Catena 136 (2016) 17–29

Contents lists available at ScienceDirect

Catena

journal homepage: www.elsevier.com/locate/catena

Assessment of Technosol formation and *in situ* remediation in capped alkaline tailings

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ARTICLE INFO

Article history:
Received 16 January 2015
Received in revised form 30 July 2015
Accepted 12 August 2015
Available online 1 September 2015

Keywords:
Pedogenesis
Technosols
Tailings management
Bauxite residue
Anthroposols

ABSTRACT

Technosols formed from mineral processing wastes such as tailings are of growing importance worldwide due to the increasing demand for mineral and energy resources. The construction and nature of tailings and capping materials in tailing storage facilities are likely to influence soil formation rates; however, these have not been evaluated to date. This study used bauxite residue (alumina refining tailings) as a model material to compare soil development and *in situ* remediation with reference to initial residue composition, landscape position, and cap type. Soil development in the capped tailings deposits was limited to accumulation of organic matter in the upper 0–2 cm of capping materials, and shallow (<10 cm deep) decreases in pH and salinity within the underlying tailing layers. Landscape position had little effect on soil development due to the formation of technic hard material through calcite cementation in sintered bauxite residues, which inhibited weathering. Weak structure development, likely due to volume shrinkage through desiccation, was observed in unsintered bauxite residue. Leaching of acidic pore waters from pyritic mine spoil caps aided in pH neutralization in unsintered bauxite residues.

The current Technosol classification was adequate for description of the soil materials within the study site; however, the introduction of a novel prefix qualifier, *Ordic*, would enable more accurate description of multilayered Technosols. Unsintered bauxite residue under a pyritic mine spoil cap was predicted to move from a Spolic Technosol to a Haplic Cambisol based on variations in structure and colour within the capping. The lack of observable weathering precluded identification of a likely pedogenic trajectory in sintered bauxite residue. Accelerating natural processes of soil development and *in situ* remediation may be achieved by application of chemically reactive caps; however, these should not preclude atmospheric gas exchange, leaching, or interaction with biota as pedogenesis will be retarded.

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1. Introduction

Technosols (similar to Anthroposols under the Australian Soil Classification; Isbell, 2002; no direct equivalent under the USDA Soil Taxonomy) are soils defined by the nature of their parent material, being derived from anthropogenic activities such as energy and minerals extraction processes, construction, and waste collection and disposal (IUSS Working Group WRB, 2014). Technosols were first recognized in the 2006 edition of the World Reference Base for Soil Resources (WRB; IUSS Working Group WRB, 2006) in light of their growing importance globally, particularly in landscapes of urban and industrial areas from which their parent material is most often sourced. Tailings represent one subset of the Technosol Reference Soil Group (RSG), comprising soils formed from waste materials produced during mineral and energy extraction and refining activities. Tailings-derived Technosols typically exhibit extremes of pH and particle size (very fine or very coarse), low nutrient availability, and high salinity. Approximately 7 Gt of tailings are generated globally each year by mineral processing and extraction activities (Mudd and Boger, 2013). Assuming an average bulk density of 1.5 t m^{-3} (Sarsby, 2000), tailings storage facilities are currently growing by an estimated 23,750 ha per year, occupying otherwise productive land. Tailings-derived Technosols are subject to the same factors of soil formation as soils derived from bedrock (climate, organisms, parent material, relief and time; Jenny, 1941) but weather rapidly due to their extreme geochemical, mineralogical, and physical properties, which places them far from equilibrium with earth surface environments. It is therefore possible to observe soil formation over relatively short (<100 years) timescales compared to soils derived from bedrock parent materials, with A horizons developing after 20 years in iron blast furnace wastes (Huot

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http://dx.doi.org/10.1016/j.catena.2015.08.006
0341-8162/Published by Elsevier B.V.

et al., 2013), formation of secondary clay minerals over 40 years in sulphidic mine spoils (Uzarowicz and Skiba, 2011), and development of a gossan layer in the upper 30 cm of sulphide tailings over 50 years weathering (Hayes et al., 2014). Although these timeframes are relatively short in comparison to rates observed in natural soils, they are relatively long when considering operational mine lives. Understanding soil development and weathering processes in Technosols is essential to improving management of these challenging materials, and ultimately accelerating soil formation through the application of targeted treatments to hasten the transition from Technosol to another soil group and return the land area occupied by tailings to a productive post-mining use.

Two major approaches are currently employed for tailings management: 'cap and store' or '*in situ* remediation'. 'Cap and store' approaches to tailings management aim to minimize infiltration of rainfall and air into the tailings, with vegetation established in the capping layers; whereas '*in situ* remediation' approaches promote and enhance contact with water and air to accelerate remediation and soil formation, with the aim of establishing vegetation directly in the remediated tailings. According to cap and store approaches, tailings are deposited in hydrologically isolated areas, capped at the top and lined at the base with impervious layers of compacted clay or plastic, and with drainage pipes at the base to collect and export leachates. Minimising water and gas exchange between the tailings and the surrounding environment limits the generation of chemically problematic leachates (very high or very low pH, high salinity, high dissolved metals concentrations) and allows any leachates generated (through gravity drainage of the 'wet' tailings) to be treated before being discharged to local waterways. *In situ* remediation approaches, in contrast, use amendments such as tillage, compost, and irrigation to enhance natural weathering processes, which does increase management inputs in the short term.

