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Accelerating soil formation in bauxite residue: a solution for long-term tailings management and storage

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Abstract

Bauxite residue is the highly alkaline (pH ~12) and saline tailings material produced during the Bayer process for alumina production. Australia is currently responsible for the management of approximately 750 million tonnes of bauxite residue and produces an additional 30 million tonnes each year. Currently only 2% of bauxite residue produced is recycled (International Aluminium Institute 2017, Di Carlo et al. 2019a); however, there is growing interest in reusing residue by transforming this byproduct into a productive soil material through in situ remediation. This aims to transform the bauxite residue into a soil-like medium, by adding amendments to the residue to reduce pH and salinity and improve other unfavourable conditions.

Our work has focused on optimising the amendment application used to accelerate soil formation, using field scale lysimeters to test a combination of treatments. Here we provide an overview of a successful year-long field trial to promote soil formation, and our key insights from the trial. Combining microbially-driven bioremediation with common amendments compost and tillage accelerated pH neutralisation of the residue and leaching of excess salts, creating a material amenable to plant growth. The trials were completed under harsh Australian climatic conditions utilising available mine site water and cost-effective amendment rates, ensuring that this technology is applicable for further industrial scale-up. The data from this study will allow development of best practice remediation strategies for bauxite residue deposits based on their climate and desired end land use and is also applicable to other alkaline tailings and waste materials. Our results with bauxite residue show that in situ remediation is an innovative solution for rapid, cost-effective long-term tailings management.

Keywords: *remediation, pH neutralisation, red mud, alumina refining, amendments, gypsum, compost, tillage, alkaline*

1 Introduction

Bauxite residue, commonly referred to as ‘red mud’, is the byproduct of alumina refining from bauxite ore. Bauxite residue is a highly alkaline and saline sodic tailings material and poses numerous challenges for long-term storage and maintenance. Australia is a major global producer of alumina, and on average Australia produces higher than global average rates of bauxite residue per tonne of alumina produced, with 1,710 kg residue/tonne alumina product compared to the global average of 1,231 kg residue/tonne product (International Aluminium Institute 2017). This results in ~30 million tonnes of residue being produced each year in Australia, adding to the ~750 million tonnes of residue already in storage (Department of Industry, Science, Energy and Resources 2020; Power et al. 2011; Santini & Fey 2016). With only 2–3% of bauxite residue being reused (International Aluminium Institute 2017; Di Carlo et al. 2019a), most residue remains in purpose-built bauxite residue storage areas (BRSAs), creating long-term legacy issues for bauxite and alumina companies.

Currently, the main approach for BRSA closure is a 'cap and store' approach, whereby high-quality topsoil, bulk fill, and impermeable layer materials are imported to create a cover to isolate the tailings from the environment and provide a plant growth medium. Although this approach may be successful in some circumstances, importing topsoil is costly, and over time the soil cap may deteriorate, resulting in breaches of the impermeable layer separating the tailings from the soil cover, breakdown of the topsoil and/or vegetation cover, capillary rise of salts, and transport of leachates or dust offsite (Wehr et al. 2006; Ren et al. 2018; Di Carlo et al. 2019a). An alternative to the conventional 'cap and store' approach is known as in situ remediation, where amendments are mixed directly into the bauxite residue to improve its chemical and physical properties and create a material amenable to plant growth. In situ remediation allows for progressive improvements to the residue properties, and thereby decreases the challenges associated with tailings storage over time.

A key control for the success of in situ remediation is the selection of amendments. Previous remediation studies in bauxite residues have commonly used either chemical (e.g. gypsum/organic matter) or physical amendments (e.g. tillage) to improve residue properties. Gypsum is known to decrease sodicity by replacing Na^+ with Ca^{2+} (Courtney et al. 2009); organic matter provides a source of organic C and nutrients (Jones et al. 2011; Zhu et al. 2017); and tillage helps to increase hydraulic conductivity and promote leaching of excess salts (Santini & Banning 2016). Effectiveness of these amendments in isolation or combined has been previously demonstrated at laboratory, glasshouse and field scale (Wong & Ho 1991; Courtney et al. 2009; Zhu et al. 2017; Bray et al. 2018; Santini & Fey 2018). A novel approach to in situ remediation is to use microbial communities to enhance remediation processes, by either stimulating native microbial communities through addition of nutrient or carbon sources (biostimulation), or through addition of a microbial inoculum (bioaugmentation) (Santini et al. 2015). Bioaugmentation and biostimulation can improve residue properties primarily through promoting the production of acidic microbial exudates, which have been shown to significantly decrease bauxite residue pH in laboratory trials (Santini et al. 2021). The next stage is to test this novel biotechnology at field scale to assess performance and combine this microbial approach with conventional chemical and physical amendments, to identify any synergistic benefits to residue properties and remediation progress.

