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The Impact of Leachate on the Quality of Surface and Groundwater and Proposal of Measures for Pollution Remediation

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Abstract

Direct discharge of municipal and industrial waste waters and leachate (originating from the illegal landfills) into recipients without prior purification is unfortunately very common practice in the region of northern Kosovo. In addition, irresponsible and incorrect selection of sites for industrial and municipal dumps, which are often located in vicinity or on actual river banks, contributes significantly to environmental pollution. Analysis of the leachate from such sites was done by direct sampling and by using TCLP (Toxicity Characteristic Leashing Procedures) method. Based on the analysis of physicochemical parameters of the filtrated water from the sites Žitkovac, Grabovac and Balaban and analysis of the samples of surface water and groundwater from the site Grabovac, possible steps for removal and reduction of the existing pollution were proposed. Potential permanent solution in form of purification of municipal and industrial waste waters as well as leachate from illegal landfills was suggested. The potential implementation of remediation with a unified system for water purification, by using Membrane Bio Reactor (MBR), which includes the process of stabilization/solidification of a residual sludge, would have as an end product a neutral powder material completely safe for the environment, suitable for a variety of applications.

Keywords

Wastewater Treatment, Membrane Bio-Reactor, Solidification, Neutral, Environment

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1. Introduction

The attitude of a society towards the environment is a good indicator of its awareness and a level of development. Scattered, illegal landfills represent one of the largest pollutants of surface and ground water, soil and atmosphere. In the region of northern Kosovo, majority of legal and illegal dumps are located on the alluvial deposits of rivers, near villages, and directly affect the spread of pollution. The environment is additionally threatened by a large number of industrial landfills, which originate from the process of Mining-Metallurgical-Chemical Plant "Trepča"—RMHK Trepča. This region has the largest lead-zinc ore deposits in Kosovo and among the largest in Europe, which have been exploited with interruptions from XIII-XXI century. RMHK Trepča lead and zinc plant was established by nationalization of property after the Second World War and it is characterized by inappropriate choice of landfill sites (mostly located on the banks of the rivers Sitnica and Ibar) and a lack of appropriate collectors for wastewater treatment. Heavy metals are known to have adverse effects on the environment and human health [1]. Highly reactive and often toxic at low concentrations, they may enter soils and groundwater, bioaccumulation in food webs, and adversely affect biota, [2] [3], also, they can cause irreversible changes in the body especially in the central nervous system [4]. High levels of heavy elements such as lead (Pb), zinc (Zn), copper (Cu), arsenic (As), cadmium (Cd) and antimony (Sb) are usually found in surface soils of areas affected by mining and basic metallurgical or smelting activities, metal factories, traffic emissions and intense agricultural practices [5]-[8]. Soil contamination by heavy elements therefore represents a worldwide environmental concern mainly because these elements can be transferred to the hydrosphere and biosphere, thereby posing a hazard to human health. Consequently the mobility and bioavailability of heavy elements in soil play an important role in the uptake of these contaminants by vegetation and animals [9]. Furthermore, filtrate water, atmospheric origin water constituent, as well as the water that is produced in the body of the landfill, form a medium in which, in the process of decomposition and degradation pollutants are released and thus define the quality of the landfill leachate (filtrate water from the landfill). In addition to the physicochemical processes that take place and external environmental conditions, composition of the landfill leachate is influenced by a host of other variable factors such as the composition of the waste, the thickness of the landfill body, fluid migration path and the possibility of intermediate layers that adsorb or absorb and minimize pollution. Formed filtrate from informal, illegal dumps is in direct contact with surface water and groundwater [10] [11], except in the case of sanitary landfills, whose standards are in accordance with the Law on Waste Management, the Law on Environmental Protection and standards and the EU regulations on waste management. Pollution of soil, surface and ground water [12] [13] may directly and indirectly enable organic and inorganic polluting substances and heavy metals to enter into the food chain [14] [15] and seriously endanger the health of people [16] [17]. The chemical composition of the leachate reflects the sum of all processes occurring within the waste body, affecting especially the organic fraction [18]-[20]. Degradation processes of municipal landfills can be globally divided into four phases. The first three phases last for a relatively short period, 180 to 200 days. In the fourth, methane phase, landfill matures i.e. comes to balancing anaerobic process in which a relatively equal amounts of CO_2 and CH_4 are released [21]-[23]. The objective of this study was to evaluate the effects of pollutants from the leachate of informal dumps on surface and ground waters in their surroundings and in view of the complexity of the problem (due to a permanent emission of the pollutants from the leachate of illegal waste landfills as well as from the industrial waste deposits, that impact the quality of surface and ground waters) to propose possible steps for reduction of the existing pollution and prevention of future pollution.

