Session LeS F.1. Mining and related cases

Environmental Risk Management of Mining Sites with Diffuse Pollution

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Summary

A complex methodology is introduced to manage the environmental risk of a former lead-zinc sulphide ore mine in Hungary: site assessment, source and pollution mapping, risk assessment, determination of target risk, differentiation between point and diffuse sources and selection of the adequate remediation methodologies.

Introduction

The mining industry in Hungary resulted waste deposits, dumps of various size, quality and distribution. The risk of these former mining sites could be assessed and reduced by an adequate combination of management tools. Differences in the management of point and diffuse pollution sources is demonstrated on a typical river catchment area of $15 \times 2 \text{ km}^2$ at the village Gyöngyösoroszi, in the Toka valley, Mátra Hills, North of Hungary (Horváth and Gruiz, 1996; Horváth et al, 1997; Gruiz, 2000)



Figure 1: Hungary and the Toka valley

The Toka catchment area was divided into sub areas according to the typical pollution transport pathways. The Northern part of the catchment with steep slopes (highest altitude: 750 m) is called the "water zone" due to the dominance of transport by surface water flows and runoffs. The area close to the village is the "sediment zone", where the main process, the sedimentation of the transported solid material in the riverbed and in the reservoirs takes place. The Southern part of the catchment is a typical flooding area, called by us the "soil zone", given that the dominant risk in this area is posed by flooding of the hobby gardens and disposal of the contaminated sediment.

Mining activity in the former lead zinc sulphide ore mine in Gyöngyösoroszi has been suspended for 20 years and final closure of the facility has not been done yet. At present, the final mine closure is being planned and remediation scenarios are evaluated. The designed complex management system is introduced in this paper.

The Toka-valley risk management system

The complex management system is shown by the scheme in Figure 2. Subtasks have partly been fulfilled and are shown in this presentation.

The primary pollution sources of the former lead-zinc-sulphide ore mine in the Toka valley are: acidmine discharge, mine wastes, flotation tailings, and lime-precipitate from the acid mine water treatment. Contaminated surface and subsurface waters, sediments and soils are considered as secondary sources from the point of view of the final receptors: ecosystem and humans.



METHODOLOGY IN GYÖNGYÖSOROSZI

Figure 2. Scheme of the integrated risk-based management system

The main pollutants: As, Cd, Pb and Zn, are transported by surface runoff and subsurface runoff partly in dissolved (ionic) form in water, or in acidic water, partly bound to the solid matrix. The runoff delivers only part of the emitted pollutants into the surface water body (both in dissolved and solid forms), the rest is getting into the soil by sorption or mixing. The model used for the whole Toka valley is shown in Figure 3., and the detailed model of a typical pollution source, a mine waste dump, in Figure 4.



Figure 3. Conceptual model of the "water zone" of Toka valley

Figure 3. shows the concept of the transport model: Toka catchment area is considered as a box with inputs and outputs of water and pollutants. Main pollution sources are mine waste dumps, which emit toxic metals and sulphuric acid. Incoming water derives from precipitation and a few springs. Part of the incoming water evaporates, other part infiltrates into deeper soil layers or supplies the Pannonian layers of the plain area (middle part of Hungary), the rest reaches the creek. Metal content of infiltrating contaminated water pollutes soil and subsurface waters.



Figure 4. Transport routes from waste deposits

Figure 4. shows a typical situation in the water zone: point and diffuse mine waste-dumps close to the surface water system. Fate and transport of the pollutants are shown on the scheme: weathering of the ore and rock, percolation and bioleaching of the pollutant, infiltration, partition in the soil and in the surface water system, sedimentation in the water and flooding.

After having the conceptual model of the catchment two further tasks were fulfilled in parallel: assessment of all point and diffuse sources and of the pollutant concentration (metal content) of the surface soil and preparation of the GIS based flow accumulation model of the water catchment.

Pollution mapping

Pollution mapping was based on *in situ* measurements and laboratory analysis of samples collected from the area. In addition, reliable historical data from our database have also been used. The extent of soil pollution around all identified pollution sources was mapped on site by the portable NITON XRF device. The pollution data were evaluated and visualised by GIS based mapping.



