Reclamation, Treatment and Utilization of Coal Mining Wastes, edited by A.K.M. Rainbow 267
Elsevier Science Publishers B.V., Amsterdam, 1987 — Printed in The Netherlands

COAL MINE SPOIL TIPS AS A LARGE AREA SOURCE OF WATER CONTAMINATION

J. SZCZEPANSKA [1] and I. TWARDOWSKA [2]

1 University of Mining and Metallurgy, Inst. of Hydrogeology and Engineering Geology, 30-059 Cracow, Al. Mickiewicza 30 / The Poland/

2 Polish Academy of Sciences, Inst. of Environmental Engineering 41-800 Zabrze, M.Skłodowskiej- Curie st.34 / The Poland/

SUMMARY

The scale of negative effects coal mine spoil tips on the water environment in the vicinity of the tip has been exemplified on the result of long-term investigations of the active tip Smolnica situated in the area of the Upper Silesian Coal Basin / Poland /.
The problem have been presented on the basis investigations of surface and underground water quality in the vicinity of the tip against the background of dynamics of soluble solids leaching from coal mine spoil deposited on the tip and chemical composition of pore solutions in vertical cross - section of the tip. The results of chemical analyses of pore solutions and effluents from the tip as well as investigations of underground and surface water have proved the negative changes of water quality in consequance of tip impact.

INTRODUCTION

Coal mine spoils have a negative effect upon the aquatic environment in the vicinity of the tips in consequence of the soluble substances leaching. The problem of water contamination as a result of coal mine spoil deposition is at moment importance, as these spoils are the biggest by quantity group of industrial solid wastes in Poland. About 60 mln m^3 of spoil are produced annualy from underground coal mines in the Upper Silesian Coal Basin / USCB / in Poland.

The deposition of so great quantitys of spoils on the limited area and frequently on a permeable bedrock, will cause the increasing threat of water environment contamination by the soluble compounds leached out from the spoil. Among these substances, the particularly negative effect is caused by the chloride leaching as well as by the oxidation of iron sulfide minerals and the subsequent hydrolisis of the reaction products to form acidic,

high sulphate and high iron and heavy metal drainages.
Taking into consideration the annual production of coal mine spoil in the USCB and the mean sulphate production estimated at the rate of 20,8 g/tonne of spoil/day the total annual sulphate production in the spoil deposited within the tips has been found to amount some 740 000 tonnes SO_4/ year.
The annual chloride load deposited on the tips has been estimated to the be about 65 000 tonnes/year.
Also the heavy metals which present in spoil in trace concentrations, are the essential factor of water contamination in the vicinity of tips.
The scale of the water contamination caused by coal mine spoil tips has been exemplified on the result of long - term investigations of water quality in the vicinity of the Smolnica-tip /USCB, Poland /, against the background of dynamics of soluble compounds leaching from coal mine spoils deposited on the tip.

MATERIAL AND METHODS

Object of investigations

The active spoil tip Smolnica covers at present some 60 hectares and has situated in the valley of the Bierawka -river / Fig.1/.
The construction of the tip proceeds towards the river bed according to the direction of the surface run off and ground water flow. In the foundation of the tip occur the permeable sand.

Spoils have been deposited since 1965 in two layers, each about 10 m thick. They have been discharged from the carboniferous strata of group 300 and 400, those being in Westphalien A and the upper Namurian C series. The spoils consists mainly of claystones / 79,5 %/, mostly of kaolinite - illite - sericite type.

The permeability to water of the spoil is generally high and ranges from $2.47 \cdot 10^{-6}$ m/s to $1.28 \cdot 10^{-3}$ m/s depending on the position in the cross-section of the tip.

Chlorides in freshly produced spoils are present in mean concentration 0,05% Cl^-. The mean total sulphur content is 0,90 % S_t. The sulphur is distributed as iron sulphide /0,81% S_p/ and in small amount as organic sulphur and in the form of sulphates / 0,009 % S_{SO_4} /.

