LOODUSLIKE RADIONUKLIIDIDE EEMALDAMINE JOOGIVEEST NING NORM JÄÄTMETE KÄITLUSVÕIMALUSED

PROJEKTI LIFE ALCHEMIA SEMINAR

3-4 märts 2020. Viimsi (Eesti)

KOKKUVÕTE

Sisukord

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Eessõna

Seminar "Looduslike radionukliidide eemaldamine joogiveest ning NORM jäätmete käitlusvõimalused" korraldati LIFE ALCHEMIA projekti raames.

LIFE ALCHEMIA on Euroopa Liidu projekt, mida kaasrahastatakse Euroopa Liidu LIFE programmist. Projekti koordinaatoriks on CARTIF tehnoloogiakeskus. Projektis osalevad ka Almería provintsi valitsus, päikeseenergia uurimiskeskus CIESOL, Tallinna Tehnikaülikool, Tartu Ülikool ning Viimsi Vesi AS.

Projekt kutsuti ellu nimetatud asutuste koostöö tulemusena, mis oli tingitud vajadusest lahendada probleemid joogivee radioaktiivsususega – Almeria provintsis (Hispaania) ning Viimsis (Eesti). Looduslikult esinevad radionukliidid, mis kuuluvad uraani ning tooriumi lagunemisritta, satuvad põhjavette seda ümbritsevate kivimite osalisel lahustumisel või läbi pinnase, mistõttu võib nende kontsentratsioon vees ületada Euroopa Nõukogu direktiivis 2013/51/Euratom ette nähtud tasemeid.

Efektiivne, ohutu, töökindel ning kulutõhus joogivee töötlemise tehnoloogia on eelduseks vee kvaliteedinõuete saavutamisel.

LIFE ALCHEMIA eesmärgiks on pakkuda sellele probleemile välja lahendus. Ühelt poolt uuritakse filtermaterjalil põhineva töötlustehnoloogia kasutamist, mis vähendab vee töötlemise kulusid kuni viis kohta. Teisalt otsitakse võimalusi radionukliidide ärastamisel tekkiva NORM jäägi/jäätme (ingl. k. Naturally Occuring Radioactive Material) koguse vähendamiseks.

Oodatavad tulemused ning kasu keskkonnale:

- vähendada radionukliidide kontsentratsiooni joogivees 75 kuni 90%;
- vähendada veetöötluse käigus tekkivaid NORM jääke/jäätmeid kuni 90% (võrreldes projekti eelsel ajal AS Viimsi Vesi veetöötlusjaamas tekkinud kogustega);
- vähendada energiatarvet 80% võrreldes enim kasutatud veepuhastussüsteemidega nagu näiteks pöördosmoos.

Eesmärkide saavutamiseks rajati neli, erineva strateegiaga, pilootjaama – kolm neist asuvad Hispaanias, Almería provintsis ning üks Eestis, Viimsis. Pilootjaamad on näidanud paljulubavaid tulemusi, mida tutvustati koos jaamade tööskeemidega toimunud seminari käigus. Seminari käigus tutvustati vee-ettevõtetele alternatiivseid, keskkonnasäästlike veetöötluse süsteeme, mille abil on võimalik veest radionukliide eemaldada.

Käesolev kokkuvõte annab ülevaate kahepäevasest seminarist ning sisaldab endas ettekannete kokkuvõtteid. Ettekanded on täismahus kättesaadavad LIFE ALCHEMIA projekti kodulehel: https://www.lifealchemia.eu/en/seminars-and-events/

Marta Gómez, CARTIF tehnoloogiakeskus, Hispaania

Maria Leier, Taavi Vaasma, Siiri Suursoo, füüsika instituut, Tartu Ülikool, Eesti

Tänusõnad

Seminari “Looduslike radionukliidide eemaldamine joogiveest ning NORM jääkide/jäätmete käitlusvõimalused“ organiseeris AS Viimsi Vesi, Tartu Ülikool ja Tallinna Tehnikaülikool. Seminari korraldamist ja läbiviimist finantseeris LIFE ALCHEMIA (LIFE16 ENV/ES/000437).

Seminari korraldajad soovivad tänada kõiki seminaril osalejaid, kes seminari eduka toimumise võimalikuks tegid.

LIFE ALCHEMIA projekti ülevaade

LIFE ALCHEMIA on Eesti ja Hispaania koostööprojekt. Projektis ette nähtud tegevustega alustati 2. oktoobril 2017 ning projekti planeeritud lõpp on 31. detsember 2020. Projekti kuuluvad järgmised partnerid:

- CARTIF tehnoloogiakeskus, Hispaania;
- Päikeseenergia uurimiskeskus CIESOL, Hispaania;
- Almeria provintsi kohalik omavalitsus (DIPALME), Hispaania;
- Tallinna Tehnikaülikool (TTÜ), Eesti;
- Tartu Ülikool (UT), Eesti;
- Viimsi Vesi AS.

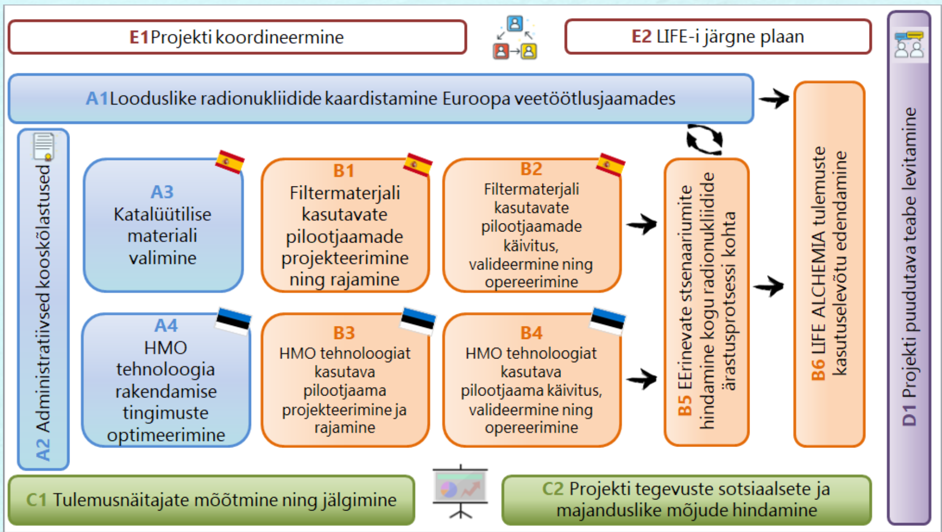
Peamised LIFE ALCHEMIA projekti eesmärgid:

⇒ Demonstreerida tehnilist ja majanduslikku suutlikkust filtermaterialil põhineva veetöötlustehnoloogia rakendamise osas, mis on optimeeritud radionukliidide ärastamiseks ning mille kasutamisel minimeeritakse NORM jääkide/jäätmete teket;

⇒ Toetada LIFE ALCHEMIA projekti raames loodud lahenduste kasutuselevõttu teistes Eurooa riikides ning edendada tehnoloogia rakendamist teistes Euroopa Liidu veetöötlusjaamades;

⇒ Edendada huvitatud osapoolte aktiivset kaasamist Euroopa Nõukogu direktiivi 2013/51/Euratom rakendamisse eesmärgiga minimeerida radionukliidide ärastamisega kaasnevaid keskkonnamõjusid veeteenuste osutamise.

