10th Eastern European Young Water Professionals Conference

BOOK of ABSTRACTS

New Technologies in Water Sector

7-12 May 2018 Zagreb, Croatia

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IWA Young Water Professionals – the international water association

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Faculty of Civil Engineering

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Zagreb 2018

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ISBN: 978-953-8168-23-9

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Zagreb 2018

Removal of Heavy Metal Ions from Landfill Leachate by Phytoremediation Using *Eichhornia crassipes*

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Abstract
The paper presents the results of the treatment of landfill leachate by applying the method of phytoremediation using hyacinth *(Eichhornia crassipes)*. The leachate was sampled from the Municipal Landfill in Novi Sad, while the common water hyacinth was purchased from the Nursery of flowers and conifers in Ugrinovci. During the research, we monitored the removal efficiency of four heavy metals, i.e., chromium, cadmium, nickel and zinc. Despite the presence and the metabolic processes of *Hirudinea*, as well as the accelerated degradation of the applied biomass, significant removal of heavy metals from the treated leachate has been achieved. After completion of the experiment, it was concluded that the removal efficiency of chromium is 81%, cadmium 80%, nickel 62% and zinc 80%.

Keywords
Municipal solid waste landfill; phytoremediation; leachate; *Eichhornia crassipes*

INTRODUCTION

Leachate represents an entity affected by a complex of factors, both within the very body of the landfill (age of the landfill, morphological composition of waste, temperature, moisture content, migration flow of liquid, waste treatment technologies before disposal, thickness of the landfill body and waste decomposition phases), and outside of it (meteorological parameters with emphasis on volume of annual precipitation and seasons changes). The process of forming a landfill filtrate includes the dissolution of solid substances in water, whereby it is percolated through the body of the landfill, and the separation of dissolved or suspended matter formed by biological and chemical processes that are inevitably occurring within the body of the landfill. The basic physical properties of landfill leachate are reflected in the dark-brown colour of the filtrate and a strong smell, while the high concentration levels of the pollutants and the BPK_5 values in the amount of 5,000 mg L^{-1} denote representative chemical parameters thereof. Pollutants found in municipal solid waste landfill leachate are classified into four groups: dissoluble organic components (volatile fatty acids, variations of humic and fulvic compounds), inorganic macro-components (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+}, Fe^{2+}, Mn^{2+}, Cl^{-}, SO_4^{2-}, HCO^{3-}), heavy metals (Cd^{2+}, Cr^{3+}, Cu^{2+}, Pb^{2+}, Ni^{2+}, Zn^{2+}) and xenobiotic organic components (hydrocarbons, phenols, chlorinated aliphatic compounds, pesticides and dioctyl phthalates) (Kjeldsen *et al*., 2002). Due to a complex chemical composition, different methods of handling and treatment of landfill leachate have been developed, and we distinguish: directing leachate (wetlands, recirculation, comparative treatment of leachate with municipal waste waters within the general sewage system), biological treatment (aerobic-aerated lagoons, active sludge systems, sequencing batch reactors (SBR), biodisc process (MBBR), drip filters; anaerobic-hybrid filtration, fluidized bed filtration, anaerobic filtration, anaerobic SBR, anaerobic digester with an upstream flow through a sludge layer), physical and chemical treatment (coagulation/flocculation, flotation, chemical deposition, adsorption, ammonia nitrogen removal by air blowing, chemical oxidation, ion exchange and electrochemical processes) and membrane filtration (reverse osmosis, nanofiltration, ultrafiltrationand microfiltration) (Abbas et al., 2009; Renou et al., 2008). An alternative method – phytoremediation proved to be especially efficient in comparison to conventional landfill leachate treatment, from the aspect of reduction of heavy metal concentration.

