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EFFECTS OF LANDFILL LEACHES ON GROUND AND SURFACE WATERS: A CASE STUDY OF A WILD LANDFILL IN EASTERN BOSNIA AND HERZEGOVINA

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ABSTRACT

Landfill leachate, due to its high total pollution, and above all due to their high organic pollution, represents a significant environmental problem. This study investigated the impacts of the Tilić ada landfill on ground and surface waters. The location of the landfill Tilić ada is extremely sensitive due to the fact that it is located right next to the Drina river bed on the border with the Republic of Serbia, and especially because the source of drinking water Tilić ada is located at a distance of approx. 500 meters.

Therefore, analyses of impact of leachate from the landfill were carried out, which indicated that the groundwater is at risk even 5 years after the landfill was closed. At the same time, water analyses from the Drina River were also carried out, which showed that the water quality was not impaired in relation to the defined water class.

Keywords**:** *landfill, leachate, groundwater, surface water.*

INTRODUCTION

The Tilić ada landfill was used for the disposal of collected mixed municipal and non-hazardous waste from the wider and narrower urban areas of the City of Zvornik in the ten-year period between 2007 and 2017 at the location of the old gravel pit. Its estimated area is approximately 2.5 ha. Given that it is a wild, unorganised, unsanitary landfill, no technical documentation based on the principles of sustainable waste management has been prepared, that is to say necessary measures were not implemented to reduce the hazardous environmental impact of the waste during its disposal at the location. No stormwater drainage system was built, and leachate went and continues to go underground in an uncontrolled manner.

The location of the Tilić ada landfill is extremely sensitive due to the fact that it is located right next to the Drina River bed on the border with the Republic of Serbia, and especially because Tilić ada potable water spring is located upstream of the landfill at a distance of approx. 500 metres. [1]. This is the reason why the landfill's impact on underground and surface water was analysed 5 years after its closure.

Landfill leachate, due to its high total pollution, represents a significant environmental problem [2,3]. Previously, a large number of studies were conducted that pointed to the leachate of waste disposal sites on the environment [4,5,6,7].

The chemical characteristics of leachate are affected by biological decomposition of biodegradable organic substances, chemical oxidation processes and dissolution of organic and inorganic substances in waste [8,9]. Landfill leachate is produced by percolation of rainwater through the landfill body, during which soluble, colloidal and suspended substances are extracted. In other words, landfill leachate is a polluted liquid, which seeped through the layers of deposited waste and thereby absorbed large amounts of pollutants, including the products of chemical and biochemical reactions that take place in the landfill body. Leachate consists of liquids that enter the landfill body from the outside, that is to say from precipitation, infiltrated groundwater, as well as water contained in the waste itself [10].

Total quantity of the landfill leachate consists of the quantity of external water that entered the landfill body and the quantity of internal water in the landfill. External waters that can reach the landfill are: rainwater from the catchment area, surface catchment waters, precipitation and ground water.

Part of the water also enters the landfill body directly with the waste (internal water), through the moisture of the waste being deposited. In untreated municipal waste, the moisture ranges between 20 and 60 wt %. However, as a rule, the moisture of untreated municipal waste is below the saturation point, so that on average the deposited waste can accept about 12 vol % of additional moisture.

The main source of landfill leachate is precipitation that comes to the landfill surface and seeps through the landfill body. Part of this water runs off as rainwater from the landfill, part returns to the atmosphere by evaporation from the upper landfill surface or vegetation (evapotranspiration), while the rest is leachate that occurs after the waste reaches full moisture saturation. The climate has a significant effect on the rate of leachate formation, as the quantity of this water is much higher in the high rainfall zone than in the low rainfall zone. Soil topography affects the direction of the water flow, as well as the quantity of water entering and leaving the landfill zone. The permeability of soil interlayers located in the landfill will affect the rate of downward movement of water. Leachate quantity decreases with increased surface water runoff, more intense evaporation of water from the landfill surface and a decrease in moisture in the overlying soil layers.

