

Evaluation of the Environmental Risk from the Co-Deposition of Waste Rock Material and Fly Ash

A. Mavrikos, N. Petsas, E. Kaltsi, D. Kaliampakos

Abstract—The lignite-fired power plants in the Western Macedonia Lignite Center produce more than $8 \cdot 10^6$ t of fly ash per year. Approximately 90% of this quantity is used for restoration-reclamation of exhausted open-cast lignite mines and slope stabilization of the overburden. The purpose of this work is to evaluate the environmental behavior of the mixture of waste rock and fly ash that is being used in the external deposition site of the South Field lignite mine. For this reason, a borehole was made within the site and 86 samples were taken and subjected to chemical analyses and leaching tests. The results showed very limited leaching of trace elements and heavy metals from this mixture. Moreover, when compared to the limit values set for waste acceptable in inert waste landfills, only few excesses were observed, indicating only minor risk for groundwater pollution. However, due to the complexity of both the leaching process and the contaminant pathway, more boreholes and analyses should be made in nearby locations and a systematic groundwater monitoring program should be implemented both downstream and within the external deposition site.

Keywords—Co-deposition, fly ash, leaching tests, lignite, waste rock.

I. INTRODUCTION

THE lignite basin of Kozani – Ptolemaida – Amyntaio is the biggest and most important in Greece. The proved lignite reserves contained in this basin are more than $4 \cdot 10^9$ t, which account for 65–70% of the country's total coal resources [1]. There are 5 lignite mines currently in operation in this area that feed 4 thermal power plants (TPPs). These plants have a total installed capacity of 4000 MW and cover 32% of the total energy generation in Greece.

As a consequence of lignite burning, large quantities of fly ash are produced. In the Western Macedonia Lignite Center (WMLC), the annual quantities of fly ash exceed $8 \cdot 10^6$ t [2]. Over 90% of this fly ash is used in exhausted open-cast lignite mines restoration-reclamation with overburden. Fly ash acts as binder for the increase of slope stabilization of the overburden in the restoration process.

Several studies have been conducted on Greek fly ashes regarding their mineralogical and chemical composition [3]–

[5], leaching behavior of trace elements and heavy metals [6], [7], as well as the correlation of trace elements and heavy metals in the lignite with the respective compositions in fly ash [8]. However, there are not any available data on the environmental behavior of mixtures of waste rock material and fly ash that are being used for the restoration of open-cast mines.

Therefore, the present study deals with the evaluation of the environmental behavior of the co-deposited waste rock and fly ash based on chemical composition and leaching potential tests. For this purpose, a large number of samples were collected from the external deposition site in the South Field mine. The site contains materials with a wide variety of composition (marl, sandy marl, sand, clay etc.), granulometry and stratigraphic origin due to the exploitation of the mine, as well as fly ash from the TPP Agios Dimitrios.

II. FLY ASH AND ENVIRONMENTAL INFLUENCE FROM BACKFILLING OPERATIONS

A. Fly Ash

Fly ash is a fine powder, consisted mainly of spherical, glassy particles that originate from burning of pulverized coal [9]. According to the European standard EN 197-1, fly ash is divided into two main categories, i.e. siliceous fly ash (V), which contains less than 10% CaO, and calcareous fly ash (W), which contains 10–35% CaO. Siliceous fly ash has pozzolanic properties, while calcareous fly ash may also have hydraulic properties.

The American standard ASTM C 618 classifies fly ash into three Classes, namely Class N, which includes raw pozzolans, with a minimum of 70% of $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$, Class F, which originates from anthracite and bituminous coals, with a minimum of 70% of $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ and Class C, which originates from sub-bituminous and lignite coals, with 50–70% $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$. Class F fly ash usually has less than 5% CaO, while Class C fly ash has a higher CaO content (10–35%).

Greek fly ash in the WMLC is calcareous (W) according to EN 197-1 and Class C according to ASTM C 618, due to its high CaO levels (Table I) [5].

Fly ashes also contain trace elements (As, Pb, Cr, Ni, Zn, Cu, Ba, Th, Se, Mo, etc.) in relatively high concentrations. Table II shows the main trace elements of fly ash in the WMLC [7].

