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Experimental research on removal of iron and ammonium ions from groundwater by natural silica sand

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An experimental study of the removal of iron and ammonium ions from groundwater by natural silica sand was carried out. Coarse sand particles were used (0.7–2.0 mm): the silica sand filter medium being 750 mm heigh. Silica sand material was washed and dried at 105 °C in an oven before using it in the filter bed; 500 l of groundwater artificially polluted with iron sulphate and ammonium chloride solutions passed through the filter charged with silica sand. The best removal of iron compounds from the water solution was obtained by using a filter medium 1 m high with the finest silica sand. Iron removal efficiency was 95% using coarse silica sand. The highest ammonium removal efficiency (94%) was achieved using 0.7–2.0 mm silica sand.

Key words: groundwater, water treatment, filtration, silica sand, fine particle size, iron and ammonium ions, removal

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INTRODUCTION

All filter media materials used for cleaning wastewater, water or air have to fit environmental protection requirements, i. e. to be easily used, harmless to the environment, biodegradable and easily utilized.

Though a high efficiency of water cleaning is reached by using expensive and high sorption capacity material, it is purposeful to focus on searching for natural and cheep, filter materials even of a lower removal capacity. Recently, in water treatment for human consumption, simple technologies based on filtration through silica sand have been elaborated.

The intensive removal of iron and ammonium ions as the main undesirable compounds of groundwater sources in Lithuania (Diliūnas, Jurevičius, 1998; Diliūnas et al., 2006; Diliūnas, Sakalauskas, 1996; Klimas, 2006) allows a potential search of filter materials and groundwater treatment possibilities. Such problems arose and began to be solved about 10 years ago in Lithuania (Sakalauskas, Šulga, 1998; Sakalauskas, 1999; Valentukeviciene, Rimeika, 2004). In 1998 followed Lithuanian Republic Higien Norm (Lietuvos…, 2003) for drinking water, and provisions of Water Law for Central and East Europe of EC directives in Lithuania were assigned as Council Directive 98/83/EC of 3 November 1998 (European Commission Council, 1998) on the quality of water intended for human consumption (OJ L 330, 5.12.1998, p. 32) corrected by C1 Corrigendum, OJ L 111, 20.4.2001, p. 31 (98/83/EC).

It is stressed in the requirements that the main problem and task for today is to reach and minimize the level of 0.2 mg/l for iron and 0.5 mg/l for ammonium ions in the water treatment equipment (World…, 2004). Groundwater contamination by iron and ammonium ions includes different levels in groundwater exceeding the requirements of the Hygiene norm. To find an optimum removal of iron and ammonium ions by silica sand media and to clean groundwater, a detailed study of iron and ammonium removal dynamics is needed.

In recent years, investigation of the usage of natural silica sand for removal of iron and manganese ions from groundwater has been started in Lithuania (Sakalauskas, Šulga, 1998). The possibility of natural silica sand to remove iron and ammonium from drinking water has been investigated comparatively recently (Valentukeviciene, Rimeika, 2004). Sand is a naturally occurring granular material composed of finely divided rock and mineral particles (Крайнов, Рыженко, Швец, 2004). As the term is used by geologists, sand particles range in diameter from 0.0625 (or $^1/_{16}$ mm) to 2 mm. An individual particle in this range size is termed a sand grain. ISO 14688 grades sands as fine, medium and coarse with the ranges from 0.063 mm to 0.2 mm, 0.63 mm and 2.0 mm. The most common constituent of sand, in inland continental settings and non-tropical coastal settings, is silica (silicon dioxide, or SiO_2), usually in the form of quartz, which, because of its chemical inertness and considerable hardness, is resistant to weathering. The chemical compound silicon dioxide, also known as silica or silox (from the Latin "silex"), is an oxide of silicon, chemical formula SiO_2, and has been known for its hardness since the 9th century. Silica is most commonly found in nature as sand or quartz witch the density 2.2 g/cm³.

For the reasons mentioned above, it is purposeful to find out how much iron and ammonium ions can be removed from groundwater using natural filter media materials (Mackie, Zielina, Dabrowski, 2003). The purpose of the experiments presented in this paper is to find out the efficiency of natural silica sand removal of iron and ammonium ions from solutions and to compare their interference dynamic (Mackie, Bai, 1993). The results of this study will be used for estimating the filter medium height needed to remove a certain concentration of iron and ammonium ions from groundwater.

