How can flow system approach help to understand the natural radionuclide content of the drinking water originating from groundwater sources?

A case study in the vicinity of a granitic complex

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József & Erzsébet Tóth

Endowed Hydrogeology Chair Fundation

Ministry of Human Capacities

Introduction

Background

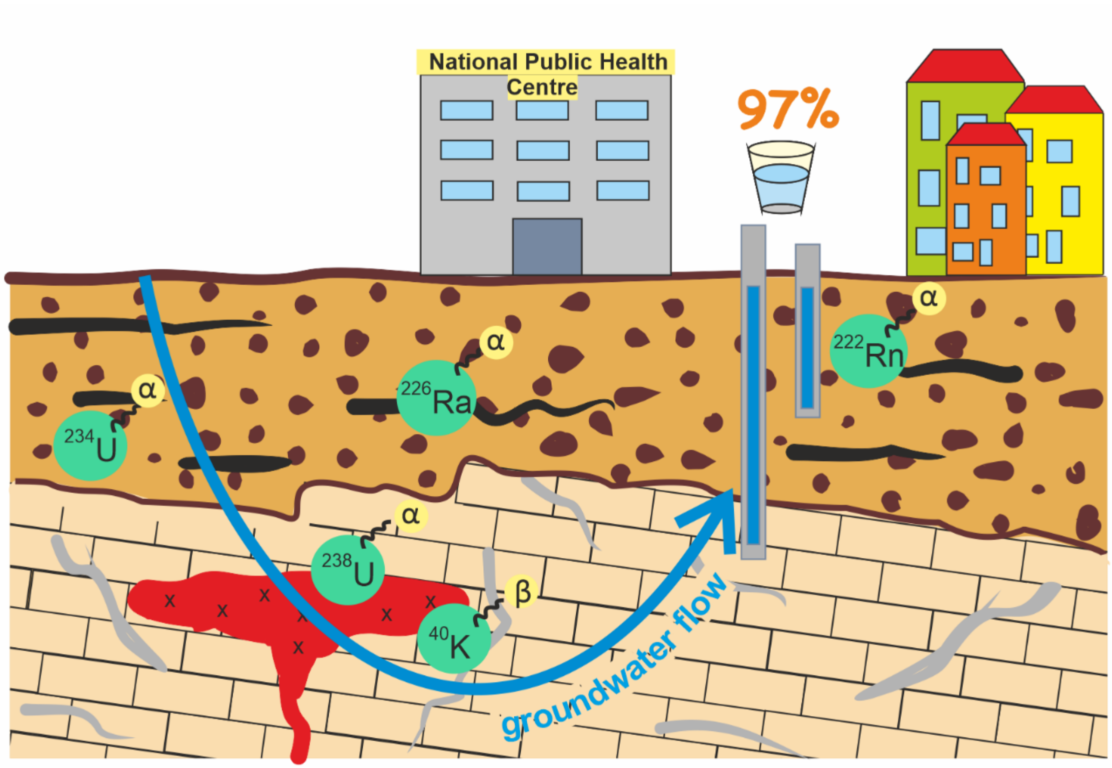
Methods

Results

Conclusion

Drinking water supply in Hungary

- Strict quality requirements based on EU regulations and WHO guidelines



Government Decree 201/2001 (25. Oct), 313/2015 (28. Oct)

- Tritium < *100 Bq/l*
- Indicative dose < *0.1 mSv/year*
- Radon < *100 Bq/l*
- Gross alpha activity < *0.1 Bq/l*
- Gross beta activity < *1 Bq/l*

