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# Temperature and pH influence on the efficiency of trace metals leaching from sewage sludge with EDTA solution

Beata Karwowska

*Faculty of Infrastructure and Environment, Department of Chemistry, Water and Wastewater Technology, Częstochowa University of Technology, Dąbrowskiego 69, 42-200 Częstochowa, Poland, Tel. +48 343250491; Fax: +48 34 3250 496; email: bkarwowska@is.pcz.pl*

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**ABSTRACT**

Sewage sludge cumulates harmful, non-biodegradable trace metals. For this reason further management could be problematic. Extraction of metals with EDTA is one of the way to decrease of their content in sewage sludge. The aim of the presented studies was an evaluation of impact of pH and temperature changes on the efficiency of selected metals (Zn, Cu, Ni, Cd and Pb) elution from sewage sludge with EDTA solution. Efficiency of EDTA was estimated after shaking of a sludge sample with 0.1 M aqueous solution. Initially with solutions adjusted to pH in the range 3.0 to 12.0 at 20°C, in the second step at different temperatures in the range 20°C–70°C at pH equal to 4.5. EDTA solution was an effective extractant for the heavy metals removal from sewage sludge. The best extracted metal was zinc and the lowest efficiency was observed for copper. Extraction of metals decreased when pH of washing solution increased. For the analyzed extraction processes, optimal pH was in the range 3.0–6.0 (efficiencies of metals removal were in the range: 30%–32%, about 2%, 26%–27%, 34%–44% and 37%–39% of total amount for Zn, Cu, Ni, Cd and Pb, respectively). For experiments at different temperatures, the amount of removed metal raised with temperature increase. The lowest amount of metal was extracted at 20°C (31%, 2%, 26%, 40% and 38% for Zn, Cu, Ni, Cd and Pb, respectively) and the highest efficiency was detected at temperature 70°C (76%, 11%, 38%, 79% and 62% for Zn, Cu, Ni, Cd and Pb, respectively).

*Keywords:* Heavy metals; Sewage sludge; EDTA extraction

## 1. Introduction

Heavy metals are common pollutants in the environment, widely spread over the world. Some metals (Zn, Cu, Se) are nutritionally essential elements at low levels, but toxic at higher levels, and others (Pb, Cd, As, Hg) have no known biological functions [1]. Trace metals are toxic for organisms in any element of the environment, because of presence in soil, they could enter into the food chain after uptake by plants, through the animal organism to the human body [2,3]. Moreover, the mobility, bioavailability and ecotoxicity of heavy metals depend on their chemical fraction in sewage sludge [4–6]. Their presence in wastewaters implicates transfer to the sewage sludge after typical treatment processes. Contamination of environment by heavy metals from wastewater as well as from sewage sludge becomes to be a serious worldwide problem. The problem of their abundance, toxicity and persistence in the environment is a target of numerous researches [3,4,7].

Wastewater treatment processes generate dramatically increasing amount of sewage sludge. It is noteworthy that the application of sludge in agriculture has many advantages. High content of organic matter improves the physical and biochemical properties of the soil. Solid matter of sludge contains nutrient elements for plants, including nitrogen, phosphorus, potassium, what could reduce the amount of

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artificial fertilizers used on farms [8,9]. Sewage sludge may contain organic, inorganic and biological pollutants from the wastewaters of households, commercial establishments and industry facilities and compounds added or formed during various wastewater treatment processes. Such pollutants include inorganic (e.g., heavy metals and trace elements), organic contaminants (e.g., PAH, PCB, dioxins, pharmaceuticals and surfactants) and microorganisms (e.g., bacteria, viruses and parasites) [10]. The presence of trace metals in sewage sludge may affect its use for natural and agricultural purposes. Unlike organic pollution that can be biodegraded with time or can be combusted, metals are not degradable, accumulate in living organisms and remain a real potential threat to the environment and human health [11,12]. Therefore, lowering the content of trace metals in sludge is a natural way to achieve environmental friendly material.

Many methods have been studied to reduce heavy metals content in sludge, including chemical precipitation, filtration, ion exchange, electrochemical treatment, adsorption on activated carbon, membrane technologies, etc. [11,13]. Chemical extraction is one of typical and popular method for trace metals removal from sewage sludge. Different compounds were used for metals removal from sewage sludge. There were reported inorganic acids such as HCl, $H_2SO_4$, $HNO_3$ [11,14], organic acids such as citric or oxalic acid [11,15] and chelating agents such as EDTA, nitrilotriacetic acid (NTA), diethylenetriaminepentaacetic acid (DTPA) [11,16–19]. Chelating agents and especially EDTA (ethylenediaminetetraacetic acid) seems to be very promising. In comparison with other chelating agents, it presents some advantages, mainly a high level of complexing capacity with respect to heavy metals and a large stability as complexing agent [16,20].