LIFE ALCHEMIA projekti raames teostatud tegevused on välja toodud joonisel 1 . Rohkem informatsiooni tegevustest ning oodatavatest tulemustest leiab projekti koduleheküljel https://www.lifealchemia.eu/



Joonis 1. LIFE ALCHEMIA projekti kuuluvad tegevused.

HMO protsess

LIFE ALCHEMIA eesmärk on demonstreerida tehnilist ja majanduslikku suutlikkust filtermaterialil põhineva veetöötlustehnoloogia osas, mis on optimeeritud radionukliidide ärastamiseks ning mille rakendamisel minimeeritakse NORM jääkide/jäätmete teket. Selle saavutamiseks on rajatud Hispaaniasse kolm pilootjaama ning Eestisse üks. Eesti pilootjaamas on valitud tehnoloogiaks HMO (ingl. k. Hydrous Manganese Oxide) ehk hüdraatmangnaandioksiidiga põhjavee töötlemine. HMO-d saab kasutada raua, mangaani ning raadiumi ärastamiseks põhjaveest.

HMO on tõestanud suhteliselt kõrget absoptsioonivõimet kahevalentsete metallide suhtes, mis on oluline Ra2+ olemasolu korral vees. Lisaks toetab MnO2 Fe ning Mn oksüdatsiooni mittelahustuvasse vormi. Nende protsesside tulemusel tekkinud sadet on võimalik väga hõlpsalt filtreerida.

HMO näol on tegemist lihtsa ning mugava tehnoloogiaga, mida on kerge rakendada, kasutades selleks mõistliku hinnaga kemikaale ning teisi komponente. Eesti pilootjaama käitamine on näidanud, et HMO on efektiivne raua, mangaani ning raadiumi eemaldamisel põhjaveest.

Kahevalentsete metallide suurim adsorptsioon toimub (neutraalses ja aluselises keskkondades) neutraatlsetes ja aluselistes pH tingimustes. Nendes tingimustes muutub MnO2 osakese pind negatiivselt laetuks hüdroksiidioonide OH- liitumise tõttu. Positiivselt laetud raadiumi ioonid on pindlaengu efekti kaudu HMO osakeste pinnal adsorbeeruvad.

Täpsemat metalliioonide adsorptsiooni oksiididele ja hüdrateerunud oksiidile saab kirjeldada järgmise valemi abil:

$$M^{n+} + x[=R\text{-}OH] \leftrightarrow M[=R\text{-}O]_x^{(n-x)+} + xH+ ,$$

kus M – adsorbeeritav metall, nt Ra2+, [=R-OH] ja [=R-O] on oksiidi välispinnaks

Mangaanoksiidi sademeosake toimib mangaani oksüdeerimise katalüsaatorina põhjavees:

$$2Mn(HCO_3)_2 + O_2 + 2H_2O \leftrightarrow 2Mn(OH)_4 + 4CO_2$$

Rauda oksüdeerub ilma katalüsaatori abita.

$$4Fe(HCO_3)_2 + O_2 + 2H_2O \leftrightarrow 4Fe(OH)_3\downarrow + 8CO_2$$

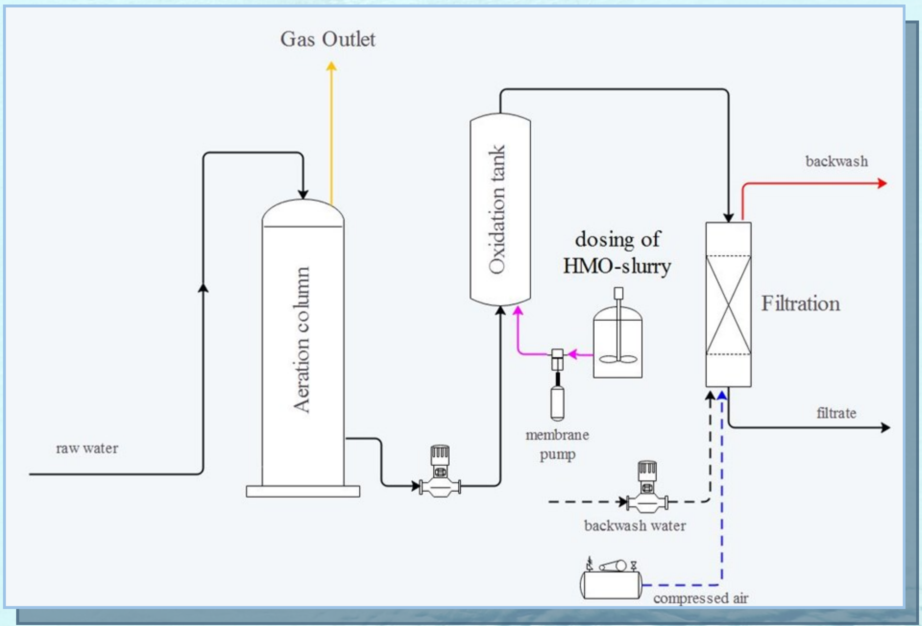
Raadium eemaldatakse adsorptsiooni teel raua ning mangaani sademele ning mangaaandioksiidile. Koossadestamine raua ja mangaaniga vähendab ka vee raadiumisissaldust.

Selle lähenemisviisi rakendamine põhineb eelnevalt valmistatud HMO suspensiooni lisamisel vette, millele järgneb filtreerimine. HMO lahuse valmistamine on lihtne, segades MnSO4 and KMnO4 soolad, kasutades stühhiomeetrilist suhet vastavalt reaktsioonile:

$$2KMnO_4 + 3(MnSO_4{\cdot}H_2O) \rightarrow 5MnO_2{\downarrow} + K_2SO_4 + 2H_2SO_4 + H_2O$$

HMO tehnoloogia koosneb järgmistest sammudest (vt joonis 2):

1) vesi juhitakse läbi aeratsioonikambri (aeratsioonikolonni);
2) vesi suunatakse oksüdeerimisreaktorisse, kuhu lisatakse HMO lahus;
3) saadud sade filtreeritakse filtrikolonnis välja;
4) mille tulemusel toimub HMO osakeste eemaldamine;



Joonis 2. HMO protsessi tehnoloogiline skeem (Eesti pilootjaam).

