STUDY REGARDING THE PEDOLOGICAL PROPERTIES OF THE SOIL IN THE "LARGA DE SUS" MINING AREA IN ORDER TO ESTABLISH THE APPROPRIATE SOIL REMEDIATION TECHNOLOGY

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ABSTRACT: *Currently, soil pollution is a critical environmental problem that concerned all humanity due to its impact on ecological environment and human health. In order to remediate the contaminated soils, some technologies, generally based on physical, chemical, thermal, and biological processes, have long been in use. However, effectiveness of these methods depends considerably on soil type and characteristics, level of pollution and mixed contaminants present in soil. Therefore, in the present study, the pedological properties of the soil collected from abandoned mining perimeter of "Larga de Sus" from Zlatna (Alba County, Romania) were investigated in order to choose the most appropriate method for soil remediation and establish the parameters of soil remediation technology. The results indicated that soil in the area is highly acidic which favors the mobility of heavy metals in soil and other environmental components. On the other hand, most of the samples are partially structured and poorly structured while some samples are well structured, very poorly structured, or present a lack of aggregates structure. Also, the coarse sandy and loam-sandy texture of the samples was identified. Thus, considering soil properties, soil washing technology may be the appropriate remediation technology for contaminated soil in the studied area considering costs and efficiency.*

Keywords: *pedological parameters; soil pollution; mining areas; soil remediation;*

1. Introduction

Currently, soil pollution is a significant environmental problem observed at local, regional, national, and global levels, attracting the attention of specialists and the whole community due to the toxic influence of pollutants on plants and animals and the risk of accumulation in agri-food products (Gămăneci and Căpăţână, 2011). In recent years, about 4 million hectares of arable land have been moderately or severely contaminated, covering about 2.90% of China's arable land. The percentage of contaminated soils that do not comply with environmental standards reaches 16.10%. Most soils are contaminated with inorganic substances (82.80%) such as Cd, Ni, Cu, As, Hg, Pb, followed by organic substances (Chao et al., 2014).

In mining areas, one of the sources that lead to soil pollution is represented by the runoff formed when rainwater reaches sulfide-bearing minerals contained by sterile dumps and other mining waste. Runoff formed is highly acidic and contains a high level of dissolved metals, sulphate, and iron (Varvara et al., 2013).

When runoff reaches the soil, some metals are dissolved and enter into solution, while others remain adsorbed and/or precipitated and move with the soil particles causing an increase in pollution that pose a significant risk to the environment and human health (Navarro et al., 2008).

Therefore, the depollution and restoration of soil quality in order to protect the environment and human health and the reintroduction of soils in the agricultural circuit are some of the major concerns considered in legislative, technical, economic, and social terms.

In this regard, in recent years, numerous techniques for depollution of polluted soils have been investigated, developed, and used (Wuana and Okieimen, 2011). However, the successful application of soil depollution technologies considerably depends on the soil characteristics and the type and concentration of contaminants.

For example, extraction of heavy metals from the soil by the washing method is influenced by the geological and hydrological characteristics of the soil, soil type and composition, its pH, the organic matter content, distribution of pollutants in the soil, and the physical and chemical characteristics of the soil (Szanto et al., 2011). On the other hand, the application of this technology may not be feasible in terms of cost if the percentage of fine fractions in the soil exceeds 50%. Moreover, the effectiveness of some washing agents used in soil depollution may be limited by certain soil constituents. For example, the use of surfactants in soil washing may not be effective if the soil has a clay content of more than 20-30% or a high amount of organic matter (Begum et al., 2016).

Consequently, the aim of the present paper is to study the pedological properties of the soil collected from abandoned mining perimeter of "Larga de Sus" from Zlatna (Alba County, Romania) in order to choose the most appropriate method for soil remediation. A series of analyzes were performed to determine the soil pedological parameters (water content, texture, structure, and pH) that influence the effectiveness of the soil remediation technologies.