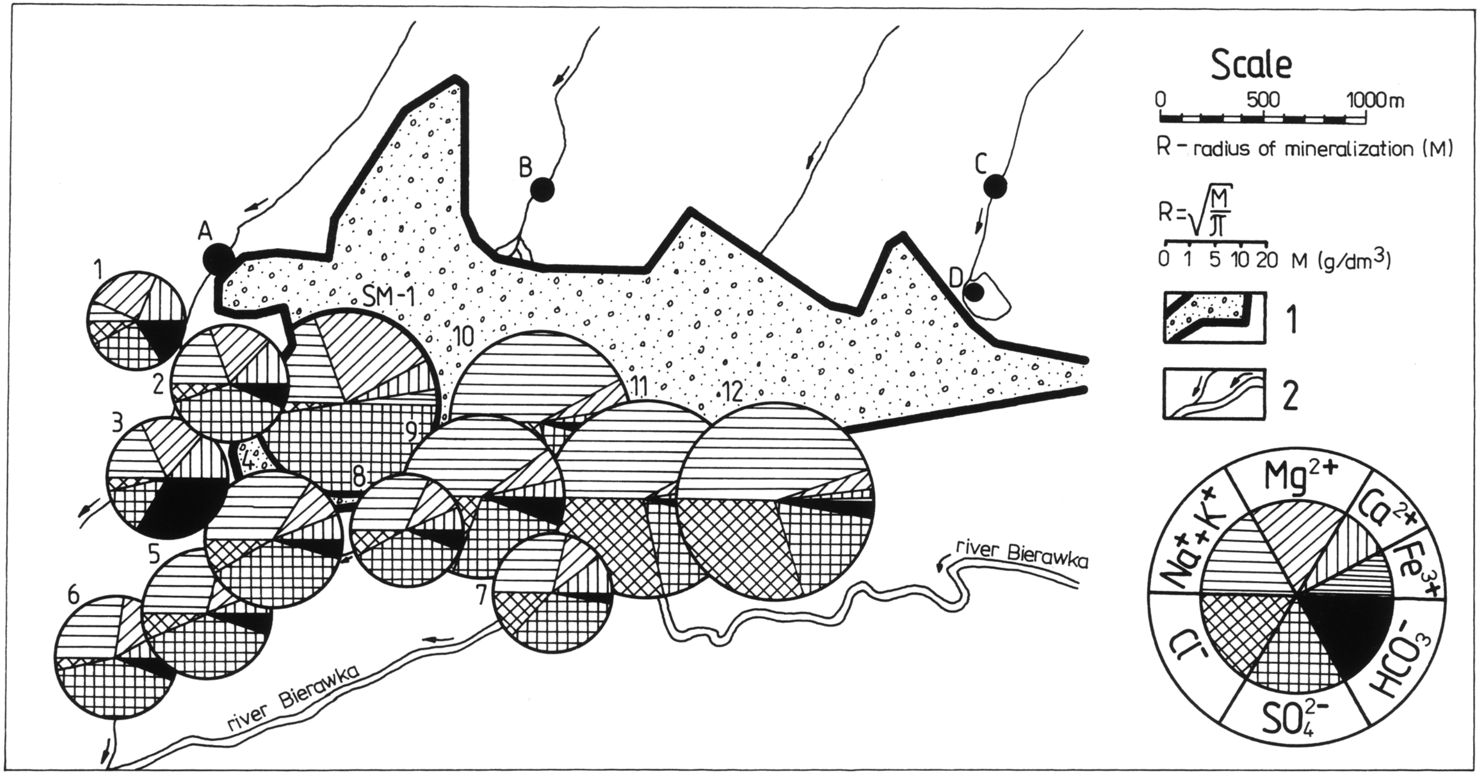

Fig.1.Hydrochemical map of the investigated area.Smolnica coal mine spoil tip-Upper Silesian Coal Basin,Poland. 1-Smolnica tip,2 - river and streams.Sites of waters sampling: A,B,C,D - streams /N/ above the tip; 1,2,3 - ponds /W/ below the tip; 4,5,6,7 - streams /S/ below the tip; 8,9,10,11,12-effluents from the tip; SM-1 - prospecting - shaft /pore solutions/.

The sulphide reactivity in the aeration zone of the tip has been described by the constant of sulphate production r_s=0,210 meq/eq G_s . day, where G_s – the pyrytic sulphur content in the spoils /eq/t/.