Seminari ülevaade

Seminar "Looduslike radionukliidide eemaldamine joogiveest ning NORM jääkide/jäätmete käitlusvõimalused" toimus 3.-4. märts 2020 Eestis, Viimsis. Seminari eesmärgiks oli anda ülevaade radionukliidide ärastamisega seonduvatest probleemidest joogivee töötlemisel ning tutvustada LIFE ALCHEMIA projekti tegevusi ning esialgseid tulemusi. Seminari osalejate hulka kuulusid erineva valdkonna inimesed, kes on joogivee radioaktiivsuse probleemiga seotud – veetöötlusjaamade operaatorid, regulaatorid, teadlased, veetöötlusega seotud ettevõtted, jäätmekäitlejad ning rahvusvahelised eksperdid. Seminar andis võimaluse kaasata joogivee radioaktiivsusega seotud aruteludesse erinevaid huvigruppe, tutvustades neile uusimaid uuringutulemusi ning valdkonnas toimunud arenguid. Tänu rahvusvaheliste ekspertide kaasamisele Soome kiirguskaitsekeskusest (STUK) ja Maailma Terviseorganisatsioonist (WHO), oli võimalus tutvuda ka teiste riikide praktikaga joogivee radioaktiivsuse küsimuste käsitluses.

Ülevaade seminari ettekannetest:

- Joogivee radioaktiivsuse hetkeolukord Eestis. Tutvustati olemasolevaid andmeid joogivee radioaktiivsuse ning sellega mõjutatud elanikkonna osas. Lisaks esitleti terviseriski hindamise metoodikat, mille eesmärgiks on hõlbustada veetöötlejate otsustusprotsessi ning käsitleti ka NORM jääkide/jäätmete teket ning tekkinud koguseid veetöötlemisel.

- Tutvustati regulaatori seisukohti joogivee radioaktiivsuse probleemi käsitlemisel.

- Anti ülevaade joogivee radioaktiivsusega seotud probleemi käsitlemisest Soomes. Samuti tutvustati planeeritavaid tegevusi seoses NORM jääkide/jäätmete tekkega Soomes.

- Ettekande tegi professor John Fawell, kes tutvustas WHO lähenemist joogivee radioaktiivsusega seotud probleemide lahendamisel tulenevalt publikatsioonist „Management of Radioactivity in Drinking-Water".

- Joogivee töötlustehnoloogia areng Viimsi Vesi AS veetöötlusjaamas. Tutvustati veetöötlusjaama senist tegevust ning tulevikuplaane.

- HMO tehnoloogia kirjeldus. Tutvustati Viimsi Vesi AS territooriumul asuva pilootjaama tulemusi raua, mangaani ja ammoniaagi ning raadiumi eemaldamisest. Tulemused näitasid, et antud tehnoloogiaga on võimalik edukalt nimetatud elemendid põhjaveest eemaldada, samal ajal vähendades radionukliidide akumuleerumist filtermaterjalis 3-4 korda. See aitab omakorda tulevikus vähendada NORM jääke/jäätmeid teket. Pilootjaamas on saavutatud optimeeritud tasakaal HMO lahuse annuste ning raadiumi eemaldamise vahel.

- Hispaania pilootjaamade tutvustus. Veetöötlusel filtermaterjali kasutavad pilootjaamad on näidanud suuremat tõhusust energia ning veetarbe osas võrreldes seni kasutusel oleva pöördosmoosi tehnoogiaga. Praegu pilootjaamades kasutusel olev tehnoloogia on optimeeritud piisava raadiumi ja uraani ärastuse ningjoogivee kvaliteedinõuete saavutamiseks.

- Tutvustati senist NORM jäätmete käitlemise praktikat Eestis. Tutvustati tavajäätmeprügilasse NORM jäätme ladustamise protessi ning sellega seotud kriteeriume.

- Seminar lõppes kulu-tulu analüüsi tutvustuse ning praktiliste juhtimianalüüsidega. LIFE ALCHEMIA projekti raames välja töötatud tööriista eesmärgiks on aidata veetöötlejaid põhjendatud valiku tegemisel sobiva veetöötlustehnoloogia leidmisel. Juhtimianalüüsid viidi läbi läbi kahe näite abil – väiksema tootlikkusega jaam (1000 m3/öp) ning suurema tootlikkusega (4500 m3/öp). Juhtumianalüüside käigus saadud tagasiside aitab muuta tööriista praktilisemaks ning lihtsamini kasutatavaks.

Seminari käigus toimusid mitmed huvitavad arutelud, mis aitasid tõsta osalejate, s.o erinevate huvigruppide, teadlikkust teema keerukuse osas. Joogiveetöötlejad näitasid üles suurt huvi, esitades küsimusi HMO tehnoloogia kohta, sellega seotud kulude ning vee puhastamise tõhususe kohta. Seminari korraldajad tänavad kõiki ettekandjaid ning osalejaid seminari eduka toimumise eest ning loodavad, et seminaril saadud teave aitab kaasa joogivee radioaktiivsuse seotud probleemide jätkuvale lahendamisele.

Seminaril ette kantud esitlused on kättesaadavad projekti kodulehel:

https://www.lifealchemia.eu/en/life-alchemia-training-seminar-3-4-march-2020-viimsi-estonia-2/

Overview of Radionuclides in Drinking Water in Estonia

Siiri Suursoo, Institute of Physics, University of Tartu, Estonia

International standards and legislative acts define three radiological parameters, which have to be monitored in drinking water. These are given as parametric values [1, 2]:

- tritium (H-3) activity concentration ≤ 100 Bq/L;
- radon (Rn-222) activity concentration ≤ 100 Bq/L;
- indicative dose (ID) ≤ 0.10 mSv/year.

The third parameter – the indicative dose (ID) – is the only one of concern in Estonia. ID is defined as the committed effective dose for one year of ingestion resulting from all the radionuclides (both artificial on natural) whose presence has been detected in a supply of water intended for human consumption, but excluding tritium, potassium-40, radon and short-lived radon decay products [1].

It is important to stress that radiological parameters are given as parametric values, not limit values. A parametric value means that an appropriate level of protection is ensured for the consumer. The risk caused by an exposure equal to 0.10 mSv/year is low enough that it is not expected to give rise to any detectable adverse health effects. Exceeding the parametric values should not be taken as a sign that the water is unsafe to drink. Instead, when the parametric value is exceeded, an assessment should be made, whether the situation poses a risk to human health which requires action.

