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PHYSICAL STABILITY AND REHABILITATION OF SUSTAINABLE AQUATIC AND RIPARIAN ECOSYSTEMS IN THE RIO GUADIAMAR, SPAIN, FOLLOWING THE AZNALCÓLLAR MINE TAILINGS DAM FAILURE

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ABSTRACT

The April 1998 breaching of the Aznalcóllar tailings dam at Boliden Apirsa's Spanish Aznalcóllar/Los Frailes Ag-Cu-Pb-Zn mine resulted in the flooding of the Ríos Agrio and Guadiamar with heavy metal- and arsenic-rich tailings and water. An interdisciplinary geomorphological-geochemical research programme is underway to: (i) monitor the recovery of the river, especially in terms of the geochemical and physical controls on the distribution of potentially hazardous elements between sediment and waters, and (ii) provide essential field data which will be used in formulating strategies for re-establishing sustainable aquatic and riparian ecosystems that are in dynamic equilibrium with their environment. The work to date has resulted in the establishment of a baseline geomorphological survey and post-clean-up sediment and water geochemistry which is the basis for ongoing research to (a) evaluate the likely pattern of river channel and floodplain adjustment to new boundary conditions (e.g. vegetation and soil removal, channel excavation, bank regrading) created by the clean-up operation and (b) highlight where in the Guadiamar intervention may be required to protect bridges, houses, irrigation systems, etc. from accelerated erosion, siltation or flooding, (c) estimate river discharges required to maintain minimum ecological flows, and (d) develop predictive models of sediment-water interactions and metal speciation.

INTRODUCTION

The Aznalcóllar tailings dam at Boliden Apirsa's Aznalcóllar/Los Frailes Ag-Cu-Pb-Zn mine 45 km west of Seville, Spain, was breached on April 25 1998, flooding approximately 4600 hectares of land along the Agrio and Guadiamar rivers with an estimated five and a half million m^3 of acidic water and 1.3 million m^3 of heavy metal-bearing tailings. Extensive cleanup work by Boliden Apirsa S.L., the Confederación Hidrografica de Guadalquivir and the Consejería de Medio Ambiente was carried out immediately after the spill until January 1999, removing most of the deposited tailings and approximately 4.7 million m^3 of contaminated soils to the Aznalcóllar open pit. To evaluate the long-term fate of sediment-borne metal contaminants, an interdisciplinary geomorphological-geochemical research programme began in December 1998. The purpose of this work is twofold: firstly, to monitor the recovery of the river, especially in terms of the geochemical and physical (erosion, sediment reworking, deposition) controls on the distribution of potentially hazardous elements between sediment and waters, and secondly, to provide baseline field data which will be used in formulating strategies for re-establishing sustainable aquatic and riparian ecosystems that are in dynamic equilibrium with their environment. This paper describes the research programme and its initial results.

PHYSIOGRAPHY, GEOLOGY AND MINING HISTORY OF THE GUADIAMAR CATCHMENT

The Guadiamar catchment (37º30'N 6º20'W) is located in southern Spain to the west of Sevilla (Figure 1). It has a catchment area of 1092 km^2 and average rainfall of c. 500mm. The Río Guadiamar rises 24 km to the north of Aznalcóllar and flows 40 km downstream to its confluence with the Río Agrio (Figure 1). The Río Agrio originally flowed through the site of an open pit at Aznalcóllar, but was diverted through diverted through the construction of a 20 million m^3 upstream dam and a 2 km tunnel (Sassoon, 1998). the construction of a 2 km tunnel (Sassoon, 1998). Downstream of the Agrio/Guadiamar confluence the river flows for another 38 km until it joins the Río Guadalquivir. The upper limit of tidal flows in the Guadiamar is located at Aznalcázar. The upper catchment is underlain by rocks of the Lower Carboniferous Volcano-Sedimentary Complex of the Iberian pyrite belt, and south of Aznalcóllar by Miocene silts and calcerinites overlain by Eocene marl beds (Almodóvar et al., 1998). The Los Frailes mine is located at Aznalcóllar at the eastern end of the Iberian pyrite belt, 45 km northwest of Sevilla. The region has a long history of mining, dating to the Roman period, with the most recent phase of large-scale mining commencing in 1979 by Andaluza de Piritas SA (APIRSA), and eventually Boliden, who acquired APIRSA in 1987. There are several ore deposits at Aznalcóllar with total massive sulphide reserves of up to 130 Mt with average grades 3.6% Zn, 2% Pb, 0.4% Cu and 65 ppm Ag. The Los Frailes ore deposit is the most recently discovered, consisting of c.70 Mt of massive sulphides with average ore grades of 0.35% Cu, 2.21% Pb, 3.87% Zn and 63 ppm Ag (Almodóvar et al., 1998). The tailings dam that was breached in April 1998 was constructed in 1978, and was used for waste materials from orebodies at Aznalcóllar from 1979 to 1997, and from the Los Frailes mine after 1997 (Sassoon, 1998).