Phytoremediation represents an economically sustainable and simple method that is defined as a designed use of green plants for the purpose of extraction, accumulation and stabilization of pollutants. Biological, chemical and physical processes taking place within phytoremediation include the absorption, accumulation and metabolism of the substrate in the plant, or remediation is carried out by means of activities of microorganisms present in the rhizosphere (Hinchman et al., 1997). The mentioned processes take place in the functional wholes of the plant, in leaves, stems, seeds and roots, whereby efficient absorption and accumulation of heavy metals, inorganic substances and organic matter is achieved. In the root zone, the degradation and immobilization of organic compounds are efficiently carried out, while accumulation and metabolism of the substrate are the characteristic processes occurring in the tissue of the sprout (Pichtel, 2007). Improvement of the efficiency of the phytoremediation method is achieved by applying hyper accumulator plant species.

Accumulators represent a group of plants that accumulate significant amounts of heavy metals, regardless of their concentrations in contaminated soil. The mechanism of the action is based on the accumulation of heavy metals in plant organs or in the form of metabolic products, due to which the negative effects of the stated activities are less harmful to the plant. Hyperaccumulators represent a group of plants that absorb toxic elements accumulating them in the aboveground part of the plant in volumes much higher than the usual concentrations, whereby the negative effects of the stated activity on the plants are minor or insignificant (Baker and Brooks, 1989). Almost all plant species from the group of hyperaccumulators that are now in use were discovered on mining pits rich in minerals. Such plants are endemic with respect to this land, indicating that hyperaccumulation represents a physiological adaptation to exposure to higher concentrations of metals (Baker and Brooks 1989). Most of the hyperaccumulators discovered so far are limited to few geographic locations. An excellent example of hyperaccumulation was discovered in the latex of the *Sebertia acuminate* tree, originating in New Caledonia, which can accumulate more than 20% of nickel. The accumulation of radioactive copper ions from contaminated soil is successfully carried out using the “copper flower” (*Haumaniastrum katangense*) (Baker and Brooks, 1989). Also, there is a significant number of plants identified the use of which successfully reduces radioactive caesium isotope 137 (Dushenkov et al., 1999; Lasat et al., 1998). Baker and Brooks (1989) identified 145 nickel hyperaccumulators, 26 cobalt hyperaccumulators, and 8 manganese hyperaccumulators. Within the phytoremediation method, some other hyperaccumulators are also used, such as the Field Pennycress (*Thlaspi calaminare*), the use of which accumulates more than 10% of zinc, Alyssum (*Alyssum bertolonii*), which is characterized by the capacity to accumulate up to 10% of nickel, accumulation up to 3 % of chromium is achieved using Bacopa (*Pimela suteri*) and Manuka (*Leptospermum scoparium*); accumulation of up to 3% of uranium is achieved by applying endemic plant species originating in New Zealand, *Uncinia leptostachya* and *Coprosma arborea*, while the accumulation of up to 1% of mercury can be achieved by using Paper birch (*Betula papyrifera*) (Pitchel, 2007). Aquatic macrophytes, water hyacinth (*Eichhornia crassipes*, the *Pontederiaceae* family) in particular, have proved to be highly efficient in the reduction and monitoring of heavy metals (Xian et al., 1989). High efficiency of the method of leachate phytoremediation from the landfill site of Moshi, India, using *Eichhornia crassipes* was achieved from the aspect of reducing the concentration of heavy metals - chromium in the amount of 51.66%, nickel in the amount of 95.65% and zinc in the amount of 92.31%, while by applying the same treatment of leachate from the landfill site of Pulau Burung, Malaysia, reduction in zinc concentrations was achieved with a 64.6% efficiency (Akinbile et al., 2012; Verma et al., 2015).