The parametric values are derived based on the linear-non-threshold model, which assumes that all radiation doses greater than zero will increase the risk of excess cancer. With the best scientific knowledge, we have today, one cannot say that a bit of ionizing radiation is a healthy stimulation to the body.

Approximately 18 % of Estonian inhabitants (230 000 people) consume drinking water where ID exceeds 0.10 mSv/yr [3]. High ID is caused by radium-226 and radium-228 in Cambrian-Vendian (Cm-V) groundwater, which is an important public water supply in North Estonia. Rear findings of ID exceeding 0.10 mSv/yr have also been found in Ordovician-Cambrian aquifer.

Radium in the Cm-V groundwater is a contaminant of natural origin. Cm-V is the deepest aquifer system available for drinking water uptake in Estonian territory and it lays on the uranium and thorium rich crystalline basement rock. The geochemical conditions in the aquifer favour the solubility of radium isotopes, while their mother nuclides – uranium-238 and thorium-232 – stay undissolved. In north-eastern Estonia, the Cm-V aquifer system is divided into two sub-aquifers by a layer of clays – Gdov (the lower) and Voronka (the upper). It has been documented that intensive water uptake in the Gdov sub-aquifer influences the groundwater quality resulting in an increase in radium and chloride concentrations [4]. Therefore, it must be kept in mind that ID in the same well is not a constant parameter – it may change over time.

Overview of Radionuclides in Drinking Water in Estonia

What does the use of radium-rich groundwater mean for a water treatment facility operator? Radium isotopes accumulate in filter material during water treatment processes. This can happen either on purpose, if the facility operator is aiming at radionuclide removal from groundwater, or unintendedly by co-precipitation with iron and manganese. In addition to radium isotopes, which are the only radionuclides of concern in groundwater, radon-222 and thorium-228 become of interest in the water treatment facility. Gaseous radon-222 is generated by the decay of radium-226 in the filter columns. This may significantly increase the indoor radon concentration in the treatment facility. The decay of radium-228 in the filter columns produces thorium-228. As the Estonian exemption level for thorium-228 is an order of magnitude lower than the levels for radium-226 and radium-228, it often becomes the first criteria why filter media needs to be treated as NORM-waste (waste containing naturally occurring radioactive elements).

Why should a water treatment facility operator bother to clean groundwater from radionuclides? Management of NORM waste can create quite some trouble. Yet, not using a treatment process for radionuclide removal does not always guarantee that NORM creation is avoided and the burden of NORM management can be forgotten. If the result of the treatment process is drinking water where ID is compliant with the $\leq$ 0.10 mSv/yr regulation, the excess risk for the consumers is optimised. The risks created by water treatment affect a much smaller part of the population. Risks concerning NORM are also better acknowledged and more controlled.

References:

[1] European Commission Directive 2013/51/Euratom.

[2] Estonian Minister of Social Affairs, Regulation No. 82 of 31 July 2001, as amended on 01.10.2019, "Quality and monitoring standards and methods of analysis for drinking water".

[3] Forte et al., 2010. Journal of Radiation Protection, 30.

[4] Suursoo et al., 2017. Science of the Total Environment, 601-602.

Creation of NORM in filter materials - why, how and what magnitude. Situation in Estonia

Taavi Vaasma, Institute of Physics, University of Tartu, Estonia

Inventory of industries related to the creation of naturally occurring radioactive material in Estonia was created within a nationwide study carried out between 2015 and 2017 [1]. Industries involved in the study were selected based on ANNEX VI of the 2013/59/EURATOM Directive, as well as through information obtained from existing research, reports and publications. This led to the investigation of the following industries:

- Oil shale industry – combustion of oil shale for electricity production; maintenance of large combustion boilers and production of shale oil;
- Cement industry and maintenance of clinker ovens;
- Rare metal processing (production of niobium and tantalum);
- Groundwater treatment plants;
- Central heating stations using solid and gaseous fuels;
- Underground oil shale mines;

Information on elevated concentrations of Ra-226 and Ra-228 in groundwater obtained from Cambrian-Vendian (Cm-V) aquifer was pre-existing through years of research work. A study [2] between 2014 and 2015 investigated the accumulation of these radionuclides in the filter material at drinking water treatment plants (DWTP). 18 DWTPs were studied, covering 47.4% of national Cm-V groundwater production and 49.8% of consumers using Cm-V aquifer whether entirely or partially.

Exemption and clearance levels (set as 1 kBq/kg for U-238 and Th-232 series radionuclides during the study period) were exceeded: a) in 15 DWTPs for Ra-226 with an average value of 7.6 kBq/kg; b) in 16 DWTPs 18 for Ra-228 with an average value of 8.0 kBq/kg; c) 11 DWTPs for Th-228 with an average value of 5.6 kBq(kg.

The total volume of filter material exceeding the exemption levels at that time was estimated to be 300 tonnes. However, there is still no clear international consensus on establishing clearance and exemption values for natural radionuclides, which are not in secular equilibrium. Currently, Estonia has set the clearance and exemption value of 10 kB/kg for Ra-226 and Ra-228, which will change the amount of filter material considered as NORM.

From 2020, NORM (filter material) created in DWTPs can be taken to a landfill for final disposal. At the moment, the only municipal landfill having the permission to accept NORM is in Jõelähtme. Work on creating improved monitoring over the creation and utilization of filter materials (potentially NORM) is ongoing.

References:

[1] Vaasma, T., Kiisk, M., Leier, M., Suursoo, S., Jantsikene, A., Putk, K., 2019. NORM-related industrial activities in Estonia – Establishing national NORM inventory. J. Sustain. Min. 18, 86–93.

[2] Leier, M., Kiisk, M., Suursoo, S., Vaasma, T., Putk, K., 2018. Formation of radioactive waste in Estonian water treatment plants. J. Radiol. Prot. 39, 1–10.

Health risk assessment - why, how and to whom?

Maria Leier, Institute of Physics, University of Tartu, Estonia

One of the parameters regarding drinking water that should be monitored, mainly in northern Estonia, are radiological. The European council directive 2013/51/EURATOM states that parametric value for annual indicative dose is 0.10 mSv. Moreover, it specifies in article 4 that when the value is exceeded, one should assess whether that poses a risk to human health which requires action from radiation protection point of view. One of the principles in radiation protection is optimisation principle which states that public and occupational exposure shall be optimised to keep the magnitude of individual doses as low as reasonably achievable (A.L.A.R.A) taking into account the current state of technical knowledge and economic and societal factors. The present work aimed to answer the following questions – how to assess the risks and how to take into account economic and societal factors?