2. Materials and methods

2.1. Study area

The Zlatna mining perimeter is located in the South Apuseni mountains (Romania) and comprises as a main exploitation "Larga de Sus" mine (Fig. 1). The "Larga de Sus" mine belongs geographically to Roatei Hill (Keri et al., 2011). The access to the "Larga de Sus" mining area is made on the road DN 74 Alba Iulia - Zlatna, on DC 108 towards Trâmpoiele, then on DC 113 Zlatna - Dealul Roatei.

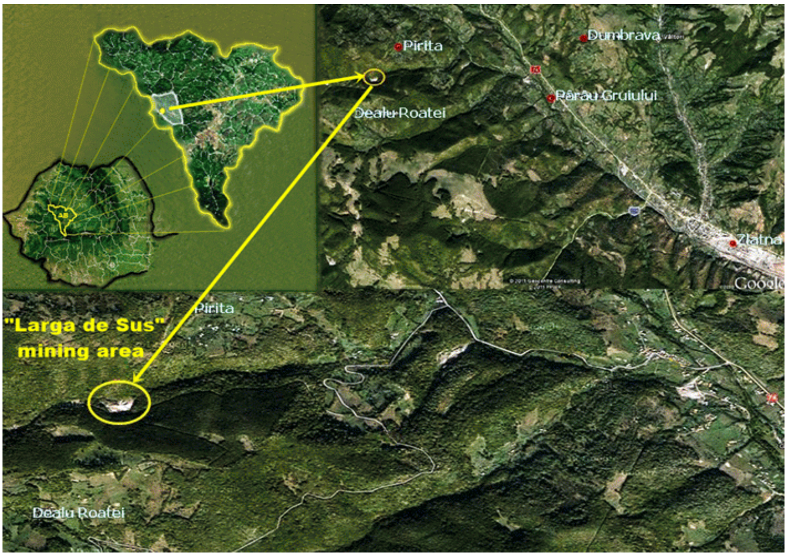

Fig.1. Geographical location of the "Larga de Sus" mining perimeter

The “Larga de Sus” mining perimeter falls within the so-called “Transylvanian Gold Quadrangular” which includes the mining perimeters of Deva, Brad, Zlatna and Caraciu (Keri et al., 2010). The “Larga de Sus”, “Haneş”, “Faţa Băii” and “Trâmpoiele” mines are located in the southeastern part of the Zlatna-Stănija volcanic alignment (Keri et al., 2011). In the Zlatna-Stănija volcanic alignment, the most important ore bodies are the pyrite concentrations from “Larga de Sus” and the tellurides and gold mineralizations from “Faţa Băii” (Luduşan, 2002).

Mineralizations are of hydrothermal origin and are present as veins, stocks, breccia bodies and lenses. The mineralization related to volcanic activity is of Au - Ag - Pb - Zn epithermal type (low sulphidation - rich sulphide). Mineralogical paragenesis consists of pyrite, chalcopyrite, blende, galena, covellite, chalcocite, tetrahedrite, antimonite, sulphonic-acid halides (petzite, calaverite, other), native gold, gangue (quartz, rhodochrosite, calcite, baryte) (Keri et al., 2010; 2011).

In the Southern Apuseni Mountains, mining activities have taken place since Antiquity, leaving their marks upon the natural environment (Duma, 2009). Although mine closure was achieved in 2006 and today is undergoing the closure and ecological rehabilitation phase (Keri et al., 2010) the soil in the area remained exceedingly polluted with heavy metals and no remediation technology was implemented till now (Keri et al., 2010; Babau et al., 2017).

2.2. Sampling and analysis

For this study, a total of 10 sampling sites were selected according to a sampling design established based on the size of the investigated mining perimeter, knowledge of the site history, the position of the main source of pollution, land topography, and visual inspections conducted on-site, as shown in Fig. 2.