To the author's knowledge, the detailed analysis of changes in efficiency of trace metal removal from sewage sludge with chelators solution in dependency on the pH of extractant or the temperature of the process is not documented. There is some information about the influence of pH but in very limited range [11,21,22] and accidental information on the temperature effect generally for bioleaching processes [23].

The purpose of the actual studies was the evaluation of the impact of changes in pH and temperature on the efficiency of selected metals (Zn, Cu, Ni, Cd and Pb) extraction process from sewage sludge using EDTA aqueous solution.

## 2. Material and methods

All chemicals used in this study were of analytical grade and were used without any further purification. Solutions were prepared using demineralized water. All presented results are the mean values from triplicate tests performed, with a standard deviation less than 5%. Statistical calculations were based on the confidence level equal or higher than 95%.

The analyzed material was dewatered final sewage sludge after filter press station. Sewage sludge samples were taken from a municipal wastewater treatment plant in central Poland in 2016. The inflow to the plant is about 9,200 $m^3/d$ and treatment is conducted by activated sludge method, with the use of nitrification, denitrification and biological dephosphatation processes. Properties of sludge samples, such as pH, hydration, dry matter (d.m.) content, ignition losses and residual, were determined following the standard analytical procedures. pH was detected using electrometric method according to the procedure described by Clesceri et al. [24], hydration and dry matter content were calculated using gravimetric methods by comparing masses of sludge sample before and after drying at 105°C, ignition loses and ignition residue were determined after combustion of dry sample at 550°C and comparing masses of sample before and after combustion.

The samples of sewage sludge were air-dried under laboratory condition (temperature 20°C), next dried in the dryer (105°C). Then sludge samples were homogenized in an agate mortar and passed through a stainless sieve with 0.4 mm diameter meshes and kept in a plastic container for further analyses. Three different samples of the same sludge were prepared for the analyses.

The characteristics of the sewage sludge used for the experiments are given in Table 1. The total content of selected heavy metals (Zn, Cu, Ni, Cd and Pb) in sewage sludge was studied after sample mineralization at 120°C with the mixture of concentrated acids: $HNO_3$ and HCl (1+3 – aqua regia) for 2 h. Extraction of selected heavy metals (Zn, Cu, Ni, Cd and Pb) from sewage sludge was carried out by washing 5 g sewage sludge samples with 50 $cm^3$ portion of 0.1 M EDTA an aqueous solution. The samples were shaken for 6 h, then the extracts were filtered through the paper filter.

The first step of studies – evaluation of the pH effect on heavy metals extraction – was carried out at temperature 20°C. Metals were washed with water and 0.1 M EDTA solutions adjusted to pH in the range from 3.0 to 12.0. pH of 0.1 M EDTA an aqueous solution equals to 4.5 and decrease of pH values was a result of acidification of solution with concentrated nitric acid. EDTA is a compound weakly soluble in water. Acidification of its solution under pH 3 caused precipitation of EDTA particles, so driving the process at lower pH seems to be unjustified. pH 6, 9 and 12 are the multiplication of first difference. Additional analysis was done under neutral environment at pH 7. The alkalization of solution was obtained by adding 2 M NaOH solution. The increase in pH was stopped at value 12 after reaching of a half of an initial extraction efficiency for majority of metals.

Table 1
Physical and chemical parameters of analyzed municipal sewage sludge

| Parameter | | Value |
|---|---|---|
| pH | | 6.8 |
| Hydration, % | | 85.3 |
| Dry matter, $g \cdot kg^{-1}$ | | 147.0 |
| Ignition losses, $g \cdot kg^{-1}$ dm | | 379.1 |
| Ignition residue, $g \cdot kg^{-1}$ dm | | 620.9 |
| Trace metals total content, $mg \cdot kg^{-1}$ dm | Zn | 2,121.0 |
| | Cu | 197.3 |
| | Ni | 39.6 |
| | Cd | 6.3 |
| | Pb | 79.2 |

d.m. – Dry matter.

In the second step of the project – evaluation of temperature dependence of extraction – 0.1 M EDTA solution with pH equal to 4.5 (without any pH adjustment) was used for leaching of analyzed metals at temperatures in the range from 20°C to 70°C. The samples were warmed up in water bath with shaking. Initial experiment was performed at 20°C (ambient temperature), the next temperature was changed by 10°C up to reaching 70°C, when the intensive evaporating of water from bath as well as from the samples was observed.

The content of metal ions in liquids after mineralization as well as after EDTA extractions was detected by an atomic absorption spectrometry method, using a spectrometer novAA 400, Analytic Jena, Germany.