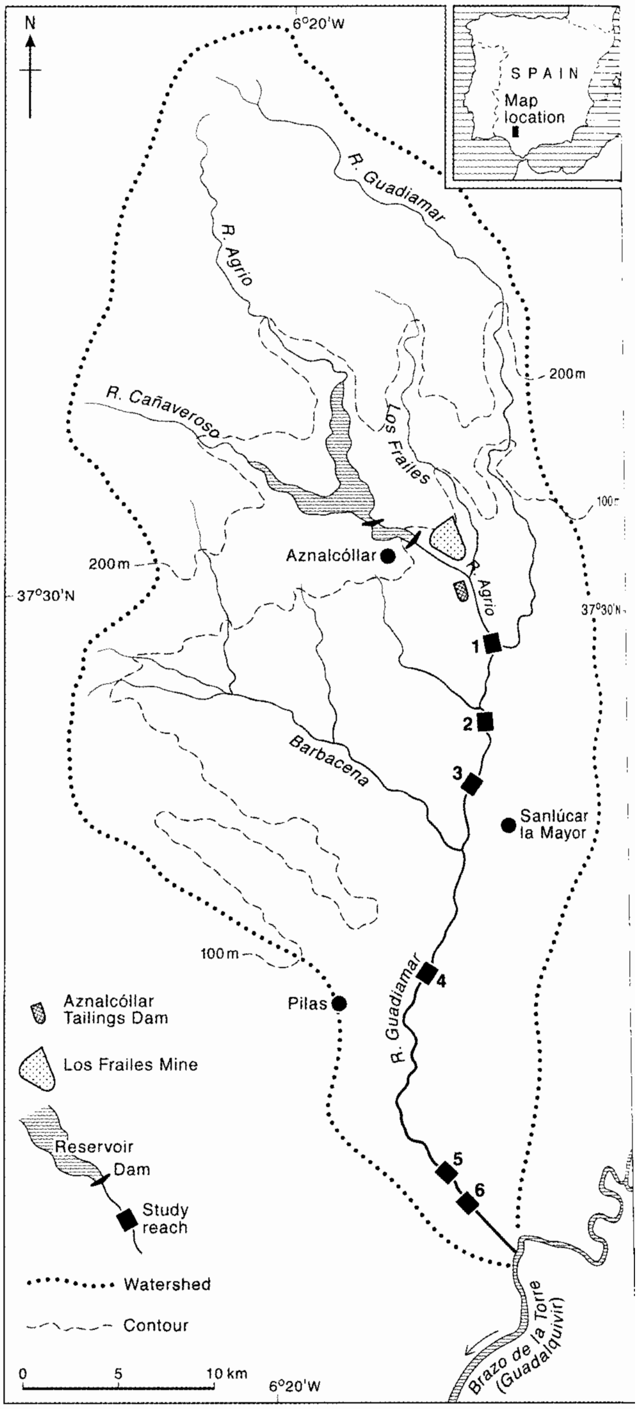

Figure 1. Location of the Guadiamar catchment, showing the locations of the six study reaches