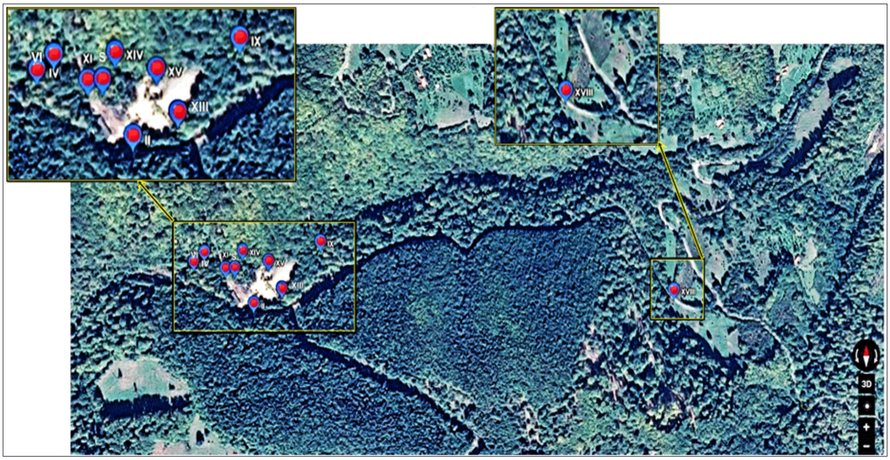

Fig. 2. Distribution of soil sampling points

Soil sample points II, IV, VI, and IX were located in the forest on the left side, upstream, right side, and downstream of the industrial area of “Larga de Sus” mine, respectively. Soil sample points XIII and XIV were located on the left side and right side of the “Larga de Sus” sterile dump. Sample point XI was located in the front of the “Larga de Sus” gallery, in the industrial area of the mine, at about 10 meters distance from the gallery. Sample point S (sediment) was located in the industrial area of the mine, in the former mining wastewaters drainage channel. Sample point XV (sterile material) was located on the "Larga de Sus" sterile dump and sample point XVIII was located on a pasture at about 800 meters downstream "Larga de Sus" mine.

According to methodological norms stipulated in STAS 7184/1-94, from every single sampling point, marked with red on the Fig. 1, were collected three soil samples corresponding to a depth of 10–30 cm, 30-60 cm, and 60-100 cm, respectively (Fig. 3).

passed through the device for 15 minutes and then the remaining amounts on each sieve were measured by gravimetry, using an electronic balance (0.1 g precision) in order to determine the percentage of fine and coarse sand.

The pH of the collected samples was measured in a suspension of soil/sediment /sterile material and distilled water at a L/S ratio (mL:g) of 2.5:1 with a Hanna HI 3512 pH-meter.

Fig. 3. Soil, sediment, and sterile material samples collected from the "Larga de Sus" mining perimeter

Collected samples were subjected to a number of analyzes in order to determine their pedological parameters. The determined pedological parameters were water content, texture, structure, and pH.

The texture of the soil/sterile material/sediment samples was determined using the RETSCH AS 200 Control sifting device which operates based on an electromagnetic propulsion system and has the role of separating the granulometric fractions by sifting with different dimensional sites: 4 mm, 2 mm, 1 mm, 500 µm, 250 µm and <250 µm. Practically, a quantity of 500 g of soil/sterile material/sediment from each collected sample, pre-dried at room temperature, was

The water content of the samples collected (soil, sediment, and sterile material) was determined by the gravimetric method. Practically, each sample was weighed with analytical balance (0.001 g precision) placed in a Petri dish and introduced in an oven at 105 °C until constant weight.

The structure of the collected samples was determined with Sekera method, which consists of the dissolution (dispersion) in water of soil/sediment/sterile material aggregates and results assessment using an indicator board with representations of stability of the aggregates according to Fig. 4 (Micle and Sur, 2012).

