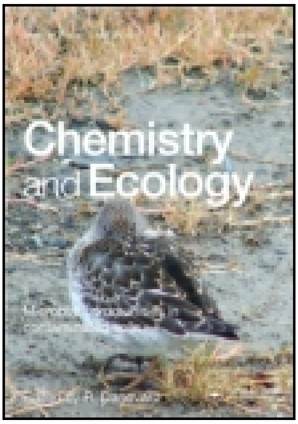

Chemistry and Ecology
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gche20

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To cite this article: D. Petruzzelli, V. Petruzzelli, T. Basile, M. Petruzzelli, A. Petrella & M. Maggiore (2011): Chemical and geochemical characterisation of a disused red brick factory area of central Italy, Chemistry and Ecology, 27:S1, 143-152

To link to this article: http://dx.doi.org/10.1080/02757540.2010.534463

Chemistry and Ecology
Vol. 27, Supplement, February 2011, 143–152

Chemical and geochemical characterisation of a disused red brick factory area of central Italy

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(*Received 22 December 2009; final version received 20 October 2010*)

Characterisation and remedial actions are proposed for a polluted site enclosed within a red brick factory, located in central Italy. Approximately 22,000 t of chromium containing tannery sludge were introduced as 'secondary raw materials' for reuse in red brick manufacture. Preliminary geo-electric tomography was carried out on the waste bank, followed by the perforation of 14 boreholes into the bottom geological stratum. Heavy metals [B, Cr_{tot}, Cr(VI), Hg, Pb, Cu, Sn, V, Zn] and polycyclic aromatic hydrocarbons [benzo(a)pyrene; phenanthrene; dibenzo(a,h)pyrene; pyrene] were determined on solid samples. Carrots were sliced to give samples from different depths in the waste bank. The following samples were determined in the waste leachate: 1,1,2trichlorethane, phenol, 2,4,6trichlorphenol, hexachlorhexane, dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyldichloroethane (DDD) and dichlorodiphenyldichloroethylene (DDE). During characterisation, laboratory experiments were also carried out on lime-treated and untreated carrot sections to evaluate heavy metal release and potential migration in the subsoil. Pollutant exposure pathways were set up, based on application of the Risk-Based Corrective Action (RBCA) model, followed by the evaluation of potential environmental and sanitary targets for site-specific risk evaluation and the proposition of scientifically based restoration scenarios. Surface capping, coupled with lateral belting of the site, were selected for the site recovery operations after extensive cost–benefit evaluations.

Keywords: industrial site clean-up; environmental characterisation; chromium tannery sludge; site-specific risk evaluation

1. Introduction

A polluted site (~1 ha) was enclosed within a red brick factory where, between March and December 1997, ~ 22,000 t (70,000 m^3) of Cr(III)-containing tannery sludge was deposited. The reference material was intended to be reused as a 'secondary raw material' in red brick production. The site was located on the right bank of the Tiber River, in Orte, Viterbo, west–central Italy (Figure 1A), where a thick sequence (10 m) of Pleistocene sedimentary clays is present. The bottom impermeable clay formations included confined layers (lentils) of gravel and sand (Figure 1B) [1].

Field data for the environmental characterisation of the site were collected in compliance with current legislation (Part IV, Title V, D.Lgs.152/2006), in accordance with the corresponding EU references [2].

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ISSN 0275-7540 print/ISSN 1029-0370 online