Environmental concerns from the presence of trace

Dr. A. Mavrikos is with the School of Mining and Metallurgical Engineering, National Technical University of Athens, Athens, 15780, Greece (phone: +30 2107722190; e-mail: mavrikos@metal.ntua.gr).

Dr. N. Petsas and E. Kaltsi were with the Generation Environment Department of Public Power Corporation S.A., Athens, 10432, Greece (e-mail: n.petsas@dei.com.gr, e.kaltsi@dei.com.gr).

Prof. D. Kaliampakos is with the School of Mining and Metallurgical Engineering, National Technical University of Athens, Athens, 15780, Greece (phone: +30 2107722211; e-mail: dkal@central.ntua.gr).

elements depend mainly on their form within the fly ash and on their mobility capability (e.g. Mo shows high leaching ability in alkaline environment). Thus, many trace elements remain entrapped in fly ash, despite the fact that they may come in contact with rain water in the deposition areas. There are, however, other circumstances that favor trace element leaching and create risks about possible degradation of groundwater quality. The issue of leaching, which could affect the environment, is of major importance for a proper evaluation of the environmental compatibility of fly ash [9].

TABLE I
CHEMICAL COMPOSITION OF FLY ASH IN THE WMLC

Main oxides	Typical range (% wt.)
SiO ₂	25,35 - 35,30
Al ₂ O ₃	9,22 - 13,51
Fe ₂ O ₃	4,98 - 6,35
CaO	29,10 - 41,00
MgO	2,85 - 4,76
Na ₂ O	0,35 - 0,62
K ₂ O	0,92 - 1,49
TiO ₂	0,47 - 0,77
SO ₃	3,55 - 6,92
LOI	3,38 - 6,17

TABLE II
TRACE ELEMENTS OF FLY ASH IN THE WMLC

Trace element	Typical range (mg/kg)
As	20,8 - 38,8
B	0,28 - 0,34
Ba	124,2 - 229,4
Cd	0,65 - 1,49
Co	14,9 - 20,7
Cr	148,9 - 201,1
Cu	30,3 - 74,9
Mn	102,3 - 246,8
Mo	5,0 - 7,3
Ni	217,4 - 265,5
Pb	20,4 - 27,2
Sb	0,82 - 1,33
Se	10,0 - 25,7
Sr	318,9 - 385,5
V	89,4 - 116,2
Zn	63,0 - 130,0

B. Fly Ash as a by-Product – REACH Regulation

It is well established that fly ash can be utilized as a by-product in the building material industry, in civil engineering, in road construction, for construction work in underground coal mining, as well as for recultivation and restoration purposes in open cast mines [10], [11]. Each producer/importer/marketer of fly ash placed in the European market must register the substance within the framework of REACH (Registration, Evaluation, Authorization and Restriction of Chemicals) Regulation that entered into force on 1st June 2007 [12]. REACH mandates that chemical substances on their own, in preparations and those which are intentionally released from articles have to be registered to the European Chemicals Agency (ECHA). The registration requires detailed

and analytical information on the properties and the potential risks of the substances, based on toxicological and ecotoxicological data.

The producers of fly ash have registered their product for use in the construction industry under REACH. All information about chemical, physical, toxicological and ecotoxicological properties was compiled and submitted in a registration dossier. Fly ash has been registered as a UVCB substance (substance of unknown or variable composition, complex reaction products or biological materials) and has been assigned with the EC number 931–322–8. All commercial uses of the substance have also been described. Details of the chemical composition and the properties of the registered fly ash are publicly available at the ECHA website (www.echa.europa.eu). Fly ash in the WMLC, according to its chemical composition as given in Table I, is covered from the substance that has been registered.

As a result of the hazard assessment and PBT/vPvB assessment it has been found that the registered fly ash does not meet the criteria for classification as hazardous (according to Directive 67/548/EEC and Regulation 1272/2008/EC) and does not meet the criteria for PBT/vPvB substances.