The influence of temperature and pH on the efficiency of EDTA solutions for metal extraction from sewage sludge was analyzed using obtained values of metal concentration in extract and related amount of removed metal in comparison with 1 kg of dry matter of sludge.

## 3. Results and discussion

### *3.1. Physical and chemical characteristics of analyzed sewage sludge*

The basic physical and chemical parameters characteristic of studied sewage sludge are summarized in Table 1. The analyzed material was neutral, with pH equal to 6.8. The hydration and organic matter content (determined as ignition losses) of the sample was 85.3% and 37.9%, respectively.

The sludge sample contained the highest amount of zinc: 2,121 mg·kg$^{-1}$ d.m., copper, nickel and lead contents were significant but relatively lower: 197.3, 39.6, 79.2 mg·kg$^{-1}$ d.m., respectively. Cadmium content was the lowest: 6.3 mg·kg$^{-1}$ d.m.

The total content of mentioned heavy metals in studied sewage sludge was lower than amounts recommended in legislative requirements for sewage sludge used in agriculture, for land reclamation to agricultural and non-agricultural purposes [25,26], but under favourable conditions in environment their mobility and bioavailability could be harmful for living organisms in ecosystem [1,12,13].

### *3.2. Temperature influence on efficiency of trace metals extraction with EDTA*

The changes of efficiency of heavy metals extraction from sewage sludge samples in dependency on temperature increase are presented in Figs. 1–5 for each metal separately and are summarized in Table 2. The amount of each metal removed from sewage sludge increased when temperature of process raised in the studied range of temperature.

Zinc removal efficiencies at different temperatures are presented at Fig. 1 as well as in Table 2. At temperature 20°C EDTA extracted 663.7 mg·kg$^{-1}$ d.m., what was 31% of total content of zinc in studied sewage sludge and with the increase of temperature up to 40°C the amount of released metal increased slightly reaching 754.5 mg·kg$^{-1}$ d.m. (36% of total zinc content). The efficiency of zinc extraction with using 0.10 M EDTA on the comparable level were reported by authors of similar papers concerning sewage sludge [18,19] and contaminated soil [27,28]. Starting from 50°C, more significant increasing of EDTA extraction efficiency was observed and at temperatures 50°C, 60°C and 70°C the amounts of zinc removed from sewage sludge were: 890.3; 1,138.9 and 1,614.4 mg·kg$^{-1}$ d.m. (42%, 54% and 76% of total), respectively. At a temperature of 70°C, EDTA solution extracted about 2.5 times more zinc than at 20°C.

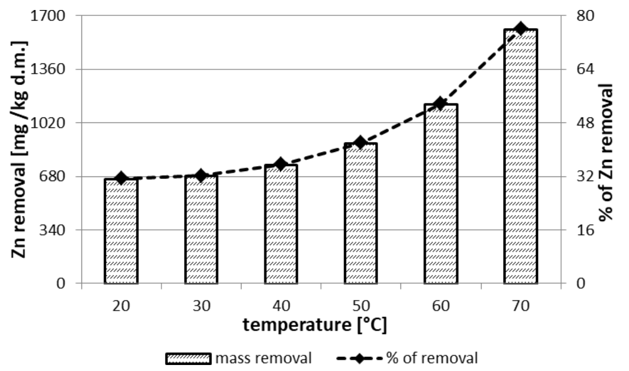


Fig. 1. Removal of zinc from municipal sewage sludge as a function of extraction process temperature.

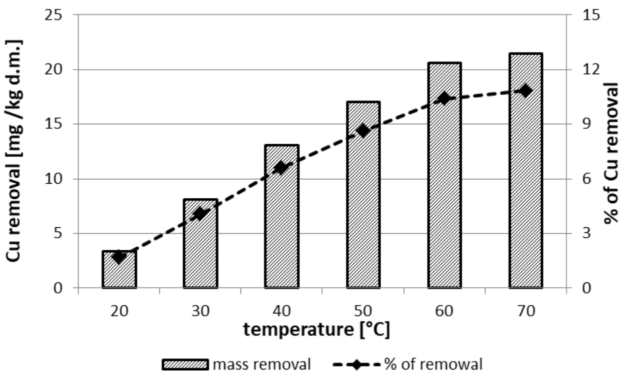


Fig. 2. Removal of copper from municipal sewage sludge as a function of extraction process temperature.