METHODOLOGY

Geomorphology

Reach Number	Location (Lat, Long)	Reach length (m)	Channel type and environment
1	Rio Agrio-Río Guadimar confluence (37º28'N, 6º13'W)	1300	Active wandering gravel bed
2	El Guijo gauging station (37º27'N, 6º13'W)	350	Active low sinuosity gravel bed
3	Upstream of Sanlucar road bridge (37º24'N, 6º14'W)	340	Active low sinuosity gravel/sand bed
4	Upstream of Aznacazar railway bridge (37º18.5'N, 6º15'W)	500	Stable low sinuosity sand/gravel bed
5	La Tiesa (37º13.5'N, 6º14'W)	930	Stable straight silt/sand bed (tidal)
6	Puente San Simone (37º11.5'N, 6º13'W)	640	Stable straight silt/sand bed (tidal)

Table 1. Description and location of study reaches

Detailed 'baseline' geomorphological surveys of the post-cleanup channel, floodplain and valley floor were carried out in January and May 1999 at six reaches representative of the types of river channel and floodplain environments in the Río Guadiamar catchment affected by the spill (Table 1). At each reach, total station survey was used to i) establish a local coordinate system, ii) survey a traverse of fixed control stations, iii) survey detail observations of channel and valley floor topography, and iv) survey monumented valley floor cross profiles.

The grain size of channel bed and bank material was also measured to assess sediment transport rates and the physical stability of the sites. Sediment and water samples (see below) were spatially referenced and integrated within a GIS (ArchInfo).

Sediment Sampling and Geochemistry

Sediment samples were taken from the channel bed and banks, the floodplain and higher river terraces to establish levels of metals in material that is likely to form sources of river sediment. All samples taken were spatially referenced and tied into the geomorphological surveys. Each sample was collected over a 12 m^2 area by combining 10 random sub-samples to c. 200-300 mg from the upper 5 cm of the sediment surface. Several samples of deposited tailings and 'pre-spill' alluvium (alluvium which was clearly older than, and stratigraphically 0.5 to several m below, the post-clean-up, surface alluvium, and was undisturbed by the incident or the clean-up operations), were also taken at all of the reaches.

The sub-samples were then homogenised, and a portion was air-dried, disaggregated and sieved to pass through a 2 mm aperture. A sub-sample of the air-dried material was then ground in an agate tema for four minutes. The samples were digested in $HF\text{-}HClO_4\text{-}HNO_3$ and analysed by ICP-MS (VG Elemental Plasma Quad II+) for Ag, As, Cd, Cu, Pb and Zn at the Institute of Geography and Earth Sciences, University of Wales, Aberystwyth. Analytical precision was determined by inserting blind duplicates to approximately 10 percent of the total number of samples analysed. Analytical accuracy was determined using the reference sediment standards GBW07311 (Office of Reference materials, Laboratory of the Government Chemist. UK). Both precision and accuracy were generally within 10 percent. Selected samples were analysed by X-ray diffraction (Philips PW1710 instrument fitted with a graphite monochromator, with Cu ($K\alpha$) radiation at 40 kV/30 Ma operating conditions) for their general mineralogical composition.

Water Geochemistry

Water sampling was carried out at the same time as the geomorphological surveying and sediment sampling in January and May 1999. Surface water samples were taken from the Río Guadiamar and, in one case, the Río Agrio (Table 3). Samples were also taken from 2 to 10 m diameter pools on the floodplain terraces within one to three days of a rainfall. Water samples were filtered in the field through 0.45 micron filters and those destined for cation analysis were acidified with 2 mL of concentrated nitric acid

At the time of submission of this paper, analytical results were not complete, and interpretation of those presented here must be considered preliminary. Aqueous SO_4 concentrations were determined by ion chromotography, As measured by HPLC-ICP-MS on unacidified, filtered samples, and pH and electrical conductivity (EC) measured in the field. A high level of reproducibility is indicated by the SO_4 concentrations of the field duplicates and was also observed in lab duplicates. Accuracy was determined by measuring SO_4 in standard waters and found to be similar within 0.1 ppm. The As values reported below represent the sum of measured As(V), As(III), the organic forms MMA and DMA. Detection limits were 0.1 to 1 ppb for As (III), DMA and MMA and 0.4 to 4 ppb for As(V). Relative standard deviations ranged from 1 to 30% with an average of 12%.

Future analytical work will include the analysis of all major and trace cations by ICP-MS, including As, Cu, Zn, Cd and Pb.