Given the identification of an unorganized landfill with different quantities of waste, type of waste, disposal period, composition and slope of the terrain, etc., where no monitoring is carried out, the quantity of leachate can be calculated in accordance with the following formula:

$$Qf = \frac{k\ x\ P}{30} \tag{1}$$

where:
Qf - daily amount of filtrate (leachate), m^3/day;
k - infiltration coefficient = 0.7 (for landfills on flat terrain) and 0.5 (for landfills on steep terrain);
P - total monthly amount of atmospheric precipitation on the given landfill area, m^3.

The infiltration coefficient is high due to the lack of data on evapotranspiration, surface runoff, the amount of moisture in the waste, the clear composition of the waste, and represents a certainty in the calculation.

A municipal solid waste landfill can be viewed as a biochemical reactor, with waste and water as inputs, and biogas and leachate as the main output components. The composition of leachate varies during the landfill exploitation and the most important factors that influence variations in the composition of leachate are [11]:

- composition of waste and its variability, which determine the rate of decomposition.;
- temperature in the landfill body oscillates according to the season and affects the growth of microorganisms and the rate of chemical reactions.;
- thickness of the deposited waste layer: thick layers of waste need more water to saturate, so the decomposition process takes longer.

The composition of leachate is particularly affected by landfill age. As the landfill ages, the concentration of organic substances decreases more than the concentration of inorganic substances, because they are decomposed and washed away, while inorganic substances are only washed away. Moisture significantly affects the degree of waste decomposition, considering that it helps with the exchange of substrates, nutrients, dilution of inhibitors and the growth of microorganisms. The most important influence on the moisture content in the landfill is the landfill construction method, the method of waste disposal and the climate. Leachate from municipal waste landfills generally contain the following impurities [12]:

- nitrogen compounds in organically bound form and in the form of ammonia: represents the highest percentage of soluble nitrogen in leachate landfill waters and is formed during the biodegradation of organic substances present;
- phosphorus compounds: they are involved in many physical, chemical and microbiological transformations. Phosphorous species are most often used in microbiological processes, complexation and dissolution.;
- heavy metals: certain concentrations of the following heavy metals occur in most filtrates from municipal landfills: Al, As, Cu, Ba, Fe, Zn, Cd, Co, Ag, Pb and Hg;
- cations: the most common cations that occur in leachate are: Na^+, K^+, Mg^{2+}, Ca^{2+}. They react with each other and with cations in waste complexes, creating complexes;
- anions: Cl^-, SO_4^{2-}, S^{2-} and HCO^{3-} are only partially transformed.;
- organic pollution: expressed through non-specific parameters BOD_5, COD and TOC;
- chlorinated hydrocarbons and pesticides;
- specific organic compounds: aromatic hydrocarbons, phenols, chlorinated aliphatic compounds, which are usually found in traces.

The filtrate is characterised by the following properties [13]:

- colour dark brown to black, unpleasant smell;
- pH in young landfills is acidic, and in old landfills basic (pH = 5.3 - 9.1);
- BOD_5 and COD very high during the acid fermentation phase, and significantly lower during the methane fermentation;
- the content of heavy metals in the acid fermentation phase is relatively high, and during methane fermentation it is almost negligible;
- relatively high chloride content in the acid fermentation phase;
- high ammonia content;
- very low phosphorus content.

The water contained in solid waste, as well as the water that infiltrates the landfill, form a medium in which all soluble substances are dissolved and which causes the movement of unreacted material downwards, towards the bottom of the landfill [14,15]. These waters are known as leachate. Landfill leachate is a medium whose composition and quantity change significantly during the landfill lifetime [16,17]. Filtrates from waste disposal sites are among the most problematic types of waste water from the aspect of toxicity. Research has shown that each landfill represents a separate system and that in this sense, the composition and amount of leachate depends exclusively on the characteristics of the landfill itself ([18,19,20].

In order to more reliably assess the expected composition of leachate from this wild and unregulated landfill, the results of leachate impact on groundwater and surface water were analysed.