DOI: 10.1080/02757540.2010.534463
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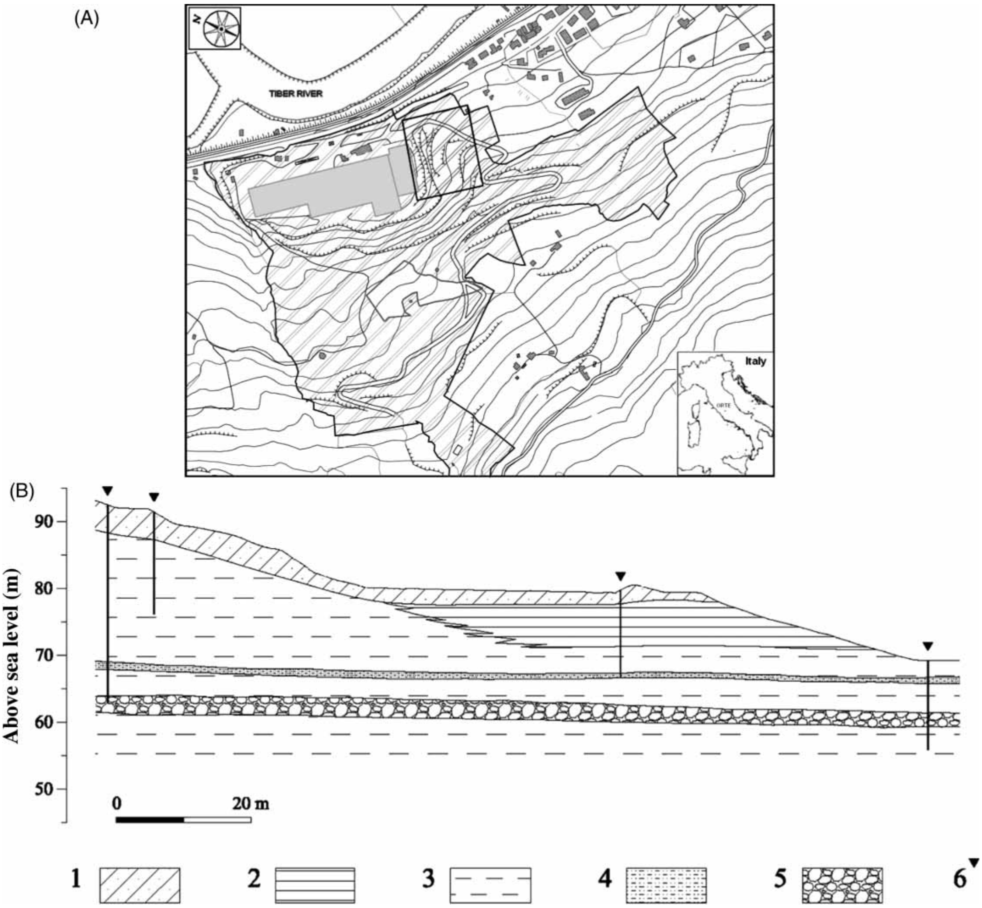


Figure 1. (A) Location of the area, the polluted site is encircled; (B) geological profile across the Orte disused industrial site. (1) Fill, (2) sludge and chromium-containing wastes, (3) Pleistocene clays, (4) interbedded sand, (5) sand/gravel interbedded layers, (6) boreholes.

The main objective of the investigation was to define the most cost-effective clean-up operations for recovery of the site. This was carried out using the Risk-Based Corrective Action (RBCA) model [3] which, after setting up the ‘environmental exposure pathways’ for the chemicals of concern (COCs), allowed us to propose scientifically based remedial actions [4,5].

Preliminary geotechnical and geophysical tests were carried with the aim of monitoring leachate ‘hot spots’ in the waste bank, and general chemical characterisation of the site was carried out by drilling 14 non-destructive boreholes (with carrot recovery), of which 6 were equipped with piezometers for leachate sampling.

To evaluate the potential release of pollutants (heavy metals, organics) from the waste material, leaching tests were carried out on lime-treated (and untreated) carrot sections sampled in the waste bank, to evaluate potential leaching and migration of the pollutants into the adjacent soil and subsoil [6].

A ‘site-specific’ risk analysis was carried out for the site using the RBCA package. Comparative technical–economic evaluations of the proposed remedial actions were carried out before the final decisions [7–9].

2. Experimental

The field investigation was based on the following operative steps: (1) geotechnical investigation of the site; (2) geophysical tests based on geo-electric tomography; (3) carrot drilling in the waste bank to the bottom geological strata; (4) installation of piezometers for leachate sampling; (5) slicing of carrots and sampling for analysis; (6) chemical determinations on solid and liquid samples; (7) leaching tests on lime-treated carrot sections; (8) setting up an environment exposure pathway for the site; (9) risk analysis; (10) proposal of remedial actions; and (11) comparative cost–benefit evaluations.

2.1. *Geophysical testing*

Geophysical tests, based on dipolar geo-electric tomography, were carried out to monitor leachate 'hot spots' in the waste bank. The more electric-conductive areas, in the presence of humidity (e.g. leachate lenses), allowed us to focus on the potential accumulation of more hydrophilic (i.e. water-soluble) pollutants, such as heavy metals and/or polar organics, formed after mineralisation of the constitutive organic matter in the waste material.

