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Recovery of the Torna-Marcal river system after the Ajka red mud spill, Hungary

Environmental Impact Statement

An increasing quantity of bauxite processing residue (red mud), a by-product of alumina refining is produced globally each year. The largest documented environmental release of this residue occurred in western Hungary in 2010 after the failure of a retaining wall of a residue impoundment. The highly caustic, metal-rich slurry had major immediate environmental impacts on the receiving water courses. This paper highlights the rapid recovery of the affected rivers after the spill due to the physical nature of the spill material (fine grained which lends itself to downstream transport) and the extensive remedial efforts undertaken in affected reaches.

Geochemical recovery of the Torna-Marcal river system after the Ajka red mud spill, Hungary

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ELECTRONIC SUPPLEMENTARY INFORMATION: Data file of total elemental concentrations for the 2010 and 2013 surveys attached separately.

ABSTRACT

The failure of the Ajka red mud depository in October 2010 led to the largest single release of red mud into the surface water environment. This study provides a comparative assessment of stream sediment quality in the Torna-Marcal-Rába catchment between post-disaster surveys (2010) and follow up surveys at an identical suite of 21 locations in 2013. The signature of red mud apparent in initial surveys with high Al, As, Cr, Na, V was only apparent at a small number of sample stations in recent surveys. These constitute <1 km of stream, compared to the >20 km reach of affected sediments in the immediate aftermath of the spill. Concentrations of red mud-derived contaminants are predominately associated with fine fractions of the red mud (<8μm). This enhances transport out of the system of

red mud-derived contaminants and, along with extensive remedial efforts, has substantially limited the within-channel inventory of potentially ecotoxic metals and metalloids.

INTRODUCTION

There have been in excess of 100 major failures of primary ore extraction or processing tailings facilities globally since 1960.[1] The vast majority of these failures occur at active mining or processing sites[2] with the most common cause cited as being extreme, or uncommon rainfall events prior to failure.[2] From a European perspective, the tailings spills at Aznalcóllar in southern Spain in 1999[3] and the Baia Mare / Baia Borşa disasters in Romania in 2000[4] played a major role in re-shaping EU legislation on the management of tailings facilities. European Union Directive 2006/21/EC on the management of waste from extractive industries (the "Mining Waste Directive"[5]) set out implementing measures to minimise the risk of further disasters within Europe. These measures included initial inventories of sites posing risk to the environment or human health and the development of disaster management plans at recognised sites.[5] Unfortunately, the implementation of the Mining Waste Directive was not sufficiently advanced to prevent the release of around 1 million m^3 of highly alkaline, saline, metalliferous red mud from an impoundment at Ajka in western Hungary in October 2010.[6] Red mud (or bauxite processing residue) is the fine fraction by-product of the Bayer process for refining bauxite for alumina production. The nature of the material in the depository also meant it did not fall under the auspices of other preventative legislature such as the Seveso II Directive. This Directive aims to minimise and prevent the occurrence of major disasters associated with certain hazardous chemical wastes. However, given NaOH (as the key constituent of the red mud liquor) is not deemed a dangerous substance at the concentrations it was present at the Ajka disposal site (category R34 under annex 1 of the Dangerous Substances Directive; 67/548/EEC), it did not fall under Seveso II reporting requirements.[7]

A number of failings in operational management and depository structure have been highlighted in official reviews since the disaster,[8] while a range of scientific studies have assessed the impacts of the spill. Although there have been large spills of alkaline wastes in the past,[9] the Ajka disaster was the single largest release of highly caustic waste to the water environment recorded, and as such, brought with it considerable uncertainty about the short and long term environmental impacts of the release. Since the disaster, a suite of studies have assessed a range of pathways and receptors for the red mud spill, the findings of which are summarised below.

Some of the key findings of the work in the spill aftermath highlight the very fine grained nature of the red mud, which as a fugitive dust puts it in a similar risk class as urban dusts,[10] while in aquatic systems lends itself to downstream transport and dilution.[11] Furthermore, the red mud is rich in various metals and metalloids of potential environmental significance.[12-13] Geochemical studies have highlighted that many of the metal(loids) immediately highlighted as a potential concern are not very mobile under ambient conditions once the leachate is neutralised.[11,13] However, in common with other alkaline residues,[14] the presence and potential mobility of metal(loids) which form oxyanions has been highlighted. These include As, Cr, Mo and V.[11,13] Biological studies have highlighted impacts of red mud on plant growth,[12,15] primary producers,[15-16] soil biota[17-18] and genotoxic effects of vanadium in higher plants.[19] However, short term impacts on human health have not been identified.[20-21] The difficulty in specifying any individual causal agent given the concomitant high salinity, alkalinity, metal(loid) concentrations and fine particle size which could all be stressors to a range of biota is highlighted in many studies.

The Hungarian government invested 38 million Forint (~€127 million) in demolition, reconstruction and environmental remediation in the aftermath of the spill.[22] Immediate interventions included acid dosing at source, the addition of around 10 000 tonnes of gypsum to affected rivers and the building of check dams to encourage buffering of waters and sedimentation.[23] Longer term measures included channel dredging and the removal of red mud from affected floodplain areas. Extensive monitoring by regulatory agencies was also carried out in the year after the disaster[24-25] while routine biological monitoring suggested wildlife recovered in the affected systems according to government press releases.[22] Recent studies based on ambient water quality monitoring data have however highlighted the persistence of As and Ni in high concentrations in the water column in the two years after the disaster.[26] Many other studies also caution of the need for longer term monitoring of affected systems for a more comprehensive risk assessment,[27] for example from the effects of sodification of soils or slow leaching of oxyanionic contaminants under ambient conditions.

