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THEMATIC SECTION: BAUXITE RESIDUE VALORIZATION

Advances in Understanding Environmental Risks of Red Mud After the Ajka Spill, Hungary

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Abstract In the 5 years since the 2010 Ajka red mud spill (Hungary), there have been 46 scientific studies assessing the key risks and impacts associated with the largest single release of bauxite-processing residue (red mud) to the environment. These studies have provided insight into the main environmental concerns, as well as the effectiveness of remedial efforts that can inform future management of red mud elsewhere. The key immediate risks after the spill were associated with the highly caustic nature of the red mud slurry and fine particle size, which once desiccated, could generate fugitive dust. Studies on affected populations showed no major hazards identified beyond caustic exposure, while red mud dust risks were considered equal to or lesser than those provided by urban dusts of similar particle size distribution. The longer-term environmental risks were related to the saline nature of the spill material (salinization of inundated soils) and the release and the potential cycling of oxyanion-forming metals and metalloids (e.g., Al, As, Cr, Mo, and V) in the soil–water environment. Of these, those that are soluble at high pH, inefficiently removed from solution during dilution and likely to be exchangeable at ambient pH are of chief concern (e.g., Mo and V). Various ecotoxicological studies have identified negative impacts of red mud-amended soils and sediments at high volumes (typically >5 %) on different test organisms, with some evidence of molecular-level impacts at high dose (e.g., genotoxic effects on plants and mice). These data provide a valuable database to inform future toxicological studies for red mud. However, extensive management efforts in the aftermath of the spill greatly limited these exposure risks through leachate neutralization and red mud recovery from the affected land. Monitoring of affected soils, stream sediments, waters and aquatic biota (fungi, invertebrates and fish) have all shown a very rapid recovery toward prespill conditions. The accident also prompted research that has also highlighted potential benefits of red mud use for critical raw material recovery (e.g., Ga, Co, V, rare earths, inform), carbon sequestration, biofuel crop production, and use as a soil ameliorant.

Keywords Bauxite processing residue · Bauxite residue · Environmental risk · Resource recovery · Red mud

Introduction

The Ajka red mud spill in western Hungary in October 2010 was the largest documented release of alumina industry by-products into the environment, with around 1 million m^3 of red mud suspension breaching the failed depository wall [1]. Although the circumstances surrounding the failure of the retaining wall of Cell X [1] 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 behavior of bauxite processing residue

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(hereafter referred to as red mud), alongside the effectiveness of remedial efforts. Furthermore, the accident focused much public attention on the alumina industry and has catalyzed efforts among leading alumina producers for collaboration and mitigation of environmental legacies associated with red mud [2, 3]. At present, around 150 million tonnes of red mud are produced per year, of which around 2–3 % is productively reused or recycled [2]. Indeed, a range of collaborative initiatives between industry and research centers are underway across the sector to increase valorization of red mud [4, 5].

In the aftermath of the Ajka spill, the Hungarian government invested 38 billion Hungarian Forint (~€127 million) in emergency and rehabilitation measures between 2010 and 2013 [6]. Emergency works on the BRDA saw the breached corner of the dam rebuilt, with the surface of the bauxite residue covered with a depth of 40 cm of humus-containing soil that had been collected from the inundated areas around Kolontár and Devecser. Emergency management of the spill material included acid dosing at source, gypsum dosing of rivers, and building of check dams to promote buffering of waters and sedimentation [7]. Longer-term measures included channel dredging, the recovery of red mud from affected floodplain areas, and the plowing of red mud into topsoil in areas of shallow (<50 mm) deposits to minimize fugitive dust generation [7, 8]. The latter was based on a three-tiered risk assessment [8] for assessing potential ecological and public health impacts, along with risks of secondary salinization of soils [9]. In the years since the disaster, a range of studies have assessed the immediate and longer-term impacts of the spill on a suite of environmental receptors, as well as considered opportunities for the valorization of both Ajka red mud and the recovered spill material (a mixture of red mud and topsoil). This paper reviews the main environmental impacts of the Ajka spill and considers both: (a) the environmental behavior of red mud, and (b) the effectiveness of remedial efforts, to highlight key lessons and opportunities for future bauxite residue management.

Methods

A comprehensive search on the Web of Science and Google Scholar databases was conducted, using as keywords "Ajka" and "red mud", "spill", "red sludge," "bauxite residue," "Hungary". The search was restricted to dates from 2010 onward.