C. Environmental Influence from Backfilling Operations with Fly Ash

The problems that relate to the vast production, combined with certain properties of the material, led to alternative ways of utilizing fly ash. Since 1930, fly ash has been used in road construction as mineral filler in asphaltic mix, in embankment soil stabilization, sub-grade base course material, as aggregate filler and bituminous pavement additive [6], [13]. Currently, fly ash applications have expanded and include cement production and related products [13], [14], restoration and remediation of polluted soil [15], [16] and production of synthetic zeolites [17]. In Greece, the main application of fly ash is the backfilling for restoration of exhausted open-cast mines. Almost 70% of the total produced quantities are used in this direction. Another 10%, with a trend to decrease lately due to the recent financial crisis, involves the utilization in other commercial applications, mainly in the cement industry, which is rather low when compared to the European average of 18% [2], [4].

Internationally, there is a widespread utilization of fly ash in filling applications of abandoned mines and embankments. In this respect, various studies have been published that examine possible influence of the groundwater, adjacent to the deposition areas, from trace element leaching. Dutta et al. reported on the leaching of ten trace elements (Fe, Mn, Ca, Na, K, Cu, Cr, Zn, As και Pb) in an attempt to assess the possibility of site contamination by release of heavy metal ions, if fly ash were to be used for backfilling of abandoned mines [18]. Various leaching tests were performed in four fly ash samples from different TPPs. They concluded that utilization of fly ash could potentially degrade groundwater, especially under acidic leaching conditions, and it was suggested that a proper mix with lime could offer a low cost practical solution.

Hansen & Christ reported on the quality of surface and groundwater in two mine sites, which had been filled with $2.2 \cdot 10^6$ t and $4 \cdot 10^6$ t of FBC ash, respectively, in order to control acid mine drainage [19]. Results showed a positive influence (pH increase) regarding surface waters. Nevertheless, there was an increase in the concentration of certain elements, such as Se and Tl. In the case of groundwater, pH ranged between 5.5–7.5, while increased As and Se concentrations were observed. It was concluded that the use of FBC ash was not adequate for controlling acid mine drainage and in some occasions led to increased trace elements concentrations downstream from the ash placement site.

Prasad and Mondal studied the influence of fly ash in groundwater quality in the case of filling an abandoned open-cast mine [20]. Results from samples within the site, as well as in the periphery of the mine, showed that all parameters were below drinking water specifications, with the exception of F and Mn. Groundwater quality at the periphery of the ash-filled zone was only slightly affected and there was no apparent effect on ground water quality 500m away from the zone, where groundwater was used for drinking water.

Similar conclusions were also drawn by Arnold et al., who examined several embankments from fly ash in UK [21]. The embankments were of different age and were comprised of fly ash with different chemical composition. The purpose of the study was to determine the extent of ground and surface water contamination in the areas adjacent to the embankments and the possible influence of fly ash. Results showed that on 3 out of 5 embankments, concentrations of sulfuric anions and four elements (Mg, Se, K, Na) were above the limits set for drinking water, possibly relating to the presence of fly ash. However, similar results were observed in an embankment that was not comprised of fly ash. The elements concentrations in groundwater samples were low. The study concluded that the use of fly ash in embankments did not pose any significant source of pollution and it was unlikely to cause any risks to human health.

Jo *et al.* also investigated the environmental feasibility of using coal ash as fill material [22]. The study focused on defining the leaching characteristics of bottom ash, lagoon ash, bottom ash–soil mixture and lagoon ash–soil mixture. The measured trace elements concentrations were below the limit values set by the Korean Ministry of Environment, US Environmental Protection Agency (EPA) and World Health Organization (WHO), except for Mn in all samples and Se in some samples. It was concluded that the tested materials may have little potential for risk to groundwater.

III. ENVIRONMENTAL BEHAVIOR OF THE EXTERNAL DEPOSITION SITE OF THE SOUTH FIELD LIGNITE MINE

A. External Deposition of the South Field Mine

South Field mine is located at the southern part of the lignite basin of Ptolemaida – Amyntaio. It includes the exploitation of the homonymous lignite mine, has an annual lignite production capacity of $18 \cdot 10^6$ – $22 \cdot 10^6$ t and covers the demands of the five units of the TPP Agios Dimitrios, which

has a total installed power capacity of 1595 MWe.

