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Impact of Red Sludge Dumps, Originating from Industrial Activity, on the Soil and Underground Water

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Abstract: In the aluminum industry, one of the most sensitive economic and environmental problems is the management of resulting waste such as slag, ash and sludge, which become potential sources of pollution. Red sludge, which results from the aluminum industry, is a mixture made up of different forms of iron and aluminum oxides, sodium and aluminum silicates, various titanium compounds, constituted in the residue left after the alkaline solubilization of alumina. The Purpose of this research is to quantify the environmental aspects involved in the storage of sludge in a landfill that has an area of 381,189 square meters and is located in the hearth of a former ballast tank in the western industrial area of the town of Oradea, Romania. The objective of the research was to determine the impact of red sludge dumps, which originated from industrial activity, on the soil and groundwater. The degree of degradation of the soil cover was highlighted by analyzing a number of 12 soil samples (4 collection points, at 3 depths). A total of 14 samples (7 samples on 2 depths) were investigated to monitor the migration mode of the sludge in the structure of the dam. In order to monitor the quality of groundwater, samples from 3 observation boreholes were analyzed. Soil monitoring results did not indicate values of the analyzed parameters above the values imposed by the national legislation on soil quality. Since the dumps were not waterproofed, the quality parameters of the water from the observation boreholes were exceeded, and gravity caused the water to drain into the underground water network in the area. Based on the samples from the observation boreholes, several measurements exceeded allowable values: pH values of the water sample taken from upstream of the dump exceeded the value limits by about 7%, and both upstream and downstream, water samples indicate an excess of 13.60% in the aluminum indicator, 267% in the sulfate ion, and 417% in the sodium ion. This shows a risk of pollution which requires additional monitoring.

Keywords: dump; red slam; environmental impact; soil; underground water; potentially significant pollution

Citation: Mintaș, O.S.; Simeanu, C.; Berchez, O.; Marele, D.C.; Osiceanu, A.G.; Rusu, T. Impact of Red Sludge Dumps, Originating from Industrial Activity, on the Soil and Underground Water. *Water* **2023**, *15*, 898. https://doi.org/10.3390/w15050898

Academic Editors: Ashwani Kumar Tiwari and Subhash Chandra

Received: 31 January 2023
Revised: 22 February 2023
Accepted: 24 February 2023
Published: 26 February 2023

1. Introduction

The management of industrial waste requires a thorough understanding of the problem [1] at several levels: industrial enterprises [2], institutions with skills in environmental management, and citizens [3–5]. The storage of historical waste from industrial activity requires proper handling and must comply with waste management legislation [6,7]. Failure to comply with these conditions can lead to serious environmental and health hazards [8–10]. Environmental impact monitoring is a necessity for the aluminum industry [4].

One of the most sensitive economic and environmental problems in the aluminum industry is the management of waste such as slag, ash and sludge in order to green the environment [11]. The efficiency of processors in the aluminum industry is closely determined by the reduction of waste quantities. In terms of the environment, waste is an additional consumer of material and energy resources, and at the same time, produces worthless residues [12] that can no longer be used and become potential sources of pollution. Waste must be stored in certain security conditions in accordance with the legislative regulations in force for the environment [13]. This problem can be solved by reducing or eliminating waste at the source and by finding recovery solutions [14].

To quantify the environmental aspects of stopping the storage of sludge in the dumps (sludge deposits) that belong to the former economic units of aluminum production, in Romania is an involved process. Economic activity owners must be constantly occupied with establish their environmental obligations [15,16], but the attention of environmental specialists [17] and the authorities [18] is also necessary. There is a lack of universally recognized research related to how to metabolize the various forms of aluminum [19–21] and understand its toxicity [22,23]; this is separate from how the evolving ways that other metals in the sludge constitute problems for the environment, soil [24–26], and water [27,28]. Red mud, which results from the aluminum industry, is a mixture of various forms of iron and aluminum oxides, sodium and aluminum silicates, various titanium compounds, etc., [29–31] and is constituted in the residue left after the alkaline solubilization of alumina. Iron oxides and hydrates are found in the red mud as predominant substances, giving it its red color [32]; specifically, they combine with sodium silicoaluminate [33] and insoluble titanate, undissolved aluminum hydroxide [34], and silica free from bauxite.

