



Environmental Impact of Sludge Dumps to the Quality of Agricultural Soils in Region Žiar nad Hronom

Tomáš TÓTH¹⁾, Jozef KULICH²⁾, Miriama KOPERNICKÁ³⁾,
Klaudia HALÁSOVÁ⁴⁾, Lenka LACKÓOVA⁵⁾

¹⁾ Doc. Ing. RNDr.; Slovak University of Agriculture, Faculty of Biotechnology and Food Sciences, Department of Chemistry, Tr. A: Hlinku 2, 949 01 Nitra, Slovak Republic; email: tomas.toth@uniag.sk

²⁾ Prof. Ing.; Slovak University of Agriculture, Agricultural Expert Institute, Tr. A: Hlinku 2, 949 01 Nitra, Slovak Republic; email: jozef.kulich@uniag.sk

³⁾ Mgr.; Slovak University of Agriculture, Faculty of Biotechnology and Food Sciences, Department of Chemistry, Tr. A: Hlinku 2, 949 01 Nitra, Slovak Republic; email: m.kopernicka@gmail.com

⁴⁾ Slovak University of Agriculture, Faculty of Horticulture and Landscape Engineering, Department of Landscape Planning and Ground Design, Tr. A: Hlinku 2, 949 01 Nitra, Slovak Republic

⁵⁾ Ing. Ph. D.; Slovak University of Agriculture, Faculty of Horticulture and Landscape Engineering, Department of Landscape Planning and Ground Design, Tr. A: Hlinku 2, 949 01 Nitra, Slovak Republic; email: lenka.lackoova@uniag.sk

Summary

In connection with the change of aluminum production technology has considerably changed the emission situation nearby source of the production at the processing plant (ZSNP a.s.). It was the reason, why the content of fluorine compounds has dramatically changed in the air and and after then in the soil. Agoregion Žiar nad Hronom has been under negative pressure acting on the agro-ecosystem since 1953, when the operation of the company ZSNP increased from the original 48 thousand tonnes to 70 thousand tonnes by gradual intensification. Production of anode, electrode and ramming masses increased from 50 thousand tonnes to 100 thousand tonnes. Ecological status around the plant after reaching the designed production capacity (aluminium, aluminium oxide, anode mass) was disastrous in the 1960's. State of the ecological production of ZSNP a.s. was the subject of the environmental audits since 1990. In the plant called SLOVAL a.s brought „B“ section to a standstill in 29th February 1996, where was realised the Soderberg's technology and production of aluminium. This technology had been used for 43 years. Significant reduction of emissions was caused by implementation of new technology "HYDRO ALUMINIA". Red and Brown sludge landfill capacity is located on an area of about 45 ha, weighted approximately 8,5 million tonnes. In the framework of the examination of the impact of the dump on the agricultural land, we carried out monitoring of the content of hazardous components in landfill. It was made at a distance of around 200, 300 and 500 metres from the front range of the landfill. Five soil samples were taken from a depth of 0,20 m in each distance (a total of 15 samples of the soil), as well as the sample was taken from the subsoil. Soil samples were analyzed in an accredited laboratory GEL, Turčianske Teplice. From the results of the determination is known that the content of Na is in the range of 78-5,270 mg.kg⁻¹, Al 2,78-5,92 mg.kg⁻¹, Pb 73-244 mg.kg⁻¹, Zn 83-298 mg.kg⁻¹, Cu 55-152 mg.kg⁻¹, Cd 0,8-3,0 mg.kg⁻¹. The total content of fluorine is a 1,965-2,217 mg.kg⁻¹ and with increasing distance from the landfill its content in soil decreases. The same result is applied in the case of the content of water-soluble F in the soil, the content of which is 28.0-46.3 mg.kg⁻¹.

Keywords: red sludge, brown sludge, heavy metals, soil contamination

Introduction

The territory around Alluminium Plant in Žiar nad Hronom has been often mentioned and evaluated in particular with regard to the contamination of soil - especially fluorine, entering the environment from the production of aluminum. It covered the period from 1953, when plant began to produce the first aluminum. Second series of electrolysis was put into processing in 1957 and and it also led to the first disastrous effects of fluorinated air pollutants to the natural surroundings.