Figure 5. Pollution maps of the Toka valley: Zn map and the distribution of As, Pb and Zn

Preliminary Qualitative Risk was assessed based on a site and problem specific questionnaire, resulting a score (point numbers) enabling us to classify the identified smaller and larger pollution sources according the following categories: 1. point pollution sources, to be removed, 2. diffuse pollution sources, to remediate, 3. pollution sources not needing further intervention, but revegetation.

Pollution source	Risk score	Tons	Comments
Tailings flotation dam, flotation tailings	99	4 000 000	isolation
Industrial reservoir, sediment	93	70 000	to remove
Ore transportation route, ore	92	30 000	to remove
Precipitate storage, lime precipitate	90,8	50 000	to remove
Agricultural reservoir, sediment	88,8	30 000	to remove
Mud retention, mixed sediment	85,5	30 000	to remove
Altáró waste dump, mine waste	84,5	1 100 000	remediation
Károly waste dump, mine waste	81,5	16 000	to remove
Gyöngyös-Rédei reservoir, sediment	81,3	30 000	to remove
Toka creek, sediment		35 000	to remove
Új Károly-gallery, mine waste I.	79,5	8 000	to remove
Új Károly-galery, mine waste II.	79,5	800	to remove
Emergency dam, various wastes	78,3	3 000	removed
Péter-Pál shaft, mine waste	75,8	16 100	to remove
Katalin gallery, mine waste	73,5	5 000	to remove
14 different waste dumps	55–70	10 000	remediation
15 different waste dumps	>50	10 000	revegetation

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Legend: mine waste, sediment, lime precipitate, various wastes diffuse pollution for remediation, diffuse pollution

The qualitative risk assessment provides a relative list of pollution sources, therefore the size of the real risk cannot be assessed from it, but only the relative order of the pollution sources. The score limits were set by experts and the scores resulted from the survey could be amended according to technological or economical reasons. The score based survey resulted 15 pollution sources to be subject to remediation and 15 more, which need revegetation.

Water Balance: quantification of the water transport of the catchment

To quantify the pollution transport by water flow, the complete water balance of the Toka-valley was prepared, taking into account the transport pathways and the processes. The annual precipitation is distributed as follows: surface flow and runoff, evaporation, infiltration, plant incorporation, leaching, erosion. The main input is the precipitation (springs are negligible), main outlet is the upper Toka creek and the mine outflow, which two come together downstream the mine. The Water Balance is the quantitative projection of the Pollution Transport Model, and we follow the calculations in details.

The average annual precipitation typical for the Mátra mountains based on 20 years monthly average (OMSZ 1982–2002) is 756 mm/year, measured by the Hungarian National Meteorological Department. The precipitation gauge measurements at the Gyöngyösoroszi site done by ourselves during the last two years gave the following yearly precipitation amounts: 792 mm in 2002 and 519 mm in 2003. According to the 20 years average (OMSZ) the yearly precipitation on the 10 km² = 10 000 000 m² area is 7 560 000 m³/year/10 km² = **20 712 m³/day/10 km²**.

Toka valley is a typical infiltration area: the andesite rock under the subsurface soil is cracked and fissured. Part of the precipitation is stored as soil moisture, other infiltrates into deeper layers through the cracks of the rock. The ratio of these two types of water depends on the intensity or the rain.

Infiltration into the upper soil layer: Water in soil down to 0.65 m depth is in the form of moisture/pore water: The water retained yearly in $3.25 \times 10^6 \text{m}^3$ volume soil ($10 \times 10^6 \text{m}^2$ area down to 0.65 m depth) was estimated to be 1 300 000 m³/year = **3 562 m³/day** (17 % of the annual precipitation). Permeability of the waste dump material is one of the most important factors of infiltration, its permeability is good medium, thus it is higher than **5***10⁶ m/sec.

Infiltration into the deeper layers: Based on several years average mine water flowrate and 26 years precipitation data, (VITUKI, 1987) 26% of the average precipitation fallen on the area got infiltrated into deeper layers. Thus, 26% of the 20 years average yearly precipitation on the 10 km² area would be 5 385 m³/day/10 km² of which approx. 2 000 m³/day is discharged via the main gallery (Altáró) as acid mine drainage. This is treated by lime at the water treatment facility. The rest of approx. 3 385 m³/day supplies the subsurface waters in the middle of Hungary as confirmed by geologists.