RESULTS AND DISCUSSION

Geomorphology

Reaches 1, 2, and 3 in the upper and middle parts of the Río Guadiamar have been totally transformed by the mine tailings spill and subsequent cleanup operations. Satellite images and aerial photographs show that, prior to the April 1998 incident, the Río Guadiamar channel upstream of the Sanlúcar bridge was tree-lined and the adjacent floodplain supported a relatively dense shrub woodland. Much of this was removed with the mine tailings along with up to 0.5 m of soil and sediment from the surface of higher river terraces, up to 2 m of material from the top of the floodplain and a considerable but unknown amount of sediment from the bank and bed of the channel. Most of the valley floor away from the river affected by the spill was stripped of vegetation with the exception of some of the largest trees. This has left very large areas of bare, compacted silty-sandy soil on terrace and floodplain surfaces, below which is set a 50-100 m floodway excavated in unconsolidated sandy gravels with a narrow (generally less than 10 m), re-sectioned channel.

Neither the new channel nor the truncated floodplain are in dynamic equilibrium with the runoff or flow regime of the Guadiamar catchment. It is very likely that, in the absence of extensive hard engineering, the next significant flood will result in very widespread channel change upstream of the Sanlúcar bridge including bank erosion, river bed aggradation behind weirs and other obstructions (e.g. bridges) and lateral movement where channels are not confined. Potentially one of the most serious consequences of large scale channel and valley floor erosion will be the remobilisation of sediment-associated heavy metals and arsenic, and their re-introduction back into the Río Guadiamar. Fortunately, the 1998/1999 winter was dry but with enough rainfall to allow vegetation to re-establish itself on floodplain and terrace surfaces, providing some protection from accelerated erosion. Even with a small c. 10 m^3 s^{-1} flood in March 1999 some reworking and adjustment has occurred in reach 1 at the Agrio-Guadiamar confluence. Channel change is largely restricted to the unregulated Río Guadiamar with up to 4 m of bank erosion and channel widening, resulting in deposition of sand and gravel splays, and channel widening downstream. Confluence zones similar to reach 1 where unregulated tributaries join the Guadiamar are likely to be the sites where channel change and adjustment rates are likely to be high, at least in the next few years.

In reaches 4, 5 and 6 downstream of the Sanlúcar bridge, cleanup operations involved the removal of c. 0.5m of tailings and topsoil material with the preservation of the pre-incident river channel and floodplain morphology. The low gradients of channels, and the cohesive nature of river banks, in these reaches make it probable that very little contaminated material will be eroded from the floodplain (or from the bank or bed of the river) provided that these areas are not disturbed by further excavation and dredging. Indeed, the middle and lower reaches of the Guadiamar are likely to be areas where a considerable proportion of contaminated sediment, reworked from the laterally and vertically active parts of the Guadiamar upstream of the Sanlúcar Bridge, will be deposited and enter long-term storage. What is unknown at present, however, is how much, and how quickly, spill material that still remains in the Guadiamar catchment will reach Doñana. This will only become apparent by monitoring sediment and contaminant fluxes over the next 2-5 years. However, by using cellular modelling (Coulthard et al., 1998) we expect to be able to provide forecasts of probable sediment-associated contaminant transfer to the Doñana wetland.

Sediment Geochemistry

A summary of the results for total sediment-associated concentrations of Ag, As, Cd, Cu, Pb and Zn for samples from the four reaches sampled in January 1999 (reaches 1, 3, 4 and 5) is presented in Table 2. Also included are average concentrations of these elements in tailings deposited during the spill, Eocene marl bedrock which underlies the Guadiamar catchment, and pre-spill alluvium. These are compared to data for other catchments elsewhere in the world affected by mining activity.

Zn concentrations in the post-cleanup Guadiamar alluvial samples are generally the highest of the elements analysed, followed by Pb, Cu, As, Cd and Ag (Table 2, Figures 2a and 2b). There is a general downstream decline in average metal and arsenic concentrations at the four reaches. At some of the reaches (e.g. reach 1, Figure 2a), concentrations of channel sediment are generally lower than those of terrace material. Average elemental concentrations in the post-cleanup alluvial sediment are 7 to 11 times lower than those in the spilled tailings material, but typically exceed concentrations in 'background', pre-spill alluvium and Eocene marl by 3 to 13 times, and 5 to 36 times, respectively. Some of the post-cleanup sediment concentrations of Zn are comparable to those reported by van Geen and Chase (1998) for alluvial sediment collected in May 1998, very soon after the tailings dam breach.