In Bihor County, in the west of Romania, there are two landfills for the storage of red sludge that has resulted from the technological process of alumina production. Both landfills are located in the outskirts of Oradea municipality (47.0465° N, 21.9189° E):

- Sludge dump C0: located in the hearth of a former ballast yard, in the western industrial area of Oradea, about 1200 m away from the residential area. It has an area of 381,189 square meters and stores about 2,000,000 t of sludge.
- Sludge dump H2: located in the outskirts of the municipality of Oradea, about 5000 m from the residential area; it includes three compartments (C1, C2, C3) and was set up in an old existing ballast tank. It has an area of 402,551 square meters and stores about 6,000,000 t of slurry.

The two storage dumps were set up in parallel to the development of technological installations for obtaining calcined alumina. The vanadium pentoxide extraction plant began operating in 1973; in 1986, the sintering plant for the processing of indigenous siliceous bauxites began operating; and in 1990, the plant for obtaining sintered alumina of the tabular type began operating. Sintering is the process of compacting and densifying a solid by applying heat treatment or pressure without reaching the melting point and liquefying the compound. Production continued until November 2006; it was then stopped, and the company's assets were transferred to conservation.

Red mud has the ability to clog and waterproof the bottom of the two dumps by filling the interstitial spaces of the granular materials. There is a lack of research regarding the metabolism of the various forms of aluminum and its toxicity outside the evolution of other metals in the sludge. The risk of pollution from the two sludge dumps shows the need for a study on environmental risk assessment regarding soil and groundwater quality.

In this context, this paper aimed to quantify the environmental aspects involved in ceasing sludge storage activity in the C0 dump, which has an area of 381,189 square meters. It focused on determining the impact of red sludge dumps that had originated from industrial activity on soil and groundwater.

2. Materials and Methods

2.1. The Composition of the Red Sludge Dump

The C0 sludge dump was built in a former ballast tank, in the pit left after the ballast was extracted, without arranging the bottom and the inner walls in any way, up to the level of the Rhine. Above the land level, a perimeter dike was built and raised several times to increase the storage capacity. Currently, this pier is 14 m high.

The transport of sludge from the technological installation to the C0 dump was carried out by pumping a pulp of sludge at a ratio of length:height = 5:1.

On the outline of the dump (left–right), there are two pipes provided with record nozzles for the uniform distribution of sludge in the dump. The sludge was deposited by decantation (gravitational), and the clear water from the surface was captured through a drain and recirculated by pumping in the company's premises; it is then reused to replenish the sludge as well as in the machinery cooling system.

The chemical composition of the red sludge stored in the landfill is [35]: aluminum oxide 12–20%; silicon dioxide 8–16%; iron oxide 22–56%; titanium dioxide 4–7%; calcium oxide 6–10%; sodium oxide 2–4%; water and other elements 7–10%.

The grain size composition of the red sludge stored in the dump is [35]: maximum grain size 1 mm; granulation below 0.075 mm is 90%; granulation between 0.16–0.075 mm is 10%. The volumetric weight in the deposited state is between 1.65–1.70 t m^{-3}. The specific weight of the slurry is 3.60 t m^{-3}.

The sludge has a colloidal structure conferred by the main components of iron oxides and hydroxides whose colloidal character is recognized in specialized literature [32,33]. Due to its physical, colloidal and surfactant properties, this sludge has the capacity to clog and waterproof the bottom of the dumps by filling the interstitial spaces of the granular materials (ballast, gravel, sand) that remain from when the ballast was extracted prior to the red mud being deposited.

