Investigation of Lead and Iron Removal from Groundwater Using Sapropel and Quartz Sand

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Abstract. In Prey Nop district of Cambodia, a district in coastal area, it was observed that concentration of iron and lead in groundwater was higher than permissible values. Lead is a highly toxic heavy metal, while iron is an element causing several problems related to the deterioration of taste and aesthetic appearance of water and the capacity reduction of water supply pipelines. Therefore, this article investigates the effectiveness of the technology for removing lead and iron from groundwater using the cheapest materials like sapropel and sand. In this study, different doses of sapropel (0.1, 0.2, 0.4, 0.5, 0.6, 0.8, 1, 2, 3, 4, 5 and 6 g/L), different durations of sorption processes (30, 60, 90, 120 and 150 min) and a laboratory bench for iron filtration filled with quartz sand were used for lead and iron ions removal. Results from the bench tests showed that both iron and lead were removed at efficiencies of 70 and 97%, and their concentrations did not exceed the permissible levels by using the lowest dose of 0.1 g/L of sapropel for sorption of lead and filtration through quartz sand filters for iron removal.

Keywords: groundwater, lead, iron, sapropel, sand.

Introduction

The Kingdom of Cambodia is located in the Southeast Asia region. Two types of water are available in Cambodia, in which surface water stands for the abundant water resources. Besides, groundwater in Cambodia is of good quality, but in some areas it contains higher level of lead, arsenic, manganese, fluoride, iron, affecting public health of people in those areas (Peng & Pin, 2002). In Prey Nop district, a district in coastal area, it was observed that concentration of iron and lead in groundwater was higher than permissible values.

Lead is one of the most dangerous heavy metal for ecosystems since it possesses strong toxic properties. Lead is a highly poisonous metal having effect on almost every organ in the body. The nervous system is the mainly affected target in lead toxicity (Akers et al., 2015; Wani et al., 2015). Lead tends to accumulate in the body and may cause adverse effects: central nervous system damage; gastrointestinal lead is absorbed by 5–10% in adults and 50% in children; 80–100% of inhaled lead enters the bloodstream; it accumulates in hair, nails and bones (Schamphelaere et al., 2014; Liu et al., 2014).

Lead is one of the most dangerous components of wastewater. Natural lead pollution occurrences from volcanic explosions and forest fire. Non-natural sources are human, from the industry and transportation (Zhang et al., 2015). Road transport, together with the exhaust, emits up to 75% of the lead contained in petrol. Renovation of old steel structures may result in the formation of large quantities of lead dust. The metallurgical industry, lead processing and fuel plants also contribute to environmental pollution by lead compounds. Other sources are from manufacturing batteries, coal burning, type casting, and older houses and buildings (Woolf et al., 2007; Zhang et al., 2009). When burning lead-containing waste – capsules, tubes, synthetic materials, anti-corrosion paint, the pollutants are released into the air and then deposit on the soil and get into water bodies. Lead contamination of groundwater could be associated to the seepage of irrigation water, corrosion of buried metallic structure (largely of iron), and leachate from the industrial dust pile (Sadiq & Alam, 1997). Sanitary landfills remain the most cost effective option for disposal of solid waste (Islam & Singhal, 2002) especially for many municipalities in developing countries. However, groundwater pollution from the leachate generated within the landfill and migrating through the liner material into the underlying aquifers is big problem (Fatta et al., 1999). Major exposure pathways of pollution also include intake of lead by consuming lead-

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contaminated water and food. Food can become contaminated with lead from the soil and water in the growing environment, or from the materials used in processing or cooking, or the containers used in storage. Lead concentrations in drinking water in China, Poland, Japan, Singapore, India, USA and Cambodia have been found to exceed the permitted level (in the EU defined as 10 µg/L by Directive 98/83/EC (The Council of the European Union, 1998)) (Chen et al., 2015; Li et al., 2015; Lu et al., 2015; Jurgens et al., 2019).

