

Surveying programme of the NORM situation of the Hungarian waterworks

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Background

- Proposal for the new EU Basic Safety Standards
- Natural radioactivity (NORM/TENORM) is to be more emphasized on the regulation of radiation protection
- To create better regulation
- Knowledge of the domestic NORM situation
- a lot of surveying of NORM/TENORM

Regulation of TENORM 1

The decree 47/2003 stipulates the following issues:

- operator has to notify about its work activity to the public health authority according to designated activities
- licensing is needed for emplacement (storage), treatment, reuse of residues
- operators are obliged to define the main radiation characteristics of the stored TENORM's and to assess the radiation burden
- in the case of new storage construction, reuse of facilities and materials, and termination of emplacement, radiological assessment has to be performed, and to be attached to the application of licence
- on the basis of the assessment the authority prescribes the quantity limit for storage and the date of termination of emplacement
- if the radiation burden to the population exceeds the authorized limit, then the utilization of the given area has to be restricted

Regulation of TENORM 2

- concerning the occurrence of TENORM the next designated activities are in Hungary:
 - uranium mining and milling
 - coal mining, coal fired power plants
 - bauxite mining, alumina industry
 - metal ore mining, metallurgy
 - exploitation of oil, gas
 - fertilizer industry, usage of phosphate ore
 - usage of geothermal energy
 - usage of zircon sand, ceramics factory
 - rare earth ore mining, milling

Waterworks were out of these designated activities

Purposes

- In Hungary, the evaluation of TENORM situation has not been performed in a full scale. The main goal is that the Hungarian regulations should meet with the EU Directive No. 96/29 (and the new EU BSS) in this field.
- A surveying project has been launched in order to collect all relevant information about the Hungarian TENORM situation. This programme covers a lot of data collection (work activities, disposal places, residue quantities) and in situ and laboratory radiological measurements. Regard with the size of the project, each task is being performed with stepwise approach.
- Nevertheless, the Hungarian situation of TENORM definitely differs from that of other countries in the aspect of occurrence forms of natural sources (or in the imported raw materials), in the quantities of exploitation, in the level of the radioactivity and in the applied technological processes.

Tasks of surveying programme

- Firstly, those work activities were chosen where huge amount of residue was produced.
- The other criteria was that the activity concentration in the majority of the given residues can be much higher than the average activity concentration of the typical Hungarian soil.
- After filtering and ranking, the following main activities were left: uranium mining and milling, coal mining, coal firing in power plants, bauxite mining and alumina production.
- Because the examined activities covered a very broad range of the amount of residues and a variety of applied technologies, the surveying programme was focused on the main processes of a given activity.
- For example at tailings ponds of the coal fired power plant, firstly, in situ dose rate measurements were carried out on the area and then a characteristic mixed sample was taken according to the distribution of dose rates.

Quantities of uranium mining residues

Ore	Quantity of ore (t)	Extracted U (kg)	U content of residues (kg)
Waste rock	19,303,515		1,029,715
Heaps for leaching	7,213,000	545,594	397,182
Acid exploitation	18,886,477	17,534,987	1,384,534
Sum	45,402,992	18,080,581	2,808,190