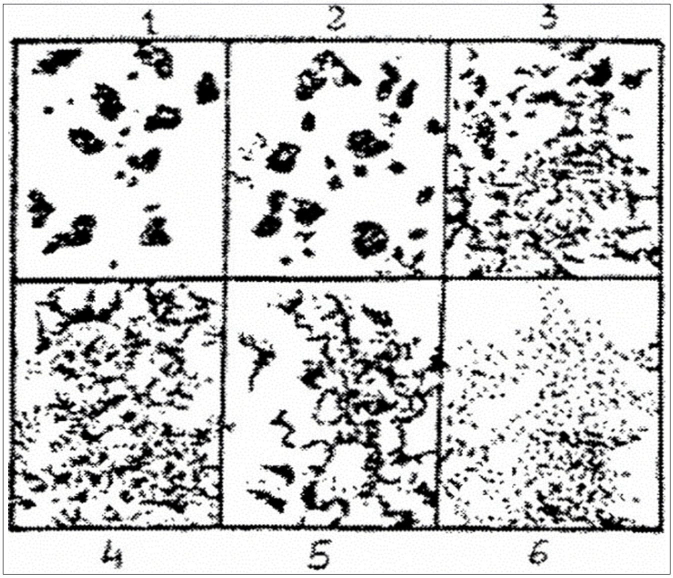

Fig. 4. Representation of aggregates stability
(Rusu et al., 2007)

The assessment of the state of the aggregates was performed according to the indicator board, and the interpretation of the results was performed as follows (Micle and Sur., 2012): i) note 1- very well structured; ii) note 2- well structured; iii) note 3- partially structured; iv) poorly structured note 4; v) grade 5- very poorly structured; vi) note 6- lack of aggregate structure.

3. Results and discussion

The humidity of the samples collected from the “Larga de Sus” mining area, determined by the gravimetric method, is presented in Fig. 5. The humidity values of the collected samples vary depending on the sampling point, and also on the sampling depth. Thus, the moisture values vary from 9.01% to 70.08%.

Considering the variation of the water content with the sampling depth, the humidity slightly increases in the case of the soil collected from sampling point II (located in the forest, upstream of the "Larga de Sus" gallery) and sterile material collected from sampling point XV (located on the sterile dump). Thus, in sampling point II, the humidity values increased from 14.61% (corresponding to a depth of 10-30 cm) to 15.24% (corresponding to a depth of 30-60 cm) and to 17.39% (corresponding to a depth of 60-90 cm), respectively. In the sampling point XV (sterile material) the humidity values increased from 9.57% to 10.95% and to 11.71%, as the depth increased from 10-30 cm to 30-60 cm, and 60-90 cm, respectively.

This increase is probably due to the texture of the soil layers in sampling point II. Deep soil layers have a higher percentage of fine particles than the surface soil layers (which is coarser), which causes more intense leaching in the surface layer and water retention in the deep layers.

The increasing trend of moisture values with depth was not observed in the case of the other sampling points. Therefore, for sampling point IX (located in the forest, downstream of the “Larga de Sus” sterile dump) the humidity decreased slightly with increasing depth, probably due to the leakage on the soil surface of the leachate coming from the sterile dumps.

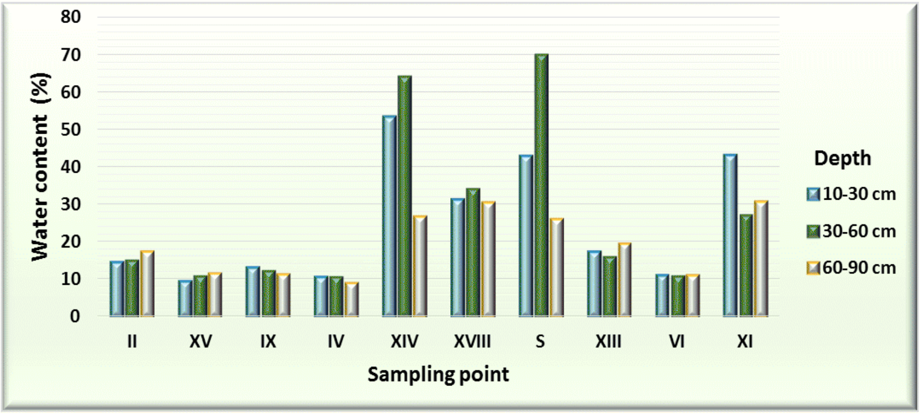

Fig. 5. Variation of the humidity of the samples collected from the "Larga de Sus" mining area depending on the depth

