

RISKS, REMEDIATION AND RECOVERY: LESSONS FOR BAUXITE RESIDUE MANAGEMENT FROM AJKA

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Abstract

This paper reviews the environmental risks highlighted in studies in the aftermath of the Ajka (Hungary) red mud spill, alongside opportunities for future management. The highly alkaline leachate was rich in many oxyanion-forming elements (e.g. Al, As, Cr, Mo, V). The oxyanionic metal(loids) were soluble at high pH, although neutralisation (by acids and gypsum) reduced their mobility downstream of the site. However, vanadium was poorly removed from solution during neutralisation to circum-neutral pH (< 16 % removed), and as such should be a key focus of for leachate management at BRDAs. Studies suggest minimal longer term legacy of the spill, which is testament to the remedial efforts and the fine-grained nature of the residue which is readily entrained and transported downstream.

Introduction

The release of ~ 1 million tonnes of saline caustic red mud suspension from the Ajkai Timfoldgyar Zrt alumina production plant in Ajka, western Hungary in October 2010 was the largest documented release of alumina industry by-products into the environment. Although the circumstances surrounding the failure of the retaining wall of Cell X¹ at the depository are not representative of modern bauxite residue disposal area (BRDA) management practices elsewhere, the numerous scientific studies in the aftermath of the spill have revealed much about the risks and environmental behaviour of red mud, alongside the effectiveness of remedial efforts. The Hungarian government invested 38 billion Hungarian Forint (~ € 127 million) in emergency measures in the three years after the spill.² This included demolition and reconstruction of the BRDA and emergency management of spill material (acid dosing at source, gypsum dosing of rivers and building of check dams) to promote buffering of waters and sedimentation.³ Longer term measures included channel dredging, the recovery of red mud from affected floodplain areas and the ploughing of red mud into topsoil in areas of shallow (< 50 mm) deposits to minimise fugitive dust generation.⁴ In the years since the disaster, a range of studies have assessed the

impacts of the spill on a suite of environmental receptors. This paper reviews the key impacts of the Ajka spill, focussing on the surrounding water environment, and considers both: (a) the environmental behaviour of red mud, and (b) the effectiveness of remedial efforts, to highlight key lessons for future bauxite residue management.

Characterisation and Environmental Behaviour of Red Mud Suspension

There are relatively few published accounts of the behaviour of red mud leachate in the environment.⁵⁻⁷ In the immediate aftermath of the spill, analysis of residual leachate release from the Ajka BRDA was characterised by extreme pH (13.1) and alkalinity (up to 7000 mg/L as CaCO₃), and enrichment of a range of potential elements of concern (Table 1).⁸ These include many oxyanion-forming elements which are soluble at high pH, such as Al, As, Cr, Mo and V. Table 1 highlights the distribution of some of these elements of concern between particulate, colloidal and dissolved phases (from sequential filtration of samples). This is revealing in showing that for the majority of elements, the bulk of the concentration was partitioned in particulate and colloidal phases in the leachate. Only for Mo and V were significant proportions partitioned in truly-dissolved phases, which would be anticipated to be more bioavailable in the environment.⁸ Furthermore, speciation analyses of V in Ajka leachate showed it to be present in its pentavalent form (as vanadate).⁹ While the concentrations of some trace elements exceed aquatic life standards in waters (e.g. V, As) and fluvial sediments (As, Cr, Ni, V), the spatial extent of these was found to be limited to the Torna Creek and part of the upper Marcal.⁸ Solid phase analyses showed many contaminants (notably As, Co, Ni and Cr) to be associated with residual hard-to-leach fractions of the red mud-affected sediments, that are unlikely to be remobilised at the circum-neutral pH of the Torna-Marcal system.⁸ This is likely to significantly limit bioavailability. In addition, speciation studies on Cr and As suggest that they are in their least toxic forms (trivalent Cr substituted into hematite and pentavalent As), although V was found in its most toxic pentavalent form in solid phases as well as leachate.⁹

There has been a burgeoning interest in recent years in the prospects for value recovery from bauxite residue.¹⁰⁻¹¹ Of particular focus has been the presence of rare earth elements and other e-tech elements crucial to future green technologies such as photovoltaics, LEDs and portable fuel cells. Like many other red muds, these e-tech elements were also ubiquitous in the Ajka deposits at modest concentrations (Table 2) and provide a good tracer of the signal of red mud throughout fluvial sediments in the affected rivers.¹² Analyses of leachate samples however shows that

a very small proportion of the e-tech element inventory in the red mud is soluble at source (gallium is a notable exception), which means prospects for the ready-recovery of these elements is limited (e.g. through passive recovery as part of legacy BRDA leachate management). Recovery approaches will therefore likely require significant energy input (sintering) or chemical transformation (e.g. using concentrated lixiviants).