Radiation levels of uranium mining residues

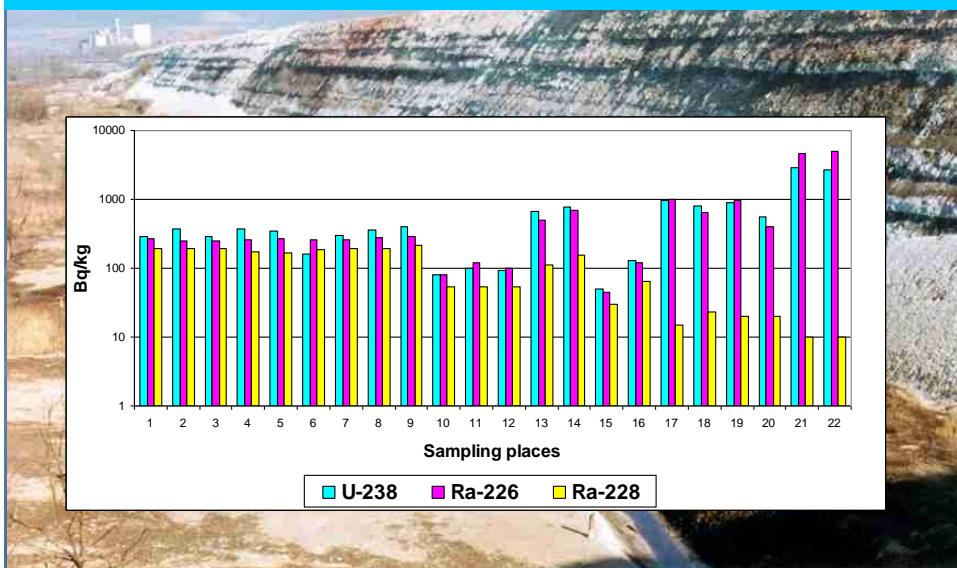
Target of measurements	Unit	Values at		
		waste rock	heaps leaching	tailings ponds
Uranium	g t ⁻¹	20-60	55-75	60-70
Ra-226	Bq g ⁻¹	0.3-1.6	0.8-2.0	9.0-14.0
Rn activity concentration (open air)	Bq m ⁻³	5-40	5-370	40-1100
Dose rate	μGy h ⁻¹	0.4-1.2-	0.4-1.5	1.2-10.0

Remediation was terminated at the end of 2008

Coal mining, coal fired power plant 1

- Mining activities were finished, but residues were remained
- In the mountain Mecsek and Vértes, the radioactivity level of the coals mined on these areas was 10-50 times higher than the worldwide average.
- The coal fired power plants have piled up in the order of magnitude of 10 million tons of fly ash, bottom ash and slag in ponds around the plants.
- The radioactivity of U-238 series of ash and slag discharged from power plant Ajka is in the range from 200 to 2000 Bq·kg⁻¹, and the radionuclides of Th-232 series varies between 20 and 40 Bq·kg⁻¹.
- The radioactivity level of U-238 and Th-232 series of ash and slag discharged from power plant Pécs is in the range of 200-300 Bq·kg⁻¹, and K-40 can be found in the ash and slag with the typical value about of 800 Bq·kg⁻¹.

Activity concentration of piles and ponds of coal mining, coal fired power plant



Bauxite mining, alumina production 2

- **The radionuclide concentrations of bauxite ores range from 200 to 300 Bq·kg⁻¹.**
- **At the refining factories, a lot of red mud has been produced with the activity concentration of 200-400 Bq·kg⁻¹.**
- **The production of alumina of 1 ton has resulted in the amount of 2.2-2.6 tons of red mud.**
- **Until now, the total quantity of red mud originated from the refining factories ranges from 35 to 40 million tons.**

Other TENORM activities

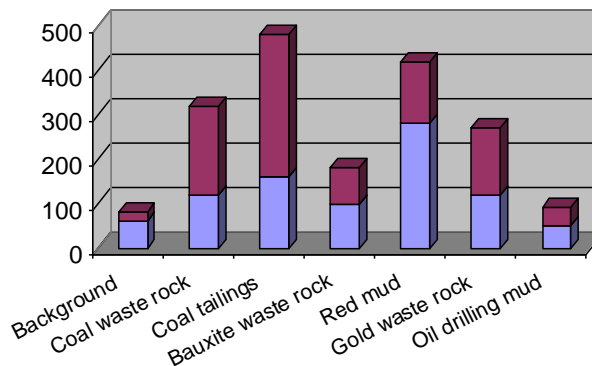
- In the iron and steel production, the total amount of slag produced during many decades was in the order of 100 million tons. The iron ores have mainly been imported, the domestic iron ore mining from mine Rudabánya was minor. The radioactivity of feedstock materials and by-products of iron/steel fabrication have been examined.
- At the oil and gas production and exploration, the drilling mud and deposition of wells were studied.
- Radioactivity of the waste rocks around copper, lead, zinc, manganese, historical gold, silver mines, and of the ores and the discharged pit water were measured.
- Both abandoned and existing waste rock piles were examined in order to get more information about the TENORM situation.
- The fertilizer and ceramic production were examined, where both the raw materials and final products were measured.