2. Materials and Methods

2.1. Study Area

Samples of the leachate originating from the municipal waste deposits were analyzed along with samples of surface and ground waters. The leachate samples were taken at the end of February 2014, while the surface and ground water samples were taken in intervals: April, 2013, September, 2013, and February 2014. In order to determine leachate characteristics with different stage of decomposition, the samples were taken from different locations and from different vertical column depth. Sampling information is given in **Table 1** and in **Figures 1-3**, together with the maps with marked locations from where the samples were taken.

2.2. The Leachate and Water Sampling

The leachate water samples from the location Balaban intended for the basic and detailed physical and chemical

Figure 1. Map of site Balaban landfill with marked sampling points [24].

Figure 2. Map of site Grabovac, the informal landfill with marked sampling points [24].

Table 1. Points of sampling leachate to sites of selected landfills.

Locality	Sampling points	Description of sampling	Depth
Landfill Balaban	Monitoring wells (Piezometer (P1))	40 m opposite the entrance to the landfill	3 m
	Monitoring wells (Piezometer (P2))	150 m to the right of the entrance to the landfill, with landfill body near the security fence	3 m
	Schacht (Š3)	Schacht leachate, 1.20 m to the right of the entrance facility for control	3 m
	Sewer (SK4)	Channel for collection leachate S1, S2, S3, S4, 50 m from the entrance	4 m
Unregulated landfill in Grabovac on the bank of river Ibar	The sample (G1)	Central part of unregulated landfill in Grabovac	1 m
	The sample (G2)	Slope unregulated landfill in Grabovac on the right bank of the river Ibar	1.5 m
Unregulated landfill in Žitkovac	The sample (Ž1)	The Northeast part of the landfill (3 m from the edge)	2 m
	The sample (Ž2)	The Southeastern part of the landfill (3 m from the edge)	2 m

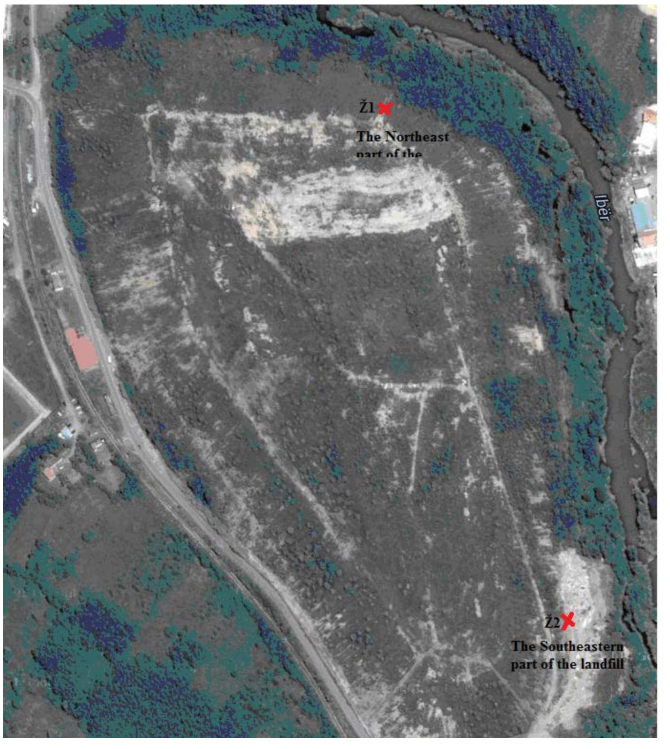

Figure 3. Map of site Žitkovac, the informal landfill with marked sampling points [24]. Note: Satellite images landfills shown in **Figures 1-3**. (are downloaded from the website www.google.rs/maps) only show sampling points, because they are old and do not show the current enormous amount of waste in the studied sites.

analysis, were collected in the plastic bottles of 2 dm^3, while the wastes from illegal landfills on the locations Grabovac and Žitkovac were collected into the plastic bags in order to simulate washout wastes and leaching waters in the laboratory (in regard to TCLP method-Toxicity Characteristic Leashing Procedures) [25]. All of the samples, after collection, were marked in according to the procedure of sampling and were stored in the refrigerator at about 4˚C until the sample preparation for the analysis. **Figure 4** shows the sampling of leachate at the landfill Balaban.

