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Use of Constructed Wetlands for Treating Mine Waste Leachates: Assessment of Longevity and Management Implications

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EPA RESEARCH PROGRAMME 2021–2030

Use of Constructed Wetlands for Treating Mine Waste Leachates: Assessment of Longevity and Management Implications

(2018-W-DS-32)

EPA Research Report

Prepared for the Environmental Protection Agency

by

University of Limerick

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The EPA Research Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.

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Executive Summary

Water resources are under continued pressure from a variety of industrial sources, including waste from abandoned mine sites. Huge quantities of mine waste and waste tailings are generated during mining operations and are typically stored in tailings management facilities. These storage facilities can, however, generate significant quantities of leachate, which, if released untreated to the aquatic environment, will contaminate surface and ground waters. It is anticipated that leachates from abandoned or closed facilities will be generated for multi-decades to centuries into the future. This presents a major long-term environmental challenge.

This project, which was carried out as part of the EPA Research Programme 2014–2020, examines the potential for constructed wetlands (CWs) to offer long-term treatment of mine waste leachates. It is primarily a desktop study, supplemented with field and laboratory analysis of a wetland field trial treating leachate waters with elevated Zn concentrations.

The three main objectives were to:

- document evidence where CWs have operated under field conditions;
- 2. identify limiting factors to long-term operation;
- identify suitable monitoring and management implications for CWs application.

Planning for the ultimate closure of modern mining and processing facilities recognises that water discharges from closed tailings management facilities will require treatment in the post-closure aftercare period. Traditional treatment methods, such as neutralisation, precipitation, adsorption, ion exchange, membrane technology and biological treatment, are not attractive in the long term because of sustained and intensive management requirements, energy inputs and operational logistics. Passive treatment methods offer an alternative approach, and CWs are widely cited as a potential treatment option; however, there are limited data on long-term field-scale applications. The application of CWs to treat mine water discharges has been documented since the 1980s, with most examples treating Fe and Al content in coal waste discharges in the USA and the subsequent implementation of the same in UK scenarios. CWs are efficient at removing Fe and Al loadings, with less success reported for Mn concentrations. Field examples of operating wetlands typically address operations over a period of 1-5 years and, although a number of studies indicate success in the longer 10-year time frame, these mainly address water quality. There is therefore a lack of data in the literature regarding the concentrations of contaminants present in wetland sediments and the forms in which the various metals exist in the wetland system. There is little information on the long-term effectiveness of wetlands in removing contaminants associated with mine tailings (Pb, Zn, Cu), although a number of short-term studies indicate effective removal rates for Pb and Zn. As with the coal waste examples, there is a lack of data on sediment quality in these wetland studies.

A 1-year monitoring study of a CW treating Zn-rich wastewater demonstrated the effective removal of Zn over this period but a follow-up assessment of sediments showed elevated total Zn content (Zn_{total}) in the front part of the CW system. Although the increased Zn content was not reflected in the macrophyte content, this study illustrates the need for sediment sampling to be included in CW monitoring and assessment.

On account of the multi-contaminant content of mine wastewaters, the implementation of a wider, more integrated, passive treatment approach may enhance the effectiveness and longevity of CWs. For example, using other passive approaches in conjunction with CWs can increase treatment effectiveness for parameters such as pH extremes, sulfate content and metals that are difficult to remove (e.g. Mn). Where the application of CWs is required, it is recommended that a full characterisation of leachates/waters is conducted to identify the contaminants of concern and inform suitable treatment options. Feasibility, including initial costs and occasional refurbishment costs, should be assessed over several years with monitoring and assessment of wetland waters and soils/sediments. Appropriate monitoring and management strategies should be identified.

1 Introduction

1.1 Mine Wastes

In excess of 6 billion tonnes of waste is generated annually by the mining industry (Table 1.1; Mudd and Boger, 2013) and this tonnage is projected to increase into the future. In the EU, for example, mine waste represents one of the largest waste streams, at 633 Mt per annum (Eurostat, 2020). Typically, mine waste is divided into two distinct physical forms: (1) coarse waste rock (usually a diameter of 2–20 cm) produced from the removal of unmineralised overburden and waste (or wall) rock with metal concentrations below grade cut-off (Tordoff *et al.*, 2000); and (2) tailings, the fine-grained (<2 mm) mixtures of crushed rock and processing fluids that remain after the extraction of economically viable metals (Lottermoser, 2010; Mudd and Boger, 2013).

1.2 Mine Tailings Production and Disposal

In modern facilities, the fine-particle tailings are typically removed from the process plant as a fineparticle, low-concentration slurry (suspension), and while some reuse is employed, significant amounts are disposed of to purpose-built tailings storage facilities or tailings management facilities (TMFs). Risks to the environment and human health are recognised where

Table 1.1. Estimated global production of mine wastes for selected commodities

Commodity	Tailings (Mt)	Waste rock (Mt)
Fe ore	700	6875
Coal	860	33,000
Bauxite	73	587
Cu	3290	8650
Pb/Zn/Ag	265	765
Ni	177	203
Au	1270	4844
U	61	112
Platinum group elements	89	98
Diamonds	85	366
Total	6870	55,500

Source: Mudd and Boger (2013).

incorrect disposal or poor management practices exist (Jones and Boger, 2012). While the implementation of sound waste management practices and closure planning has led to improved environmental performance, it has also highlighted the challenges for regulators and the industry regarding the long-term release of contaminated leachate from solid waste and tailings (Hansen et al., 2008). Furthermore, the global prevalence of inactive or abandoned mine sites with potential for the heavy metal contamination of watercourses and surrounding land is well recognised (Tordoff et al., 2000; Xie and van Zyl, 2020). It is anticipated that the contaminated wastewaters and leachates from abandoned or closed facilities will be generated in the timeline of multiple decades to centuries; this presents a major environmental challenge (e.g. Crafton et al., 2019).

1.3 Drainage Waters – Classifications

Leachates from water percolating through mine workings (pits and underground works), tailings and/or waste rocks can be contaminated with elevated metal content. Depending on the geochemistry of the tailings, mine drainage can have an acid, alkaline or neutral pH (Heikkinen *et al.*, 2009; Mayes *et al.*, 2009). Specific characteristics of the leachates will vary considerably and are strongly influenced by the geochemistry of mined wastes and deposits (Park *et al.*, 2019).

1.4 Acidic Leachates

Acid mine drainage (AMD) describes low-pH waters that are generated through a combination of chemical and biological processes converting sulfides in mine waste to leachate high in sulfates and with increased mobility of toxic metal(loid)s (Lottermoser, 2010; Skousen *et al.*, 2017). Sulfidic ores are usually chemically stable when *in situ* or when in saturated environments without contact with oxygen and water. However, exposure of sulfide tailings to oxygenated environments causes the sulfides to oxidise, producing acidic conditions with associated elevated levels of sulfate, heavy metals and metalloids (Lottermoser, 2010; Fernando *et al.*, 2018; Xie and van Zyl, 2020). Significant volumes of sulfidic wastes (e.g. tailings, waste rock) can result following the processing of sulfide-bearing ores, such as metallic ore deposits (Cu, Pb, Zn, Au, Ni, U, Fe), phosphate ores, coal seams, oil shales and mineral sands (Lottermoser, 2010; Xie and van Zyl, 2020).

Pyrite (FeS₂) is normally the most abundant sulfide mineral present, although other iron sulfides (e.g. marcasite, pyrrhotite) or sulfides having Fe as a major constituent (e.g. chalcopyrite) may be present (Lottermoser, 2010; Cerqueira *et al.*, 2012). As the most abundant of the sulfide minerals, oxidation of pyrite has been most studied and the direct and indirect oxidation processes are shown in equations 1.1 and 1.2, respectively (Lottermoser, 2010; Parbhakar-Fox and Lottermoser, 2017):

$$\begin{split} \text{FeS}_{2} (s) + \frac{7}{2} O_{2} (g) + H_{2} O (I) &\rightarrow \text{Fe}^{2+} (aq) \\ &+ 2 S O_{4}^{2-} (aq) + 2 H^{+} (aq) + \text{energy} \end{split}$$

$$\begin{array}{l} 4 \text{FeS}_2 \left(\text{s} \right) + 14 \text{O}_2 \left(\text{g} \right) + 4 \text{H}_2 \text{O} \left(\text{I} \right) \rightarrow 4 \text{FeSO}_4 \left(\text{aq} \right) \\ + 4 \text{H}_2 \text{SO}_4 \left(\text{aq} \right) + \text{energy} \end{array} \tag{1.2}$$

The rate of pyrite weathering (oxidation) is influenced by several factors, including its mineralogical properties (e.g. particle size, porosity, surface area, crystallography and trace element content) and external factors, including oxygen and carbon dioxide concentrations (Lottermoser, 2010).

Generation of AMD from pyritic tailings has been associated with elevated concentrations of metals in the surrounding water and terrestrial environment (Figure 1.1) and presents a significant environmental concern. In the USA, it is estimated that there are over 300,000 active and historical AMD sites (Wilkin, 2008). In the east of the USA alone, > 10,000 km of streams and > 72,000 ha of lakes and reservoirs have been adversely affected by AMD (Herlihy *et al.*, 1990). The estimated cost for the total worldwide liability associated with the current and future remediation of acid drainage is approximately US\$100 billion (Tremblay and Hogan, 2001).