Fluvial sediments provide a good indication of the long term exposure of a river system to both aqueous and particulate contaminants as well as identifying sinks of contaminants that could be potentially remobilised to the water column in the future. In the aftermath of major base metal mining tailings failures elsewhere, sediment studies have highlighted the longevity of the pollution issues and also the timescales for recovery of the systems. In a review of river system recovery after major sediment spills, Bird et al.[28] highlight how local geomorphology and remedial efforts can have a major influence on long term sediment concentrations. In affected river systems confined to narrow valleys, sediment metal concentrations can rapidly recover to pre-spill conditions within a year.[28] However, in systems with larger floodplain systems, episodic reworking of floodplain sediments means that sediment contamination after major spills can remain readily identifiable 2-3 years after the major spill, as was highlighted in the Vişeu River, Romania, after the Baia Borşa and Novat-Roşu tailings failures in 2000.[28-29] Surveys of the Ríos Agrio and Guadiamar over a year after the Aznalcóllar spill in southern Spain highlighted the signal of the metal and sulphide-rich tailings in fluvial sediments[30] while other studies also highlighted the bioaccumulation of As, Cd, Cu and Pb in grasses to potentially toxic levels 18 months after the spill.[31] However in the Aznalcóllar case, the extensive removal of contaminated material from floodplain areas is thought to have aided recovery relative to other spills.[28]

This study aims to assess the changes in sediment quality across the river systems affected by the spill, the Torna Creek, Marcal River, Rába River and Mosoni-Duna, which form a major tributary of the Danube. Through comparative assessment of sediment quality this study aims to (a) highlight the distribution of any residual red mud-derived contaminants in the Torna-Marcal system, and (b) assess the effectiveness of both the natural attenuation and remedial efforts in the system since the disaster.

METHODS

Study site

Sample stations along the course of the Torna Creek, Marcal, Rába and Mosoni-Duna rivers were sampled as in [11] (Figure 1). These sample locations covered reference sites on the Torna Creek (code: T2), Marcal (M1) and Rába (R1) rivers as well as 18 sample stations directly affected by the spill as it propagated downstream. Bedrock geology in the upper catchment is dominated by dolomites and limestones of Triassic age which lie beneath a sequence of fluvial marls, slates and interbedded sands of Miocene age.[32] Land use in the catchments is predominantly agricultural with some heavy industry in the towns of Ajka and Győr, while the Torna, Marcal and Rába are all extensively channelised with levees minimising the extent of floodplain, particularly downstream of Pápa.

Sediment samples

At each station triplicate bulk (≈500g) sediment samples were collected by aggregating three randomly collected sub-samples from a 12m^2 area of stream bed (9 separate locations sampled at each reach to give three replicates). Sediments were homogenized, air-dried, disaggregated gently and sieved (2mm aperture) prior to microwave-assisted total digestion (aqua regia and HF) following standard methods.[33] Elemental concentrations in digests were analysed using a Perkin Elmer Elan Inductively Coupled Plasma Optical Emission Spectrometer (Optima 5300 DV ICP-OES) for all elements quoted hereafter. Selected dried and disaggregated samples were also prepared for particle size analysis.

Particle size distributions on selected samples were determined through taking approximately 0.2g sample which was then shaken with 5ml DIW in a 10ml tube. The sample was then ultrasonicated for 30 mins, soaked for 24 hours and ultrasonicated again for 30 minutes before analysis on a Malvern Mastersizer 2000E laser granulometer. Prior to each sample addition background laser intensity (> 80%) was determined separately and subsequently subtracted from sample data. For analysis the dispersed slurry was then added dropwise to a Malvern Hydro SM small sample dispersion unit (pump speed 1500 rpm) until a laser obscuration value of 10-12%

was achieved. Sample data were calculated from the mean of three separate scans. Particle size data were analysed using the GRADISTAT v.4 program.[34]

Statistical and spatial analyses

All statistical analyses were undertaken in Minitab v15. Data were not normally distributed even after log-transformation (Kolmogorov-Smirnov $p>0.05$) so non-parametric methods were used to compare sediment metal(loid) concentrations between years and explore relationships between metal(loid) concentration and particle size. Principal Component Analysis (PCA) was undertaken on standardized sediment element concentration data.

Spatial patterns in sediment metal(loid) concentrations were assessed using spatial interpolation (kriging) tools in ArcGIS v.9.3. This allowed estimates of the length of channel reach above a range of published threshold values to be computed for each of the sample years.

RESULTS

The downstream trends in selected elements enriched in red mud at the Ajka site are shown in Figure 2 for the two respective survey years. The general patterns apparent for all elements are that the exceptionally high concentrations of red mud-derived elements in source areas (K1-3: 0-5km from the spill site), areas of preferential deposition in the Torna Creek (T5-6) and the upper Marcal River (M2-3) in 2010 (25-30km downstream of the spill site) are not apparent in the 2013 survey. For V and As, which were both highlighted as highly mobile in the affected rivers shortly after the spill,[11] the total sediment concentrations in the lower parts of the Marcal and Raba are consistently lower (and often close to detection limits for As) in the present survey than in 2010. Only for Na, do sediment concentrations in the lower reaches of the Marcal appear to be higher in 2013 than in the post-spill surveys (Figure 2).