Here we hypothesised that combining conventional amendments (compost and tillage) with a novel microbial amendment, would accelerate the remediation of bauxite residue into a material amenable to plant growth, by decreasing pH and salinity to a greater extent than compost and tillage or microbial amendments in isolation. To test our hypothesis, a field trial was conducted using 10 m × 15 m × 2 m plots containing bauxite residue and selected amendments, and changes in the geochemical and physical properties of the residue were measured and evaluated over the course of 12 months. In this work we aimed to: (a) test and quantify the effectiveness of microbial and compost and tillage amendments, in isolation and combination, on improving bauxite residue properties at field scale and under field climatic conditions; and (b) determine the optimal amendment, or combination of amendments, to accelerate in situ remediation of bauxite residue. This paper will present the key preliminary results from a year-long field trial, assessing in situ remediation of bauxite residue using common amendments compost and tillage, with novel microbial technology, to accelerate 'soil' formation.

2 Methodology

2.1 Field trial location and setup

Four lined and under drained plots (10 m × 15 m × 2 m deep) were set up at Alcoa's Kwinana residue storage area, located 40 km south of Perth, Australia. The bottom 150 cm of each plot was filled with unamended, fresh filter press residue mud. The upper 50 cm of each plot received gypsum amended (5 wt.%) filter press residue.

Three amendment treatments were applied to the plots using a factorial design (microbial, compost and tillage, and combined microbial and compost and tillage), alongside a control plot (Figure 1). The microbial (M) plot had the upper 5 cm of residue replaced with residue pre-neutralised in a bioreactor pit (Figure 2).

Residue (gypsum amended) in a bioreactor pit received 5 wt.% topsoil as microbial inoculum, 7.5 wt.% dextrose and 2.25 wt.% spent brewing yeast as carbon and nutrient sources to fuel microbial fermentation and organic acid production, and was incubated for 32 days before use in the microbial plot. pH was monitored periodically during incubation and reached a minimum of 9.02. The compost and tillage (CT) plot had 1.67 wt.% compost tilled into the upper 15 cm of residue and received subsequent fortnightly tillage to 15 cm depth (Figure 2). The combined microbial and compost and tillage (M+CT) plot had the upper 5 cm replaced with pre-neutralised bioreactor pit residue, which was then tilled into the upper 15 cm with 1.67 wt.% compost and received subsequent fortnightly tillage (Figure 2). Daily irrigation (approx. 1–2 mm above daily evaporation rate) was applied to all plots in summer months using available onsite recycled water (pH 9–9.5 and electrical conductivity (EC) ≥ 1 mS cm^{-1}).