Lead concentration in water can be reduced by nanofiltration, membrane filtration (Gholami et al., 2014). Adsorption is often used for lead removal (Bhatia et al., 2017). Adsorption using nanosorbents has a higher efficiency in removing chemical elements (Ngulube et al., 2017). However, majority of these techniques cause several undesirable effects namely chemical requirements, complexity, low efficiency, and usually large amounts of sludge produced causing other environmental problems. Compared with other treatment methods, adsorption is generally the most effective method due to its merits of efficiency, free-sludge (Gupta et al., 2011). For this reason, numerous studies have been investigating the adsorption by using low-cost sorbents. Various sorbents are used to remove heavy metals: biomass, activated carbon, red clay, zeolites, magnetite, aluminium, carbon nanotubes, chitosan, copper, iron, manganese and zinc oxides, aluminium sorbents, bentonite (Bhatia et al., 2017; Badawi et al., 2017; Golbad et al., 2017; Kocaoba & Arisoy, 2018; Zou et al., 2019). Oak bark, mahogany bark, lignin, rice shells, clay, modified peat, bamboo pulp, sawdust, active sewage sludge, modified wool, palm tree waste fibres, saffron flower waste etc. can be also used as sorbents (Mishra & Patel, 2009; Alhogbi et al., 2019; Khohsang & Ghaffarinejad, 2018). It has also been found that lead is effectively removed by sapropel. Sapropel is a silty sediment or sediment formed on the bottom of fresh, slack water bodies. The organic material consists of lignin-humus mix, carbohydrates and bitumens (Stankevica et al., 2014). Depending on sapropel composition, it is categorized into organic (50–90% of organic matter by dry weight), calcareous (30–60% of calcium carbonate), siliceous (25–45% of silicon dioxide) and mixed varieties (Bakšienė, 2006). Sapropel has the property of sorbing heavy metals as much as up to 97.4% from solution, depending on contact time and concentration of heavy metals in the solution (Birgėlaitė et al., 2016). An enhancement of efficiency in removing lead from aqueous solution or groundwater is a crucial point that most of the scientific researchers have been developing. Keeping this in mind, environmental and physicochemical conditions for conducting an experiment have been carefully considered in order to reach an optimal efficiency. Maximum removal capacity of Pb ions was affected by its initial concentration, adsorbent dose, contact time, pH, metal speciation, temperature, and presence of NO_3^- and Cl^- (Mouflih et al., 2006; Liu & Zhang, 2009; Mishra & Patel, 2009; Fan et al., 2017).

Investigations of health professionals have shown that higher levels of iron in water do not adversely affect human health (Lieu et al., 2001), but cause a number of other problems in the water supply for consumers: the interaction of soluble divalent iron compounds with oxygen in the air produces low-soluble brownish trivalent iron hydroxides, and the water becomes cloudy, its taste and aesthetic appearance deteriorate. These clouds settle on various plumbing fixtures and need to be removed periodically. Another problem is that aerated iron water flowing through the pipelines causes the accumulation of iron bacteria, that immobilize trivalent iron hydroxide, on these pipelines. As these substances accumulate, the diameter of the pipelines and their throughput are reduced. Bacterial activity products can give water an unpleasant odor. As the water pressure fluctuates in the system, more iron sediments appear from the tap (Vreeburg & Boxall, 2007; Marjani et al., 2009). For these reasons, the World Health Organization (WHO) recommends that the concentration of iron in drinking water supplied to consumers should not exceed 300 µg/L. Under Council Directive 98/83/EC, the concentration of total iron in drinking water must not exceed 200 µg/L. Iron is removed from groundwater by the conversion (oxidation) of soluble divalent iron into insoluble trivalent iron compounds which are then filtered. The rate of iron oxidation depends on water temperature, pH, alkalinity, hydrogen sulfide content and other parameters. The higher the pH value of the water, the faster the divalent iron is oxidized to the trivalent. It is also necessary to consider the grain size of the filters, their composition, uniformity, porosity, chemical and mechanical resistance. Various filters are used for iron removal. Catalytic filters such as Nevtraco natural dolomite filler are used as well as granular filtration fillers based on manganese dioxide based Pyrolox, Greensand, Birm. The simplest, cheapest and most widely used is quartz sand (Veressinina et al., 2000; Lee et al., 2009).

The purpose of this article is to investigate the effectiveness of the technology for simultaneously removing lead and iron from groundwater using the cheapest materials such as sapropel and sand.

