

## Sustainable sludge management - its use for remediation of degraded lands

<https://doi.org/10.21698/rjeec.2020.224>

Proceedings Paper

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### Abstract

*The sludge resulted from the urban wastewater treatment plants, can be used, from a circular economy, as a fertilizer for agricultural and degraded lands, for horticulture and forestry.*

*This paper aims to propose an alternative for the use of this type of sludge in a sustainable way, with a low impact on the environment. The study consisted of experiments done to recover the stabilized sludge from the urban wastewater treatment plants in the process of degraded land rehabilitation, using the energetic willow, *Salix sp.**

*The experimental results showed that the use of stabilized sludge as fertilizer for naturally and anthropogenic degraded lands determined the improvement of nutritional characteristics of the lands cultivated with *Salix sp.*, and allowed the growth, development, and maintenance of this plant. Taking into consideration the characteristics of the cultures and the quantities of metals accumulated by *Salix sp.*, it can be concluded that these plants were adapted to the environmental conditions and the substrate toxically composition. The results obtained on two degraded lands indicate that *Salix sp.* can be used for its introduction in the economic circuit, both for salty soils and ash dumps.*

**Keywords:** *sludge, degraded land, circular economy*

### INTRODUCTION

The European Environmental Legislation raises extremely complex issues on how to approach the sludge disposal. The Urban Wastewater Directive (91/271/EEC) requests the treatment of effluents for inhabited areas of a certain size, this approach leading to an increase of the sludge volume generated from treatment plants [1].

At the EU level, there are different options for sludge disposal from sewage treatment plants. The most common methods consist of sludge deposition in non-hazardous waste landfills (50-70%), use as organic fertilizer, incineration/co-incineration, co-processing (in cement factories), etc.

Processing of sludge from wastewater treatment plants is following EU policy on the application of the circular economy principles and the National Sludge Management Strategy [2-4]. The sludge recovery options target two directions, *energy recovery, and agricultural use*. Due to the content of organic matter, macro- and micro-nutrients, the sludge is suitable to be used as fertilizer.

The Directive no. 86/278 / EEC [5] transposed by Order no. 344/708 on the approval of technical norms for the environmental protection and especially of soils, when using the sludge from wastewater treatment plants in agriculture, provides limits for the sludge used in agriculture as well as for the soils on which it is stored [6]. To establish the doses of sludge applied as organic fertilizer, it is necessary to know the land quality by performing periodic analytical control.

Degraded soils are poor in nutrients and organic matters and may contain toxic elements, and also may present high acidity or alkalinity [7, 8]. For the restoration of these lands, the soils can be covered with fertile soil, and/or mineral fertilizers can be used. These methods are too expensive. A cheaper and successful alternative to these processes is the use the sludge from urban wastewater treatment plants as fertilizer [9-11].

Starting from these aspects, in this paper are presented the results based on the experimental studies related to the use the sludge, resulting

from urban wastewater treatment plants, as a fertilizing agent for naturally salty lands and anthropogenic degraded areas (ash dumps) [12, 13]. For the restoration/rehabilitation of the degraded lands, it was decided to apply the phytoremediation process using the culture of

the energetic willow, *Salix sp.* The advantage of this method is that *Salix sp.* can remove the soluble salts and the metals (Zn, Ni, Cu, Fe, Mn, Pb) from soil depth due to its root system and, in the same time, can be used as a renewable energy source [14, 15].

## EXPERIMENTAL PART

Experimental studies on the restoration/rehabilitation of degraded soils were performed in-situ on two types of land: one natural degraded land (salty land) and on the surface of an ash dump.

To increase the rate of organic matter and nutrients (NPK mainly), the experimental lots were fertilized with stabilized sludge from the urban wastewater plant and manure from a pig farm. The characteristics of the stabilized sludge and the manure are presented in tables 1 and 2. The experimental lots were fertilized with 40 t/ha of stabilized sludge and 80 m<sup>3</sup>/ha

of manure. The incorporation of the fertilizers in the superior layer (0-30 cm) was realized on the same day.

The meaning of experimental lots: LD – experimental lot on salty land and fertilized with manure; LN – experimental lot on salty land and fertilized with stabilized sludge; HD – experimental lot placed on ash dump fertilized with manure and HN – experimental lot placed on ash dump fertilized with stabilized sludge. Each experimental lot was divided into three sublots.

