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Leachate of Landfill Smiljevići (Sarajevo, B&H) and their Environmental Status

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Abstract: Leachates are produced as a filtrate from waste landfills as a result of highly polluted waters. Organic substances that are mostly present are: phenolic compounds, halogen organic substances, oils and fats. The presence of nitrogen substances is significant, followed by phosphoric substances, sulfates, chlorides, and heavy metals. Sanitary landfill "Smiljevići", created 60 years ago, is placed on the hill area of Novi Grad municipality, Sarajevo. In that time area around landfill was very sparsely populated. Following the legislative, the leachate from the Smiljevići landfill does not have a good environmental status. On the other hand, according to global - typical values, contaminant concentrations are much closer to low than average values. The content of contaminants with long retention in nature and tendency to bioaccumulation is within legal limits. Most parameters that exceed the permitted values include usually biodegradable compounds that the stream of the river Bosnia can absorb better than the small ecosystem of the Lepenički creek. The problem is further aggravated bz the fact that the area surrounding the creek is now relatively densely populated, the stream has a small water capacity, especially in the summer, and the contaminants are mostly volatile. Finally, this problem also can be attributed to irresponsible activity of urban planning.

THE ORIGIN AND HAZARDOUS OF LEACHATE

The problem of human-caused waste dates back to early civilizations. In the Bible (Old Testament) and Ibn Khaldun's writings (14th century) the problems of waste disposal and resulting hazard (Morling, 2007) are mentioned. Accordingly, in the ancient time and today, the most common method to handle solid waste made by human action has been and still is to collect and place it in landfill. An inevitable problem created by the disposing the solid waste is the leachate left on the landfill that could causing significant water pollution. Leachate is produced as a filtrate from a municipal waste disposal site, which is created by the action of various factors, often combination of several of them. They are mostly caused by the passage of external water, most often by precipitation, through the layers of soil where the waste is deposited. Part of the water can evaporate, depending on the air temperature and the intensity of solar radiation. The share of individual sources certainly varies among different landfills, and often changes within the same landfill during the year, depending on the hydrological situation. Figure 1 presents a schematic picture of the water balance in a landfill (Bozkurt, 2000).

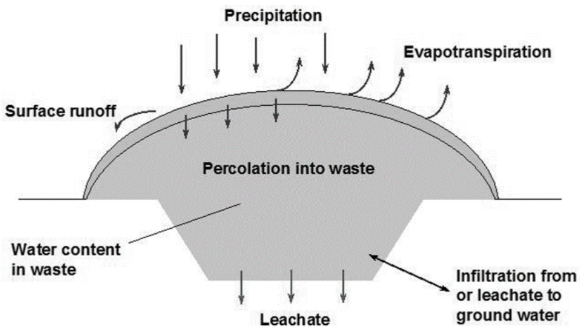

Figure 1. Scheme of leachate generate, (Bozkurt, 2000).

Unlike clearly visible waste that locally contaminates the soil with its presence, or an unpleasant odor from burning/evaporation that contaminates the air, landfill leachate is less noticeable and can contaminate a wider

Kolanka, 2013; Zornoza, Moreno-Barriga, Acosta et al., 2016). These compounds are formed by aerobic decomposition on the surface or anaerobic decomposition in deep deposits of waste in the soil that come into contact with water and create a leachate. Organic substances are mostly present: aromatic compounds (mainly phenolic compounds), halogen organic substances, oils and fats. The presence of nitrogen substances is significant, primarily and mostly in the form of ammonia compounds, then phosphoric substances, sulfates, chlorides and heavy metals (Reinhart and Grosh, 1998; Vaverkova, Elbl, Koda et al., 2020). Waste is decomposed in several stages, so the composition of the leachate is also affected by which stage of decomposition the waste contaminated the water. Factors of landfill water contamination include: method of deposition, depth of deposition, geological characteristics of the area where the waste is deposited, temperature, hydrological situation of the area such as precipitation, alluvium,and presence of groundwater (Christensen, Kjeldsen, Bjerg et al., 2001). The composition and concentration of substances present in leachate also changes with the age of the deposited materials. As landfills age, degradable organic substances slowly undergo anaerobic degradation, which results in leachate from older landfill having a more stable chemical composition and lower concentrations of harmful substances, especially organic substances. Young leachate mainly contains low molecular weight organic compounds often with oxygenated functional groups (e.g. carboxyl and alcoholic groups). Old leachate has organic compounds with a wide range of molecular weight, more complex structures with nitrogen, sulfur,and oxygen functional groups. Generally, leachate contamination gradually increases for the first 5 to 10 years and then decreases (Cheremisinoff, 1997; Calace, Liberatori, Petronio et al., 2001).