The spoils are low-buffered /$\xi_{buf.}$ =0,71/,so they are predisposited to turn acidic. The carbonate minerals are mainly sideroplesite and minor quantities of calcite and dolomite. Trace concentrations of the heavy metals: zinc, copper and lead are also present.

Sampling and investigation methods

Dynamics of soluble components leaching from coal mine spoils has been observed in situ as a function of the age of spoils. The spoils in the outer, 100 cm thick layer of this tip of various age since the moment of deposition / 0 – 12 years/ have been sampled. The soluble components have been derived from the spoil by means of the successive 3-time repeated standard water extractions of 1 : 5 / rock: water/.

The chemical composition of the pore solutions in the vertical cross-section of the tip have been analysed too. The samples for investigation of pore solutions were taking from the prospecting shaft / Fig.2/ about 11 m deep. The upper parts of profile up to the depth of 7,5 m contained spoils deposited 10 years ago. The spoils in the bottom part were 15 years old.
Spoil of natural moisture content have been sampled every 0,5 m. From these samples the pore solutions have been extracted using pressure method / ref.1 / and concentrations of major ions have been determined.

The changes in hydrogeochemical conditions in the vicinity of the tip have been observed by comparing the chemical composition of the adjacent natural waters flowing towards the tip with the water quality below the tip; drainages ditches, effluents from the tip, ponds, streams / Fig.1/. The chemical composition of the water samples have been analysed by standard methods.

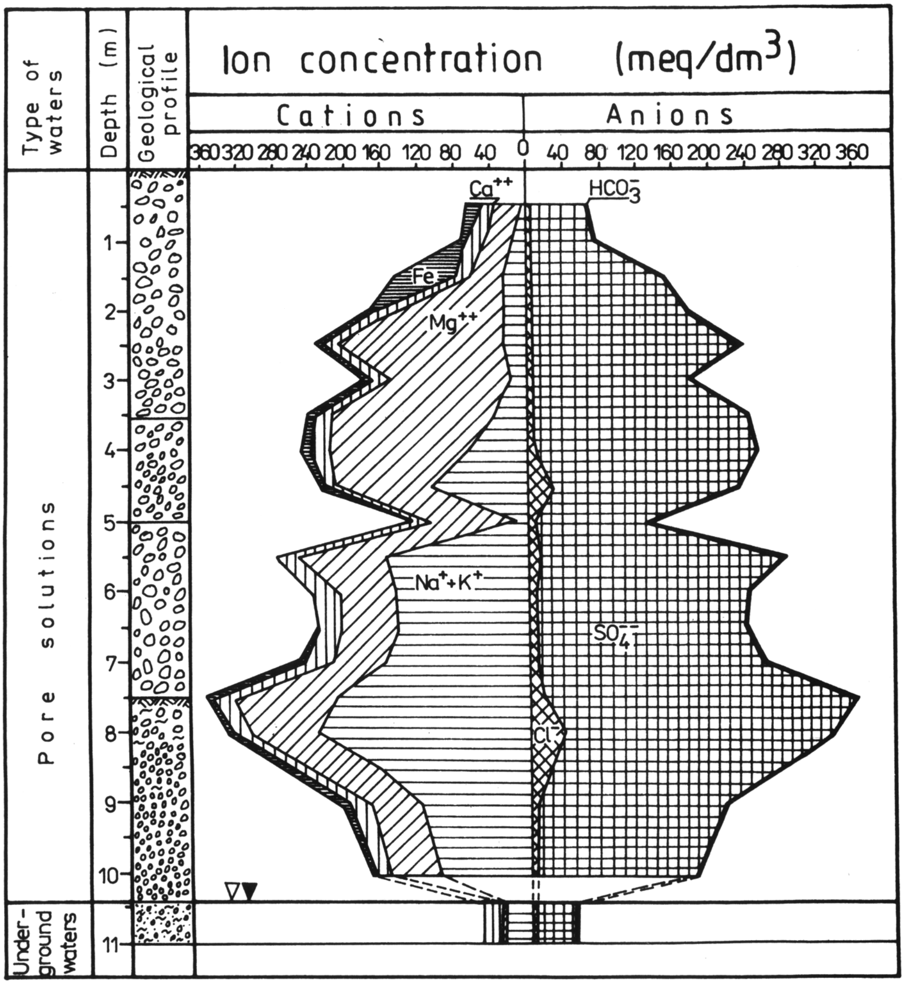

Fig.2. Hydrochemical profile of the pore solutions extracted from the spoil deposited on the Smolnica tip. The prospecting - shaft SM-1.