The external deposition of the South Field mine began in 1981 (Fig. 1). Initially, only waste rock material from the mine was deposited, such as marls, sands, overburden and topsoil with very low calorific value. Deposition of fly ash initiated in 1984, in the area between the villages Klitos and Akrini. Additionally, fly ash from the auxiliary deposition site near TPP Agios Dimitrios was also transferred with trucks. In early 1985, the main branch of conveyor belts was installed for the transfer of fly ash from the TPP Agios Dimitrios. Currently, the deposition site covers an area of approximately 5.4 km^2 , has an average height of 80–90m and it is estimated that the total quantity of waste rock material and fly ash that have been deposited comes up to $460 \cdot 10^6 \text{ m}^3$, with an average fly ash content of 7–10%.

For the generation of the external deposition site in the South Field mine, loader, spreaders and conveyor belts were used. There was no specific provision regarding the kind, the quantities or the mixing of the materials and there was a constant succession of overburden layers which were intercepted by fly ash layers. The irregular alternation of the layers and the different extent of compression of the deposited materials have contributed to the formation of a particularly complex embankment, both from geotechnical as well as from hydro–geological point of view. These specific characteristics, as mentioned below, have a significant influence on the chemical composition and the leaching potential of the samples and, consequently, on the environmental behavior of the deposition.

B. Sampling and Analyses

For the evaluation of the environmental behavior of the co-deposition of waste rock materials and fly ash in the external deposition site of the South Field mine, one borehole (G1 in Fig. 1) was drilled within the site, with 96m length and 101 mm diameter. The borehole was deep enough to fully penetrate the deposition and at least 10m of the underlying natural soil. Sampling was performed under dry conditions, in order to avoid leaching of the samples from the boring water. The core of the borehole was cased in PET bags so as to maintain the natural moisture of the sample until the chemical analyses and leaching tests.



Fig. 1 Location of the borehole (G1) on the external deposition site of the South Filed mine

It was not possible to classify the layers of the deposition into specific geological formations, while the materials were only generally categorized (e.g. deposition materials, clay, tailings etc). The upper layer of the borehole (0–35m) consisted of overburden and fly ash. In the 35–44m layer, there were clays and tailings, while in the 44–53m layer there were only clays. Another layer of deposited overburden was found between 53–75 m (lower layer) and between 75m until the bottom of the borehole there were natural undisturbed formations, i.e. conglomerates (75–78m and 90–93m) and clays (78–90m and 93–96m).

86 samples were taken totally from the borehole. Macroscopic and laboratory analyses of the samples were performed for the zones 0–35 m and 53–75m (depositions), as well for the zone 75–96m (underlying natural soil).

C. Results from Chemical Analyses

The average chemical composition of the materials of the borehole, in total and also based on the division in two deposition zones and natural soil, is shown in Table III.

TABLE III
AVERAGE CHEMICAL COMPOSITION OF THE BOREHOLE (mg/kg)

Trace element	Total	Upper zone (0 - 35 m)	Lower zone (53 - 75 m)	Underlying natural soil
As	11,43	12,11	8,19	11,28
B	250,21	247,10	321,50	194,45
Ba	1,01	1,05	1,06	0,75
Cd	31,18	32,21	26,75	30,45
Co	309,64	330,50	231,00	284,00
Cr	28,34	29,43	20,70	30,55
Cu	0,016	0,016	0,017	0,014
Mn	675,21	685,20	589,50	711,00
Mo	2,39	2,71	1,22	1,55
Ni	269,14	285,60	208,50	247,50
Pb	15,33	15,74	13,10	15,50
Sb	< 5	< 5	< 5	< 5
Se	< 5	< 5	< 5	< 5
Sr	155,69	160,26	172,50	116,05
V	90,11	93,96	70,25	90,75
Zn	68,44	69,40	57,35	74,75

The above results show a distinction between the two zones

of the deposited materials. The upper and, therefore, more recent zone exhibits increased concentrations of Mo (122%), As (48%), Cr (43%), Cu (42%), Ni (37%) and V (34%) when compared to the lower zone. In comparison to natural soil, the upper zone has increased concentrations of Mo (75%), Cd (41%) and Sr (38%).

The lower zone has increased concentrations of Ba (65%), Sr (49%) and Cd (42%) in relation with the underlying natural soil. The latter shows higher concentrations in all other trace elements by 20–30%. This observation could be attributed to a certain “enrichment” of the undisturbed formations of the underlying natural soil, probably due to leaching phenomena from the overburden and fly ash depositions.

