



Removal of radionuclides from Estonian groundwater using aeration, oxidation, and filtration

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Abstract. Groundwater in the north-eastern suburb of Tallinn was analysed to determine the content of iron, manganese, sulphides, ammonia, and radionuclides (Ra^{226} , Ra^{228}) and total radioactivity. It was established that for several wells of the Cambrian–Vendian water layer the annual effective dose would exceed the EU guideline for drinking water (0.1 mSv/yr). The purpose was to find a technology for simultaneous purification of groundwater from iron, manganese, sulphides, ammonia, and some radionuclides (Ra^{226} , Ra^{228}). A pilot plant consisting of a Venturi-type aeration unit GDT (Gas-Degas Technology, Mazzei Corp., USA), an oxidation tank, and two-stage filtration columns was constructed. Several non-catalytic (Everzit Special Plus, sand) and catalytic filter materials (Filtersorb FMH, Pyrolox) were tested. Along the flow sheet from the aerator to the II stage filter outlet the pH, dissolved oxygen and carbon dioxide, turbidity, the content of iron, manganese, and ammonia were monitored, mainly by using spectrophotometry (HACH DR/2000). Radioactivity of water samples was determined by the Estonian Radiation Centre using the liquid scintillation counting method and γ -spectrometry. It was established that by intensive aeration of groundwater followed by oxidation for a certain contact time and appropriate selection of filter materials of different properties, it was possible to remove together with iron and manganese also radium isotopes. The total effectiveness of the process was 90% removal of gross-alfa and 70% removal of gross-beta activity of groundwater. Since the uranium content in the well water was marginal and radon was almost totally (99%) removed in the degas separator, the total effective dose was calculated by Ra^{226} and Ra^{228} . It was about 0.067 mSv/yr, which is lower than the EU DWD guideline (0.1 mSv/yr). The theoretical assumption that radionuclides were already removed with $\text{Fe}(\text{OH})_3$ flocks in the first filtration step was verified by examining wash water. The radioactivity of wash water containing precipitate and the filtered wash water were measured. Results showed 4.6 times higher gross-alfa and 5.3 times higher gross-beta activity in the water containing precipitate. The co-precipitation process, where MnO_2 and $\text{Fe}(\text{OH})_3$ flocks played an essential role, resulted in simultaneous removal of radium isotopes. Co-precipitation of radium with MnO_2 was more effective than with $\text{Fe}(\text{OH})_3$.

Key words: groundwater, iron, manganese, radium isotopes, aeration, filtration.

INTRODUCTION

Groundwater is usually considered as the best drinking water. In many cases this is true, but not always. Raw groundwater very often contains many undesired natural admixtures such as iron, manganese, hydrogen sulphide, ammonia, fluoride, chloride, sulphate, boron, barium, radionuclides (radon, radium, uranium), etc. Groundwater radioactivity is especially dangerous. Contamina-

tion with radionuclides is attributed to the contact of groundwater with uranium- and thorium-containing minerals and rocks. The majority of the best-known radionuclides (Rn^{222} , Ra^{224} , Ra^{226} , U^{238} , U^{234}) are α -emitters as opposed to Ra^{228} , which is a β -emitter. The two radium isotopes of primary concern are Ra^{226} and Ra^{228} because of their potential carcinogenic impact.

The WEKNOW (Web-based European Knowledge Network on Water) investigation in 2005 revealed increased content of radionuclides in groundwater in 18 European countries: Austria, Bulgaria, Czech Republic,

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Germany, Spain, France, Finland, Greece, Ireland, Italy, Hungary, Sweden, Slovakia, Slovenia, Luxemburg, Portugal, Poland, and Estonia [1].

About 2/3 of the drinking water in Estonia is produced from groundwater. Removal of radionuclides from groundwater is at present an especially 'hot' problem in Estonia. Systematic studies on groundwater radioactivity in Estonia were started in 1994–1998, followed by some complementary studies in 2001–2009 [2]. The summarized radiation activities, gross-alpha and gross-beta, were determined as well as the ^{222}Rn content. It was established that for more than 70% of the wells in the Cambrian–Vendian water layer, the special activity index (I) was higher than 1.0. This means that the consumption of 2 L/day of water will lead to up to a 2.7 times higher effective dose than the WHO guideline of 0.1 mSv/year. According to the groundwater studies, about 90% of groundwater radioactivity in Estonia is caused by the content of radium isotopes Ra^{226} and Ra^{228} [3].

