Constructed Wetlands For Treatment Of Bauxite Residue Leachate: 7.5 Years Of Monitoring

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Abstract

Constructed wetlands (CWs) have been proven as an effective, nature-based alternative for effective treatment of predominantly acidic waters. Alkaline wastes and leachates, including bauxite residue, have received far less focus, with long term studies limited to around 5 years.

The work presented demonstrates that a CW can effectively treat bauxite residue (BR) leachate from a pH ca. 11.2 to pH <9 over a period of 7.5 years, providing evidence CWs may be a viable treatment method for BR leachate.

Keywords: Bauxite residue leachate, constructed wetlands, longevity, alkaline leachates.

Introduction

The extraction and refining of metals has led to the accumulation of tailings and waste deposits globally, amounting to over 100 billion tonnes, with drainages and leachates from such stockpiles potentially of environmental concern if not managed appropriately (Mishra *et al*. 2022). Of this global stockpile, the alumina industry is estimated to have produced and stored over 4 billion tonnes of bauxite residue (BR), increasing by 150 MT annually (International Aluminium Institute 2022). BR leachates are inherently alkaline owing to the Bayer process utilising NaOH, with leachate pH between 9 and 13.2, as well as elevated Al, As, V, and Na (Burke *et al*. 2013; Higgins *et al*. 2017)

As BR leachates and other drainages can have the potential to form for decades post-closure, long term management can pose a challenge to operators and environmental regulators (Higgins *et al*. 2017; Riley and Mayes 2015; Roadcap *et al*. 2005). Both active and passive technologies can be employed, but each site and its individual chemical, geographical and climatic parameters may determine the appropriate technique to be applied. Globally, increasing attention to water quality and a shift to passive, nature-based technologies has resulted in the increased application of constructed wetlands (CWs) for leachate and drainage treatment as performance has been found comparable to active chemical treatment, whilst also being cost effective (Chen *et al*. 2021; Hedin 2020).

While CWs have been extensively utilised for the treatment of acidic and neutral mine drainage (Acharya and Kharel 2020; Ighalo *et al*. 2022), their use for the remediation of alkaline drainages has received far less research focus (Gomes *et al*. 2019; Hudson *et al*. 2021). Although efficacy has been assessed in acidic and neutral mine drainages for up to 20 years (Hedin 2020), published longevity studies for CWs mostly cover operational periods of 1-5 years, with a limited number covering over 10 years. Monitoring of CWs used for treatment of alkaline leachates is particularly lacking (Gomes *et al*. 2019; O'Connor and Courtney 2020). This study presents results of an ongoing pilot scale CW treating BR leachate for over 7.5 years.

Methods

Site Description.

A 4 m × 11 m pilot scale constructed wetland (CW) was constructed at a bauxite residue disposal area (BRDA) at Aughinish Alumina, Ireland. Local soil was transplanted for use as the substrate with the following characteristics: pH = 6.7, EC = 188μS cm^{-1}, CEC 38 cmol kg^{-1}, and 2.8% organic C content. The system was planted with Phragmites australis, Typha latifolia and Sparganium erectum supplied locally (FH Wetland Systems Ltd) and acclimatised with fresh water for 6 months.

A PLC mixing system was installed to feed the CW at a pre-determined pH and flow rate as per modelling of a closed BRDA (Residue Solutions 2007). The system consists of three 1000 L tanks. Alkaline leachate and water were pumped to a mixing tank until a target pH ca. 11 was reached. The mixed influent was then conveyed to a dosing tank and discharged to the CW at 45-55 L hr^{-1} (summer loading) and 10-30 L hr^{-1} (winter loading). Seasonal variation in loadings were due to increased winter precipitation. Initially, the system was operated using tap water to dilute the leachate, but due to the formation of calcite deposits within the mixing and dosing system that compromised efficiency, system was converted to utilise deionised (DI) water for leachate dilution (Higgins *et al*. 2018; Higgins *et al*. 2017). Additionally, the conversion to DI water allowed the investigation into the use of CW for a system which would have a low to negligible Ca content. The system has been receiving the deionised water leachate since May 2015.

Monitoring and Sampling Campaigns.

Inflow and outflow pH was measured daily using a field probe, with monthly averages reported over the 7.5-year study period. Wetland inflow and outflow samples were also taken for elemental analysis, filtered to 0.45 µm and determined by inductively coupled plasma mass spectrometry (ICP-MS).