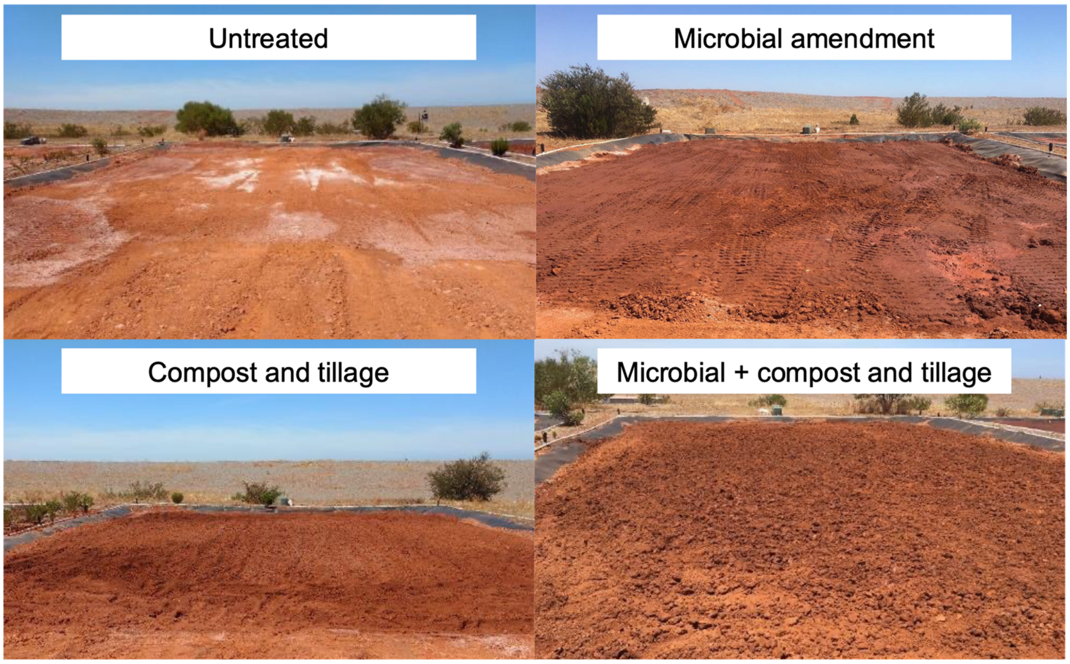

Figure 1 Photos of bauxite residue plots with selected amendment treatments (Untreated (U), microbial (M), compost and tillage (CT) and combined microbial and compost and tillage (M+CT) at time 0 of the field trial

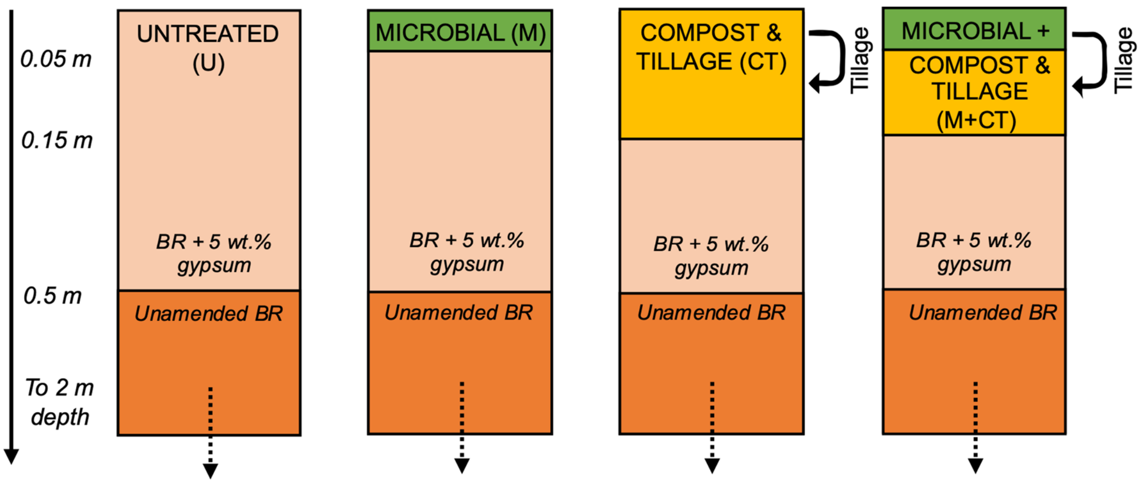

Figure 2 Depth profiles of amended bauxite residue plots (untreated (U), microbial (M), compost and tillage (CT) and combined microbial and compost and tillage (M+CT)

2.2 Sample collection and analyses

Three replicate cores were collected from each field plot at days 0, 90, 180, 270 and 360. Cores at day 0 were collected to 40 cm depth using PVC pipes hammered into the residue. From day 90 onwards, the coring method was improved by using a percussion corer and reached a depth of 90 cm. Cores were sub-sectioned into 0–5 cm, 5–10 cm, 10–20 cm, 20–30 cm, 30–40 cm, 40–50 cm, 50–60 cm, 60–70 cm, 70–80 cm and 80–90 cm intervals for analysis.

Residue samples were dried at 40°C until constant weight. The pH and EC of dried samples were measured in a 1:5 soil/water extract (Rayment & Higginson 1992; Santini et al. 2013). Replicate core samples were combined and finely ground into one composite sample for X-ray diffraction (XRD) analysis. Synchrotron XRD patterns were collected at the Australian Synchrotron Powder Diffraction beamline 10BM1, using a wavelength of 0.7746 Å over a range of 3–90 °2θ, with a count time of 4 min per position. XRD patterns were analysed quantitatively using the Rietveld method (Rietveld 1969) in TOPAS Academic V3.