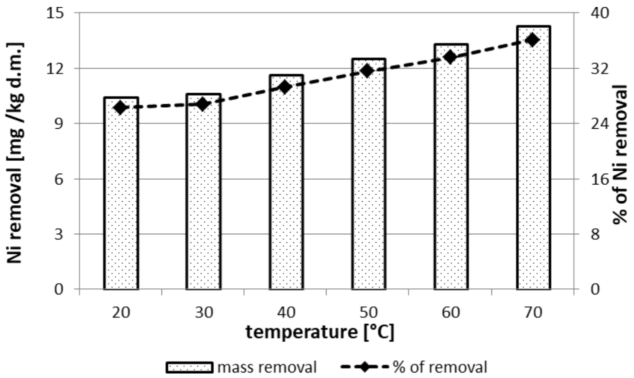


Fig. 3. Removal of nickel from municipal sewage sludge as a function of extraction process temperature.

Fig. 2 shows the tendency in the copper amount removed from sewage sludge according to increasing in temperature. Detailed values of mass and percentage efficiency of Cu removal are presented in Table 2. At 20°C, the observed quantity of copper was 3.5 mg·kg$^{-1}$ d.m., what was about of 2% of total metal concentration in analyzed material. Initially changes of copper content eluted from sludge were significantly higher. The change in temperature from 20°C to 30°C caused more than two times increase in extraction efficiency, next temperature steps resulted in a lower increase of washed metal amount reaching 8.0, 13.0 and 17.1 mg·kg$^{-1}$ d.m. at temperatures 30°C, 40°C and 50°C, respectively. At next steps, much lower rises were observed and at 60°C and 70°C the amounts of extracted copper were 20.5 and 21.1 mg·kg$^{-1}$ d.m. what was 10% and 11% of the total copper content. The values obtained at temperatures 60°C and 70°C were very similar and could be concluded that further increase in temperature would be non-effective for copper extraction and optimal temperature for mentioned metal extraction was 60°C.

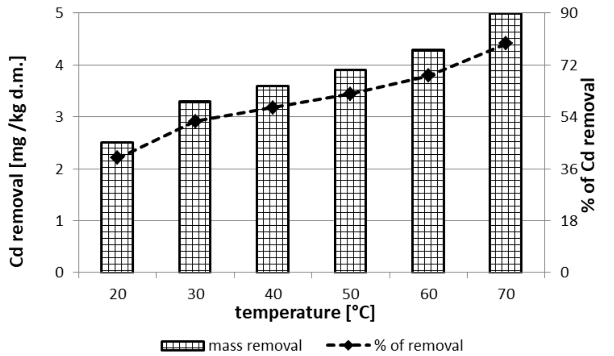


Fig. 4. Removal of cadmium from municipal sewage sludge as a function of extraction process temperature.

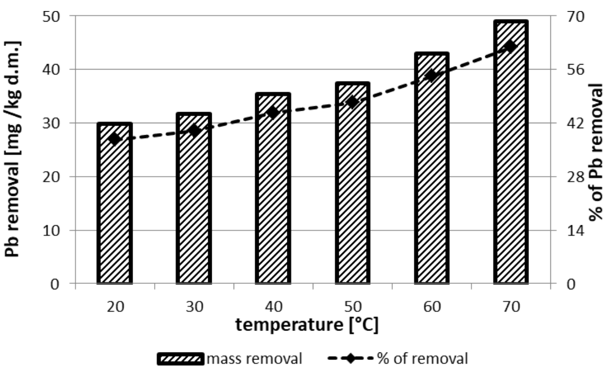


Fig. 5. Removal of lead from municipal sewage sludge as a function of extraction process temperature.

The efficiency of nickel removal (Fig. 3, Table 2) changes was similar to zinc behaviour. At a temperature of 20°C the detected amount of metal washed from sewage sludge was 10.4 mg·kg$^{-1}$ d.m., what was 26% of the total nickel content. At temperature 30°C the released amount of nickel was very similar to previous: 10.6 mg·kg$^{-1}$ d.m. (27% of total Ni content). Next temperature steps were more effective and at temperatures 40°C, 50°C, 60°C and 70°C the washed contents of Ni were: 11.6, 12.5, 13.3 and 14.3 mg·kg$^{-1}$ d.m. (29%, 32%, 34% and 38%), respectively. The change at the whole temperature range was approximately 40%, what suggests, that energy expenditures in a case of Ni were not effective.

Cadmium removal efficiency increased systematically with the increase of extraction process temperature as shown in Fig. 4 and Table 2. At 20°C the amount of washed metal was equal to 2.5 mg·kg$^{-1}$ d.m. and was 40% of initial cadmium content in studied sewage sludge sample. Comparable efficiency for cadmium removal was reported earlier for sewage sludge [18,19], bottom sediments [15–17] as well as soil [27,28]. The highest change in amount of removed nickel was observed after first temperature step (0.8 mg·kg$^{-1}$ d.m.), next temperature steps increased the amount of leached nickel by about 0.3–0.4 mg·kg$^{-1}$ d.m. and at 70°C the observed quantity of removed metal was almost twice its initial value, reaching 5.0 mg·kg$^{-1}$ d.m. (79% of total nickel content in sewage sludge). It has to be noticed that total amount of cadmium was at the insignificant level and the interpretation of its behaviour is only an approximation.

