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Feasibility of a HMO-process in drinking water treatment technology for removing natural radioactivity and avoiding generation of NORM

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INTRODUCTION

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- *Ca*. 18 % of Estonian inhabitants (230 000 peoole) consume drinking water where **indcative dose (ID) exceeds 0.10 mSv/yr *** (Forte *et al*., 2010)

  * Parametric value given by Directive 2013/51/Euratom.

- High ID caused by **^{226}Ra** and **^{228}Ra** in Cambrian-Vendian groundwater.
- Groundwater treatment by filtration results in **NORM accumulation**.
- **More than 300 tonnes** of filter media with ^{226}Ra, ^{228}Ra, ^{228}Th ≥ 1 kBq/kg

  (Leier *et al*., 2018)

  Average activity concentrations ^{226}Ra – 7.6 kBq/kg,
  ^{228}Ra – 8.0 kBq/kg
  ^{228}Th – 5.6 kBq/kg

- No national NORM management strategy.
- Potential for uncontrolled NORM waste disposal.

AIMS AND OBJECTIVES

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- Ideal solution: a NORM-free water treatment technology.
- LIFE Alchemia „**Towards a smart & integral treatment of natural radioactivity in water provision services**“ (2017-2020) https://www.lifealchemia.eu/en/
  - Objective: to demonstrate the technical and economic feasibility water treatment technologies to remove radioactivity from water and to minimize the generation of NORM.
  - 3 pilot plants in Spain and 1 pilot plant in Estonia.
  - Technology chosen in Estonia – **the HMO process**.

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THE HYDROUS MANGANESE OXIDES PROCESS

- For Fe, Mn, and Ra removal
- 3-stage process: aeration → HMO oxidation → filtration
- **HMO suspension**:
  $2KMnO_4 + 3(MnSO_4 \cdot H_2O) \rightarrow 5MnO_2\downarrow + K_2SO_4 + 2H_2SO_4 + H_2O$
- **Removal mechanisms:**
  - $4Fe(HCO_3)_2 + O_2 + 2H_2O \leftrightarrow 4Fe(OH)_3\downarrow + 8CO_2$
  - $2Mn(HCO_3)_2 + O_2 + 2H_2O \leftrightarrow 2Mn(OH)_4 + 4CO_2$
  - Ra adsorption to and co-precipitation with Fe and Mn particulates and MnO_2