A study done in 2017 showed that radiological analyses were done in approx. 50% of the waterworks that had regular consumers and from these, in 36 water treatment plants, the annual indicative dose was exceeded with an average value of 0.253 mSv. Using nominal risk coefficients provided by the International Commission on Radiological Protection (ICRP), cancer cases were calculated for four different scenarios, concluding that preventable loss to the society would be one cancer case per year. Knowing that and using the Global Burden of Disease data from WHO and IMPACT study data from the transportation field, the loss to the society in monetary value was calculated.

Knowing the acceptable cost for achieving the parametric value mentioned before, the calculation can be therefore used for a representative group of consumers. Taking into account the results of this work and also assessing the costs for improving the existing technology, implementing new technology or finding entirely different alternatives (e.g. changing the water source), one can find an answer to the question whether the ALARA principle is followed.

Estonian regulations on practices involving naturally-occurring radioactive material (NORM)

Karin Muru, Environmental Board of Republic of Estonia

The Radiation Act lays down the basic safety requirements for the protection of people and the environment against the adverse impact of ionizing radiation as well as regulates radiation practices and activities where natural radiation sources may cause a significant increase of the exposure for workers or members of the public. It includes an indicative list of NORM-related activities in which natural radiation sources may cause annual effective dose to workers or members of the public of more than 1 mSv. The Radiation Act provides requirements for radiation practices and radiation practice licence, and defines exemption and clearance criteria for radioactive sources and practices as well as establishes exemption and clearance levels for radionuclides. When activity concentrations of radionuclides are above exemption levels, the requirements of the Radiation Act should apply. It is then necessary to draw up a radiation safety assessment to assess exemption based on the dose criterion. To use natural resources like water, the groundwater filtration facilities must have an environmental permit for the specific use of water. The requirements for the application and content of the environmental permit are set out in the General Act of the Environmental Code. The Water Act sets the requirements for the quality standards and inspection requirements for drinking water, and the methods of analysis. In the case of drinking water from the Cambrian-Vendian and Ordovician-Cambrian aquifers, the concentration of radium in drinking water must be evaluated and remove radium may require at water filtration facilities. The Environmental Board issue environmental permits and radiation practice licenses, and the Environmental Inspectorate inspects the compliance with requirements of these. The Health Board inspects the quality of drinking water. Estonia has some industries involving NORM, including groundwater filtration facilities. For the latter, the main issues are the accumulation of radionuclides in the filter material and proper management of the filter material in relation with disposal or possible recycling, based on a study conducted by the Institute of Physics at Tartu University in 2014-2015. Since water purification is a continuous process and the concentrations of radionuclides in groundwater can vary from location to location, it is difficult to estimate the rate of contamination with radionuclides in filter material and the time when the filter material becomes a radioactive source in the purpose of the Radiation Act. Therefore, to monitor the practices better, the control measures are going to be established in the environmental permit to assess radioactivity in filter material and requirement for a radiation safety assessment. The primary target group is groundwater filtration facilities using raw water from the Cambrian-Vendian and Ordovician-Cambrian aquifers to produce drinking water. Granting a radiation practice licence is rather a specific case and the practices are assessed on a case-by-case basis. The need for radiation practice licence is assessed mainly in the case with activities related to the removal of filter material from equipment and the storage. When activities are regulated with the radiation practice licence, the further management of removed filter material has to be assessed to dispose of it as a waste or foresee possible recycling. Currently, there is one waste management company who has the right to receive and dispose of the filter material with higher radioactivity than the exemption level. The conditions, including special clearance levels, method of landfilling, environmental sampling, for this activity has set out in the integrated environmental permit and are based on radiation safety assessment. The requirements for transport, including dangerous goods, on-road are regulated with the Road Transport Act and its regulations. Upon carriage of dangerous goods, including radioactive material, the European Agreement on International Carriage of Dangerous Goods by Road (ADR) requirements must be followed.

Drinking water radioactivity and NORM issues in Finland

Niina Leikoski, Radiation and Nuclear Safety Authority (STUK), Finland

There are approximately 1600 water suppliers in Finland, and they vary greatly in production volumes. 60 % of the household water produced by water utilities is groundwater or artificial groundwater. The surveillance of radioactivity in drinking water is done by municipal health protection and STUK. The quality parameters of the drinking water are set in the Decree of the Ministry of Social Affairs and Health relating to the quality and monitoring of water intended for human consumption 1352/2015. Requirements are set for radon-222, total indicative dose and tritium in addition to uranium.

Radioactivity and uranium concentrations mostly meet the requirements in the supplied water. In the groundwater treatment plants, the water is most often groundwater from the soil and the uranium or natural radionuclide concentrations are usually not problematic and not a reason for treatment. If radon needs to be removed, it is often done by aeration.

Most of the exposure to drinking water is caused by radon-222 followed by Pb-210 and Po-210. In Finland, 10 % of the population has a private well. STUK has estimated that 20 000 people use water from a drilled well with radon concentration more than 1000 Bq/l, which is 10 % of people using drilled wells. It was also estimated that 26 000 people are using water from a drilled well which has uranium more than 30 µg/l. No clear health effects have been reported for the people using water from drilled wells, despite the long-term exposure to high uranium concentrations. In private drilled wells the water may be treated and occasionally wastes with radioactivity need to be disposed of. STUK has made a guide for private households on disposal of wastes arising from the treatment of drinking water for its radioactivity.

The groundwater filtration facilities are listed as industrial sector involving naturally occurring radioactive materials (NORM) in the Directive 2013/59/Euratom. If groundwater is treated, NORM may accumulate in the process. The Finnish Radiation legislation was recently renewed. New obligations apply to NORM industries such as groundwater treatment plants. Groundwater treatment facilities are obligated to assess exposure to natural radiation. In addition to this, if the activity concentrations of natural radionuclides in the solid wastes are higher than clearance levels, approval from STUK must be acquired for waste processing.

The accumulation of NORM at groundwater treatment plants has not been characterized in Finland. The activated carbon filters are known to bind natural radionuclides. In other water treatment processes, the accumulation is still unknown. Based on the average level of the naturally occurring radionuclides in Finnish groundwater and typical groundwater treatment plant, very high exposure to other natural radionuclides than radon is not expected for the worker. Radon in the indoor air of some water treatment facilities is known to be high. Typical treatment processes in which NORM can be expected to accumulate are different filtrations. There is an ongoing survey at STUK, which aims to characterize the NORM in Finnish groundwater treatment plants. Based on the results Finnish water suppliers will be informed later this year about NORM, the need for radiation protection and how to fulfil their obligations based on the Radiation act.