For the purpose of testing the leachate from the illegal dumps the samples were taken from dumps in Grabovac and Žitkovac, and TCLP method was applied. Nowadays there are a number of methods that can be used to simulate the process of leaching of waste in a landfill or in the environment in order to assess the content of pollutants, or to determine the characteristics of the leachate in contact with the test material. Depending on the alkalinity and buffering capacity of waste two different extraction solutions were used in the applied TCLP method: solution 1 and 2 with pH of 4.92 and 2.88, respectively. In the first step, pH levels of the samples were determined in order to select the appropriate extraction solution for leaching. The solution was mixed together with 5 g of crushed and homogenized waste sample (with diameter less than 1 mm) and then mixed for 5 minutes with 96.5 cm^3 of distilled water. In line with the US procedure EPA [25], for the pH of the obtained sample solution lower than 5, extraction solution 1 was used for leaching whereas in case of the pH higher than 5, extraction solution 2 was used. The samples were crushed and sieved, so for the analysis only fractions smaller than 9.5 mm were used. After that, the extraction solution was added according to the procedure [26] at a 1:20 mix of sample and solvent. For example, in this particular case a TCLP jug contain 100 g of sample and added solution to 2 dm^3 (liter); (prepared of solution: Extraction fluid 1: Add 5.7 ml glacial CH_3CH_2OOH to 500 ml of reagent water, add 64.3 ml of 1N NaOH, and dilute to a volume of 1 dm^3 (liter). When correctly prepared, the pH of this fluid will be 4.92 + 0.05. Extraction fluid 2: Dilute 5.7 ml glacial CH_3CH_2OOH with reagent water to a volume of 1 dm^3 (liter). When correctly prepared, the pH of this fluid will be 2.88 + 0.05). The leachate mixture is sealed in extraction vessel for general analytes, and tumbled for 18 hours to simulate an extended leaching time in the ground. The solution was subsequently separated by filtration through a glass fiber filter (pore size 0.7 microns) under vacuum (suction) and then analyzed.

P1

P2

Š3

SK4

Figure 4. Sampling of leachate at the landfill Balaban.

2.3. Chemical Analysis

Qualitative and quantitative analysis of physical-chemical parameters of the leachate and waters was carried out using multiparameter equipment for water sampling Multi 340i Wissenschaftlich-technische Werkstatten GmbH (for the measurement and the determination of several parameters: pH/oxygen, conductivity and temperature), Dissolved oxygen (DO), biochemical oxygen demand (BOD) were measured with Winkler aside method. Most of parameters leachate are expressed in milligrams per cubic decimeter (m g/dm^3), except pH, EC (µ S/cm) and temperature (˚C). As soon as the samples of leachate and water were brought to the laboratory they were preserved with 1ml of concentrated nitric acid (HNO_3), filtered and stored in dark at an ambient temperature until microwave acid digestion following [27]. The digestion of 50 ml was performed with 4 - 5 ml HNO_3 65% and 1 ml HCl 35%. Total metal samples were filtered through 0.45 µm nylon filters after digestion. The concentrations of Cr, Ni, Pb, Cd, Zn, Hg, Fe, and Cu were measured using inductively coupled plasma-optical emission spectrometry (ICP-OES Optima 2100 DV) in accordance with the standard method [28] and are expressed in nanograms per cubic decimeter (µg/dm^3). Three independent replicates were performed for each sample and blanks were measured in parallel for each set of analyses using the same procedure. All reagents used in this work were analytical or HPLC grade and used without any further purification. Indicator of the loading with biodegradable organic water pollutants or water activity is expressed as an Index of Phosphates Activity (IPA—(µmol/s/dm^3) p NP, 30˚C). Based on this index the water quality is characterized according to the categorization [29] [30].

3. Results and Discussion

3.1. The Impact of Leachate on the Quality of Surface and Groundwater

General and detailed analysis of physical and chemical properties of the leachate, surface and ground waters included: determination of the dissolved oxygen concentration, hydrogen ions concentration (pH values), electrical conductivity and hardness of water, as well as determination of parameters of the organic pollution such as chemical oxygen demand and biological oxygen demand in five days (BOD_5). The results of the basic and detailed physical and chemical analysis of the samples of landfill leachate Balaban are given in **Table 2**.

Based on the data presented in the **Table 2**, regarding the quality of the leachate from the landfill Balaban, it can be concluded it is extremely polluted and cannot be discharged into the recipient without prior treatment.

Table 2. Physicochemical parameters of the leachate from the landfill Balaban.