THE HYDROUS MANGANESE OXIDES PROCESS

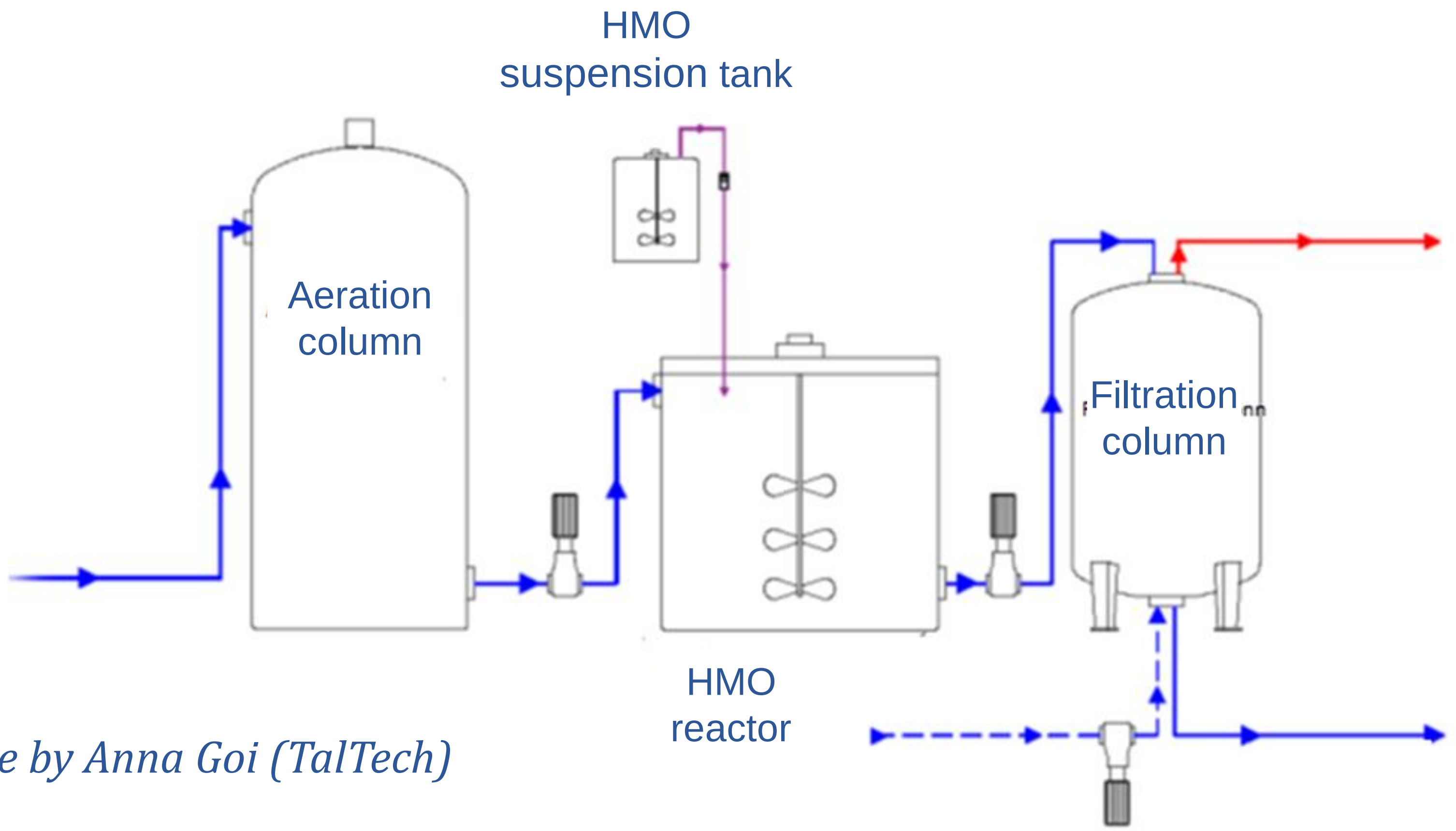


Figure by Anna Goi (TalTech)

Best removal rates in **lab-scale experiments**:

- Mn – 81%
- Fe – 90%
- Ra – 86%

Schemes tested on the pilot plant:

a) Aeration → HMO oxidation → Filtration [gravel–sand–anthracite], periodic dosing

b) Aeration → HMO oxidation → Filtration [gravel–sand–anthracite–HMO precipitates], periodic dosing

c) Aeration → HMO oxidation → Filtration [gravel–sand–anthracite], continuos dosing

THE PILOT PLANT

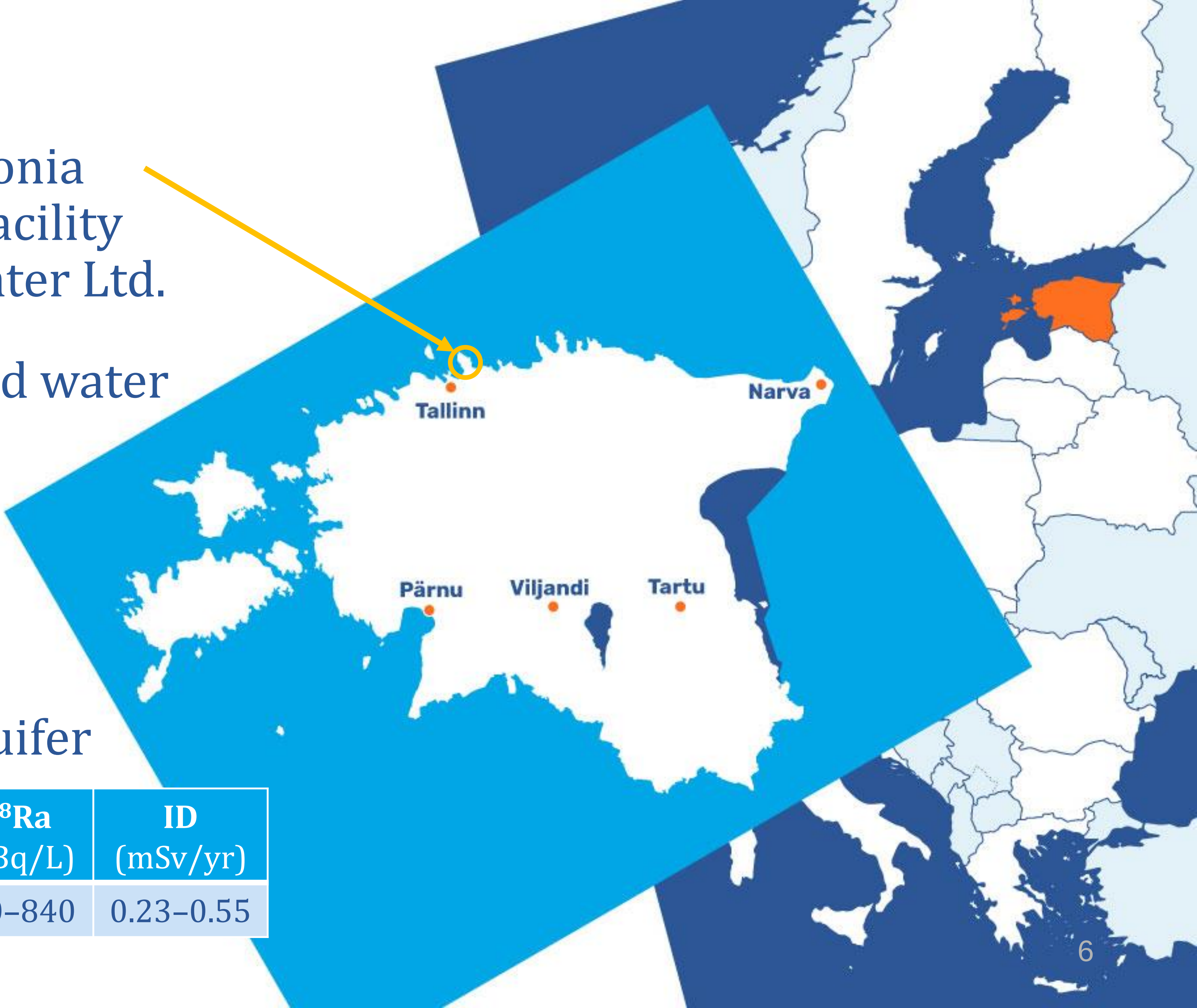
- Situated in Viimsi, Estonia in a water treatment facility operated by Viimsi Water Ltd.