Subsurface runoff: The Water Balance sheet for the water zone will include **20%** infiltrated subsurface runoff from the annual precipitation (20 712 m³/day/10 km²), which is **4 142 m³/day/10 km²** and which amount of water does not follow the regular route of the infiltrated water or of the runoff flow but disappears suddenly in the cavities of the mountain.

The average surface runoff is collected by the Toka creek (645 m³/day/10 km²). As an additional amount the runoff produced by heavy rain events (medium and extreme) is estimated as follows: we assumed yearly two medium size rainfall events and one extremely heavy event every 5 years (recurrence valid for the last 15 years). According to our measurements and to the available documents extreme rainfall event means a rainfall over 100 mm and a medium size event means 60–70 mm rainfall. The yearly runoff is estimated from the two medium size events and 1/5 of the extreme one. The flow rate and the amount of the runoff have been calculated from the amount of the rain in mm: Medium size event: rain: 68 mm, for 3 hours. Assuming 14 mm (20%) infiltration, but neglecting other

transport routes, the runoff amount is 54 mm, which is **7%** of the yearly average precipitation. Extremely heavy event: rain: 105 mm, for 3 hours. Supposing 14 mm (14%) infiltration, but neglecting other transport routes, the runoff is 91 mm, which is **12%** of the yearly average precipitation.

Water flow and erosion: Current Water Balance sheet does not consider transport by erosion. Water incorporation by plants: This water volume is part of the produced biomass during the vegetation period and it is in addition to the evaporated amount. The plants' water uptake was estimated based on the yearly biomass production of decituous forests (35 kg/m²/year) and their water content (80 %). The water amount taken up by vegetation (forests) per day in the area of 10 km² was estimated to be 767 m³/day (3.7% of annual precipitation).

Evaporation: according to 24 years (1978–1991) data from the National Meteorological Station in the Mátra Kékestető (OMSZ) the average annual evaporation is 57.58 mm, which is **7.6%** of the average annual average precipitation amount (756 mm). Given that the Northern catchment of Toka the rain events are frequent and the forest area and vegetation cover is almost uniformly covering it we increased the evaporation percentage to 10%. Thus, the evaporation on the 10 km² area from the daily precipitation (20 712m³/day) would be **2 071 m³/day**.

Toka flow rate: the water flowrate has been determined by measuring the flowrate downstream to the outlet of the studied area (water zone). According to our measurements the flowrate relevant to the 12 hours maximum water level is 2.8*645 m³/day=1806 m³/day. The average flowrate of the Toka river in our model is made up of the average daily flowrate (**645** m³/day) and the maximum flowrate (maximum water level in the river) for 12 hours (1806 m³/day) (based on field observations), giving in total **2 451** m³/day. A summary of the above detailed Water Balance is presented below in Table 2.

INCOMING	Denomination	% of total	Relevant in-	Water form	Processes	Data source
WATER	of the incom-	incoming	coming water	and compo-	involved	
	ing water	water	m³/day/10 km2	nents		
	Precipitation	100%	20 712	Rain, snow		Hydrological
						Meteorological
						data
	Infiltrated	43.31%	8 972			Hydrological
	water					data
	N	17.00%	3 562	pore water,	Infiltration into	Microcosm test
				soil mois-	upper soil	<i>In situ</i> meas-
				ture	layer	urements
					- Partition	
	<u> </u>		34		- Plant uptake	
		0,18%	39 m°/	Contamina	Infiltration into	Microcosm
			day/68 506 m ⁻	ted acidic	mine waste	tests
		\backslash		leacheate	- bioleaching	In silu meas-
			E 20E	Cubaurfaaa	Infiltration into	urements
		20.00%	5 365	Subsurface	Inilitration into	
				water	deeper layers	uala
			2 000	Acidic mine	Mine outflow	Hydrological
			2 000	water		data
			3 385	Drinking	Pannonian	Hydrological
		▼	0.000	water	lavers	data
	Subsurface	15.68.00%	3 248	Runoff	Underground	Observation
	runoff			water	brooks	Hydrological
						data
	Surface	15.65 %	3 241	Runoff wa-	- Runoff	Modelling run-
	runoff			ter	- Partition	off: measure-
						ments
						and hydrologi-
						cal data
				K _d		
	Erosion			Transfer 1 11		
					- Partititon	Nodelling ero-
				and sedi-	- Bioleaching	SION: GRASS
	Water in	2 70 %	767	Dient weter	- Plant uptake	"Atlag Egglagy"
	> water in	3.70 %	101	Plant water		Allas Ecology
	510111055				incorporation	
	Vapour	10 00 %	2 071	Evano-	Evano-	Meteorological
	- Tapour	10.00 /0	2.071	transpiration	transpiration	data
						5010
OU FLOW	Outflow	11.83 %	2 451		Surface flow	Flow model and
				Toka	201000 11017	measurements
from the						
WATER ZONE				creek		
20112		100.17%	20 718			