Comparison of sediment metal(loid) concentrations with sediment quality guidelines[35-36] shows that far shorter reaches of channel are in breach of potential ecotoxicological thresholds in 2010 than 2013. Table 1 shows the length of the river system affected by the spill in breach of a range of target values based on spatial interpolation between successive sample locations. The Threshold Effects Levels (TEL) and Predicted Effects Levels (PEL[36]) are being used in some EU states as informal guidance on fluvial sediment quality. The TEL marks the lowest concentration at which negative effects on aquatic biota are apparent in toxicological tests, while the higher Predicted Effects Levels (PEL) gives the concentration above which negative impacts on sediment-dwelling organisms would be anticipated.[36] However, red mud is characterised by potential contaminants for which such formal fluvial sediment contamination guidelines have not been formulated such as Co and, notably, V which has been highlighted as a particular concern given its solubility in pentavalent form as vanadate under the ambient, circum-neutral pH conditions of the affected systems.[13] For these elements, initial screening against the Dutch Intervention Values for contaminated soils, which are generally less precautionary than the TEL/PEL approach. It must be stressed that these guidelines offer nothing more than simple screening tools for total sediment metal(loid) concentrations and offer no indication of whether negative impacts on aquatic biota would occur in this system, given that is dependent on the form and bioavailability of the contaminants, alongside the nature of exposed communities and their local physico-chemical environment. However, they are useful in assessing relative enrichment of certain metal(loids) in the system and the temporal trends since the spill. For most thresholds, a substantial decrease in the length of the system exceeding guideline values is apparent between the two survey years (Table 1). For example, some of the key contaminants of concern in the Ajka red mud such as V and As show falls from around 20km of stream being affected in initial surveys, to less than 1km of channel being in breach of Dutch Intervention Values in 2013. When comparing against the TEL and PEL values, it is also apparent that overall reaches above prescribed thresholds has fallen for the majority of contaminants listed in Table 1. However, the TEL values should be used with caution in this case, given thresholds are exceeded for As, Cr, Pb and Ni at some of the reference sites in the system.

Principal component analysis (PCA) was found to be a useful tool in highlighting the end members of sediment elemental composition in the immediate aftermath of the spill[11] and is useful here in highlighting the change in chemical signature of the fluvial sediments over time. In updated analysis, incorporating all samples from 2010 and 2013, three important factors are identified, which account for 73.0% of the variance in the data (from scree analysis and taking a 10% variance threshold[37]). The signature of the red mud is apparent through enrichment of a range of major and minor elements. Na, Al, Fe and Ti are all in relatively high concentrations in red mud, while enrichment of V, Cr, As, Ni, Co, Ga and Zr are apparent here. The red mud is apparent as one end member of the sediments in the Torna-Marcal system; plotting to the right hand side of Figure 3 with high values for factor 1. The series of samples that plot along the line roughly parallel to PCA 1 represent mixing of red mud with the unaffected sediments in the catchment which plot to the left hand side of Figure 3. The only sample sites in the recent survey that suggest red mud enrichment are those from locations M1 and M2 in the upper Marcal River. The former is something of a curiosity given this site on the Marcal River upstream of the confluence with the Torna Creek (in which the slug of red mud passed). However, consultation of 10m Digital Terrain Models indicates the possibility of the spill material backing up to this location given the low gradient nature of the system. Such a signal was not apparent in initial surveys at M1.

The unaffected sediments are relatively enriched in K, Mg and Ba which are indicative of lithogeneous weathering in the catchment which is underlain by Triassic dolomites. Extensive gypsum smothering of benthic habitats was apparent after the spill as part of emergency remediation efforts. These sediments comprise gypsum as well as carbonate-dominated secondary precipitates[38] that in original surveys formed a distinct population of fluvial sediment samples that plot to the upper left of Figure 3 with high values on factor 2. This population is characterised specifically by elevated S content (142-152 g/kg). While Ca is abundant in the gypsum (and associated secondary carbonates), it is a poor predictor of gypsum smothering given it is also present in red mud (concentrations in the region of 50 g/kg) as well as unaffected sediments in the system (range 10-50 g/kg). Figure 3 also shows two of the more recent samples in a very small tributary of the Torna Creek from close to the red mud impoundment appearing geochemically distinct to the lower centre of the plot. These samples were characterised by enrichment of many elements indicative of unaffected sediments (e.g. Ba and Mg) but are enriched in Mo (range: 60-119 mg/kg), albeit at levels lower than Dutch Intervention Values (200 mg/kg).

Particle size

Analysis of grain size of the sediments also highlights the physical distinctiveness of the red mud from reference sediments in the system. Figure 4 shows a bimodal particle size distribution in the red mud which can be classified as a poorly sorted mud.[39] This contrasts with reference sediments upstream of the Ajka site which are categorised as unimodal, poorly sorted muddy sands, with a D_{50} of 83µm and comprising predominantly silicates.[11] The bulk (94.9%) of the distribution of reference sediments is across fractions coarser than 8µm (Figure 4). The affected sediments in the Torna Creek system show a markedly different particle size distribution between the two sample occasions, with significantly (Mann Whitney: W: 876, d.f.: 46, P <0.001) larger D_{50} in the recent survey (median: 84.4mm, range: 7.5 – 516.5) than in 2010 (median: 4.1; range: 1.8 – 8.6: Figure 5). Caution should be heeded when assessing differences in particle size distribution between two isolated sampling campaigns, especially given the different sample months (November 2010, September, 2013). However, given the average flow conditions at the time of sampling (11.5 m^3/s in 2010 compared with 5.5 m^3/s in 2013 at a permanent gauging station at M7) were more conducive to fines transport in 2010, the patterns are likely controlled by the red mud in the system shortly after the spill.