Leachates are one of the major problems in attempts to protect the population and the environment from the harmful effects and consequences of waste disposal. They directly threaten surface and underground water in the landfill area and surround (Wiesmann, Choi, Dombrowski, 2007). In order to eliminate or alleviate this problem, it is necessary to process the leachate to the quality of the effluent prescribed by legal acts, before discharging it into the natural recipient. Legislation is mainly based on EU directives regarding waste management. EU directives set strict requirements for the degree of purification and the maximum allowed discharge of harmful substances into natural watercourses. Application of modern purification methods (microfiltration, ultrafiltration, nanofiltration and reverse osmosis) are gaining more and more importance (Brennan, Healy, Morrison et al., 2016; Directive EU, 2018). In Bosnia and Herzegovina, the level of purification is determined according to the source of water discharge, and the allowed concentrations of certain harmful substances are prescribed by the “Regulation of the conditions for the discharge of waste water into the environment and public sewage systems” (Official Gazette FBiH, 2020). Regarding leachate from waste landfills, the contamination parameters prescribed by law that have to be determined are: pH, suspended substances, BOD (biological oxygen demand), COD (chemical oxygen demand), ammonia and total nitrogen, organic halogens, total organic carbon, total phosphorus, fats and oils and heavy metals.

SMILJEVIĆI LANDFILL

Sanitary landfill "Smiljevići", created in the 1960s, is placed on the locality of Buća Potok, municipality of Novi Grad, City of Sarajevo. All the waste that is collected in Sarajevo Canton is deposited at this landfill, and that includes: municipal waste, industrial waste, hospital waste, sewage sludge, waste as a result of traffic, slag and ash from waste incinerators and other waste. The area of the landfill is 122400 m². In the wider area of the Smiljevići landfill, the drainage of surface water and seepage takes place through the Lepenički creek, which flows directly into the Bosna river. Due to the configuration of the soil and tributaries, water level of the creek often varies during the year (KJKP Rad, 2013).

Leachate and rainwater produced in the area of the landfill must be treated in a treatment plant, thus achieving the required level of purification, and then discharged as a purified effluent of satisfactory quality into the natural recipient - Lepenički creek, in accordance with current regulations (Official Gazette FBiH, 2020). In the Smiljevići landfill, there is a pool infrastructure and there is a membrane bioreactor as a water purifier, but it has been out of order for a long time, so the leachate is discharged into the Lepenički creek without any treatment. Considering that the leachate from the Smiljevići landfill has not undergone any treatment, it can be considered as a general picture of the leachate of any landfill, regardless of the differences in the landfills (deposited material, soil structure and composition, environmental conditions, etc.).

THE STATUS OF LEACHATE FROM THE SMILJEVIĆI LANDFILL COMPARED TO OTHER LANDFILLS

In addition to the legal obligation of water purification, which is not carried out, landfill managing also includes monitoring water pollution parameters, which the landfill manager KJKP Rad performs regularly and publishes the results publicly on the website. Tables 1 and 2 show a cross-section of analysis results for the period of 2021.

Table 1. Contents of the main parameters of contamination of leachate from the Smiljevići landfill during 2021 (KJKP Rad, 2022), legal emission limit values and characteristic values with regard to the age of the landfill (Renou, Givaudan, Poulain et al., 2008; Knežević, Cukut, Dunović, 2012; Oreščanin, 2014; Mukherjee, Mukhopadhyay, Hashim et al., 2015; Brennan, Healy, Morrison et al., 2016).

Parameter	Landfill Smiljevići			Typical or global average values			Official Gazette FBiH 26/20, 96/20 emission limit values
	annual average	annual max.	annual min.	average	young landfill*	old landfill*	
pH	8.0	8.6	7.6	7.8	5.6 - 9.1	7.0 - 11.5	6.5 - 9.0
COD mg/L	1922	2640	1066	13000	1400 - 79000	100 - 10000	187.5**
BOD_5 mg/L	363	480	280	6000	90 - 27000	3 - 800	37.5**
ammonia mgN/L	541	609	399	1000	10 - 13000	1 - 1600	15.0**
total nitrogen mg/L	563	624	422	1000	70 - 13000	5 - 1700	22.5**
total phosphorus mg/L	4.7	7.4	3.3	30	5 - 100	5 - 10	3.0**
TOC mg/L	511	1830	210	6000	1500 - 20000	80 - 160	45.0**
conductivity µS/cm	11330	15570	7660	15000	3000 - 28000	2600 - 10500	-
flow m^3/day	318	380	260				
temperature °C	14.6	18.5	10.8				

* Renou et al. 2008 classify a young landfill as still active or closed max. 5 years ago, closed over 10 years ago is considered old landfill, closed 5 to 10 years ago is intermediate.
** values are according Appendix 19 and Article 22. of Official Gazette FBiH 26/20

Compared to the legal provisions (Official Gazette FBiH, 2020), the leachate from the Smiljevići landfill is considered very polluted because most of the parameters from Table 1 are above the permitted values, which is recorded in the report of the company that manages the landfill (KJKP Rad, 2022).