**Table 1.** Characteristics of the stabilized sludge used for fertilization

Parameter	Measurement unit	Average determined value	Limit values for metal concentration in sludge [6]
pH	-	7.7	-
Humidity	%	71.4	-
Volatile matter	%	64.2	-
Total N	%	3.15	-
Total P	%	0.904	-
Cd	mg/kg d.m.	2.64	10
Cr	mg/kg d.m.	35.4	500
Cu	mg/kg d.m.	47.7	500
Fe	mg/kg d.m.	418	-
Mn	mg/kg d.m.	1107	-
Ni	mg/kg d.m.	38.8	100
Pb	mg/kg d.m.	41.4	300
Zn	mg/kg d.m.	298	2000
K	mg/kg d.m.	2784	-
BTEX	mg/kg d.m.	<0.01	-

Note: d.m. = dry matter

**Table 2.** The manure characteristics from a pig farm

Parameter	Measurement unit	Average determined value	Parameter	Measurement unit	Average determined values
pH	-	7.6	Cd	mg/kg d.m	<0.5
Humidity	%	99.2	Cr	mg/kg d.m	2.1
Volatile matter	%	51.6	Cu	mg/kg d.m	164
Total N	%	0.14	Ni	mg/kg d.m	10.2
P <sub>2</sub> O <sub>5</sub>	%	0.12	Pb	mg/kg d.m	<0.5
K <sub>2</sub> O	%	0.18	Zn	mg/kg d.m	513

The stabilized sludge used for the fertilization of experimental lots presents an average content of organic matter, nutrients, and the metal concentrations within the limits provided by Order 344/708/ 2004.

The manure used was liquid, with an organic matter content of 51.6 % and the nutrients content smaller than in the stabilized sludge. The metal concentrations are within the limits provided by Order 344/708 [6].

Metals such as Zn and Cu found in the manure come mainly from the additives and

#### Methods of analysis

The soil samples were collected from five points on each experimental subplot, from a depth between 5-20 cm.

The soil samples were processed by mineralization with aqua regia and analyzed by atomic absorption spectrophotometry according to SR ISO 11466:1999 [17] and SR 13315-96/C91:2008 for Fe [18, 19] methods, using an

#### Method of calculation

The bioaccumulation (transfer) factor, BF, represents the bioavailability of metals in the upper soil layers and their transfer to the plants

$$BF = C_t / C_s$$

where  $C_t$  – concentration of metal in the *Salix*

#### Statistical analysis

The values of the metal concentrations determined in the soil and the *Salix* sp. stems

## RESULTS AND DISCUSSION

During the experiments, was determined the concentration of metals (Fe, Mn, Zn, Ni, Cu,

#### Determination of metals in *Salix* sp. cultivated on salty soil

Figures 1 and 2 show the quantities of metals determined on *Salix* sp. in the 2<sup>nd</sup> year of vegetation (after 12, 15, and 18 months from cultivation) on a salty land and fertilized with manure and stabilized sludge.

The amounts of metals accumulated in *Salix* sp. plants increased with the increasing time of vegetation. During the 2<sup>nd</sup> year of vegetation, it was found that *Salix* sp. cultivated on salty lots fertilized with manure had a high accumulation

supplements used in pig food. The capacity of Cu absorption in soil determines the toxic action on the plants. As for Zn, it presents reduced toxicity for animals and can be accumulated easily and in a large proportion in the green parts of the plants [16].

Due to the content of nutrients (nitrogen, phosphorus, potassium, microelements) and the organic matter concentration, the stabilized sludge and the pig manure are considered very useful as soil fertilizers.

Avanta atomic absorption spectrophotometer.

To determine metals in *Salix* sp. the stem was chosen because this part of the plant is a better pollution indicator than leaves, due to the longer period of exposure to heavy metals. At the same time, the stems of *Salix* sp. can be used as a source of renewable energy.