1.5 Neutral Leachates

Generation of neutral pH leachate can occur when the waste material is low in sulfides, when sulfide oxidation is weak or when sufficient neutralisation



Figure 1.1. Example of AMD generation and colour leachate. Source: S. Callery.

occurs as a result of the carbonate content of the material (Heikkinen *et al.*, 2009). Although such waters can be characterised with a neutral pH range (c.pH 6–10), many potentially toxic metals, such as Ni, Zn, Co, As and Sb, are soluble at near-neutral pH and can potentially contaminate mine effluents. These are commonly referred to as contaminated neutral drainage or neutral mine drainage (Pettit *et al.*, 1999; Heikkinen *et al.*, 2009; INAP, 2009; Lindsay *et al.*, 2009).

Elevated concentrations of elements, such as As, Cr, Co, Cu, Fe, Mn, Ni, Pb and Zn, and sulfates in neutral to alkaline mine waters have been documented in several studies (Balistrieri *et al.*, 1999; Schmiermund, 2000; Younger, 2000; Ashley *et al.*, 2003; Rollo and Jamieson, 2006; Heikkinen *et al.*, 2009; Lindsay *et al.*, 2009; Sracek *et al.*, 2010). For example, pore water concentrations in carbonate-rich tailings in Lavrion, Greece, showed high concentrations of Cd, Pb and Zn (Xenidis *et al.*, 2003). High concentrations of elements were also found in seepage water from a waste pile in a carbonate-rich mining area in Guizhou, China (up to 6780 µg L⁻¹ and 324 µg L⁻¹ of Pb and Zn, respectively) (Zhang *et al.*, 2004). While the environmental impact associated with neutral drainage is often viewed as less severe than that associated with AMD, the potentially elevated metal contents still present risks that may continue for decades after mine closure (Pettit *et al.*, 1999; Heikkinen and Räisänen, 2008). Consequently, their management may call for the implementation of control and remediation measures comparable to those of highly acidic tailings (Heikkinen *et al.*, 2009).

1.6 Alkaline Leachates

Alkaline leachates, considered to be hyperalkaline at pH 10–12, are less common than those of an acidic nature. Alkaline leachates arise from the disposal sites of industrial by-products such as steel slag, chromite ore processing residue, bauxite residue, coal ash and municipal solid waste incinerator ash (Sakata, 1987;

Mayes *et al.*, 2008; Whittleston *et al.*, 2011; Gomes *et al.*, 2016; Higgins *et al.*, 2018; O'Connor and Courtney, 2020). The high alkalinity of such leachates is typically due to reagents used in the extraction processes, such as sodium hydroxide (NaOH) and lime (CaO), which can persist in the residue and release OH⁻ (Mayes *et al.*, 2008) (see equation 1.3).

$$Ca(OH)_2 \to Ca^{2+} + 2OH^- \tag{1.3}$$

Often, alkaline leachates are enriched with trace metals, such as As, Cr, Mo, Se and V, and present a pollution risk where fugitive emissions occur (Mayes *et al.*, 2006, 2008; Olszewska *et al.*, 2016; Higgins *et al.*, 2018; Hua *et al.*, 2018; Gomes *et al.*, 2019). Remediation options for alkaline leachate sites have historically comprised intensive and sustained capital input, such as aeration and acid dosing (Mayes *et al.*, 2008).

2 Environmental Pollution from Historical Sites

Although waters containing elevated levels of metals have been reported for closed mine sites (e.g. Herr and Gray, 1997; Aslibekian et al., 1999; O'Neill et al., 2015), these have mostly been for abandoned sites or closed sites with minimal remediation works. AMD can have a direct and dramatic impact on receiving watercourses, resulting in elevated metal concentrations in surface waters (Aslibekian et al., 1999), stream/river ecology and sediment loading (Herr and Gray, 1997; O'Neill et al., 2015). The low pH and osmotic potential of AMD, the presence of toxic metals and metalloids, and the formation and deposition of particulate materials can be toxic to aquatic organisms, resulting in the stress and death of indigenous populations (Johnson and Hallberg, 2005). AMD can ultimately lead to the precipitation of ochreous encrustations that degrade the aquatic habitat (Winland et al., 1991; Bigham and Nordstrom, 2000; Kirby and Cravotta, 2005). An estimated 1500 km of rivers in England and Wales have been polluted with a variety of metals, including As, Cd, Cu, Fe, Pb, Mn, Hg and Zn, caused by historical mines (Early, 2020). In Ireland, water contamination from historical mine sites occurs to a lesser extent than in England and Wales and can be locally problematic. Case studies of sediment and surface water contamination are noted below. Furthermore, the generation of metal-rich waters from historical mine sites is predicted to continue over multiple decades post closure.

2.1 Tynagh

At the legacy Tynagh site in County Galway, where mining activities ceased in 1980, assessment of surface water and sediment samples identified high levels of potentially toxic elements (O'Neill *et al.*, 2015). Concentrations of Cd, Ni, Mn, Pb and Zn in surface water ditches within the tailings facility and from adjacent streams exceeded guideline values (Government of Ireland, 1989; Government of Ireland, 2009a; O'Neill *et al.*, 2015). Additionally, the study found that sediment samples from nearby streams had concentrations of As, Pb, Cu, Cd, Mn, Hg and Zn that exceeded Canadian Soil Quality Guidelines (CSQG). The findings highlight the risk of surface water bodies as a significant pathway of contaminant transport to downstream areas, with high concentrations of minerelated metals detected almost 3 km downstream.

2.2 Avoca

Avoca, County Wicklow, has a long mining history and the most recent intensive mining for copper and pyrite recommenced at the existing sites during 1958-1962 and 1969-1982. Since the cessation of mining operations, leachate stream discharges from the abandoned mine to the Avoca river have resulted in severe contamination with toxic metals and the formation of ochreous deposits on the substrate, and this has also contributed significantly to the reduction of both invertebrates and vertebrates in the Avoca river. Gray (1998) calculated that the annual discharge of metals in AMD is in the region of 90t of Zn, 230t of Fe, 5t of Cu and 0.25t of Cd, and has resulted in Fe, Cu and Zn concentrations in sediment greatly exceeding average background levels for unpolluted waters (Herr and Gray, 1997). The impacts on freshwater ecology from metal loading to the Avoca river include ochre formation and the elimination of macrophytes, leading to loss of habitat diversity. Macroinvertebrates are largely absent, though tolerant species are present at some downstream sites. With the exception of small eels caught 10 km downstream of the mines, fish are absent along the 15.6 km length of the river (Gray, 1998). Further sampling of surface and sub-surface sediment metal concentrations in 2001 generally found no significant differences to those reported during the mid-1990s. The exception was the concentrations of Fe, which were significantly reduced and attributed to a significant reduction in the concentration of Fe discharged in the AMD (Gaynor and Gray, 2004). However, based on assessment of pyrite from the site it is predicted that AMD generation will persist well into the next century with the continuing pollution of the river system.

A. Hudson et al. (2018-W-DS-32)



Figure 2.1. Surface water ponding on tailings at the Silvermines site.

2.3 Silvermines

The Silvermines area in County Tipperary has a long and extensive history of mining, with the most intensive mining activity occurring between 1968 and 1982. A number of waste heaps, tailings facilities and mine adits still remain after the cessation of mining operations (see Figure 2.1). Monitoring of surface water at the abandoned mine site showed elevated concentrations of Pb (169 mg L⁻¹), Zn (82 mg L⁻¹) and Cd (0.6 mg L⁻¹), while analysis of the adjacent Yellow river found concentrations of Cd, Fe, Mn, Pb and Zn in exceedance of the maximum allowable concentration (MAC) (EU, 1975; Aslibekian *et al.*, 1999). Surface water from stream samples downstream of the abandoned mine site also contained Cd, Mn and Fe concentrations that exceeded MAC values. Elevated metal content in downstream sediments was also reported. For instance, high Zn concentrations in sediments (up to 1000 mg L^{-1}) were detected at a distance of 10 km from the mine. Soil samples taken in floodplains were found to be comparable to those from within the mine sites, with concentrations up to 20 (Pb) and 50 (Zn) times greater than threshold values (Aslibekian and Moles, 2003).

Some remediation of the Silvermines area was undertaken between 2006 and 2011, notably the rehabilitation of the surface of the Gortmore TMF. Treatment of contaminated water with passive treatment systems has been highlighted as a part of the overall rehabilitation planning and management of the site (Connelly *et al.*, 2005; Connelly, 2009).