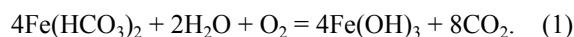
Radium is present in water in the form of a large 'hydrophobic' cation and can be removed by strong acid cationite (SAC). Uranium is in the common range of $\text{pH} = 6\text{--}8$ present in the form of anion $\text{UO}_2(\text{CO}_3)_2^{2-}$, and consequently, should be removed by anion exchange through strong basic anionite (SBA) [4,5]. Studies carried out in the USA have proved that the best solution for simultaneous removal of radium and uranium from water is filtration of water through SAC and SBA. An alternative to the ion exchange process is reverse osmosis [6]. Significant disadvantages of both processes along with their relatively high costs are the requirement of proper pretreatment of groundwater to remove iron, manganese, hardness, organics, etc. as these cause ionite inactivation and fast fouling of the membrane. Furthermore, application of ion exchange is accompanied by the periodic discharge of salted radioactive regeneration solutions and reverse osmosis, and even worse – by continuous discharge of highly salted radioactive retentate (15–25% of the input).

The selection of groundwater treatment processes depends on the raw water quality. A typical flow sheet consists of a pre-aeration (pre-oxidation) stage followed by filtration/adsorption and post-disinfection stages. Some very interesting studies [7,8] pointed to possibilities involving more economical ways for radium removal from groundwater than ion exchange and reverse osmosis: by natural separation processes of dissolved iron and manganese or by external formation of hydrous manganese oxide (HMO) or by using catalytic filtration materials containing MnO_2 .

The main aim of the present study was elaboration of an economical and possibly chemical-free technology for radium removal from iron- and manganese-containing groundwater using aeration and filtration processes.

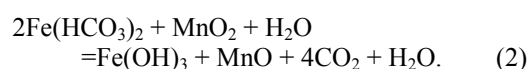
During the planning of our study, we started from the following hypotheses. Iron and manganese are natural constituents of soil and rocks. Usually natural waters have an iron content that is greater than the manganese content. Waters that have iron levels greater than 10 mg/L or manganese levels greater than 2 mg/L are seldom found [9]. Iron exists in water in the +2 or +3 oxidation states, whereas manganese exists in either +2, +3, +4, +6, or +7 oxidation states. However, iron(III) and manganese(IV) are the only stable oxidation states found in waters containing oxygen. Before iron and manganese can be filtered (usually through sand or anthracite) they need to be oxidized to a state in which they can form insoluble complexes.

Ferrous iron (Fe^{2+}) is oxidized to ferric iron (Fe^{3+}), which readily forms the insoluble iron hydroxide complex $\text{Fe}(\text{OH})_3$. Manganese (Mn^{2+}) is oxidized to Mn^{4+} , which forms insoluble MnO_2 [8]. For example, iron oxidation by dissolved oxygen of air:



The oxidation rate depends on the pH, alkalinity, organic content, and oxidant concentration. It is very difficult to oxidize Fe^{2+} in the presence of humic and tannic acids [10]. Both of them retard the oxidation of Fe^{2+} significantly. In spite of the fact that ferric iron complexes in water are charged positively, their large flocks can also remove some positively charged radium cations as a result of sweep coagulation. According to research information [11], manganese dioxide (MnO_2) particles created after divalent manganese oxidation in water form in neutral, and especially, in basic media negatively charged flocks, which should possess higher potential for the removal of radium cations than the ferric hydroxide flocks. All oxidation reactions (with air, ozone, permanganate, chlorine, etc.) liberate CO_2 , which together with the gases (CO_2 , Rn , CH_4 , H_2S , etc.) initially dissolved in groundwater may cause serious problems in filter bed operation.

Contemporary methods for the removal of iron, manganese, and hydrogen sulphides are mainly based on catalytic filter materials containing MnO_2 (*Birm*, *Manganese Greensand*, *Filox*, *Pyrolox*, *Everzit-Mn*, etc.). In the oxidation/reduction reaction MnO_2 is reduced to MnO , and ferrous iron is oxidized to ferric iron forming ferric hydroxide, which is precipitated [9]:



Manganese dioxide works most efficiently at a pH of 6.5 to 9.0. It should not be used on waters that contain bacterial or organic iron. It was expected that adsorption of radium cations onto the MnO_2 layer of the catalytic filter materials can serve as a third option for radium removal.