EXPERIMENTAL PART

Material and chemicals

The leachate used in the research was collected from the Municipal Landfill in Novi Sad. For the purposes of the experiment, 15 L of leachate from a lagoon within the landfill site were sampled. The sampled leachate was stored in three containers of 6.25 L each made of materials for commercial use (polyvinylchloride). Due to the variability and susceptibility to external effects, the measurement of two parameters, conductivity and pH value, was realized in-situ using a portable multiparameter device for measuring temperature, pH value, conductivity and quantity of diluted oxygen "Multi 340i" produced by WTW GmbH. After implementing the procedure, samples were transported in two portable refrigerators to the accredited Laboratory for landfill, wastewater and air monitoring of the Faculty of Technical Sciences.

For the purpose of the experiment, 1.5 kg of the biomass of water hyacinth was delivered from the Nursery of flowers and conifers in Ugrinovci. Water hyacinth was grown in sanitary lagoons of the Nursery over a 14-day interval, at a temperature range of 15 to 25°C The delivered biomass was packed in a polyvinyl chloride bag, which, after closing, was additionally secured with a layer of thin polyethylene foil with the aim of minimising evapotranspiration and circular water cycle in the package, after which the package was placed in a cardboard box to prevent fracture.

Before setting up the experiment, the preparation of sampled leachate was carried out with the aim of adjusting the parameters, such as the pH value and the concentration of heavy metals. During the in-situ measurement of the parameter values, after sampling of leachate, the conductivity value was at 2.94 mS cm^{-1}, while the measured pH value amounted to 8.95. The pH adjustment to the value of 6.37 was carried out with the aim of adapting the stated parameter to the characteristic values of the "young" leachate and improving the process of phytoextraction of heavy metals by means of the present plants. Adjustment of the pH value of the leachate sample was carried out in a barrel of 15 L capacity made of high-density polyethylene using diluted hydrochloric acid (HCl 1%) and diluted ammonium hydroxide (NH4OH 1%), whereby the monitoring of the movement of the value of the said parameter was carried out using the portable multiparameter device "Multi 340i", manufactured by WTW GmbH.

The primary task of the experiment was to monitor the efficiency of the method of the leachate phytoremediation using *Eichhornia crassipes*, from the aspect of phytoextraction of four heavy metals - cadmium, chromium, nickel and zinc. Due to low concentrations of heavy metals in the initial sample, as well as the defined limit of detection of the flame atomic absorption spectrophotometer "Thermo Scientific S Series", which for cadmium amounts to 0.0015 mg L^{-1}, for chromium 0.006 mg L^{-1}, for nickel 0.01 mg L^{-1}, and for zinc 0.001 mg L^{-1}, spiking was performed. The process of spiking was carried out after the process of adjusting the pH value by adding 2 mg L^{-1} of the standard solution of each - cadmium, chromium, nickel and zinc of 1000 mg L^{-1} initial concentrations, manufactured by Proanalytica.

After adjusting the parameters, 15 L of leachate was poured into a plastic trough made of high-density polyethylene, of 54 x 23.5 x 38.5 cm in size. Finally, 1.5 kg of water hyacinth biomass was placed in the plastic trough containing the sample of leachate.

Course of the experiment
The experiment was carried out in laboratory conditions, over a 14-day interval. After the experiment was set, 100 mL of treated leachate was sampled on the third, the seventh, the eleventh and the fourteenth day, after which the stabilization with concentrated nitric acid was applied, followed by storing in the refrigerator until the analysis.

Applied methods
For the purpose of determining the pH value and conductivity on a daily basis, within the time interval of conducting the experiment, a portable multiparameter device "Multi 340i", was used. For the spectrophotometric determination of the concentration of sulphate ions and nitrate ions within the experiment, UV/VIS spectrophotometer, the DR 5000 model - portable, manufactured by Hach-Lange, was used. The flame atomic absorption spectrometer "Thermo S Series" with flame based on air combined with acetylene, manufactured by ThermoFisher Scientific, was used to determine the concentration of four heavy metals in the leachate sampled during the experiment on four occasions.