Additional sampling campaigns were conducted to determine pH, EC and trace metal concentrations (Al, As, Ca, Mg, Na & V) within the wetland cell in the 4th and 5th years of operation in both the supernatant and sediments (air-dried and sieved <2 mm). Microbial analyses were also conducted in the 5th year. Detailed analyses from the 4th and 5th year are available separately (Hudson *et al*. 2023; O'Connor and Courtney 2020).

Results

Monthly average pH (Figure 1) was reduced from an influent pH ca. 11.5 to 7.1 over the 7.5-year study period. Within the additional sampling campaigns, it was shown that the reduction to pH < 9 threshold was obtained between 3.5 m and 5 m into the cell in the 5th year of sampling (Figure 2). The primary objective of the CW is to obtain a pH < 9, which the system has been successfully maintaining throughout its operation.

In addition to pH, EC and trace elements have also shown reductions from inlet to outlet. Reductions in average Al were 98% and 84% in year 4 and 5 respectively, with corresponding average V reductions of 90% and 57% respectively (Hudson *et al*. 2023; O'Connor and Courtney 2020). The dominant cation in the effluent across the 7.5 years was Na. Reductions in effluent Na have decreased over time, with a 31% removal reported over the first year by Higgins *et al*. (2017), versus 13% reported in year 5 (Hudson *et al*. 2023).

Sediment analysis indicated elevated trace element content in addition to microbial biomass in the first 5 m of the wetland. Trace elements Al, As and V were found to be mostly recalcitrant (>94% of total content), with Na content found to be mostly present in the water-soluble fraction (48–62% of the total). Microbial respiration rate utilising endemic Phragmites australis matter also showed an elevated rate in the first 5 m of the CW.

Discussion

Interest in CWs and implementation of nature-based treatment solutions has been increasing, with their potential to provide a cost effective and passive treatment solution appealing to operators. Most available studies are focused on acidic drainage sites and for short operating periods (<5 years) (Pat-Espadas *et al*. 2018).

The primary treatment objective for this CW was to reduce leachate pH to below a threshold of pH 9. The sustained decreases in pH over 7.5 years demonstrates that this CW has been successful in providing an effective treatment for alkaline BR leachate. Further investigations within the cell in the 5th year showed pH reduction to below threshold was achieved by 5 m (less than halfway) into the CW, implying sufficient capacity for another 7.5 years approximately. Obtaining pH < 9 threshold is required in several alkaline

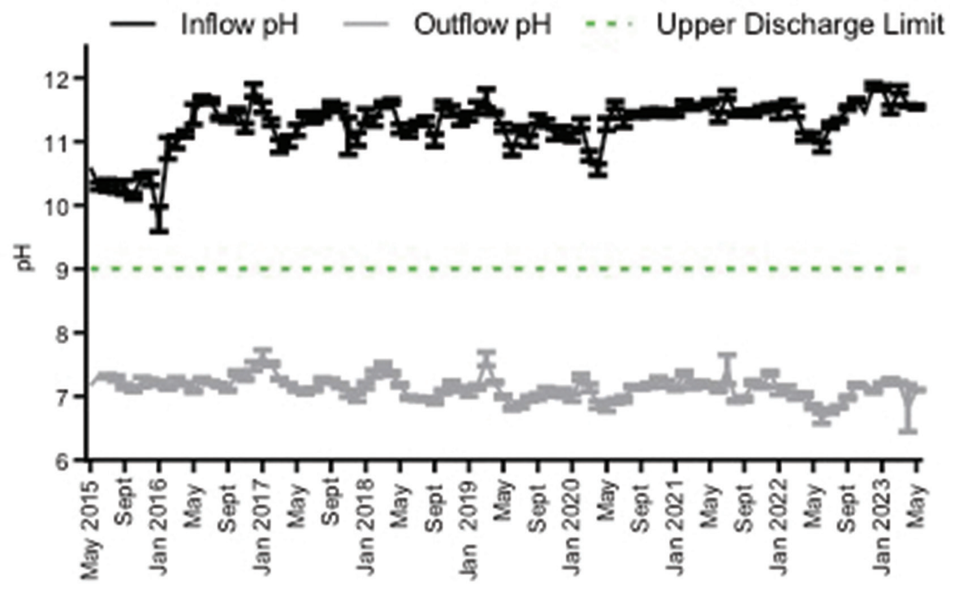

Figure 1 Average monthly inflow and outflow pH of a constructed wetland treating bauxite residue leachate, measured from May 2015 to January 2023.