Radiological evaluation of residues

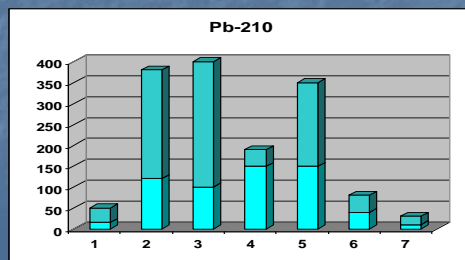
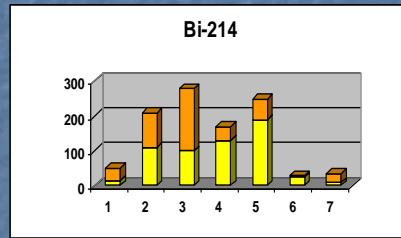
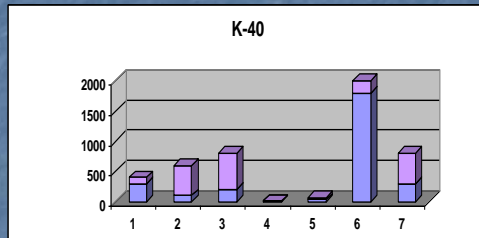
Radiological evaluation comprises the followings:

- re-evaluation of the amount of residues,
- measurements of radioactivity of residues,
- sampling and measurement of radioactivity of water and vegetation,
- gamma dose rate and radon exhalation measurements,
- modelling of transportation of dominant radionuclides,
- model experiments,
- establishment of radiological criteria system,
- proposal for re-use and remedial action of residues.

Typical dose rate range (nSv/h)

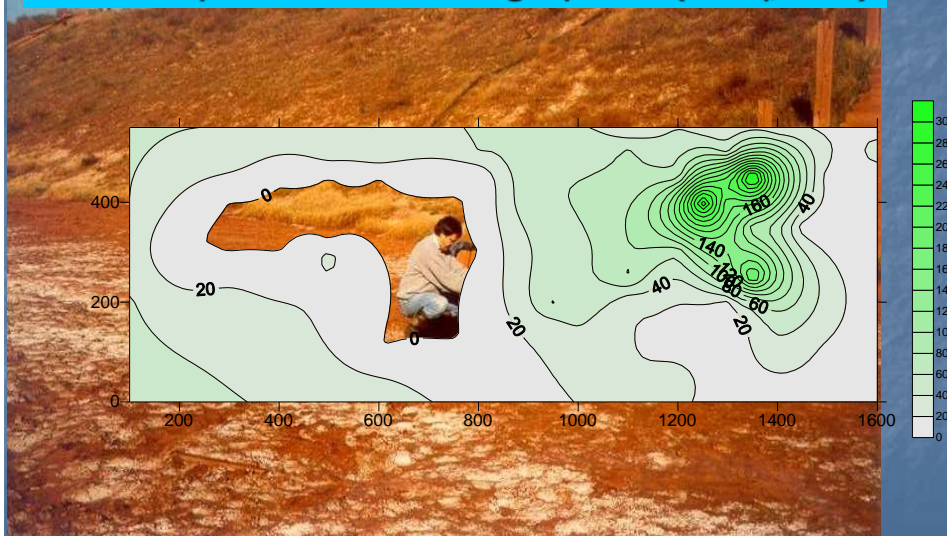


Activity concentration of TENORM samples (Bq/kg)



- 1 Background**
- 2 Coal waste rock**
- 3 Coal tailings**
- 4 Bauxite waste rock**
- 5 Red mud**
- 6 Gold waste rock**
- 7 Oil drilling mud**