Thus, our study was focused on the optimization of the groundwater pilot plant treatment scheme for iron, manganese, ammonia, and sulphides removal, containing the stages of aeration, oxidation, and filtration/catalytic filtration, varying the dissolved oxygen concentration, residence time for oxidation, number and composition of the I and II stage filter materials (including thickness of layer, diameter of particles, contact time, etc.).

MATERIALS AND METHODS

Pilot plant studies were carried out with raw groundwater ($Q = 3 \text{ m}^3/\text{h}$) from the Viimsi Water Treatment Plant (WTP; 6 km from Tallinn city). The initial contents of iron, manganese, ammonia, and radionuclides exceeded the guidelines of the EU DWD [12] (maximum permissible concentrations, MPC, are in brackets): 0.1–2.6 (0.2) mg/L; 0.004–0.183 (0.05) mg/L;

0.07–1.4 (0.5) mg/L, and 0.08–0.73 (0.1) mSv/year. The flow sheet consisted of a pre-aeration unit GDT (Gas–Degas Technology) with an injector, an intermediate oxidation tank to provide the needed residence time for iron, and especially, for manganese oxidation, a centrifugal degas separator, and one or two filtration columns ($D = 180 \text{ mm}$; $H = 2200 \text{ mm}$) with different catalytic filter materials (Fig. 1). Different filter materials (Table 1) were tested during pilot studies; variations were made in their sequence and heights.

The pilot plant was constructed to enable changes in the order of technological steps and flow rates of air and water. Groundwater was pumped through the injector at a rate of 2500 L/h, to achieve air and water ratio $2\text{--}6 \text{ m}^3/1 \text{ m}^3$ and to enable the GDT separator to work at the maximum degas regime. The air inlet to the injector was regulated with a ball-cock; direct measurements were not carried out, whereas the necessary amount of air was derived from the water dissolved oxygen content, which preferably was 6 mg/L. Preliminary tests

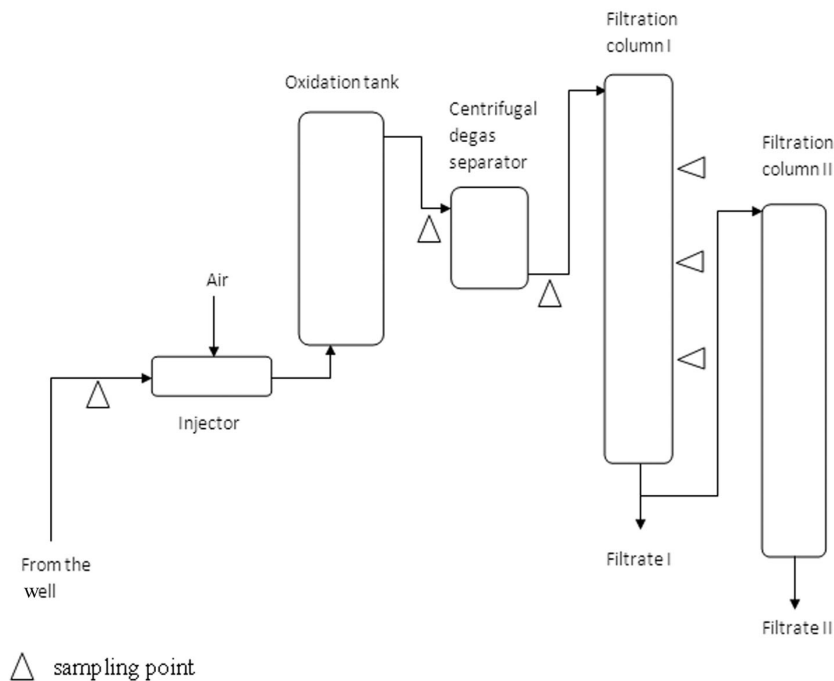


Fig. 1. Pilot plant flow sheet.