Post-cleanup concentrations of Cd, Cu and Zn in Guadiamar alluvium are similar to those found in surface soils from Doñana National Park collected in May 1990 (Ramos et al., 1994). This suggests that the remedial operations have removed enough contaminated material to achieve pre-incident, surface sediment/soil Cd, Cu and Zn levels. Pb concentrations, however, appear to have been elevated considerably. Whether this was directly the result of the 1998 tailings dam failure, or contamination from mining or other sources in the early 1990s is not clear. Ramos et al. (1994) reported that increased mining activity since the early 1980s had resulted in the considerable metal enrichment of the Doñana soils, and that the pollution had increased from 1983 (as reported by González et al., 1990) to 1990, with a 'front' of metal contamination advancing steadily towards the Doñana Park. Although Ramos et al. (1994) attributed elevated sediment and soil metal concentrations mainly to mining activity, they also suggested that effluents from olive oil production along the Río Guadiamar, fertilizers from extensive rice cultivation bordering the northeastern portion of the park, and discharges from untreated sewage from neighbouring villages, may have made significant contributions to the total metal load. The relative contributions of these sources to the alluvial sediment need to be more fully evaluated.

Sample/ Area	Ag	As	Cd	Cu	Pb	Zn
Post-cleanup (January 1999) Guadiamar alluvium						
Reach 1 (n=29)	0.4-7.4 (2.8)	48-540 (300)	0.5-12 (4.4)	81-490 (290)	130-2500 (1000)	410-2600 (1200)
Reach 3 (n=12)	0.2-6.0 (1.8)	36-1000 (220)	0.6-9.3 (4.1)	110-730 (310)	120-2200 (610)	270-3200 (1200)
Reach 4 (n=12)	0.5-4.5 (2.4)	46-690 (290)	0.1-5.6 (2.7)	58-370 (190)	71-1600 (820)	270-1700 (920)
Reach 5 (n=9)	0.6-2.2 (1.1)	30-180 (100)	0.7-6.9 (2.8)	45-300 (160)	75-750 (320)	190-1800 (720)
Pre-spill Guadiamar alluvium, Ecocene marl and tailings						
Pre-spill alluvium (n=8)	0.3-1.2 (0.7)	13-59 (41)	0.1-0.8 (0.3)	21-220 (96)	47-530 (190)	70-150 (110)
Eocene marl (n=3)	0.4-0.6 (0.5)	14-24 (20)	0.3 (0.3)	15-18 (17)	10-30 (22)	94-110 (100)
Tailings (n=10)	9.2-35 (26)	1800-3700 (2500)	12-76 (28)	990-3000 (1700)	3700-12000 (8400)	3700-23000 (7600)
Rio Guadiamar 1990 alluvium (n=3) [f]	n.r.	n.r.	4.72-13.1 (9.5)	90.4-1340 (649)	35.4-126 (76)	948-4200 (2690)
Rio Guadiamar May 1998 alluvium (n=6) [g]	n.r.	n.r.	n.r.	n.r.	n.r.	390-12400 (5330)
Doñana Park 1990 soils (n=15) [f]	n.r.	n.r.	0.23-7.1 (2.0)	1.62-46.8 (23.0)	3.15-44.3 (22.9)	5.13-1077 (210)
Alluvium in other mining-affected catchments elsewhere in the world						
Rio Tinto, Spain (n=14) [b]	<9-30 (4.4)	110-750 (370)	n.r.	75-1500 (300)	490-3100 (1800)	<20-1200 (200)
River Nent, UK (n=20) [e]	n.r.	n.r.	2-160	17-230	2800-13000	790-38000
River Swale, UK (n=36) [a]	n.r.	<2-8 (4)	n.r.	10-37 (18)	240-4500 (1700)	140-3000 (640)
River Vistula, Poland (n=3) [c]	n.r.	n.r.	11-46 (23)	n.r.	15-230 (150)	190-751 (434)
Rio Pilcomayo, Bolivia (n=7) [d]	n.r.	62-6800 (1268)	<1-4 (1)	<1-385 (116)	290-3042 (935)	4-22473 (4140)

All values are in mg kg^{-1}. Values in brackets are means. n – number of samples analysed; n.r. – not reported; [a] Hudson-Edwards et al., 1999a, [b] Hudson-Edwards et al., 1999b; [c] Macklin and Klimek, 1992; [d] Macklin et al., 1996; [e] Macklin et al., 1997; [f] Ramos et al., 1994; [g] van Geen and Chase, 1998.