Range of Studies and Approaches Adopted

To date, there have been 46 published scientific papers and conference papers directly relating to the Ajka spill or experimenting with red mud from the site. The majority of these focus on environmental impacts of the spill and cover the range of potential pathways and receptors for red mud (Fig. 1). Many of the longer-term studies have aimed to model potential impacts on the soil–water environment, given that any longer-term impacts of the spill are most likely to affect these pathways, rather than the more short-lived air and water risks (Fig. 1). Of the environmental studies, in addition to a broad coverage in environmental pathways and potential receptors across a range of trophic levels, there have also been some novel approaches to environmental risk assessment adopted. These include remote sensing studies assessing the extent and thickness of the spill material [10], high-frequency mobile monitoring

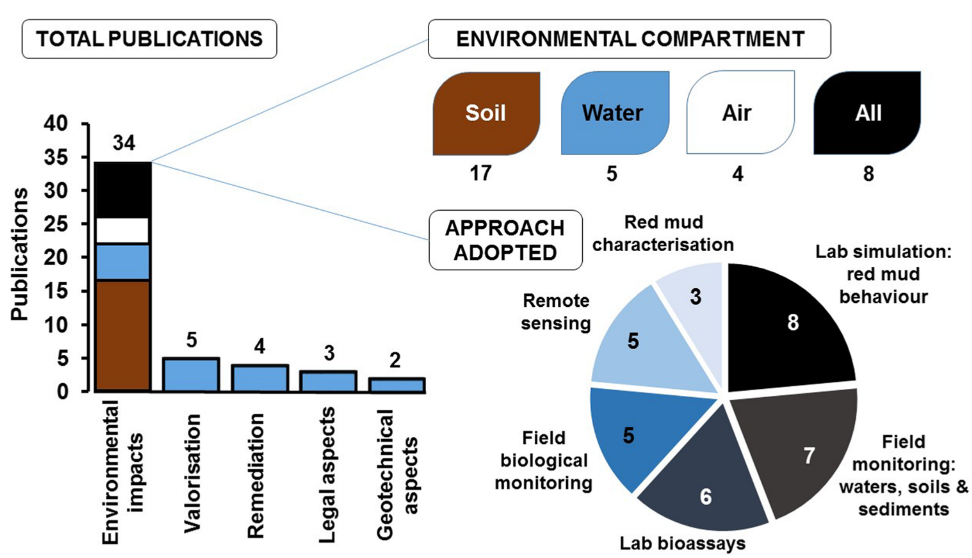

Fig. 1 Summary of categories of scientific papers and conference proceedings after the Ajka spill. Broad area of focus shown in the *lower left*, with separation by environmental compartment studied in the *upper right* (sediments are included with soil). The *lower right image* highlights the principal approach to data collection (i.e., some studies may have combined field monitoring with laboratory experimentation, but the key focus of the study is taken here)

of the affected streams and air [7], and speciation studies on important contaminants [11, 12] for which few prior published accounts for red mud were available.

Environmental Pathways After the Spill

Air: Fugitive Dusts

Key studies in the immediate aftermath of the spill focussed on the risk of fugitive dusts. Although the spill occurred in October, before a period of extended winter snow cover, the risk of dust generation, particularly during clean-up operations was a concern for the Hungarian authorities. Physical characterization of the spill material showed a particle size distribution that fell predominantly within the PM_{10} class centered around 3–8 μm. Only a small portion of the red mud was distributed within finer aerosol classes ($PM_{2.5}$ and PM_1: with diameters less than 2.5 and 1 μm, respectively [13]). This gave the spill material a similar distribution to urban dusts, with researchers concluding that no risks above that typically encountered with urban dusts would be anticipated [13]. Continuous (15 min interval) monitoring of PM_{10} fraction aerosols at two locations within the spill zone for a year after the spill did not show any breaches in air quality limits other than those that could be ascribed to dusts from domestic biomass burning in winter months [7].

Water: Red Mud Leachates

Information on the quality of leachates from red mud is relatively scarce in the published literature but is imperative for assessing risks posed by environmental red mud releases and for long-term BRDA management [14]. In the immediate aftermath of the spill, analysis of residual leachate release from the Ajka BRDA was characterized by extreme pH (13.1) and alkalinity (up to 6600 mg L^{-1} as Na_2CO_3), and enrichment of a range of potential elements of concern (Table 1, [15]). 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, P, and V were significant proportions partitioned in truly dissolved phases, which would be anticipated to be more bioavailable in the environment [15]. Furthermore, speciation analyses of V in Ajka leachate showed it to be present in its most toxic, pentavalent form (as vanadate [11]). Residual releases of leachate continued for a month after the spill, although acid-dosing stations were in place to minimize downstream impacts [7]. While the concentrations of some trace elements exceed aquatic life standards in waters (e.g., V, As, etc.) and fluvial sediments (As, Cr, Ni, V, etc.), the spatial extents of these were found to be limited to the Torna Stream and part of the upper Marcal due to a combination of dilution and remedial measures [15]. Phosphorus concentrations have not been widely reported for red mud leachate, but similar to other oxyanion-forming elements, P is present in elevated concentrations (Table 1). While it is predominantly in particulate form, the dissolved and colloidal fractions are at environmentally significant levels and may pose a long-term issue for increasing nutrient status of receiving waters and soils (Table 1).