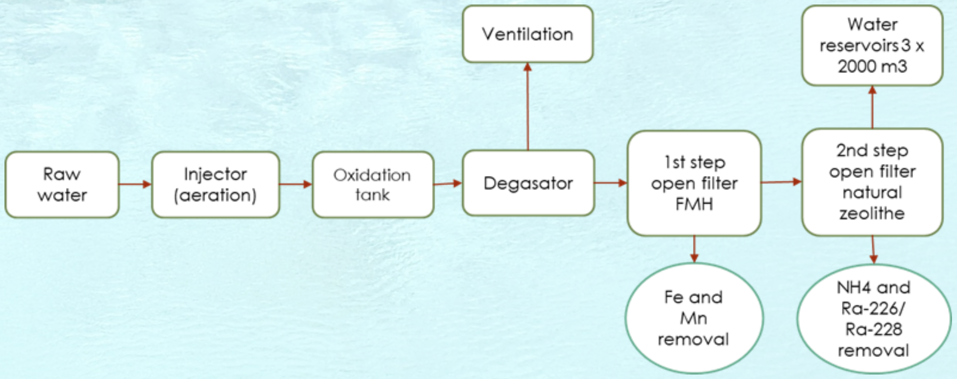
The evolution of drinking water purification technology at Viimsi drinking water treatment plant

Nele Nilb, VIIMSI VESI Ltd., Estonia

Ltd Viimsi Vesi is a water company in Northern Estonia. It has 24 employees and is owned by Viimsi municipality. Viimsi has ca 21 000 inhabitants.

Viimsi Water Treatment Plant started to operate in February 2012 to purify the water to the required standard. Water quality problems were iron, manganese, ammonium and radionuclides. The water is pumped from 80-100 m Cambrian-Vendian wells (11 pieces). The production rate of the water treatment plant is ca 3,300 m^3/d, in summer ca 4200 m^3/d.

Technological scheme of the Viimsi water treatment plant:



Viimsi Vesi was the first water company in Estonia to purposefully remove radionuclides from drinking water. The Cambrian-Vendian aquifer contains natural radionuclides (Ra-228 and Ra-226 isotopes). Based on the results of the analysis of these isotopes, the indicative dose of raw water reaching Viimsi water treatment plant is 0.31 mSv /y (the parametric value is 0.1 mSv/y). Natural zeolite is used to remove radionuclides (cleaning efficiency over 90%).

The main problem of the above-described technology is that the zeolite adsorbs the radionuclides and the filter material becomes radioactive over time (NORM waste). Another problem is that the filter material has an effective life cycle of only a few years.

A few years after the treatment plant was commissioned Viimsi Vesi needed to reconstruct one of the water treatment lines and remove filter material that had already become NORM waste from two filter tanks. At that time, there was no NORM waste management practice in Estonia. Today, Viimsi Vesi has been able to take this NORM waste to the landfill.

In 2020 Viimsi Vesi will probably start the reconstruction of the water treatment plant because the existing filter tanks are corroded and the existing water treatment system produces NORM waste. There is a plan to implement open aeration system and HMO technology as it is the best available technology for radionuclide removal. BUT the question remains, is it even necessary to remove radionuclides from the water and make a major investment?

Other constituents in the drinking water impacting its quality and their removal needs. Overview of the HMO technology and the pilot set up at Viimsi DWTP

Juri Bolobajev, TalTech, Estonia

Besides the presence of radionuclides, groundwater may consist of other inorganic constituents, which lower its overall quality and comprise several challenges to water suppliers during water treatment. In this overview, particular attention will be paid to iron (Fe), manganese (Mn) and ammonium cation (NH_4^+). In Estonia, the threshold limit for Fe, Mn and NH_4^+ in drinking water are 0.2, 0.05, and 0.5 mg/l, respectively.

Despite NH_4^+ is not considered as a toxic substance, this ion is a major constituent of many contaminated aquifers and it is known to negatively influence the quality and usability of groundwater. For example, at certain conditions, the presence of the ammonium cation in raw water may result in drinking-water containing nitrite. The situation is complicated by the fact that there is no conclusive evidence for NH_4^+ -consuming reactions (nitrification or anammox) in the anoxic core of aquifer. As a result, NH_4^+ tends to persist in deep aquifers.

Groundwater is commonly rich in Mn and Fe ions as a result of continuous percolating and movement through the layers of rock and sediments containing these elements. In the absence of dissolved oxygen, both elements tend to exist in the aqueous phase in the form of soluble Fe(II) and Mn(II) forms. In anaerobic groundwater, Fe and Mn could be present at concentrations of up to several mg/l without any discolouration or turbidity of water. In drinking-water supplies, Fe(II) and Mn(II) get oxidized to their insoluble forms as a result of inevitable contact with atmospheric oxygen. Iron is precipitated as insoluble iron(III)hydroxide, which settles out as rust-coloured silt. Staining of laundry and plumbing may occur at concentrations above 0.3 mg of iron per litre. Iron also promotes undesirable bacterial growth ("iron bacteria") within a waterworks and distribution system, resulting in the deposition of a slimy coating on the piping. At concentrations as low as 0.02 mg/l, manganese causes brown or black spots on laundry and plumbing fixtures as well. Concerning the organoleptic properties of drinking water, at concentrations exceeding 0.1 mg/l Mn and Fe impart an undesirable taste to beverages.

Many treatment methods are widely used for the reduction of Fe, Mn and NH_4^+ content in drinking water. Due to the complexity of water chemistry, the applicability of any removal system is site-specific and there can be significant variations in each particular treatment scheme.

The use of different forms of MnO_2 has proven its ability to remove Fe and Mn from water. It is a relatively inexpensive tool, which has been largely used in water treatment processes. MnO_2 is capable of doing two important operations. First, oxidize Fe and Mn into insoluble forms. Second, MnO_2 has relatively high sorption capacity towards divalent metal ions including Ra^{2+}.

Despite the HMO-technology is not intended to deal with NH_4^+ in drinking water, the experiments on the pilot-setup installed at Viimsi DWTP demonstrated, however, high ability in the removal of ammonium cation. NH_4^+ concentration was decreased below the threshold limit. The most likely cause was the development of nitrifying bacteria in aerator and sand filter. The hypothesis on bio-oxidation of ammonium-nitrogen was supported by ion-chromatography analyses, where a remarkable rise of NO_3^- was detected indicating, therefore, the nitrification process.

Abstraktid

Other constituents in the drinking water impacting its quality and their removal needs. Overview of the HMO technology and the pilot set up at Viimsi DWTP

Contemporary MnO_2 based treatment includes the use of catalytic filter materials, i.e. either MnO_2 coated media with a support base or MnO_2 solid mined ores. Another approach of implementing the catalytic oxidation is the injection of preformed hydrous manganese oxide (HMO) slurry into the water and subsequent filtration of resultant suspension. This method has several advantages among other MnO_2 based technologies. Radium-containing particles are accumulated in the upper part of the filter, so they could be easily removed using the backwashing procedure. The backwashing could be accomplished using only water and compressed air, so there is no need to use expensive chemicals for the regeneration of filtration media. Also, HMO slurry could be synthesized on-site using relatively inexpensive chemicals such as $KMnO_4$, $MnSO_4$ and NaOH.