D. Results from Leaching Tests

Leaching tests were performed according to ELOT EN 12457-1 and ELOT EN 12457-2 standards. For comparison reasons only, trace elements concentrations in the leachates were evaluated in relation to the limit values for waste acceptable at landfills for inert waste, according to the Council Decision 2003/33/EC [23]. Tables IV and V show the results of the leaching tests. Even though the results based on ELOT EN 12457-2 are different in absolute values, general conclusions are not affected.

Results showed that concentrations in the leachates are lower compared to the limit values for inert waste at landfills for the trace elements As, Ba, Cd, Mo, Cu, Hg, Ni, Pb, Sb, Se and Zn, as well as for Cl⁻, F⁻ and Dissolved Organic Carbon (DOC). On the contrary, Cr (only in the upper zone) and SO₄²⁻ exceeded the limit values, but, in total, only SO₄²⁻ concentrations were above the limits. For the extraction of average values, the detection limit was considered for all samples with concentrations below the limits of instrumental analysis.

TABLE IV
AVERAGE LEACHING VALUES OF THE BOREHOLE (mg/kg, ELOT EN 12457-1)

Trace element	Limit value*	Total	Upper zone (0 - 35 m)	Lower zone (53 - 75 m)	Underlying natural soil
Al	—	3,1443	0,7634	4,1284	8,2180
As	0,1	<0,1	<0,1	<0,1	<0,1
B	—	0,2035	0,3618	<0,1	<0,1
Ba	7	0,1026	0,0974	0,0763	0,1575
Cd	0,03	<0,004	<0,004	<0,004	<0,004
Cr	0,2	0,1161	0,2102	0,0333	0,0773
Cu	0,9	0,0129	0,0106	0,0153	0,0151
Fe	—	3,8389	3,5093	2,7020	5,0341
Hg	0,003	<0,001	<0,001	<0,001	<0,001
Mn	—	0,2300	0,0809	0,2206	0,5837
Mo	0,3	0,0480	0,0941	0,0146	0,0210
Ni	0,2	0,0669	0,0399	0,0476	0,1521
P	—	0,5250	0,5045	0,3600	1,1400
Pb	0,2	0,0299	<0,02	0,0254	0,0308
Sb	0,02	<0,02	<0,02	<0,02	<0,02
Se	0,06	<0,04	<0,04	<0,04	<0,04
Sr	—	0,5393	0,7634	0,4800	0,3692
V	—	0,0417	0,0817	0,0125	0,0168
Zn	2	0,0337	0,0289	0,0399	0,038
Cl ⁻	550	25,8667	30,0889	15,8	8,8
F ⁻	4	1,0836	1,1825	1,0	0,67
SO ₄ ²⁻	560	2000,0	2234,4444	2147,0	574,0
DOC	240	42,5383	44,0222	67,25	40,02

*: inert waste, 2003/33/EC

TABLE V
AVERAGE LEACHING VALUES OF THE BOREHOLE (mg/kg, ELOT EN 12457-2)

Trace element	Limit value*	Total	Upper zone (0 - 35 m)	Lower zone (53 - 75 m)	Underlying natural soil
Al	—	18,6067	1,9582	7,7887	71,0684
As	0,5	<0,5	<0,5	<0,5	<0,5
B	—	0,6885	0,9709	0,5087	<0,5
Ba	20	0,3302	0,2827	0,2290	0,6342
Cd	0,04	<0,02	<0,02	<0,02	<0,02
Cr	0,5	0,2772	0,2898	0,0983	0,5788
Cu	2	0,0699	0,0546	0,0720	0,1074
Fe	—	18,3516	7,7069	7,4715	55,7764
Hg	0,01	<0,005	<0,005	<0,005	<0,005
Mn	—	0,9731	0,5749	0,4162	2,6331
Mo	0,5	0,0914	0,1346	0,0603	0,0663
Ni	0,4	0,3595	0,1802	0,1613	1,0392
P	—	1,0329	1,2818	0,7391	1,2368
Pb	0,5	0,1576	0,1012	<0,1	0,3584
Sb	0,06	<0,05	<0,05	<0,05	<0,05
Se	0,1	<0,1	<0,1	<0,1	<0,1
Sr	—	1,2129	1,9582	0,7835	0,8184
V	—	0,2939	0,4482	0,1015	0,2129
Zn	4	0,1682	0,1746	0,1003	0,2835
Cl ⁻	800	35,16	43,3	41,0	13,25
F ⁻	10	5,36	5,9571	4,55	3,7
SO ₄ ²⁻	1000	2934,1	3624,4286	2155,0	620,0
DOC	500	119,2	108,1714	250,4	132,95