The ecological status of the biota in the vicinity of the plant after reaching the specified production capacity (aluminium, aluminium oxide, anode mass) was disastrous in the 1960s. Issued by high-risk pollutants accounted for approximately 23 000 tonnes per year in 1960, in 1970 about 20 000 tonnes per year, 12 500 tonnes in the year 1190 and 4505 tonnes in the year 2000 (Antalová, 1991). State of the ecology of the production ZSNP a. s. was the subject of environmental audits since 1990. In the years 1991 - 1992, the company Agiplan from Austria has carried out an audit

under "Concept to improve the state of the environment of the regions of the upper Nitra (Žiarska kotlina)". The company called Haskoning from Netherlands in 1993 and the company Iwaco in 1994 carried out an ecological audit on behalf of the European Bank for investment development. It was decided to modernise the electrolytic production of aluminum in advance burning anodes and air pollutants generated fluorine by dry absorption. The contract for the modernisation of production of aluminium was signed by Ardal company from Sundal and Verk a.s. This company took off and has teamed up with the company Hydro Aluminium. The amount of pollutants emitted and generated was reduced by 7613 tonnes, representing 62.8% and the volume of goods of primary production remained on a higher level than in 1990.

The emission situation has changed significantly also in the issues, that had not yet entered into force in 1990. It is highlighted as in Table 1.

The results of the transformation of the production technology of aluminium and implementation of greening

Tab. 1 The quantity of pollutants emitted into the atmosphere by the plant ZSNP in 1990 and 1996 [t.year⁻¹]

Tab. 1 Ilość zanieczyszczeń emitowanych do atmosfery przez elektrownie ZSNP w latach 1990 i 1996 [t.rok⁻¹]

Type of emission	Year	
	1990	1996
emissions of fluor	900	60
SO ₂	7000	3100
CO ₂	1700	400
organic carbon	900	50
solid fallout	2000	300

project are very reflected on the reduction of emissions into the air, water and showed the reduction of waste. The development of pollution situation has very positive impact and improve the environment in Žiarska kotlina.

Material and methods

The monitored area surrounding Žiar nad Hronom is located on the middle reaches of the river Hron and subarea medium high valleys of Slovak highlands, specifically in Žiarska kotlina. Žiarska kotlina represents the tectonic depression between Vtáčnik, Kremnické and Štiavnické vrchy. Žiarska kotlina constitutes almost a closed basin, with ideal conditions for unwanted emission dispersion to the agricultural and forestry land, as well as villages. The territory, which is damaged by the immediate effects of harmful substances leaking from ZSNP Žiar nad Hronom

is located in the north narrowed and in the S-SW expanded, so has roughly triangular shape. Air masses containing waste gases mainly SO₂ and HF is evidenced, in particular, to the south, in the municipalities situated in the immediate vicinity of the plant and at the foot of Štiavnické vrchy (Kobza, et al, 2008). Soil forming processes in the conditions of the three basic soil types: luvisol, fluvisol, cambisol (ilimerizate, alluvial, brown).

Žiarska kotlina is quite a large volcanic depression and hydrologically the numerous breaks give a presumption of groundwater and surface water to create water formations to great depths with the possibility of thermal and hyperthermal waters in the permissible depths. Basin falls within the warm climate area, slightly humid zone with mild winters. In terms of the type of climageographical type is a warm - slightly warm climate of depression and slightly warm -



The soil samples collected about 200 metres from the front of sludge (1A, 2A, 3A, 4A, 5A), 300 metres from the front of sludge dump (1B, 2B, 3B, 4B, 5B) and 500 metres on the front of sludge dump (1C, 2C, 3C, 4C, 5C).