The contrasting particle size distributions between the red mud source term and reference sediments (Figure 4) allows particle size distribution to be used as an additional tracer of the dispersal and legacy of red mud across the Torna-Marcal catchment. There are very strong (Spearman Rank correlation coefficients: r_s: 0.69-0.86) and significant (P <0.001) positive relationships between the proportion of sample that is fine fraction and metal(loid) concentration across all sample sites and sample years. These are displayed in Figure 6, with the percentage of fine material in the sample (taken in this case as being fine silt and finer, <8µm: given this covers 94.9% of the particle size distribution of red mud, and only 6.1% of reference samples: Figure 3) plotted against metal(loid) concentrations. The aggregated data for the two sample years also reinforces the patterns of generally higher metal(loid) concentration in the earlier 2010 survey.

DISCUSSION

The hotspots of red mud deposition in initial (2010) surveys were typically in the lower Torna Creek and upper Marcal River. These were reaches characterised by a largely natural channel planform, slow flow and dense riparian and marginal macrophytes (dominated by *Phragmites australis)* conducive to deposition of fine sediments. This was unlike the upper Torna Creek which is heavily channelized (straight engineered channel with trapezoidal cross section) and velocity typically two to three times that of the lower Torna Creek.[11] The sites where red mud accumulated in the lower Torna Creek and upper Marcal were subject to intensive red mud removal and dredging.[23] Around 80km of the Torna Creek and Upper Marcal were subject to dredging which was completed by the end of 201,[23, 40] and removed around 60,000 m^3 of red mud and red mud-contaminated sediments from the affected rivers.[23]

Previous workers have highlighted the characteristic fine nature of the red mud, with Gelencser et al.[10] showing the distribution of resuspended red mud peaking marginally above a 1µm aerodynamic diameter. Other analyses of red mud from the impoundment shortly after release showed a peak of particles centring on 0.7µm corresponded with nano-particulate heamatite aggregates,[11,13] while the coarser peak (centring on 3µm) was identified as cancrinite, a common sodium aluminosilicate mineral in red mud.[41] These fractions are typically finer than those documented in systems impacted by tailings spills from base metal refining. For example, sediment-borne contaminants are concentrated in fractions of the order of 10-500µm after the Aznalcóllar spill in southern Spain in 1999[30] and the signal of metal contamination remained in the river system several years after the spill.[42]

The strong, significant correlations between metal(loid) concentration and proportion of fines in the sample (Figure 6) demonstrate: (a) the significance of metal(loids) within the fine fraction of the sediments, and (b) that the fine fraction red mud is the predominant source term and vector for many contaminants through the system. Previous analyses showed many contaminants (notably As, Co and Cr) to be associated with residual hard-to-leach fractions of the red mud-affected sediments which is likely to limit and therefore are less likely to be remobilised from the fine fraction red mud.[11] Other workers[13] found Cr to be present predominantly in trivalent form substituted into haematite in source material, so while Cr is present in total concentrations in sediments that are above contamination screening guidance values, under the ambient circum-neutral pH of the Torna-Marcal system,[11] remobilisation to the water column would be minimal. As such, while the red mud will be readily entrained in the water column, the impacts of contaminated sediments during transit and on downstream systems is likely to be tempered significantly by the limited bioavailability of many of the contaminants present in the red mud.

While the dominant trends are similar between contaminants considered in Figure 6, there are some subtle differences between elements, which may reflect their occurrence or behaviour in the Torna-Marcal system. For example, the correlation between As concentration and the percentage of very fine silt fraction is slightly noisier than or other red mud-derived contaminants. This may suggest a more widespread occurrence of As throughout lithogeneous sediments in the catchment or possibly increased mobility and cycling of As within the fluvial system given the As elevations are not consistent with other red mud-derived elements. The former would be anticipated given the presence of the Csabpuszta bauxite deposits and Ajka Coal Formation (Cretaceous) in the upper catchment,[43] which can have modest to high As concentrations. The latter would be consistent with the elevated aqueous As concentrations observed in the Marcal catchment since pre-spill conditions[26] which may be a consequence of leaching of As from red mud affected soils (e.g. the reductive dissolution of As in organic-rich sediments). This has been shown in experimental conditions to be enhanced by mixing red mud with organics (for example where thin layers of red mud were ploughed into topsoil); a release that is largely dependent on the final pH of the soil after mixing with red mud.[44]