Compared to the typical - usual values of most landfills, the contamination parameters of the Smiljevići landfill are not high, they are mostly in the range of values of older landfills. Total phosphorus values are mostly below typical minimum values while pH is within average, with small oscillations within one pH unit, that is acceptable. Slightly higher values for total organic carbon (TOC) were detected. The values for all parameters, with the exception of conductivity, do not show large oscillations during the year, showing the characteristic of well-managed sanitary landfills. Conductivity is otherwise a parameter that could oscillates even regardless of the content of conducting substances, so change of temperature can change the conductivity of the same solutionIn addition change of pH has significant influence on conductivity.

The values of chemical oxygen demand (COD) and biochemical oxygen demand (BOD), as parameters that are the most important for assessing the contamination of landfill leachate, exceed the values established by country regulations (Official Gazette FBiH, 2020). However, they are below the average of typical values and are within the limits of older landfills. The reason for that is certainly the sanitary management of waste at Smiljevići landfill. The data from the table are at the global level where many landfills, especially in underdeveloped countries, are not subject to sanitary disposal measures, and the values of contaminants can be extremely high, especially COD and BOD. In addition to BOD and COD values, their ratio is an important parameter in assessing the landfill status, and it is a frequently used indicator of the degree of waste decomposition, i.e. stable values of leachate contaminants. Stable landfills are considered the ones with a BOD/COD ratio below 0.1 (Pohlard and Harper, 1986; Ehrig, 1989; Reinhart and Grosh, 1998). Hovewer this is a necessary but not sufficient parameter to consider that the waste is well decomposed, i.e. the content of contaminants in the leachate is stable (Barlaz, Rooker, Kjeldsen et al., 2002). In the case of the Smiljevići landfill, based on the annual average, the value is 0.19, so it cannot be considered that waste is well-degraded, regardless of the fact that the parameters values do not fluctuate significantly during the year. Otherwise according to Kjeldsen, Barlaz, Rooker et al. (2002) values of the BOD/COD ratio range from 0.02 to 0.8.

According to some authors ammonia compounds are the most significant long-term pollutants of leachate. The reason is the poor degradability of ammonia compounds in anaerobic - methanogenic conditions, which results in accumulation in leachate (Robinson, 1995; Burton and Watson-Craik, 1998; Christensen, Kjeldsen, Bjerg et al., 2001). Interestingly, in unregulated landfills, the ammonia content in leachate can be lower than usual because ammonia compounds are faster degradable in contact with air meaning that significant part of the ammonia evaporates into the atmosphere. Inversely, in the conditions of sanitary landfills potential-total ammonia has a low degree of elimination through air, mostly below 10%, depending on climate, temperature and general weather conditions (Barlaz, Rooker, Kjeldsen et al., 2002). In the leachate of the Smiljevići landfill, ammonia and total nitrogen showed lower concentrations compared to the global average or typical values. Both parameters are within the limits of leachate from older landfills, however they are much above the values allowed by regulations (Official Gazette FBiH, 2020) When compared to the typical values they are higher in concentrations and among the pollutants with the higher value compared to other pollutants. th. The ammonia/total N ratio in landfill leachate in the European Union is on average 0.75 (Brennan, Healy, Morrison et al., 2016). In the case of the Smiljevići landfill, that ratio is higher, and amounts is about 0.9 (Table 1), probably due to possible higher presence of quaternary ammonium compounds, which have been found to inhibit biological processes and prolong staying of ammonia, even in the treatment process of waste water (Tezel, 2009). The situation is similar with total phosphorus in both cases, when comparing with the typical values and legal provisions.