2.3 pH and EC remediation targets

To measure the pH neutralisation performance of amendments, a target of pH ≤9 and EC of ≤4 mS cm^{-1} within the upper 20 cm was set. These values were based upon targets set within previous similar studies investigating alkaline, saline sodic soils or bauxite residue revegetation (Gräfe et al. 2011; Gräfe & Klauber 2011; Santini et al. 2015; Di Carlo et al. 2019b).

3 Results and discussion

3.1 Microbial amendments promote pH neutralisation

Combining the microbial treatment with compost and tillage (M+CT) resulted in synergistic benefits to neutralisation of residue pH. Addition of the microbial treatment (M or M+CT) immediately decreased surface pH (Figure 3), and lower surface pH within M and M+CT was then maintained throughout the trial. Over the one-year field trial, treatments containing the microbial amendment returned a significantly lower average pH (M: pH 8.96; M+A: pH 9.21) in the upper 0–5 cm than the untreated control (U) or CT; and M+CT had a significantly lower pH (9.28) in the 5–10 cm depth than all other treatments. Lower average pH in the microbial treatments was maintained despite onsite irrigation water fluctuating between pH 9.1–9.3 in all treatments during summer months. By day 360, pH in the upper 20 cm of M was 9.0 and in the M+CT treatment 9.5, both significantly lower than the untreated control or CT plots.

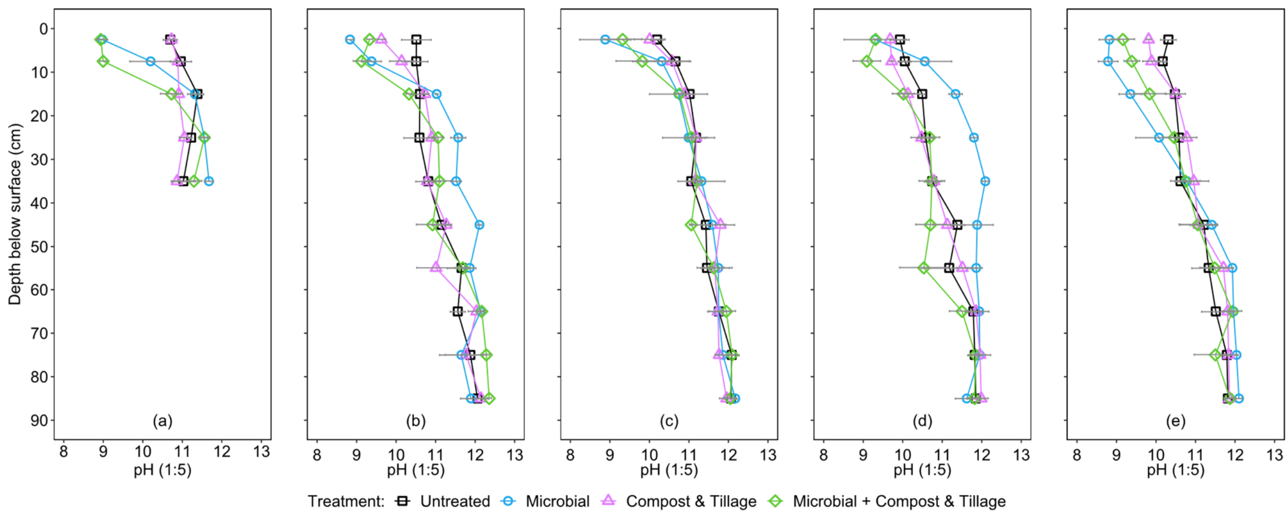

Figure 3 Depth profile showing the change in pH within the untreated control (U), microbial (M), compost and tillage (CT) and combined microbial and compost and tillage (M+CT) plots at times: (a) 0 days, (b) 90 days, (c) 180 days, (d) 270 days, and (e) 360 days. NB Coring at 0 days only reached a depth of 30 cm

Depth of pH neutralisation remained shallow despite amendments (Figure 3). Significant differences in pH compared to the untreated control were limited to the upper 10 cm for M+CT and the upper 5 cm for M, when considering all time points. Below 50 cm depth, residue pH remained >11 within all plots. This is expected given the minimal irrigation rates and the dry Perth Mediterranean climate, resulting in limited leaching. Glasshouse trials also only achieved pH ≤9 in the upper 15 cm after 5 months of leaching (Santini et al. 2019). However, the pH neutralisation front did advance downwards during the trials. Average pH across all treatments was significantly lower in the upper 0–5 cm than below 5 cm at day 0; and significantly lower in the upper 0–20 cm than below 20 cm at day 360, reflecting the downwards advance of the pH neutralisation front over time.