One of the other key characteristics of the fluvial sediments in the immediate aftermath of the spill, was the signal of gypsum amendment (to aid neutralisation of the highly alkaline leachate). These gypsum affected reaches were apparent around source areas (K2-3) but most notably in the middle reaches of the Marcal system (samples M7, M10-M11: Figure 3) from where neutralisation operations were coordinated. Scavenging of mobile metal(loids) from the water column in secondary neo-formed carbonate precipitates, which were a product of gypsum dosing, was highlighted as being at least a transient sink for As and V in previous studies.[13,38] Coupled with the lack of evidence of any persistence of gypsum-dominated sediments in the system (Figure 3), the low As and V concentrations in these gypsum-dosed reaches in the recent surveys suggests that risks of remobilisation of large inventories of metal(loids) from remediated sediments is not a long term issue. Given that many sediment sample sites were in the vicinity of the road bridges over the streams which were also the deployment locations for gypsum dosing, the lack of high Ca and S sediments in these reaches is good evidence that the remedial efforts themselves have left no lasting negative legacy on the system. Furthermore, there were no observable hardpans at any of the sample stations in 2013. Concerns were raised for example of the effects of carbonate hardpan formation[15] which can limit oxygen diffusion through benthic sediments and negatively affect invertebrate communities as has been demonstrated in other freshwater systems subject to benthic smothering with mineral precipitates and fines.[46-47]

Conclusions and management implications

Given the scale of the red mud release from the Ajka site, and the international concern surrounding the potential long term environmental impacts of the spill,[47] it is encouraging that recent surveys show that the geochemical signal of the red mud is largely absent from the affected downstream river systems. This is also consistent with official monitoring data on water quality and biological indices of river system health.[23] The extensive and rapid remedial efforts at Ajka, led by Hungarian scientists and authorities, served to neutralise and contain residual water releases from the site in the short term, while longer term within-channel dredging and recovery of thick red mud deposits in riparian areas has significantly limited the prospect of remobilisation of contaminated floodplain sediments. The recovery of the Torna-Marcal system has been assisted by the fine-grained nature of the red mud, and importantly the concentration of most contaminants of concern in the red mud (e.g. As, Cr, Ni, V) within the very fine fraction of red mud. As studies immediately after the spill suggested, the fine-grained nature of the spill material lends itself to downstream transport and dilution.[11] The very strong relationship between particle size and metal(loid) content highlighted here reinforces this notion. Ongoing monitoring of the system would still be prudent and should focus on (1) the risks associated with potential increased salinity and alkalinity of affected soils and waters (e.g. sodification), (2) the fate of metal(loids) in areas where red mud was ploughed into soils, and (3) any long-term biological response to red mud exposure. However, the recent surveys undertaken here highlight the rapid removal of much of the contaminated material released from the Ajka red mud spill through either remedial efforts or dilution. As such the long term impacts of contaminant metal(loids) released into the Torna-Marcal system are unlikely to be as recalcitrant as in other systems subject to notable tailings impoundment failures.