The red sludge resulted from the technological installations as a liquid phase with 240–450 g L^{-1} solid suspensions; it reaches a solid content between 40–60% after settling in the landfill. The humidity of the red sludge is about 40%.

From a physical point of view, the sludge or "red mud" consists of two phases: solid and liquid.

The liquid phase of the red sludge has a residual alkalinity determined by variable concentrations of NaOH, Na_2CO_3, and sodium aluminate; this remains after the washing–settling–filtering operations but can be controlled by the parameters of the washing–treatment operations of the sludge before storage.

From a mineralogical point of view, the solid phase of the red mud is a mixture formed by different forms of iron and aluminum oxides, aluminum and sodium silicates, and various titanium compounds; all of these were formed by the alkaline leaching of bauxite containing metals predominant in bauxite.

The dangerous raw material used in the alumina manufacturing process, and which contributes to the generation of "red sludge" waste, is the caustic soda solution. In sludge, this is mostly found as a combination of the minerals contained in bauxite, which generate complex compounds, such as hydroxides, basic salts, basic oxides, and relatively stable compounds, in the alkaline environment. The excess alkaline solution, unreacted, is commonly found in the liquid phase from the initial composition of the sludge.

Before storing the red sludge in the landfill, it was subjected to technically feasible treatment operations. These operations consisted of [35]:

- removing and recovering the alkaline solution through five stages of successive washing followed by the separation of the phases through decanting operations, I—thickening, I—filtration;
- reducing the vanadium content in the stored waste to the minimum possible during the processing of bauxite with vanadium content by valorizing it in the "vanadium pentaoxide production facility";
- capturing the clarified water from the dumps through drains and recirculating it by pumping in the company's premises, reusing it to both pulp the sludge and in the equipment cooling system; thus, the red sludge dumps were an integral part of the alumina manufacturing technological flow, and the amount of alkaline solution stationed in the dump was reduced to the residual moisture of the sludge;
- sprinkling water on the surface of the dumps, especially during dry periods, to reduce air and environmental pollution from particles carried by the wind.

2.2. Location of the Red Sludge Dump

Geographically, the area around the dumps belongs to the major depression geological structure of the Pannonian Plain [36]. The geological succession is given by the complex of Pannonian clays and sands of grey-violet color; recent formations are discordantly scattered over them: terrace sands and gravels as well as clay-loam-sandy alluvial formations of Pleistocene–Holocene age were identified during the facility construction. The aquifer layer appears in the upper part of the Pliocene age formations, about 150–200 m deep.

In the deeper layers, calcareous marl and sandstone formations from the Miocene age are found, and calcareous formations of Mesozoic age are found from 1050–1100 m.

The microrelief shows the upper terrace of the Crisul Repede river (Figure 1), a relatively flat land [37], which geologically belongs to the upper and lower Pleistocene; this is represented by sands and gravels 5–15 m thick, and alluvial deposits are placed over them. The layer of gravels with fine and medium sands is slightly clayey, with rare boulders; its has variable thicknesses between 6.40–10.10 m. Under the gravel, Pannonian deposits are represented by sandy clays, clayey sands, and sandy dusts. The gravel layer is an aquifer.