R E S U L T S

Dynamics of soluble components leaching from the outer layer of the tip.

Spoils of various age / from 0 to 12 years/ sampled from the outer layer of the tip / 0 – 1 m / reveal a very high dynamics of chloride leaching.The rate of chloride removal was found to range 80 % just in the first year after deposition / Table 1/. In the following years chloride concentrations show only negligible changes. High susceptibility of chlorides to leaching off is the result of their occurence in the liquid phase i.e. in pore solution infilling the carboniferous rocks / ref.2 / as well as the consequence of the lack of equillibrium limitations.

The dynamics of sulphate leaching shows the diffrent character.The sulphate concentration in the spoils is a resultant of two processes: the sulphate generation by the action of iron sulphide oxidation and the sulphate leaching by infiltration water.

On the analysed tip during the 5-6 years long period, after spoil deposition, constant increase of sulphate concentration in spoils have been observed / Table 1 /. It indicates the predominance of production over the leaching off process. In the following years the tendency of sulphate concentration decrease in spoil is marked. It evidences a gradual sulphide depletion in the spoils.
At the same time, the spoils, become acidic in reaction just in the first year after deposition / pH 3.3 – 3.7/ in consequence of their low buffering capacity.

The major component of the freshly produced spoils are chlorides / Table 1 /. After a lapse of a year, the dominant anions become sulphates. In the period from 1 to 6 years after spoil deposition, the sulphate concentration in the outer layer of the tip ranges from 160 to almost 500 % of the initial content in the freshly produced spoils.

The results of field investigations of the dynamics of the spoil leaching imply that the risk of contamination of the aquatic environment by chlorides is determined by the annual load of chlorides deposited on the tip together with coal mine spoils.

The hazard of water contamination by sulphates is defined by the total quantity of spoils filed in the aeration zone of the

Table 1

Retention of chloride, sulphate and total dissolved solids in the spoil deposited on the Smolnica tip

Approx. age of tip at sampling point /years/	Depth of sample from surface /cm/	pH	Chloride as Cl /dry weight/		Sulphate as SO_4 /dry weight/		Total dissolved solids TDS /dry weight/	
			g/t	% chloride content of fresh wrought spoil	g/t	% sulphate content of fresh wrought spoil	g/t	% TDS content of fresh wrought spoil
Fresh wrought spoil		7,90	531,1	100,0	281,0	100,0	1462,0	100,0
1 - 2	0 - 20	3,33	70,5	13,3	450,0	160,2	725,8	49,6
	70 - 100	6,06	101,0	19,0	722,9	257,3	1214,5	83,1
3	0 - 20	3,59	70,5	13,3	452,5	165,4	775,6	53,0
4 - 6	0 - 20	3,75	88,2	15,5	658,6	234,3	1004,9	68,7
	70 - 100	3,14	94,0	17,7	1396,3	496,9	2020,0	138,2
10 - 12	0 - 20	3,38	72,8	18,7	278,9	99,1	489,6	33,5
	70 - 100	6,20	86,9	15,3	1143,2	406,8	1700,8	116,3

tip in the condition of a relatively free acces of oxygen.The total surface area of the tip is great importance too.It decides of the quantity of leaching water, and of the size of the outer layer of tips where the conditions for generation of soluble components and their leaching off are the best.

Chemical composition of pore solutions in the aeration zone of the tip.

In the vertical profile of the prospecting-shaft SM-1 /Fig.2/ the distinct differentiation of the chemical composition of pore solutions has been marked. It has been caused of the two-layer construction of the tip body. The highest concentration of sulphates and total dissolved solids in the pore solutions occurs on the contact of the upper 10 years old with the lower 15 years old spoil layer i.e. on the depth of about 7,5 m.
Total dissolved solids concentration is there as high as 24 300 mg/dm^3. Sulphates have been present in concentrations 16 500 mg/dm^3. Pore solutions are of SO_4 -Na-Mg type.Up the profile of the upper layer, the concentrations of dissolved components gradually decreased. Chemical type of pore solutions changes from SO_4- Mg in the layer 5,0-1,5 m to SO_4 - Mg- Fe in the most washed out outer layer 1,5 - 0 m. In this layer also the highest acid potential of spoils and the lowest the buffering capacity has been recorded.