[20, 21]. With the use of the bioaccumulation factor, one can evaluate the phytoremediation potential of certain plants.

$$(1)$$

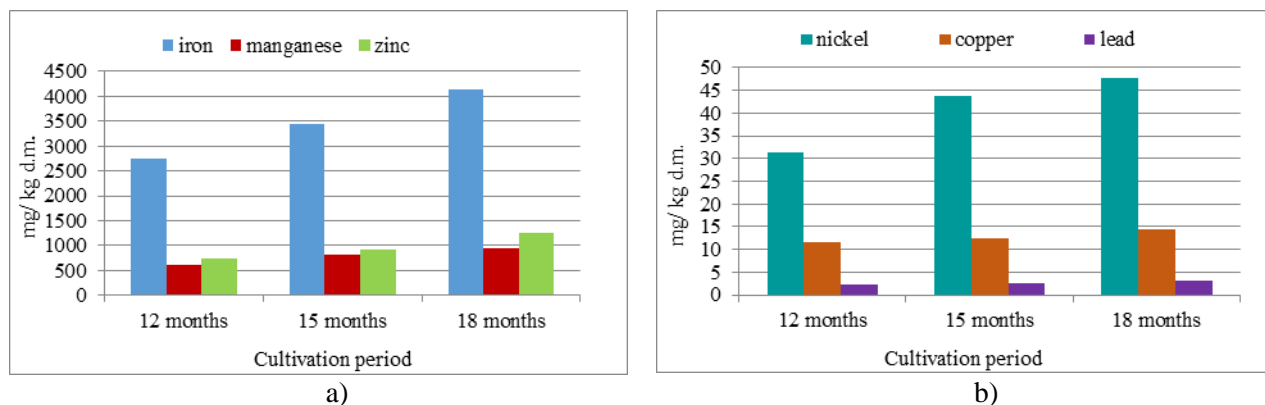
sp. stem, and  $C_s$  – concentration of metal in soil.

represent the average of five samples taken from each subplot.

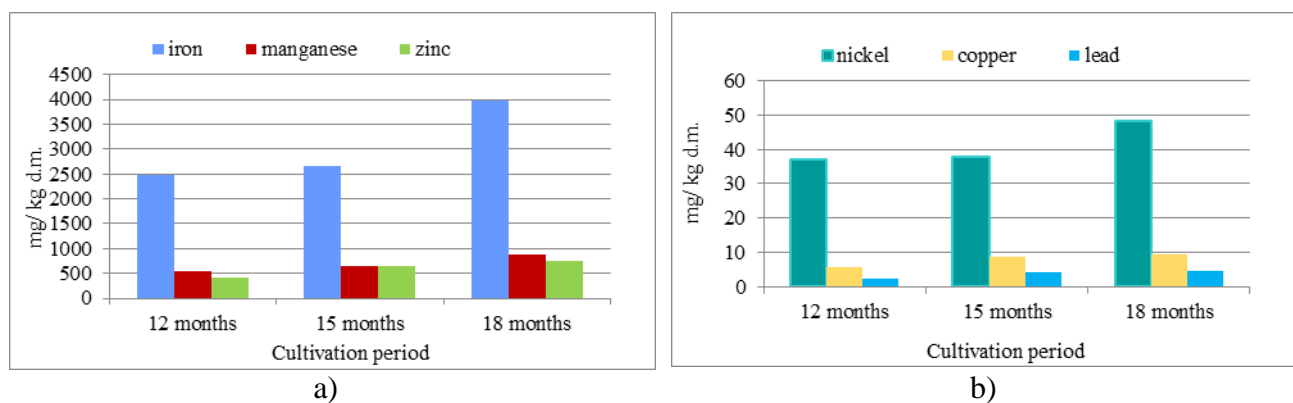
Pb) accumulated by *Salix* sp., during two phenological stages of vegetation.

capacity for Zn, Mn, and Fe. The amount of Zn accumulated after 18 months from cultivation shows an increase of about 41-45% in comparison with the stems harvested after 12 months. For Mn and Fe the increase was of 31-35% and 33-37%, respectively.

The quantities of Cu, Ni, and Pb bioaccumulated in *Salix* sp. were under 100 mg/kg d.m., the smallest amount being about 5 mg/kg d.m for Pb.