- Target values in treated water

Fe (µg/L)	Mn (µg/L)	ID (mSv/yr)
≤ 200	≤ 50	≤ 0.10

- Raw water from Cambrian-Vendian aquifer

Fe (µg/L)	Mn (µg/L)	^{226}Ra (mBq/L)	^{228}Ra (mBq/L)	ID (mSv/yr)
40–420	50–230	240–620	360–840	0.23–0.55



RESULTS – WATER

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Removal efficiencies (%) for Fe, Mn, and Ra isotopes

Green – complient with drinking water quality parameters.

Red – target value exceeded.

	Scheme	*Fe*	*Mn*	*^{228}Ra*	*U(^{228}Ra), k=2*
a)	Aeration → HMO → Filtration [gravel–sand –anthracite], periodic dosing	79	79	49	10
		73	89	37	14
		85	83	63	7
		88	90	64	10
b)	Aeration → HMO → Filtration [gravel–sand –anthracite–HMO precipitates], periodic dosing	83	47	45	13
		88	66	71	9
		52	95	64	11
c)	Aeration → HMO → Filtration [gravel–sand –anthracite], continuos dosing	**92**	**92**	**79**	**4**
		87	**87**	**74**	**7**

RESULTS – BACKWASH WATER

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Activity concentration of ^{226}Ra and ^{228}Ra in filter backwash water

	Scheme	*^{226}Ra (Bq/L)*	*$U(^{226}Ra)$, k=2 (Bq/L)*	*^{228}Ra (Bq/L)*	*$U(^{228}Ra)$, k=2 (Bq/L)*
a)	Aeration → HMO → Filtration [gravel–sand –anthracite], periodic dosing	**5.48**	**0.14**	**7.89**	**0.33**
		5.87	**0.19**	**8.08**	**0.38**
		3.12	**0.10**	**4.23**	**0.21**
b)	Aeration → HMO → Filtration [gravel–sand –anthracite–HMO precipitates], periodic dosing	3.24	0.11	4.32	1.65
		3.83	0.13	6.23	0.31
		2.43	0.08	3.23	0.16

RESULTS – FILTER MATERIAL

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Accumulation rate of ^{226}Ra, ^{228}Ra, and ^{228}Th, in filter material

	Filter material	*^{226}Ra (Bq/kg yr^{-1})*	*^{228}Ra (Bq/kg yr^{-1})*	*^{228}Th (Bq/kg yr^{-1})*
Pilot plant	Sand	160	220	28
	Anthracite	1300	1500	130
Current technology in Viimsi Water Ltd. (Hill *et al.*, 2018)	*Filtersorb®FMH*	*1400*	*1860*	
	Zeolite	*3400*	*4760*	

TO BE CONTINUED...

Experiments will be continued:

- Continuos dosing
- Scheme with 2-stage filtration

  Aeration → HMO oxidation →
  → Filtration I [gravel–sand–anthracite] →
  → Filtration II [gravel–zeolite]

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Thank you!

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