Although the cap and store approach to tailings management simplifies management in the short term, it poses several challenges in the long term. Minimizing water and gas exchange between the tailings and the surrounding environment is likely to retard soil development in the tailings material by reducing the influence of the soil forming factors of climate and organisms (Jenny, 1941), and the cap and store strategy relies heavily upon the integrity of linings and cappings to keep tailings isolated from the surrounding environment. In the long term, impoundments can fail, with catastrophic consequences. Such failures comprise the majority of environmental incidents associated with mining (Franks et al., 2011). Although cap and store approaches aim to minimize infiltration of leachates and interaction with vegetation covers (Mohan et al., 1997; Wehr et al., 2006), leaching and contact with plant roots can be beneficial in remediating some of the undesirable chemical and physical properties of tailings (Mendez and Maier, 2008; Rodriguez-Vila et al., 2014; Santini and Fey, 2015; Solis-Dominguez et al., 2012). Cap and store and *in situ* remediation strategies are not mutually exclusive. Any *in situ* remediation of tailings material underneath caps (*e.g.* through use of permeable caps (Santini and Fey, 2015) or increased permeability of the cap over time during weathering) helps to decrease future risks associated with potential impoundment failure. Soil development is closely tied to *in situ* remediation of tailings because in the absence of targeted remediation strategies, pedogenic weathering processes such as rainfall leaching and desiccation cracking are the main processes by which *in situ* remediation of tailings will occur.

Bauxite residue is a highly alkaline (pH 10–13), saline (EC 2–18 dS m^{-1}), fine grained (sandy clay loam to clay texture) tailings material produced as a byproduct of the Bayer process used for alumina refining. Around 120 Mt of bauxite residue are produced globally each year, adding to the estimated 3 Gt already in tailings storage facilities (Power et al., 2011). An increasing proportion of global bauxite residue production (mainly in China, where energy costs are low) is sintered with lime and/or soda ash prior to disposal to increase aluminium recovery from the Bayer process through subsequent leaching of sodium and calcium aluminates formed during sintering (Liu et al., 2009; Smith, 2009):

$$Al_2O_{3(s)} + Na_2CO_{3(s)} \rightarrow 2NaAlO_{2(s)} + CO_{2(g)} \tag{1}$$

$$12CaCO_{3(s)} + 7Al_2O_{3(s)} \rightarrow 12CaO \cdot 7Al_2O_{3(s)} + 12CO_{2(g)}. \tag{2}$$

Unsintered and sintered bauxite residues present similar challenges for remediation, although sintered bauxite residues have a tendency to form cemented, concrete-like materials as they settle and dry (Liu et al., 2009). Addressing the high alkalinity and salinity of bauxite residue is the key to minimizing environmental consequences of potential tailings storage failures (Ruyters et al., 2011). Permeable caps have previously been demonstrated to have beneficial effects on both soil development and vegetation establishment in bauxite residue (Santini and Fey, 2015) by allowing for neutralization and export of alkaline, saline pore waters, amongst other effects. However, the effects of minimally permeable capping materials and 'reactive' capping materials (such as those releasing acidic or alkaline leachates to buffer the underlying tailings) on soil development has not been evaluated for tailings-derived Technosols to date. Caps can be generally expected to retard soil development due to the insulating effect that they confer from surficial weathering processes; however, this is dependent on their physical properties (coarse-textured caps will allow greater interaction with air, water, and biota than fine-textured caps due to their higher permeability) and the manner in which they are deposited (*e.g.* compaction, use of geotextile liners, *etc.*).

The cap and store approach to tailings management was implemented at the Alcoa–Reynolds Arkansas bauxite residue (alumina refining tailings) storage facility. The residue has weathered for up to 25 years since the cessation of residue deposition, with leachates collected and treated, and a grass cover established in capping layers. The Alcoa–Reynolds Arkansas residue storage area contains subsites with different caps, residue compositions, and landscape positions, and was therefore investigated to: (a) compare effects of initial bauxite residue composition (parent material), landscape position (relief), and capping on soil formation and *in situ* remediation; (b) identify a likely pedogenic trajectory for bauxite residue tailings based on observed soil properties; and (c) evaluate the suitability of the *Technosol* classification within the WRB for describing soils formed from capped tailings.

2. Materials and methods

2.1. Site history and sample collection

The Alcoa–Reynolds Arkansas site contained a bauxite mine as well as two alumina refineries, one operated by Reynolds Metal Company (active 1942–1984) and the other operated by Alcoa (active 1898–1990), each of which had their own tailings storage areas (Fig. 1). These tailings storage areas varied in terms of initial residue composition (unsintered or sintered) and cap type (sand and clay, sand only, and pyritic mine spoil) (Table 1). Immediately after the cessation of residue deposition, capping layers were hydraulically placed over the residues: the Alcoa sintered residue area was capped in 1999; the Alcoa unsintered residue area in 1990; and the Reynolds sintered residue area in 1984. After capping, sites were fertilized at the same rates with a general purpose agricultural fertilizer containing N, P, and K and seeded with a mixed grass cover. Additional sites were sampled along an elevation gradient within the Alcoa sintered residue area to investigate the potential influence of lateral pore water discharge on residue properties (Table 1).

The Alcoa–Reynolds Arkansas site is under a humid subtropical climate (Cfa) according to the Köppen–Geiger climate classification system (Peel et al., 2007), and all bauxite residue storage areas hosted a grass cover dominated by *Avena* sp. and *Festuca* sp. with no apparent symptoms of nutrient deficiencies or metal toxicities. Within each