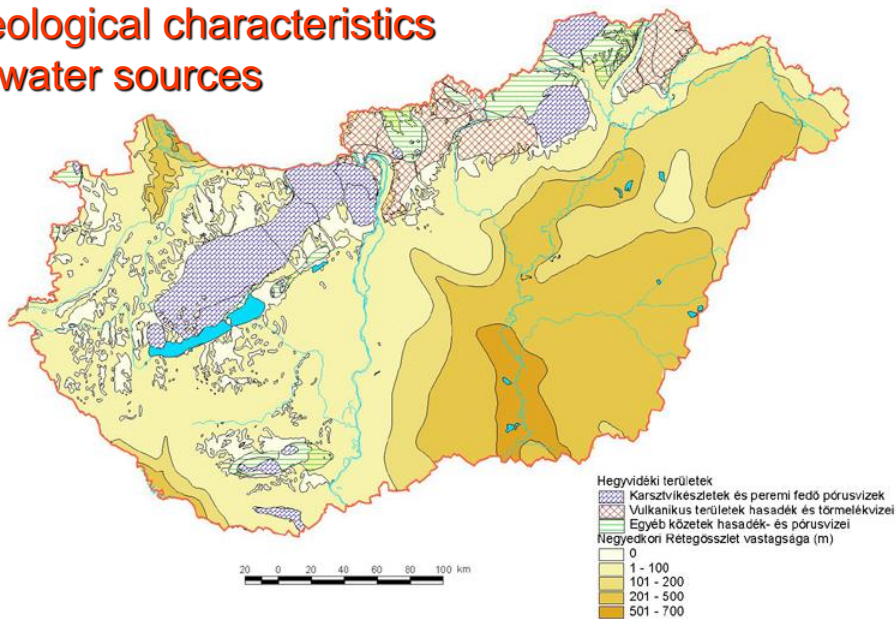
Measurements of radon exhalation at Neszmély red mud tailings pond (mBq/m²s)



Water sources

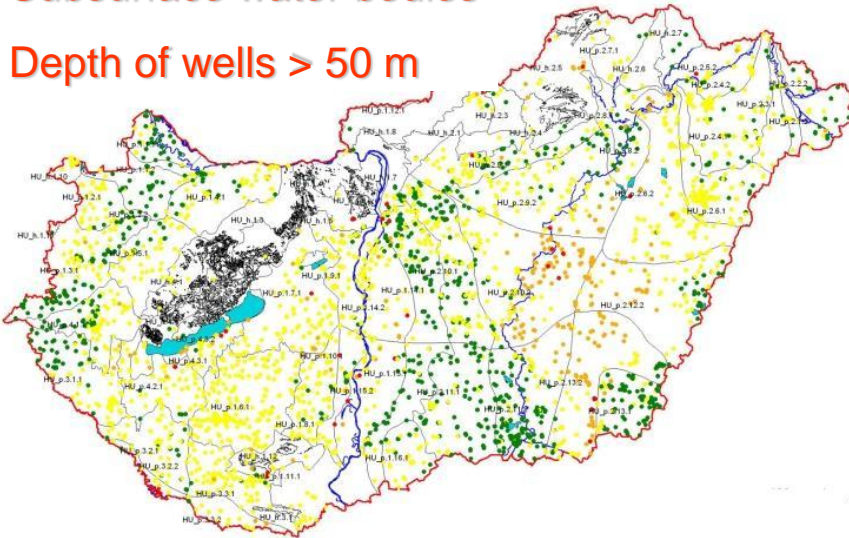
- Drinking water can be derived from
 - deep aquifer
 - surface water bodies
 - karstic water
 - groundwater
- In Hungary about 4000 pieces of wells and 120 waterworks have been established.