PA - soil sample 200 metres from the front of sludge dump - depth sampling 0,8 m

PB - soil sample 300 metres from the front of sludge dump - depth sampling 0,8 m

PC- soil sample 500 metres from the front of sludge dump - depth sampling 0,8 m

Fig. 1 Localization of soil sampling points

Rys. 1 Położenie miejsc pobierania próbek gleby

Tab. 2 Chemical composition of brown and red sludge and its real impact on the soil

Tab. 2 Skład chemiczny brązowego i czerwonego szlamu oraz ich wpływ na glebę

Component	Sludge [mg.kg ⁻¹]	Soil (mg.kg ⁻¹)	
		In the range of alkalization by sludge and ground water	In the range of long-term contamination by emissions
Al	55200	3420-6240	6800-11200
Fe	97880	2346-2914	4900-5200
Mn	524,7	297-314	260-320
Cu	21,5	79,2-83,4	6,5-31,2
Zn	55,9	60,2-81,6	14,1-51,2
Cr	154,9	5-7,5	2,9-14,2
Pb	112,6	81,6-112,4	26,8-62,4
Cd	1,2	1,1-1,2	0,3-0,7
As	15,6	10,2-52,4	15,2-51,6
Na	1358,8	490,0-650,0	690-1500
Mg	315,0	390,0-480,0	206-492,1
P	trace amount	16,2-62,4	60,2-92,1
K	540,5	93,6-206,8	143,6-206,2
F water-soluble		9,16-31,2	8,6-28,1
pH H₂O	10,4	7,14-7,58	5,0-5,6
pH KCl			4,7-5,1

slightly cool mountain climate. Basin is a little ventilated, has a high incidence of still air (meadow of river Hron up to 41%).

The most important annual climatic characteristics of surroundings Žiar nad Hronom:

- average of temperature per year: 8.3°C, for July 18.2°C, -0.3°C for January
- annual rainfall: 706 mm,
- occurrence of calm is 41%.

One of the most important meteorological factors for the localization of emissions in the entire site is the airflow, because in many cases has a main role in the length, intensity and frequency of exposures of the place against toxic substances out of the plant. Dispersion and dislocations in the area are affected by discharges of direction and wind force. Diffusion of aerosols in the atmosphere depends on the wind speed and temperature stratification from the air. In turbulent fluxes are transferred only by the wind, but during convention will have been received in the higher layers.

The territory is characterized by the largest percentage of the annual average in the north-west (NW) of the winds (20,6%). Depending on the configuration of the terrain in addition to the NW direction occurs an increased percentage of 17.6%) and NE (Eastern winds (15.3%), which allows the presence of elevated concentrations of discharges in the SE, NW and west direction proceed.

The accompanying phenomena in the production of ZSNP a.s. was the creation of a sludge landfill, placed in close proximity to the plant. The chemical composition of brown and red sludge is shown in table 2. Red and Brown sludge landfill capacity in the area of about 45 ha, weight of approximately 8.5 million tonnes, with the real content of iron and aluminum and the presence of other strategic metals after the “canning” is still a raw material for further processing “in situ”.

In the framework of the examination of the impact of

the dump on the agricultural land, we carried out monitoring of the content of hazardous components in landfill, and it is at a distance of around 200, 300 and 500 m from the front of the landfill.

In each distance were taken 5 soil samples from a depth of 0.20 m (a total of 15 samples of the soil), as well as the sample was taken from the soil from the depth of 0.80m (3 samples). Soil samples were analyzed in an accredited laboratory GEL, Turčianske Teplice. In the framework of the analysis have been carried out the following analysis: soil pH (H₂O), pH (KCl), Ca, Mg, Na, Al, Pb, Zn, Cu, Cd, F (the total content) and F (water-soluble). Concentrations of heavy metals were analysed in infusion 2 M HNO₃. The content of the monitoring indicators was subsequently evaluating with the legislative regulations for their maximum content in the soil.

Results and discussion

The results of the analyses of the elements are shown in table 3. Higher content of cadmium in the soil in increased amounts can be determined either by sedimentation derived from geochemical anomalies, imissions from industrial and energy-producing fossil fuel thermal power plants, application of superphosphates or from transport.