It is known that the removal capacity is increased by crushing a filter medium because the surface area increases. Therefore, the smallest naturally obtained fraction (imported from Poland) of natural silica sand was used in this study.

MATERIALS AND METHODS

The filter media investigated in this study was silica sand naturally obtained from a Polish sand excavation quarry. Different researchers studied the removal of iron and ammonium from solutions by filtration through silica sand filter media (Diliūnas, Sakalauskas 1996; Sakalauskas, Šulga, 1998). Silica sand can be regarded as a microorganisms' growing surface for biological filtration if we consider the biological removal of iron and ammonium ions as dominant, or / and silica sand coated with iron oxides can be the catalyst for chemical removal processes.

A pilot test bench was constructed for the study (Fig. 1). 500 L of groundwater (5) artificially polluted with iron sulphate and ammonium chloride solutions was poured into a metallic container (4). The most frequent range of ammonium ions (0.3–2.0 mg/l) at Lithuanian water treatment plants (Klimas, 2006) was selected. This mixture was stirred hydraulically and pumped (6, 7) into the filtration column (∅ 100 mm) in which iron and ammonium ions were removed (2). Samples were taken from the filtration column through a sample tap (3) at different heights of the filter medium to define the iron and ammonium concentration in the different points. The flexible hose connected the filtration column to different pipelines (10, 14, 15, 16, and 20). On the top of the filtration column, water was aerated and then filtered through a silica sand layer 1.00 m high. The filtrate samples were taken from the bottom of the filtration column to determine iron and ammonium concentrations.