Microbial fermentation of glucose in the microbial treatments enhanced pH neutralisation in the microbial treatments, as has previously been demonstrated in laboratory and glasshouse studies (Santini et al. 2016, 2021), and now for the first time here at field scale. Concurrently, microbial treatments also enhanced dissolution of alkaline Bayer process mineral tricalcium aluminate (TCA; $Ca_3Al_2O_6$), therefore enabling greater pH neutralisation than the control or A. TCA was completely removed from M+CT and M at 0–5 cm by day 90, 5–10 cm by day 270 and 10–20 cm by day 360, while TCA was only removed from the untreated control and CT at 0–5 cm by day 180, and was still present at >5 cm depths at day 360 (Figure 4). TCA assists in buffering residue pH at ~10 (Khaitan et al. 2010), and its removal is necessary for dissolution of other alkaline minerals (such as sodalite and calcite) to proceed effectively (Khaitan et al. 2009). Ensuring the removal of solid phase alkalinity and its subsequent buffering capacity is key to achieving and maintaining circumneutral pH during the long-term remediation process.

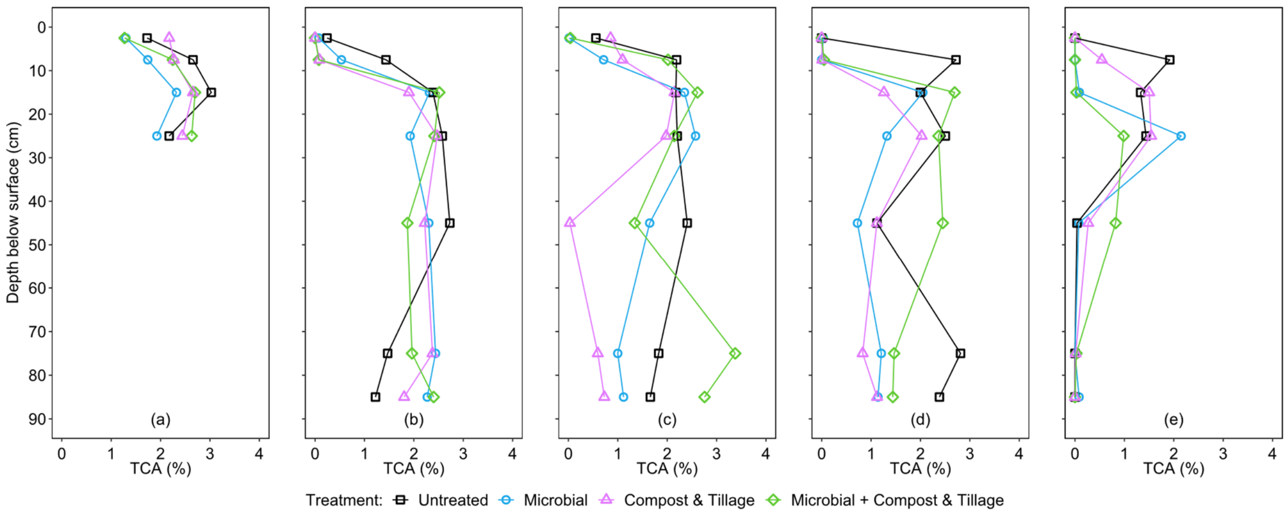

Figure 4 Depth profile showing the change in TCA content (%) within the untreated control (U), microbial (M), compost and tillage (CT) and combined microbial and compost and tillage (M+CT) plots at times (a) 0 days, (b) 90 days, (c) 180 days, (d) 270 days, and (e) 360 days. NB Coring at 0 days only reached a depth of 30 cm

Gypsum ($CaSO_4.2H_2O$) addition within the upper 50 cm may be suppressing TCA dissolution through a common ion effect. At day 360, enhanced removal of TCA at 50 cm depth and complete removal at 70–80 cm depth was observed, in addition to removal within microbial treatments in the surface 0–15 cm (Figure 4). Wissmeier et al. (2011) also observed that gypsum suppressed TCA dissolution during predictive modelling simulations of gypsum amended bauxite residue sand under field conditions. Here, the microbial amendments were able to promote TCA dissolution in the upper 50 cm, despite the presence of gypsum. Without the presence of gypsum, dissolution of TCA below 50 cm was likely able to proceed in all plots due to a higher average moisture content throughout the trial, compared to the upper 50 cm which was frequently affected by wet-dry cycles (reflected by capillary rise of salts; see Section 3.2). Despite the removal of TCA, residue pH remained high and indicates that further leaching of alkaline porewater and salts is still required.