The plot of amount washed lead vs. temperature of the extraction process is presented in Fig. 5 as well as in Table 2. The efficiency of lead removal from the analyzed sewage sludge was close to cadmium and zinc. The shape of changes could be also compared with zinc and cadmium. At an initial temperature of 20°C, the amount of extracted lead was equal to 29.8 mg·kg$^{-1}$ d.m., what constituted about 38% of its total concentration. The removal of lead by EDTA on the level about 40% was determined previously by another groups of researches [15–19,27,28]. The increase in temperature of the process caused a gradual raise in efficiency and at final temperature 70°C reached 49.0 mg·kg$^{-1}$ d.m. (62% of total value). The amount obtained at 70°C was more than 1.5 times higher than observed at 20°C. Moreover the changes observed at two last temperature steps were slightly higher than at lower temperatures.

The results of the EDTA extraction efficiency vs. temperature indicated general tendency of increasing of eluted

Table 2
Efficiency of the selected heavy metals extraction from sewage sludge with EDTA 0.10 M solution in dependency of the process temperature in mg·kg$^{-1}$ d.m. (dry matter) and percentage of the total metal content

| Metal | Zn | | Cu | | Ni | | Cd | | Pb | |
|---|---|---|---|---|---|---|---|---|---|---|
| Temperature, °C | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % |
| 20 | 663.7±3.8 | 31 | 3.5±0.3 | 2 | 10.4±0.3 | 26 | 2.5±0.1 | 40 | 29.8±0.3 | 38 |
| 30 | 684.3±3.5 | 32 | 8.0±0.4 | 4 | 10.6±0.2 | 27 | 3.3±0.1 | 52 | 31.7±0.3 | 40 |
| 40 | 754.5±4.1 | 36 | 13.0±0.3 | 7 | 11.6±0.5 | 29 | 3.6±0.3 | 57 | 35.4±0.1 | 45 |
| 50 | 890.3±3.9 | 42 | 17.1±0.3 | 9 | 12.5±0.3 | 32 | 3.9±0.1 | 62 | 37.5±0.2 | 47 |
| 60 | 1,138.9±3.8 | 54 | 20.5±0.5 | 10 | 13.3±0.4 | 34 | 4.3±0.1 | 68 | 43.0±0.3 | 54 |
| 70 | 1,614.4±4.3 | 76 | 21.1±0.6 | 11 | 14.3±0.4 | 38 | 5.0±0.3 | 79 | 49.0±0.3 | 62 |

metal amount with increasing in temperature of process. But the observed shapes of the dependencies were different. Most of the physical and chemical processes, and especially processes based on the solubility of substances, the rise in temperature causes the rise in the rate of process and in the consequence the higher value of efficiency. In the case of presented experiments, for zinc and cadmium, initial increase in temperature caused slight rise in efficiency. From the temperature of about 50°C, the efficiency increased much more intensively. Such behaviour indicates probably that energetic barrier of extraction was crossed and the rate of metal extraction had increased rapidly. For nickel and lead, the observed dependence was different. In the whole range of studied temperature, the mentioned dependency was almost linear, what could suggest that energetic barriers are lower than for zinc and cadmium and was crossed at temperature under studied range. Next for copper, initial increase in temperature caused intensive rise in amount of washed metal. After reaching a temperature of 50°C, the changes in observed efficiency of copper removal were more insignificant. Such behaviour and fact that copper was extracted by EDTA in the lowest ratio among analyzed metals suggests that energetic barriers for Cu elution were at higher temperature.

### *3.3. pH influence on efficiency of trace metals extraction with EDTA*

The changes of efficiency of heavy metals extraction from sewage sludge samples in dependency on the pH increase are presented in Figs. 6–10 for each metal separately and are summarized in Table 3.

Efficiency of zinc removal from municipal sewage sludge rises with decrease of pH of 0.1 M EDTA solution (Fig. 6 and Table 3). The highest amount of zinc (684.9 mg·kg$^{-1}$ d.m.) was extracted at pH equal to 3.0 and constituted 32% of total content of metal in the analyzed sludge sample. At pH 4.5, natural for aqueous solution of EDTA, the efficiency of Zn removal was very similar and reached 663.7 mg·kg$^{-1}$ d.m. – 31%. Content of washed zinc decreased slightly up to pH 7.0 (631.8 mg·kg$^{-1}$ d.m. – 30%). Further increasing the pH value decreased amount of removed Zn significantly and obtained 492.5 mg·kg$^{-1}$ d.m. (23% of total zinc content) at pH 12.0. The removal of zinc with acidified or alkalized water was much lower than 1% of total zinc amount and was stated as negligible in comparison with EDTA aqueous solution. It indicates that chemical forms of zinc in studied sewage sludge were unavailable for water extraction.