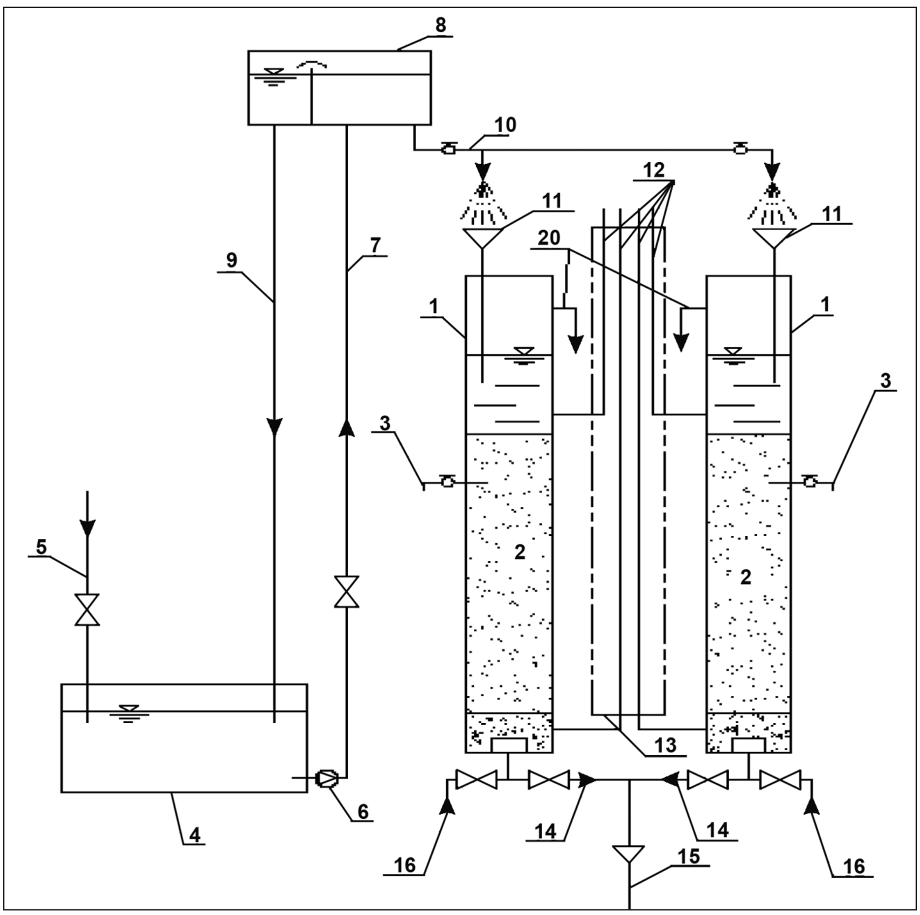

Fig. 1. Scheme of the experimental model:
1 – filter column, *2* – silica sand filter medium, *3* – sampling tap, *4* – groundwater reservoir, *5* – iron sulphate and ammonium chloride solutions, *6* – water pump, *7* – water supply pipeline, *8* – equalisation reservoir, *9* – water supply pipeline, *10* – water supply for aeration stage, *11* – water aeration, *12* – piezometers, *13* – piezometers scale, *14* – filtered water pipelines, *15* – filtered water outlet, *16* – backwash water supply

1 pav. Eksperimentinio stendo schema:
1 – filtro kolona, *2* – filtro užpildas iš kvarcinio smėlio, *3* – ėminio ėmimo čiaupas, *4* – požeminio vandens rezervuaras, *5* – geležies sulfato bei amonio chlorido tirpalai, *6* – vandens siurblys, *7* – vandentiekio vamzdynas, *8* – išlyginimo talpa, *9* – vandentiekio vamzdynas, *10* – vandens tiekimas aeravimui, *11* – vandens aeravimas, *12* – pjezometrai, *13* – pjezometro skalė, *14* – filtruoto vandens vamzdynai, *15* – filtruoto vandens ištekėjimas, *16* – vandens tiekimas plovimui

Coarse size (following ISO 14688) grades of sand particle ranges were used in this study. The test material was washed and dried at 105 °C in an oven before using it in the filter bed.

Typical water quality parameters for the source water were as follows: for WTP Antaviliai: pH 7.33–7.52; conductivity ~514 μS/cm; Fe_{total} 0.3–2.86 mg/l; NH_4^+ 0.19–0.59 mg/l; Mn 0.13–0.19 mg/l; Na ~ 13 mg/l; Ca 57.8–74.5 mg/l; and dry residual 306 mg/l. The effectiveness was evaluated by residual concentrations of iron and ammonium ions removed by filtration through quartz sand filter media. The filtration process was started at a filtration rate of 5 m/h, and experiments were stopped when marginal pressure losses (measured with piezometers, 20) were reached and the quality of filtrate was not acceptable. Three replicated experiments were performed for each operational scenario. Iron and ammonium ion concentrations in the experimental water before filter and filtrate samples were measured continuously, five times each. Merck system Aquaquant tests were used for a fast determination of iron, manganese and ammonia concentrations. A water quality analysis was made, and certain technological parameters were determined for the control and evaluation of technological processes using international standard methods: total iron concentration mg/l, ammonium ion concentration mg/l, filtration rate m/h, backwash time min. Taps for water sampling were equipped on the pilot size filter column at different heights. The filter medium was backwashed with treated water, and special taps regulated the experimental pilot size plant. Equipment in the untreated water supply pipelines and the filtrated water outlet pipeline, and the water flow was measured by the volume method.

RESULTS AND DISCUSSION

The results of the experimental research using coarse natural silica sand for iron and ammonium removal from water are shown in Table and Fig. 2–4.

The different periods of mixing and filtration were varied by using different filter medium levels. The water filtration cycle using the filtration rate of 5 m/h was the shortest. Because of the high level of iron and ammonium concentration, the filter medium was obstructed and the filtration rate became too low;

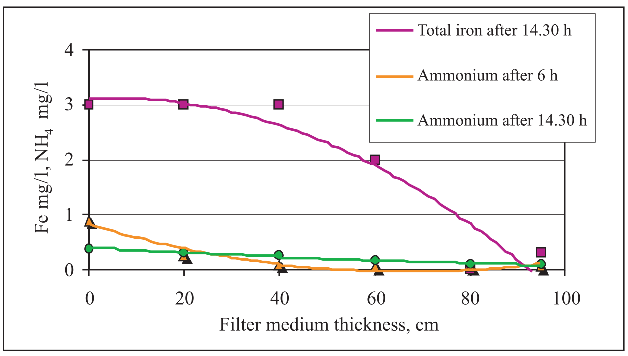

Fig. 2. Removal of iron and ammonium ions by natural silica sand fraction 0.7–2.0 mm at the primary filtration rate $V_0 = 5$ m/h