Hazardous phenolic compounds and organic halogens appear in landfill leachate mainly through the decomposition of deposited plastic materials. The share of these materials in the total balance of waste can be large, especially in landfills where sorting and recycling of plastic waste is not carried out. In recent years, the public company that manages the Smiljevići landfill has a program for separating plastic waste and sending it for recycling, but this partly depends on the conscience of the citizens who use the landfill. Otherwise, the content of these substances is increased in older landfills compared to younger ones, which was expected and recorded (Gibbons, Dolan, May et al., 1999), the reason being of course the slow decomposition of plastic substances. The content of phenols in landfill leachates listed in the literature ranges from 0.6 μg/L to 1200 μg/L, the same data for organic halogens ranges from 200 μg/L to 5000 μg/L (Albaiges, Casado, Ventura, 1986; Gintautas, Daniel, Macalady, 1992; Oman and Hynning, 1993; Robinson, 1995; Reitzel and Ledin, 2002; Baun, Ledin, Reitzel et al., 2004). Organic halogens in the leachate of the Smiljević landfill range from 370 to 690 μg/L, i.e. an annual average of 534 μg/L, while phenols are in the range of 30 μg/L to 80 μg/L, with an average of 45 μg/L (KJKP Rad, 2022). Both parameters have relatively low and stable values, i.e. they are much closer to the minimum values given in the literature as typical. Although the content of these substances in the leachates of most of the examined landfills is at the level of μg/L, it is slowly but steadily decreasing due to the increase in the recycling trend (Kjeldsen, Barlaz, Rooker et al., 2002).It is understood that the same trend will be with the Sarajevo landfill.

The content of heavy metals in the leachate of the Sarajevo landfill compared to typical values is much closer to the minimum values (Table 2), it is within the legal provisions (Official Gazette FBiH, 2020). Heavy metals could be very hazardous contaminants and their presence in landfill leachate must be investigated, but they rarely exceed the limit values in sanitary landfills, especially in areas where there are no large fluctuations in precipitation during the year, e.g. areas of the continental climate (Barlaz, Rooker, Kjeldsen et al., 2002). More often, high concentration of heavy metals appears in arid regions with sudden periods of precipitation, e.g. in subtropical areas (Siddiqi, Al-Mamun, Sana et al., 2022). Similar to the content of phenols and organic halogens, materials that are waste sources of heavy metals are more susceptible to recycling day by day, so it is to be expected that in the future there will be no significant hazardous concentrations of metals in the leachate of the Smiljevići landfill. On the other hand, the content of heavy metals can be influenced by several external factors: pH, the presence of complexing or precipitating substances, oxygen that oxidizes anions into more soluble forms of metal salts (e.g. sulfide into sulfate) etc., so there is always a possibility of changing the content, regardless of the quantity of deposited material, which are sources of heavy metal contamination.

Table 2. Typical values (global range) of content of heavy metals (Lin and Chang, 2000; Silva, Dezotti, Sant'Anna, 2004; Morling, 2007; Li, Yun, Li, et al., 2008; Salem, Hamouri, Djemaa et al., 2008; Kulikowska and Klimiuk, 2008; Oreščanin, 2014), phenols and organic halogens (Albaiges, Casado, Ventura, 1986; Gintautas, Daniel, Macalady, 1992; Oman and Hynning, 1993; Robinson, 1995; Reitzel and Ledin, 2002; Baun, Ledin, Reitzel et al., 2004) in leachate, values from leachate of Smiljevići landfill during 2021 (KJKP Rad, 2022) and legal emission limit values.

Parameter	Landfill Smiljevići (mg/L)			Global range	Official Gazette FBiH 26/20 emission limit values
	annual average	annual max.	annual min.		
As (mg/L)	0.021	0.03	0.01	0.001-0.38	0.05
Cd (mg/L)	0.017	0.03	0.01	0.015-0.13	0.05
Pb (mg/L)	0.043	0.06	0.03	0.002-3.49	0.10
Cr (mg/L)	0.07	0.10	0.02	0.012-0.748	0.15
Hg (mg/L)	<0.001	<0.001	<0.001	0.0002-0.012	0.005
organic halogens (μg/L)	534	690	370	200-5000	1000
phenols (μg/L)	45	80	30	0.6-1200	100

The leachate from the Smiljevici landfill consists of three waters:
1. Shallow drainage, the first collector, collects leachate from the multi-barrier protection through layers of mostly fresh disposal waste;
2. Deep drainage, the second collector, collects water from deep layers of waste under the multi-barrier protection, it is old waste that was not disposed of according to sanitary landfill rules.
3. Calota drainage, the third collector, leachate collected at the lowest points of the landfill, underground water that stream through the tunnels of the landfill;

The company that manages the landfill does not have the obligation to investigate individual waters, but the collective water that flows into the ecosystem of the Lepenički creek, but in the report of Prazina, Đug, Mahmutović et al. (2022) stated that water from shallow drainage from fresh waste is the most polluted. The same water is dominant in volume compared to the other two, and it is the main source of pollution of the creek. This is expected considering that it is known that fresh waste is the main source of landfill leachate pollution (Mukherjee, Mukhopadhyay, Hashim et al., 2015). Only nitrates are close in values in all three waters, and this is usual for this parameter when it is about leachates of young and older waste (Knežević, Cukut, Dunović, 2012).