Nr.	The investigated parameters	Identification methods	unit of issue	Monitoring wells of leachate			
				P1	P2	Š3	SK4
1.	The water temperature on the ground	P-IV-I[3]	˚C	7	10	27	13
2.	The air temperature		˚C	4	7	4	4
3.	Electrical conductivity	P-IV-II[3]	µS/cm	2660	8550	20,800	19,540
4.	pH value	P-IV-6 (A)[3]	-	7.91	7.68	8.59	8.65
5.	The concentration of dissolved O_2	P-IV-12 (B)[3]	mg/dm^3	7.22	7.11	0.19	0.21
6.	Biological oxygen demand BOD_5	P-IV-13[3]	mg/dm^3	26	32	784	1275
7.	Consumption of $KMnO_4$	P-IV-9a[3]	mg/dm^3	3.73	6.76	446.82	459.46
8.	Hardness of water	LCK 327	^{0}dH	4.68	12.9	74.3	72.4
9.	Nitrites	8507	mg/dm^3	0.010	0.011	0.310	0.400
10.	Nitrates	8171	mg/dm^3	0.2	<0.1	40	37
11.	Phosphorus	LCK 349	mg/dm^3	<0.15	<0.15	13.6	6.85
12.	Potassium	8049	mg/dm^3	3.7	3.6	515.78	491.85
13.	Chrome	LCK 313	mg/dm^3	<0.03	0.036	1.08	0.306
14.	Iron	P-V-17/B[3]	mg/dm^3	7.395	7.477	18.320	10.095
15.	Zink	ISO 8288/86	mg/dm^3	0.619	1.367	1.444	1.517
16.	Cadmium	LCK 308	mg/dm^3	<0.02	<0.02	0.12	0.19
17.	Nickel	ISO 8288/86	mg/dm^3	3.947	3.998	4.613	5.029
18.	Calcium	P-V-22/B	mg/dm^3	15.152	58.542	51.613	58.150
19.	Magnesium	P-V-22/B	mg/dm^3	11.290	20.829	293.284	281.432

Very low concentration of the dissolved oxygen in all tested samples, within the interval of 0.19 - 7.22 mg/dm^3, point to the burdening of these leachate with organic matters, while very high conductivity range (2660 - 20,800 µS/cm) suggests that the total content of the dissolved salts is very high. Considering that extremely high heavy metals concentrations (Ni, Fe, Zn) were detected which are several times higher than Maximum allowed concentration (MAC) stated in the Directive on waters classification of the Republic of Serbia [30]-[32], these waters are considered extremely polluted and categorized as out of the legal limits.

Physical and chemical characteristics of the leachate waste from the illegal dump in Grabovac, obtained after treatment of waste samples using TCLP method are given in **Table 3**.

Based on the data shown in the **Table 3**, it can be concluded that the leachate from this illegal landfill in Grabovac, are detected of high heavy metals concentrations (Fe, Zn, Cd), also loaded with organic matter, and that the concentration of the dissolved salts is very high (the concentration of dissolved O_2 0.15 - 0.18 mg/dm^3; electrical conductivity values were 820 - 918 µS/cm respectively). Although the results of physical and chemical testing show less pollution of the leachates compared to the collected leachates Š and SK from the closed waste landfill Balaban, this is not conclusive result. Due to leaching of the deposited waste with atmospheric precipitation and direct leak in the environment and the neighboring river Ibar, it was necessary to analyze surface and ground waters in the vicinity of the landfill (**Table 5**).

The tailings deposit of the Mining-Metallurgy Chemical Company "Trepča" in Žitkovac (north of the town Zvečan), is located on the left bank of the river Ibar. It occupies surface of 150,000 m^2 and was originally 20 m high. Although initially flotation tailings from the mineral processing in Zvečan, originating from processing of ores from mines "Stari Trg", "Belo Brdo", "Crnac", "Ajvalija", "Badovac", "Kišnica" and "Novo Brdo") were deposited there, in recent years it has become an illegal deposit of the municipal waste of the municipalities of

Table 3. Physicochemical parameters of the leachate from the illegal waste landfill in Grabovac on the bank of the river Ibar.