1. Materials and methods

According to one investigation of the groundwater quality in Prey Nop district of Cambodia, a district in coastal area, it was observed that concentration of iron and lead in groundwater was higher than permissible values (Table 1).

The first type of test water was deionized water contaminated with lead(II) ions with the concentration of lead(II) ions of 0.07 mg/L. This concentration of lead in water has been chosen by taking into account the chemical composition of the water taken from the Prey Nop area of Cambodia.

Stock solutions of lead(II) (1000±2 ppm, Buck Scientific, USA) were used for GFAAS calibration. Standard solutions with the required lead(II) ion concentrations were prepared by appropriate dilution with deionized water. The pH of every solution was maintained at a desired value by adding 0.01 M NaOH or 0.01 M HNO_3.

Test water was prepared from dilution of stock solution of Pb (II) in one-litre measuring flasks with deionised water. 6 flasks of 1 L with 0.07 mg/L Pb (II) of test water were used for the tests.

The temperature has the impact on the adsorption process. As the temperature increases, the adsorption process can be activated due to the increase of surface area of the sorbent. The optimum temperature for the adsorption process is between 30 °C and 70 °C (Badawi et al., 2017). Experimental adsorption tests were performed at room temperature at 20±2 °C.

Table 1. Quality data of groundwater in Prey Nop district, Cambodia

Parameters	Amount
pH	5.25
Electrical conductivity, μS/cm	32.2
Turbidity, NTU	6.95
Dissolved oxygen, mg/L	2.52
Total dissolved solid, mg/L	1660
Ammonia, mg/L	0.05
Nitrite, mg/L	0.001
Iron, mg/L	0.958
Lead, mg/L	0.0702

The raw sapropel was extracted from 2–3 m depth of the Apslavas lake located in Stabulankliai village, Leliūnai local municipality, Utena district, Lithuania. The sapropel sample was taken to analyse its composition and to conduct the further experiments. The chemical (elemental) composition of sapropel was investigated by X-ray fluorescence spectrometer with wave variance Axios MAx (manufacturer PANalytical, Netherlands). An X-ray source of 4 kW power with Rh (rhodium) anode. The chemical composition was calculated using the betalon methodology and Omnian software (PANalytical, Netherlands). The mineral composition was investigated by powder X-ray diffraction (Powder X-ray diffraction). Used X-ray diffractometer SmartLab (manufacturer Rigaku, Japan). Analysis was made using the EVA software (Bruker AXS) and X-ray database PDF-2. The carbon content was determined by the carbon and sulphur analyser CS-2000 (Manufacturer ELTRA, Germany). The composition of sapropel is shown in Table 2.

Table 2. Chemical composition data of the sapropel

Element	[%] w/w	Compound	[%] w/w
C	6.616	CO_2	24.240
O	56.325	O	0.105
Na	0.363	Na_2O	0.489
Mg	0.279	MgO	0.462
Al	2.602	Al_2O_3	4.916
Si	30.383	SiO_2	65.000
P	0.040	P_2O_5	0.091
S	0.165	SO_3	0.413
Cl	0.011	Cl	0.011
K	1.422	K_2O	1.712
Ca	0.890	CaO	1.246
Ti	0.147	TiO_2	0.245
Cr	0.027	Cr_2O_3	0.040
Mn	0.010	MnO	0.013
Fe	0.657	Fe_2O_3	0.940
Ni	0.003	NiO	0.003
Cu	0.001	CuO	0.001
Zn	0.003	ZnO	0.003

End of Table 2

Element	[%] w/w	Compound	[%] w/w
Rb	0.005	Rb_2O	0.006
Sr	0.005	SrO	0.006
Y	0.002	Y_2O_3	0.002
Zr	0.024	ZrO_2	0.032
Ba	0.013	BaO	0.015
Pb	0.007	PbO	0.008

The sorbent for the adsorption studies were washed twice with deionized water, dried for 3 hours in a drying oven at 110±2 °C. The dried substance was crushed in a porcelain mortar to a homogeneous mass and spread through a 0.2 mm sieve.

Dry sapropel in the doses of 1, 2, 3, 4, 5 and 6 g was weighed with analytical scales. The prepared sorbent was added to the prepared 1 L six containers with test water contaminated with lead(II) ions. The test water was mixed with sorbents in a magnetic stirrer at 200 rpm.