**Fig. 1.** Quantities of metals accumulated by *Salix sp.* cultivated on salty land fertilized with manure a) for Fe, Mn, Zn, b) for Ni, Cu, Pb



**Fig. 2.** Quantities of metals accumulated by *Salix sp.* cultivated on salty land and fertilized with stabilized sludge a) Fe, Mn, Zn; b) Ni, Cu, Pb

The *Salix sp.* planted on the experimental lots fertilized with pig manure accumulated higher quantities of metals in comparison with those cultivated on the experimental lots fertilized with stabilized sludge.

The quantities of metals accumulated in the stems of *Salix sp.* harvested from the experimental lots are in correlation with the data from other studies [21].

#### Determination of metals in *Salix sp.* cultivated on ash dump

Figures 3a and 3b show the metal concentrations in *Salix sp.* cultivated on the ash dump in the I and II phenological stages of vegetation.

The quantities of Fe, Zn, and Cu in the stems of *Salix sp.* were higher for the ash lots fertilized with manure. The samples of *Salix sp.* collected from the lots fertilized with stabilized sludge presented higher quantities of Ni, Mn, and Pb comparing with those collected from the lots fertilized with manure. An explanation for these differences may be the fact that the upper layers of soil have various concentrations of metals

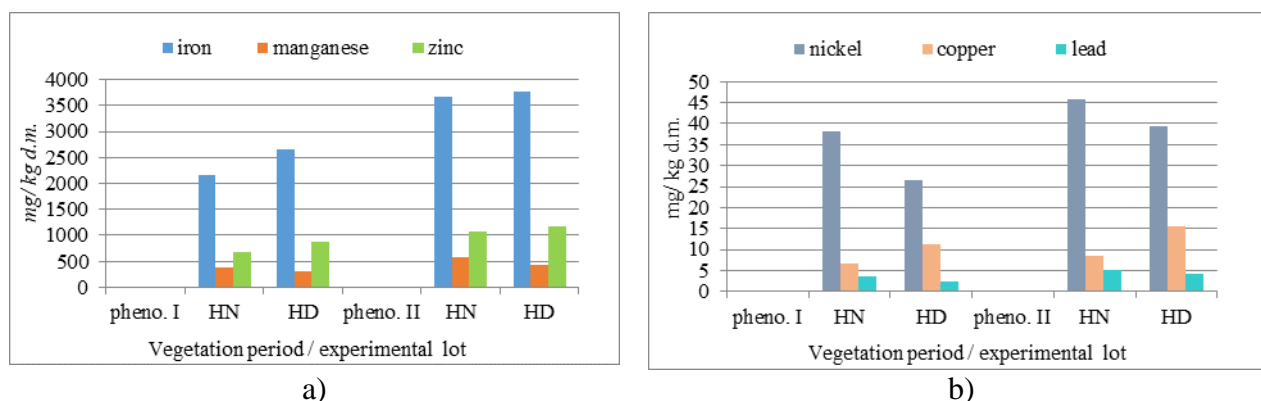
The capacity of *Salix sp.* to accumulate metals varies with the type of willow, the development phase of the plant, and the type of soil. According to previous studies, the concentrations of metals in *Salix* present a wide range of variations: 145-6900 mg/kg d.m. for Fe, 28-980 mg/kg d.m. for Mn, 41 - 2087 mg/kg d.m. for Zn, 5.6-24 mg/kg d.m. for Cu and 0.4-29 mg/kg d.m. for Pb [22- 25].

and the quantities accumulated by *Salix sp.* depend on the degree of bioavailability of the metals and nutrients in the soil and on the climatic conditions [21].

The biggest accumulations are of Fe, Zn, and Mn. The quantities of Fe accumulated in the stem of *Salix sp.* after the II<sup>nd</sup> phenological stage, are of 3667-3782 mg/kg d.m., which are smaller than the concentrations reported by Wojtun et al. in 2019 (value of 6900 mg/kg d.m.) [21].

Ni, Cu, and Pb present much lower concentration, situated under 45.8 mg/kg d.m.

(Fig. 3b), results being similar to the concentrations reported by the same study [21].