Table 1: Concentration of selected oxyanion-forming elements in Ajka leachate during sequential filtration (all values in mg/l).⁸

Element	Total	< 0.45 µm	< 10 kDa
Al	678.70	539.60	24.70
As	3.40	2.90	0.50
Cr	0.27	0.06	0.03
Mo	5.60	5.20	4.30
V	5.60	5.30	3.30

Table 2: Concentration of selected e-tech elements in Ajka red mud (solid phase, air-dried) and leachates (at pH 13.1).¹²

Element	Use	Solid (ppm)	Aqueous (total, ppm)	Aqueous (< 0.45 µm, ppm)
Ce	Catalysts, low energy bulbs	405	0.42	<0.01
Co	Magnets, high strength alloys	97	0.21	<0.01
Ga	LEDs, photovoltaics	79	2.44	2.30
Ge	Semiconductors	27	0.02	<0.01
La	Hybrid fuel cell component	150	0.75	<0.01
Nb	High strength alloys and semiconductors	85	0.26	<0.01

Management

The emergency management efforts at Ajka comprised red mud removal from land and water, extensive dosing with acids (HCl and H₂SO₄) and gypsum dosing of affected streams.^{2,4} Downstream trends in water quality during the emergency efforts highlight the effectiveness of this management in limiting the downstream impacts of the residual releases from the site (Figure 1).⁸ Water pH was rapidly buffered to within water quality guidelines (pH 6-9) within 2 km of the BRDA through acid dosing. As would be anticipated, this rapid neutralisation of pH is accompanied by associated attenuation of some key elements of concern, such as Cr and Al (Figure

1). For V however, which retains mobility as vanadate throughout the ambient circum-neutral pH range of the affected catchment (underlain by Triassic dolomites and limestones).¹³ The fall in V concentration downstream is driven largely by dilution as opposed to pH adjustment. Indeed loadings measurements taken at the site suggested no major change in V load in the 40 km downstream of the spill zone.⁸ These field data are supported by laboratory experimentation which assessed metal removal during neutralisation by different methods for Ajka samples.¹⁴ These laboratory data highlight that V, along with Mo, is not efficiently removed during neutralisation, unlike other key contaminants such as Al and As (Table 3).

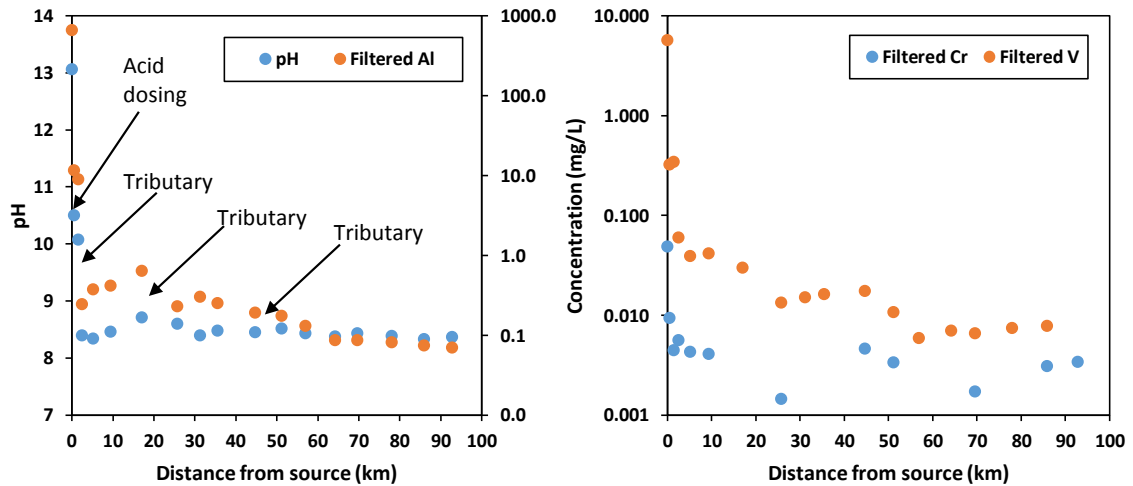
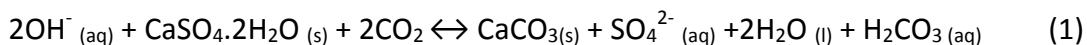