3 Environmental Legislation and Management of Leachates

The EU Water Framework Directive 2000/60/EC (EU, 2000), which requires Member States to ensure that water bodies achieve good chemical and ecological status, is the overriding legislation that requires the mitigation of watercourse contamination. Environmental management and pollution control of mining operations, including waste arising, is controlled through Integrated Pollution Control (IPC) licences issued by the Irish Environmental Protection Agency (EPA). All current tailings facilities in Ireland are covered by IPC licences and before granting these the EPA must be satisfied that emissions from the activity will not have any significant adverse environmental impacts. Correct implementation of licences aims to prevent or reduce emissions to air, water and land, reduce waste and use energy/resources efficiently (EU, 1996).

Both the Directive on the Management of Waste from the Extractive Industry (EU, 2006) and the relevant Irish Statutory Instrument (Government of Ireland, 2009b) require that there is sufficient maintenance, monitoring and control and that there are corrective measures in place for the after-closure phase of a mine site. This includes an evaluation of the leachate generation potential from a waste site, including contaminant content of the waste leachate during the operational and after-closure phases of the waste facility. Furthermore, Member States are required to develop and periodically update an inventory of legacy sites "which cause serious negative environmental impacts or have the potential of becoming in the medium or short term a serious threat to human health or the environment" (Government of Ireland, 2009b).

Strategies are therefore required for preventing or minimising leachate generation and surface water or groundwater from being contaminated by the waste. The provision of adequate collection and treatment of contaminated water and leachate from the waste facility to the appropriate standard is required for their discharge. As the generation of leachates is anticipated to be in the multi-annual time frame, the potential for passive treatment approaches is desirable and the implementation of strategies such as soil cover or reactive barriers, to inhibit sulfide oxidation in the impoundment or to reduce and treat discharge loading from TMFs, needs to be considered. In addition, detailed site-specific studies are needed to describe the processes initiating the mine drainage to select the best practice for each mine site (e.g. Heikkinen and Räisänen, 2008). For licensed facilities in Ireland, tailings site operators have an obligation to plan and cost ultimate closure and to develop tailings rehabilitation approaches during the operational lifetime of the tailings facility. An example of such a licence condition for the tailings facility at Galmoy, County Kilkenny, is IPC licence condition 7.6.20, which states that "The licensee shall establish, monitor, and maintain for the duration of the mining operation a test facility for the purposes of validating the TMF closure proposals" (EPA, 2013). This should include rehabilitation of surface tailings and strategies for treating leachates and wastewaters.

Mining ceased at Galmoy in 2012, and this was followed by the restoration of the TMF. As part of the tailings restoration an integrated constructed wetland (CW) was implemented within the TMF footprint to treat any runoff from the closed TMF. In 2009, a wetland trial comprising two cells was constructed adjacent to the TMF. Both cells were planted with four types of wetland species [*Glyceria maxima* (main species), *Carex riparia, Cladium mariscus* and *Alisma plantago-aquatica*] and irrigated using water from the TMF spillway and surface runoff to assess their impact on the wetland plants and on the parameters of interest. See Table 5.6 for examples of its efficiency.

Modelling predicts that, following closure and rehabilitation of the tailings area at the Lisheen Mine, all water flow from the TMF rock fill cap will be suitable for discharge without any treatment. However, passive systems may be required over a longer period for water attenuation, and the possible need for treatment is also acknowledged (Lisheen Mine, 2016). Similar approaches have been implemented at the Tara Mines, where trial wetlands are being explored for treating waters from the tailings facility (Boliden Tara Mines, 2017).

4 Mine Waste Leachate Treatment

4.1 Active Treatment Technologies

Once generated, suitable remediation techniques are required for the treatment of mine waste leachates. Several active and passive techniques, such as neutralisation, precipitation, adsorption, ion exchange, membrane technology and biological treatment, are employed (Fernando *et al.*, 2018; Park *et al.*, 2019).

For active treatment technologies, large capital investments in material handling, equipment and the continuous supply of reagents, and long-term use of energy and/or maintenance are typically required (Chockalingam and Subramanian, 2006; Sheoran and Sheoran, 2006; Kefeni et al., 2017; Kaur et al., 2018). In addition, sludges, which are generated as a by-product from the treatment process, can contain elevated concentrations of potentially hazardous elements and may require disposal (Kefeni et al., 2017; Dhir, 2018). As the generation of leachates with elevated metal content is anticipated to continue in the timescale of multiple decades to centuries (Mayes et al., 2008; Whittleston et al., 2011; Gomes et al., 2016, Åhlgren et al., 2020), the application of active treatment methods becomes very costly and unsustainable in the long term (Gazea et al., 1996; Taylor et al., 2005).

4.2 Passive Treatment Technologies

Following mine closure, passive methods can be attractive for the treatment of mine drainage water, as they are anticipated to operate with an almost negligible requirement for maintenance and supervision. Compared with active treatment, less input is required for the construction, external energy and operational costs of passive treatment techniques or systems (Taylor et al., 2005; Sheoran and Sheoran, 2006; Hedin, 2020). Passive treatment relies on natural biological and biochemical processes to precipitate or otherwise remove dissolved metals, sulfates and other chemicals from water. These processes are passive insofar as they rely on gravity flow, require little or no addition of chemicals and require a low level of maintenance (Wiseman and Edwards, 2004); however, some level of monitoring

and possible maintenance is required to operate successfully over their design lives to maintain effective removal efficiency (PIRAMID Consortium, 2003; Johnson and Hallberg, 2005; Vymazal and Kröpfelová, 2008).

Acid drainage remediation by passive systems was first documented in the early 1980s during studies conducted by Huntsman et al. (1978) and Wieder and Lang (1982), which found amelioration of water quality in natural Sphagnum moss wetlands receiving AMD. These observations stimulated the idea that engineered wetland systems might be used for the intentional treatment of acid mine waters. During the last decade, much research has been conducted to evaluate the feasibility of these schemes. Most of these systems have been developed to treat acidic coal mine drainage (e.g. Hedin et al., 1994), and observations of macrophyte colonisation and improved water guality have led to the employment of wetlands as a method for treating leachate (Gazea et al., 1996; Mayes et al., 2009).

4.3 Constructed Wetlands

Several studies have shown the capabilities of CW systems as a strategy for treating and purifying wastewaters through interactions between the wetland's soil matrix, vegetation and microbial communities (e.g. Scholz and Lee, 2005; Moreau et al., 2013). The potential for natural wetlands to remove metals and metalloid contaminants from wastewaters has been extensively documented (e.g. Dinges, 1983; Sobolewski, 1999; Mayes et al., 2006; Vymazal et al., 2007) and they can also be optimised for one specific function at the expense of others (Mitsch and Gosselink, 2000). All components of a CW, such as flow, substrate, water depth and vegetation, can be adjusted or selected on the basis of site-specific geology, chemistry and hydrology, and this flexibility has facilitated the potential of CWs to perform specific, predetermined objectives at greater treatment capacities (Mays and Edwards, 2001). Pollutants can be removed through a variety of mechanisms, and each CW should be designed to match the treatment

requirements of each individual site. Therefore, CWs exploit the natural functions of wetland vegetation, soil media and their associated microbial populations to treat water (Knight and Kadlec, 2000; Johnson and Hallberg, 2005) and they present popular passive treatment strategies for mine water treatment (PIRAMID Consortium, 2003).

CWs can be distinguished from other passive treatment approaches by the incorporation of vegetation (e.g. PIRAMID Consortium, 2003; Pilon-Smits, 2005), which can contribute significantly to the metal removal processes (Ge et al., 2015; Hua et al., 2018). Typically, CW systems have shallow water depths (typically <250 mm) and can be categorised according to the various design parameters, including hydrology (open water-surface flow and sub-surface flow), type of macrophytic growth (emergent, submerged, free-floating and floating-leaved) and flow path in sub-surface wetlands (horizontal and vertical) (Vymazal and Kröpfelová, 2008; Kadlec and Wallace, 2009). For the purposes of this review, "CW" is used generically, as not all studies provide details on the specific wetland type in operation.

4.4 Metal Removal Mechanisms in Constructed Wetlands

Although wetlands are capable of removing large quantities of trace elements from wastewater, there is considerable variation among metals and between wetlands in the degree to which each metal is removed (Ye et al., 2001). The extent of metal removal using CWs is determined by a complex interaction of several processes, including settling, sedimentation, sorption, co-precipitation, cation exchange, photodegradation, phytoaccumulation, biodegradation, microbial activity and plant uptake (Sheoran and Sheoran, 2006; Kadlec and Wallace, 2009; Marchand et al., 2010). There are limited quantitative data available regarding the relative importance of the different mechanisms in the overall performance of the wetlands, and it can be difficult to identify the specific removal processes of metal contaminants within the wetland, as the processes are interdependent. The degree of removal or reaction will depend on the influent guality, composition of the wetland substrate and vegetation dynamics. The extent to which these reactions occur depends on the composition of the substrate, sediment pH, nature of wastewater and plant species. The

hydrology or hydraulic loading within the wetland system is also an important determinant supporting specific wetland processes (Kadlec and Wallace, 2009). Different removal mechanisms may also prevail in different parts of a wetland system. When metals are present in high concentrations, wetlands do not generally provide effective metal removal on account of minimal oxidation (Sikora *et al.*, 2000).