Table 1. Filter materials used during pilot plant studies

Material	Composition	Density, g/cm^3	Particle size, mm	Bed height, mm
Filtersorb [®] FMH	MnO ₂ coated dolomite	1.56	0.8–1.26	1000
Everzit Special Plus	92% carbon	1.3	0.8–2.0	800
Zeolith N	Natural zeolith, up to 72% SiO ₂ and up to 13% Al ₂ O ₃	1.6–1.8	1.0–2.5	1100
Pyrolox [™]	Natural MnO ₂	1.9	0.2–0.4	300–600

indicated that as a result of iron and manganese oxidation reactions, intensive liberation of CO₂ started already during the flow through the Venturi-type injector. To promote the removal of gases, an oxidation tank (a contact column with alterable volume) was installed after the injector. The primary purpose of the oxidation tank was to enable sufficient contact time for iron and manganese oxidation, but it also worked as a preliminary gas separator, removing some of the injected air, and produced CO₂ while the majority of gases were released to the atmosphere from the centrifugal degas separator. The volume of the oxidation tank was altered between 94 and 47 L (contact time varied between 2.4 and 1.2 min). The aerated water was filtered at a rate of 200 L/h (maximum filtration rate 10 m/h) while the rest of the water was channelled. Despite the intensive degas separation before the filters, some gases still accumulated in the filter body, causing an extra pressure drop. Since the filters were opened to the atmosphere, the filtration rate strongly depended on the pressure drop, and accumulated gases caused the specific filtration rate to fall by up to 40% (from 8 to 5 m/h).

In addition to filtrate quality monitoring, some water parameters were determined in different points of the pilot plant. Samples were taken from the well, after the injector, after the oxidation tank, after the degas separator, from the filter body (various heights), and from filtrate I and filtrate II to measure temperature, pH, dissolved oxygen (electrode Marvet Junior 156), and dissolved carbon dioxide (HACH colorimetric express method, phenolphthalein and NaOH solution). In both filtrates important physical and chemical parameters, such as colour (spectrophotometer HACH DR/2000, method 8025, APHA Pt-Co method, $\lambda = 455$ nm), turbidity (spectrophotometer HACH DR/2000, method 8237, $\lambda = 510$ nm), total iron (colorimeter MaxiDirect, 1,10-Phenanthroline method, $\lambda = 530$ nm), manganese (colorimeter MaxiDirect, PAN method, $\lambda = 560$ nm), and ammonia (colorimeter MaxiDirect, indophenol blue method, $\lambda = 610$ nm) were measured as well.

Radioactivity of the water samples (gross-alfa and gross-beta activity in Bq/L) was determined by the Estonian Radiation Centre (liquid scintillation counting method). Forte et al. [3] pointed out that over 90% of the expected effective radiation dose in Estonian Cambrian–Vendian groundwater is caused by Ra²²⁶ and Ra²²⁸. The effective dose assessment for the accuracy of Ra²²⁶ and Ra²²⁸ content in filtrate I and filtrate II was also performed in the Estonian Radiation Centre (liquid scintillation counting and γ -spectrometry). Effective annual doses were calculated from WHO recommendations [13].

Filters were backwashed with raw water 1–2 times a week; flow rates were chosen according to filter materials. As in regular backwash of filters, the average

radioactivity of backwash water (0.41 Bq/L) was significantly lower than the permissible clearance level in Estonia (10 Bq/L), the backwash water was directly discharged to the sewerage. Also the radioactivity of the filter bed, which had been in operation for about a month, was checked using scanning electron microscopy. No traces of radium fixed to grains were discovered, which means that the radium bound to the iron hydroxide and manganese dioxide flocks was actually removed together with them during filter backwash.

RESULTS AND DISCUSSION

Ferrous iron (Fe²⁺) was oxidized to ferric iron (Fe³⁺), which readily formed the insoluble iron hydroxide complex Fe(OH)₃. These flocks formed a blockage, which increased with time. In addition, insoluble gases (injected air and carbon dioxide formed during the oxidation process) accumulated in the filter body, but were released when forming larger bubbles. After 48 hours of continuous work, accumulation of gases stopped and the pressure drop remained almost constant. It gave us reason to believe that the saturation of gas accumulation was achieved. Still, we could register an increasing pressure drop at the top layer of the filter media in filter I (70–260 mm from the surface), which represented Fe(OH)₃ accumulation.

To understand the origin of gases, the oxidation tank was removed from the flow sheet, so that the Venturi-type injector was followed by the degas separator and filters. Results of this change showed that without the oxidation tank, the pressure drop increased by about 50%. This demonstrates clearly the importance of detention time and maximum liberation of carbon dioxide prior to the filtration step.

The dissolved oxygen concentration in well water was less than 2 mg/L. Fast saturation with air oxygen was achieved in the Venturi-type injector, where dissolved oxygen concentration rose above 8 mg/L. In the oxidation and filtration steps the concentration decrease was around 6–7 mg/L in the filtrate, whereas dissolved carbon dioxide concentration increased due to oxidation, but then again dissolved oxygen separated from the liquid phase forming a gas blockage in the filter body (Fig. 2).