Figure 2B shows a typical output from the geo-electric tests: clearer areas correspond to more conductive areas, darker and shaded areas correspond to areas of lower humidity. The geo-electric

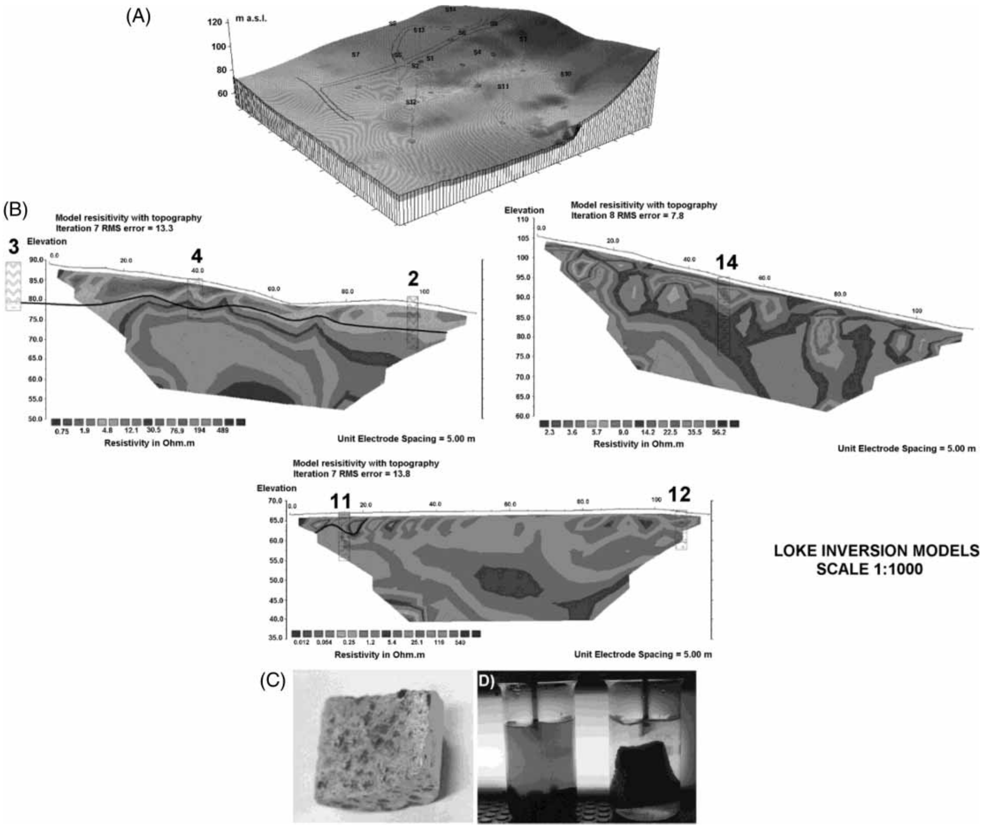


Figure 2. (A) Three-dimensional reproduction of the waste bank with positioning of the boreholes. (B) Geo-electric dipolar tomography of waste bank sections indicating the presence of moisture (dry areas appear darker, wet areas appear lighter). (C) Lime-treated consolidated sample. (D) Corresponding leaching (jar) test.

data served as indicators for geognostic tests, in terms of the extent and depth of drilled boreholes, which were intensified in 'hot spots' where pollutants were more likely to be present.

2.2. *Non-destructive drilling and chemical determinations on carrot samples*

Based on chemical data about the disposed of waste, the geomorphology of the site and geophysical tests, how far the polluted area extended ($\sim$1 ha) was estimated, with an average depth of the waste bank not exceeding 10–15 m (Figure 1B). Based on this, 14 non-destructive boreholes were drilled (6 equipped with piezometers) to an average depth $>$15–20 m, at which point the (almost) impermeable bottom clay layers were reached. Figure 2A shows a three-dimensional reconstruction of the area under investigation with the positioning of the boreholes.

Samples to be used in chemical determinations were withdrawn from carrots at 1, 3, 5, 10, 15 and 20 m below the surface and delivered to the laboratory for analytical determination of the heavy metals [Cr_{tot}, Cr(VI), Hg, Pb, Cu, Sn, V, Zn and B] and polycyclic aromatic hydrocarbons [PAHs; benzo(a)pyrene, phenanthrene, dibenzo(a,h)pyrene, pyrene]. In addition to the above-mentioned parameters, 1,1,2trichloroethane, phenol, 2,4,6trichlorophenol, hexachlorocyclohexane (lindane), DDT, DDD and DDE were also determined in the leachate samples.