Nr.	The investigated parameters	Marking methods	unit	Leachate water	
				G 1	G 2
1.	Temperature of waste in the field	P-IV-I[3]	˚C	7	7
2.	The air temperature		˚C	3	4
3.	Electrical conductivity	P-IV-II[3]	µS/cm	820	918
4.	The pH value	P-IV-6 (A)[3]	-	7.39	7.41
5.	The concentration of dissolved O_2	P-IV-12 (B)[3]	mg/dm^3	0.18	0.15
6.	Biological oxygen demand BOD_5	P-IV-13[3]	mg/dm^3	198	201
7.	Chemical oxygen demand	P-IV-10[3]	mg/dm^3	376	382
8.	Hardness of water	LCK 327	˚dH	71.3	70.4
9.	Nitrites	8507	mg/dm^3	0.037	0.040
11.	Phosphorus	LCK 349	mg/dm^3	5.61	5.85
12.	Potassium	8049	mg/dm^3	45.78	39.85
13.	Chrome	LCK 313	mg/dm^3	1.03	0.96
14.	Iron	P-V-17/B[3]	mg/dm^3	13.124	10.095
15.	Zink	ISO 8288/86	mg/dm^3	1.024	1.307
16.	Cadmium	LCK 308	mg/dm^3	0.24	0.19
17.	Nickel	ISO 8288/86	mg/dm^3	0.643	0.529
18.	Calcium	P-V-22/B	mg/dm^3	47.313	51.150
19.	Magnesium	P-V-22/B	mg/dm^3	284.384	279.432

Zvečan and Mitrovica North. Physicochemical characteristics of the leachate from the waste deposited in the illegal dump in Žitkovac, obtained after treatment of waste samples by using TCLP method are presented in **Table 4**.

The observed very low concentration of the dissolved oxygen in the investigated samples (**Table 4**) from illegal deposit in Žitkovac (0.15 and 0.16 mg/dm^3) and high values of conductivity (1120 - 1870 µS/cm) indicate also loading of these leachates with organic matters and dissolved salts. Furthermore, concentrations of some heavy metals, similarly to some of the previously investigated locations are above the legal limits of MAC. Considering the fact that the municipal waste is deposited on the former tailings deposit from mineral processing plants, with direct leaking of the leachate in the environment and the river Ibar, some special attention was paid to investigation of presence of heavy metals in surface and ground waters running down flow from Grabovac, Žitkovac and deposit Balaban. The results of these investigations are presented in the **Table 5**.

Constant and complex pollution of the area to which the north part of Kosovo belongs, due to the presence of a large number of industrial landfills and illegal municipal solid waste landfills located in close vicinity or on the banks of the local rivers and the determined presence of heavy metals from industrial plants and landfills, and other hazardous and noxious substances, which are very high on the list of pollutants in this area due to their toxicities, has initiated the examination of the content of heavy metals in the river Ibar and groundwater's downstream from Grabovac and the landfill site Balaban (Pb, Cr, Ni, Cd, Zn and Hg). The test results are given in **Table 5**.

Conductivity is an important and fast method that measures the total dissolved ions and is directly related to total solids. The electrical conductivity (EC) of water samples in the Ibar river and groundwater ranges from 224 to 798 µS/cm. Conductivity is also affected by temperature. The warmer waters have higher values of the conductivity. The water temperature is also a significant parameter that controls the inborn physical qualities of the

Table 4. Physicochemical parameters of the leachate from the illegal waste landfill in Žitkovac.

Nr.	The investigated parameters	Marking methods	Unit	Leachate water	
				Ž1	Ž2
1.	Temperature of waste in the field	P-IV-I[3]	˚C	9	8
2.	The air temperature		˚C	4	4
3.	Electrical conductivity	P-IV-II[3]	μS/cm	1120	1870
4.	The pH value	P-IV-6 (A)[3]	-	7.95	8.01
5.	The concentration of dissolved O_2	P-IV-12 (B)[3]	mg/dm^3	0.15	0.16
6.	Biological oxygen demand BOD_5	P-IV-13[3]	mg/dm^3	438	572
7.	Chemical oxygen demand	P-IV-10[3]	mg/dm^3	212	172
8.	Hardness of water	LCK 327	˚dH	65.3	72.4
9.	Nitrites	8507	mg/dm^3	0.227	0.234
11.	Phosphorus	LCK 349	mg/dm^3	12.6	6.85
12.	Potassium	8049	mg/dm^3	437.62	399.31
13.	Chrome	LCK 313	mg/dm^3	0.39	0.43
14.	Iron	P-V-17/B[3]	mg/dm^3	13.25	15.90
15.	Zink	ISO 8288/86	mg/dm^3	1.51	2.13
16.	Cadmium	LCK 308	mg/dm^3	0.03	0.05
17.	Nickel	ISO 8288/86	mg/dm^3	3.561	4.181
18.	Calcium	P-V-22/B	mg/dm^3	49.28	53.75
19.	Magnesium	P-V-22/B	mg/ dm^3	187.2	202.4

Table 5. Physicochemical parameters and concentration of heavy metals in the surface water at the sites of Grabovac and Balaban and in well water at the sites of Grabovac.