Table 1 Concentrations of selected oxyanion-forming elements in Ajka leachate during sequential filtration (all values in mg L^{-1})

Element	Total	Colloidal and dissolved (<0.45 μm filter)	Truly dissolved (<10 kDa ultrafiltered)
Al	678.7	539.6	24.7
As	3.4	2.9	0.5
Cr	0.27	0.06	0.03
Mo	5.6	5.2	4.3
P	13.8	0.6	0.2
V	5.6	5.3	3.3

After [15]

Soils and Sediments

The low-lying agricultural lands in the region of 800–1020 ha on the floodplains of the Torna Stream and Marcal River were impacted by the spill, alongside up to 120 km of river channel where the red mud was also deposited [9, 10]. Solid-phase analyses of the red mud showed it to be having a high Na content (5.8 % as Na_2O) [16]. The pH of red mud diminishes over time with carbonation, and this, along with buffering capacities of affected soils, has a major bearing on controlling pH and subsequent trace element mobility [17, 18]. The red mud is enriched in many potential contaminants (notably Al, As, Co, Ni, and Cr [16]) which were associated with residual hard-to-leach fractions of the red mud-affected sediments [15, 19]. Therefore, these contaminants are unlikely to be remobilized at the circumneutral pH values of soil pore waters and those of the Torna-Marcal system, which limits bioavailability significantly. Also, speciation studies on Cr and As suggest that they are in their lower toxicity forms (trivalent Cr substituted into hematite and pentavalent As), unlike the V which is present in solids in pentavalent form similar to leachates [11].

Numerous researchers have assessed the impact of red mud mixing with agricultural soils (Fig. 1). Key issues highlighted during laboratory modeling studies of soil columns with surface amendments of red mud (10 cm thick) were the downward migrations of Na and Mo to depths of 80 cm [20]. The authors cautioned that secondary salinization of the soil was an important risk if thick red mud layers were not removed from the soil [20]. Another key aspect of the affected soil systems is the mixing of red mud with organic matter. The addition of red mud was found to dramatically increase the soil pH and concentrations of dissolved organic carbon (DOC) in soil pore waters [20, 21]. DOC is an important vector for many trace elements in the soil environment, and studies in the aftermath of Ajka spill have suggested enhanced leaching of As, Cu, and Ni from red mud when it is in contact with organic-rich media [20, 22, 23]. A further issue in both streams and the affected soil environments is the potential for the long-term cycling of metals and metalloids in the soil–water environment due to anion-exchange reactions. Enhanced release of As from soil-red mud mixtures under both aerobic and anaerobic conditions were reported with phosphate addition [18]. Given the ubiquity of phosphate in soils and surface waters in agricultural settings, long-term monitoring of areas where the residual red mud was plowed into the soil was recommended.

One final risk was identified: the issues associated with enrichment of red mud with radionuclides. The Ajka red mud was already well studied in advance of the spill with regard to the presence of isotopes of uranium, radium, and thorium, which could limit possible reuse of red mud in building materials [24]. Post-spill studies assessed the presence of radionuclides in the red mud and the effective dose of red mud collected from floodplain areas, as well as in situ monitoring of radon concentrations. Analysis of the spill material showed activity concentrations for ^{238}U, ^{236}Ra, ^{232}Th, and ^{40}K below the prescribed clearance levels [25].

Receptors: Assessing the Risks Associated with the Spill

Humans

The immediate tragic effects of the spill caused ten fatalities and injured at least 200 people [16, 26]. Comprehensive reviews by Hungarian authorities reviewing work from different groups on the exposed populations found that, except for the health hazard provided by the caustic nature of the spill material through dermal contact, no other major risks were identified (e.g., evidence of effects on immune systems, or toxicity) [27]. The key short-term pathway after the spill for human exposure was via fugitive dusts. Laboratory bioassays on rats exposed to short-term (2 h/day over 2 weeks) desiccated red mud dusts to assess such hazards [28] supported the prognosis from physico-chemical characterization of dusts by other workers [13]. Mild respiratory symptoms developed following exposure of healthy individuals, but these were deemed to pose no greater respiratory hazard than urban dust at comparable concentrations [28]. Encouragingly, direct monitoring of exposed potentially exposed populations (i.e., clean-up workers) showed no short-term cytogenetic or genotoxic effects of exposure to the spill material [26]. However, it is notable that exposure levels differed greatly in the exposed population group (<10 to >300 h), and these authors emphasized the need for longer-term assessments should there be latency in response to those exposed to the red mud, which are currently ongoing [26].

The other key potential pathway for human exposure identified in the aftermath of the spill was potential contamination of groundwater supply wells [29]. However, extensive monitoring (120 samples) of groundwater supply wells in the Ajka area showed no evidence of the red mud plume affecting controlled groundwaters [29], given the relatively confined area of the spill away from important aquifers. This was consistent with analyses of leachate from red mud-amended soil columns in laboratory studies showing pore waters with concentrations of most contaminants below drinking water-quality thresholds [20].

The assessments of technologically enhanced naturally occurring radioactive materials (TENORM) in the red mud highlighted a potentially higher external or internal (if inhaled) dose as a result of gamma emitting radionuclides [25]. However, the authors cautioned that this modest increase in exposure risk is negligible when compared to average annual background radiation [25].