5 water samples were taken in the quantities of 25 ml after different contact times: 30, 60, 90, 120 and 150 min (Ö. Gerçel & H. Gerçel, 2007). After adsorption, the solution was filtered through a membrane filter (47 mm diameter, pore size 0.45 μm). After filtration the concentrations of lead(II) ions were determined by using atomic absorption spectrometry (AAS; model Buck Scientific 210 VGP, USA) operated with graphite furnace, a slit of 0.7 mm at 283.2 nm wavelength. A chemical modifier ($NH_4H_2PO_4$) was used for possible alleviation of matrix interferences, which are normally severe during graphite furnace analysis for lead.

Due to the comparatively low concentration of lead after removal with high doses of sapropel – a more detailed study with lower doses of sapropel is required. The second test was designed to determine the conditions under which sapropel adsorbs lead(II) ions best at low doses of sorbent – 0.1, 0.2, 0.4, 0.5, 0.6 and 0.8 g/L, and the concentration of lead (II) ions in water was measured after 30, 60, 90, 120 and 150 minutes (Ö. Gerçel & H. Gerçel, 2007).

A laboratory batch experimental setup of iron removal, as shown in Figure 1, was developed to measure the filtration of iron with filter media. To construct filter column, 1.20 m in length and diameter of 0.055 m of plastic column was prepared and connected with faucet to regulate and stabilize the flow rate of filtration. Inside of the column, gravel ranging from 2–2.5 mm in size was put into the column at 10.5 cm from bottom of column as supporting layer and the sand was then gradually added into to the column to achieve a 80 cm filter depth from upper gravel layer.
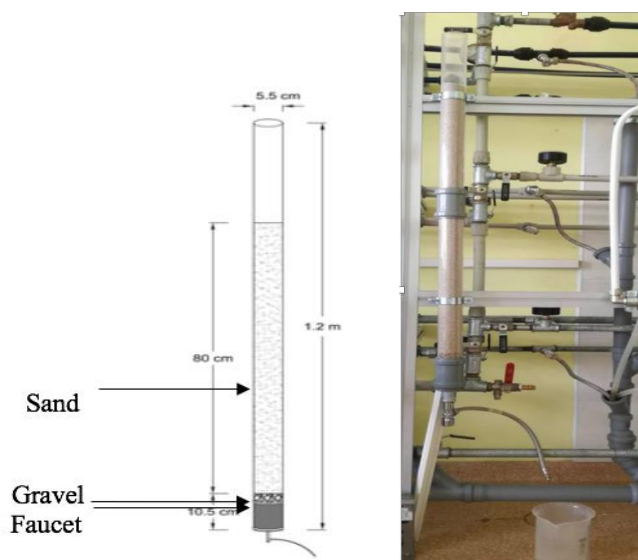

Figure 1. Experimental setup of iron removal

To establish the efficiency of combined steps including adsorption with sapropel, aeration during stirring and sand filtration in removing both lead and iron ions from water the second type of synthetic water was used in the experiments (solution of lead(II) and iron(II) with concentrations of 0.07 mg/L and 0.9 mg/L, respectively). The 1 L of synthetic water was mixed with the lowest dose of sapropel, 0.1 mg/L. The solutions were then stirred using magnetic drives connected with straight blade impeller at 200 rpm under room temperature for 30, 60 and 90 min. Filtration

column filtrated 1 L of stirred solutions with flow velocity of 8.5 $m^3/m^2/h$. Lead and iron concentrations in filtrate were measured using Atomic Absorption Spectrophotometer and photometer, respectively.

Water quality assessment tests after mixing were performed based on the following standards:

Determination of lead concentration in water was carried out according to ISO 15586 Water quality – Determination of trace elements using atomic absorption spectrometry with graphite furnace (International Organization for Standardization [ISO], 2003).

Determination of total iron concentration in water was carried out according to ISO 6332 Water quality – Determination of iron – Spectrometric method using 1,10-phenanthroline (ISO, 1988).

According to Council directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption the result is acceptable, when Trueness and Precision of method is less than 10%.