Lesley *et al.* (2008) suggested that Fe removal at the front end of the wetland system was due to oxidative processes and the formation of iron hydroxides, with an unclear mechanism in the latter stages. Removal of Fe depends on pH, the oxidation–reduction potential, the presence of various anions and sufficient retention time (Sheoran and Sheoran, 2006). Effective removal rates for Fe from coal mine discharges are frequently reported, both in the short term and over several years, and redox-sensitive Fe is preferentially removed as sedimentary sulfide minerals (FeS, FeS₂) (e.g. Hedin *et al.*, 1994; Younger, 2000; Younger *et al.*, 2002).

The rate of removal and retention of mine water contaminants existing as cations, for instance Cu, Zn, Pb, Ni and Cd, will be governed by the cation exchange capacity (CEC) of the wetland substrate, and can be increased in certain substrates with increasing clay and organic matter content (Matagi *et al.*, 1998; Sheoran and Sheoran, 2006). The retention of metals can differ; the retention of Pb, Cu and Cr by adsorption is generally greater than that of Zn, Ni and Cd (Sheoran and Sheoran, 2006). These weaker-held metals may therefore be more labile and bioavailable.

Metals such as AI and Mn may be removed through hydrolysis and/or oxidation and through the formation of insoluble oxides, oxyhydroxides and hydroxides (Sheoran and Sheoran, 2006; Kröpfelová *et al.*, 2009; Marchand *et al.*, 2010). AI removal is governed by pH (Hedin *et al.*, 1994; Higgins *et al.*, 2018) and a 97% removal efficiency of AI in a wetland resulted in increased concentrations in carbonate, oxidebound and organically bound AI in the substrate (Higgins *et al.*, 2017). Owing to its complex chemistry, removal of Mn from mine waters is more challenging than for many other metals (PIRAMID Consortium, 2003), with the removal efficiency influenced by temperature, Fe removal, pH and Ca content (Matthies *et al.*, 2010). In a study of a full-scale passive treatment system to examine metal removals from acidic coal mine discharges, Matthies et al. (2010) reported a removal efficiency for Fe and AI of 84-87%, whereas for Mn it was 23% in the first 2 months of operation; thereafter, however, the concentration in output waters exceeded that of the input. Similarly high removals (c.98%) have been reported for Cd, Cu, Pb and Zn, with much lower rates observed for Mn (Reisman et al., 2009; Neculita and Rosa, 2019). Removal of Mn is reported to be between 20 and 40 times slower than for Fe in a range of systems (Hedin et al., 1994; Lesley et al., 2008) and is generally relatively weakly and sometimes reversibly sorbed (Wieder, 1993). Thus, the provision of a wetlands system that has a size 20-40 times that required to remove Fe under similar conditions has been suggested for the removal of Mn (Hedin et al., 1994; Lesley et al., 2008). Alternatively, for improved efficiency it has been suggested that Fe and Al are removed first, followed by the removal of Mn in a final treatment cell (Sikora et al., 2000; Skousen et al., 2017).

4.5 Hybrid Passive Systems

Different passive treatment strategies are often combined to utilise specific mechanisms and maximise

contaminant removal; thus, the lifetime of CWs may be enhanced by the incorporation of other passive technologies within the overall treatment system (Vymazal, 2005; Gusek, 2008). Treatment of coal mine waste discharges in the UK has seen the installation of around 70 passive (and hybrid passive) treatment systems (Coal Authority, 2017). Integrated hybrid passive treatments using wetlands as a component have been employed and incorporate sedimentation tanks and various wetland types, for instance anaerobic, aerobic, and reducing and alkalinityproducing system (RAPS) (e.g. PIRAMID Consortium, 2003; Wiseman and Edwards, 2004; Matthies *et al.*, 2010).

In the treatment of alkaline leachate, the implementation of cascade systems such as those reported by Gomes *et al.* (2017) can reduce pH in alkaline leachate by 1–1.5 pH units and offer a potential additional treatment to the wider passive technology approach. Carbonation of bauxite residue leachate can consume alkalinity through the precipitation of carbonates and generate conditions conducive to (contaminant) metal co-precipitation (Higgins *et al.*, 2018).

5 Evidence of Long-term Use of Constructed Wetlands for Metal Treatment

A literature review was conducted to document evidence of CWs operating over time frames greater than 12 months and under field conditions. The accompanying database (available at www.epa.ie) documents these studies and this chapter summarises the findings.

5.1 Coal Waste Leachate

A number of passive treatment approaches, including CWs, have been implemented at several sites globally since the 1980s (e.g. Brodie, 1988; Hedin et al., 1994; Gazea et al., 1996; Stottmeister et al., 2006). Initially, many were constructed at sites in the USA for treating coal waste leachates. An overview provided by Hedin et al. (1994) reported an effective treatment for a number of wetlands treating Fe over a time frame of 2–10 years (Table 5.1). Based on criteria for coal mine treatment, the service life for the systems was estimated at 20 years. Similarly, Brodie et al. (1988) reported the effective Fe removal in a range of wetlands for up to 12 years; however, most of these were short-term treatment operations of 1-3 years, with only one site operating over a longer time frame (12 years) (Table 5.1). These studies chiefly address the removal of Fe and Mn from coal waste discharges

and report high rates of Fe removal at a range of sites. Removal of Mn is much more variable. Although some sites recorded high Mn removal rates (80–98%), low rates were also reported (8.8–14%). Furthermore, some sites showed the wetland system to be a net exporter of Mn (Table 5.1). The data summarised address the metal contaminant removal, but no information on changes in the wider wetland system, soil substrate and vegetation was reported.

In a study to investigate the performance of a CW to treat coal waste leachate over a 10-year period, Ye et al. (2001) reported the effective removal of a range of trace elements with maximum concentration reduction rates of 99% for Fe, 91% for Cd, 63% for Zn, 61% for S, 58% for Mn and 50% for B. Seasonal variation in metal loading and corresponding removal efficiencies of 34-63% for Zn, 7-33% for Mn, 62-99% for Cd and 54-99% for Fe were also reported. Sediment sampling from the two wetland cells in the same study showed higher concentrations for Fe, B, Cd and Zn in the upstream cell (cell 1) than in the downstream cell (cell 2). Separation of sediment into three different depths (0-5, 5-10 and 10-15 cm) also found the highest levels of metals in the 0-5 cm fraction. It was determined that the wetland sediment was a major pool of trace elements removed by the

Age of CW at time of	Fe (mg L⁻¹)			Mn (mg L⁻¹)		
report (years)	Influent	Effluent	Decrease in Fe (%)	Influent	Effluent	Decrease in Mn (%)	
2	150	6.4	95.7	6.8	6.2	8.8	
1	153	0	100	4.9			
3	135	3	97.8	24	4	83.3	
1	11	0.5	95.5	9	0.2	97.8	
1	45.2	0.8	98.2	13.4	0.2	98	
2	40	3.4	91.5	13	14	-7.7	
2	13	0.8	93.8	5	1.9	62.0	
2	17.9	3.3	81.6	6.9	5.9	14.5	
12	12	1.1	90.8	8	1.6	80.0	
3	30	0.9	97.0	9.1	2.1	76.9	
1	0.7	0.7	-	5.3	13.5	-154.7	

Table 5.1. Summary of Fe and Mn removal in CWs from US coal mine sites

Sources: Brodie et al. (1988); Hedin et al. (1994).

wetlands. The authors concluded that the CWs were still able to efficiently remove metals in the long term (i.e. > 10 years after construction).

Similarly, Hedin (2020) reported a sustained metal removal efficiency over 14 years (2004–2018) for two different passive treatment systems comprising vertical flow ponds (VFPs) and CWs treating metal-rich acidic leachates from coal waste (Table 5.2). System 1 comprises four parallel VFPs, followed by a single polishing pond, whereas System 2 treats water with four parallel VFPs, followed by a series of three CWs. Greater treatment efficiency is evident for effluent waters from the CW than from the polishing pond system.

Passive treatment technology for mine waters was introduced to the UK during the 1990s, using guidelines developed in the USA (e.g. Hedin et al., 1994; Younger, 2000). As with the US examples, CWs have mainly been used for coal mine drainage. CWs (passive treatment technologies) for the remediation of discharges from active and abandoned mines in the UK were implemented at 25 sites (Younger, 2000), with examples shown in Table 5.3. The Fe concentrations in coal wastewaters reported by Wiseman (2002) demonstrate the effectiveness of CW in treating Fe removal over a 2-year period (Figure 5.1), with removal rates of 82–96% reported. Effective metal removal using wetlands at the former Whitworth facility over a longer time frame of 7 years was also reported (Wiseman, 2002). The system comprised four different wetland cells operating either aerobically or anaerobically and planted with Juncus or

Table 5.2. Average metal content (mg L⁻¹) of effluent waters from two treatment systems between 2004 and 2018

Metal	Influent	VFP effluent	Final effluent
System 1			
Fe	5.9	5.3	1.1
Mn	7.2	6.2	3.3
AI	10.7	0.4	0.3
System 2			
Fe	32.6	17.3	0.5
Mn	6.0	5.0	1.9
AI	30.7	0.5	0.2

Source: Hedin (2020).