Geological characteristics of water sources



Subsurface water bodies

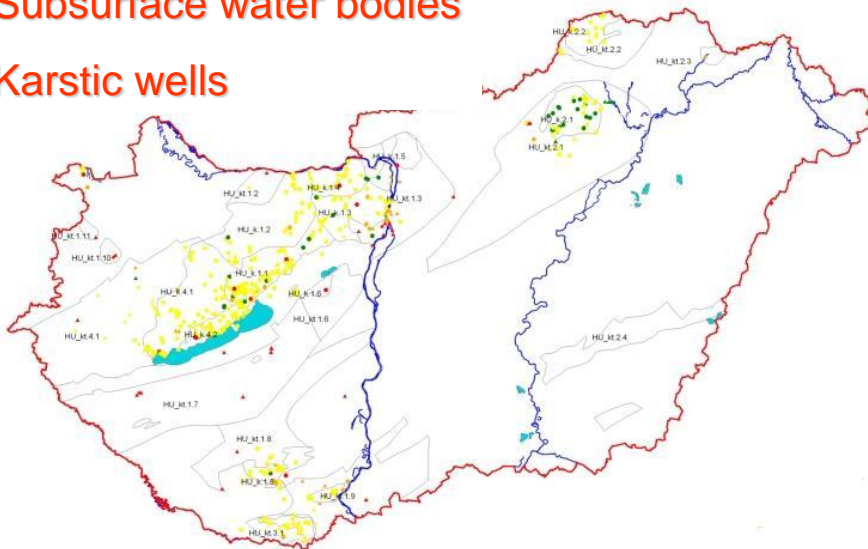
Depth of wells > 50 m



Ministry for Environment and Water

Subsurface water bodies

Karstic wells

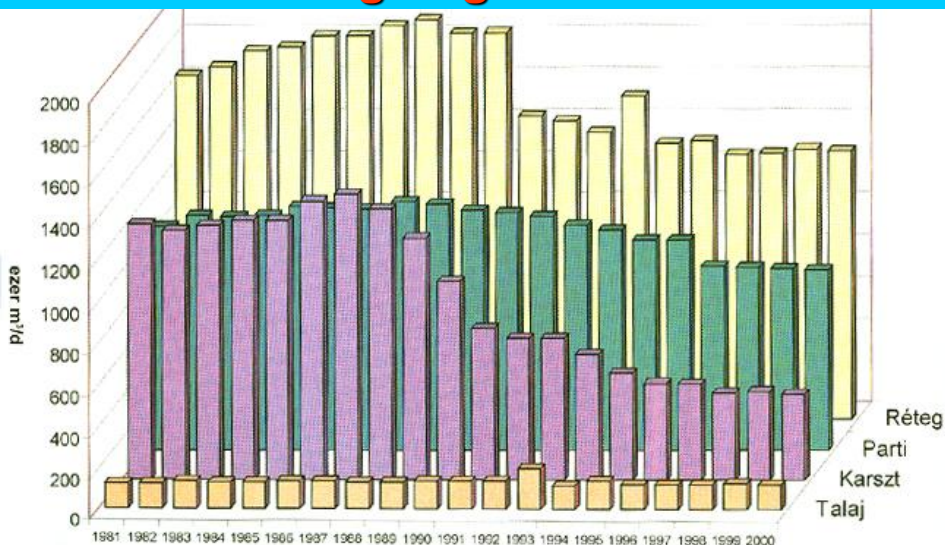


Ministry for Environment and Water

Distribution of water consumption

- The groundwater is less supplied for drinking, because pollution can often take place in this, so the water supplying from groundwater is below 5 %
- The karstic water for drinking water is rated in about 10% of the total supplied water.
- The rate of near surface water supplied for drinking is about 25 % of the total water consumption, and this water mainly arises from the bank parts of the rivers after natural filtering.
- In Hungary, more than 60% of the drinking waters are supplied from the deep aquifer through drilled wells. The typical depth of these wells is in the range of 50 m to 250 m.