Aquatic Biota

The immediate acute impacts of the spill on the receiving watercourses eliminated all aquatic life in a 71 km stretch of the Torna-Marcal system according to some accounts [30]. In electrofishing surveys, 2 weeks after the spill, in affected reaches of the Marcal river (25–100 km downstream), only four species of fish and 59 specimens were surveyed (compared to 22 species and present in prespill 2008 surveys, at a similar time of year) highlighting the impact of the caustic pulse on fish populations [31], which would likely have been sensitive to the sudden change in ionic strength and pH [32]. Further downstream still, some revealing assessments came during the initial movement of the red mud flood wave through the Danube. Monitoring of planktonic rotifers (microscopic freshwater zooplankton) at the Danube in Budapest (258 km downstream of the spill site), in the immediate aftermath of the spill, showed a decline in median species richness and species diversity (Shannon–Weaver) to zero after the arrival of the contamination wave [33]. The pH of the contamination wave was well buffered by this distance downstream, which would have limited metal(loid) availability, with only elevated conductivity (dominated by NaOH) being the signature of the spill. While rotifer assemblages recovered after 3 weeks, initial levels of diversity and abundance were not reached during this monitoring interval [33].

Studies on the impacts of the spill on fluvial hyphomycetes (fungi), which play a major role in leaf decomposition and subsequent in-stream nutrient cycling, highlighted differences before and after the spill in the Torna Stream [30, 34]. Reduced rates of leaf litter decomposition were apparent after the spill, and that was ascribed to diminished fungal colonization relative to reference sites [34]. It was suggested that gypsum smothering of substrates was a more important factor in diminishing fungal growth and species richness than the red mud itself [30]. The larger particle size of the gypsum lent itself to deposition unlike the finer red mud, which remained suspended in the water column [30].

Laboratory assays have also been used to assess the toxicity of red mud in fluvial settings [35]. These have included various trophic levels from aerobic bacteria, Common Duckweed (*Lemna minor*) through to sediment-dwelling ostracods (*Heterocypris incongruens*). There were strong positive correlations between many red mud-related contaminants and *Lemna minor* growth inhibition in the affected fluvial sediments [35]. However, a theme among studies on the biological impacts of the spill was that there were multiple potential stressors in the habitats affected by red mud, and as such, determining causal factors for the negative responses is difficult. In some cases, there was no obvious impact of red mud, with tests on ostracods showing no clear relationship between red mud content of the sediment and assay response [35]. Aerobic bacteria colony assays showed a tenfold to 100-fold increase in bacterial colony number with modest red mud amendment of sediments compared to unaffected reference sites [21, 35]. This may have been a feature of underlying contamination in the Torna-Marcal catchment from other industrial sources [36] and the possibly beneficial effect of red mud in limiting availability of preexisting contaminants (e.g., metals) [35].

Terrestrial Biota

Some of the first published results after the spill assessed the impacts of red mud on plant growth in laboratory experiments. Spring Barley (*Hordeum vulgare*) was used as a test species [16]. At applications of red mud to soil above 5 %, shoot yield was significantly diminished. Salinity, not trace metal concentration, was concluded to be the main immediate concern for red mud in soil, given trace element uptake in test species were not at levels deemed to be phytotoxic [16], and similar growth inhibition was apparent with NaOH-amended reference treatments [16]. Similarly, negative response in the yield (shoot and root growth) of White Mustard (*Sinapis alba*) in sediments with significant red mud content (typically >5 % volume) were reported in other studies [35]. Other plant-red mud response studies have identified molecular-level impacts on test species. Studies on *Tradescantia* (Trad-MN assay) and with root tip cells of *Allium cepa* (A-MN assay) assays showed symptoms of chromosomal damage and diminished root growth when exposed to high concentrations of red mud [37]. Follow-up studies replicated the impacts with vanadate solutions (albeit at higher concentrations than those reported in most of the affected areas) suggesting that pentavalent vanadium could be the causal agent for these observed genotoxic and cytotoxic effects [37].

Assessments of terrestrial fauna affected by the spill were limited to field monitoring and associated laboratory assays on communities of springtails (Collembola: a diverse group of omnivorous hexapods). Changes in community structure of Collembola were studied in affected soils following the disaster, although no adverse effect was observed in overall Collembola abundance [38]. However, laboratory assays using *Folsomia candida* (a soil-dwelling springtail: Collembola) did show significantly enhanced mortality above reference treatments in surface (0–30 cm) layers and subsurface (30–50 cm) soil layers covered in red mud [21].