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FIGURES

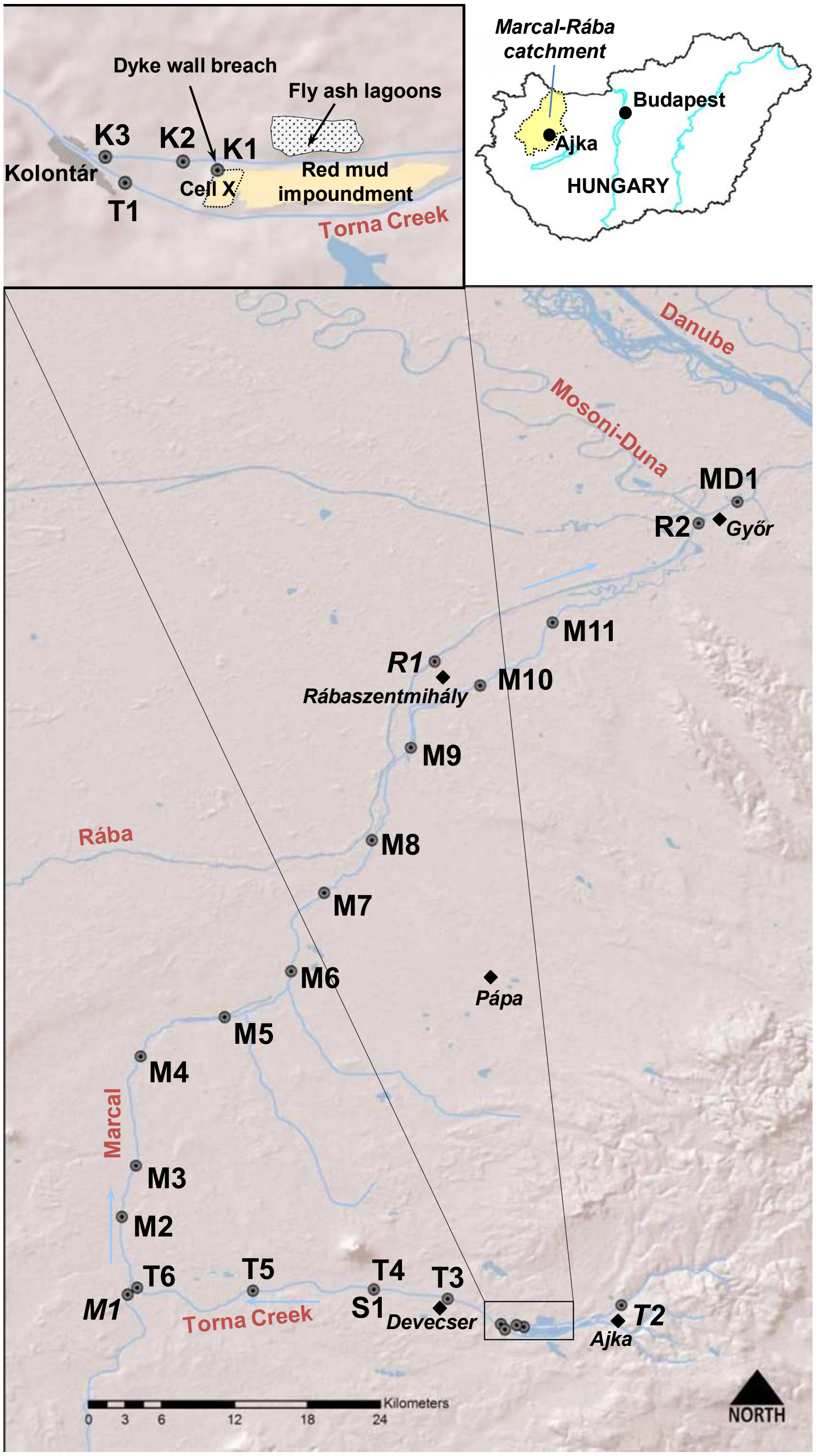

Figure 1. Location map of sample stations throughout the Torna Creek (sample prefix T), Marcal River (M), Raba (R) and Mosoni-Duna (MD). Location of K1 (source sample) is Lat 47^{0}05'20N Long 14^{0}29'43E.

Figure 2. Downstream trends in concentrations of key major and minor elements through the Torna-Marcal system between 2010 and 2013 surveys

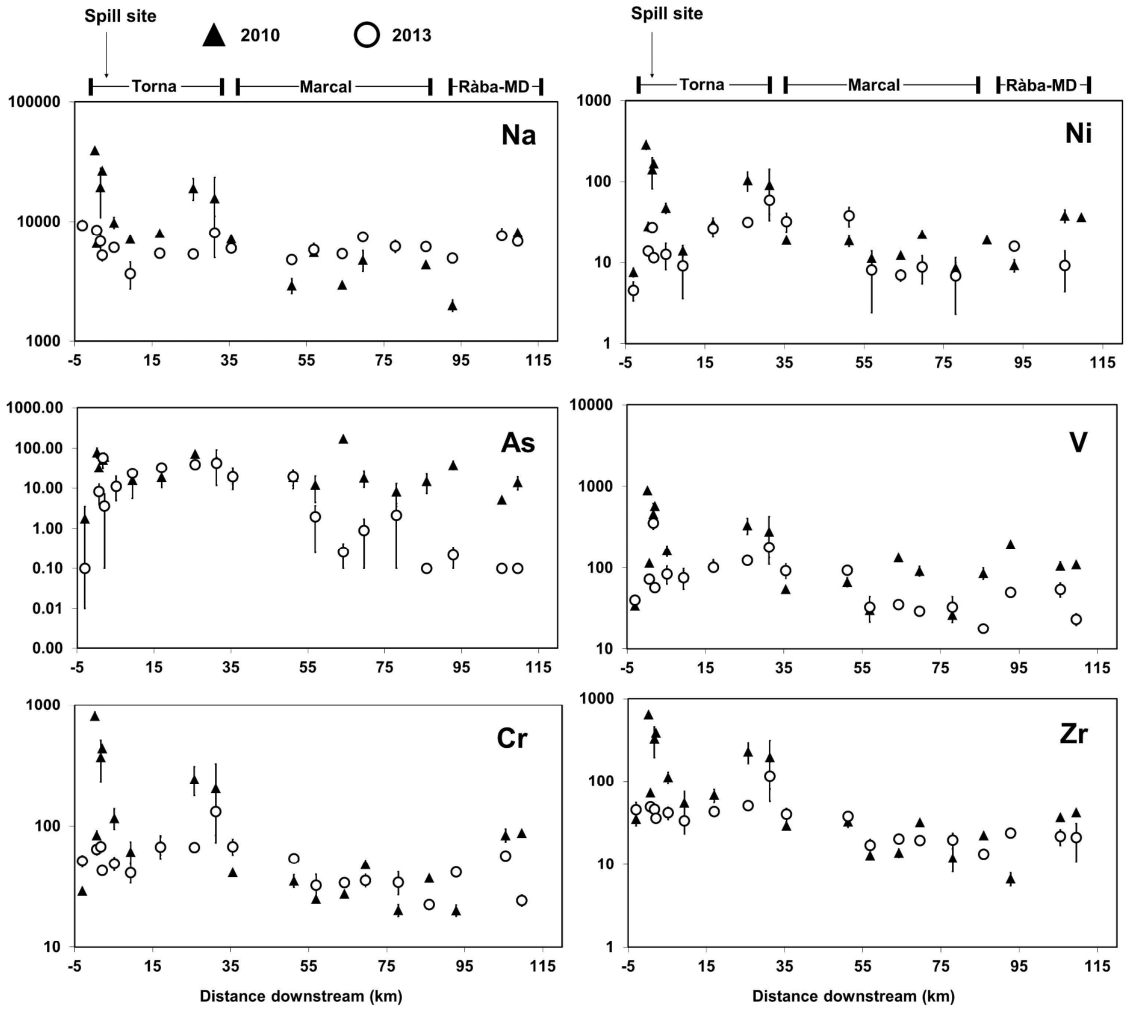

Figure 3. Principal Component Analysis of elemental concentrations in sediments in 2010 and 2013 surveys by site (left panel) with eigenvectors for key elements (right panel).