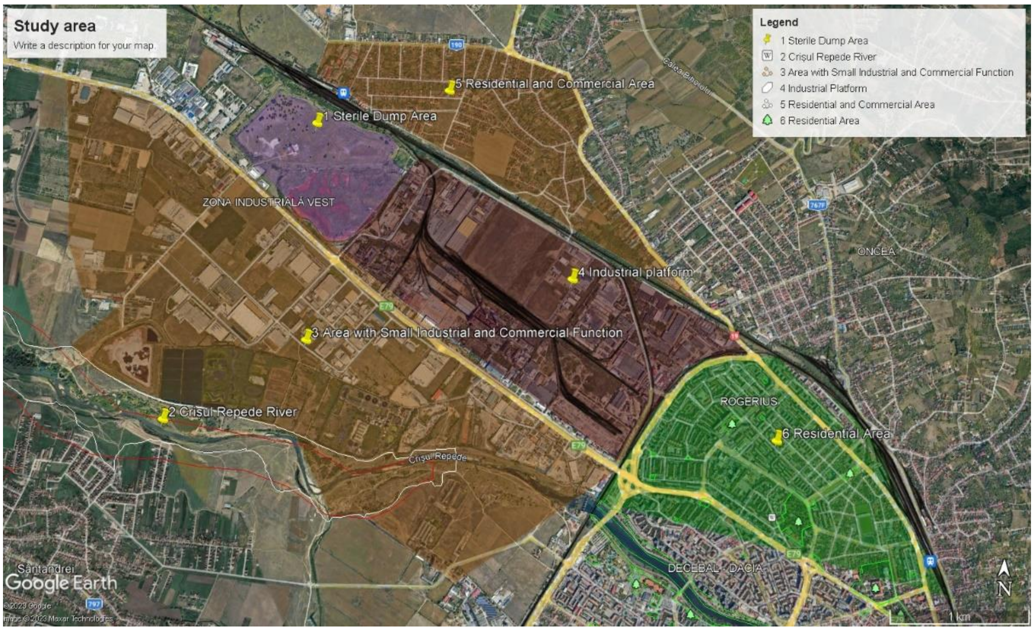

Figure 1. Research area and location of the red sludge dump.

From the analysis of the data gathered from 4 boreholes [35] made on the site of Halde C0, the lithological structure of the area is characterized as follows: the vegetal soil is encountered up to a depth of 3 m; a borchis floor (terrace) up to a depth of 12.50–16 m; marl to a depth of 21.40–25 m.

Since the exploitation of the ballast was intense at the time of drilling, the elevation of the land related to the dump was lowered by about 4–5 m compared to the rest of the region.

The average flow of the Crişul Repede river as recorded at the Oradea hydrological station is 19.60 mc s^{-1}; the minimum value recorded was 0.81 mc s^{-1} (1953), and the maximum was 820 mc s^{-1} (1932) [35].

The hydrogeological research carried out in the area [35] highlighted the phreatic horizon, confined in the Pleistocene–Holocene formations of the Quaternary, the complex of meadows (Pleistocene) and terraces (Holocene) of the Crişul Repede, and a deep aquifer complex confined in the Pannonian formations.

Storing large amounts of water in the area was favored because of the permeable formations at different levels in both in the Quaternary and the Pannonian.

The phreatic aquifer was well-defined and investigated by means of a series of boreholes [35] at the alluvial deposits of the meadow and terrace (gravel, sand, and boulders).

The aquifer layers contained in Quaternary age formations that make up the discharge cone of the Crişul Repede river can provide appreciable flows, reaching about 10–15 L s^{-1} downstream of the Oradea municipality, and much lower flows (0.88–1.50 L s^{-1}) upstream of Oradea.

The hydrographic regime is present through the Crișul Repede river, a western Pericarpathian type river. The Crișul Repede river presents average daily flows, annual minimums (mc s^{-1}): 1.40 with 97% insurance; 1.50 with 95% insurance; 1.86 with 90% insurance; 2.18 with 80% insurance; 2.45 with 70% insurance.

The hydrological regime is characterized by an increase in water in February–March and a decrease in August–September. This hydrological regime is under the influence of the oceanic masses, especially in winter when there is river heating and even rains. The snow melts around mid-February. The winter runoff is even higher than the spring runoff, reaching 30–40% of the total water volume and causing 2–6 floods, some of them very large.

The spring floods are from the rains and are somewhat smaller; those in the summer are usually even smaller, and in the fall, small floods occur but are more significant than those in neighboring areas. Due to the relatively large distance from Crișul Repede (about 1.50 km), the waste site is not subject to the risk of floods.