The high risk may include in its moving to the deeper parts of the soil profile and groundwater in acidic soils with a low sorption capacity (Kobza et al, 2008). The median value of cadmium content of the sampling points at a distance of 200 metres from the front sludge dump is 1.7 mg.kg⁻¹, at a distance of 300 metres from the front sludge dump is 1.4 mg.kg⁻¹, and at a distance of 500 from the front sludge dump is 2.0 mg.kg⁻¹, as compared to limit value for its content in the soil (0.3 mg.kg⁻¹) set in 2-3 mol.dm⁻³ exceeded limit value by 5 to 6.5 times. When soil reaction is in the range of neutral to alkaline, cadmium accessibility is limited. That is concluded by Hronec et al (2002, 2010), Tomáš et al. (2007, 2011). Cadmium in alkaline soils is slightly limited to moving and the few available for crops.

Alkalization of soil environment largely contributes to high levels of Ca, Mg, Al, Na. Regardless of the distance from the sampling point of the soil samples can be their content considered extremely high, disruptive physico-chemical processes in the soil, which may caused chemodisrupcion of soil, with changes of the physical, chemical and biological properties of the soil. The lead content in the soil can be considered extremely high, where the minimum content (73 mg.kg^{-1}) exceeds the limit value for its content in the soil (3.00 mg.kg^{-1}) by 24 times, and maximum content (244 mg.kg^{-1}) by 77 times. In the case of cadmium content in the soil, we did not record dependency of lead in soil from distance from the front of sludge dump. Lead and its compounds in soils with $\text{pH} > 5$ or with a high content of organic matter is slightly soluble. With the decline of the value of soil reaction $\text{pH} < 4.5$, its solubility rises gradually. This finding highlights the need for liming of soils. High levels of lead in the soil may reduce the total number and variety of microflora in soil. Microbial activity is more inhibited in sandy soils than in clay soils (Beneš, Pabianová, 1987). In terms of zinc content is excess of the limit value for its content in the soil (40 mg.kg^{-1}) in the range of 2.07 to 7.5 times of exceeding. Zinc in the soil with alkaline soil reaction is also just a slight migration capability, since it is bound to organic matter, as well as the Fe oxide forms a complex compounds in the soil, respectively, which does not allow her to receive plants. High levels of zinc in soils are found mainly near urban agglomerations in the vicinity of min-

ing heaps and water treatment plants (Hronec et al., 2010). The degree of accumulation of zinc in the soil is similar to that of cadmium and lead. Outputs represent an average of 81% of the inputs, which is about 50% zinc and another about a third of the collection is “take zinc away” by erosion. In soils full of phosphates, its acceptability by plants has been reduced. It is known to be a positive correlation between the content of zinc and contents of Cu, Mn, Fe in soil. An excess of Fe increased the income of Zn (Tomáš et al., 2011). The copper content in the soil is in the range of $55\text{-}152 \text{ mg.kg}^{-1}$. The limit value for the copper content in the soil is 20 mg.kg^{-1} , and this content has been exceeded by 2 to 7 times. Works of Tóth et al., (2002, 2009, 2011) and Tomáš et al., (2007) point to the significantly positive effects of sodium humate and application of zeolites, such as adsorbents of heavy metals in soil and the subsequent reduction of the content and exposure to heavy metals and harmful elements in plants to significantly contaminated soils of Slovak Republic. Copper has a relatively high affinity for adsorption by organic and inorganic colloids. It is therefore common that may be present in relatively high concentrations in the soil solution, without toxic effects on plants. The major soil properties affecting the availability of copper has the largest effect of pH, organic matter and grain size composition of soil. Organic substances, such as soil humus, reduce accessibility of copper for plants by formation of stable complexes, in which copper can be so strongly strongly bound copper, and is inaccessible to the