As regards soil sampling points IV, XIV, XVIII, and S (sediment) the humidity increases in the middle layer (corresponding to the depth 30-60 cm), and remains low in the other two layers. This may be due to the higher percentage of fine fractions contained in the soil layers located at a depth of 30-60 cm compared to the other two layers (with a lower content of fine fractions). On the other hand, as regards sampling points XIII, VI,

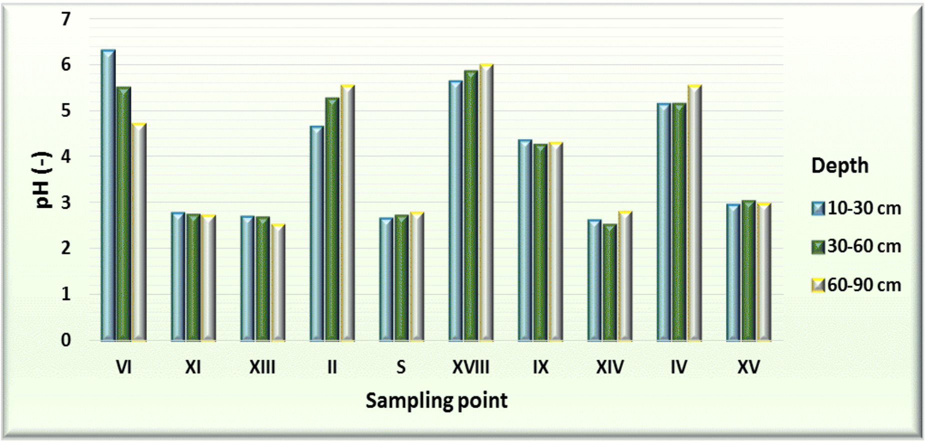

Fig. 6. pH variation of the samples collected from the "Larga de Sus" mining area

and XI the humidity decreases in the soil layer located at a depth of 30-60 cm compared to the surface layer, after which it grows again in the soil layer located at a depth of 60-90 cm. In this case, too, a possible explanation could be related to the content of fine fractions in the soil layers which favors water retention.

The soil collected from the abandoned "Larga de Sus" mining perimeter is characterized by pH values between 2.54 and 6.32. The pH variation of the samples collected from the "Larga de Sus" mining area, depending on the depth is indicated in Fig. 6.

For more than a half of the soil samples collected from the "Larga de Sus" mining area, the pH value was below 3.5. This indicates an extremely acidic environment.

On the other hand, five samples were

classified as very strongly acidic and strongly acidic as they showed values between 3.6 and 5.0 pH units, and seven samples were moderately acidic with pH values between 5.1 and 5.6.

Only a number of three samples showed pH values between 5.86 and 6.32, thus being classified as weakly acidic. Therefore, as expected, the samples collected from the "Larga de Sus" mining area have pH values corresponding to the extremely strongly acidic to weakly acidic range.

Regarding the pH variation with the depth, for sampling point VI, XI, and XIII the pH decreases slightly with increasing depth. For sampling points II, S, and XVIII the pH increases slightly with increasing depth.

Another trend of pH variation was observed in the case of sampling points IX, XIV, and IV where the pH decreased slightly in the soil layer corresponding to a depth of 30-60 cm as opposed to the soil layer corresponding to a depth of 10-30 cm, after which it grew slightly in the soil layer corresponding to a depth of 60-90 cm.

The pH variation from layer to layer does not show significant values, the differences between the measured pH values being between 0.01 and 0.82 pH units. However, when the first and the last sampling layer are considered, the pH values vary even by 1.61 pH units. Thus, considering the first and last layer, two trends of pH variation were observed. Therefore, in 4 sampling points (IX, VI, XI, and XIII) the pH decreased with depth, and in 6 sampling points (XIV, S, IV, II, XVIII, and XV) the pH was found to increase with depth. Depending on the available data from the analyzes performed and the ones from the field observations, a connection could not be made between the sampling point and the variation of the pH.