Table 2. Ranges and mean values (in mg kg^{-1}) of sediment-borne heavy metal and arsenic concentrations for sediment samples from the Guadiamar catchment before and after the tailings dam spill, and other mining-affected river basins.

The ranges of post-cleanup Guadiamar alluvial sediment metal and arsenic concentrations are not dissimilar to those in other alluvial river systems elsewhere in the world affected by mining activity (Table 2). Of particular interest is the nearby Río Tinto catchment, c. 30 km to the west of the Río Guadiamar, which also drains massive sulphide mines of the Iberian Pyrite Belt. Alluvium in the Tinto catchment is considerably enriched in Ag, As, Cu, Pb and, to some extent Zn, due to intense mining activity since the mid-nineteenth century (Hudson-Edwards et al., 1999b). In the Río Tinto and many of the catchments listed in Table 2, unlike the Guadiamar little remedial activity has been carried out.

Although many of the post-cleanup Guadiamar alluvial sediments exhibit elevated total metal and arsenic concentrations, these concentrations give little information about their speciation and mobility. Preliminary field evidence and XRD analysis has shown that the tailings are composed mainly of pyrite [FeS_2], which produces secondary hydroxide and sulphate minerals including goethite [$FeO{\cdot}OH$], jarosite [$KFe_3(SO_4)_2(OH)_6$], hydronium jarosite [$Fe_3(SO_4)_2(OH)_5{\cdot}2H_2O$] and melanterite [$FeSO_4{\cdot}7H_2O$]. These minerals are effective scavengers of metals and arsenic, and some, such as melanterite, are highly soluble (Jamieson et al., in press). In the Río Tinto catchment, for example, soluble sulphate minerals such

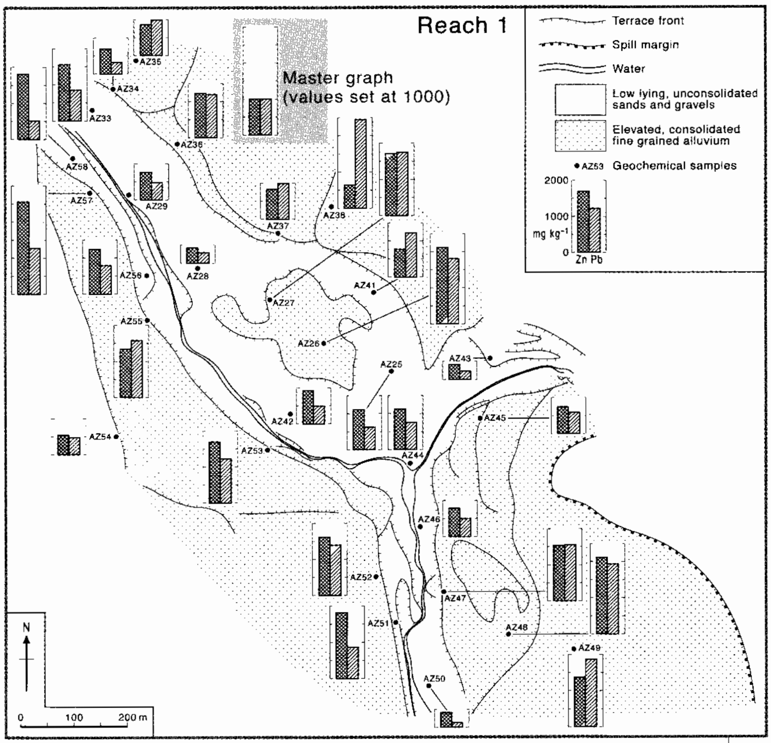

Figure 2a. Geomorphology and Zn-Pb geochemestry of Reach 1.

as copiapite $[Fe_5(SO4)_6(OH)_2 \cdot 20H_2O]$ form on the alluvium and are a temporary store of metals, but are dissolved during periods of high rainfall or flooding, releasing metals to the aqueous system (Alpers et al., 1994; Hudson-Edwards et al., 1999b). Work on the post-cleanup alluvial sediment mineralogy is presently being carried out to determine metal and arsenic potential mobility, and bioavailability, within the Guadiamar catchment.