Typha. Summary data (Table 5.4) demonstrate overall decreases in dissolved Fe from around 21.8 mg L^{-1} to just over 3 mg L^{-1} (86% decrease) and an AI removal

Table 5.3. Summary of Fe loading and removal from passive treatment approaches, including CWs treating coal wastewater

	Fe conc (mg L⁻¹)	entrations	
Site	Inflow	Outflow	Removal (%)
Pelenna III	69	4.2	93.9
Gwynfi	7	<1.0	85.7
Nailstone	46	14.6	68.3
Dodworth	30	<1	93.9
Woolley Colliery	10	<1	93.9
Old Meadows Drift	5	<1	93.9
St Helen's Auckland	3	3	93.9
Bowden Close	3	3	93.9
Bowden Close ^a	45.3	5.9	93.9
Edmondsley Yard Drift	27	1	93.9
Quaking Houses	10	2	93.9
Oatlands	85	25	93.9
Shilbottle	100	25	75.0
Dalquharran	200	50	75.0
Monktonhall	10	<1	
Minto Colliery	18	<1	
Blairingone	38	2.3	

^aFrom Matthies *et al.* (2010). Source: Younger (2000).





Table 5.4. Changes in metal content (mg L⁻¹) through the Whitworth treatment system (1995–2002)

Source	Mn _{total}	AI _{total}	Fe _{total}	Fe _{dissolved}
Mine water	1.8	0.41	23.3	21.8
Outlet	1.1	0.09	3.81	3.1

Source: Wiseman (2002).

rate of 80.5%. Removal of Mn was much lower, with an overall removal rate of 39%. The most efficient cells were those operating aerobically and resulting in the precipitation of predominantly metal oxides.

Similar reductions in Fe content in wetlands operating over 2–6 years were reported by Younger (2000) and are summarised in Table 5.3. The reductions show general agreement with US data (Hedin, 2020). Further investigations of the Quaking Houses site demonstrate seasonal variation in the removal rates for Fe of <10% up to 90% over a 14-month period (Jarvis and Younger, 1999; Batty and Younger, 2004), with averages of 45% for Fe and 63% for Al. Similar seasonal variation in removal was also reported by Ye *et al.* (2001).

Monitoring of waters at the closed Bowden Close facility by Matthies *et al.* (2010) indicated that removal rates of the main contaminants, although seasonal, were constant over 5 years of monitoring. However, compared with data presented by Younger (2000), the Fe concentration in influent water was much higher, at 45 mg L⁻¹, which decreased to 5.9 mg L⁻¹ in the wetland effluent (equating to a 87% removal rate) (Table 5.5). Monitoring over the period 2003–2009 demonstrated that average water qualities were also improved for Al (90%) and Zn (57%), with no overall reductions in Mn.

Table 5.5. Summary of average influent and wetland effluent metal concentrations (mg L⁻¹) at the Bowden Close facility

Metal	Influent	Wetland effluent	Removal (%)
Fe	45.3	5.9	87
AI	19.6	1.9	90
Mn	4.4	4.4	0
Zn	0.7	0.3	57

Source: Matthies et al. (2010).

Generally, the literature for coal waste leachate reports high removal rates for Fe (typically around 70% to >90%), with less efficiency for Mn (8–90%). Indeed, at some sites higher Mn concentrations were found in the wetland outflow than in the inflow. This suggests that the CW systems for coal leachate treatment may be more suited to Fe removal and that a complementary treatment system may be required for the removal of Mn.

Specific removal mechanisms, particularly the possible accumulation of trace elements in the wetland substrate sediment and vegetation, are not always reported; however, this type of data is significant when assessing the longevity of CW systems. For example, the findings of Ye et al. (2001) indicate the potential accumulation of trace elements in wetland sediments, with trends of front-loading evident. Wiseman et al. (2002) and Matthies et al. (2010) concluded that the inclusion of a routine maintenance programme is an essential component for future treatment schemes. Similarly, the long-term (c.15 years) performance reported by Hedin (2020) included continued monitoring and maintenance at the site where organic substrates were periodically inspected to assess their reactivity and replaced before their failure. However, no further detail on the inspection criteria or frequency of the replacement was included in this study.

5.2 Other Mine Waste Leachates

5.2.1 Lead/zinc tailings

The efficacy of wetlands to treat waters from Pb/ Zn tailings have been reported at the trial stage and for closed facilities. In a pilot trial operating over a 3-year period, O'Sullivan *et al.* (2004) reported that Zn was removed by up to 70 mg m⁻² day⁻¹ (99% of the loading) and Pb by up to 6.6 mg m⁻² day⁻¹ (64% of the loading). Similarly, Devoy *et al.* (2018) reported sustained removal rates for Pb and Zn to well within the emissions limit value (ELV) over a 3-year period for a wetland installed as part of a planned closure at a Pb/Zn tailings facility (Table 5.6). The wetland reported by Devoy *et al.* (2018) is reported to have a long lifespan design (50–100 years), with capacity to deal with high variations in hydraulic loading.

Evidence for longer-term efficiency for wetland treatment of Pb and Zn tailings leachate was reported by Yang *et al.* (2006). Here, 10 years of monitoring

	Discharge quality (μg L⁻¹)				Removal (%)		
Metal	2015	2016	2017	ELV (µg L⁻¹)	2015	2016	2017
Pb	1	1	1	7.2	85	90	94
Zn	32	22	56	100	99	99	98

Table 5.6. Wetland discharge quality and efficiency results for Pb and Zn (2015–2017)

Source: Devoy et al. (2018).

data for a CW system in Fankou, Guangdong Province, China, demonstrated long-term success in decreasing metal concentrations from Pb/Zn waste. Data summarised in Table 5.7 show high removal efficiency for both Pb and Zn, with lower rates recorded for Hg. Interestingly, concentrations determined in outlet water were below the environmental standards set by the regulator (Chinese environment quality standard).

Although not designed as a wetland treatment facility, macrophyte colonisation of surface spillways surrounding the historical Pb/Zn TMF facility at Gortmore, Silvermines, Ireland, indicates successful treatment of metal waters emanating from tailings. Figure 5.2 shows the data over a 6-year period from runoff and seepage water reported for the wetland area (inflow) as well as the outflow data for two subsequent wetland cells. As highlighted in section 2.3, surface water and soils surrounding the Gortmore TMF show evidence of historical contamination from mine workings. It is worth noting that the TMF received substantial remediation works between 2006 and 2011, including the installation of a capping and soil cover on much of the tailings surface. Effective remediation of tailings/waste pile surfaces has implications for both the volume and metal loading of waters requiring treatment.

5.2.2 Tin/zinc tailings

Wetlands have been used to treat leachate at former mine sites where closure has been conducted in the absence of research-driven remediation or where legacy remains. At the Wheal Jane, Cornwall, UK, Sn and Zn facility, Younger (2000) reported significant removal rates for As and Fe, with lower rates for Cu and Zn. As with results reported for coal wastes, the Mn removal rate was low at 20% (Table 5.8).

5.2.3 Nickel waste

Eger and Kairies Beatty (2013) presented data for a wetland system for treating Ni in leachate (influent Ni concentration of $c.5.1 \text{ mg L}^{-1}$), which achieved removal rates of greater than 90% during the first 3 years of operation. The Ni removal capacity in the wetland peat substrate was calculated at 1400 kg and by dividing this by the average annual Ni input (170 kg y⁻¹) a lifetime of 8 years was calculated. Subsequent capping of the stockpile decreased the leachate inflow and Ni concentration entering the wetland, resulting in a 90% decrease in Ni loading, and the lifetime of the wetland was revised upwards to 100 years.

In a study assessing Sb removal efficiency in two peat-based natural wetlands operating for 8 and 10 years, Khan *et al.* (2020) reported that the wetland

	Element								
Sample	Pb	Zn	Hg	Cd	As				
Inlet (mg L ⁻¹)	11.49±7.70	14.47±8.37	0.0003 ± 0.0002	0.05±0.04	0.077±0.024				
Outlet (mg L ⁻¹)	0.11 ± 0.05	0.39 ± 0.17	0.0001 ± 0.0000	0.003 ± 0.001	0.016±0.010				
Removal rate (%)	99	97	71	94	79				

Table 5.7. Summary data (mean±standard deviation) for metal concentrations in wetland inlet and outlet samples and average removal rates for CW treating Pb/Zn tailings leachate (1991–2000)

Source: Yang et al. (2006).



Figure 5.2. Metal removal over a 6-year monitoring period from the "wetland system" at the legacy Gortmore tailings facility: (a) Pb, (b) Zn, (c) Cd and (d) Ni. Data from CDM Smith (2018).

	Typical influent	Typical effluent	
Element	(mg L⁻¹)	(mg L ⁻¹)	Removal (%)
As	2.7	0.01	99.6
Fe	141	19	86.5

45

20

0.2

43

50

20

Table 5.8. Summary data for trace element removalat the Wheal Jane Sn and Zn facility

Source: Younger (2000).