2 pav. Geležies bei amonio jonų šalinimas 0,7–2,0 mm frakcijos gamtiniu kvarco smėliu, kai pradinis filtravimo greitis $V_0 = 5$ m/h

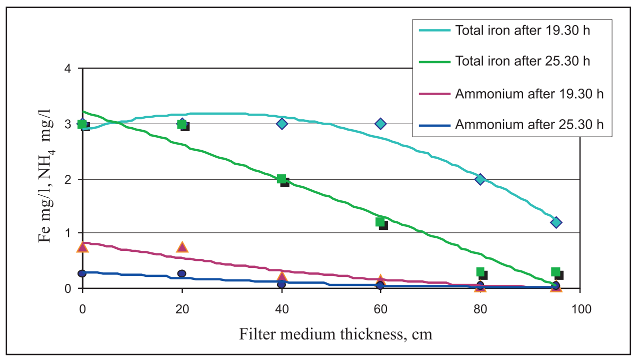

Fig. 3. Removal of iron and ammonium ions by natural silica sand fraction 0.7–2.0 mm at the primary filtration rate $v_0 = 3$ m/h

3 pav. Geležies bei amonio jonų šalinimas 0,7–2,0 mm frakcijos gamtiniu kvarco smėliu, kai pradinis filtravimo greitis v_0=3m/h

Table. **Removal of iron and ammonium ions by filtration through silica sand fraction 0.7–2.0 mm**

Lentelė. **Geležies bei amonio jonų šalinimas filtruojant pro 0,63–1,0 mm frakcijos kvarco smėlį**

Start / Stand by time	Filtration run, h	Filtration rate, m/h	Fe_{tot} C_1, mg/l	Fe_{tot} C_f, mg/l	Removal effectiveness, %	NH_4^+ C_1, mg/l	NH_4^+ C_f, mg/l	Removal effectiveness, %
2008 02 28	6.0	5.0	2.0	0.2	90	0.9	0.05	94
	16.0	3.0	2.0	0.1	95	0.9	0.05	94
2008 03 01	20.0	5.0	2.0	0.3	85	0.9	0.05	94
	28.3	3.0	3.0	0.3	90	0.4	0.1	75
2008 03 04	33.3	3.0	3.0	1.2	60	0.8	0.05	94
	39.3	3.0	3.0	0.3	90	0.3	0.04	87
2008 03 08	44.3	3.0	3.0	0.8	73	0.9	0.54	40
2008 03 12	51.3	3.0	2.0	0.8	60	1.1	0.74	33
	60.3	3.0	2.0	0.2	90	0.8	0.33	52
	68.3	3.0	0.7	0.1	86	2.08	1.38	34
2008 03 18	72.3	3.0	2.0	0.1	95	1.72	1.57	15

Note. Primary total iron concentration in the mixing unit was C_1 2–3 mg/l; ammonium solution concentration was 2.08–0.3 mg/l in the mixing unit; C_f = solution concentration in filtered water, mg/l.

Pastaba. Pirminė bendrosios geležies koncentracija maišymo talpykloje buvo C_1 2–3 mg/L; amonio tirpalo koncentracija maišymo talpykloje buvo 2,08–0,3 mg/L; C_f – koncentracija filtrate mg/L.

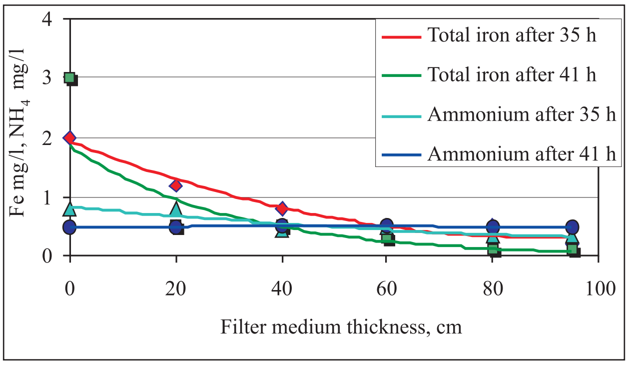

Fig. 4. Removal of iron and ammonium ions by natural silica sand fraction 0.7–2.0 mm at the primary filtration rate $v_0 = 3$ m/h after long-term filtration