Table 2. Water Balance: quantification of the water transport of the catchment

Meteorological data: OMSZ, National Meteorological Service, 2002; Hydrological data: Terramed Bt. Risk Reduction Plan, 1996 Heinrich, D. and M. Hergt: Atlas Ecology, Springer,, Budapest, Berlin, 1995

For our Water Balance the **acid drain water volume** produced during bioleaching was calculated from the yearly precipitation (756 liter). Given that from the 756 liter/m² annual precipitation 26% is infiltrated into deeper layers, 10% is evaporated, 3.7% is taken up by vegetation, 17% is preserved as soil mois-

ture, 15,68% is infiltrated subsurface runoff resulting hidden temporary creeks and assuming that the rest is surface run off that turns into sulphuric acid solution produced during bioleaching, the amount of acid drain water is **208.8 liter/m²/year**.

Total surface area of the waste dumps (point sources) in the Northern catchment of Toka creek is 44 506 m² its volume is: 653 752 m³, and the relevant mine waste tonnage is 1 634 380 tonnes. According to our estimation the tonnage of diffuse pollution sources in the area is 20 000 tonnes, resulting 12 000 m³ volume of material and 24 000 m² surface area.

Total sulphuric acid solution produced/year by bioleaching on the total area of the waste dump (point sources) is 44 506 m²*208,8 liter/m²/year = $9,29*10^6$ liter/year = $9,292 \text{ m}^3$ /year = 25.45 m^3 /day The total sulphuric acid solution produced by bioleaching on the total area of point and diffuse pollution sources will be 68 506 m²*208.8 liter/m²/year = $14.3*10^6$ liter/year = 14300 m^3 /year = 39 m^3 /day.

Flow accumulation



Figure 6. Flow accumulation in the Toka water catchment and location of the pollution sources

Pollutant transport: emission from the point and diffuse sources

Pollution Transport by water was quantified by the quantitative water balance and by the GIS based Flow Accumulation Model of the catchment, which allows for reading of the surface area and watershed of every waste dump and contaminated sub-area. Table 3. gives the surface area of the point and diffuse sources and their watershed area read from the GIS Flow Accumulation Model.

Waste dump	Surface	Watershed	Cell size	Surface area of the waste
	area	of the dump		dump watershed
	m ²	cell number	m ²	m²
Total point sources	197132	8 228	100	822 800
Toka total waste dumps	44 506	4 109	100	410 900
Diffuse sources	24 000	2 000	100	200 000
Residue from point + diffuse	68 000	6 220	100	622 000
Total Toka water catchment		250 000	100	25 000 000

On the basis of the leaching microcosm experiments the contaminant concentration of the runoff originating from direct precipitation and indirect water flow through the waste dumps was calculated. This type of pollution mapping and the Flow Accumulation Model makes possible the estimation of the effect of the removal of certain waste dumps and pollution sources or the reduction of their emission, substantiating environmental risk based decision making.

The total pollutant emission is calculated from the water ratio that runs off the polluted areas according to the Water Balance and the maximally leachable metal content measured in the leaching tests. The emission from the pollution sources calculated by the model gives a realistic view on the emitted metal amount and these data can be used for direct decision making in spite of the fact that the natural risk reduction potential (e.g. filtering effect of the soil) will not be taken into account, therefore, the pollution load of the surface water is highly overestimated.

Table 4. Metal concentration of the leacheate from the microcosm test

Leached metal	g/m ³
As	0,741
Cd	1,20
Cu	4,71
Pb	3,58
Zn	163,53

Leaching of metals from pollution sources and the characteristic parameters of the process were set based on a complex (physical-chemical-biological) leaching test (microcosm). using the most polluted mine waste material in the area. As a pessimistic approach, all pollution sources were considered to have the same pollution emission as the most toxic one. To estimate the metal amount transported by the runoff the metal concentration of the leacheate from the microcosm experiment was used.