Tab. 3 Content of monitored indicators in soil in mg.kg^{-1}

Tab. 3 Zawartość monitorowanych wskaźników w glebie w mg.kg^{-1}

label	pH H ₂ O	pH KCl	Ca	Mg	Na	Al	Pb	Zn	Cu	Cd	F ¹	F ²
1A	7,72	7,05	7120	2480	201	3280	108	156	96	1,5	2217	16,3
2A	7,61	7,96	5840	2080	150	3160	119	154	112	1,6		
3A	8,73	7,49	6570	2010	1830	4580	121	125	118	1,2		
4A	8,77	7,42	14390	3260	2700	5920	121	191	88	1,1		
5A	7,62	6,58	4650	1580	1180	4980	158	83	97	3,0		
Average 1A-5A	8,09	7,30	7714	2282	1212	4384	125	141	102	1,7		
P-A	7,65	7,49	6890	3060	154	3230	176	214	136	2,2		
1B	7,62	6,97	6690	2840	78	2780	137	192	105	1,9		
2B	7,59	7,01	7650	3360	176	3040	157	219	121	2,0	1976	28,4
3B	8,33	7,29	10000	2980	1880	5400	109	127	119	1,3		
4B	9,81	7,85	7280	2160	6520	4420	129	119	80	1,0		
5B	8,33	7,05	5890	2060	1470	4650	73	110	55	0,7		
Average 1B-5B	8,33	7,23	7502	2680	2024	4058	121	153	123	1,4		
P-B	7,72	7,95	5990	1730	5270	5210	106	90	119	0,9		
1C	7,60	7,60	8710	4130	119	3220	215	298	137	2,5		
2C	7,84	6,83	5670	2530	214	3250	179	197	121	2,1		
3C	7,62	6,95	5840	2130	280	3690	244	286	151	2,5		
4C	7,99	6,93	5870	2160	368	4105	153	162	152	1,7	1965	28,0
5C	7,74	6,95	6780	2440	357	4650	142	140	151	1,3		
Average 1C-2C	7,75	7,05	6574	2678	267	3783	186	216	142	2,0		
P-C	7,73	6,93	6210	2280	557	5420	86	85	126	0,8		
Min	7,59	6,58	4650	1580	78	2780	73	83	55	0,7		
Max	9,81	7,96	14390	4130	6520	5920	244	298	152	3		

F1 total content of F (NaOH fusion)

F2 content of water-soluble F (aqueous extract 1:20)

Ca, Mg, Na, Al, Pb, Zn, Cu, Cd – in 2 mol.dm^{-3}

plants. The subsequent mineralisation of soil organic matter by microorganisms, the copper is released in the form of soluble salts.

Industrial emissions are the main anthropogenic source of fluorine, which may be in a gaseous or solid form to receive into the atmosphere, and thus secondary to contaminate soil, water and vegetation.

The average values of gaseous fluorides moved around the aluminium plant in Žiar nad Hronom in the period 1985-1989, was in the range of 2.6 to 6.1 ng.m⁻³ with maximum value of 35.1 ng.m⁻³ (Antalová, 1991). The average value of the water-soluble fluorine is 24.23 mg.kg⁻¹ and high value exceeds the limit value (5 mg.kg⁻¹) (Act No. 220/2004 Coll.). These soils have historically been significantly affected by the F-emissions resulting expression of a high concentration of emitted element (elements) in their surface, which is caused by the strong textural differentiation of the soil profile. The total content of fluorine in soil from the dump decreases with the distance of 200 meters from the sludge dump at 2217 mg.kg⁻¹, 300 metres from the sludge dump at 1976 mg.kg⁻¹ and 500 metres from the sludge dump at 1965 mg.kg⁻¹. Fluorine, as very reactive element, have been attributed to the properties and already exists in the very evaluations in general position. Many works have already been associated with specific data on the burden of the region in the form of water-soluble fluorine, but also the dynamics of the interface shown above the threshold and below the threshold level in the atmosphere.

Conclusion

Monitored area is often referred to and assessed particularly in relation to the contamination of soils, in particular fluorine, which is received into the surrounding environ-

ment from the production of aluminum, and it virtually since 1953, since it first began to producing of aluminum. When in 1957, a second series of electrolysis was put into service, there were also the first disastrous effects of fluorinated discharges to the natural surroundings. In spite of the fact that the emission situation after 1990 started to significantly improve in particular the modernisation of production technology, which quickly began to manifest itself on the quality of the air, on the other hand long-term soil pollution in parts of the soil layer Žiarska kotlina carries the adverse consequences of even at the moment.