2.3. Research Methods Regarding the Impact of the Landfill on the Soil and Groundwater

The examination of the dykes from the C0 slurry dump, which is the subject of this paper, demonstrates the characteristics of the soil from both the dams and the natural terrain. The research discussed in this study was conducted by performing open surveys on the banks of the dykes and exploratory drilling as well as dynamic cone penetrations inside the sludge deposit. The dynamic cone penetrations were carried out up to the maximum depth of 9.00 m, compared to the crown of the dikes or the sedimentation level of the slurry.

A stratification of the soil and dendritic draining materials, both from the dikes and from the natural terrain, resulted from the drilled holes. The geotechnical characteristics of the cohesive and non-cohesive soil layers were determined from samples that were taken at different depths from both dikes and the natural terrain.

In places, the detrital material of the dikes is predominantly gravel with boulders, and the sandy fraction is reduced. Instead, leachate impregnation of the ballast layers occurred by washing the cohesive soil layers over time, which otherwise ensured the degree of compaction and the bond between the aggregates. It was found that these dykes are made on natural ground, and the surveys and drillings executed on the outer side of the dykes highlighted this aspect.

Regarding the material inside the warehouse, the tailings resulting from the technological process of obtaining alumina, is granulometrically made up of silty or clayey sands, sandy dusts, and the color is red, generally light. In some places and at certain depths, the color ranges from dark red to brick.

Relative to the height of the crown of the dikes, the sludges are about 15 m thick. This thickness cannot be considered homogeneous for the entire area of the deposit.

To assess the impact of red sludge storage from the C0 dump on the soil and groundwater, the national methodology (Romania) specified by the Order of the Minister of Water, Forests and Environmental Protection 184/1997 was applied [38]. Given its constant source, i.e., the production of alumina from bauxite, the chemical composition of the slurry inside the compartments is approximately constant.

The degree of degradation of the soil cover, determined by the activity carried out in the C0 Oradea sludge deposit area, was highlighted by analyzing 12 soil samples (four sampling points at three depths), as follows:

- Sampling point 1—P1—from a depth of 0–20 cm, marked P1-1 (0–5 cm), P2-1 (5–10 cm), P3-1 (10–15 cm), P4-1 (15–20 cm);

– Sampling point 2—P2—from a depth of 20–40 cm, marked P1-2 (20–25 cm), P2-2 (25–30 cm), P3-2 (30–35 cm), P4-2 (35–40 cm);
– Sampling point 3—P3—from a depth of 40–60 cm, marked P1-3 (40–45 cm), P2-3 (45–50 cm), P3-3 (50–55 cm), P4-3 (55–60 cm).

When taking the samples, from the first depth, the natural action of environmental and climatological factors was taken into account, which led to the partial covering of the sludge with fertile soil, where various plant species developed spontaneously. This aspect was also considered when fixing the sampling points and depth, which were carried out by this research team between 2005–2018. In 2019, a new monitoring campaign for analysis of this landfill was begun; it was not completed until 2021 because of the COVID-19 pandemic. The results of this last study are presented in this paper.

A distribution of 4 points was planned, one central and three at the ends of the dump downstream and upstream of the central point.

In order to monitor the mode of sludge migration in the structure of the dikes, samples were collected and analyzed from the dikes (D). A total of 14 samples were taken (7 samples at two depths), as follows:

– Sample D1—from the depth of 100 cm, marked D1-1, D2-1, D3-1, D4-1; D5-1; D6-1; D7-1;
– Sample D2—from the depth of 200 cm, marked D1-2, D2-2, D3-2, D4-2; D5-2; D6-2; D7-2.

The analyzed soil quality indicators were pH, dry matter and water content, sulfates, iron, and aluminum. Initially, the samples were dehydrated; then, they were analyzed using the methods presented in Table 1. The concentrations of polluting elements in the soil were reported as normal, alert, or intervention thresholds established by the Order of the Minister of Water, of Forests and Environmental Protection 756/1997 [39] for soils with sensitive use.