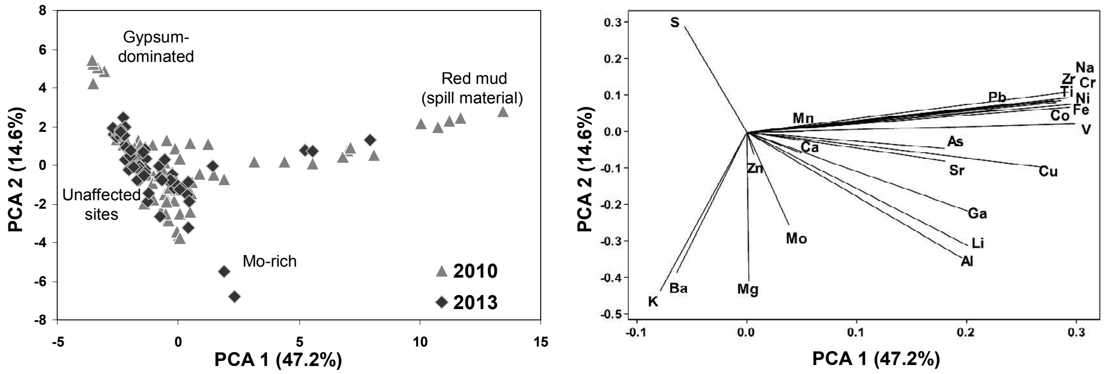

Figure 4. Mean particle size distribution of red mud (sample K1) and reference sediments in the Torna Creek (sample T2). Error bars show standard deviation (n = 3). Where not visible error bars are plotted within the sample symbols.

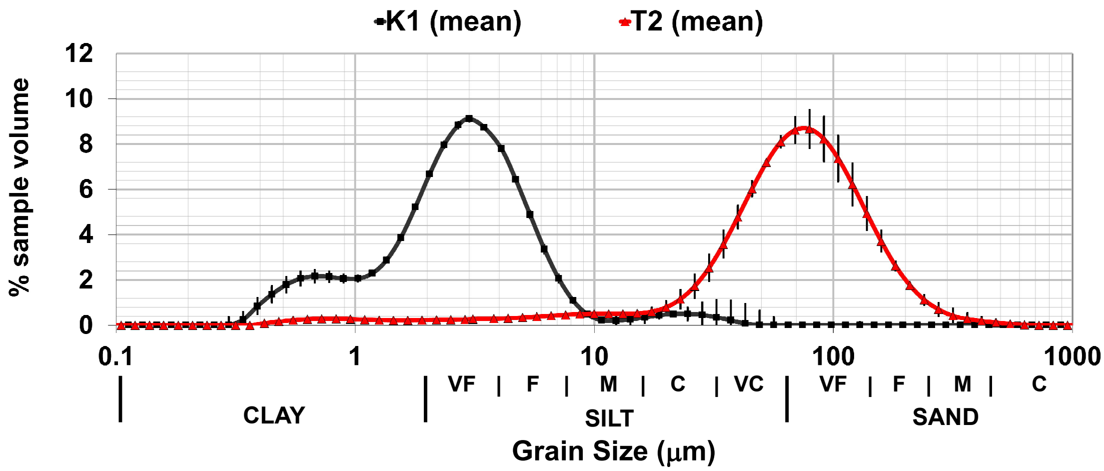

Figure 5. Distribution of D_{50} values for sediments in the Torna Creek in 2010 and 2013 surveys (n = 24 for each year).

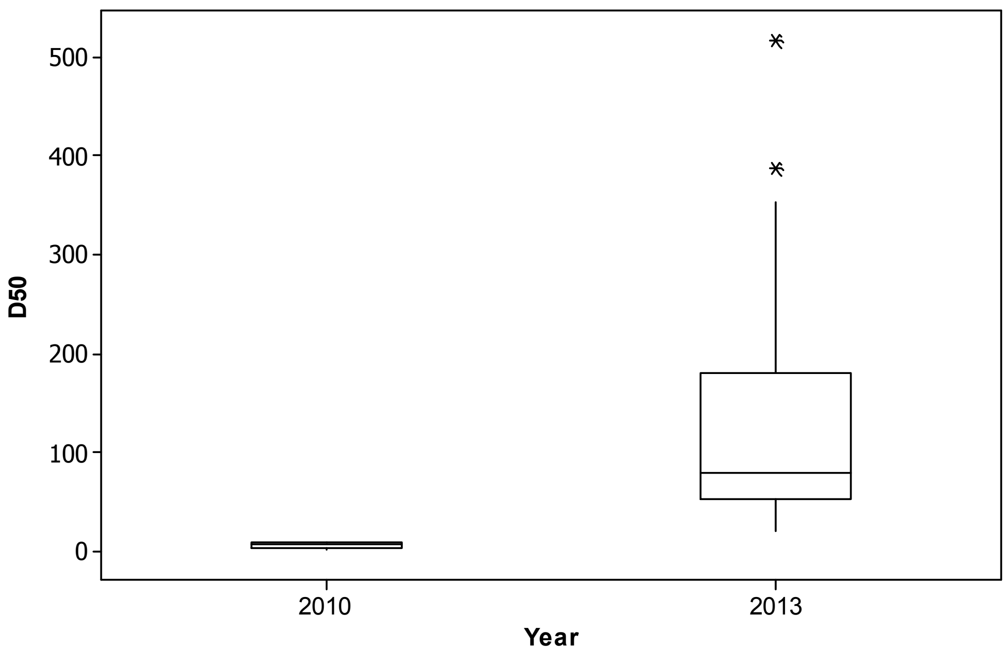

Figure 6. The relationship between the proportion of very fine silt (<8μm) in fluvial sediments and metal(loid) concentrations through the Torna-Marcal system in 2010 and 2013.