The results obtained from the pilot plant confirmed that HMO- technology may serve as an effective tool to deal with the problems related to the elevated concentrations of Mn, Fe, NH_4^+ and Ra in groundwater.

Results of the HMO pilot plant - radionuclide removal

Siiri Suursoo, Institute of Physics, University of Tartu, Estonia

As a part of the LIFE ALCHEMIA project, a pilot water treatment plant using the HMO (hydrous manganese oxide) process is being tested in Viimsi, Estonia.

The pilot device is situated on the premises of Viimsi Water Ltd. The plant is fed by Cambrian-Vendian groundwater with elevated radium content. Depending on the combination of wells used, the activity concentration of radium-226 ranges from 240–620 mBq/L, while radium-228 activity concentration is between 360–840 mBq/L. The indicative dose of the raw water exceeds the parametric value (0.10 mSv/yr) 2.3 to 5.5 times.

The technological process consists of three stages: first, water is aerated, then the HMO solution is added, and finally, the resulting precipitates are filtered out (see chapter "The hydrous manganese oxide (HMO) process" for more details). Lab-scale experiments of the HMO process demonstrated removal rates above 80% for manganese, iron and radium. According to the results of lab experiments, two schemes were chosen for testing on the pilot plant:

A) Aeration → HMO oxidation → Filtration [gravel–sand–anthracite], periodic dosing (8h per day)

B) Aeration → HMO oxidation → Filtration [gravel–sand–anthracite], continuous dosing (24h per day)

From the viewpoint of radiation protection, three aspects of the treatment process need to be monitored: cleaned water, filter material, and backwash water.

Cleaned water. Despite positive results on lab-scale, periodic dosing was not able to remove enough radium on the pilot plant. The average removal efficiency of radium was only 55%. More promising results were obtained with continuous dosing. Depending on the HMO dose rate, values near 100% may be achieved. Continuous operation of the pilot plant is a key issue in achieving a stable treatment process – radium removal efficiency decreased significantly when the treatment process was temporarily stopped for two months.

Filter material. During the observation period (15 months) radium activity concentrations in anthracite have reached ca. 1000 Bq/kg. Although the technology is not NORM-free, the accumulation rate is significantly lower than with the previous technology used in the Viimsi water treatment plant. Moderate accumulation of radium is observed in the sand.

Backwash water. Some of the radium is removed from the filter material by regular backwash with clean water. However, radium concentrations in the backwash do not get high enough to require specific attention from the perspective of radiation protection.

Even though NORM-free water treatment is difficult to achieve when Cambrian-Vendian groundwater is used, HMO technology is a big step closer to an optimised solution for radionuclide removal from groundwater.

NORM waste Handling on Jõelähtme landfill

Terje Luure, Tallinn Waste Recycling Center Ltd., Estonia

Tallinn Waste Recycling Center is a waste management company owned by the city of Tallinn. It started operation in 2003, at present the company has 86 employees. The main activities of Tallinn Waste Recycling Center are waste transport, waste fuel production, bio-waste composting, bottom ash handling and ageing and landfilling.

Since August 2019, Tallinn Waste Recycling Center is the first and only waste management company in Estonia having a permit for NORM waste handling. The NORM waste is taken to the Jõelähtme landfill situated about 20 km east from Tallinn. It covers 67 hectares and serves more than 500 000 people in western and northern Estonia, including the West-Viru county.

Preparations to receive the permit for NORM waste handling took two years. Preparatory work by Tallinn Waste Recycling Center included the following:

- Radiation safety assessment for the disposal of filter material from the water treatment industry contaminated with radium-226, radium-228 and thorium-228 on Jõelähtme landfill.
- Formulation of NORM waste management procedures on the landfill and purchase of measuring instruments for monitoring the work.
- Training for workers.
- Observation of radium and thorium background in the leachate water of the landfill, monitoring will continue throughout the operation of the landfill.

The first load of NORM waste reached Jõelähtme landfill in January 2020. 40 tonnes of NORM waste from the Viimsi Water Ltd. water treatment plant was put into a pit and covered with mixed municipal waste. The area on the landfill was marked with a radioactivity hazard sign. Doses to workers stayed well below a micro-Sievert as the operation time was very short.

WHO´s Approach to Managing Radioactivity in Drinking water

Prof John Fawell, Cranfield University, Water Science Institute, UK

The WHO Guidelines for Drinking-water Quality are the scientific point of departure for setting national standards. The Guidelines are now based around a Framework for Safe Drinking-water at the heart of which are Water Safety Plans, a proactive, preventive approach to managing drinking-water quality. These are supplied specific and require hazards to be identified from source to tap, risk assessed and managed. Radioactivity is one of the potential hazards covered in chapter 9 of the Guidelines and supported by a stand-alone document that is presented in plain language for non-experts in a question and answer format. The Guidelines cover non-emergency situations. (https://www.who.int/water_sanitation_health/publications/management-of-radioactivity-in-drinking-water/en/)

The guidelines recommend screening values for gross alpha (0.5 Bq/L) and gross beta (0.1 Bq/L) activity based on a conservative IDC (Individual Dose Criterion) of 0.1 mSv from one-year consumption of 2 Ls drinking-water per day. The Guidelines apply to natural radionuclides of the thorium and radium decay series; they do not cover radon which is considered separately as a gas. They also apply to human-made radionuclides such as caesium-134 and 137, strontium-90, iodine-131, tritium and carbon-14. However, levels in drinking-water are generally very low. For screening supplies, it is useful to have samples at different periods (seasons) to establish variability.

Should a screening value be exceeded, the first step is to repeat the analysis. If it is still exceeded, then a detailed analysis is required to identify the actual radionuclide/s responsibly. The levels can then be compared to the individual Guidance levels for each radionuclide. Where more than one radionuclide is involved then the sum of the measured activity concentration above the respective limit of detection of each radionuclide divided by the guidance level for that radionuclide, calculated using the default assumptions (adult, 2 L/day) should not exceed 1. If that is the case then options for reducing exposure should be considered but it should always be remembered that an adequate supply of safe drinking water is vital and one hazard should not be exchanged for a more serious one, such as waterborne microbial pathogens. Neither the screening values nor the IDC should be interpreted as a limit above which drinking-water is unsafe for consumption.

Screening is not useful for some elements, e.g. lead-210 and radium-228 due to low energy of their beta particles. This is rare and requires individual analysis, where indicated.