Table 1. Methods used for soil quality indicators.

No. Crt.	Quality Indicator	Method of Analysis [40]
1	Granulometric analysis (sieving)	SR EN ISO 14688-1: 2018
2	pH (potentiometric)	SR 7184/13-2001 PTL-19
3	Dry matter and water content	SR ISO 11465-1998 PT-63
4	Sulphates	SR ISO 11048-1999 PTL-23
5	Iron	EPA 6200 PTL-37 Ed.5 rev.0
6	Aluminum	SR ISO 11047-1999 PTL-68

The red sludge dump did not have a rainwater collection system, and the land related to it was not waterproofed. The infiltration waters from the body of the C0 deposit are evacuated by gravity into the local hydrographic network. Meteoric waters fall on the body of this deposit, infiltrate into the sludge, and then infiltrate into the soil and basement of the deposit.

The large distance of the site from Crișul Repede excludes the risk of its contamination with pollutants specific to the sludge storage activity.

In order to monitor the quality of groundwater, samples were taken from 3 observation boreholes (2 more observation boreholes are being executed and they are currently plugged), as follows:

– Drilling F1—25 m deep, is located in the north-eastern part, upstream, outside the boundary of the premises;
– Drilling F2—depth 25 m, upstream is located outside the dump in the eastern part of it;
– Drilling F3—depth 25 m, downstream is located outside the dump in the southeast part.

To establish the parameters that need to be investigated, the following were taken into account: the physical–chemical parameters that characterized the red mud and the monitoring provisions imposed by the regulatory acts that applied to the company.

The boreholes were cleaned prior to collecting the samples. The samples were placed in brown glass containers and transported to the laboratory for analysis.

The analysis methods used to determine the groundwater quality indicators are presented in Table 2.

Table 2. Methods used for groundwater quality indicators.

No. Crt.	Quality Indicator	Method of Analysis [40]
1	pH	SR ISO 10523/12 PTL-19
2	Total iron	SR ISO 6332/C91-2006 PTL-14
3	Sulfur	EPA 375.4 PTL-23 ed.5 rev 0
4	Aluminum	SR ISO 12020-2004 PTL-33
5	Sodium	STAS 3223/2-1980 PTL-36

3. Results

3.1. The Influence of the Red Sludge Dump on the Quality of the Soil

The results of the soil sample analysis are presented in Table 3. The monitoring results did not indicate values of the analyzed parameters above the values imposed by the national legislation on soil quality.

Table 3. Influence of C0 sludge dump on soil quality.

No. Crt.	Quality Indicator	Depth, cm	Sample Collection Points			
			P1	P2	P3	P4
1	pH	05–20	9.88	7.94	8.72	8.54
		20–40	9.69	8.84	8.72	8.59
		40–60	9.78	9.09	8.74	8.97
2	Dry matter and water content, %	05–20	99.26	94.38	98.47	92.60
		20–40	97.69	82.58	99.30	94.95
		40–60	98.48	75.65	85.65	83.00
3	Sulfates, mg kg^{-1} dry substance	05–20	108.4	50.40	426.5	233.5
		20–40	130.6	<50	120.9	77.19
		40–60	119.5	<50	86.38	<50
4	Total iron, mg kg^{-1} dry substance	05–20	24,754	32,113	24,612	19,962
		20–40	17,974	19,207	74,638	23,569
		40–60	21,364	21,960	45,002	53,708
5	Aluminum, mg kg^{-1} dry substance	05–20	44,312	61,328	47,858	57,953
		20–40	46,672	56,773	51,866	54,675
		40–60	45,492	67,087	59,280	43,104

Results of the analysis of the samples collected from the dike are presented in Table 4. The pH is usually lower at a depth of 100 cm and slightly higher at a depth of 250 cm, but there are exceptions. The same trend is maintained for all determined quality indicators.