Summary of Identified Risks

This wide range of potential pathways and receptors highlighted by the various studies demonstrate the requirement for the integrated risk assessment approach adopted by authorities [8]. These potential risks are summarized in Table 2 by the different facets of red mud physicochemical composition. The acute impacts identified in the water environment were relatively short-lived, once the red mud pulse had moved through the system and pH was effectively managed. In many of the laboratory bioassays, while there were some negative effects apparent, these were at high rates of red mud dosage. Given the extensive remedial efforts (summarized below) such dose rates/concentrations represent either conditions that did not persist for longer than several months after the spill, or spatially covered very small areas longer term. However, a very useful array of information is provided from the bioassays on the potential ecotoxicity of red mud in various environmental compartments and justifies the management responses detailed below. Given the relative dearth of

Table 2 Summary of key environmental risks identified after the Ajka red mud spill with an overview of estimated timescales of potential impact and current level of scientific understanding

Aspect of red mud creating potential risk	Key pathway(s)	Key receptor(s)	Timescale of potential impact				Level of understanding	Comment
			Days	Weeks	Months	Years		
Fine particle size distribution (fugitive dusts)	Air	Humans	■	■			M	Short-term risks similar to urban dusts. Possible latency in response—long-term studies on exposed populations ongoing
	Water	Aquatic biota (e.g., benthic smothering)	■	■			L	Particle size a possible contributor to negative response in aquatic bioassays. Not well quantified
Salinity/high ionic strength	Water	Aquatic biota	■				L–M	Longevity of issues in water minimized by dilution/remediation
	Soil	Terrestrial plants	■	■	■	■	M–H	Long-term salinization of soils identified as key risk in soil studies and of detriment to plant growth. Red mud removal efforts significantly limited spatial extent
High alkalinity	Water	Aquatic biota Humans (dermal contact)	■				M	Associated issues for metal(loid) mobility at high pH; buffering by dilution (water) or carbonation (soils/waters) minimizes timescale of issue
	Soil	Terrestrial ecosystems	■	■			M	Carbonation (via atmospheric CO_2 and respiration of soil organisms) likely to limit in surface layers within weeks
Metal(loid) enrichment	Water	Aquatic biota	■	■	■		L–M	Short-term issues coupled with pH. Key risks for those not efficiently removed during dilution (e.g., V and Mo). Emergency efforts effective at buffering
	Soil	Terrestrial plants	■	■	■	■	L–M	Possible genotoxic effects on plants in high dose rates. Residual issues surround anion exchange in soil–water systems. Red mud removal considerably limited this extent
Phosphorus enrichment	Water Soil	Aquatic/terrestrial primary producers/ecosystems	■	■	■	■	L	Residual issues for long-term cycling in soil–water environments and eutrophication risk. Largely unquantified
Radionuclide presence	Soils/air	Terrestrial ecosystems/humans	■	■			M–H	Risks well characterized and minimized by red mud removal

H high, *M* medium, *L* low

information about ecotoxicity at the alkaline end of the pH spectrum [39], these studies make available a valuable basis for informing future research in this area, alongside BRDA rehabilitation efforts.

Remediation and Recovery

Effectiveness of Short-Term Remedial Works

The emergency management efforts at Ajka comprised red mud removal from land and water, extensive dosing with acids (acetic acid, HCl, and H_2SO_4), and gypsum dosing of affected streams [7, 15]. Downstream trends in water quality during the emergency efforts highlight the effectiveness of this management in limiting the impacts of the residual releases from the site [15], while continuous monitoring of pH in the Torna Stream showed compliance with pH standards (<9) for the year after the spill [7]. Indeed, a key strategy of the neutralization, coupled with the building of check dams, was to prevent any pollution of the Danube and, in particular, transboundary impacts [7]. 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 neutralization of pH was accompanied by associated attenuation of some key elements of concern, such as Cr and Al [15]. V, however, retains mobility as vanadate throughout the ambient circumneutral pH range of the affected catchment (underlain by Triassic dolomites and limestones [40]). The fall in V concentration downstream is therefore 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 [15]. These field data are supported by laboratory experimentation that highlight that V, along with Mo, is not efficiently removed during neutralization, unlike other key contaminants such as Al and As (Table 3, [12]).

Around 10,000 tonnes of gypsum was used to dose waters downstream of the site for pH neutralization [7]. However, analyses of secondary deposits revealed that there are potential benefits of gypsum dosing, not just for neutralization of waters (Eq. 1), but also for carbon sequestration (and a potential industrial offset), and metal immobilization.

$$
\begin{aligned}
&2OH^{-}_{(aq)} + CaSO_4 \cdot 2H_2O_{(s)} + 2CO_2 \\
&\leftrightarrow CaCO_{3(s)} + SO^{2-}_{4(aq)} + 2H_2O_{(l)} + H_2CO_{3(aq)} \qquad (1)
\end{aligned}
$$

Secondary deposits at the gypsum-dosed sites were dominated by carbonate minerals [41]. Within these, there was a modest uptake of contaminants, notably As, Cr, and Mn [41]. This is consistent with laboratory simulations that also show a significant scavenging of As in secondary deposits (possibly as Ca-arsenate phases [12]). C and O stable isotope ratios of carbonate precipitates formed through gypsum dosing were used to quantify the importance of the neutralization process in sequestering atmospheric carbon dioxide. This process was particularly pronounced at the sites most affected by gypsum addition, where up to 36 % of carbonate-C appears to be derived from atmospheric in-gassing of CO_2 [41]. It was suggested that at such rates of sequestration, widespread gypsum dosing could offer a 3–4 % of the direct CO_2 emissions offset for the alumina industry through secondary carbonate precipitation [41]. However, full cost–benefit analysis and Life Cycle Assessment would be required for thorough evaluation of this, which may critically depend on gypsum sources (i.e., flue gas desulfurization or primary extraction). Gypsum dosing as a wider management practice in BRDAs [42, 43] therefore holds possible scope, not just for well-documented improvements in soil nutrient content and structure, but also in providing a modest emissions' offset for the industry, and potentially in limiting metal(loid) availability.