Water Geochemistry

The relatively low pH values and high concentrations of As and SO_4 in most of the standing waters (Table 3) indicate that these ephemeral pools contain low-quality water that may result from the interaction of meteoric water and residual tailings. The significant difference in As concentration measured in field duplicates W4a and W4b may indicate that a substantial fraction of the As is bound to colloidal Fe hydroxide. The ratio of water to suspended sediment differed in the subsamples and the waters remained turbid even after filtering. Complete trace element data will clarify this. Although these waters are not expected to make a volumetrically significant contribution to river and ground water they pose a local, short-lived environmental hazard. The analysis of sediment samples taken from these pools will provide an opportunity to understand how the interaction of meteoric water with sediment contributes to the chemical evolution of the waters.

Table 3 indicates that, based on data available to date, there is greater variation between the river samples collected in January and May than between the sampling locations. pH is lower and SO_4 is higher in May relative to January for reasons that are not understood at present. The As concentrations are consistently below 50 ppb (US-EPA guideline for drinking water), but some of the SO_4 concentrations exceed 250 ppm, the highest admissible concentration in drinking water advised by the World Health Organization.

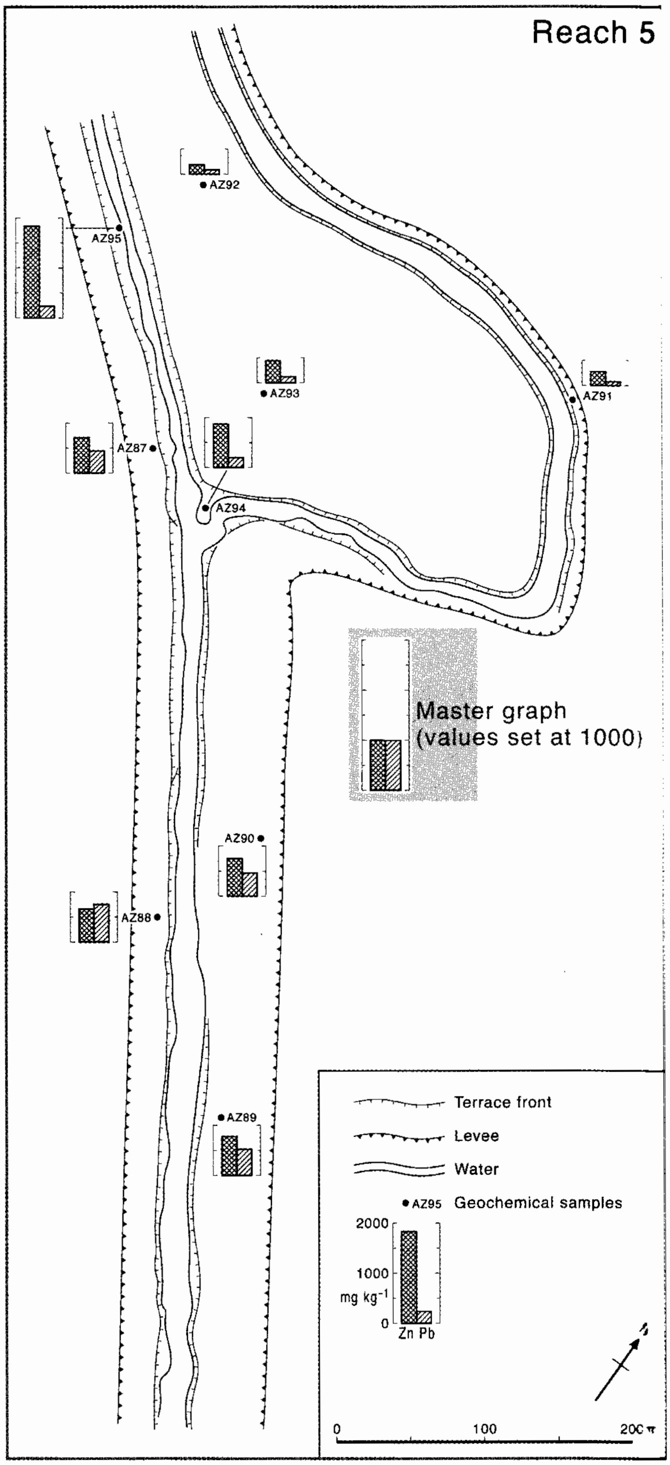

Figure 2b. Geomorphology and Zn-Pb geochemestry of Reach 5.

	pH	EC, mS/cm	Temp (ºC)	SO_4, ppm	As, ppb
REACH 1					
Standing waters					
9901-A-1004	2.3	9.07	15.3	11876	NA
9905-W1	2.3	20.2	29.1	35694	13500
9905-W2	5.9	3.27	26.6	2395	<10
9905-W3	3.4	3.72	25.1	2697	<10
9905-W4a	2.7	4.54	21.3	3359	70
9905-W4b	NA	–	–	3360	140
9905-W5	2.5	5.30	30.1	4496	320
Surface waters					
9901-1003	NA	0.53	10.6	183	NA
9905-R1AGR	4.4	1.49	26.9	833	<10
9905-R1GUA	7.9	0.61	28.3	112	<10
REACH 2					
Surface waters					
9901-1002	7.0	0.74	13.8	299	NA
9905-R1b	6.2	1.79	23.9	1020	10-50
REACH 3					
Surface waters					
9901-1006	7.6	0.62	10.1	212	NA
9901-SMB-1001	6.8	0.72	NA	257	NA
9905-R2	6.1	18.24	21.4	1081	<10
REACH 4					
Surface waters					
9901-1007	7.3	0.79	10.7	271	NA
9905-R3	7.1	1.76	21.2	815	10-50
REACH 5					
9905-R4	7.4	1.81	22.6	457	10-50