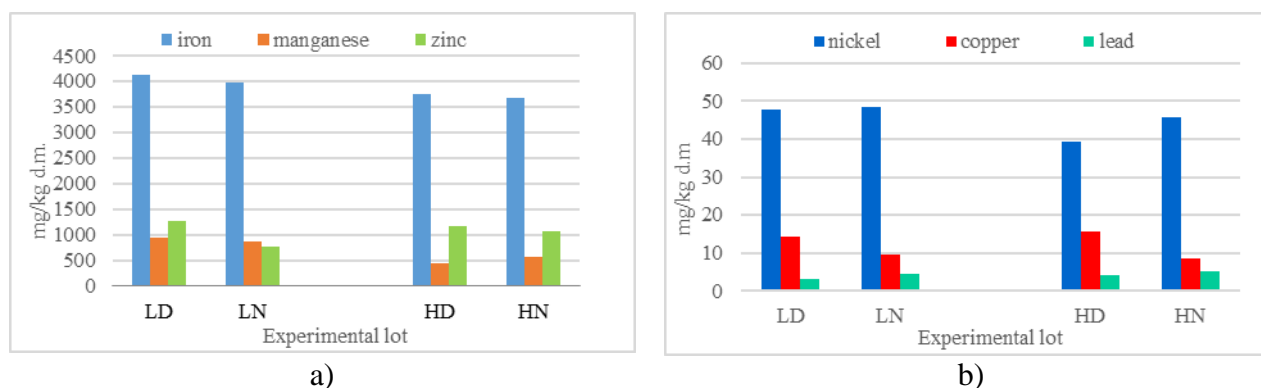


**Fig. 3.** Quantity of metals in *Salix sp.* cultivated on ash dump fertilized with stabilized sludge and manure  
a) Fe, Mn, Zn; b) Ni, Cu, Pb

#### Variation of the metal content in *Salix sp.*

Figures 4a and 4b present a comparison between the quantities of metals accumulated in the *Salix sp.* planted on the salty land and in the

ash dump, in the II<sup>nd</sup> phenological stage of vegetation.



**Fig. 4.** Quantities of metals in *Salix sp.* cultivated on salty land and ash dump  
a) Fe, Mn, Zn; b) Ni, Cu, Pb

The concentrations of metals accumulated in the stem of *Salix sp.* was higher in the case of the experimental lots placed on salty land and fertilized with manure in comparison with that one from the ash dump fertilized with stabilized sludge.

The experimental studies confirmed that *Salix sp.* presents a high capacity of absorption for Zn, Mn, and Ni.

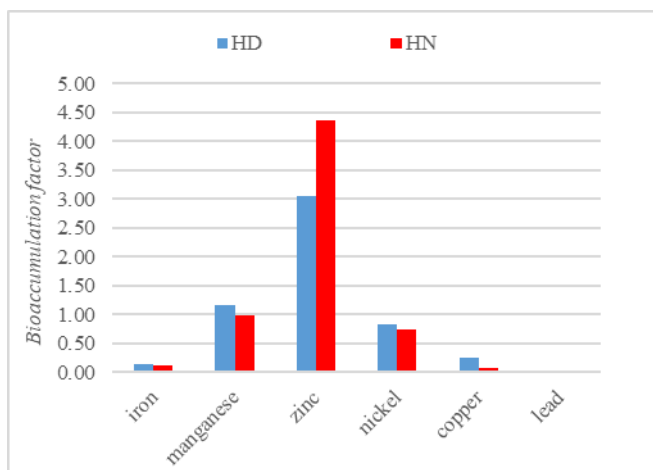
The quantity of Zn in the stem of *Salix sp.* cultivated on the 4 types of experimental lots varied from 762 to 1264 mg/kg d.m., the biggest quantity of Zn being registered on the salty lot

fertilized with manure. The accumulated quantity is situated in the interval reported by the previous studies, between 41-2087 mg/kg d.m [21]. The biggest quantity accumulated in the stem of *Salix sp.* was that of Mn, of 937 mg/kg d.m., also for the salty lot fertilized with manure. The biggest quantity of accumulated Ni was registered on the salty lots fertilized with manure and stabilized sludge (47.6 and 48.3 mg/kg d.m.). The domains of accumulation for Mn and Ni are 28-980 mg/kg d.m. for Mn and 2.1 – 45 mg/kg d.m. for Ni, as reported in previous studies [21].