The investigated parameters	unit	Surface water						Well water			The quality of water by classes based on parameter values				
		Grabovac Apr./13	Grabovac Sep. /13	Grabovac Feb. /14	Balaban Apr./13	Balaban Sep./13	Balaban Feb./13	Grabovac Apr./13	Grabovac Sep./13	Grabovac Feb./13	I	II	III	IV	V
Scent		the smell of rot	the smell of rot	the smell of rot	the smell of rot	the smell of rot	the smell of rot	without	without	without					
Colors	NTU	2.1	2.0	2.2	2.1	1.9	2.1	no color	no color	no color					
Suspended solids	mg/dm^3	21	18	19	9.5	14	11	4	4	4	25	25	-	-	-
The water temperature in the field	˚C	12	16	9	8	15	6	9	10	9					
The air temperature	˚C	10	15	7	7	14	3	10	15	8					
Electrical conductivity	mg/dm^3	442	508	451	442	395	369	236	224	230	<1000	1000	1500	3000	>3000
The pH value	mg/dm^3	7.9	8.1	8.2	7.8	8.1	7.9	7.8	7.9	7.9	6.5 - 8.5	6.5 - 8.5	6.5 - 8.5	6.5 - 8.5	<6. or >8.5
The concentration of dissolved O_2	mg/dm^3	5.4	5.3	5.4	6.1	6.3	6.2	8.9	8.7	8.8	-	7	5	4	< 4

Continued

Biological oxygen demand BOD_5	mg/dm^3	7.2	6.9	7.1	5.8	5.8	5.7	3.9	4.2	4.5	-	5	7	25	> 25
Chemical oxygen demand	mg/dm^3	17.3	16.5	17.2	12.8	13.7	12.6	5.9	5.5	5.7	5	10	20	50	>50
Hardness of water	^{0}dH	198	189	170	201	203	199	175	178	175					
Nitrates	mg/dm^3	1.2	0.8	0.65	0.39	0.47	0.34	0.02	0.02	0.02	0.01	0.03	0.12	0.3	>0.3
Phosphates	mg/dm^3	0.1	0.07	0.06	0.1	0.08	0.05	0.03	0.03	0.02	-	0.1	0.2	0.5	>0.5
Sulfates	mg/dm^3	95.50	94.66	88.45	81.74	76.63	75.41	43.72	56.80	59.67	50	100	200	300	>300
Chloride	mg/dm^3	18.4	18.3	18.1	18.3	18.7	18.1	10.3	10.5	10.1	50	100	150	250	>250
Chrome	$\mu g/dm^3$	0.09	2.58	2.76	0.28	0.27	2.61	0.05	< 0.05	0.05	25	50	100	250	>250
Nickel	$\mu g/dm^3$	1.8	6.59	3.12	0.10	0.09	3.28	0.18	0.16	0.17	20	25	50	100	>100
Lead	$\mu g/dm^3$	193.22	148.31	132.15	54.68	39.90	27.62	12.97	18.85	13.81	50	100	100	200	>200
Cadmium	$\mu g/dm^3$	5.0	17.68	0.10	0.06	0.06	13.12	0.06	0.05	0.05	5 and <	30 and <	100 <	200	>200
Zink	$\mu g/dm^3$	3.24	7.24	3.21	0.06	< 0.05	0.81	0.08	0.09	0.10	10	100	200	500	1000
Mercury	$\mu g/dm^3$	0.01	0.35	0.43	0.03	0.02	0.37	5.98	4.99	5.48	5	5	<10	10	>10
Iron	$\mu g/dm^3$	94	75	80	74	75	56	47	40	38	100	200	500	1000	2000
Copper	$\mu g/dm^3$	112	124	310	85	57	65	18	22	19	10	100	200	500	1000
Calcium	mg/dm^3	64.7	67.6	65.8	85.6	70.7	69.2	60.5	57.3	67.4	-	-	-	-	-
Magnesium	mg/dm^3	17.4	19.2	18.7	26.5	21.4	20.8	14.1	18.4	11.9	-	-	-	-	-
Phenols	mg/dm^3	0.008	0.006	0.004	0.006	0.005	0.004	0.002	0.001	0.002	0.001	0.001	/	/	/