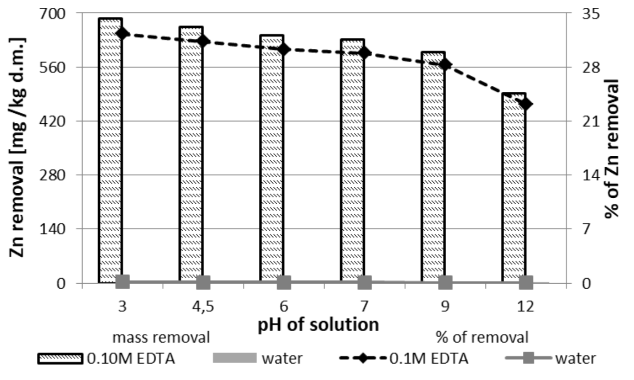


Fig. 6. Removal of zinc from municipal sewage sludge in dependency on pH.

The amount of copper extracted from 1 kg of sludge dry matter in dependency of extracting solution pH is shown in Fig. 7.

Extraction efficiency of copper from municipal sludge increased with decreasing pH. The best results were observed in the range 3.0–6.0. The maximal removal: 3.7 mg·kg$^{-1}$ d.m. was observed at pH 3.0 and minimal: 1.7 mg·kg$^{-1}$ d.m. at pH 12.0, what was 1.9% and 0.9% of the whole copper amount in studied sludge, respectively. The water influence on Cu

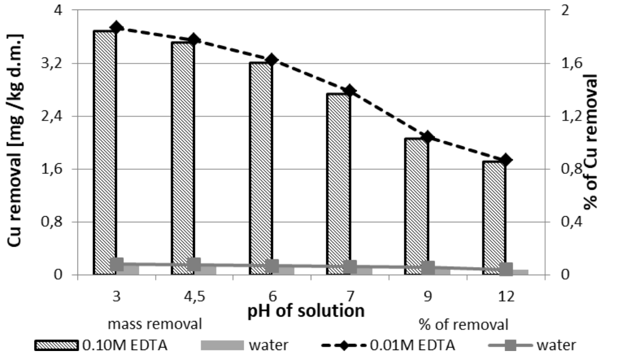


Fig. 7. Removal of copper from municipal sewage sludge in dependency on pH.

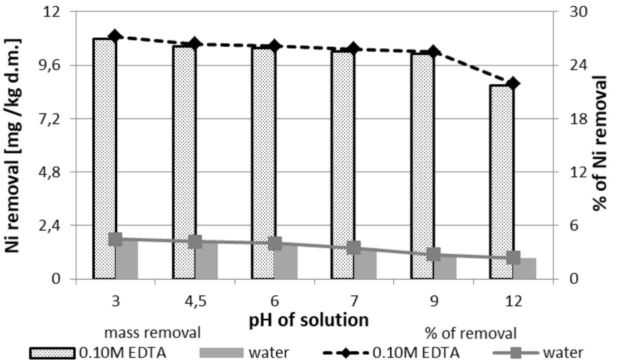


Fig. 8. Removal of nickel from municipal sewage sludge in dependency on pH.

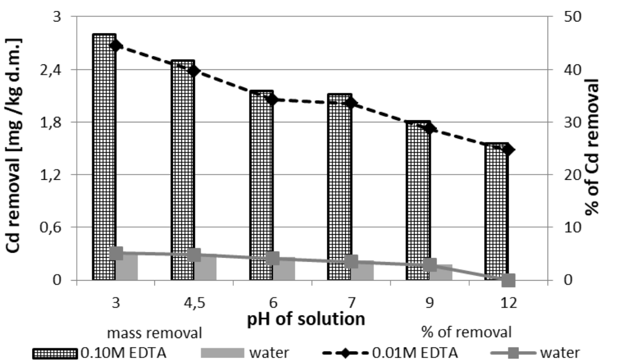


Fig. 9. Removal of cadmium from municipal sewage sludge in dependency on pH.

extraction, similarly for zinc extraction, was detected on the level lower than 0.1% of total copper content and was assumed as negligible at studied pH range.

Nickel extraction from municipal sewage sludge was pH affected similarly to previously described. The changes of nickel content eluted with EDTA in dependency on pH of the solution are presented in Fig. 8.