2.3. *Leaching tests for heavy metal release*

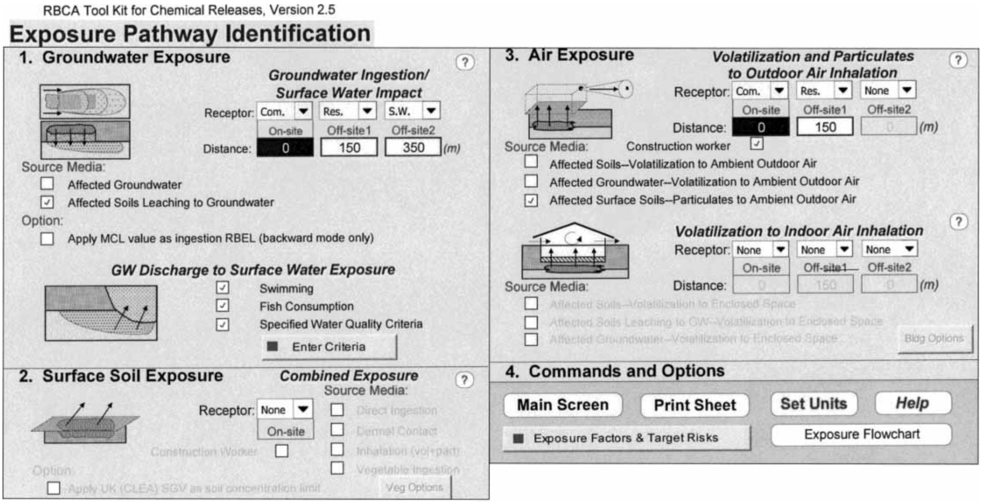
To verify the potential leaching of heavy metals from drilled samples, laboratory tests were carried out. To this end, discrete sections (diameter: 10 cm; height: 5 cm) of sludge carrots were embedded into calcium hydroxide (1 : 1 w/w) (Figure 2C). Lime-treated samples were aged (10 days) and, after consolidation, underwent leaching tests in demineralised water, according to standard EU protocols [10] (Figure 2D). For comparison, corresponding untreated carrot samples (taken from adjacent sections of the same carrot) were submitted to the leaching test, keeping all the experimental conditions constant.

2.4. *The Risk-Based Corrective Actions (RBCA) model*

The RBCA model (Groundwater Software Co., Groton, MA, USA) was applied throughout the investigation [3]. The RBCA is a modelling and risk-characterisation software package for supporting risk-based corrective actions on site clean-up operations. It is designed to evaluate the potential risk associated with the presence of chemicals of concern (COCs) in environmental compartments such air, soil, ground and surface water after man-made activities (i.e. industry, waste management practice). RBCA focuses on protecting human health and the environment, and promotes cost-efficient remedies to address risk. The approach is applicable to chemical release for site-specific evaluations located in different geographical settings and managed by various regulatory authorities [11].

2.5. *Exposure pathways and site-specific risk evaluation*

Sanitary targets were associated directly with humans, whereas the environmental target was associated with the riverbank of the Tiber, as well as with nearby civil infrastructures, i.e. arable soil, wildlife, etc. [12]. Specifically, a site may be considered as polluted, or potentially polluted, when the following are present: (1) a source of contamination; (2) routes for the effective migration of pollutants into the environmental compartments; and (3) targets, e.g. ecologic and/or sanitary targets, 'intercepted' by pollutants [13–15]. Figure 3 shows the exposure pathways for the system



RBCA Tool Kit for Chemical Releases, Version 2.5

Exposure Pathway Identification

1. Groundwater Exposure

Groundwater Ingestion/ Surface Water Impact

	On-site	Off-site1	Off-site2	
Receptor:	Com.	Res.	S.W.	
Distance:	0	150	350	(m)

Source Media:

☐ Affected Groundwater

☑ Affected Soils Leaching to Groundwater

Option:

☐ Apply MCL value as ingestion RBEL (backward mode only)

GW Discharge to Surface Water Exposure

☑ Swimming

☑ Fish Consumption

☑ Specified Water Quality Criteria

2. Surface Soil Exposure

Combined Exposure

Receptor: None (On-site)

Construction Worker ☐

Source Media:

☐ Direct Ingestion

☐ Dermal Contact

☐ Inhalation (vol+part)

☐ Vegetable Ingestion

Option:

☐ Apply UK (CLEA) SGV as soil concentration limit

3. Air Exposure

Volatilization and Particulates to Outdoor Air Inhalation

	On-site	Off-site1	Off-site2	
Receptor:	Com.	Res.	None	
Distance:	0	150	0	(m)

Source Media:

Construction worker ☑

☐ Affected Soils--Volatilization to Ambient Outdoor Air

☐ Affected Groundwater--Volatilization to Ambient Outdoor Air

☑ Affected Surface Soils--Particulates to Ambient Outdoor Air

Volatilization to Indoor Air Inhalation

	On-site	Off-site1	Off-site2	
Receptor:	None	None	None	
Distance:	0	150	0	(m)

Source Media:

☐ Affected Soils--Volatilization to Enclosed Space

☐ Affected Soils Leaching to GW--Volatilization to Enclosed Space

☐ Affected Groundwater--Volatilization to Enclosed Space

4. Commands and Options

Figure 3. Exposure pathway identification flowchart.

at hand, from which it can be seen that the involved environmental compartments are groundwater and air exposure. Two off-site compliance points were taken at 150 and 350 m along the groundwater flow toward the riverbank. Groundwater discharge into surface water would have an effect on recreational uses such as swimming and other sporting activities.

3. Results and discussion

3.1. *Chemical characterisation of the waste bank*

Table 1 reports the chemical composition of solid samples (carrots) and Table 2 shows the corresponding data for the leachate in the waste bank. The data confirm the essential presence of tannery sludge in the waste material.

Figure 4 shows the three-dimensional distribution of chemicals (e.g. Hg, Pb, Cr_{tot}) at different depths in the waste bank, whereas Figure 5 shows the distribution Al, B, Cr_{tot} and Cr(VI) in the leachate lenses. From a comprehensive analysis of the chemical data, relevant information on pollutant distribution in the waste bank was obtained, thus allowing for its rationalisation in terms of ‘environmental exposure pathways’, a flowchart for which is given in Figure 6 .

3.2. *Flowchart of the environmental and sanitary exposure pathway for the site*

By considering the fate and transport phenomena of single pollutants in the involved environmental compartments (air, water, soil/subsoil), the following main factors were considered to be responsible for the migration of pollutants out of the site: (1) surface run-off of rainwater onto the waste bank, followed by leaching through the permeable materials; and (2) migration of underground pollutants to reach the Tiber riverbank, 500 m from the site boundary. Air dispersion of volatile compounds, although very active during disposal operations, was disregarded because of: (1) sludge mixing with clays during disposal operations; and (2) an assumption of quantitative mineralisation of the waste organic matter. Considering the local hydrogeology, partial permeability of the underground environment was reasonably assumed, after the presence of interconnected sand/gravel lenses in the otherwise compact Pleistocene clays underneath the

Table 1. Average analytical data for heavy metals present in the sludge at different depths.

Parameter (mg · kg^{-1})	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13	S14	MAC for residential areas (mg · kg^{-1})	MAC for commercial/ industrial areas (mg · kg^{-1})
Boron (B)	12.43	22.1	46.13	11.3	16.63	19.45	19.07	20.87	16.65	8.17	23.27	15.7	9.87	10.23	150	800
Chrome tot	40.10	69.60	573.57	30.95	46.77	55.95	46.3	35.33	53.78	12.23	24.30	15.60	33.13	32.65	150	800
Chrome VI	0.19	0.29	2.20	0.18	0.13	0.13	0.38	0.35	0.35	0.39	0.46	0.13	0.13	0.27	2	15
Mercury (Hg)	0.13	0.11	0.22	0.11	0.11	0.14	0.13	0.1	0.12	0.10	0.10	0.10	0.11	0.17	1	5
Lead (Pb)	6.45	15.63	21.53	9.8	7	8.85	8.47	13.63	8.33	1.83	9.23	9.1	8.97	11.2	100	1000
Copper (Cu)	17.7	32.23	16.8	18.8	21.67	24.2	21.93	21.3	21.05	11.47	22.17	15.03	18.17	17.15	120	600
Tin (Sn)	0.11	0.11	0.30	0.11	0.26	0.11	0.20	0.16	0.21	0.10	0.18	0.19	0.20	0.12	1	350
Vanadium (V)	21.55	61.97	15.47	17.55	26.67	39.38	26.2	20.02	33.8	13.4	25.83	8.77	18.3	26.80	90	250
Zinc (Zn)	48.18	73	1481.53	90.45	58.77	64.58	42.27	62.88	66.05	18.57	47.07	24.43	25.07	46.48	150	1500
Benzo(a)pyrene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–	<0.1	<0.1	–	–	–	0.1	10
Phenanthrene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–	<0.1	<0.1	–	–	–	0.5	10
Dibenzo (a,e)pyrene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–	<0.1	<0.1	–	–	–	0.1	10
Pyrene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	–	–	<0.1	<0.1	–	–	–	5	50

Notes: S1–S14, drilled boreholes; MAC, maximum allowable concentration.