The impact of the different period at spoil deposition has been marked in the lower 15 years old layer of spoils by the decrease of the total dissolved solids to the level of 13 000 mg/dm^3. It has been caused by the partial, substantial washing out of this layer before the deposition of the upper spoil level.On the other hand the chemical type of water SO_4 - Na - Mg remains the same.

In the whole profile a very low chloride concentrations amounting to 70 mg/dm^3 Cl^- have been observed.
The concentration of this ion in the pore solutions extracted from freshly produced spoils have been found to be about 7000 mg/dm^3.Reduction of chloride content which accounts for about 99 % has been the result of the multiple washing out of the spoils in the whole analysed profile and the high dynamics of chloride

leaching.

The observed increase of dissolved components in the vertical profile of the tip is a consequence of the influence of two factors, i.e. a vertical redistribution of salt loads / ref.3 / and a various degree of washing out of the each particular spoil layer in the profile of the tip body. The extent of vertical redistribution of dissolved component loads is limited by the geochemical constrains imposed by equillibrium conditions.Chlorides redistribute the most intensive because of the lack of equillibrium limitations within the observed range of concentrations. The rate of vertical redistribution of sulphates depends on the buffering capacity of spoils contributed by calcium carbonates, which limits a sulphathe solubility to the level of equillibrium with gypsum.

Because of the low buffering capacity of spoils analysed, the equillibrium limitations has a little effect thus the vertical redistribution of sulphate loads is significant.

Besides of the vertical redistribution loads process,the chemical composition of pore waters in the vertical profile of the tip is influenced also by the different degree of washing out of spoil layers in the vertical cross-sections of the tip body, determined by flow rate of infiltration water. For the analysed tip, the period of one-fold washing out including transport of contaminants leached off from the outer layer of the tip by percolating water to the waters beneath the tip toe, lasts some 5 years. In this period the outer layer 1 m thick will be washed out about 20 times. The changes of chemical composition of pore solutions in the vertical profile of the tip have reflected the different age of infiltration waters. In the lower part of profile waters of the chemical composition corresponding to the erlier stages of leaching process are present.It is testified to the occurence of sodium ions. In the upper, outer layer sodium is replaced by iron and hydrogen ions.

Chemical composition of underground and surface waters in the vicinity of tip

The results of analyses of drainages from the spoil tip as well as investigations of the adjacent under ground and surface

Table 2

Chemical composition of pore waters extracted from the spoil deposited on the Smolnica tip and surface and underground waters in the vicinity of this tip

Type of waters	Location of sampling points		pH	Total dissolved solids content TDS /mg/dm^3/	Chloride content Cl^- /mg/dm^3/	Sulphate content SO_4^{2-} /mg/dm^3/
Pore solutions	Prospecting-shaft SM-1 0 - 7,5 m /10-years old spoil/		4,0 - 5,2 5,0	4162,4 - 24287,5 13553,9	84,8 - 931,8 247,6	2829,3 - 16510,8 9559,0
	Prospecting-shaft SM-1 7,5 - 10 m /15-years old spoil/		4,6 - 5,2 5,0	12249,2 - 22854,0 16275,9	129,5 - 1276,2 494,1	8490,6 - 14511,6 10880,9
Under-ground waters	Prospecting-shaft SM-1 /10,43 - 11 m/		7,0 - 7,9 7,5	3610,5 - 3637,4 3624,0	148,4 - 168,2 158,3	1787,3 - 1799,3 1793,3
Surface waters	Fresh waters	Streams /N/ above the tip	7,4	123,6 - 363,3 228,8	23,1 - 34,4 29,2	50,0 - 131,7 100,4
	Polluted waters	Ponds /W/ below the tip	7,6 - 9,3 8,7	3693,3 - 4929,8 4354,1	34,4 - 172,0 124,9	133,9 - 2708,9 1340,2
		Streams /S/ below the tip	5,8 - 7,9 7,1	5755,9 - 8128,2 7230,8	153,7 - 845,1 591,2	3045,1 - 4450,4 4064,9
		Effluents from the tip	7,4 - 8,2 7,7	6457,8 - 19236,8 13919,4	742,0 - 5886,5 3776,8	3856,6 - 6341,2 4866,7