4 pav. Geležies bei amonio jonų šalinimas 0,7–2,0 mm frakcijos gamtiniu kvarco smėliu, kai pradinis filtravimo greitis $v_0 = 3$ m/h po ilgo filtravimo periodo

after 16 h the filtration stopped. The longest (35 h) filtration time was used for a 3 m/h filtration rate. The filter was obstructed more slowly compared with the filtration rate of 5 m/h, but the effectiveness of ammonium removal from groundwater during the latter filtration stage decreased to only 33%. Using the filtration rate of 5 m/h, the efficiency for ammonium was 94%. The filtration rate was constant up to the end of the cycle when water treatment effectiveness was up to 75%.

During the experiment, the constant primary concentration of solution was established by using a mixture of groundwater from the laboratory tap with a solution of iron sulfate, and ammonium chloride concentration in the mixing unit (first reservoir) was less than in the filter intake, but it became higher during the mixing and aeration processes because of a different solubility of added ingredients of iron and ammonium compounds in water. A brown colour of oxidized iron compounds appeared in the mixing reservoir of water in the filtration unit intake, and the layer of oxidized iron compounds was not removed from the filter medium during filtration. During the long-term period, some part of oxidized iron compounds from the first filter medium layer passed into the deep silica sand layers, and iron removal efficiency increased. Conventional units of iron removal are operated with a coated silica sand layer on the top of the filtration unit, and when the accumulated quantity of oxidized iron compounds exceeded the allowable level it had to be removed because of the possibility to increase iron concentration in the filter unit outlet. The experimental conditions were adjusted to real conditions. The concentrations of iron compounds in supplied water and filtered water are presented in Fig. 2–4.

Dependency result was evaluated following the polynomial model when the statistical determination coefficient was $R^2 = 0.87$ for ammonium removal and $R^2 = 0.96$ for the removal of iron compounds. The concentration of ammonium ions increased during the longer filtration period. Figure 1 shows that the filtration at a 5 m/h filtration rate lasted no longer than 16 h when the achieved ammonium concentration in treated water did not exceed the levels permitted by HN 24 : 2003. When the primary ammonium concentration was below 1.0 mg/l, filtered water could contain <0.5 mg/l of ammonium ions.

The removal of iron and ammonium ions in filter media was tested for comparison, and the results are presented in Fig. 2–4.

Iron concentration in filtered water increased during a longer filtration period. The results were statistically evaluated with $R^2 \approx 0.94–0.99$.

The best results of iron removal from water were obtained when ammonium ions were removed up to 0.5 mg/l (Figs. 2–4).

Results presented in Fig. 2 show that during the long-term treatment process the efficiency decreased to 75% using a 5 m/h filtration rate. The best removal efficiency was obtained by filtration for no longer than 16 h when the efficiency decreased to 85%. When a silica sand filter medium with a particle size of 0.7–2.0 mm was used, the highest ammonium removal efficiency was 94%.

Using a slow (3 m/h) filtration rate, treatment efficiency after 8 h was 90% and after 12 h decreased to 60%. The highest treatment efficiency was 95%. Using a slow filtration rate, ammonium removal after 18 h was 33%, and the highest treatment efficiency was 94%.

A comparison of results of different filtration rates for iron and ammonium removal is presented in Fig. 2–4.

The biggest quantity of removed iron ions was obtained at a 3 m/h filtration rate through silica sand as a filter medium, and the filtration cycle was the longest compared with the 5 m/h filtration rate. A comparison of the quantity of removed ammonium compounds shows that the largest amount of adsorbed ammonium ions using 3 m/h was by 57% bigger than at the filtration rate of 5 m/h, and iron removal effectiveness was nearly the same for both 3 m/h and 5 m/h. The same comparison of the decreased concentrations of iron and ammonium ions was made for different filtration cycles of silica sand filter media with the primary iron compound concentration below 3 mg/l and ammonium ion concentration below 2 mg/L concentration after the aeration unit.