Table 2. Heavy metals present in the leachate.

Parameter	Unit	S3	S4	S6	S7	S8	S11	MAC ($\mathrm{mg \cdot L^{-1}}$)
Aluminium	$\mathrm{mg \cdot L^{-1}}$	1.20	1.00	0.013	0.016	0.016	0.016	0.20
Boron	$\mathrm{mg \cdot L^{-1}}$	0.55	0.41	0.33	0.69	0.48	0.22	1.00
Cadmium	$\mathrm{mg \cdot L^{-1}}$	0.0091	0.0148	0.0096	0.0089	0.0098	0.00	0.005
Chrome tot	$\mathrm{mg \cdot L^{-1}}$	0.07	0.03	0.019	0.022	0.033	0.026	0.05
Chrome VI	$\mathrm{mg \cdot L^{-1}}$	<0.01	<0.01	<0.01	0.015	0.032	<0.01	0.005
Mercury	$\mathrm{mg \cdot L^{-1}}$	<0.0001	<0.0001	<0.0001	0.00007	0.000016	<0.0001	0.001
Lead	$\mathrm{mg \cdot L^{-1}}$	0.01	0.01	0.02	0.186	0.025	0.02	0.01
Copper	$\mathrm{mg \cdot L^{-1}}$	0.0139	0.0126	0.0078	0.0077	0.0142	0.0084	1.00
Manganese	$\mathrm{mg \cdot L^{-1}}$	0.04	0.02	0.01	0.01	0.01	0.01	0.05
Nickel	$\mathrm{mg \cdot L^{-1}}$	1.30	0.91	0.07	0.327	0.124	0.17	0.02
Zinc	$\mathrm{mg \cdot L^{-1}}$	0.12	0.07	0.04	0.05	0.05	0.06	3.00
COD	$\mathrm{mg \cdot L^{-1}}$	5380.20	871.90	55.80	52.30	64.50	44.30	–
PAHs								($\mathrm{\mu g \cdot L^{-1}}$)
1,1,2-trichloroethane	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0002
2,4,6-trichlorophenol	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.005
2-chlorophenol	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.18
2,4-dichlorophenol	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.11
Pentachlorophenol	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0005
Lindane	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0001
DDT	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0001
DDD	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0001
DDE	$\mathrm{\mu g \cdot L^{-1}}$	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	0.0001

Notes: S3–S11, piezometers; MAC, maximum allowable concentration.

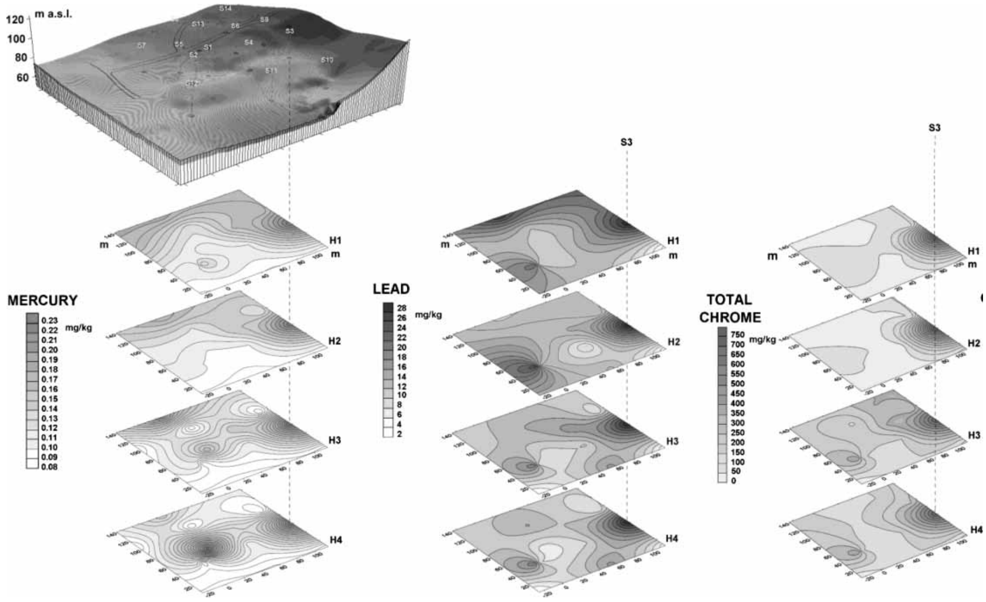


Figure 4. Three-dimensional isoconcentration profiles for Hg, Pb and Cr_{tot} species at different deepness in the waste bank (higher concentrations in darker areas).