*: inert waste, 2003/33/EC

The aforementioned results from leaching tests are consistent with the respective results from chemical analyses, since they clearly demonstrate the different behavior of the two zones of the deposited materials and the underlying natural soil. Leaching of Cr is six times higher in the case of the upper zone, while the respective leaching of Mo is seven times higher when compared to the lower zone and four times if compared with natural soil. Moreover, the lower zone of the deposited materials shows no significant differentiation from natural soil, with the exception of SO₄²⁻.

This lack of uniformity in the behavior of the deposited materials in relation to depth is shown in Figs. 2–6. Based on these diagrams:

- Mo and Cr exhibit similar leaching behavior in relation to borehole depth.
- At the lowest point of the upper zone (32–35m), there is a layer of materials with alkaline pH, high conductivity and more intense trace elements leaching, which can probably be attributed to a plain fly ash layer.
- Below 40m, both Cr and Mo leaching seem to decrease and practically (with only very few exceptions) drop to zero. This observation, combined with the analyses of the deeper layers of the deposition and the natural soil, suggests the absence of fly ash below this level.
- The most intense leaching behavior of all the examined parameters is observed between 0–20m.

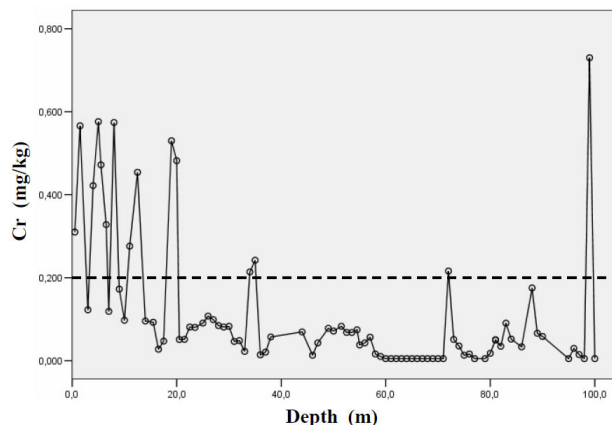


Fig. 2 Leachate concentrations of Cr for various depths (EN 12457-1). The dotted line represents the limit value for waste acceptable at landfills for inert waste

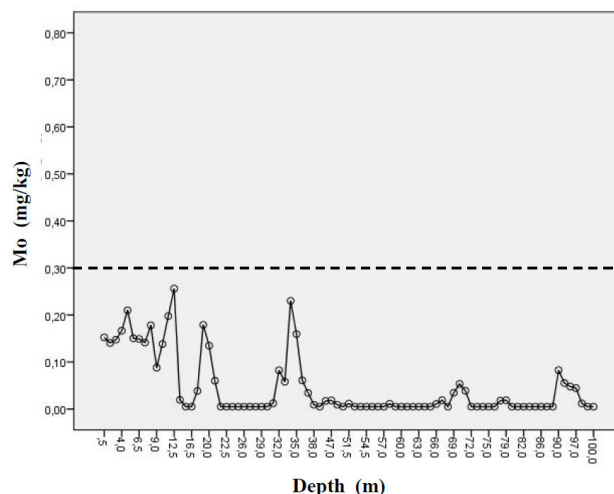


Fig. 3 Leachate concentrations of Mo for various depths (EN 12457-1). The dotted line represents the limit value for waste acceptable at landfills for inert waste