Combining the microbial amendment with CT minimised the potential for capillary rise and pH rebound, which was observed in the M only plot at day 270 (Figure 3). The pH increase was likely caused by insufficient leaching of alkaline porewater and salts. M had lower permeability in general than other treatments, likely resulting from exudation of extracellular polymeric substances (EPS) by resident microbial communities during fermentation processes (Santini et al. 2015) that clogged fine pores. Incorporation of tillage and compost in M+CT avoided this reduced hydraulic conductivity, and subsequent pH rebound; likely by encouraging EPS to form around aggregates and bind particles together into larger, stable aggregates that enhanced drainage. Tillage also likely minimised pH rebound through accelerated atmospheric carbonation, which contributes to overall pH neutralisation.

3.2 Tillage and compost enhance salt leaching

Rainfall and irrigation driven leaching of saline porewater decreased salinity in all treatments over the course of the trial. Significant decreases in EC compared to day 0 within the upper 30 cm were observed for all treatments by day 180 (Figure 5). By day 360 there were no significant differences in EC among treatments. However, periodic capillary rise and precipitation of salts at the surface was observed. The microbial (M) plot in particular was heavily impacted by capillary rise throughout the trial. This is likely due to increased production of microbial EPS, leading to the overall lower hydraulic conductivity in this treatment, and therefore less leaching of salts and higher potential for capillary rise. This is evident through small increases in EC within M between days 0–90 and 180–270, followed by a significant decrease in EC between days 90–180 and 270–360. The generally higher EC in M compared to M+CT throughout the trial indicates that tillage was essential for effectively exporting salts from the system, particularly in treatments with greater pH neutralisation and subsequently higher dissolved salts in porewater.

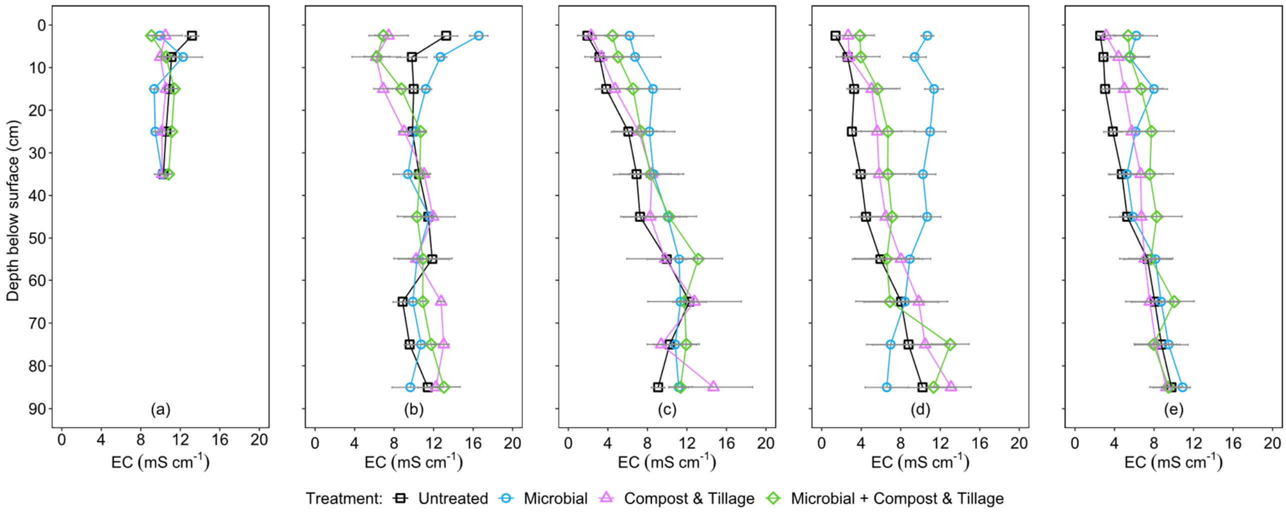

Figure 5 Depth profile showing the change in electrical conductivity (EC) within the untreated control (U), microbial (M), compost and tillage (CT) and combined microbial and compost and tillage (M+CT) plots at times (a) 0 days, (b) 90 days, (c) 180 days, (d) 270 days, and (e) 360 days. NB Coring at 0 days only reached a depth of 30 cm