Figure 3 illustrates the changes in ammonia, total iron, and manganese content in the two-stage filtration system. The overall removal of radioactivity is shown in Fig. 4. The co-precipitation process, where MnO₂ and Fe(OH)₃ flocks play an essential role, results in radionuclide removal [14]. During Fe(OH)₃ formation, hydrophobic Ra cations associated with charged ferrous ions and ferrous intermediates form a precipitate. The second mechanism is believed to be the adsorption onto Fe(OH)₃ flocks and MnO₂ grains. Water radioactivity

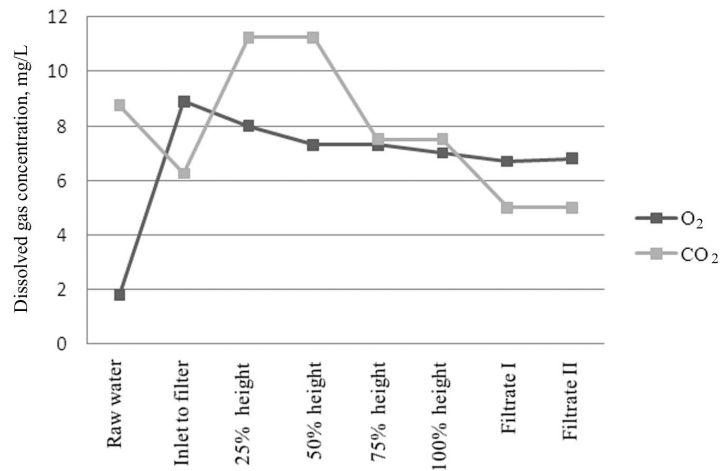


Fig. 2. Changes in dissolved oxygen and carbon dioxide concentration.

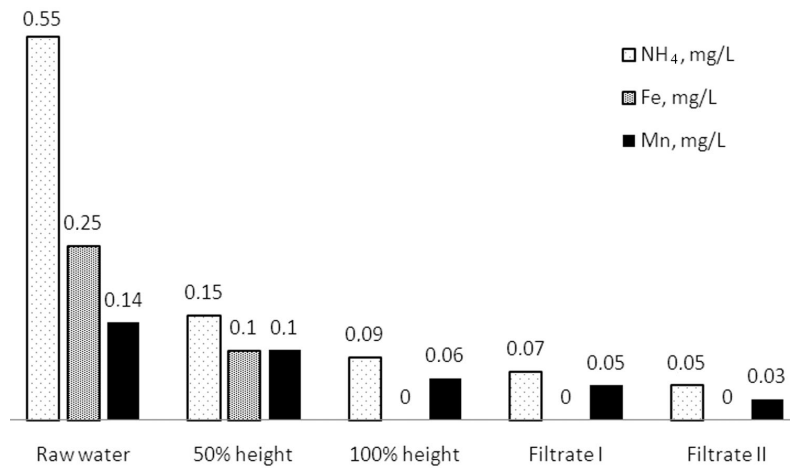


Fig. 3. Changes in ammonia, total iron, and manganese content in the two-stage filtration system.