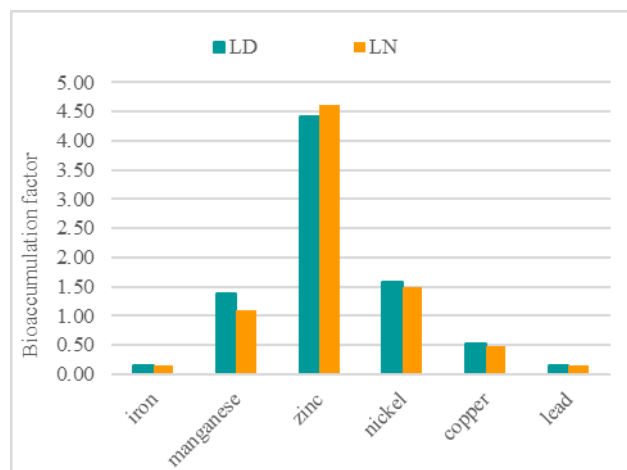
#### The degree of bioavailability of metals in soil

The increase of metal quantities in *Salix sp.* is mainly associated with the degree of bioavailability of metals in soil. Figures 5 and 6

present the bioaccumulation factors (BF) for Fe, Mn, Zn, Ni, Cu, and Pb in the experimental lots.



**Fig. 5.** The bioaccumulation factor of metals in the stems of *Salix sp.*, in the ash dump



**Fig. 6.** The bioaccumulation factor of metals in the stems of *Salix sp.*, in the salty land

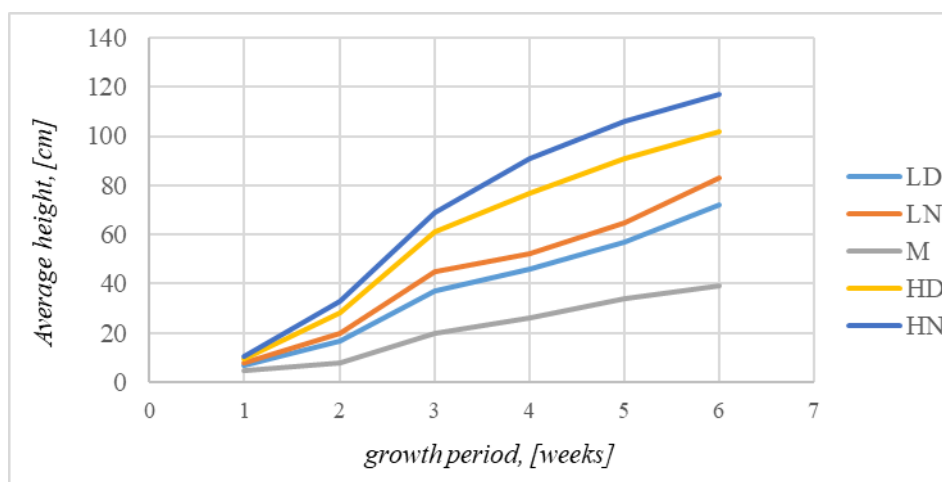
BF values for Zn, Mn, Ni, Cu, Fe, and Pb were higher in the case of the salty lands fertilized with pig manure. In the case of the lots situated on the salty land and in the ash dump, the bioaccumulation factors for Zn and Mn were higher than 1, with values situated between 3.06-4.42 for Zn and 1-1.39 for Mn.

BF for Ni was situated in the range 1.4 to 1.55 in the case of the salty land and between 0.73 - 0.84, in the case of the ash dump.

The bioaccumulation factors for Cu, Fe, and Pb were lower than 0.5. The lowest values were registered for Pb, of 0.02, and, in the case of the lots in the ash dump, of 0.03. The obtained results are similar with the data from previous studies, which present the following values for BF in *Salix polaris*: Zn 0.9-6.2; Mn 0.14-1.3; Ni 0.2-1.6; Cu 0.09-0.8; Fe 0.01-0.1; Pb 0.001-0.2 [21, 24, 25].

The BF difference between various fertilization treatments may be an effect of the pH in the rhizosphere zone. This is in correlation with previous studies that report that the soil pH greatly influences the concentrations of metals accumulated in the plants [26-28].