waste bank (Figure 2B). Although scarcely permeable, such lenses may represent a preferential route of migration for pollutants toward the Tiber riverbank. This latter situation was assumed to be the most important environmental impact at the site, and was taken into consideration during the restoration project.

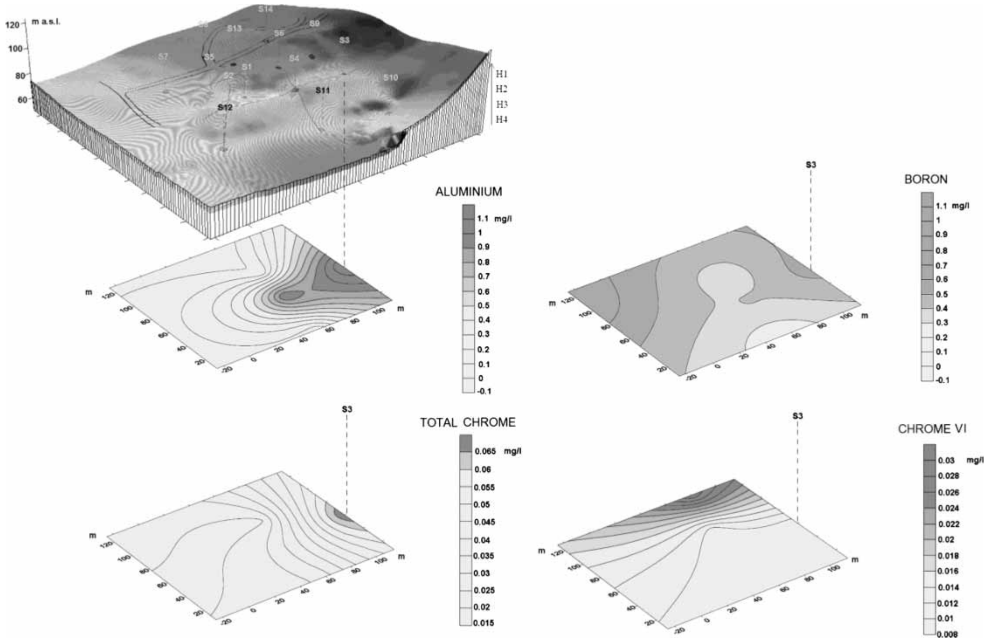


Figure 5. Three-dimensional isoconcentration profiles for Al, B, Cr_{tot} and Cr(VI) species in the leachate (excess concentrations with respect to maximum allowable concentrations are indicated as darker areas).

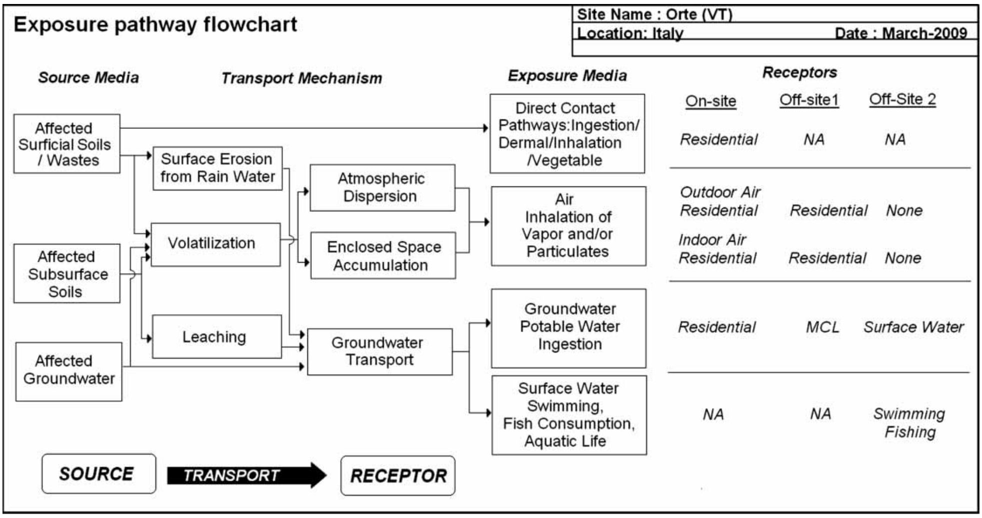


Figure 6. Exposure pathways flowchart for the site.