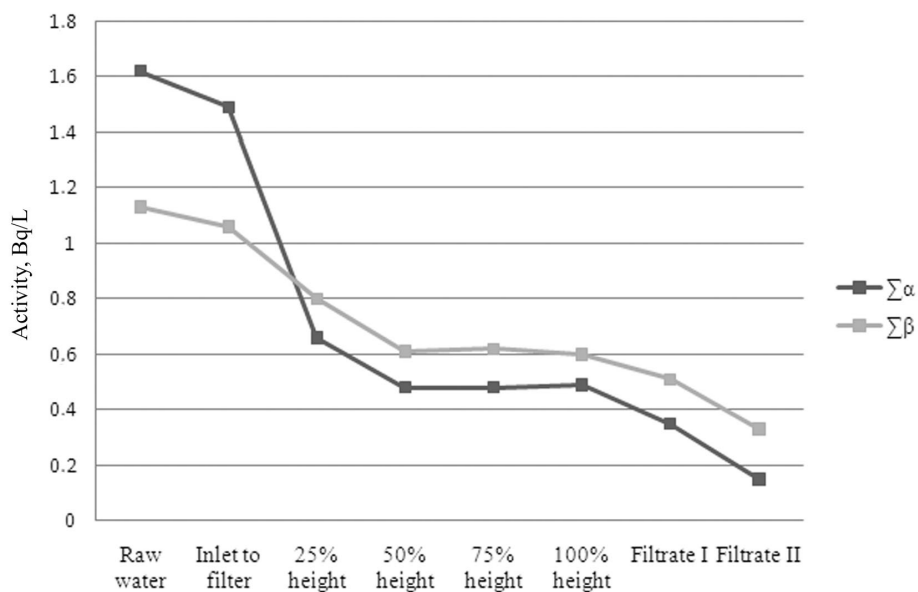


Fig. 4. Changes in the radioactivity of water.

and iron were removed concurrently during filtration and precipitation in backwash water, which was of higher radioactivity. This proved simultaneous removal of iron, manganese, and radionuclides. Patel and Clifford [8] found that adsorption onto MnO_2 is more effective than onto $\text{Fe}(\text{OH})_3$, but since in our case the natural Fe^{2+} content of well water was significantly higher than its manganese content, pilot studies mainly showed the importance of $\text{Fe}(\text{OH})_3$ in the radionuclide removal mechanism. Even if co-precipitation with MnO_2 is more effective, it is reasonable to make use of iron available in raw water, which also has to be removed.

In the top layer of filter I (400 mm) 70% of gross-alfa and 45% of gross-beta activity and most of the iron (72%) were removed. In filter II 55% of gross-alfa and 35% of gross-beta activity was removed compared to filtrate I. The total effectiveness was 90% removal of gross-alfa and 70% removal of gross-beta activity. Gross-beta activity includes K^{40} (in Estonian groundwater on average 0.24 Bq/L [3]), which was not taken into account in total effective dose calculation because it does not accumulate in human tissues. Since the uranium content in well water was marginal and radon was almost totally (99%) removed in the degas separator, the total effective dose was calculated by Ra^{226} and Ra^{228} and was about 0.067 mSv/a.

The theoretical assumption that radionuclides are removed with $\text{Fe}(\text{OH})_3$ flocks in the first filtration step was verified by examining the wash water. The radioactivity of the wash water containing precipitate and that of the filtered wash water were measured. The results showed 4.6 times higher gross-alfa and 5.3 times higher gross-beta activity in the water containing precipitate.

Removal of radionuclides was achieved simultaneously by three different mechanisms: co-precipitation with iron hydroxide flocks, adsorption onto in situ formed manganese dioxide, and filtration through MnO_2 layered filter material.

After groundwater saturation with air oxygen in the pre-aeration unit, ferrous iron (Fe^{2+}) was oxidized to ferric iron (Fe^{3+}), which formed insoluble positively charged iron hydroxide complexes or oligomers $\text{Fe}(\text{OH})_n^+$. Divalent manganese, Mn^{2+} , was oxidized to Mn^{4+} , which in water with $\text{pH} > 7$ usually forms negatively charged insoluble flocks of $\text{MnO}_2 \cdot n\text{H}_2\text{O}$. Iron hydroxide flocks swept part of the radium cations in the process of coagulation and settling. Especially effective was radium removal with MnO_2 flocks formed in water in situ as a result of dissolved divalent manganese oxidation. Additional removal of radium was achieved in the process of filtration of water through filter materials covered with MnO_2 layers of various concentration. The following configuration of materials gave the best results:

- Filtration column I: Filtersorb[®]FMH $h = 1000$ mm; Pyrolox[™] $h = 300$ mm
- Filtration column II: Zeolith N $h = 1100$ mm.

This configuration enabled us to achieve water quality that meets the requirements of the EU DWD in one filter only, leaving the second filter for a 'polishing step'.