Experimental data presented in Figs. 2–4 were analysed independently of different heights of the filter medium (the same volume of water filtered through a silica sand medium was used). The results show that silica sand filter medium has the biggest capacity for iron compound removal. The 0.7–2.0 mm particle size of silica sand has a higher removal capacity for iron compounds than for the removal of ammonium ions in the same filter medium.

According to our results, natural coarse silica sand filter medium can be used for both iron and ammonium removal from groundwater. The obtained characteristics of iron and ammonium removal can be useful for the estimation, project design, and the equipment start up procedure, renovation and maintenance of existing water treatment plants. The preliminary experimental results of this research need more extensive studies in the future with evaluation of suspended matter, different chemical ingredients, pH and other substances that possibly influence the removal process.

CONCLUSIONS

1. During the laboratory experiment with a pilot test bench, the possibility to use two different filtration rates (3 m/h and 5 m/h) of natural silica sand with a particle size of 0.7–2.0 mm for iron and ammonium removal from groundwater was achieved.

2. The highest iron compounds removal efficiency (95%) was achieved at a 3 m/h filtration rate. When the concentration of

primary iron compounds in water solution was 2 mg/l, the total iron residual in filtered water was 0.1–0.8 mg/l.

3. From the two filtration rates, the highest removal of iron and ammonium ions from groundwater solution was shown by the slow filtration rate. Removal efficiency was 85–90% and 60–95%, respectively, for 5 m/h and 3 m/h filtration rates. When the concentration of primary ammonium ions was 0.3–2.0 mg/l, the content of residual ammonium ions was 0.04 mg/l and 1.38 mg/l in filtrated water, respectively.

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Marina Valentukevičienė

EKSPERIMENTINIAI GELEŽIES BEI AMONIO JONŲ ŠALINIMO IŠ POŽEMINIO VANDENS GAMTINIU KVARCO SMĖLIU TYRIMAI

Santrauka

Eksperimentiniai geležies bei amonio jonų šalinimo iš požeminio vandens tyrimai atlikti naudojant gamtinio kvarco smėlio užpildą. Buvo naudojamas 0,7–2,0 mm frakcijos 750 mm aukščio kvarco smėlio užpildas. Prieš eksperimentą gamtinis kvarco smėlis išplautas ir išdžiovintas krosnyje apie 105 °C temperatūroje. Pro filtro įkrovą praleistas eksperimentinis tirpalas iš 500 l požeminio vandens ir geležies sulfato bei amonio chlorido. Lyginant gautus eksperimento rezultatus nustatyta, kad filtravimas pro 1 m kvarco smėlio užpildą efektyviausiai šalina geležies junginius iš požeminio vandens. Geležies junginių šalinimo efektyvumas siekė atitinkamai 95%, kai buvo naudojamas stambios frakcijos kvarco smėlis. Geriausiai amonio junginius šalino 0,7–2,0 mm kvarco smėlis – šalinimo efektyvumas siekė 94%.

Марина Валентукявичене

ЭКСПЕРИМЕНТАЛЬНОЕ ИССЛЕДОВАНИЕ УДАЛЕНИЯ ЖЕЛЕЗА И АММОНИЙ-ИОНОВ ИЗ ПОДЗЕМНОЙ ВОДЫ КВАРЦЕВЫМ ПЕСКОМ

Резюме

Экспериментальные исследования удаления железа и аммоний-ионов из подземной воды проводились загрузкой природного кварцевого песка.

Применялась загрузка кварцевого песка крупной (0,7–2,0 мм) фракции, высотой 750 мм. Природный кварцевый песок был промыт и высушен при температуре 105 °С. Через фильтрующую загрузку протекал экспериментальный раствор, изготовленный из 500 л подземной воды с растворами сульфата железа и хлорида аммония. Сравнением результатов эксперимента определено, что фильтрование через 1 м загрузки кварцевого песка наиболее эффективно удаляет из подземной воды соединения железа. Эффективность очистки соединений железа кварцевым песком крупной фракции достигала 95%. Эффективность очистки соединений аммония кварцевым песком, размеры зерен которого составляли 0,7–2,0 мм, достигала 94%.