MATERIAL AND METHODS

Sampling and physical and chemical analysis of leachate at the Tilić ada landfill location was carried out on November 2022. Sampling was carried out at the following locations (Figure 1.):

- three deep piezometers with a depth of 15 metres installed directly next to the landfill body (marked PB-1, PB-2 and PB-3),
- two deep piezometers with a depth of 10 metres installed in the landfill body (marked PB-4 and PB-5)
- two shallow piezometers with a depth of 6 metres installed in the landfill body (marked PB-6 and PB-7).

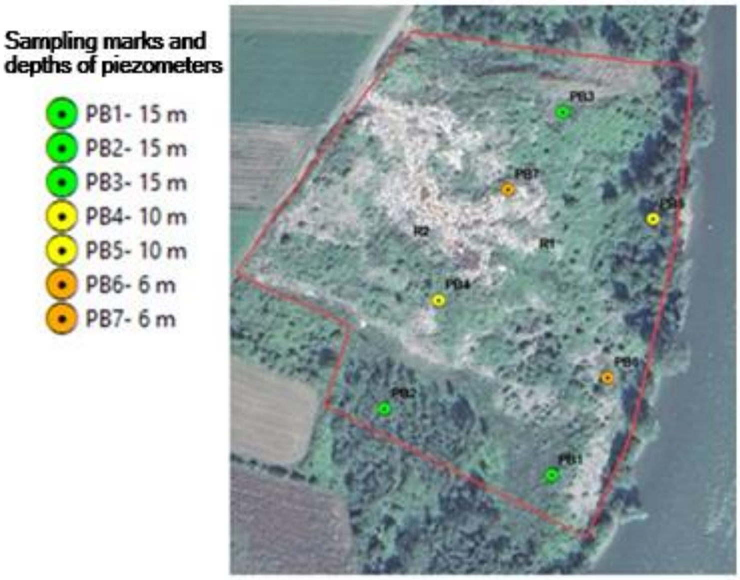

Figure 1. Position of piezometers from which groundwater sampling was performed

Sampling and physical and chemical analysis of the Drina River water quality was carried out at three locations on November 2022. Sampling was carried out at the following locations (Figure 2.):

- upstream of the landfill location (marked D1),
- directly next to the landfill location (marked D2)
- downstream of the landfill location (marked D3).

Figure 2. Sampling sites of the Drina River water

The analysis of water quality from piezometers and the Drina River included the following parameters: pH, electrical conductivity (EC), total dissolved solids (TDS), total suspended solids (TSS), biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), total nitrogen, total phosphorus, sulphates, chlorides, lead (Al), iron (Fe), manganese (Mn) and aluminium (Al). Four metals were chosen because of their availability in landfill leachates All parameters were analysed in accordance with the current water regulation [21,22]. The classification of surface waters according to the current water regulation is shown in the Table 1.

Table 1. The classification of surface waters with maximum permissible concentration (MPC)

Parameter	Unit of measure	MPC for surface waters				
		Class I	Class II	Class III	Class IV	Class V
pH	pH units	6.80-8.50	6.80-8.80	6.5-9	6,5-9,5	<6,5;>9,5
EC at 20°C	μS/cm	<400	400-600	600-800	800-1500	>1500
TDS	g/m^3	<300	300-350	350-450	450-600	>600
TSS	g/m^3	<2	2-5	5-10	10-15	>15
BOD_5	$g\ O_2/m^3$	<2	2-4	4-7	7,0-15	>15
COD	$g\ O_2/m^3$	<12	12-24	22-40	40-50	>50
Total nitrogen	$g\ N/m^3$	<1	1-6	6-12	12-30	>30
Total phosphorus	$g\ P/m^3$	<0.01	0.01-0.03	0.03-0.05	0,05-0,100	>0,100
Sulphates	g/m^3	<50	50-75	75-100	100-150	>150
Chlorides	g/m^3	<20	20-40	40-100	100-200	>200
Pb	mg/m^3	<0.1	0.1-0.5	0.5-2.0	2,0-5,0	>5,0
Fe	mg/m^3	<100	100-200	200-500	500-1000	>1000
Mn	mg/m^3	<50	50-100	100-200	200-400	>400
Al	mg/m^3	<20	20-50	50-200	200-500	>500