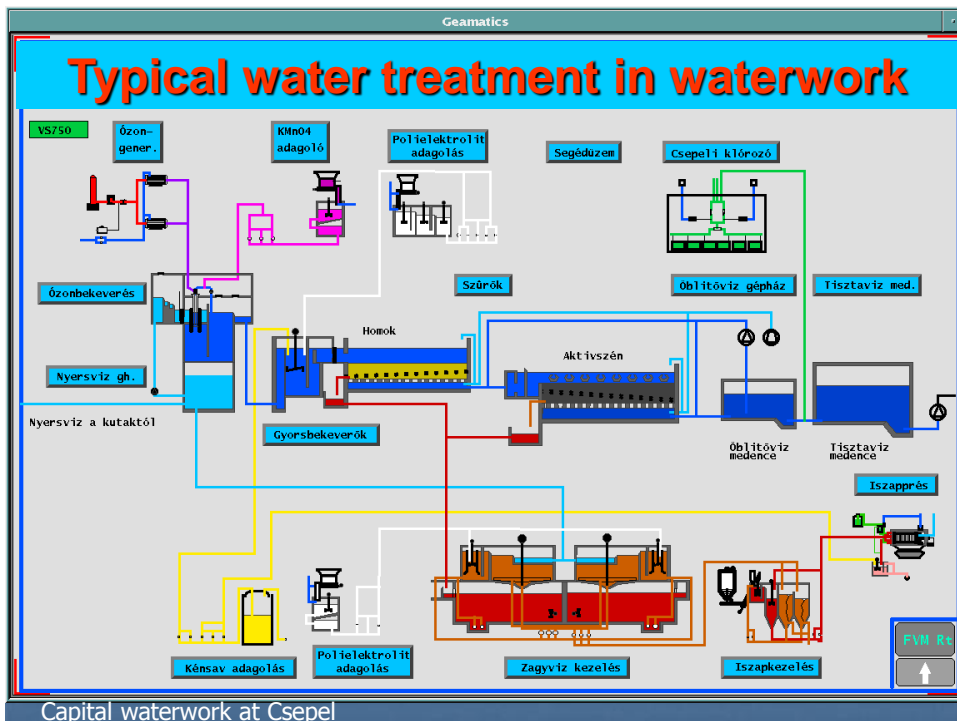
Distribution of water consumption concerning origin of raw water



Ministry for Environment and Water

Water treatment

- The water by the aid of the complex hydrogeology transport processes reaches the different source points, and owing to these transport processes the content of solved elements in the water is very altering at each source well.
- Close to the whole part of the raw waters obtained from different sources (wells) is to be purified and treated. This treatment is firstly performed to remove the pollutants, like manganese, iron, and arsenic. The treatment includes several filtration processes applying different filters (charcoal, gravel bed) and finally more or less sludge and used filter material arise



Waterwork with gravel bed filters



Residues of water treatment

- The filtration process for the pollutants takes out also the great parts of radioactive elements containing the raw water, therefore the sludge and used filter materials comprise different radionuclides, too. So these residues of waterworks can be regarded as a NORM.
- Different amount of water is treated and supplied by the different waterworks, so the amount of the residues is very diverse. The range of the volume of the residues can be from 1 m³ to 100 m³ in a year. The operators of the waterworks emplace firstly the residues in a landfill, but many operators reuse these residues, such as gravel bed for backfilling, if it is possible concerning chemical pollutants,.

Surveying of waterworks

- In the survey program, firstly, the waterworks supplying water from drilled wells were focused on, and secondly few waterworks using surface water were involved in the survey.
- The examination of karstic water and groundwater is to be planned in the next step.
- In general, the waterworks are operated by private firms, and the operators have a little dislike to any radioactive examination, but finally, firms from the planned representative areas could be found for the cooperation with the surveying programme.
- Firstly, 25 waterworks represented from different parts of the country were chosen for the survey programme



Sampling

- The content of radioactive elements depends on the NORM content of the given pathways (strata, layers) through which the water has infiltrated, so the occurring radionuclides and their activity in the water can change from well to well
- At 25 waterworks represented from different parts of the country, raw water, used filter materials and sludge samples were taken.