3.3. *Leaching tests*

Table 3 shows the leaching test data for the lime-treated (consolidated) and untreated samples. From the data, heavy metal ‘immobilisation’ by the lime treatment can clearly be seen. On this basis, it is likely that metals are precipitated in the form of insoluble oxide/hydroxide, thus minimising their mobility to off-site migration. From the operative point of view, these results led to the adoption of ‘on site’ lime treatment on the waste bank. For this purpose, drilled boreholes

Table 3. Analytical data referring to leaching tests carried-out on lime treated untreated samples withdrawn from the same carrot (A–D).

Carrot	Metal	Untreated ($mg \cdot L^{-1}$)	Lime treated ($mg \cdot L^{-1}$)
A	Cr(III)	2.70	0.1
	Cr(VI)	0.02	0.02
	Cd	0.04	0.015
	Cu	1.90	0.25
	Ni	3.30	0.10
	Pb	0.50	0.15
	Zn	5.20	0.50
B	Cr(III)	3.20	0.10
	Cr(VI)	0.60	0.55
	Cd	0.05	0.01
	Cu	2.1	0.20
	Ni	4.70	0.10
	Pb	0.30	0.10
	Zn	3.10	0.50
C	Cr(III)	0.9	0.20
	Cr(VI)	0.01	0.01
	Cd	0.02	0.01
	Cu	1.01	0.20
	Ni	1.50	0.20
	Pb	0.10	0.30
	Zn	2.20	0.40
D	Cr(III)	0.50	0.10
	Cr(VI)	0.02	0.01
	Cd	0.01	0.01
	Cu	0.80	0.10
	Ni	2.12	0.10
	Pb	0.21	0.15
	Zn	3.15	0.51

were filled with hydrated calcium hydroxide to minimise heavy metal migration to the adjacent soil/water compartments.

3.4. *Remedial actions*

The criteria for remedial actions were based on the following factors: (1) final destination of the site; (2) residual pollutant concentrations; and (3) technical/economic (cost–benefit) evaluations.

For the case at hand, the following remedial actions were identified as potentially applicable: (1) removal and transfer of the waste bank to a nearby controlled landfill site (the closest installation was located 500 km from the site); (2) reuse of tannery sludge in red brick manufacture, with qualitative limitations on the final product; and (3) on-site confinement of the waste bank through the installation of cement/bentonite liners followed by surface capping.

Based on a comparative cost–benefit evaluation, this latter solution gave the best compromise in terms of feasibility, reliability and economy. Among the key points pointing toward the proposed action was the avoidance of waste transfer (road/train/ship) to nearby controlled landfills, with the associated environmental and sanitary impacts.

Specifically, surface capping of the site was carried out using bentonite clay liners, coupled with lateral belting with cement/bentonite mixtures, both compacted at a porosity $<10^{-7}\,cm \cdot s^{-1}$, in compliance with current EU legislation [1,4,16]. The bottom of the site was assumed to be reasonably impermeable, although the mentioned sand/gravel lenses might represent a potential risk. This latter point was properly managed by the installation of deeper belts extending well beyond the bottom of the waste bank (>15 m), thus reaching the deeper impermeable clay layers.

4. Conclusions

After a comprehensive chemical, physico-chemical and geophysical characterisation of a polluted site filled with Cr(III) from the tannery industry, an environmental and sanitary exposure pathway of the area was set up, thus allowing for the best targeting of the potential risks. On these bases, scientifically based remedial actions were selected and, after a comparative cost–benefit evaluation, the most efficient solutions from both a technical and an economic point of view were proposed. The final solution was based on surface capping and lateral belting of the waste bank using compacted ($<10^{-7}$ cm $\cdot$ s^{-1}) bentonite clays and cement/bentonite mixtures.

Acknowledgements

Centro Laterizi Nazionale S.p.A., Lucera, Foggia, Italy, is kindly acknowledged for the kind cooperation and allowance for publication of the above field data.

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