All leachate samples were collected in 5l polypropylene carboys, transported to the laboratory, stored at 4°C and analyzed within 3 days. The samples were collected, preserved, and analyzed according to the Standard Methods for the Examination of Water and Wastewater:

- Water quality - Sampling - Part 1: Guidance for designing sampling programs and techniques sampling BAS EN ISO 5667-1:2008*, BAS EN ISO 56671/Cor1:2008
- Water quality - Sampling - Part 3: Preservation I handling of water samples BAS EN ISO 5667-3:2019
- Water quality - Sampling - Part 6: Guidelines for water sampling from rivers and streams BAS ISO 5667-6:2017
- Water quality – Sampling – Part 11: Guidelines for groundwater sampling BAS ISO 5667-11:2010.

On-site measurements of pH and electrical conductivity (EC) were conducted during the sampling process using a digital pH meter and a digital EC meter. Specifically, for heavy metal analyses, samples were individually gathered in pre-washed polypropylene containers with a capacity of 100 ml. To prevent metal precipitation, the samples were acidified on-site. Physical-chemical parameters such as total dissolved solids (TDS), major anion such as chlorides (Cl-) of leachate and groundwater samples were analysed by titrimetric methods. Chloride is included in water quality assessment because it measures the extent of dispersion of leachate into the groundwater body [23]. Sulfates in groundwater samples were analyzed by nephelometric turbidity method.

Nitrates and determination of total organic carbon (TOC) in groundwater samples was performed using a spectrometer. The assessment of chemical oxygen demand (COD) was done by closed reflux titrimetry, while biochemical oxygen demand (BOD) was calculated by determining oxygen by Winkler titration for a preserved leachate sample. Heavy metals such as Mn Pb concentrations in leachate and groundwater samples were analyzed using an atomic absorption spectrophotometer.

RESULTS AND DISCUSSION

Discussion of physicochemical parameters of groundwater

Physicochemical parameters of groundwater quality in the investigated area were determined on water samples from 7 piezometers located as mentioned earlier. The results of testing water quality from piezometers PB-1, PB-2 PB-3, PB-4, PB-5, PB-6 and PB-7 are presented in Table 2. The obtained measurement results were compared with the wastewater discharge limits.

According to literature data, the pH of the leachate varies depending on age of the landfill [5]. As a rule, leachates from younger landfills show lower values (less or more acidic), while with age, the pH value moves towards basic values. As we can see from Table 2., pH values of the tested samples range from 7.35 to 7.60, indicating that leachate are in intermediate or semi-matured stages.

Table 2. Comparative presentation of test results and limit values prescribed by the Rulebook [21]

Parameter	Unit of measure	Result							LV–wastewater discharge limits
		PB-1	PB-2	PB-3	PB-4	PB-5	PB-6	PB-7	
pH	pH units	7.41	7.35	7.48	7.45	7.40	7.40	7.60	6.5-9.0
EC at 20°C	µS/cm	1326	469	2704	4610	1326	1201	5320	-
TDS	g/m^3	0.96	1.84	1.21	3.75	0.73	13.5	26.2	-
TSS	g/m^3	0.62	1.43	0.11	3.14	0.62	0.30	0.41	35
BOD_5	$g\ O_2/m^3$	1	4	7	6	1	2	50	25
COD	$g\ O_2/m^3$	8	12	16	12	6	6	72	125
Total nitrogen	$g\ N/m^3$	1.171	1.171	1.171	1.2	12.88	1.64	1.15	15
Total phosphorus	$g\ P/m^3$	0.027	0.1	0.089	6.80	0.022	0.14	1.36	3
Sulphates	g/m^3	0.0915	0.042	0.14	0.27	0.16	0.15	0.25	200
Chlorides	g/m^3	0.071	213	2.284	1668.5	1.07	142	2982	250
Pb	mg/m^3	n.d*	n.d	n.d	n.d	n.d	n.d	n.d	10
Fe	mg/m^3	n.d	40	821	1800	710	130	3350	2000
Mn	mg/m^3	10	380	1270	750	920	320	400	500
Al	mg/m^3	n.d.	10	40	n.d	80	80	250	1000

*n.d.-not detected

EC and TDS values are attributable to the presence of dissolved inorganic materials. Higher values of EC, especially on PB-7, agree with the highest values of dissolved inorganic materials in that sample (chlorides, Fe).