Council Directive 2013/51/EURATOM

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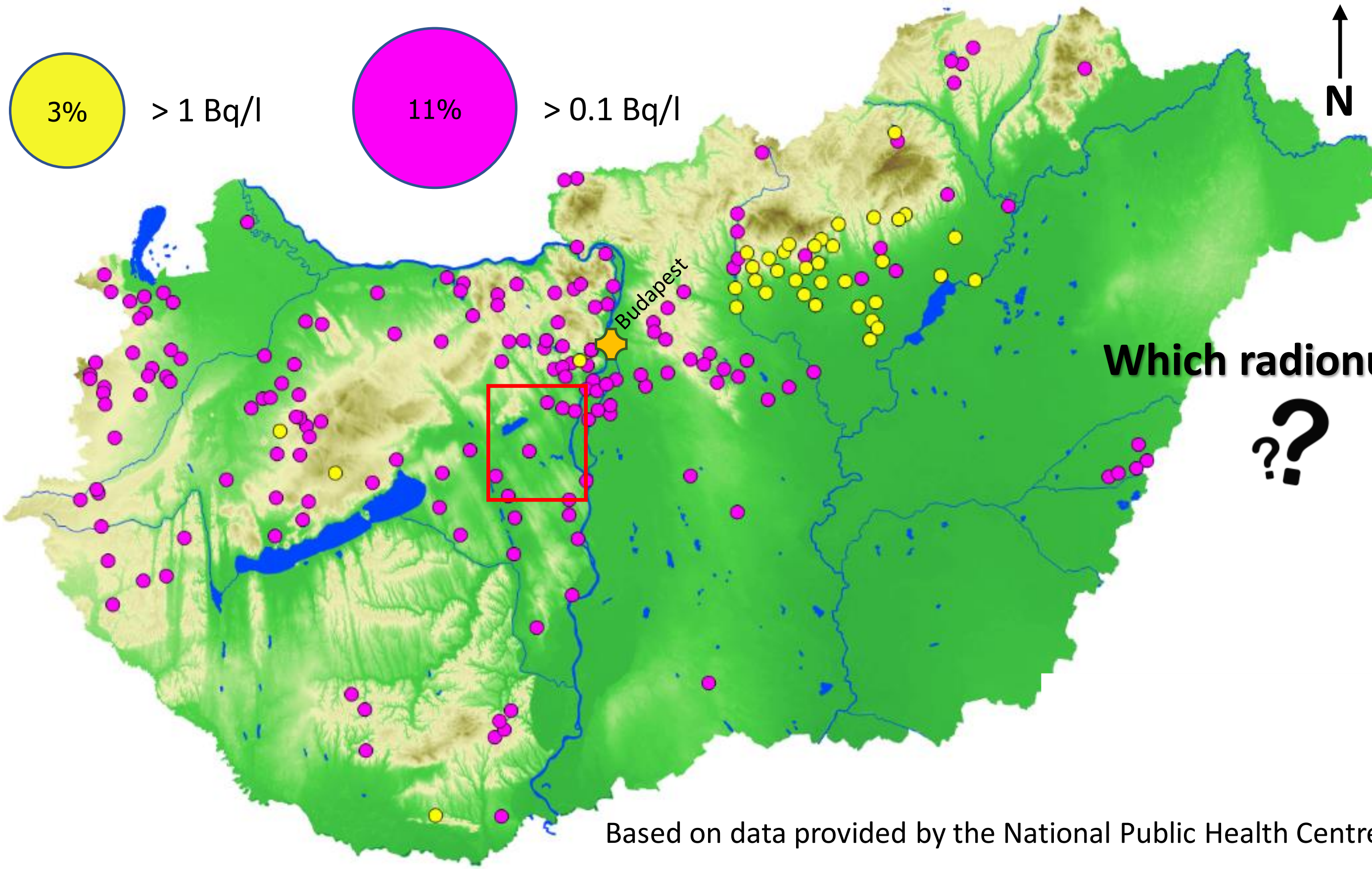
Council Directive 98/83/EC