3.2. The Influence of the Red Sludge Dump on the Quality of Underground Water

The red sludge heap influences the quality indicators, which were determined differently depending on the indicator and collection point. Values recorded by us were compared with the values allowed and the limits identified in Law 458/2002 [41] and the additions from Law 311/2001 [42]. Table 5 shows the results of the groundwater samples collected from the observation wells located around the dumps.

Table 4. The influence of the C0 sludge dump on the samples collected from the dump dam.

No. Crt.	Quality Indicator	Depth, cm	Sample Collection Points						
			D1	D2	D3	D4	D5	D6	D7
1	pH	100 250	7.20 7.45	7.32 7.37	7.47 8.16	7.31 7.45	7.39 7.17	7.11 7.37	7.47 7.34
2	Dry matter and water content, %	100 250	96.93 98.58	98.05 97.46	96.17 98.28	96.82 98.09	97.73 98.69	96.91 98.54	97.79 97.29
3	Sulfates, mg kg^{-1} dry substance	100 250	<50 <50	<50 106.34	173.06 122.85	<50 74.48	66.34 193.88	<50 <50	<50 69.04
4	Total iron, mg kg^{-1} dry substance	100 250	23,809 14,529	18,738 18,718	17,448 20,634	34,721 12,125	18,962 12,922	14,678 10,365	16,744 12,687
5	Aluminum, mg kg^{-1} dry substance	100 250	15,229 8973.4	13,544 14,978	15,736 12,294	16,842 10,498	14,332 8846.6	15,528 8850.5	15,557 13,115

Table 5. The influence of the C0 sludge dump on the samples collected from the groundwater.

No. Crt.	Quality Indicator	Sample Collection Points			Admissible Limit cf L458/2002 [41] with Compl. L311/2001 [42]
		F1	F2	F3	
1	pH	10.16	6.69	6.46	6.50–9.50
2	Total iron, mg L^{-1}	0.160	0.240	0.060	<0.200
3	Sulfates, mg L^{-1}	918.01	14.35	44.53	<250
4	Aluminum, mg L^{-1}	27.41	3.55	4.53	0.200
5	Sodium, mg L^{-1}	1035	114	75.20	200

4. Discussion

The results of soil analysis do not exceed the values and limits imposed by law, but we can appreciate that they have an impact on the environment [41]. This means the considerable negative modification of the physical, chemical and structural characteristics of natural environmental elements and factors [42,43]; such an impact can be identified in the present or may manifest in the future, which is considered unacceptable by the competent authorities [24,44]. The analysis of the quality indicator values of the samples collected from the dump dam highlighted the presence of the vegetative layer that was spontaneously created over a partial surface of the dump; metabolism by plants in that layer may contribute to a mitigation of the changes in the iron, aluminum, and sulfate content, as well as the pH [21]. Specifics of the deposit, including the red sludge and the residue left after the alkaline solubilization of alumina, were determined to include a high content of iron and aluminum oxides, as well as sodium and aluminum silicates, but in an unexpected way and with a high content of sulfate in the upper soil layer at P3 and P4. Sulfate and iron ions were found in greater quantity, especially in P3; in our opinion, their positioning at different depths, i.e., 05–20 cm for sulfate and 20–40 cm for iron, was influenced by the composition granulometry of the soil.

The results of the water analysis were compared with the limit values allowed for drinking water (STAS 1342/1991—Drinking water quality), and Law 458/2002 [45], amended and supplemented by Law 311/2004—Annex 1 [46]. Since the dumps were not waterproofed, the drinking water quality parameters were exceeded, and as the water drains gravitationally into the underground water network in the area, the same issue was found in similar situations [47,48].

The study of the composition of groundwater from 2019–2021 indicated that concentrations of Fe, Al, and Na all exceeded the values stipulated in Law 458/2002, both upstream and downstream, and that the pH value was below the limits provided by the normative act downstream; upstream showed overshoots in the flow direction of the aquifer layer.