The structure of the samples collected from the “Larga de Sus” mining area is indicated in Fig. 7, depending on the

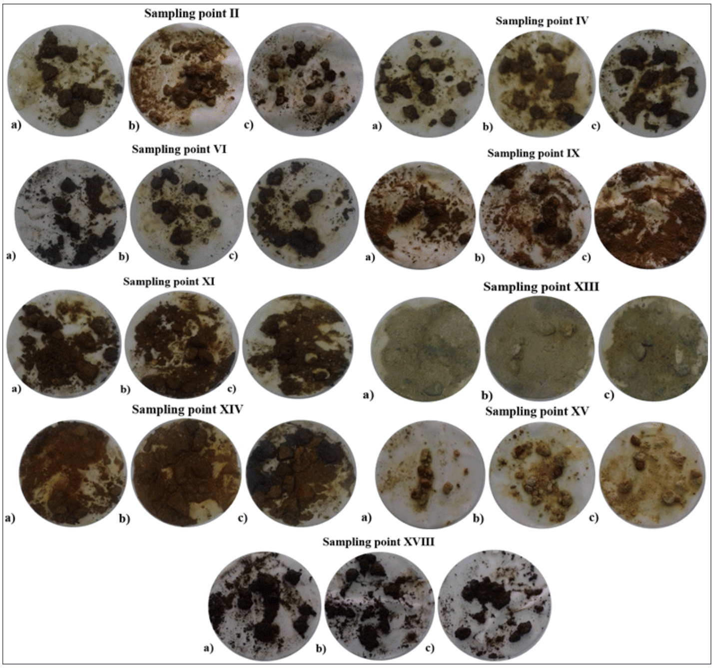

Fig. 7. The structure of the samples collected from studied area depending on the sampling point and sampling depth: a) 10-30 cm; b) 30-60 cm; c) 60-90 cm.

sampling point and sampling depth.

Most of the samples are partially structured and poorly structured. On the other hand, some samples are well structured, very poorly structured, or present a lack of aggregates structure.

The variation of the fine and coarse sand percentage in the collected samples is indicated in Fig. 8 and Fig. 9.

one sample, namely the sample collected from the sampling point S from a depth of 30-60 cm. In this case, the percentage of fine sand is higher than the coarse sand percentage, which is possibly due to the different nature of this sample, namely sediment collected from the former mining wastewaters drainage channel.

The analyzes carried out indicated a

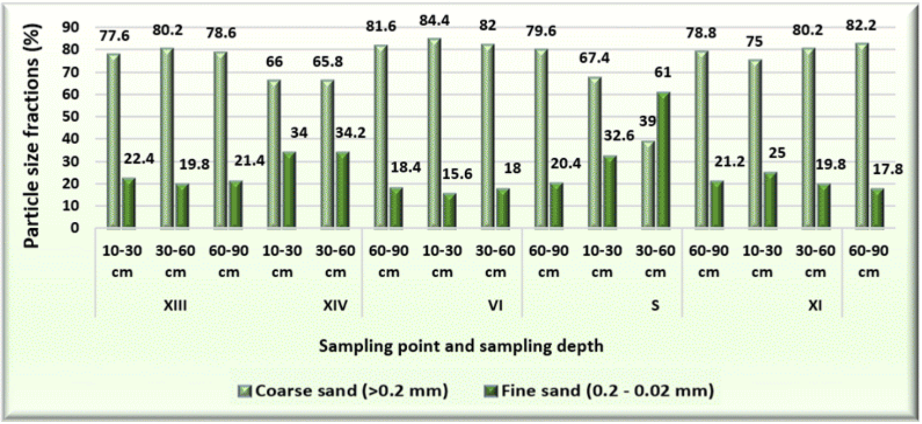

Fig. 8. The variation of the fine and coarse sand percentage for the samples collected from sampling points XIII, XIV, VI, S and XI depending on the sampling depth