RESULTS AND DISCUSSION
Conductivity and pH values of the tested leachate were monitored in the period from the third to the seventh day and from the 10th to the 14th day of the experiment. Comparison of the measured values of the parameters at the specified time intervals is shown in Figure 1 and Figure 2.

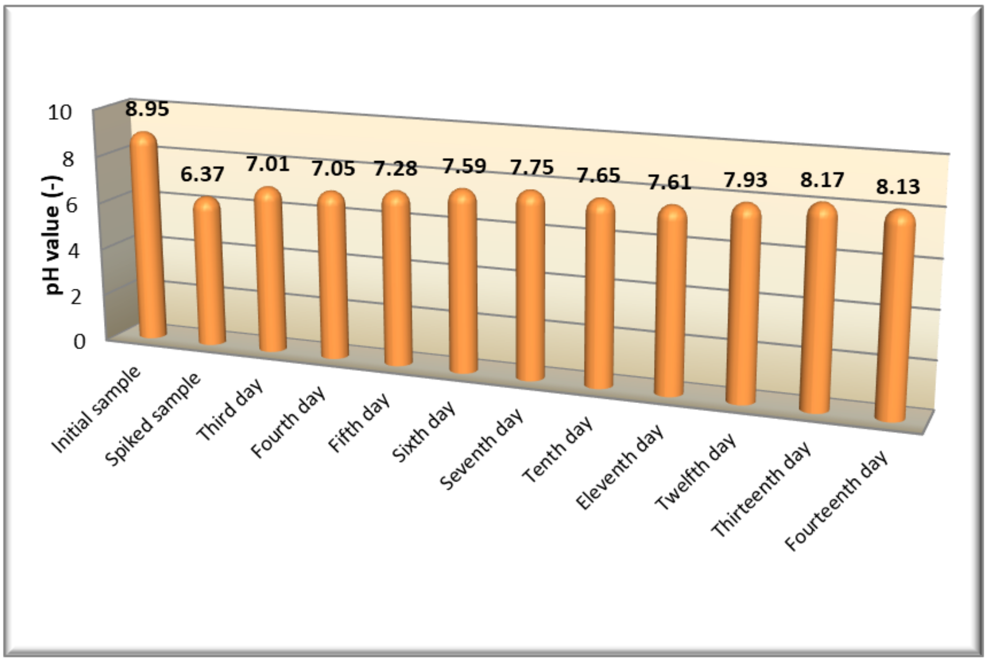

Figure 1. Measured pH values over a 14-day interval of research

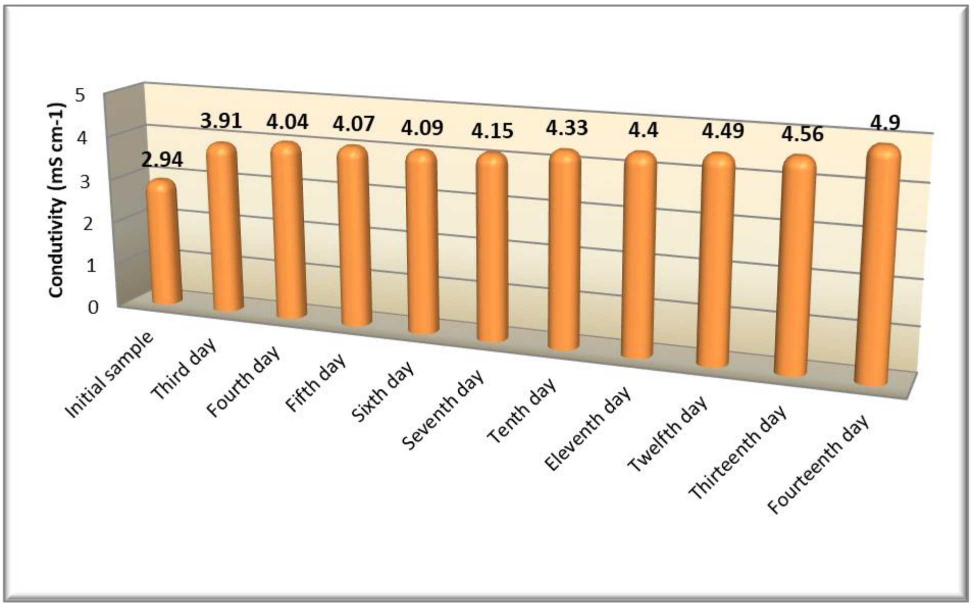