Analyzed values show that the allowed limits were exceeded [41,45], for pH in F1 (10.16), for total iron in F2 (0.240 mg L^{-1}), for sulfates in F1 (918.01 mg L^{-1}), for aluminum in F1, F2, F3 (3.55–27.41 mg L^{-1}), and for sodium only in F1 (1.035 mg L^{-1}). The pH values of the water sample taken from the F1 borehole, i.e., upstream of the dump, indicated a value that was over its limit by about 7%. Water samples taken from well F1 indicate an excess of 13.60% in the aluminum indicator, 267% in the sulfate ion and 417% in the sodium ion. The samples taken from wells 2 and 3 also showed an excess of the aluminum ion concentration by 1.67% and by 2.16%, respectively.

At the same time, there is a risk of potentially significant pollution from concentrations of pollutants in the environment that exceed the alert thresholds provided in the environmental pollution regulations [49]. These values define the level of pollution at which the competent authorities consider that a site may have an impact on the environment and establish the need for additional studies [50] and measures to reduce pollutant concentrations in discharges [51].

The final rehabilitation of tailings dumps can be achieved through the following measures: excavation of bypass trenches [52,53]; landfill consolidation [54,55]; marking the perimeter of the surface of the dumps; ensuring the flatness of the dumps [56]; placing a protective cover against dust; covering with a final layer [57,58]; and revegetation of the cover layer [59]. It is mandatory to maintain the access roads, drainage systems, and vegetation (including revegetation if necessary) [60] during the entire monitoring period of the dumps.

The limitations of the study are related to the characteristics of the studied area, namely, the current characteristics of the soils in the study area are defined mainly as a result of long-term anthropogenic activities [61] and have very diverse characteristics. In these conditions, careful monitoring of resources [62,63], as provided for by national legislation, is very important in the future.

5. Conclusions

The examined sludge dump was not provided with a rainwater collection system, and the land related to it was not waterproofed, which have impacts on the environment. The infiltration waters are evacuated by gravity into the local hydrographic network. Meteoric water falls on the body of this deposit, infiltrates into the sludge and further infiltrates into the soil and basement of the deposit.

The landfill monitoring studies for the period 2005–2021 show that most of the analyzed samples have an alkaline pH, which is due both to the initial non-development (not waterproofing) of the bottom of the landfill, its inner walls, and the dikes, as well as of other industrial activities in the area. The analysis of the evolution of iron concentrations shows decreasing concentrations in the sludge and, at the same time, increasing concentrations in the underground water. Analysis of the evolution of the aluminum concentration values shows increasing concentrations in the sludge and, at the same time, decreasing concentrations in the underground water. The analysis of the values of the sludge's quality parameters revealed that the presence of the spontaneously created, partial vegetative layer on the surface of the dump contributed to a metabolization of the content of iron, aluminum, sulfates and attenuation of the pH. The great distance of the site from Crișul Repede removes the risk of the river's contamination with pollutants specific to the sludge storage activity.

Since the concentration of one or more pollutants exceeds the alert threshold, additional monitoring is required as well as the adoption of measures to reduce the concentrations of pollutants from the discharges.

Author Contributions: Conceptualization, O.S.M.; methodology, O.S.M. and T.R.; software, C.S. and D.C.M.; validation, O.B. and A.G.O.; formal analysis, O.S.M., D.C.M. and C.S.; investigation, O.S.M., T.R. and C.S.; data curation, T.R., D.C.M. and A.G.O.; writing—original draft preparation, T.R. and O.S.M.; writing—review and editing, C.S., O.B. and D.C.M.; visualization, T.R., O.B. and A.G.O.; supervision, T.R. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Acknowledgments: In our research, we benefited from the support of Acormed companies.

Conflicts of Interest: The authors declare no conflict of interest.

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