The highest value was obtained at pH 3.0 (10.8 mg·kg$^{-1}$ d.m.). It was approximately 27% of the total nickel amount in analyzed sample. The content extracted at pH 4.5, characteristic of EDTA 0.1 M solution, was approximately the same: 10.4 mg·kg$^{-1}$ d.m. (26% of total content). Next increasing in pH caused the decrease in the amount of washed metal up to pH 12.0, reaching 8.7 mg·kg$^{-1}$ d.m. (22% of total concentration). The extraction of nickel was not sensitive for pH influence and the optimal pH for nickel removal was in the range 3.0–9.0.

The behaviour of nickel for water extraction was different in comparison with zinc and copper. Water, adjusted to pH in the range 3 to 12, caused much significant removal of mentioned metal. The amount of washed nickel was 1.8 mg of Ni per 1 kg d.m. and 0.9 mg of Ni per 1 kg d.m. at pH 3.0 and 12.0, respectively. The content of water extracted nickel was on the level 2%–5%. That behaviour indicated that Ni could be mobilized much easier than Zn and Cu.

Fig. 9 illustrates the plot of cadmium removal vs. pH of extractant. The total content of cadmium in sewage sludge was the lowest and amounts of metal in the extracts were almost at the limit of detection, thus indicated values of content are approximate and probably burdened with the high uncertainty level. 0.1 M EDTA solution extracted 2.8 mg Cd·kg$^{-1}$ d.m. at pH 3.0. Efficiency of cadmium removal decreased slowly with pH rising and minimal value 1.6 mg Cd·kg$^{-1}$ d.m. was determined at pH 12.0. The mass percentage of removal was 44.5 and 25.5, respectively. For cadmium extraction significant efficiency of water was detected. Water extracted cadmium at a level of 3%–5%, showed the high mobility and availability of the mentioned metal.

The changes of lead content eluted from studied sewage sludge under different pH conditions are presented in Fig. 10. Similarly to other metals, the highest amount of extracted metal was observed at pH 3.0 and was equal to 30.9 mg·kg$^{-1}$ d.m. The value corresponded to 39% of total lead concentration in the analyzed municipal sewage sludge. The increase in pH caused insignificant decrease in removal efficiency up to pH 6. At pH 6 the amount of washed Pb was equal to 29.0 mg·kg$^{-1}$ d.m. (37% of total Pb content). A further rise in pH decreased the amount of extracted metal and at pH 12.0 reached 20% of total content – 15.8 mg·kg$^{-1}$ d.m. The efficiency of lead extraction at pH 12.0 was approximately a half of its initial value observed at pH 3.0.

The water efficiency in lead removal from municipal sludge was lower than 0.1% and was mentioned as practically negligible. That behaviour of lead suggested its strong affinity to sludge matter and immobilization of metal in organic matter, the chemical forms of lead were unavailable for water.

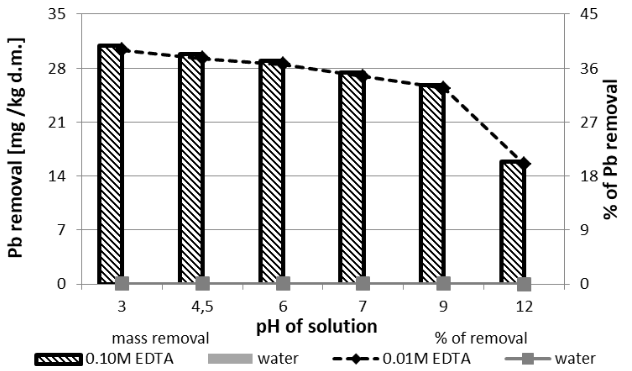


Fig. 10. Removal of lead from municipal sewage sludge in dependency on pH.

Table 3
Efficiency of the selected heavy metals extraction from sewage sludge with EDTA 0.10 M solution in dependency of extractant pH in mg·kg$^{-1}$ d.m. (dry matter) and percentage of the total metal content