The calculated total metal amount emitted from the identified pollution sources during the first year resulted from the following data:

- \Box Total surface area of the mine waste dumps plus diffuse minus the flotation tailings dump: 68 000 m²
- □ Total tonnage of the mine waste plus diffuse material: 1 654 380 tonnes
- Direct flow (rain) on the total surface of the waste dumps and diffuse sources: 22 168 m³/year, calculated from the GIS model.

□ Indirect flow through the total surface of the mine waste dumps and diffuse sources: 202 772 m^3 /y. The total calculated metal load (kg/year) transported in the 1st year by the direct + indirect rain running off and through the total surface of the mine waste dumps and diffuse sources is:

As: 91 kg; Cd: 148 kg; Cu: 556 kg; Pb: 442 kg; Zn: 20 203 kg.

The emission and consequently the water transported metal load will decrease in time exponentially. This aspect was not taken into account in our approach. The calculated emitted metal amount was used as a quantitative parameter to characterise the risk of the existing point and diffuse pollution sources.

Natural risk reduction of the catchment area

The calculated emitted metal amount was the basis for determining the contribution to the total risk of each point and diffuse pollution source. The calculated emission was used as a semi-quantitative relative risk value for differentiation between the sources. If we would like to estimate the pollutant flow along one of the pathways into the Toka creek, we have to take into account the risk reducing capacity of the site (dilutions, sorption in the soil, partition between phases, changes in the chemical form and as a consequence changes in the mobility of the metals, bioaccumulation, etc.) (Gruiz, 2002).

The risk reduction efficiency of the site was calculated by comparing the calculated emitted metal content (based on the results of the microcosm experiments) with the actual metal concentration measured in the surface water. The total metal emission of the pollution sources in the total water catchment compared to the final measured metal load of the outflow, represented by the Toka creek, gives the natural risk reduction efficiency (NRRE) of the Toka-valley "box". The NRRE of the Toka box = emitted metal concentration/measured metal concentration in the outflow of the box.

	Table 5. Calculation of the Misk Neduction Enclency of the Toka-box										
Waste concent	dumps emi tration (esti microcosn	tted lea mate ba n test)	acheate ased on	Toka creek (measured concentration)			Risk Reduction Efficiency (NRRE) of the Toka box				
As	Zn	Cd	Pb	As	Zn	Cd	Pb	As	Zn	Cd	Pb
µg/lit	µg/lit	µg/lit	µg/lit	µg/lit	µg/lit	µg/lit	µg/lit				
741	163 530	1200	3580	96	813	2	118	8	201	600	30

Table 5. Calculation of the Risk Reduction Efficiency of the Toka-box

This tool is suitable 1. to calculate future concentration in the Toka creek after having reduced the emission (removal of point sources and reduction of emission from the residual sources) or 2. to plan the removal of the sources, if target concentration in the Toka creek is known (see below). The target concentration in the Toka creek was set by experts based on site-specific environmental guality criteria taking into account also the different effect based guality criteria in various countries.

	-		, ,	
Effect based environmental quality	As	Zn	Cd	Pb
criteria for surface water	µg/lit	µg/lit	µg/lit	µg/lit
HU standard for subsurface water	25	200	0.5	10
Holland	8,6	6	0.35	10
Canada	50	30	0,01–0,06	1–7
US-EPA	190	110	1.1	3.2
Swedish	0,45–9	4,5–9	0,045–0,09	0,6–1.2
Danish	4	86–110	2,5	5,6–9,2
UK	50	8–50	-	4–20

Table 6. EBQC: Effect Based Quality Criteri

Knowing the target EBQC of the Toka creek (set by experts) and the effect of the risk reduction box (calculated), the targeted emission from the diffuse and residual sources in the Toka catchment could be determined. This value should ensured by the remediation procedure.