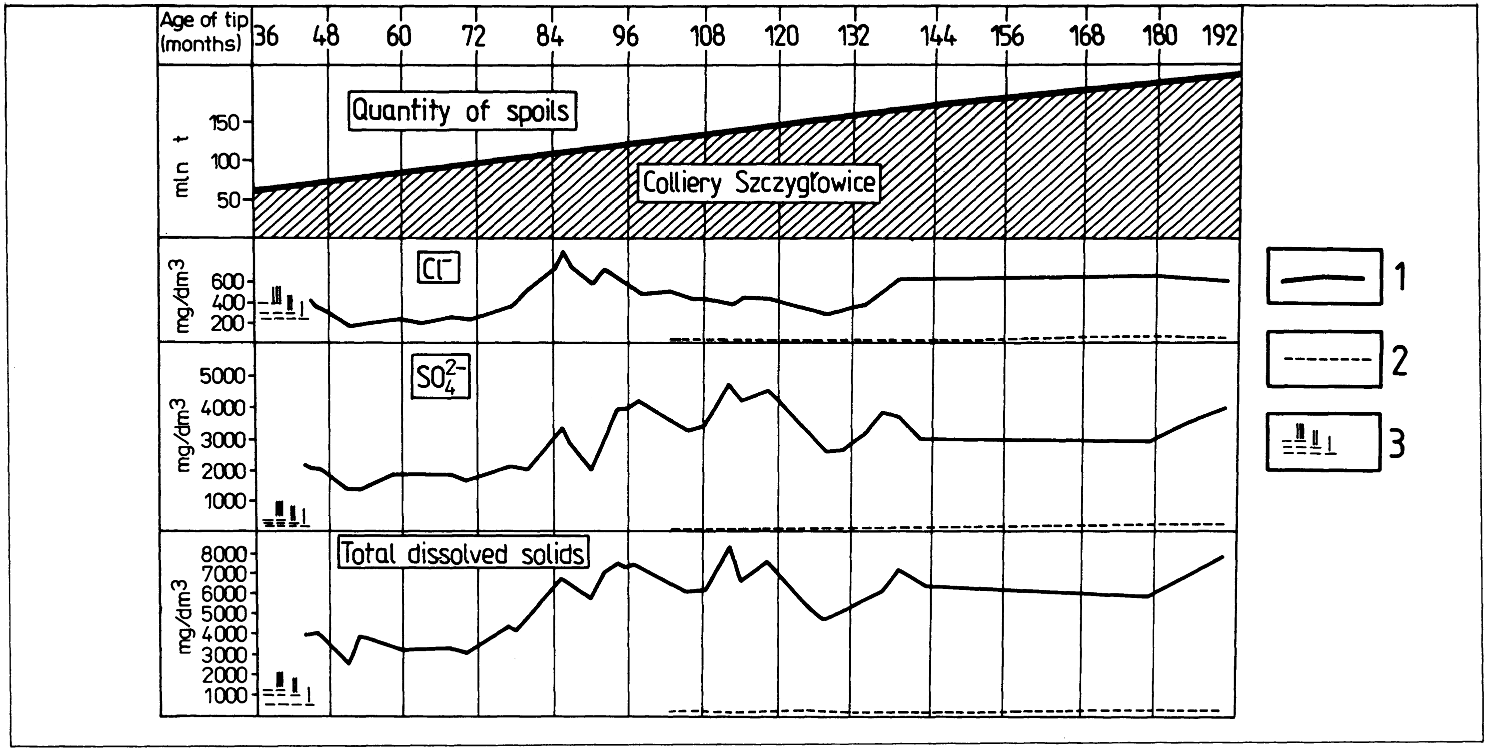

Fig.3. Variability of chloride /Cl^-/, sulphate /SO_4^{2-}/ and total dissolved solids in surface waters caused by coal mine spoil deposition. Smolnica spoil tip. 1 - drainage ditch from the tip /sampling point-4/; 2- stream above the tip / sampling point - A/; 3 - classes of water quality / I,II, III/.