Trueness is a measure of systematic error, i.e. the difference between the mean value of the large number (10 measurements) of repeated measurements and the true value. Stock solutions of lead(II) is 0.01 mg/L and iron 200 μg/L according to Council directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption parametric value of lead is 10 μg/L and iron 200 μg/L. Precision is a measure of random error and is usually expressed as the standard deviation (within and between batches) of the spread of results from the mean. Acceptable precision is twice the relative standard deviation (Table 3).

Table 3. Trueness and precision of method

	Trueness of lead	Precision of lead	Trueness of iron	Precision of iron
Average value, μg/L	9.7	10.12	196.5	202.2
Assigned value, μg/L	10.0	10.0	200	200
Standard deviation, μg/L	0.24	0.39	3.16	3.03
Relative standard deviation, %	2.4	3.9	1.61	1.82
Acceptable trueness, %	4.8	7.8	3.22	3.64

The obtained result of the analysis is the arithmetical average of concentrations of three samples, when distribution does not exceed 5%, if higher tested were repeated. 95% confidence interval was calculated.

2. Results and discussion

In the first stage of the study, the task was to determine how the presence of sapropel in aqueous solution affected the quantity of lead. The sapropel dose is a main variable for the quantitative removal of lead ions from model groundwater. Optimization of the dose of the sapropel used was performed with a model lead solution of concentration about 0.07 mg/L which is similar to those in a natural groundwater in Cambodia. The process was conducted for various contact times in the range of 30, 60, 90, 120 and 150 min at ambient temperature 20±2 °C using various quantities of sapropel, 1, 2, 3, 4, 5 and 6 g/L. The experimental data obtained are shown in Figure 2.

Having completed the analysis of the lead ion sorption using different doses of sapropel and different sorption times, Figure 2 shows that using all doses of sapropel from 1 to 6 g/L and the sorption process times from the shortest one of 30 min to the longest time of 150 min, the lead concentration is effectively reduced to 100% and no longer exceeds the allowable limits. The most efficient removal of lead ions by sorption at different times occurs at a sapropel dose of 6 g/L. At different doses of sapropel, lead ion concentration is most effectively reduced by the longest sorption process of 150 min. This study suggests that sapropel doses of less than 1 g/L can be used to remove lead ions.

Figure 3 shows the removal of lead ions using different sapropel doses (0.1, 0.2, 0.4, 0.5, 0.6 and 0.8 g/L) and at different sorption times (30, 60, 90, 120 and 150 min).