| Metal | Zn | | Cu | | Ni | | Cd | | Pb | |
|---|---|---|---|---|---|---|---|---|---|---|
| pH | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % | mg·kg$^{-1}$ d.m. | % |
| 0.10 M EDTA | | | | | | | | | | |
| 3.0 | 684.9±4.1 | 32 | 3.7±0.2 | 2 | 10.8±0.4 | 27 | 2.8±0.1 | 44 | 30.9±0.6 | 39 |
| 4.5 | 663.7±3.8 | 31 | 3.5±0.3 | 2 | 10.4±0.3 | 26 | 2.5±0.1 | 40 | 29.8±0.3 | 38 |
| 6.0 | 642.5±3.3 | 30 | 3.2±0.2 | 2 | 10.4±0.4 | 26 | 2.2±0.3 | 34 | 29.0±0.4 | 37 |
| 7.0 | 631.8±3.6 | 30 | 2.7±0.2 | 1 | 10.2±0.1 | 26 | 2.1±0.1 | 34 | 27.4±0.3 | 35 |
| 9.0 | 599.3±4.0 | 28 | 2.0±0.3 | 1 | 10.1±0.3 | 25 | 1.8±0.2 | 29 | 25.8±0.3 | 33 |
| 12.0 | 492.5±3.3 | 23 | 1.7±0.1 | 1 | 8.7±0.2 | 22 | 1.6±0.2 | 25 | 15.8±0.1 | 20 |
| Water | | | | | | | | | | |
| 3.0 | 2.8±0.1 | 0.13 | 0.16±0.05 | 0.08 | 1.8±0.2 | 4.5 | 0.32±0.03 | 5.1 | 0.03±0.006 | 0.03 |
| 4.5 | 2.0±0.1 | 0.09 | 0.15±0.06 | 0.07 | 1.7±0.1 | 4.2 | 0.30±0.03 | 4.8 | 0.03±0.000 | 0.03 |
| 6.0 | 1.9±0.3 | 0.09 | 0.13±0.06 | 0.07 | 1.6±0.1 | 4.0 | 0.26±0.1 | 4.1 | 0.03±0.005 | 0.03 |
| 7.0 | 1.4±0.1 | 0.07 | 0.12±0.06 | 0.06 | 1.4±0.1 | 3.5 | 0.22±0.03 | 3.5 | 0.01±0.001 | 0.02 |
| 9.0 | 0.5±0.2 | 0.02 | 0.11±0.04 | 0.06 | 1.1±0.2 | 2.7 | 0.18±0.03 | 2.9 | 0.01±0.004 | 0.02 |
| 12.0 | 0.3±0.2 | 0.01 | 0.08±0.04 | 0.04 | 0.9±0.1 | 2.4 | 0.00±0.00 | 0.0 | 0.00±0.000 | 0.00 |

The direction of efficiency changes with the rising of pH value was in agreement with previously reported tendency [11,27,28]. For Cu, Cd and lead at pH 12, the detected efficiency of metal EDTA extraction reached about a half of its initial value observed at pH 3.0. Zn and Ni were not such sensitive for increase in pH, but their efficiencies decreased significantly. Further increase of pH value would cause next fall in amount of washed metals. Increase of pH decreases mobility of heavy metals [29]. At higher pH values studied trace metals have a tendency to form hydroxides and oxohydroxides insoluble in water. The optimal pH range for elution of the analyzed group of metals was 3.0–6.0, but the changes of efficiency of the extraction process in the mentioned above range were insignificant, therefore it is justified the driving an extraction process at pH without the correction, it means at pH equal to 4.5.

High efficiency of heavy metals removal with EDTA solution was in agreement with data from another group of scientists obtained for extraction of trace metals with chelating agents [15–19].

## 4. Conclusions

Chelators, among them EDTA, are known as effective washing compounds for trace metals in solid samples of soil, bottom sediments and sewage sludge. Presented results confirmed the usefulness of 0.1 M EDTA aqueous solution for extraction of Zn, Cu, Ni, Cd and Pb from municipal sewage sludge samples. The 0.1 M EDTA solution extracted metals from analyzed sewage sludge with different efficiency. The amount of eluted metal decreased with increase of pH. For pH in the range from 3.0 to 6.0, the fall in the amount of each extracted metal was insignificant, for a further pH increase much indicative, so pH range from 3.0 to 6.0 for heavy metals removal was detected as optimal. pH of aqueous 0.1 M EDTA solution equals to 4.5 and is included in the optimal range. Therefore in practice in technical scale heavy metals extraction process could be driven without any pH adjustment.

Analysis of extraction process efficiency vs. temperature demonstrated that temperature rise caused the increase in amount of eluted metal for all studied trace metals. Further increase in temperature over 70°C should improve efficiency of the analyzed EDTA extraction process, but in the studied range of temperature, the highest observed efficiency was at 70°C. Deeper analysis indicated that the optimal temperature for trace metals extraction from studied sewage sludge was in the range 40°C–50°C. Increase in temperature over mentioned range causes very insignificant improvement in process efficiency usually. Consider the properties of municipal sewage sludge and slight rise in effect of extraction process, it could be stated that driving the process in the higher temperatures, especially in practice in technical scale, seems to be burdensome and consuming additional costs of energy used for heating of extraction system. Usually heavy metals extraction process occurring even under ambient conditions, yields to removal efficiency on the level, what secures forming the new material, with the heavy metals content under limitations given in proper legislative acts [24,25].

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