Table 3. Preliminary results of water analysis. 9901= sample taken in Jan '99, 9905= sample taken in May '99. W4a and W4b are field duplicates, NA – not analysed.

CONCLUSIONS

In just under 18 months following the Aznalcóllar incident, the mining company Boliden Apirsa S.L., the Confederación Hidrográfica del Guadalquivir and the Consejería de Medio Ambiente working on behalf of the regional government have successfully removed somewhere in excess of 90% of the tailings material deposited in the river channel and valley floor. Although this is a considerable achievement, and our geochemical analyses show that contaminant levels in many parts of the floodplain and river channel are now similar to those before the spill, significant areas of the Guadiamar valley upstream of the Sanlúcar Bridge still have elevated levels of As, Cd, Pb and Zn. The long-term behaviour and fate of these is now the key issue that faces environmental protection agencies responsible for the conservation and management of the Guadiamar catchment and Doñana wetland. It is our view that further removal of contaminated material from the valley floor, especially dredging of river channels, will be highly counterproductive and would result in the large scale remobilisation of contaminant metals and As. Most of the tailings that remain in the Guadiamar channel are likely to be trapped upstream of weirs reconstructed or constructed after the spill. If these sites are left undisturbed it is very probable that contaminated sediment will become buried.

Our work has resulted in the establishment of a baseline geomorphological survey and post-clean-up sediment and water geochemistry. We plan to carry out a monitoring and research programme for at least the next three years to:

a) evaluate the likely pattern of river channel and floodplain adjustment to new boundary conditions (e.g. vegetation and soil removal, channel excavation, bank regrading) created by the clean-up operation;

b) highlight where in the Guadiamar intervention may be required to protect bridges, houses and irrigation systems from accelerated erosion, siltation or flooding;

c) estimate river discharges required to maintain minimum ecological flows; and

d) develop predictive models of sediment-water interactions (c.f., Coulthard et al., 1999) and metal speciation, involving more extensive geochemical and mineralogical analysis on sediment and water samples collected in 1999 and in future years.

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