Landscape and Catchment Scale Recovery

Longer-term monitoring of the downstream aquatic environment, as well as low-lying terrestrial environments inundated by red mud, provides a good insight into the broader-scale success of remedial efforts after the spill. The recovery of red mud from affected fields (where a continuous >5 cm depth was apparent) has been shown to be vindicated by the various laboratory studies modeling the

Table 3 Concentrations of selected oxyanion-forming elements in Ajka leachate before and after neutralization with gypsum and HCl (0.2 µm filtered: after [12])

Determinant	Red mud leachate	6 M HCl (~5 mL L^{-1})	Gypsum (30 g L^{-1})
pH	13.1	8.3	9.8
Selected elements (mg L^{-1})			
Al	352	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*)

Values in parenthesis show percentage removed migration of red mud contaminants into underlying soil and their resultant toxicity [20, 21]. Systematic field sampling of affected floodplain soils after red mud removal has shown that no major residual signal of the inundation could be identified [9]. Measurements of pH, along with various key red mud indicators (Na and trace metals such as As, Co, Cr, and Mo), showed that less than 7 % of the flooded area had concentrations of metals that breached guidelines for the production of food and fodder crops, suggesting minimal leaching/mixing between red mud and the underlying arable soils before recovery works [9]. Even in the affected areas, the effective development of short-rotation coppice plantations for biofuel crops has been demonstrated by researchers in the years after the spill [44]. A three-stage process including composting, subsequent planting of annuals (for nutrient cycling and reestablishment of biological activity), followed by sapling planting of highly productive energy crops such as willow and poplar was recommended [44]. While the red mud did not restrict the potential planting of crops for food production, given the proximity of a nearby power station in Ajka, along with possible psychological concerns of local populations, biofuel production was deemed a particularly viable strategy for landscape restoration [44]. Other studies have shown phytoremediation using giant reed (*Arundo donax*) could reduce electrical conductivity of red mud by 25 % and removed trace metals (83 % Cd, 75 % Fe, 62 % Ni and 60 % Pb) in red mud contaminated soil from Ajka [45].

In the water environment, long-term monitoring (in the 3 years post spill) demonstrated that the concentrations of dissolved trace metal(loid)s downstream of the site did not exceed either European or US aquatic life criteria values (excluding one sample for Cd [46]). However, continued, albeit modest, elevations of As and Ni were apparent in the Marcal River 2 years after the spill compared to that observed in the predisaster period [46]. These elevations of As and Ni are consistent with the enhanced leaching from red mud when in contact with organics (e.g., where the red mud was plowed into agricultural soils [18, 21, 22]) which may suggest a residual signal of the red mud. However, further monitoring of the river systems would be desirable over the full range of hydrological conditions occurring in the affected streams to assess any such long-term trends. 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.

Stream sediments provide a better measure of potential long-term secondary sources of contamination within rivers affected by major tailings spills than solute samples [47]. Comparative sediment surveys between 2010 and 2013 showed that the signal of red mud was only apparent at a small number of locations in small channels near the site in 2013 (Fig. 2, [6]). Vanadium provides a good indicator of this recovery 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 (Fig. 2). This was considered to be a function of both the extensive dredging efforts (in the region of 80 km of the channel) and the fine particle size of the red mud (D_{50} of 3.4 μm) which lends itself to entrainment and downstream transport out of the system [6].

This physicochemical recovery has been coupled with rapid biological recovery. Some of the early colonizers (or possible survivors of the spill pulse) in the affected rivers were aquatic fungi (notably *Tricladium angulatum*) and diatoms (unicellular algae) observed in stream foam samples 1 month after the spill [30]. By this point, the acid dosing operations maintained the Torna stream pH to values <9 [30]. Longer-term monitoring of aquatic hyphomycetes colonization using leaf bags showed slightly slower rates of recolonization and lower species richness at impacted sites compared to unaffected reference sites [30], which may have been due to gypsum smothering. Nonetheless, a discernible recovery was noted in species richness by the spring 2011 [30]. The gypsum smothering was also highlighted as being an impediment to macroinvertebrate recolonization in the Torna stream [30]. Further downstream in the Marcal, official monitoring, however, showed the full recovery of macro-invertebrate communities in the year after the spill [48]. Crucially, fish populations also showed a recovery to prespill levels of species richness within 1 year of the accident in the Marcal River, providing an excellent indicator of aquatic ecosystem-level recovery [31], albeit some 25 km downstream of the most impacted areas of the Torna Stream. Rapid recolonization of native species saw species richness increase consistently during the 2 years after the spill, along with total abundance that recovered from <60 specimens 2 weeks after the spill to over 2000 individuals in the summer 2012 [31].