In the current study, BOD ranged between 1 and 50 g/m^3 and COD values ranged between 6 and 72 g/m^3. The BOD_5/COD ratios (0.13-0.69) confirmed the leachate semi-matured stages. Usually, landfill leachate with BOD_5/COD ratio less than 0.1 is considered to be toxic and are typical for younger landfills.

The chloride concentration in the leachate varied from 1000 to 8000 g/m^3 for young leachate samples and 1000–6,00 g/m^3 for stabilized samples. [23].

Heavy metals have significant impact on groundwater as well as surface water quality even if it found in traces amount. Pb was not detected in any sample examined, and the concentration of Al is below the limit value. The metal's concentration in the tested samples are mostly below the safe limit except for Fe (3350 mg/m^3 in PB7) and Mn (1270 mg/m^3 in PB3, 920 mg/m^3 in PB5 and 750 mg/m^3 in PB4). It can be noted that, especially in the case of Fe, the higher concentration values are in the piezometers located in the body of the landfill. Figure 3 shows Fe and Mn concentrations by sampling location.

Factors that affect groundwater quality are either natural or anthropogenic. The content of Fe and Mn in piezometer waters can be of natural origin and originate from the dissolution of rocks. Unfortunately, we did not have any data on the quality of groundwater in this area before the start of exploitation of this landfill. This is the reason why we cannot say with certainty that, especially in the case of enormously high concentrations of Fe and Mn, it is not possible to say with certainty that it is a purely anthropogenic factor.

However, bearing in mind that the highest concentrations of Fe were found in the samples from piezometers in the body of the landfill, we must not ignore the anthropogenic factor.

Moreover, the samples from shallow piezometer installed in the landfill body (PB-7) have a high value for the most parameters (EC, TDS, BOD_5, COD, chlorides, Fe, Al).

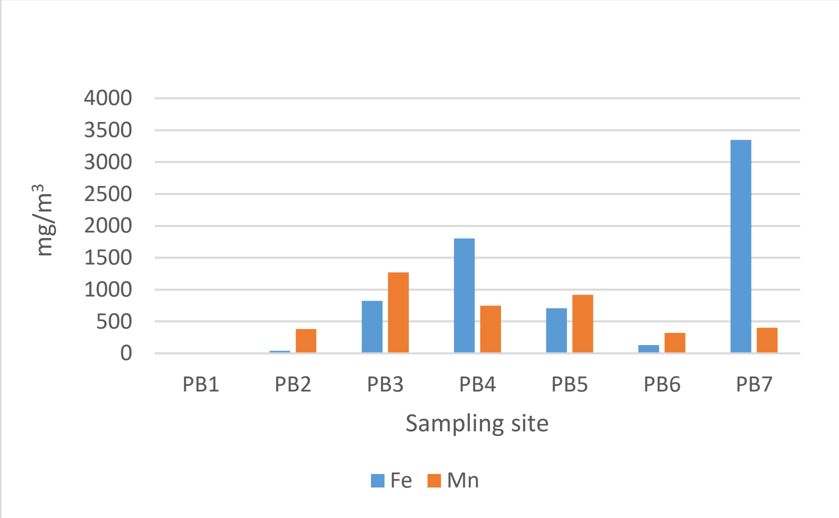

Figure 3. The concentrations of Fe and Mn by sampling location

Discussion of physicochemical parameter of surface water

Testing of the river Drina covered the basic groups of physical-chemical parameters of the water sample. The obtained measurement results were compared with the limit values, which are defined by the Regulation [22]. Results of the Drina River surface water analysis are shown in Table 3.