Figure 2. Conductivity values measured over a 14-day interval of research

Comparison of the initial (spiked) and final values of the concentration of four heavy metals - chromium, cadmium, nickel and zinc, as well as the efficiency of the removal of metal over a 14-day interval of research is shown in Figure 3 and Figure 4, respectively.

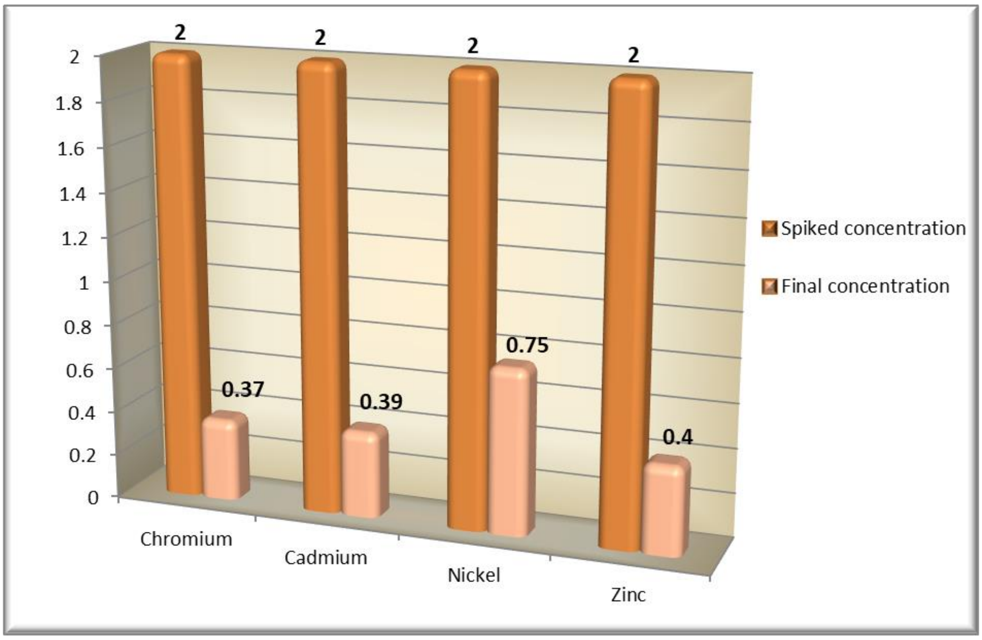

Figure 3. Initial (spiked) and final values of concentrations of four heavy metals, chromium, cadmium, nickel and zinc