Research Efforts to Promote Red Mud Afteruse and Valorization in Hungary Since the Spill

The spill at Ajka put an immediate regulatory focus on risk assessments at other red mud depositories within the territory to limit potential environmental hazards. A key site identified in these reviews [1] was Almásfüzitő in northwestern Hungary where dated residue disposal practices from a closed refinery have led to documented issues with environmental releases [48] due to the floodplain location and inappropriate lining [1, 49]. The spill has also prompted efforts to find alternative solutions to disposal

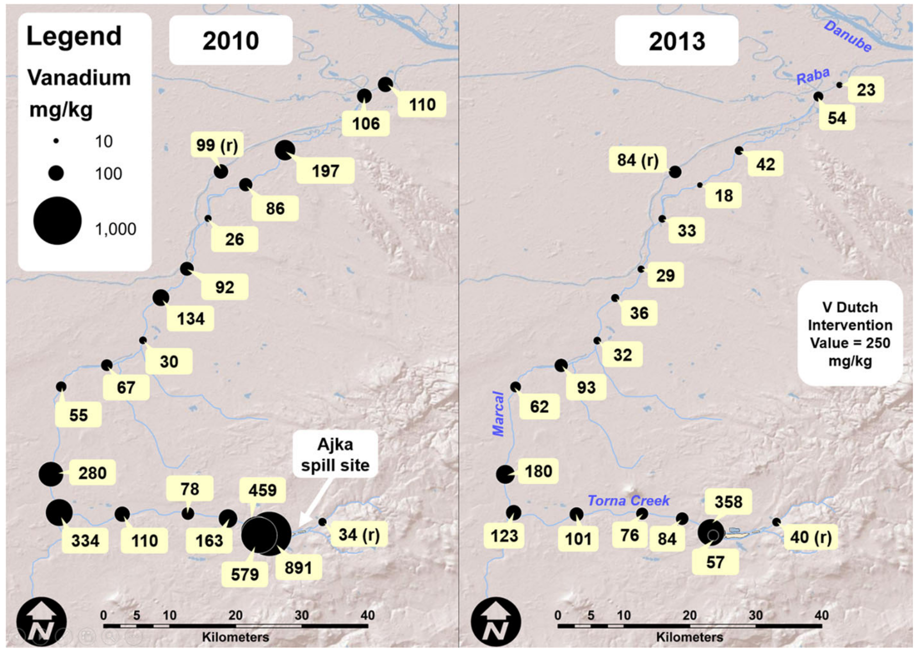

Fig. 2 Comparison of average vanadium concentrations ($n = 3$) in the Torna–Marcal system between 2010 and 2013 surveys. Values with (*r*) show reference sites on unaffected tributaries. After [6]

Table 4 Concentrations of selected critical raw materials in Ajka red mud (solid-phase, air-dried) and leachates (at pH 13.1)

Element	Example of use	Solid (ppm)	Aqueous (total, ppm)	Aqueous (<0.45 µm, ppm)
Ce	Catalysts, low-energy bulbs	368–405	0.42	<0.01
Co	Magnets, high-strength alloys	97–100	0.21	<0.01
Ga	LEDs, photovoltaics	79–89	2.44	2.3
Ge	Semiconductors	27	0.02	<0.01
La	Hybrid fuel cell component	130–150	0.75	<0.01
Nb	High-strength alloys and semiconductors	85	0.26	<0.01
Sc	Al–Sc alloys for aerospace applications	89	0.17	<0.01

After [53, 54]

and encourage efforts for red mud valorization. There has been a growing interest in recent years in value recovery from bauxite residue [50, 51]. Like many other red muds, Ajka deposits are enriched in a range of Critical Raw Materials [52] at modest to high concentrations (Table 4, [53, 54]). Analyses of leachate samples show that only a small proportion of the valuable element inventory in the red mud is soluble at source (gallium is a notable exception—Table 4), which means the recovery of these elements is likely to require significant energy input (sintering) or chemical transformation (e.g., using concentrated lixiviants).