3.3 Performance of amendments against remediation targets

The microbial amendment accelerated pH neutralisation through increased dissolution of TCA and neutralisation of porewater alkalinity, likely from the production of acidic microbial fermentation products (Santini et al. 2016, 2019, 2021). Neutralisation of bauxite residue pH is arguably the most important goal of remediation works and is essential for any revegetation works to proceed; pH is known to influence a range of soil and plant properties and availability of key nutrients (Hazelton & Murphy 2007). The pH remediation target of ≤9 was only met in M, however M+CT was close to this target at day 360.

The addition of CT in combination with the microbial amendment was essential for effective leaching of salts and structure development, which was limited in M due to the enhanced aggregation likely caused by microbial EPS. The EC target of <4 mS cm^{-1} was met by the untreated control and CT (at 0–10 cm); however, attainment of the EC target in these treatments was likely only due to their weaker pH neutralisation performance which released lower concentrations of soluble neutralisation salts than M+CT and M. Additionally, no significant differences in EC between treatments was observed at day 360, suggesting that M+CT and M are likely to reach the EC target shortly.

While the results are not presented here, the addition of both a microbial amendment and CT likely contributed to improving other important residue properties, such as bulk density, organic C and N, exchangeable sodium percentage, sodium absorption ratio, major and trace element levels, and removal of additional alkaline minerals such as sodalite and calcite. Changes in these residue properties due to the addition of microbial, CT, or combined amendments will be quantified in future works.

Although only two key remediation targets (pH and EC) were assessed here, it is clear that the microbial treatments outperformed the untreated control and CT after one year. M had the highest extent of pH neutralisation; however, the combined M+CT treatment showed significantly greater pH neutralisation than the control while also maintaining lower EC than M. This demonstrates that combining both microbial and CT amendments can provide synergistic benefits, and lead to overall greater success during in situ remediation of bauxite residue.

3.4 Implications for bauxite residue and alkaline tailings remediation going forward

The combined microbial and CT treatment utilised here could be applied at other bauxite residue or alkaline tailings storage facilities. For the CT amendment, compost is likely to be readily available across a range of sites (i.e. wood chips from land clearing activities, treated sewage sludge from onsite facilities). While the microbial treatment does require an incubation period, the amount of soil used for the microbial inoculum is far lower than would be required for a ‘cap and store’ technique. Additionally, using an in situ remediation approach (compared to ‘cap and store’) allows for the progressive remediation of tailings, and minimises risks associated with storage over time.

Once applied, the extent and rate of remediation using a combined microbial and compost and tillage treatment is likely to change depending on individual site conditions. The progression of remediation here was limited by climatic conditions and water restrictions within the Perth region of Western Australia. Leaching of saline porewater and neutralisation salts from the residue is key for improving residue properties, but low levels of rainfall and irrigation limited these processes during the one-year field trial. During the summer months, irrigation levels were restricted to countering evaporation rates (approx. 1–2 mm above evaporation rates); however, capillary rise was still noted within the field plots. Additionally, the trial irrigation water was sourced from onsite recycled water sources, in order to reduce costs and demonstrate the potential for scale up and was consistently between pH 9–9.5 which likely inhibited the remediation process. Higher irrigation rates during the initial stages of remediation, with freshwater if available, are recommended when applying this approach for closure of other storage areas, in order to accelerate the remediation process. Leveraging favourable seasonal conditions (wetter and cooler periods) is recommended to maximise remediation success. Using this remediation approach within wetter climates would likely result in accelerated remediation progress compared to observations in this study.

4 Conclusion

Bauxite and alumina companies are increasingly seeking ways to further improve the sustainability of their industry and develop innovative solutions for tailings storage and management. The outcomes from this study provide evidence for in situ remediation as a rapid and effective solution for long-term tailings management. Here we have demonstrated that: (a) microbial amendments provide an effective means for neutralising bauxite residue pH at field scale; and (b) that combining microbial biotechnology with commonly used amendments (compost and tillage) for bauxite residue remediation supports the attainment of pH and

EC remediation targets, thereby contributing to creating a soil-like medium from the residue, and is more effective than either a microbial or compost and tillage approach alone.

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