FINAL CONSIDERATIONS

Leachates belong to highly polluted waters and represent a great danger to the environment. They can be environmentally more harmful than industrial waters because they have a variable and unpredictable quantitative and qualitative chemical composition, even in highly regulated waste management systems. A number of measures are implemented in order to protect the environment from the hazardous impact of landfills and its leachate: sanitary method of disposal, recycling, composting or incineration of biodegradable waste and treatment of leachate. The application of these measures in waste management in the Sarajevo region is not complete, hence the unsatisfactory status of leachate in the scope of legislative. Parameters whose concentration exceeds legal limits (COD, BOD, ammonia, total nitrogen, TOC, total phosphorus) are mainly generated in leachate from the biodegradable waste. Burning and/or composting this type of waste, which is a trend in the European Union, the concentration of these parameters in the leachate of the Smiljevići landfill would be significantly reduced, probably under legal limit values even without treatment of water. Compounds which make up these parameters mainly are biodegradable so the greater ecosystem of river Bosna is able to accept those without serious hazardous consequences. However, the hazard of these parameters is strongly expressed in the area of the small ecosystem of the Lepenički creek, which has become densely populated in the last 20 years, so the responsibility for the exposure of the population to contaminants can also be attributed to the municipality authorities that allowed the urbanization of this area. Parameters that have a high retention in nature and the potential for bioaccumulation in the living organism (heavy metal and organic halogens) are within the limits of legal provisions, which probably indicates good management in that segment of measures that are applied - sanitary waste disposal and recycling. This is positive data, as these substances are potentially the most dangerous contaminants of leachate for the local and wider ecosystem, and their elevated presence in leachate is difficult and long term solvable, in the short term they cannot be eliminated without the use of a water treatment system. On a global scale, the parameters of the leachate contamination of the Smiljevići landfill are closer to the lowest limits than the average values, even those parameters with the highest values that are above the limit values prescribed by law. But this is certainly not a reason for lack of concern and passivity because the leachate from the Smiljevići landfill flows through a relatively large settlement that is not built illegally but urbanized and the population is partially agriculturally active. Another reason which enhance the mentioned problem is small capacity of the Lepenički creek, whose water mass is significantly contributed by the leachate of the landfill. This problem especially increases during the summer period, with high temperatures and drought the water mass of creek becomes significantly reduced, even the air in this area becomes polluted due to the evaporation of contaminants.

Certainly, the best solution is to close the existing Smiljevići landfill because it is quite old, around 60 years. Another reason for the need to close the landfill is the fact that when the landfill was put into use, the surrounding of the landfill and the surrounding of the leachate stream were very rarely populated, today it is the opposite.

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Summary/Sažetak

Procjedne vode koje nastaju kao filtrat s odlagališta komunalnog otpada, spadaju u vrlo onečišćene vode. Najviše su zastupljene organske tvari: fenolni spojevi, halogene organske tvari, ulja i masti. Značajna je prisutnost azotnih tvari, najvećim dijelom u obliku amonijačnih spojeva, zatim fosfornih tvari, sulfata, hlorida i teških metala. Sanitarna deponija "Smiljevići", nastala prije oko 60 godina, nalazi se na brdskom području općine Novi Grad, Sarajevo, koje je u tom periodu bilo vrlo rijetko naseljeno. U okviru zakonskih akata koji propisuju maksimalne koncentracije zagađujućih tvari, procjedne vode deponije Smiljevići nemaju dobar okolinski status. S druge strane, u svjetskim okvirima (tipične vrijednosti), koncentracije kontaminanata su mnogo bliže niskim nego prosječnim vrijednostima. Sadržaj kontaminanata sa dugom retencijom u prirodi i sklonosti bioakumulaciji (teški metali, organski halogeni i sl.) su u zakonskim granicama. Parametri koji prelaze dozvoljene vrijednosti uglavnom čine biorazgradive komponente koje širi ekosistem rijeke Bosne uglavnom može absorbovati, no hazardne su za okolinu primarnog toka - područje Lepeničkog potoka. To je tok malog kapaciteta vode pa je problem dodatno pogoršan, osobito ljeti jer su kontaminanti obično volatilni. Ovo područje je sada relativno gusto naseljeno, stanovništvo je djelimično poljoprivredno aktivno pa se ovaj problem može pripisati i lošoj i neodgovornoj djelatnosti iz oblasti urbanizma.