79

24

0.4

Zn

Cu

Mn

operating for 8 years had a mean removal efficiency of $67\% \pm 32\%$, compared with $14\% \pm 32\%$ in the older wetland. This was attributed to the lower loading to the younger wetland, typically < $100 \ \mu g \ L^{-1}$ for the younger wetland compared with typically > $200 \ \mu g \ L^{-1}$ for the older wetland. A reduction in treatment capacity over time was also noted for the younger wetland, with the Sb having a removal efficiency of > 80% in the first 3 years of operation, which declined to about 75% thereafter. Although not from mine discharge sites, evidence provided over a 2-year period by Kröpfelova *et al.* (2009) evaluated the removal of trace elements from municipal sewage in three horizontal flow CWs in Czechia (Table 5.9). In a separate CW study to treat motorway runoff, Gill *et al.* (2017) reported Pb removal efficiencies of 31%; however, earlier studies showed a removal efficiency of 86% when sampling inlet and outlet effluent from discrete storm events.

5.2.4 Bauxite residue leachate

The use of CW for buffering alkaline pH and removing trace element content in bauxite residue leachates has recently received attention (Buckley *et al.*, 2016; Hua *et al.*, 2015, 2018; Higgins *et al.*, 2018; O'Connor *et al.*, 2020).

The potential of CW to buffer alkaline pH in the long term, c.5+ years, was evidenced in field trials (O'Connor *et al.*, 2020), where the inflow of pH 9.2–12.2 was consistently decreased to pH 6.6–8.3. Moreover, by the fifth year of operation,

	CW 1		CW 2	CW 2			
Element	Inflow	Outflow	Inflow	Outflow	Inflow	Outflow	Removal efficiency (%)
Al	3748	328	5658	84	938	186	80.2–98.5
Zn	186	26	232	22	72	30	58.5–90.5
Pb	13.2	2.9	15.6	2.46	3.66	2.72	25.7–84
Cd	0.33	0.1	0.32	0.07	0.1	0.1	0–78
Cr	11.3	3.66	6.75	1.99	2.88	2.1	27–70

Table 5.9. Removal of selected trace elements (µg L⁻¹) from municipal sewage in three CWs during the period March 2006–June 2008

Source: Kröpfelova et al. (2009).



Figure 5.3. Al and V content in wetland inflow and outflow samples in years 1 and 5 of operation [based on data from Higgins *et al.* (2018) and O'Connor and Courtney (2020)].

levels of the trace elements AI and V showed an effective and sustained decrease, with removal rates of 96% and 98% for AI and of 86% and 90% for V in years 1 and 5, respectively (Figure 5.3). Similarly, effective AI removal rates are reported for other wetlands (e.g. Kröpfelová *et al.*, 2009).

Intervention and/or management may be required should metals accumulate in wetland sediments to levels exceeding threshold guideline values (Gomes *et al.*, 2019). After 5 years of operation, the values for AI_{total} and V_{total} in wetland sediment were not deemed excessive and there was no evidence of increased bioaccumulation in wetland vegetation (O'Connor and Courtney, 2020).

5.3 Case Study – Constructed Wetland Trial for Treating Zinc Tailings Leachate

A CW trial was implemented to investigate the potential to treat elevated Zn content from tailings waters. The trial wetland was composed of four cells, each 60 m^2 , and planted with *Glyceria maxima* and *Typha latifolia*. The inflow and outflow of the wetland complex were monitored over a 14-month period (2019–2020) for Zn content (Figure 5.4). After this period, the wetland continued to operate for a further year before a series of sediment (at depths of 0–10 cm) and vegetation samples were taken from within the wetland cells. Samples were taken



Figure 5.4. Inflow and outflow Zn content from trial wetland treating tailings water over a 14-month period.

from the front (F) and back (B) sections of each cell (Figure 5.5).

5.3.1 Inflow and outflow zinc concentrations

Over the 14-month period, concentrations of inflow Zn ranged from 2.4 to 8.6 mg L^{-1} , while outflow was

in the range $0.01-1.13 \text{ mg L}^{-1}$, resulting in an overall reduction of approximately 96% (Figure 5.4). This is consistent with other reported reductions in Zn loading to wetlands (O'Sullivan *et al.*, 2004; Yang *et al.*, 2006; Devoy *et al.*, 2018). The Pb content in inflow waters was below the limit of detection (data not shown).



Figure 5.5. Selected soil physico-chemical parameters of soils from four wetland cells: (a) soil pH, (b) soil salinity measured as electrical conductivity, (c) soil CEC and (d) per cent carbon (C %).

5.3.2 Sediment and vegetation

Following 2 years of receiving tailings leachate, there was no discernible effect on soil pH (Figure 5.5a).

The lowest electrical conductivity was recorded for the fourth cell, but in the absence of baseline data it cannot be determined if the higher values in cells 1–3 were the result of the leachate inflow Zn content (Figure 5.5b). Within the wetland cell system, most of the Zn removal appears to occur in cell 1, which is consistent with other studies (e.g. Jordan *et al.*, 2021).

The Zn_{total} content in the CW soil (Figure 5.6a) was elevated in (1) cell 1, where it exceeded the guideline limit values for spreading of sludges and metal accumulation (Government of Ireland, 1998), and (2) the front section of cell 2. For some samples, the Zn_{total} content is close to or higher than the Dutch intervention levels of 720 mg kg⁻¹ (MHSPE, 2000).

Most Zn removal in wetlands is attributed to hydrous metal oxides of Mn and Fe precipitating under reducing conditions (Reddy and DeLaune, 2008). Zn is also known to interact with hydrogen sulfide to form zinc sulfide (Kadlec and Wallace, 2008), and further study is warranted to examine Zn fractionation studies in the sediment. The amount of exchangeable Zn (Zn_{exchangeable}) ranged from 0.004 to 0.04 meq 100 g⁻¹ (Figure 5.6b) and represented 2.3% of the total Zn content overall, with a maximum of 5.7% Zn_{exchangeable} in cell 1B. Although this represents an available form of Zn, the elevated content in the sediment samples is not reflected in the plant content, where Zn levels remained low and consistent across all cells (Figure 5.6c). Zn_{exchangeable} also represents < 0.05% of the effective CEC.

Finally, the carbon content (% C) in soils remained relatively consistent across the CW system, in the range of 6–8%. The wetland soil CEC was significantly correlated with the carbon content (r=0.72), indicating that organic matter supply may contribute a role to metal absorption.

In summary:

- Wetlands were effective in reducing Zn loading over a 14-month period.
- There was an elevated Zn_{total} content in the front cell of the wetland after 30 months of operation.
- There was no trend of changes in soil pH, electrical conductivity or CEC following the operation period.
- There was no trend of elevated Zn content in wetland vegetation.



Figure 5.6. Zn content in wetland cell soil and vegetation samples: (a) total Zn, (b) exchangeable Zn, (c) *Glyceria* sp. vegetation. Note, cell 2B is *Typha* sp.

6 Wetland Longevity

The operational lifetimes of wetlands for treating mine waters are highly variable and depend on wetland dimensions and type, the influent water quality and loading rates, and the specific mechanism of metal removal processes (Sheoran and Sheoran, 2006; Kadlec and Wallace, 2009; Marchand *et al.*, 2010; Palmer *et al.*, 2015).

The lifespan of CWs is often based on the predicted saturation indices for the contaminant of interest and, although removal processes vary according to metal types, typically they are a combination of precipitation, adsorption, ion exchange and complexation with the organic substrate (Eger et al., 1994; Kadlec and Wallace, 2008). Depending on substrate type, Wieder (1993) reported a removal efficiency over 2 years of 9-81% for Fe and of between -28% and 56% for Al. Most removal occurs in the top 20 cm of the wetland substrate (Eger and Kairies Beatty, 2013; Eger et al., 1994), and removal performance is governed by the number of available removal sites within the rhizosphere. For sustainable and effective treatment, it is essential that new removal capacity is generated within the wetland so that the metals can be retained within the substrate.

Mass balances calculated on wetland test cells demonstrated that >99% of the removed metals were associated with the substrate and that <1% of the total removal occurred in the vegetation (Eger *et al.*, 1994). Sequential extraction tests, conducted on a series of substrate samples collected from test cells constructed at the Dunka Mine, demonstrated that only 1–2% of the Ni was water soluble and could, therefore, be easily removed from the substrate (Eger *et al.*, 1994). Higgins *et al.* (2017) reported that the content of Al, As and V in wetland soil receiving bauxite residue leachate after 1 year was mostly in the recalcitrant fractions (oxide, organic, residual).

While a nominal design life of 15–20 years for CWs treating coal mine discharges has been proposed (Hedin *et al.*, 1994; PIRAMID Consortium, 2003; Sheoran and Sheoran, 2006), it is recognised that many leachate sources will comprise a range of elements of varying concentration and be subject to different removal mechanisms. Furthermore, variation

in seasonal performance has been highlighted at a number of sites (e.g. Matthies *et al.*, 2010).