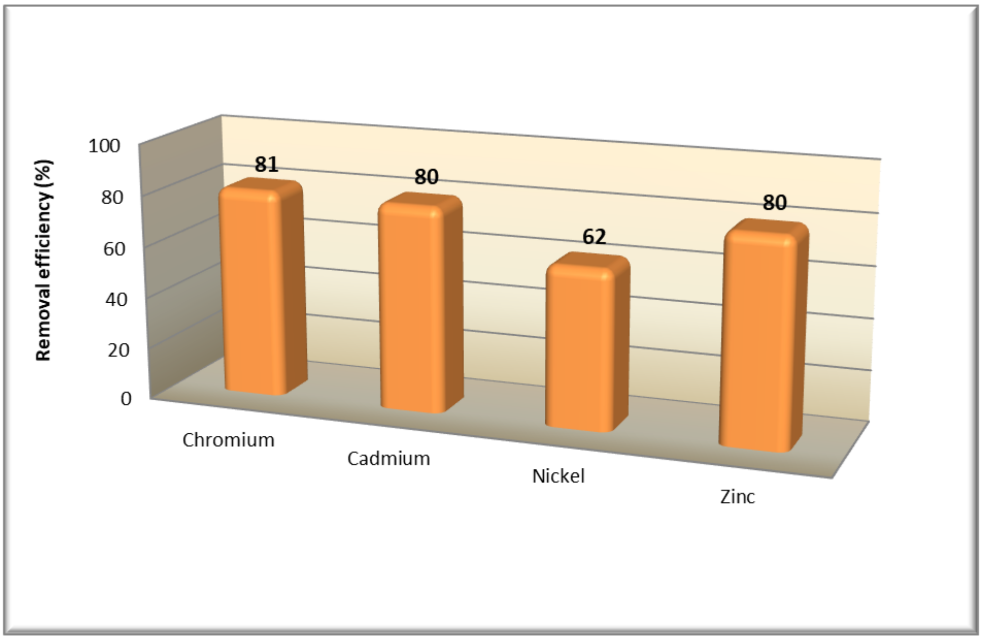

Figure 4. Efficiency of the removal of four heavy metals, chromium, cadmium, nickel and zinc, from the landfill leachate over a 14-day interval of research

The maximum efficiency of metal phytoextraction from landfill leachate using *Eichhornia crassipes* was achieved on the third day of the experiment carried out. The degradation mechanism of water hyacinth biomass, which is an inevitable consequence of the process of phytoextraction of heavy metals present, that is, their accumulation in the root system, as well as the distribution to sprouts and leaves of the plant species, has been further accelerated by the presence of biomass of leeches using the plant species as food. In accordance with the above, the phenomenon of gradual degradation of water hyacinth biomass was noticed on the fourth day of the research. A continuous increase in the pH value and conductivity over the time interval of the experiment is a consequence of the degradation of water hyacinths, as well as the release and dissolution of their constituent elements due to the application of the plant species for the purpose of feeding the present biota.

Despite the presence and the metabolic processes of *Hirudinea*, as well as the accelerated degradation of the applied biomass, significant removal of heavy metals from the treated leachate has been achieved. After completion of the experiment, it was found that the efficiency of chromium removal amounts to 81%, cadmium 80%, nickel 62% and zinc 80%.

CONCLUSION

After completing the research on applying the method of phytoremediation of leachate from the municipal solid waste landfill using *Eichhornia crassipes*, the following conclusions can be drawn:

- Solid waste landfill leachate can be treated by applying the stated method;
- The presence of macrobenthic invertebrates (*Hirudinea*) in treated leachate increases the intensity of degradation and the release of the constituent elements of the used biomass, resulting in an increase in the value of the concentration of sulphate ions and ammonium nitrogen, and consequently the values of parameters such as conductivity and pH value;
- Reduction of the concentration of the present heavy metals is the most intensive during the first three days of the experiment, with the maximum absorption capacity of the applied biomass still not affected by *Hirudinea*;
- The efficiency of the removal of heavy metals continuously, yet less intensely, was increasing until the end of the experiment.

The efficiency of the method of leachate phytoremediation can be improved in several ways. By applying a number of different hyperaccumulator species, the maximum effect of removing heavy metals at a shorter time interval is achieved. Despite the necessary temperature range from 20°C to 30°C, the application of phytoremediation systems is also possible in the winter period by using plant species with dominant overground biomass, which, as a cover layer, has a protective role for the root system. The research on the efficiency of applying certain plant species in phytoremediation of landfill leachate contributes to the improvement of existing knowledge in the mentioned area and to designing future phytoremediation systems.

ACKNOWLEDGMENTS

This environmental study has been financially supported by Ministry of Education, Science and Technological Development, Republic of Serbia (Project III46009 and TR34014).

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