Having completed the analysis of the lead ion sorption using different doses of sapropel and different sorption times, Figure 3 shows that using all doses of sapropel from 0.1 to 0.8 g/L and the sorption process times from the shortest one of 30 min to the longest time of 150 min, the lead concentration is effectively reduced and no longer exceeds the allowable limits. Lead removal efficiency ranges from 91% to 99%. Lead ion sorption of 30 min shows the tendency that with the increase of the sorbent dose the efficiency of lead ion removal decreases. At longer sorption times, this tendency disappears. For shorter sorption times (30 and 60 min), the most effective dose of sapropel is 0.1 g/L. With increased sorption time (90, 120 and 150 min), the most effective dose of sapropel is 0.8 g/L. Lead ions are removed very efficiently (91 to 99%) at all sorption times. Since all doses of sapropel have very high lead removal efficiency and lead is removed to the allowable limits, the smallest sapropel dose of 0.1 g/L will be used for further economic calculations.
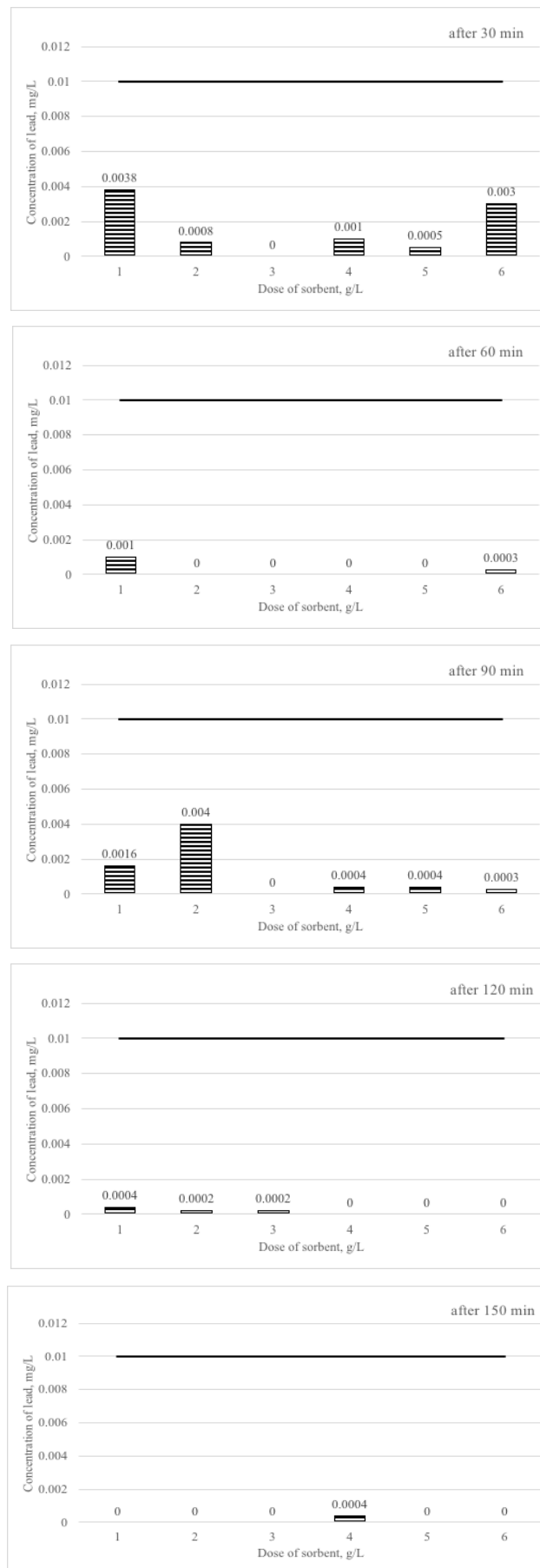
Figure 2. Effect of adsorbent dosage and contact time on adsorption of Pb ions. Initial lead concentration 0.07 mg/L
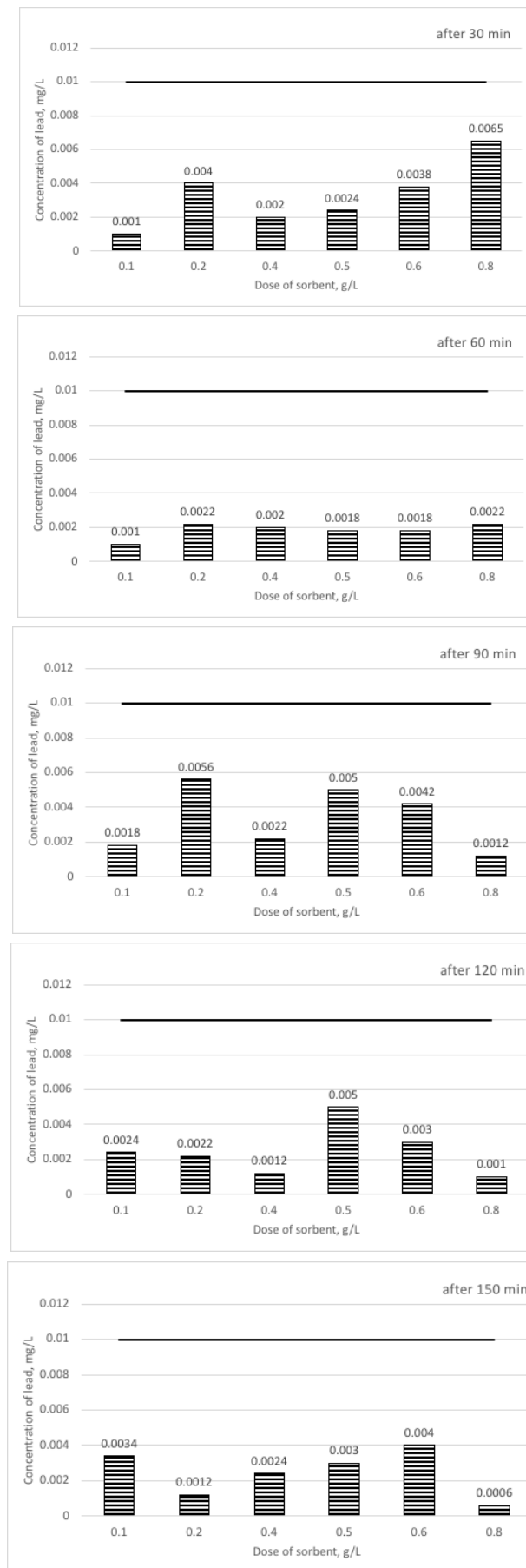
Figure 3. Effect of adsorbent dosage and contact time on adsorption of Pb ions. Initial lead concentration 0.07 mg/L