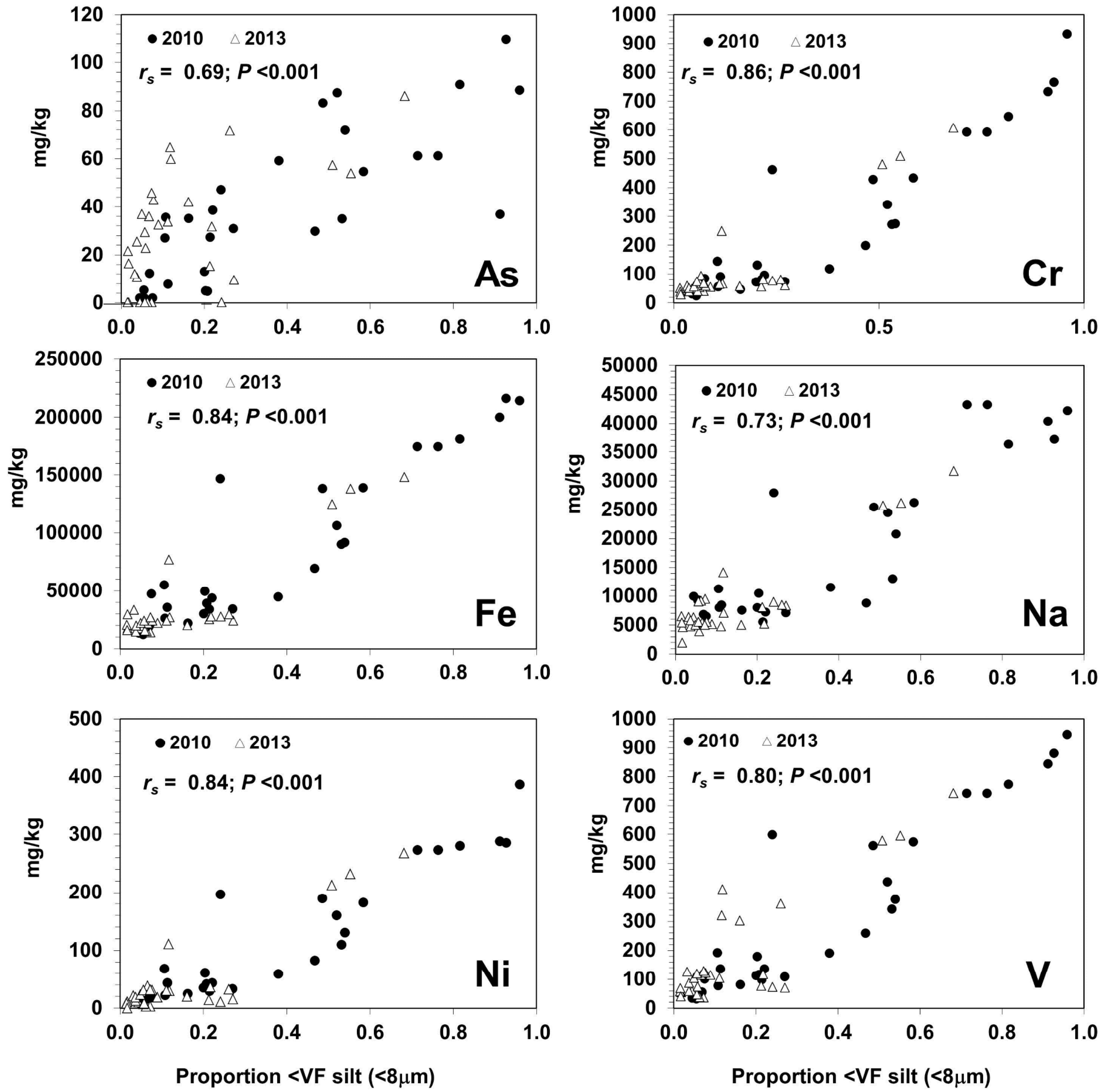

Table 1. The length of stream channel with average sediment metal(loid) concentrations in excess of standard soil and sediment contamination screening guidance in 2010 and 2013. (-): no prescribed threshold value.

		As	Co	Cr	Cu	Ni	Pb	V	Zn
Dutch Intervention Values (mg/kg)		55	240	380	190	210	530	250	720
PEL (mg/kg)		17	-	90	197	36	91.3	-	315
TEL (mg/kg)		5.9	-	37.3	35.7	18	35	-	121
2010 (km)	**Dutch**	**23.1**	**0**	**1.1**	**0**	**0.2**	**0**	**18.3**	**0**
	PEL	*71.8*	-	*31.1*	*0*	*33.8*	*2.2*	-	*0*
	TEL	105.2	-	68.2	2.0	64.4	40.2	-	1.8
2013 (km)	**Dutch**	**0.1**	**0**	**0**	**0**	**0**	**0**	**0.4**	**0.6**
	PEL	*45.7*	-	*12.4*	*0*	*19.0*	*1.7*	-	*0*
	TEL	54.1	-	56.6	0	43.5	18.4	-	0.7