The number of active adsorptive sites can increase or decrease over time, depending on, for example, the quality of influent (Ronkanen and Kløve, 2009; Palmer et al., 2015) while removal efficiencies may decrease if the retention capacity of the wetland is exceeded (Wieder, 1993). Eger and Wagner (2013) reported that peat accumulation of 1 mm per annum for northern wetlands added an additional 7 kg of Ni removal capacity each year, and when coupled with the remediation works on reducing load from the waste pile, that lifetime of the wetland was further revised upwards to about 290 years. Similar long-term time frames have been proposed for sediment within a wetland receiving wastewater from an abandoned Pb-Zn mine (Beining and Otte, 1996). In this study, it was calculated that after c.120 years of metal loading to a natural wetland system, only up to 30% of the estimated total capacity of the substrate to retain metals had been used and longevity of the system was predicted to be in the order of centuries.

In a study of an aerobic wetland with a limestone bed to treat coal mine-derived AMD in the Huff Run watershed of eastern Ohio, USA, over a 13-year monitoring period, consistent performance of Fe and Al removal was partly attributed to circumneutral pH of emergent water (compared with acid pH inflow) (Crafton *et al.*, 2019). Although the system received no maintenance in the timeline, the potential to continue treatment in the longer term was uncertain.

Lesley *et al.* (2008) investigated a wetland treating coal leachate at Whittle, UK, and reported a 98% Fe removal efficiency after 2 years, a slight improvement on the 97% removal reported for its first year of operation, and demonstrated continued function. However, the area-adjusted removal rate was less than reported for other wetlands by Hedin *et al.* (1994) and was attributed to a larger "over-engineered" wetland system in the Whittle study. Furthermore, the wetland with the greater removal rate in the study by Hedin *et al.* (1994) was supplemented by additions of limestone and/or organics. Some studies indicate that CWs have a finite lifespan with respect to metal retention and that they could eventually fail to remove some elements (Weider, 1993; Horne, 2020). For example, the capacity of wetlands to retain Fe (primarily as oxides) might eventually be exhausted (Stark et al., 1995) and while summary data presented in Chapter 5 show the effective removal of AI and Fe content in wetlands, low values for Mn removal were typically reported. Removal of Mn in CWs can be complex, with some reports of higher concentrations in the outflow than the inflow (Matthies et al., 2010). The incorporation of limestone beds was proposed as necessary for Mn removal (Crafton et al., 2019). Although the causes of increased Mn levels in outflow waters are uncertain, they may be partly due to variability in the inflow quality (Neculita and Rosa, 2019).

Khan *et al.* (2019) noted the risk of Sb mobilisation and leaching from peat sediment, as evidenced by the decrease in its concentration in surface samples. This, along with the decline in the capacity of peatland to absorb Sb concentration peaks, suggested that the removal process should not be used indefinitely for treatment of Sb-contaminated waters, for example as a long-term treatment system after mine closure. Large areas and small input loads are recommended in particular for long-term metal removal in CWs (Khan *et al.*, 2020), but a sudden improvement in inflow water quality after a number of years can lead to deterioration of treatment efficiency, even in large wetlands. Conversely, Hedin (2020) reported long-term success in treating metal removal over a 14-year period and attributed this, in part, to improvements (i.e. a lower metal content) in the influent water and lower contaminant loading to the systems.

Palmer *et al.* (2015) found that the fraction of easily mobilised (i.e. available) metal in peatlands after 7 years of operation varied depending on the element. For As, 9–40% of the total amount was available whereas for Sb and Ni this was in the range of 3–10% and 6–9%, respectively. Khan *et al.* (2019) reported leaching of As from soils and suggested that it was symptomatic of the decreasing As-retaining capacity of peatlands.

The accumulation of metals within wetland sediment typically necessitates intervention and management, and the lifespan of a wetland could be assessed by considering guideline values (Palmer *et al.*, 2015). Palmer *et al.* (2015) determined metal levels in wetland soils. By comparing these with lower guideline values, they predicted an exceedance for As and Ni within 1 and 11 years, respectively; using higher guideline values for As, Sb and Ni, they predicted



Figure 6.1. Accumulation of iron oxide and hydroxide on the surface of a RAPS. Reproduced from Wiseman (2002). Contains Environment Agency information. © Environment Agency and database right.

exceedance within 6, 10 and 18 years, respectively. Similarly, sediment from wetlands used to treat steel slag leachate exceeded threshold effect levels for As, Cd, Cr, Cu, Ni and Zn and the predicted effect level was exceeded for As, Cr and Ni (Gomes *et al.*, 2019).

In addition to reduced treatment efficiency, there is risk of contaminant leaching from wetland soil when contaminants have accumulated to high concentrations (Wieder, 1993; Ronkanen and Kløve, 2009; Palmer *et al.*, 2015). Some contaminants, particularly mobile metals such as Ni or Zn, can be leached more readily than Cu from wetland soils (Nieminen *et al.*, 2002; Palmer *et al.*, 2015) and waters rich in anions can exchange with adsorbed elements. Furthermore, sudden changes in the wetland hydraulic regime, such as storm events, may bring about changes in element mobility (Khan *et al.*, 2019, 2020). Therefore, monitoring for maintenance and performance are critical for longevity in the wider application of passive treatment systems. For example, Wiseman and Edwards (2004) reported that the projected lifespan of 15–20 years for Fe removal and retention is achievable but will be determined by the space within the system. Accumulation of ochre within the system is evident in the RAPS treatment step (see Figure 6.1) and ultimately this will require the removal and controlled disposal of metal-contaminated substrate and vegetation.

7 Conclusions and Recommendations

The application of CWs has demonstrated variable success in removing trace elements associated with mine wastewaters. Effectiveness is determined by several variables, including the specific metal types, the quality and quantity of wastewater, removal mechanisms and the duration of wetland operation. Typically, removal of the contaminants Fe, Al and Zn is highly effective in both the short (1–3 years) and longer term (c.10 years). However, other metals, such as Mn, are more problematic and lower removal rates are frequently reported.

Although studies have demonstrated success in improving water quality in the 10-year time frame, data on wetland soil/sediment changes and potential bioavailability are much less readly available. In some cases, there is evidence of metal loading to soils being excessive and requiring intervention. Certainly, monitoring of wider wetland system parameters, not just water quality, is recommended to fully assess the effectiveness of the treatment approach.

For enhanced CW efficiency, the incorporation of other passive treatment approaches may be appropriate. Positive results on this approach in the coal industry may have application for the wider mine waste sector for parameters such as pH adjustment, sulfate removal and the removal of specific metals. Such an approach

can offer enhanced effectiveness and increased longevity through minimising loading to the wetland component.

Ultimately, the application of CWs for the treatment of mine wastewater will be site specific and warrants preliminary investigations to fully characterise water quality and identify appropriate removal mechanisms.

In sites where the application of CWs technology is anticipated or is operational, the following is recommended.

- Full characterisation of mine waste leachate is conducted to identify contaminants of concern and to identify the most suitable wetland approach and appropriate hybrid system, where applicable.
- The feasibility is assessed at the field scale over a minimum of 5+ years to determine treatment effectiveness and assess the fate of contaminants.
- In addition to water quality monitoring, the element content within the wetland system, particularly the determination of metal concentrations and availability in wetland soils/sediments, must be assessed.
- When loss of effectiveness and/or accumulation of elements is observed, the appropriate management and intervention strategies should be identified.

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Abbreviations

AMD	Acid mine drainage
CEC	Cation exchange capacity
CW	Constructed wetland
ELV	Emissions limit value
IPC	Integrated Pollution Control
MAC	Maximum allowable concentration
RAPS	Reducing and alkalinity-producing system
TMF	Tailings management facility
VFP	Vertical flow pond

AN GHNÍOMHAIREACHT UM CHAOMHNÚ COMHSHAOIL

Tá an Ghníomhaireacht um Chaomhnú Comhshaoil (GCC) freagrach as an gcomhshaol a chaomhnú agus a fheabhsú mar shócmhainn luachmhar do mhuintir na hÉireann. Táimid tiomanta do dhaoine agus don chomhshaol a chosaint ó éifeachtaí díobhálacha na radaíochta agus an truaillithe.

Is féidir obair na Gníomhaireachta a roinnt ina trí phríomhréimse:

Rialú: Déanaimid córais éifeachtacha rialaithe agus comhlíonta comhshaoil a chur i bhfeidhm chun torthaí maithe comhshaoil a sholáthar agus chun díriú orthu siúd nach gcloíonn leis na córais sin.

Eolas: Soláthraímid sonraí, faisnéis agus measúnú comhshaoil atá ar ardchaighdeán, spriocdhírithe agus tráthúil chun bonn eolais a chur faoin gcinnteoireacht ar gach leibhéal.

Tacaíocht: Bímid ag saothrú i gcomhar le grúpaí eile chun tacú le comhshaol atá glan, táirgiúil agus cosanta go maith, agus le hiompar a chuirfidh le comhshaol inbhuanaithe.