Based on the findings can be provided in the vicinity of a sludge dump site in Žiar nad Hronom in terms of contamination of soils classified among the most burdened by the territory of Slovak Republic, despite already quite favorable immission situation.

The content of cadmium in the soil exceeds the limit value by 5 to 6.5 times the lead 24 to 77 times, zinc 2 to 7.5 times, copper 7.6 times, water soluble fluorine 2.7 to 4 times. The average content of total (F) soils is 2052.6 mg.kg⁻¹. The findings are a direct consequences of long-term contamination of the biosphere, and soils in particular. The land is unsuitable for the agricultural purposes, requiring permanent monitoring, the monitoring of the processes of soil degradation, with the possibility of renewal measures and including rehabilitation. It will be necessary to implement the change of use of agricultural land for other purposes.

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Wpływ środowiskowy zrzutów osadów na jakość gleb uprawnych w regionie Žiar Nad Hronom

W związku ze zmianą technologii produkcji aluminium znacznie zmieniła się sytuacja emisji w pobliżu źródła produkcji w zakładzie przetwórstwa (ZSNP a.s.). Było to powodem, przez który znacznie zmieniła się zawartość związków fluoru w powietrzu, a co za tym idzie, również w glebie. Agroregion Žiar nad Hronom znajdował się pod negatywnym wpływem wywieranym na agro-ekosystemie od 1953 roku, kiedy to produkcja firmy ZSNP w ramach stopniowej intensyfikacji wzrosła z pierwotnych 48 tysięcy ton do 70 tysięcy ton. Produkcja anod, elektrod i form ogniotrwałych wzrosła z 50 tysięcy ton do 100 tysięcy ton. Stan ekologiczny wokół zakładu po osiągnięciu zaprojektowanej zdolności produkcyjnej (aluminium, tlenek aluminium, masy anodowe) w 1960 roku był fatalny. Osiągnięcie stanu ekologicznej produkcji w ZSNP a.s. było przedmiotem audytów środowiskowych od 1990 roku. W zakładzie o nazwie SLOVAL a.s. spowodowało to zamknięcie sekcji "B" 29 lutego 1996, w której realizowana była technologia Soderberga i produkcja aluminium. Technologia ta była wykorzystywana przez 43 lata. Znacząca redukcja emisji była spowodowana wdrożeniem nowej technologii "HYDRO ALUMINA". Składowisko czerwonych i brązowych osadów zajmuje powierzchnię około 45 ha i waży około 8,5 mln ton. W ramach badania wpływu składowiska na grunty rolne, przeprowadzono monitoring zawartości składników niebezpiecznych w składowisku. Został on wykonany w odległości około 200, 300 i 500 metrów od przedniej granicy składowiska. Pięć próbek gleby pobierano z głębokości 0,20m w każdym kierunku (w sumie 15 próbek gleby), pobrano także próbki z podłoża. Probki gleby przeanalizowano w laboratorium akredytowanym GEL, Turčianske Teplice. Na podstawie wyników oznaczania wiadomo, że zawartość Na znajduje się w zakresie 78-5,270 mg.kg⁻¹, Al 2,78-5,92 mg.kg⁻¹, Pb 73-244 mg.kg⁻¹, Zn 83-298 mg.kg⁻¹, Cu 55-152 mg.kg⁻¹, Cd 0,8-3,0 mg.kg⁻¹. Całkowita zawartość fluoru wynosi 1,965-2,217 mg.kg⁻¹ i jego zawartość w glebie zmniejsza się wraz ze wzrostem odległości od składowiska. Ten sam wynik jest odnosi się do przypadku zawartości rozpuszczalnego w wodzie F w glebie, którego zawartość wynosi 28,0-46,3 mg.kg⁻¹.

Słowa kluczowe: czerwony szlam, brązowy szlam, metale ciężkie, skażenie gruntu