Figure 1: Downstream trends in pH and selected metals during emergency management of residual releases from the Ajka BRDA (November, 2010).⁸

Table 3: Concentration of selected oxyanion-forming elements in Ajka leachate before and after neutralisation with gypsum and HCl (0.2 µm filtered¹⁴). Values in parenthesis show percentage removed

Determinand	Red mud leachate	6 M HCl (~5 ml/l)	Gypsum (30 g/l)
pH	13.1	8.3	9.8
Selected elements (mg/l)			
Al	352.0	1.2 (99.7)	4.9 (86)
As	8.1	6.2 (24)	1.6 (81)
Mo	11.6	10.5 (9)	11.0 (4)
V	15.6	14.8 (5)	13.1 (16)

Around 10,000 tonnes of gypsum was used to dose waters downstream of the site for pH neutralisation.² Analyses of secondary deposits revealed that there are potential benefits not just for neutralisation of waters (Equation 1), but also for carbon sequestration (and a potential industrial offset) and metal immobilisation.



Secondary deposits at the gypsum-dosed sites were dominated by carbonate minerals.¹⁵ Within these there was a modest uptake of contaminants, notably As, Cr and Mn.¹⁵ This is consistent with laboratory simulations which also show a significant scavenging of As in secondary deposits (possibly as Ca-arsenate phases).¹⁴ C and O stable isotope ratios of carbonate precipitates formed as a result of gypsum dosing were used to quantify the importance of the neutralisation process in sequestering atmospheric carbon dioxide. This process was particularly pronounced at sites most affected by gypsum addition, where up to 36 % of carbonate-C appears to be derived from atmospheric in-gassing of CO₂.¹⁵ While cautioning for the need for full cost-benefit analysis, it was suggested that at such rates of sequestration, widespread gypsum dosing could offer a 3-4 % of the direct CO₂ emissions offset for the alumina industry through secondary carbonate precipitation.¹⁵ Gypsum dosing as a wider management practice in BRDAs¹⁶⁻¹⁷ therefore holds scope not just for well-documented improvements in soil nutrient content and reduced sodicity, but also in providing a modest emissions offset for the industry and potentially in limiting metal(loid) availability. Future studies on the importance of gypsum on these processes in rehabilitated red mud would therefore be desirable.

Biological Impacts and Recovery of the Marcal-Torna System

Table 4 summarises the key findings of studies assessing the biological response of various organisms to red mud exposure. Short-term impacts on aquatic organisms were highlighted in numerous studies showing negative impacts on zooplankton and ostracods,¹⁸⁻¹⁹ while documented impacts on terrestrial plants have ranged from stunted growth, salt-stress and suspected genotoxic effects in high red mud dosage rates.¹⁹⁻²¹ A theme amongst the various studies on the biological impacts of the spill was that there are multiple potential stressors in the terrestrial and aquatic habitats affected by red mud. These include the metal concentrations, fine particle size distribution, high pH, and crucially the salinity, which is regarded as the key short-term stressor in various studies.^{20, 22} Human health studies have highlighted no short-term impacts on local populations and emergency workers exposed to red mud (notably via fugitive dusts), although longer term studies are ongoing (Table 4).²³⁻²⁴