Sampling from sludge collector pool



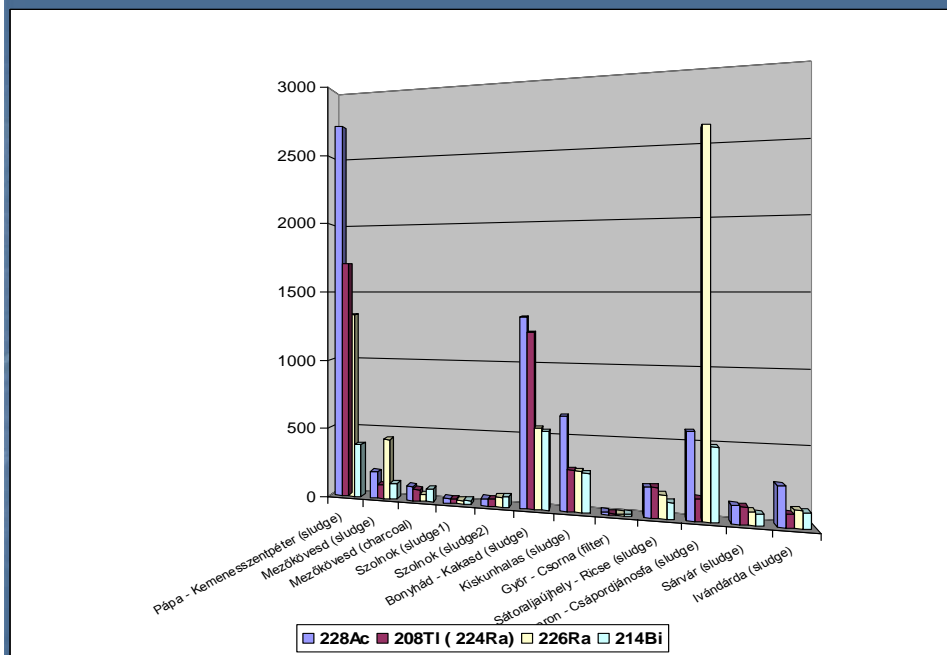
Measurements

- The activity of samples (sludge, filter) was measured by using gamma-ray spectrometry
- The radionuclides of ^{226}Ra and ^{228}Ra in the raw waters were separately analyzed, too.

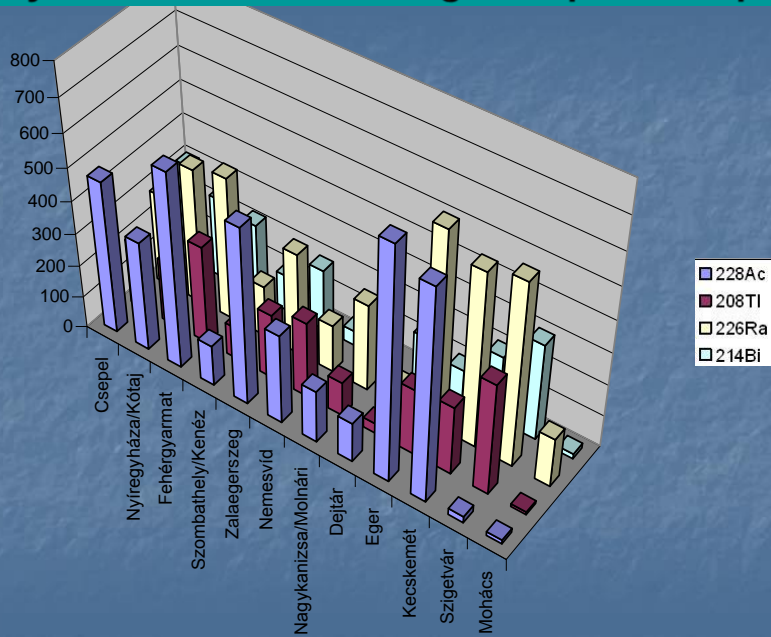
Activity concentration of the representative samples in Bq/kg