EBQC of Toka creek				Efficiency of the risk reduction box			Targeted emission from the dif- fuse and residual sources							
As	Zn	Cd	Pb	pН	As	Zn	Cd	Pb	pН	As	Zn	Cd	Pb	pН
µg/lit	µg/lit	µg/lit	µg/lit						incr.	µg/lit	µg/lit	µg/lit	µg/lit	
3.0	5.0	0.03	1.0	7,5	8	201	600	30	2 units	24	1000	18	30	5,5

Table 7. Targeted emission from the Toka-catchment after remediation

Site remediation

As demonstrated previously, the target emission after remediation of the diffuse sources is determined by the surface water quality and the risk reduction potential of the site. Remediation is conducted on the basis of the results of the laboratory and field remediation experiments.

Risk reduction consists of excavation and confinement of point sources and chemical + phytostabilisation of diffuse sources and of the residual waste after removal of point sources (Lelie et al, 2001). The main objective in case of Gyöngyösoroszi is to reduce the pollution flux from the pollution sources, which involves mitigation of the leaching process and stopping erosion of the polluted soil. The choice of the remediation process for diffuse pollution is based on: the toxic metal concentration (Zn, Pb, As), the toxic metal (Zn, Pb, As) emission flux by runoff, the access to the pollution source, cost evaluation. According to the Difpolmine approach, if the diffuse source is barely vegetated phytostabilisation combined with chemical stabilisation is performed. Laboratory tests are performed on real soil samples. These tests enable to determine the amendments (immobilising agents like fly ash, iron oxide) to be added to the soil and the plant species able to grow on the substrate. Then, the field plots are implemented on selected sites and the water emission and pollution load remaining after the implementation of the remediation is measured at different locations and at different time periods.

The risk reduction efficiency of the combined chemical and phytostabilisation will be measured by these experiments. A Remediation Risk Reduction Efficiency factor (RRRE) will be created and used for the

design of the remediation. The microcosm experiments on chemical stabilisation demonstrated that 3% fly ash addition to the polluted soil resulted 95% reduction in the water soluble metal content of the soil. The field experiments on combined chemical and phytoremediation are being planned and will start this year. The implementation of this remediation alternative will reduce the emission and the pollution load in the surface waters of the area, however the subsurface runoff and mine water will have to be treated separately, by an adequate water treatment methodology (Younger et al., 2002).

Relation to the Difpolmine, Life Environment Demonstration project

Risk management in the Toka-valley is connected to the DIFPOLMINE Project, which focuses on the treatment of Diffuse Pollution from Mining Activities and demonstrates that an integrated and adaptive approach using runoff control (Pottecher et al., 2002) and phytostabilisation (Vangronsveld et al., 1995), reduces the pollution transfer to surface waters on former mine sites.

Gyöngyösoroszi in Hungary is a demonstration site for a risk management methodology, which is an adaptation of the French Difpolmine approach worked out in the frame of the EC Life Difpolmine Project (DIFPOLMINE, 2002–2004). The Difpolmine site is in Salsigne, France at a former gold mining area, where the pollution sources are wastes from hydrometallurgy, pyrometallurgy, mine wastes, sludges. The main pollutant is arsenic, that is transported by ground water and/or runoff to the surface water bodies. After removal of the highest risk sources, the Salsigne site undergoes remediation by a combined chemical and phytostabilisation technology.

The two sites are different from the point of view of the transport routes and the transported metals. An other difference is, that in Hungary both point and diffuse pollution sources should be managed, while in Salsigne point sources have already been removed. The mine closure and remediation planning in Gyöngyösoroszi started in 2004, therefore the results of the Difpolmine project can be utilised.

Summary

A complex risk management tool was established for the Toka-valley site utilising the results of the DIFPOLMINE project. GIS based pollution mapping and flow modelling supports the risk management system. The assessed and mapped characteristics and emission of the sub-areas and point sources help to estimate and handle the risky components of the site. The Toka-valley box-model helps to calculate and estimate emission on catchment scale. Leaching Capacity, Natural Risk Reduction Efficiency and Remediation Risk Reduction Efficiency factors were determined by laboratory and field experiments and measurements and were used for planning and risk calculations.

After having completed the differentiation between the point and diffuse pollution sources, the point sources will be excavated and transferred onto the top of the tailings dump. Given that the erosion of the polluted soil leads to the highest toxic metal emission from the pollution source, once the point sources are excavated and confined, the emission of the toxic metal will diminish considerably. The residual pollution will be treated as diffuse pollution together with the already existing diffuse pollution sources, applying the complex approach of chemical and phytostabilisation.

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