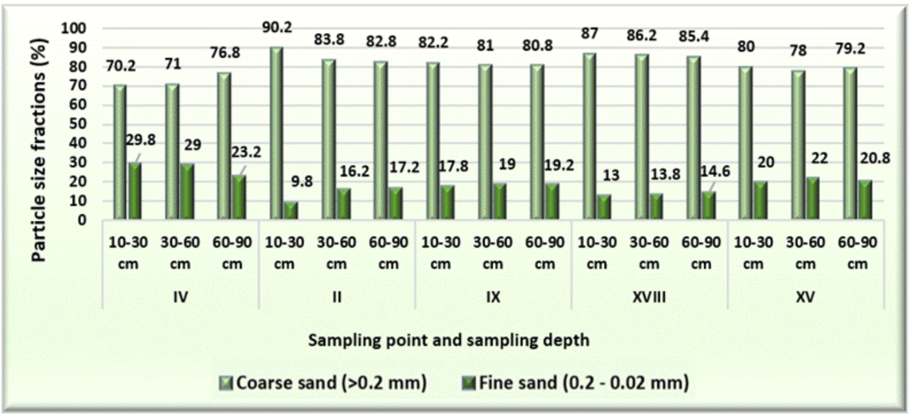

Fig. 9. The variation of the fine and coarse sand percentage for the samples collected from sampling points IV, II, IX, XVIII and XV depending on the sampling depth

As can be seen from Fig. 8 and Fig. 9, the percentage of coarse sand is higher than the fine sand percentage for all samples collected from the "Larga de Sus" mining area and for all sampling depths, except for

percentage between 39% and 90.2% coarse sand and a percentage between 9.8 and 61% fine sand. Thus, the coarse sandy and loamy-sandy texture can be noticed.

For the samples collected from sampling point XIII (located next to the access road, on the left side of the "Larga de Sus" sterile dump) or from sampling point VI (located in the forest, on the right side of the "Larga de Sus" gallery) the percentage of coarse sand is between 77.6% and 80.2% and 79.6% and 84.4%, respectively, indicating a coarse sandy texture.

In the case of samples collected from the sampling point S (sediment) the percentage of coarse sand is between 39% and 78.8% indicating a loamy-sandy texture. For the samples taken from sampling point XIV the percentage of coarse sand is between 65.8% and 81.6% indicating a sandy-loamy texture.

The samples taken from sampling points XI and XV have a coarse sandy texture, the percentage of coarse sand being between 75% and 82.2% and between 78% and 80%, respectively.

With regard to the samples taken from point IV, the percentage of coarse sand is between 70.2% and 76.8%, which indicates a coarse sandy texture. Also, the samples collected from sampling points II, IX, and XVIII present a coarse sandy texture, the percentage of coarse sand being between 80.8% and 90.2%.

4. Conclusions

In the present study, the pedological parameters (water content, texture, structure, and pH) of the soil collected from abandoned mining perimeter of "Larga de Sus" from Zlatna (Alba County, Romania) were investigated in order to choose the most appropriate method for soil remediation and to establish the operating parameters of soil remediation technology.

The effectiveness of the available soil remediation technologies considerably depends on the acidity and alkalinity of the soil, the percentage of fine fractions, organic matter, humidity etc.

The results indicated that the soil in the studied area is highly acidic, with pH values between 2.54 and 6.32, which favors the mobility of heavy metals in soil and other environmental components.

Most of the samples are partially structured and poorly structured while some samples are well structured, very poorly structured, or present a lack of aggregates structure.

Also, the coarse sandy and loam-sandy texture of the samples was identified, the percentage of coarse and fine sand being situated between 80.8 - 90.2 % and 13 -16.6 %, respectively.

Therefore, among available soil remediation technologies, soil washing may be the appropriate soil remediation technology for polluted soil in the studied area considering costs and efficiency.

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