water. In this study, the surface water temperature ranged between 6˚C and 16˚C, while the temperature of the ground water was in the range of 9˚C - 10˚C. The pH is a measurement of the acidity or basic quality of water. Extremely high or low pH levels have a significant effect for most aquatic organisms. The pH of the water changes even slightly. In this study the pH levels range from 7.8 to 8.2. Turbidity is another important parameter of water pollution. The present study shows the turbidity of river in the range of 19 - 98 NTU, while the turbidity of groundwater is 1 NTU. The highest desirable limits for turbidity are 5 NTU and maximum permissible limit 50 NTU [29] [32]. In all locations the value of turbidity present are not within the desirable limit. It reveals that the river pollution is high. Dissolved oxygen in water is an essential and important parameter for aquatic life. Deficiency of dissolved oxygen comes as a result of anaerobic decomposition of organic waste [30] [33]. DO in our study ranges from 5.3 to 8.9 mg/dm^3. Biological Oxygen Demand (BOD) is an important parameter used to assess the organic pollution. When surface water is having BOD more than 5 mg/dm^3, it is unsafe for domestic use. In our study, all samples of surface water with BOD above permissible limits (5.7 - 7.2 mg/dm^3). Therefore, a high value of BOD confirms the present situation of highly contaminated river Ibar.

Based on the results presented in **Table 5** and the references [30]-[32] and exceeding limits of certain physical and chemical parameters (concentration of dissolved O_2, COD, BOD_5, Sulphates, Nitrites and Phenols) and considering determined extremely high concentration of lead and copper in three of the samples of surface water and cadmium in the two, the surface waters of the investigated locations are classified in the III, IV or V class, which designates poor quality of the water, unusable for irrigation of agricultural crops. According to reference [34] water quality of the river Ibar is decreasing. What is particularly worrying is that increased contents of mercury and phenols were found in well water at the location in Grabovac, which is used for drinking. Activity in thermal power plants, coal mines, and impact of existing ash, industrial and municipal landfills, pesticides, electrical equipment (batteries, lamps, switches) smelting and fossil-fuel combustion, result in wastewater pollution, that are subsequently discharged directly into the rivers Sitnica and Ibar without any prior treatment [35]. Existence of such complex pollution requires immediate action, especially given that polluted water can cause serious consequences for human health, plants and animals, and the whole environment. Advanced wastewater treatments such as classification, treatment, flocculation and filtration of the discharged waters would enable their re-use and minimize the pollution.

Comparing the results of the analysis of the leachate collected in schacht (S3) and sewer (SK4) in the landfill Balaban, where there is no direct contact between leachate of landfill and surface and ground waters in the environment, with the results for leachate of illegal and informal dumps (Grabovac and Žitkovac) and analysis of the investigated parameters of surface and ground waters (**Table 5**), it can be concluded that the waste, disposed of in this and other similar locations, is washed by atmospheric precipitation that leach toxic and harmful substances and release the leachate into the immediate environment: soil, surface water and groundwater, and agricultural crops, plants and animals and finally, the human body. Pathways of metal transfer in the terrestrial food web along with the selected references for each pathway [1] are shown in **Figure 5**.

3.2. The Measures for the Rehabilitation of Existing and Preventing Future Pollution

As a first step it would be necessary to find an adequate location for the sanitary landfill of municipal waste and to change the existing attitude towards waste as waste now represents "the raw material in the wrong place", also to apply hierarchy approach to waste management (reduce, reuse, recycle, recover and residual management) and to implement an adequate methods for rehabilitation and for prevention of future pollution.

Construction of a drainage channel at borders of a landfill (the so-called collector), which would be connected to a network of municipal waste water and drainage pipes placed into the body of illegal dumps, could prevent the pollution of surface and ground water, and thus contamination of crops as well as spread of pollution in the region. A schematic presentation of links of leachate from illegal dumps with a network of urban wastewater is presented in **Figure 6**.

Figure 7 shows examples of drainage pipes placement into the landfill body and collecting the filtrate water in the collection basin lined with an impermeable foil.