The possibilities to recover rare earth elements (REEs) from Hungarian red mud with combined acid leaching and liquid–liquid extraction have been assessed in laboratory trials [54]. Furthermore, the possibility for heterotrophic bioleaching by benchmarking mineral acids versus small molecular weight organic chelators was determined [53]. The leaching tests for Hungarian red mud showed that La and Ce leaching efficiencies were the highest with hydrochloric acid (HCl) in comparison with nitric acid (HNO_3), sulfuric acid (H_2SO_4), oxalic acid ($C_2H_2O_4$), or citric acid ($C_6H_8O_7$). The REEs' leaching efficiencies using $C_6H_8O_7$ were comparable to those for mineral acids, albeit requiring a higher process temperature above 90 °C (vs. 25 °C, resp.) using the same contact times [53]. When different HCl concentrations are compared, the maximum metal leaching efficiency was achieved while using 6 M HCl (for instance, the leaching efficiencies were La ~ 98 % and Ce ~ 74 %, respectively). Liquid–liquid extraction using D2EHPA (di-(2-ethylhexyl)phosphoric acid) was applied to purify REEs from the leachate, and the achieved extraction-efficiency percentages were 96 % for Ce and 92 % for La [54]. Aluminum recovery from red mud using the organic acids generated by microorganisms was also studied. The bioleaching of Al from red mud from

Ajka using 17 species of filamentous fungi showed that the *Aspergillus niger* G-10 strain was the most efficient, extracting 141 mg L^{-1} of Al from 0.2 g dry weight red mud ($\sim$1 % Al in residue [55]). However, further research is needed to scale up and optimize the process to use bioleaching in heaps in BRDA management.

Opportunities for bulk reuse of red mud have also been assessed in several soil-restoration studies. One such microcosmic study aimed to reveal the beneficial effects of the Ajka red mud, as soil ameliorant, on a specific acidic sandy soil in Eastern Hungary [56]. It was reported [56] that red mud applied at 5 % w/w had positive effects on the degraded acidic sandy soil, significantly increasing soil pH, thereby improving soil texture and water-holding capacity. In addition, according to the ecotoxicological test results, 5 % w/w red mud had no significant adverse effects on *Aliivibrio fischeri* (bioluminescent bacterium), *Sinapis alba* (plant), *Triticum aestivum* (plant), and *Tetrahymena pyriformis* (unicellular animal) as assays of the soil ecosystem [56]. The applicability of Ajka red mud and soil mixture (RMSM) in the surface-layer component of landfill cover systems was also investigated in a field study in Hungary [57]. RMSM was removed from the flooded soil surface after the accident in Hungary where the slurry broke free, flooding the surrounding areas. Based on metal composition, it approximates to a 1:4 mixture ratio of red mud to the affected soils [15, 57]. The RMSM was mixed with low-quality subsoil originally used as a surface layer of an interim landfill cover in the two-step field study (lysimeters and in field plots). According to [57], the addition of RMSM to the subsoil at up to 20 w/w % did not result in any ecotoxic effect on the studied test organisms (*Aliivibrio fischeri*, *Sinapis alba*, *Triticum aestivum*, and *Folsomia candida*), but it increased the water-holding capacity, and the microbial substrate utilization became three times that of subsoil after 10 months. The soil amelioration in combination with metal recovery may thus represent a viable approach for red mud management, from both environmental and economic points of view.

Conclusions and Opportunities for Future Management

Given the size of Ajka spill and the immediate international concern, the scientific studies in its aftermath have offered a largely positive prognosis on the long-term impacts of the spill on terrestrial and aquatic environments. The short-term impacts of the highly caustic, saline, and metal(loid)-rich sludge were effectively managed around source areas, while fugitive dust issues associated with the fine particle size did not show demonstrable impacts beyond those expected for similar fine urban dusts. The numerous biological studies in the affected areas suggest short-term impacts were dominated by salinity stress and the caustic nature of the spill material, but these rapidly passed in the water environment. Metal(loid) availability appears to be a secondary issue, but is the one regarding which 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), or those that are subject to potentially long-term cycling in soil–water systems (e.g., As, Cu, and Ni). Other longer-term issues highlighted include salinization of soils and potential nutrient enrichment as a result of phosphorus in the red mud suspension. However, the areas affected are relatively small given the effective red mud-recovery works. Large-scale landscape remediation using rapidly growing short-rotation coppice for biofuel generation has also demonstrated the productive use of affected agricultural land [44] and may be an appropriate avenue for investigating for BRDA rehabilitation and afteruse.

The behavior of red mud leachate at Ajka offers some insight into approaches adopted for long-term leachate management at BRDAs. Neutralization (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 neutralization (notably V and Mo) and, as such, should be a particular focus of studies looking at leachate treatment (e.g., bioremediation using wetlands [58]). Gypsum dosing may offer advantages beyond neutralization through limiting metal availability and via sequestration of modest quantities of atmospheric carbon.

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 [45]. Improvements to the physicochemical environment in the affected and downstream areas have been reported, alongside the biological ones. Furthermore, the spill has also catalyzed research efforts to find appropriate end-uses for red mud, both within Hungary and further afield. As with many other red muds, significant inventories of Critical Raw Materials were apparent at Ajka, with initial studies demonstrating effective leaching and recovery technologies for a range of elements [55]. Bulk reuse of recovered red mud has also been effectively tested under pilot field conditions as an ameliorant to poor, acidic sandy soils, and in landfill cover systems without negative biological impacts. As such, the body of scientific information that has arisen after the Ajka spill has not only provided valuable information on the environmental risks associated with red mud that can feed into more robust BRDA management plans elsewhere, but also reinforced the potential for value recovery from red mud depositories.

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