If there is a need to manage radioactivity in the supply the first step would be to consider the possibility of alternative supply if a safe supply was available. The second step is to consider whether blending with a low activity supply is possible, again making sure that the supply is safe. The third step is to consider installing or modifying treatment. Some treatment trains applied to surface waters are effective at removing suspended radionuclides and adsorptive treatments may be used for groundwater. However, proper consideration must be given to safe disposal of waste streams, which consist of concentrated radioactive material compared to the source water.

In emergencies, the IAEA Safety Standards Series should be consulted.

Overview of natural radioactivity issues in drinking water in Spain. Preliminary results obtained from the LIFE ALCHEMIA project

I.M. Rodríguez Ruano, Provincial Government of Almeria, European Initiatives and Economic Development Section, Spain

J.L. Casas López, Solar Energy Research Centre (CIESOL), University of Almeria; Spain

At the beginning of the 20th century, there was a total lack of knowledge about the parameters that could be considered beneficial or not in the quality of the drinking-water destined for the Spanish population; in fact, the term radioactivity on the labels of bottled water was synonymous with "miracle cure for everything". The increase in health protection in general, and in particular of water for human consumption, has allowed a constant evolution through European and Spanish legislation, which has led us to achieve a precautionary principle to ensure a high level of protection in the health of the population.

The consolidated text in force that transposes the Directive 2013/51/Euratom into the Spanish legislation is the Royal Decree 140/2003, which establishes the health criteria for the drinking-water quality. However, natural radioactivity is not being systematically analysed at the national level and gross alpha activity is one of the parameters with the lowest percentage of compliance. The magnitude of the problem can only be analysed in small areas of the Spanish geography since only 19% of 10.160 supply areas registered in the Spanish National Drinking Water Information System (SINAC) reported data on gross alpha activity in 2018, being Almería one of the pioneer provinces in the control of natural radioactivity.

Since 2003, it has been installed 15 drinking water treatment plants based on reverse osmosis to eliminate natural radioactivity from groundwater intended for human consumption in Almeria. However, these treatments have a high water and energy footprint associated, generating high volumes of reject water and complex management of their consumables.

The LIFE ALCHEMIA UE project (LIFE16 ENV/ES/000437) aims to eliminate the natural radioactivity present in groundwater destined for human consumption through bed filtration systems with different granular materials, ensuring compliance with the parametric values established in the legislation in force (Royal Decree 140/2003). Furthermore, to minimise the generation of NORM wastes (Naturally Occurring Radioactive Materials) associated with this type of installations that require non-conventional management (Directive 2013/59/EURATOM, pending transposition to the Spanish legal system).

In the province of Almeria, three pilot plants have been designed and built with a treatment capacity of 10.8 m^3/h each, installed in series between the water catchment wells and the reverse osmosis plants in service. The versatility of the designed pilot plants allows to change the order of filtration between their tanks, dosage of reagents and to carry out a water and energy analysis. This will allow making a comparison between the pilot plants under study and the current treatment systems.

After several months of operation, the first analytical results conclude that it has been possible to reduce the total indicative dose by over 65%, achieving percentages of elimination of the radioisotopes Ra-226 and Ra-228 of 98% and 86%, and of 42% and 40% in the case of U-234 and U-238. The current reverse osmosis treatment systems have rejection volumes that range from 36-42%, compared to 9-12% obtained with the pilot plants of the LIFE ALCHEMIA project, that represent a water consumption mean reduction of 80%. In the same way, the energy consumption has been reduced by 87%.

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Introduction to Cost Analysis tool for DWTP operators

Maria Leier, Institute of Physics, University of Tartu; Estonia

To follow the ALARA principle in the field of drinking water treatment, one must also consider technical knowledge and economic factors. The aim of the Cost Analysis (CA) tool is to provide a helpful step to make a reasonable choice between technology in use and technology developed during LIFE ALCHEMIA. Reasonable is here considered as an appropriate decision to lower costs and reduce NORM. The tool was developed taking into account that it can be used independently and only economic aspects are considered, therefore, in the final phase, social and other related aspects should also be thought of. CA tool parameters are divided into two – local conditions of the WTP and also selected criteria which are required during LIFE ALCHEMIA project but also from the initial feedback from the operators. Parameters and costs are described with a unit production cost in €/m^3. This is calculated for implementation costs and operating costs such as one-time implementation costs, filter material, backwash and chemical costs.

Lisa 2. Seminari kava

Day 1 (3rd of March)

08.30 – 09.00	Arrival and registration
09.00 – 09.15	Introduction to the seminar (*Taavi Vaasma, UT*) and overview of the LIFE ALCHEMIA project (Marta Gomez, CARTIF)
09.15 – 09.45	Overview of radionuclides in drinking water in Estonia (*Siiri Suursoo, UT*)
09.45 – 10.15	Creation of NORM in the filter materials – why, how and what magnitude. The situation in Estonia. (*Taavi Vaasma, UT*)
10.15 – 10.45	Health risk assessment – why, how and to whom? (*Maria Leier, UT*)
10.45 – 11.15	Coffee break
11.15 – 11.45	Management of NORM – monitoring, notification, responsibilities and regulatory aspects. (*Karin Muru, The Environmental Board, radiation department*)
11.45 – 12.15	Drinking water radioactivity and NORM issues in Finland (*Niina Leikoski, STUK*)
12.15 – 13.30	— LUNCH —
13.30 – 14.00	Results obtained on the HMO pilot at Viimsi DWTP: purification efficiency; reduction in NORM generation etc. (*Siiri Suursoo, UT*)
14.00 – 14.30	Other constituents in the drinking water impacting its quality and their removal needs. Overview of the HMO technology and the pilot set up at Viimsi DWTP. (*Juri Bolobajev, Taltech*)
14.30 – 15.00	The practice and requirements of accepting NORM to Jõelähtme landfill (*Terje Luure, TJT*)

— END OF FIRST DAY —

Day 2 (4th of March)

08.45 – 09.00	Arrival and registration
09.00 – 09.30	WHO guidelines based on the document "Management of radioactivity in drinking water" (*John Fawell, technical expert, independent consultant for WHO*)
09.30 – 10:00	Overview of natural radioactivity issues in drinking water in Spain. Preliminary results obtained from the LIFE Alchemia project. (*Isabel Rodríguez, Provincial Government of Almeria & Jose Luis Casas, CIESOL*).
10.00 – 10:30	Coffee break
10.30 – 11.00	Introduction to Cost Analysis tool for DWTP operators*. (*Maria Leier, UT*) Practical exercise part 1 – evaluating implementation and operational costs during transitioning from one purification technology to another; a case study. Practical exercise part 2 – implementation of the Cost Analysis tool during different scenarios, case studies **For more active participation in the practical exercise, please bring your laptop.*
11.00 – 11.15	Concluding the seminar.

— END OF THE SEMINAR —

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