fraction organic carbon contents in red mud aggregates. Heavy fraction organic carbon accounted for 97.24%–99.11% of total organic carbon, dominated by coarse particulate and mineral-combined forms. Aggregate POC content ranged from 20%–30% and showed an increasing trend with stacking age. Within aggregates, mineral-combined organic carbon had the highest distribution ratio (40%–50%), while free POC had the lowest. The increasing mineral-combined organic carbon content and distribution ratio with stacking age indicate enhanced carbon stabilization. These findings demonstrate that natural weathering processes improve red mud quality and promote carbon sequestration, providing a theoretical basis for sustainable red mud remediation.

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