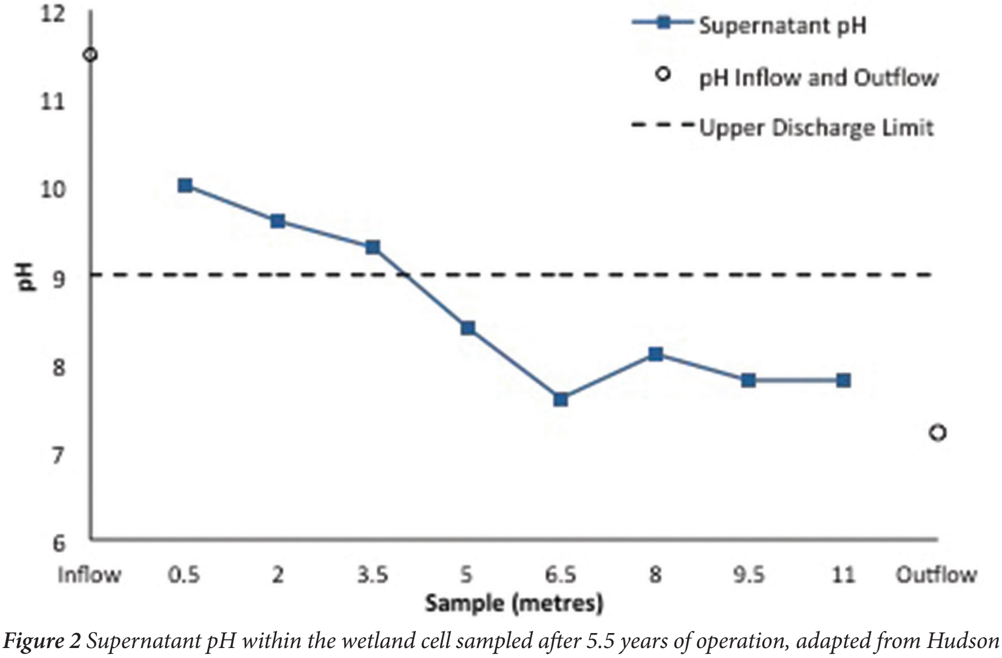

Figure 2 *Supernatant pH within the wetland cell sampled after 5.5 years of operation, adapted from Hudson* et al. *(2023).*

waste streams including steel slag leachates. Although CWs treating steel slag leachate have also achieved pH reductions of 2.5 units, from 12.8 to 10.3, they still exceeded the pH 9 threshold (Gomes *et al*. 2019). Mechanisms suggested to be responsible for pH reduction include microbial respiration, carbonation and the production of organic acids (Mayes *et al*. 2009b; Mayes *et al*. 2009a; O'Connor and Courtney 2020), and is supported by the elevated microbial respiration and biomass found in the first 5 m of the BRDA CW cell (Hudson *et al*. 2023).

Reduction in trace elements from the leachate demonstrates that CWs can also reduce contaminant load in BR leachate. Al and As removal of over 90% and 86% respectively is comparable with or exceeds that reported in CWs treating municipal wastewater (Kröpfelová *et al*. 2009; Vymazal and Krása 2003). V removal in this cell (57%) was also greater than that reported for steel slag leachate (Gomes *et al*. 2019).

Water quality has been the focus of most monitoring projects; however, CW sediment must also be investigated when considering the trace element removal. In this study, sediment Na has increased with time, particularly in the first 5 m of the wetland, following a front-loading pattern. Water-soluble Na increased from 9% in the baseline soils to >48% at all sampling points within the cell in the 5th year of operation. The reduction in Na removal from 31% to 13% could be attributed to the sediment base becoming saturated or the release of water-soluble Na due to displacement from the cation exchange surface. Although Na is elevated, effluent total Na remains below the 200 mg L-1 limit set by the EU Drinking Water Regulations (Environmental Protection Agency 2014). Further monitoring of the Na concentration in the cell sediment is needed to determine why Na removal rate has reduced, and to monitor its influence on nutrient uptake in vegetation (Ruyters *et al*. 2011).

Conclusions

This monitoring campaign provides evidence that a CW can successfully maintain pH<9 (from pH ca 11.5) in BR leachate for over 7.5 years. In addition, trace elements Al, As and V in the leachate were reduced, with sediment concentrations of these elements mostly in residual forms. To date, this study provides one of the longest datasets for CW treatment of alkaline leachates. Further monitoring is needed to assess performance as the system ages and to determine additional design optimisations that may improve its longevity.

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