Table 4: A summary of key biological studies in the aftermath of the Ajka spill

Pathway / Receptor studied	Approach and key findings
Contaminated floodplain soils / plants ²⁰	<ul style="list-style-type: none"> - Red mud characterisation and toxicity for plants in small-scale red mud/ non-contaminated soil mixtures. - Salinity, not trace metal concentration were the main concern for red mud in soil.
Waters / planktonic rotifers ¹⁸	<ul style="list-style-type: none"> - Planktonic rotifer assemblages of the Danube were investigated at Budapest during the spill. - Median species richness decreased to zero, Shannon-Weaver diversity to 0 after the arrival of the contamination wave. - Rotifer assemblages recovered after 3 weeks, but initial levels of diversity and abundance were not reached.
Fluvial sediments / bacteria, ostracods, aquatic plants, terrestrial plants ¹⁹	<ul style="list-style-type: none"> - Series of bioassays on fluvial sediments collected from across Torna-Marcal system. - Negative response of some bioassays in presence of red mud or gypsum-dosed sediments. - Difficult to discern individual stressor given concurrent salinity, metal availability and small particle size.
Dusts / human health ²³	<ul style="list-style-type: none"> - Study of short-term genotoxic effects on inhabitants either with burns or on those inhaling desiccated dust. - Red mud exposure did not pose an immediate genotoxic hazard on residents. Long-term hazards of ultra-fine fugitive dust particles with alkalinity of residual NaOH in red mud are still unknown.
Soil / Collembola ²⁵	<ul style="list-style-type: none"> - Changes in community structure of <i>Collembola</i> were studied in soils following the disaster. - No adverse effect observed in <i>Collembola</i> abundance. - Change in pH with soil re-acidification highlighted as potential long term risk due to metal re-mobilisation.
All possible risk posing pathways / human health ²⁴	<ul style="list-style-type: none"> - Summary of results from different groups on toxic metal concentration of urine, extensive screening of locals, chromosoma aberration tests on exposed people, effects on the immune system, lung and digestive system, carcinogenic and toxic effects on fish. - Except for the health hazard posed by the caustic nature of red mud, no other major risk was identified.
Dusts / rats ²⁶	<ul style="list-style-type: none"> - Exposure to airborne red mud dust at high concentration (2 weeks, 2 h/day) with control (room air). Airway resistance and respiratory tissues mechanics measured, histopathology performed. - Mild respiratory symptoms developed following short-term exposure of healthy individuals. These pose no greater respiratory hazard than urban dust at comparable concentrations.

Long term monitoring of the affected river systems has revealed a fairly rapid physico-chemical recovery. Sediment surveys in the immediate aftermath of the spill highlighted the characteristic signal of red mud (fine particle size, enriched in Al, As, Cr, Ni, V in particular) throughout the Torna Creek and in some hotspots of

deposition in the upper Marcal River where the channel planform lent itself to deposition (low gradient, wide channel with abundant macrophytes⁸). Follow up surveys in 2013 showed that the signal of red mud was only apparent at a small number of locations in small channels in the immediate vicinity of the site (Figure 2).² V provides a good indicator of this positive trend and it was observed that a reduction of channel length exceeding prescribed sediment quality standards for V declined from over 18 km in 2010 to less than 0.5 km in 2013 (Figure 2). This was considered to be a function of both the extensive dredging efforts (in the region of 80km of channel) and the fine particle size of the red mud (D_{50} of $3.4 \mu\text{m}$) which lends itself to entrainment and downstream transport out of the system.²