The pH of the lots fertilized with manure was between 5.4 and 5.7, which leads to the increase of metals mobility and availability for plants. According to Qaswar et al. [16], the process of manure degradation in the soil may reduce the pH [16]. Therefore, *Salix sp.* absorbs the polluting metals instead of macroelements vital for growth and development, so it appears as a nutritional imbalance. This is manifested by slowing down the plant growth rate, drying and premature fall of the leaf, reduction of the photosynthesis process, etc. All these effects lead to a culture of *Salix sp.* less developed on salty land (Fig. 7).



**Fig. 7.** The average height variation of *Salix sp.* plants in six months, on the experimental lots and the witness lot

The pH of the lots fertilized with stabilized sludge was between 7.3 and 7.7. At these pH values the possibility of the formation of insoluble hydroxides and carbonates increases. In this case, the metals are less accessible to plants.

The stabilized sludge used as a fertilizer is relatively rich in organic matter, therefore it contributes to the soil's organic matter increase and also the slower and gradual release of heavy metals for plants, due to the absorption

#### *The Salix sp. cultures on the experimental lots*

The monitoring of the *Salix sp.* cultures took place over six months of vegetation, by the "average height" indicator (Fig. 7). For comparison, there is given the growth variation of *Salix sp.* on an unfertilized lot in the ash dump (the witness sample - M).

From analyzing the *Salix sp.* cultures over six months of vegetation was established that the stabilized sludge determines a more vigorous growth and development of the plants in comparison with the plants from the lot fertilized with manure. This development of the plants is correlated also with the higher content of nutrition elements (N, P, K) in the lot fertilized with stabilized sludge. After the II<sup>nd</sup> phenological stage of vegetation, the *Salix sp.* culture in the ash dump was better developed than the one raised on the salty land. The plants were vigorous and showed a healthy aspect, presenting no symptom of the sickness of the willow leaves or plants.

Following the evolution of the plants, we can

## CONCLUSIONS

The results of the experimental studies confirm that *Salix sp.* presents a high accumulation capacity for Zn, Mn, and Ni. For all experimental lots, the bioaccumulation factors for Zn, Mn, and Ni presented values higher than 1. The maximum values of the bioaccumulation factor were found for Zn, between 3.06 and 4.42. These results recommend *Salix sp.* as a bioaccumulator of Zn, Mn, and Ni, the plant could be used for the bioremediation of polluted soil.

The bioaccumulation factors for Cu, Fe, and Pb were lower than 0.5. The smallest values were for Pb, 0.02 and 0.03, for the lots situated in the

and complexation processes of metals on colloidal surfaces or of other components from the sludge. According to previous studies, this might be the reason for the decrease of the bioaccumulation factor of metals in the case of the ash deposit fertilized with stabilized sludge [29-31]. This is explained by the fact that the culture of *Salix sp.* from the ash dump fertilized with stabilized sludge is more vigorous and has a healthy aspect (Fig. 7).

conclude that the effect of the salinity stress actions on the *Salix sp.* plants is higher or smaller depending on the soil, humidity, precipitations level, the quantity of sodium over the total quantity of absorbed ions, fertilization level and the ratio between the nutrition substances present in the soil.

The cultures on the witness experimental lots developed harder, the average heights of the plants were 52-76 % shorter than in the case of the cultures raised on the lots from the ash dump fertilized with stabilized sludge. The culture on the witness lots dried out in 9-12 months from its plantation.

Applying stabilized sludge and manure over a long period determines the raise of the nutrient content in the soil and enhances its physical and chemical characteristics (pH, soil porosity, the capacity of retaining water, etc.), determining an optimal growth and development of cultures [26, 27].

ash dump.

After the II<sup>nd</sup> phenological stage of vegetation, the culture of *Salix sp.* from the ash dump was much better developed than that one grown on salty soil. The plants were vigorous and presented healthy aspects, no symptoms of the disease of leaves on willow plants were observed.

The salty land causes a higher accumulation of metals in *Salix sp.* This increase is due to the pH values of the soil in the rhizosphere area, which favors the increase of mobility and the availability of metals for plants.

## ACKNOWLEDGEMENTS

The present research was financially supported by the Romanian National “Nucleu” Program (PN 16 25 03 08).

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