Figure 4 shows the simultaneous removal of iron and lead ions in a laboratory bench using three different times of sorption process (30, 60 and 90 min) at a sapropel dose of 0.1 g/L and filtration of water through a sand filter at 8.5 m^3/m^2/h speed.
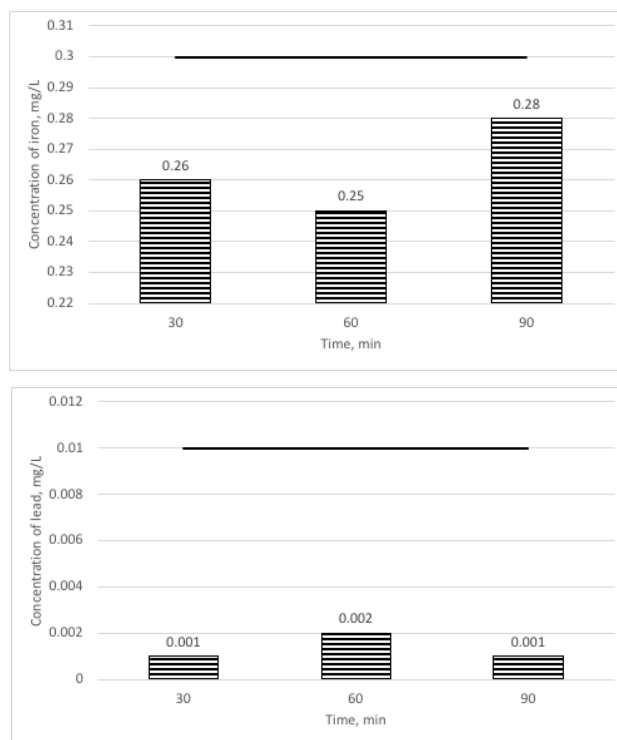

Figure 4. Iron and lead removal efficiency in experimental setup at different lead sorption times. Initial iron and lead concentrations are 0.9 and 0.07 mg/L

Iron and lead ion removal bench tests, using different sorption times, showed that by using a minimum sapropel dose of 0.1 g/L for lead sorption and filtration through quartz sand filters for iron removal, both iron and lead were removed efficiently (70 and 97%) and their concentrations did not exceed the permitted limits. At different sorption times, the removal efficiency of iron and lead ions is practically unchanged, so we can use the shortest sorption time of 30 minutes in this technology.

Conclusions

1. The lead ion sorption study using different doses of sapropel and different sorption times showed that using all sapropel doses from 1 to 6 g/L and the sorption process from the shortest time 30 min to the longest time 150 min, the concentration of lead ions is very effectively reduced to 100% and no longer exceeds the allowable limits.
2. The lead ion sorption study using different doses of sapropel and different sorption times showed that using all sapropel doses from 0.1 to 0.8 g/L and the sorption process from the shortest time 30 min to the longest time 150 min, the concentration of lead ions is very effectively reduced and no longer exceeds the allowable limits. Lead removal efficiency ranges from 91% to 99%.
3. For shorter sorption times (30 and 60 min), the most effective dose of sapropel is 0.1 g/L. With increasing sorption time (90, 120 and 150 min), the most effective dose of sapropel is 0.8 g/L.
4. Iron and lead ion removal bench tests, using different sorption times, showed that by using a minimum sapropel dose of 0.1 g/L for lead sorption and filtration through quartz sand filters to remove iron, both iron and lead were removed efficiently (70 and 97%) and their concentrations did not exceed the permitted limits.

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