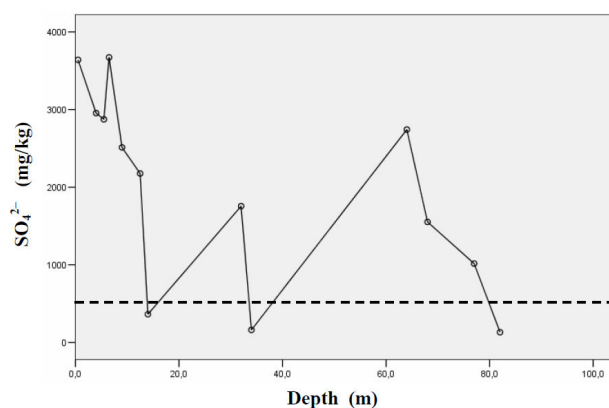


Fig. 4 Leachate concentrations of SO₄²⁻ for various depths (EN 12457-1). The dotted line represents the limit value for waste acceptable at landfills for inert waste

Regarding pH values (Fig. 5), they are alkaline in the upper zone and neutral in the rest of the samples. At 35m, pH is highly alkaline and Mo leaching is also intense. However, below 40m, pH values are similar as in natural soil samples. This fact clearly demonstrates the retention or precipitation of all elements from the clays and verifies the assumption that there is no fly ash deposited below this point. Conductivity values (Fig. 6) follow the same trend as pH values. The peaks of the plot coincide, regarding depth, with the respective peaks from the Mo leaching diagram.

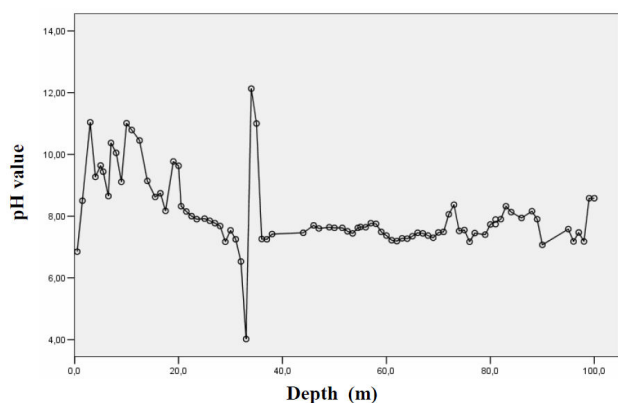


Fig. 5 pH values in various depths (EN 12457-1)

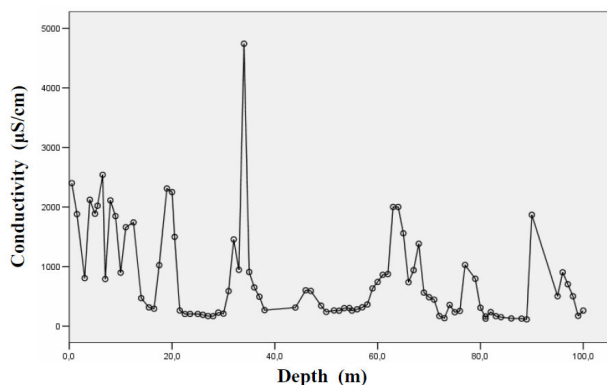


Fig. 6 Conductivity in various depths (EN 12457-1)

IV. DISCUSSION AND CONCLUSIONS

Trace elements concentrations in leaching tests, which are of particular importance primarily for the assessment and evaluation of potential risks in the area's groundwater, exceed only in certain few occasions the limit values set for Cr in landfills for inert waste, mainly in the upper and more recent layers of the deposition. Below 40m, Cr (and Mo) leaching decreases and is practically depleted. Several samples showed exceedances of the limit values set for sulfates. Nevertheless, the latter are above the limits even in the underlying natural soil samples.

Therefore, it can be concluded that only minor environmental issues might arise from groundwater pollution due to leaching. Besides, hydraulic properties of fly ash create artificial barriers that isolate the subjacent materials for

rainwater flow. The results of the present study confirm the fact that, in practice, inorganic trace elements and heavy metals are leached only to a small degree. It should be noted that these assessments are also verified from chemical analyses in groundwater samples downstream of the deposition. However, since the behavior of the whole deposition is highly complex because of the dissimilarities and heterogeneity of the materials, there should be a systematic monitoring of the groundwater quality in the area and downstream of the deposition on a long-standing basis. Furthermore, it is essential that more boreholes should be made in order to verify the results of this study and a quantitative risk assessment study should be performed for the area's groundwater pollution, based on the "source-path-receiver" principle.

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