CONCLUSIONS

This study focused on the optimization of the groundwater pilot plant treatment scheme for the removal of iron, manganese, ammonia, and sulphides and contained the stages of aeration, oxidation, and filtration/catalytic filtration, by varying the dissolved oxygen concentration, residence time for oxidation and filtration, and the number and composition of the I and II stage filter materials (incl. thickness of layer, diameter of particles, etc.). Ferrous iron (Fe^{2+}) was oxidized to ferric iron (Fe^{3+}), and divalent manganese (Mn^{2+}) to manganese dioxide (MnO_2). The co-precipitation process, where MnO_2 and $\text{Fe}(\text{OH})_3$ flocks played an essential role, resulted in simultaneous removal of radium isotopes. Co-precipitation of radium with MnO_2 was more effective than with $\text{Fe}(\text{OH})_3$. In the case of water with high radioactivity, the last polishing stage of the process can be adsorption of Ra^{2+} onto MnO_2 containing surface of catalytic filter material or application of zeolites. The detention time of water in the oxidation tank played a key role in the chemical reactions implementation and gas (CO_2) removal to avoid a sharp increase in the pressure drop of the filters. The total effectiveness of the described technology was 90% removal of gross-alfa and 70% removal of gross-beta activity. To protect rights to the technology, applications for an Estonian patent (P201000052) and international patent (PCT/EP2010/061529) have been submitted.

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Radionukliidide kõrvaldamine Eesti põhjaveest õhustamise, oksüdeerimise ja filtrimisega

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Tallinna äärelinna põhjavees määrati raua, mangaani, sulfiidide, ammooniumi ja radionukliidide (Ra^{226} , Ra^{228}) sisaldust ning summaarset radioaktiivsust. Tehti kindlaks, et mitmete Kambriumi-Vendi põhjaveekihi puurkaevude joogiveest saadav aastane efektiivdoos ületab Euroopa Liidu kehtestatud piirnормi (0,1 mSv/a).

Antud töö eesmärgiks oli välja töötada tehnoloogia, mis võimaldaks põhjaveest kõrvaldada samaaegselt raua, mangaani, sulfiidide, ammooniumi ja mõningaid radionukliide (Ra^{226} , Ra^{228}). Konstrueeriti pilootseade, mis koosnes Venturi-tüüpi inžektorist, õhustamisest GDT (Gas-Degas Technology, Mazzei Corp., USA), oksüdatsioonikambrist ja kaheastmelisest filtrist. Seadmel katsetati erinevaid mittekatalüütilisi (Everzit Special Plus, liiv) ja katalüütilisi (Filtrisorb FMH, Pyrolox) materjale. Pilootseade oli varustatud proovivõtukraanidega, mis olid paigutatud iga tehnoloogilise astme vahele ja filtrikorpusse. Spektrofotomeetriga (HACH DR/2000) määrati raua, mangaani ja ammooniumi sisaldust ning hägusust, lisaks määrati pilootseadme erinevates punktides pH-d, lahustunud hapniku ja süsinikdioksiidi sisaldust. Vee radioaktiivsus määrati Keskkonnaameti kiirgusosakonnas, kasutades vedelikstintillatsiooni meetodit ja γ -spektromeetriat. Määrati kindlaks, et intensiivse õhustamise, piisava oksüdatsiooni ja sobiva materjalide valikuga on võimalik samaaegselt kõrvaldada veest raua ning mangaani koos raadiumi isotoopidega. Protsessi efektiivsuseks saavutati 90%-line summaarse α -kiirguse ja 70%-line summaarse β -kiirguse vähenemine põhjavees. Kuna uraanisisaldus uuritavas põhjavees oli marginaalne ja radoon eemaldatakse peaaegu täielikult (~99%) õhustusseadmes, arvutati joogiveest saadav aastane efektiivdoos Ra^{226} ning Ra^{228} põhjal ja filtraadi puhul saadi tulemuseks 0,067 mSv/a, mis jääb alla Euroopa Liidu kehtestatud piirnормi (0,1 mSv/a). Teoreetiline eeldus, et suur osa radionukliidide eraldub koos $\text{Fe}(\text{OH})_3$ flokkidega juba esimeses filtreerimisastmes, leidis kinnitust filtrite pesuvee uurimisel. Radioaktiivsust mõõdeti nii sadet sisaldava kui filtreeritud pesuvee puhul ja tulemused näitasid sadet sisaldavas pesuvees 4,6 korda kõrgemat summaarset α -kiirgust ning 5,3 korda kõrgemat summaarset β -kiirgust. Koossadenemise protsess, kus MnO_2 ja $\text{Fe}(\text{OH})_3$ osakestel on tähtis roll, tõi kaasa raadiumi isotoopide samaaegse eraldumise. Raadiumi kaassadenemise protsess on efektiivsem MnO_2 sademel, võrreldes $\text{Fe}(\text{OH})_3$ sademega. Lähtevee kõrgema radionukliidide sisalduse puhul on võimalik kasutada teise astme filtris vajalikku katalüütilist adsorbenti või ionii.

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