Place (sample)	Th-series		U-series		
	Ac	Tl (Ra)	Ra	Bi	Pb
Pápa - Kemenesszentpéter (sludge)	2725±46	1719±31	1336±51	369±10	374±7
Mezőkövesd (sludge)	189±4	95,8±2,8	430±16	113±3	117±3
Mezőkövesd (charcoal)	100±3	80,4±2,9	46,2±6,2	90,6±2,3	92,3±1,9
Szolnok (sludge 1)	32,5±1,3	31,2±1,4	25,4±2,9	28,8±1	29,8±0,7
Szolnok (sludge 2)	49,3±1,8	51,3±2,6	68±14	74,9±2,6	62,8±3,1
Bonyhád - Kakasd (sludge)	1328±22	1224±35	562±49	543±17	560±12
Kiskunhalas (sludge)	654±11	286±9	282±20	270±6	259±5
Győr - Csorna (filter)	17,6±0,4	9,2±0,4	11,8±1,1	14,5±0,3	14,4±0,3
Sátoraljaújhely - Ricse (sludge)	204±4	205±5	158±8	112±3	116±2
Sopron - Csápordjánosfa (sludge)	557±12	147±8	2994±408	490±11	589±23
Sárvár (sludge)	121±3	113±3	88,1±4,6	75,2±1,7	78,4±1,4
Siklós – Ivándárda (sludge)	267±9	87,8±5,7	116±6	104±3	109±3

Activity concentration of sludge and filter samples in Bq/kg



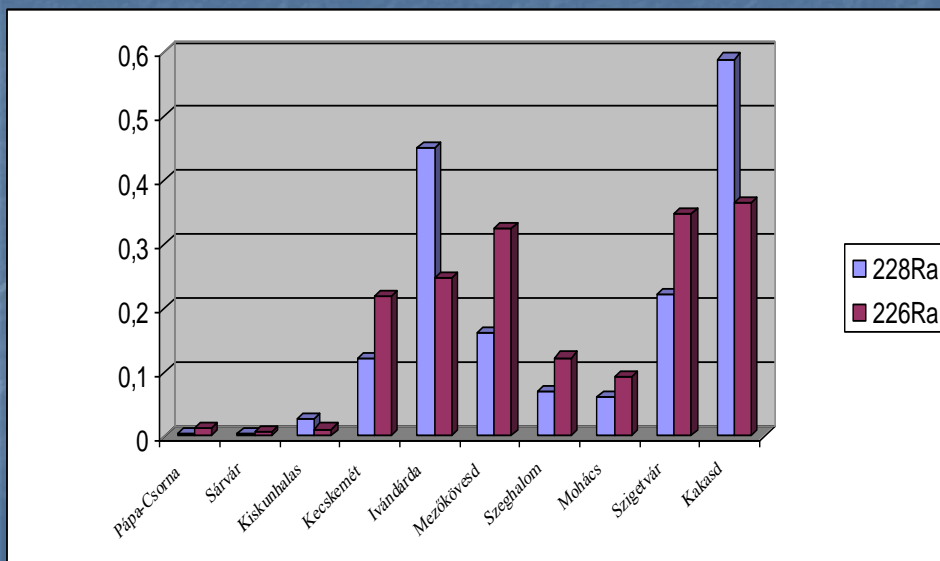
Activity concentration of sludge samples in Bq/kg



Radium activity concentration of raw waters in Bq/l

Place	Ra	Ra
Pápa-Csorna		
Sárvár		
Kiskunhalas		
Kecskemét		
Ivándárda		
Mezőkövesd		
Szeghalom		
Mohács		
Szigetvár		
Kakasd		

The radium activity concentration of the representative raw waters in Bq/l



Results

- Activity concentration of some sludge and used filter material samples are higher by a factor 10 to 15 to the average soil in Hungary
- Few sludge samples contain elevated radium activity concentration that are higher than 1 Bq/g
- Radium activity concentration of few samples of raw waters is close to the 1 Bq/l

Conclusion

- Before the modification of the Decree 47/2003, a complementary investigation is to be carried out for the waterworks of karstic water and for the other waterworks, where the drinking waters are derived from the surface water bodies and groundwater.
- It is concluded that to date, very high elevated activity concentration of the NORM arising from the waterworks have not been measured and the amount of the residues is not too much.
- It is also remarked that lately in Hungary, the consumption of the mineral waters is more and more increasing, and these waters are also derived from the deep aquifers, so an extension of the survey of the NORM is considered in the future to this way.