waters have proved the negative changes of water quality in consequence of tip impact. / Table 2, Fig. 1 - 3/.
Total dissolved solids concentrations in waters beneath the tip have increased from 6 to over 14 times, in comparison with those in adjacent natural waters flowing towards the tip on its uphill side.
Sulphate concentrations increase from 11 to more than 33 times, and chloride content from 2,5 to over 25 times.The highest fluctuations of chloride content have been observed in adjacent waters beneath the tip. It corresponds with the degree of washing out the spoil on the particular parts of the tip of different age according the dynamics of chloride leaching.

In the adjacent waters beneath the tip /on its the downhill side/ the distinct increase of heavy metals, mainly copper, zinc and lead have been observed, too. / Table 3 /.

The chemical type of waters changes from HCO_3 - Ca- Mg for natural waters flowing toward the tip to SO_4 - Na or SO_4 - Cl - Na in waters beneath the tip.

Natural waters quality flowing towards the tip fulfill the standards for pot water. Water quality in drainages, watercourses and ponds below the tip exceeds not only the standard limits for pot water, but also the standards for the lowest III quality class for the inland waters.

C O N C L U S I O N

On the basis of the studies and long-term investigations of coal mine spoil tip Smolnica / Upper Silesian Coal Basin / has been concluded:

Coal mine spoil tip are found to be the serious source of contamination of natural underground and surface waters in the vicinity of the tip.

In the studied area the waters, yielding over normative amount of dissolved matters, sulphates, chlorides and heavy metals. The pollution is due to leaching of soluble matters / chloride, sulphate and heavy metal / in the process of percolation of precipitation water through the spoil deposited on the tip.

Table 3

Heavy metals content in the spoil deposited on the Smolnica tip and in the waters in the vicinity of this tip /Approx. age of tip 7 - 16 years/

Parameter	SPOILS /Content - g/t/	W A T E R S /Content - mg/dm³/					
		Fresh waters		Polluted waters /drainage ditch from the tip/			
		Stream /N/ above the tip		Ditch W		Ditch S	
		min	max	min	max	min	max
Chromium - Cr	50	0,00	0,09	0,00	0,10	0,00	0,10
Nickel - Ni	60	0,00	0,78	0,11	0,45	0,11	0,80
Copper - Cu	30	0,00	0,001	0,00	0,99	0,001	1,03
Zinc - Zn	50	0,00	0,11	0,10	2,70	0,30	1,89
Cadmium - Cd	—	0,00	0,001	0,00	0,02	0,00	0,05
Lead - Pb	14	0,00	0,10	0,00	1,07	0,00	0,80
Cobalt - Co	80	0,00	0,25	0,06	0,12	0,05	0,08

On the tip with high permeability of spoil generation and leaching of soluble solids have been observed in the full profile of aeration zone.

The waters percolate through the spoil tip becaming saturated with soluble components. Chemical composition of infiltration waters in the vertical profile in the aeration zone of the tip is a consequence of influence of two factor:
vertical redistribution of salt loads and a various degree of washing out spoil layer in the profile of the tip body determined by rate flow of infiltration water.

According to dynamics of chloride leaching from the spoils the risk of contamination of the water environment by chlorides is determined by the annual load of chlorides deposited on the tip together with coal mine spoil.

The water pollution by sulphates is defined by the total quantity of spoil in the aeration zone of the tip in the conditions of air penetration, to the spoil, which is a factor limiting sulphide oxidation.

R E F E R E N C E S

1. J. Szczepańska, Chemistry of pore waters from marine and continental clays of Tertiary formation, Geologia, t.8, z.3, Z. Nauk. AGH, Kraków, 1982, pp. 105 / in Polish, sum. English/
2. J. Herzig, J. Szczepańska, St. Witczak, I. Twardowska, Chlorides in the Carboniferous rocks of Upper Silesian Coal Basin, Fuel, 0172 / 1986 /.
3. I. Twardowska, Mechanism and dynamics of Carboniferous spoils leaching on the tips, Prace i Studia IPIS - PAN No 25, Wrocław, 1981, pp.206 / in Polish, sum. English /.