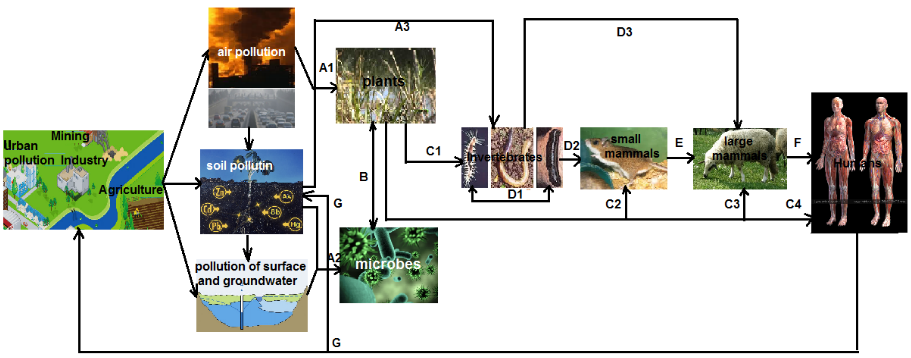

Figure 5. Pathways of metal transfer in the terrestrial food web and selected references for each pathway (A1–G) [1]: (A1) [36]-[38]; (A2) [39]-[41]; (A3) [42] [43]; (B) [44]-[46]; (C1) [47]-[51]; (C2) [52]-[54]; (C3) [55]-[57]; (C4) [58]-[63]; (D2) [64]-[66]; (D3) [67]; (F) [68]; (G) [69]-[72].

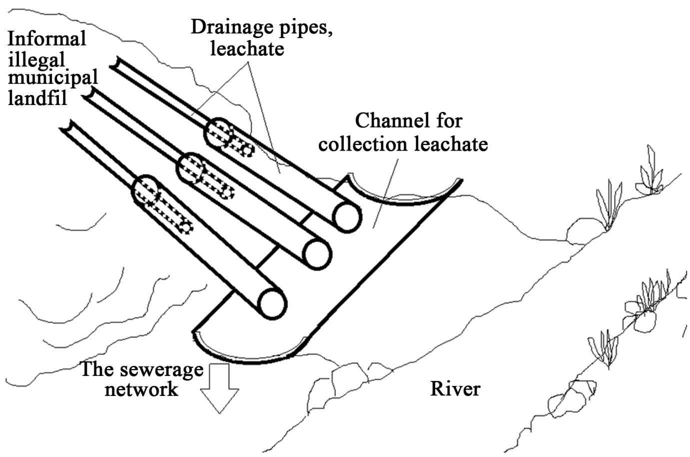

Figure 6. Landfill leachate drainage system.

(a) (b)

Figure 7. Examples of remediation: (a) drainage pipes placement and (b) impermeable thin substrate.

Including of landfill leachate into the system of municipal waste water offers a possibility for purification of municipal wastewater, industrial wastewater and leachate using unique Membrane Bio Reactors (MBR) technology. Performed analysis shows that, after the MBR wastewater treatment, quality of an effluent is better than legally required [73] [74].

In the process of treating waste and leachate with the MBR technology, the excess waste sludge is minimal, it is adequate and well stabilized, which enables its further processing. The amount of sludge in other processes is about 0.6 - 0.7 kg per kg of BOD, depending on temperature, sludge age and effluent quality.

Technological procedure of sludge stabilization is based on the process of solidification, with a purpose to convert the waste and sludge (hereinafter, on the basis of the definition of waste, used only the term waste) [75] [76], with at least one dangerous characteristic, to the state in which its constituents are immobilized in the way that doesn't make them dangerous for the environment [77] [78].

This technological process is a physical and chemical procedure in which a reaction takes place between waste and additives based on calcium (calcium oxide CaO and calcium hydroxide $Ca(OH)_2$. During this reaction, the waste molecule is permanently transformed into a new neutral product. The process takes place in a reactor under sub-pressure, so there is no emission of pollutants into the air and thus no pollution of the environment. The final product of the permanent transformation of the waste mixture is completely neutral and it can be used for various applications [79].

4. Conclusion

Results of physicochemical parameters and determined contents of heavy metals leaching from dumps in the northern part of Kosovska Mitrovica suggest an exceptional pollution. High contents of organic pollutants and heavy metals found on sites near illegal waste dumps in Grabovac and Žitkovac, as well as the concentrations of these metals in groundwater (well) in Grabovac. Particularly is disturbing increased mercury content in groundwater in Grabovac, content of copper, especially, in surface water in Grabovac and multiple increased concentrations of phenol in samples of surface and ground water. Given that the sampling was performed in three different periods (April 2013, September 2013 and February 2014), it can be concluded that pollution is present in all experimental periods. Constant, growing environmental degradation requires urgent interventions in waste management and implementation of accessible and effective method for reducing and preventing environmental pollution and the harmful and hazardous substances from entering the food chain. In this paper, we proposed a method of rehabilitation and prevention of further pollution by applying of Membrane Bio Reactor (MBR) plants for treatment of municipal, industrial and leachate from informal landfills, which include the process of stabilization of residual sludge. This technological procedure is considered as one of allowed and recommended technologies in Europe for industrial and municipal waste and leachate recycling, as well as the best available technology that does not require any excessive costs of waste management and does not pollute the environment.

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