Table 3. Comparative presentation of the Drina River test results and the limit values prescribed by the Regulation [22]

Parameter	Unit of measure	Result/class of surface waters (Table 1.)		
		D1	D2	D3
pH	pH units	8.54/II	8.34/I	7.43/I
EC at 20°C	µS/cm	297/I	302.15/I	237/I
TDS	g/m^3	0.25/I	0.22/I	0.22/I
TSS	g/m^3	0.13/I	0.13/I	0.21/I
BOD_5	$g\ O_2/m^3$	1/I	2/II	2/II
COD	$g\ O_2/m^3$	4/I	5/I	5/I
Total nitrogen	$g\ N/m^3$	1.155/I	1.1569/II	1.1533/II
Total phosphorus	$g\ P/m^3$	0.01/II	0.012/II	0.013/II
Sulphates	g/m^3	0.39/I	0.20/I	0.14/I
Chlorides	g/m^3	0.11/I	0.14/I	0.11/I
Pb	mg/m^3	n.d./I	n.d./I	n.d./I
Fe	mg/m^3	20/I	20/I	30/I
Mn	mg/m^3	n.d./I	n.d./I	n.d./I
Al	mg/m^3	n.d./I	n.d./I	n.d./I

*n.d.-not detected

Based on the analyses performed and the results obtained, it can be observed that the Drina River has a good quality of the quality class II. The Tilić ada landfill is located in an area that does not meet the prescribed criteria for a location where waste can be deposited, considering the engineering-geological characteristics of the terrain and the fact that it is located in the very Drina River bed, and that it is in a narrow zone of sanitary protection of Tilić ada spring. Waste is disposed of without operational guidelines regulating this field.

The location is also not fenced off, and during the period of exploitation, the types and quantities of disposed waste were not recorded, nor the compaction and covering of waste with inert material. A special problem is the complete absence of emissions control and prevention of leachate filtering through soil layers, as well as degassing through the biothorn system.

In addition to negative impacts on human health and the environment, generally speaking, all uncontrolled landfills are characterised by the problem of unpleasant odours and a negative aesthetic effect, because they are often located in close proximity to roads and populated areas [24,25]. The problem with this landfill is the occurrence of torrential floods, which consequently lead to the lifting of and carrying away large amounts of waste during the withdrawal of water from the landfill site, to which border municipalities in the Republic of Serbia located downstream from the landfill site are exposed.

Based on the above, it can be concluded that the unsanitary way of waste management does not entail great costs during the period of use of the site for waste disposal, but on the other hand, apart from being contrary to the legal regulations, it entails complex environmental impact and significant costs during rehabilitation and recultivation. In order to minimise harmful effects, landfills as sources of pollution require closure and rehabilitation, but certainly one of the biggest obstacles in developing countries is the lack of financial resources.

CONCLUSION

This study investigated the impacts of the Tilić ada landfill in eastern Bosnia and Herzegovina on ground and surface waters, as typical example of wild and unorganized waste disposal sites. The landfill was active for a ten-year period, and the research presented in this study were conducted five years after the landfill was closed. Physico-chemical parameters of water quality were analysed on samples of underground water from the body of the landfill and immediately next to it, as well as on samples of surface water of the Drina River.

The results presented in the paper showed high concentrations of Fe and Mn in groundwater taken from piezometers located in the body of the former landfill. Although the origin of these metals in groundwater cannot be defined with certainty due to the lack of data from earlier times, anthropogenic influence cannot be excluded.

The results obtained from the analysis of water samples of the Drina River are classified into appropriate classes of water to which a certain value belongs, according to the Regulation on water classification and watercourse categorisation. Based on the analyses performed and the results obtained, it can be observed that the Drina River has a good quality of the quality class II.

Wild and unregulated landfills are one of the biggest potential polluters of underground and surface waters. The results presented in this article show that the impact of landfill leachates does not stop even after their closure, especially if no remedial measures have been taken.

Considering the performed analysis and categorisation of the landfill in question, based on the risk to the environment and human health, it is necessary to take steps towards its rehabilitation and recultivation, and prioritise planning and finding funds for these needs.In addition, hydrochemical monitoring is a necessary measure to control groundwater pollution in the research area.

The findings of this study can be used as initial data for the next phase of the research, which will investigate the possible further impact of the landfill's leachates, as well as ensuring permanent hydrogeological monitoring.

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