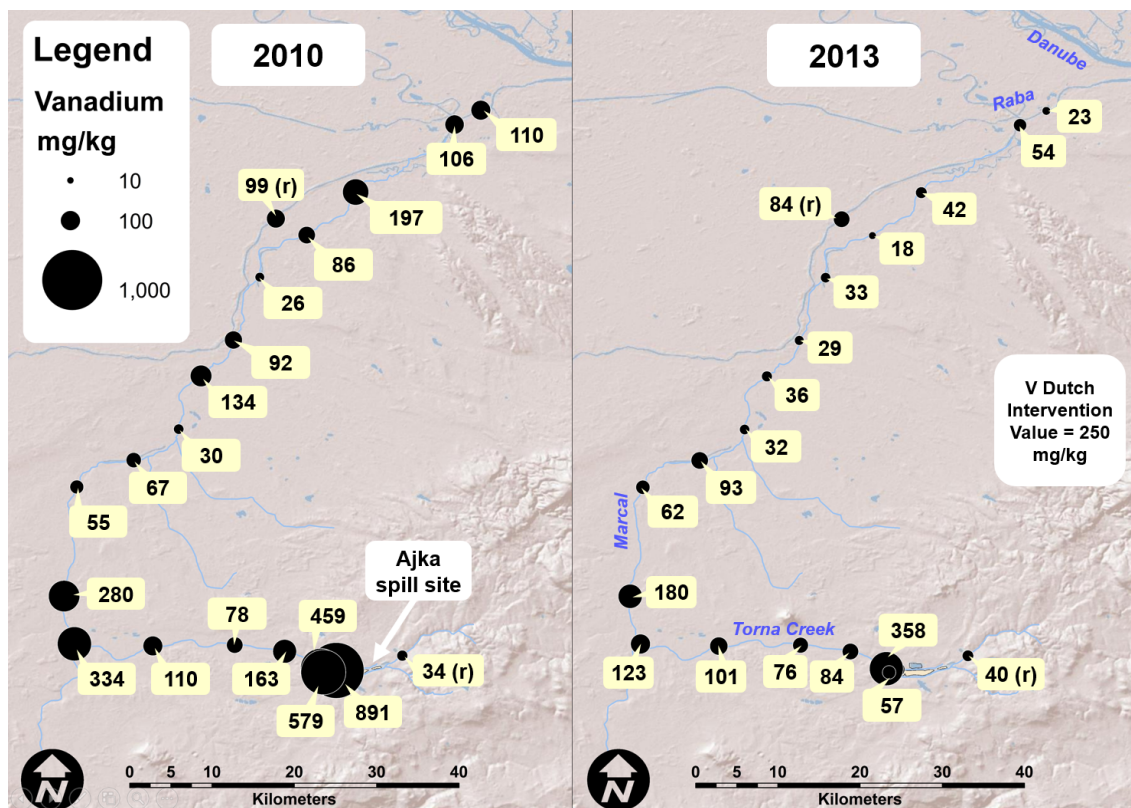


Figure 2: Comparison of average vanadium concentrations ($n = 3$) in the Torna-Marcal system between 2010 and 2013 surveys. Values with (r) show reference sites on unaffected tributaries.^{2, 8}

Water quality monitoring highlighted the initial success of the dosing operations in reducing pH around source areas. Longer term monitoring suggested that the concentrations of dissolved trace metal(loid)s downstream of the site did not exceed the European water quality standards and the US Environmental Protection Agency aquatic life criteria values (excluding one sample for Cd).²⁷ This is also consistent with

official monitoring of biological indices of river system health (e.g. macroinvertebrate samples).³ However, continued elevations of As and Ni were apparent in the Marcal River 2 years after the spill compared to that observed in the pre-disaster period.²⁷ This elevation of As and Ni is consistent with the enhanced leaching of these from red mud when in contact with organics (e.g. where red mud was ploughed into agricultural soils²⁸⁻²⁹), but further monitoring of the river systems would be desirable over the full range of hydrological conditions apparent in the affected streams to assess any such long term trends. Furthermore, the absence of vanadium water quality data in the official monitoring efforts is something of an omission, given its presence in both red mud and leachates highlighted above.

Conclusions and Opportunities for Future Management

Given the size of Ajka spill and the immediate international concern, the scientific studies in the aftermath have offered a largely positive prognosis on the long term impacts of the spill on terrestrial and aquatic environments. The highly caustic, saline and metal(loid)-rich, the alkalinity was effectively managed around source areas where the majority of the metal(loid) inventory in the red mud leachate is associated with particulate and colloidal phases. The numerous biological studies in affected areas suggest short term impacts were dominated by salinity and the caustic nature of the spill material. Metal(loid) availability appears to be a secondary issue, but an issue that numerous workers have cautioned for the need for longer-term monitoring, mainly due to the presence of some mobile oxyanions (e.g. Mo and V) in potentially toxic forms (in the case of vanadate).

Behaviour of red mud leachate at Ajka offers some insight for approaches to long term leachate management at BRDAs. Neutralisation (either through gypsum addition or acid dosing) is effective at limiting the availability of many of the potentially hazardous metal(loids) present in red mud leachate. However, some are less efficiently removed during neutralisation, notably vanadium and molybdenum, and as such should be a particular focus of studies looking at passive leachate treatment (e.g. bioremediation using wetlands). The fact that vanadium was omitted from official regulatory monitoring in Hungary was a key oversight in some of the published studies after the spill and should be incorporated into all monitoring suites at BRDAs. Gypsum dosing may offer advantages beyond neutralisation through limiting metal availability and via sequestration of atmospheric carbon. Ajka provided a large scale analogue for the potential use of gypsum for emissions offsetting in active BRDAs and the field data suggests that the magnitude of the offset could be significant for the industry. The longer term studies of the affected systems suggest that the lasting legacy of the Ajka red mud spill appears to be relatively minor

compared to the numerous well-documented tailings spills from base metal mining facilities. Both water quality improvements and sediment quality improvements have been reported which are testament to the extensive remedial operations by the authorities and the physical nature of the red mud (fine grain size) which lends itself to downstream transport and dilution.

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