Ár bhFreagrachtaí

Ceadúnú

Déanaimid na gníomhaíochtaí seo a leanas a rialú ionas nach ndéanann siad dochar do shláinte an phobail ná don chomhshaol:

- saoráidí dramhaíola (m.sh. láithreáin líonta talún, loisceoirí, stáisiúin aistrithe dramhaíola);
- gníomhaíochtaí tionsclaíocha ar scála mór (m.sh. déantúsaíocht cógaisíochta, déantúsaíocht stroighne, stáisiúin chumhachta);
- an diantalmhaíocht (m.sh. muca, éanlaith);
- úsáid shrianta agus scaoileadh rialaithe Orgánach Géinmhodhnaithe (OGM);
- foinsí radaíochta ianúcháin (m.sh. trealamh x-gha agus radaiteiripe, foinsí tionsclaíocha);
- áiseanna móra stórála peitril;
- scardadh dramhuisce;
- gníomhaíochtaí dumpála ar farraige.

Forfheidhmiú Náisiúnta i leith Cúrsaí Comhshaoil

- Clár náisiúnta iniúchtaí agus cigireachtaí a dhéanamh gach bliain ar shaoráidí a bhfuil ceadúnas ón nGníomhaireacht acu.
- Maoirseacht a dhéanamh ar fhreagrachtaí cosanta comhshaoil na n-údarás áitiúil.
- Caighdeán an uisce óil, arna sholáthar ag soláthraithe uisce phoiblí, a mhaoirsiú.
- Obair le húdaráis áitiúla agus le gníomhaireachtaí eile chun dul i ngleic le coireanna comhshaoil trí chomhordú a dhéanamh ar líonra forfheidhmiúcháin náisiúnta, trí dhíriú ar chiontóirí, agus trí mhaoirsiú a dhéanamh ar leasúchán.
- Cur i bhfeidhm rialachán ar nós na Rialachán um Dhramhthrealamh Leictreach agus Leictreonach (DTLL), um Shrian ar Shubstaintí Guaiseacha agus na Rialachán um rialú ar shubstaintí a ídíonn an ciseal ózóin.
- An dlí a chur orthu siúd a bhriseann dlí an chomhshaoil agus a dhéanann dochar don chomhshaol.

Bainistíocht Uisce

- Monatóireacht agus tuairisciú a dhéanamh ar cháilíocht aibhneacha, lochanna, uiscí idirchriosacha agus cósta na hÉireann, agus screamhuiscí; leibhéil uisce agus sruthanna aibhneacha a thomhas.
- Comhordú náisiúnta agus maoirsiú a dhéanamh ar an gCreat-Treoir Uisce.
- Monatóireacht agus tuairisciú a dhéanamh ar Cháilíocht an Uisce Snámha.

Monatóireacht, Anailís agus Tuairisciú ar an gComhshaol

- Monatóireacht a dhéanamh ar cháilíocht an aeir agus Treoir an AE maidir le hAer Glan don Eoraip (CAFÉ) a chur chun feidhme.
- Tuairisciú neamhspleách le cabhrú le cinnteoireacht an rialtais náisiúnta agus na n-údarás áitiúil (m.sh. tuairisciú tréimhsiúil ar staid Chomhshaol na hÉireann agus Tuarascálacha ar Tháscairí).

Rialú Astaíochtaí na nGás Ceaptha Teasa in Éirinn

- Fardail agus réamh-mheastacháin na hÉireann maidir le gáis cheaptha teasa a ullmhú.
- An Treoir maidir le Trádáil Astaíochtaí a chur chun feidhme i gcomhair breis agus 100 de na táirgeoirí dé-ocsaíde carbóin is mó in Éirinn.

Taighde agus Forbairt Comhshaoil

• Taighde comhshaoil a chistiú chun brúnna a shainaithint, bonn eolais a chur faoi bheartais, agus réitigh a sholáthar i réimsí na haeráide, an uisce agus na hinbhuanaitheachta.

Measúnacht Straitéiseach Timpeallachta

 Measúnacht a dhéanamh ar thionchar pleananna agus clár beartaithe ar an gcomhshaol in Éirinn (*m.sh. mórphleananna forbartha*).

Cosaint Raideolaíoch

- Monatóireacht a dhéanamh ar leibhéil radaíochta, measúnacht a dhéanamh ar nochtadh mhuintir na hÉireann don radaíocht ianúcháin.
- Cabhrú le pleananna náisiúnta a fhorbairt le haghaidh éigeandálaí ag eascairt as taismí núicléacha.
- Monatóireacht a dhéanamh ar fhorbairtí thar lear a bhaineann le saoráidí núicléacha agus leis an tsábháilteacht raideolaíochta.
- Sainseirbhísí cosanta ar an radaíocht a sholáthar, nó maoirsiú a dhéanamh ar sholáthar na seirbhísí sin.

Treoir, Faisnéis Inrochtana agus Oideachas

- Comhairle agus treoir a chur ar fáil d'earnáil na tionsclaíochta agus don phobal maidir le hábhair a bhaineann le caomhnú an chomhshaoil agus leis an gcosaint raideolaíoch.
- Faisnéis thráthúil ar an gcomhshaol ar a bhfuil fáil éasca a chur ar fáil chun rannpháirtíocht an phobail a spreagadh sa chinnteoireacht i ndáil leis an gcomhshaol (*m.sh. Timpeall an Tí, léarscáileanna radóin*).
- Comhairle a chur ar fáil don Rialtas maidir le hábhair a bhaineann leis an tsábháilteacht raideolaíoch agus le cúrsaí práinnfhreagartha.
- Plean Náisiúnta Bainistíochta Dramhaíola Guaisí a fhorbairt chun dramhaíl ghuaiseach a chosc agus a bhainistiú.

Múscailt Feasachta agus Athrú Iompraíochta

- Feasacht chomhshaoil níos fearr a ghiniúint agus dul i bhfeidhm ar athrú iompraíochta dearfach trí thacú le gnóthais, le pobail agus le teaghlaigh a bheith níos éifeachtúla ar acmhainní.
- Tástáil le haghaidh radóin a chur chun cinn i dtithe agus in ionaid oibre, agus gníomhartha leasúcháin a spreagadh nuair is gá.

Bainistíocht agus struchtúr na Gníomhaireachta um Chaomhnú Comhshaoil

Tá an ghníomhaíocht á bainistiú ag Bord lánaimseartha, ar a bhfuil Ard-Stiúrthóir agus cúigear Stiúrthóirí. Déantar an obair ar fud cúig cinn d'Oifigí:

- An Oifig um Inmharthanacht Comhshaoil
- An Oifig Forfheidhmithe i leith cúrsaí Comhshaoil
- An Oifig um Fianaise is Measúnú
- Oifig um Chosaint Radaíochta agus Monatóireachta Comhshaoil
- An Oifig Cumarsáide agus Seirbhísí Corparáideacha

Tá Coiste Comhairleach ag an nGníomhaireacht le cabhrú léi. Tá dáréag comhaltaí air agus tagann siad le chéile go rialta le plé a dhéanamh ar ábhair imní agus le comhairle a chur ar an mBord.

EPA Research Report 400

Use of Constructed Wetlands for Treating Mine Waste Leachates: Assessment of Longevity and Management Implications



Authors: Ashlene Hudson, John Murnane and Ronan Courtney

Identifying Pressures

Mine waste storage facilities can generate significant quantities of wastewaters and leachates with extreme pH and elevated metal contents. If released untreated to the surrounding environment, these leachates will contaminate soils and surface and ground waters. Of particular concern is the generation of leachates from legacy sites, which are anticipated to exist in the timeline of multiple decades to centuries; this presents a major long-term environmental challenge. When planning for the closure of modern facilities and remediation of legacy sites, the use of constructed wetlands as a passive treatment system is viewed as an effective approach for the long-term treatment of mine waters, including for pH buffering and metal removal. However, the potential for wetlands to operate effectively in the long term is relatively unknown, with limited information on metal removal efficiencies over time and uncertain management implications for continued effectiveness.

Informing Policy

Modern mine waste (tailings) facilities operating under licence from the EPA have constructed wetlands as a component of their Integrated Pollution Control (IPC) licences; Industrial Emissions licences; Closure, Restoration, Aftercare Management Plan (CRAMP) documents; and closure plans. Furthermore, remediation of legacy sites require low-cost, passive strategies to treat mine waters emanating from waste facilities. Although many bench-scale and short-term trials have demonstrated great potential for constructed wetland technology to effectively treat mine waters, there is limited information on the long-term, field applications of such approaches. This study documents evidence of long-term applications of constructed wetlands within the mining industry and identifies key processes requiring monitoring and management for continued operation. Long-term effective treatment fulfils obligations to achieve "good quality" water status and informs sustainable development.

Developing Solutions

This study provides evidence of the effectiveness of constructed wetlands for treating mine waters from different settings over time frames of several years. It is shown that for constructed wetlands (1) metal removal processes are complex, variable and dependent on many properties within the wetland system; (2) wastewater characterisation should inform the design of an appropriate wetland system; (3) long-term treatment is possible; (4) monitoring of the wider wetland substrate and vegetation is required to assess long-term effectiveness; and (5) appropriate intervention and management strategies should be identified based on monitoring data.



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