- Tritium < 100 Bq/l
- Indicative dose < 0.1 mSv/year

Water quality monitoring - radionuclides

- Gross alpha and gross beta measurements



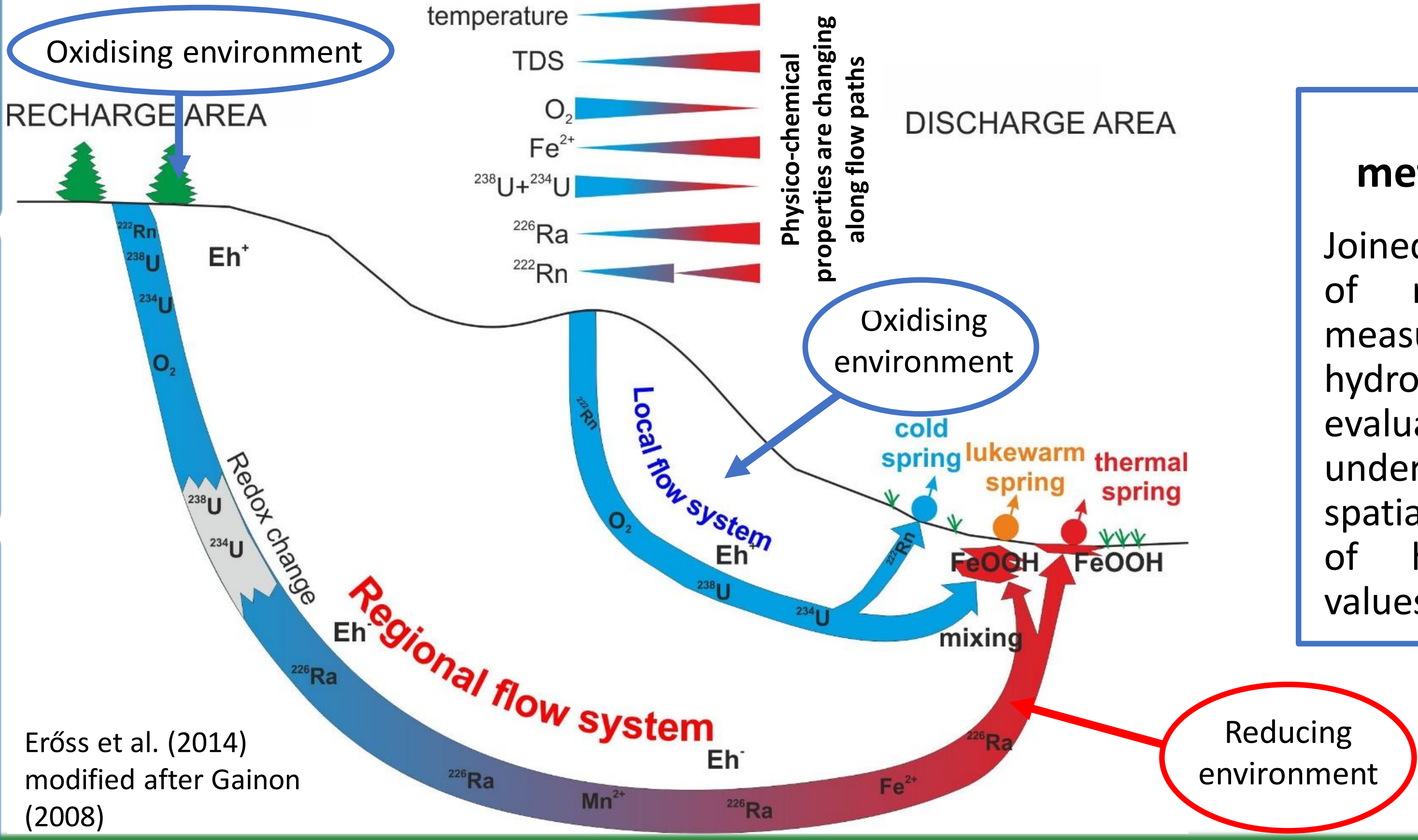
Which radionuclide ??

Based on data provided by the National Public Health Centre (Febr 2019)

- Gross alpha activity: total activity of all alpha particle emitters
- Gross beta activity: total activity of all beta particle emitters excluding tritium, although other weak beta emitters are also excluded using most screening measurement techniques
    - Expressed in terms of unit of activity per unit of volume (e.g. Bq/l).
    - The gross screening measurements do not provide the identity of or activity concentration of specific alpha-emitting radionuclides.

Radionuclides in the gravity driven groundwater flow system

- ^{40}K and the members of the natural ^{238}U decay chain (^{238}U, ^{234}U, ^{226}Ra, ^{222}Rn) are the most common (through groundwater-rock interaction e.g. dissolution,desorption, diffusion, atomic recoil)
- Uranium and radium mobilization is affected by the redox conditions



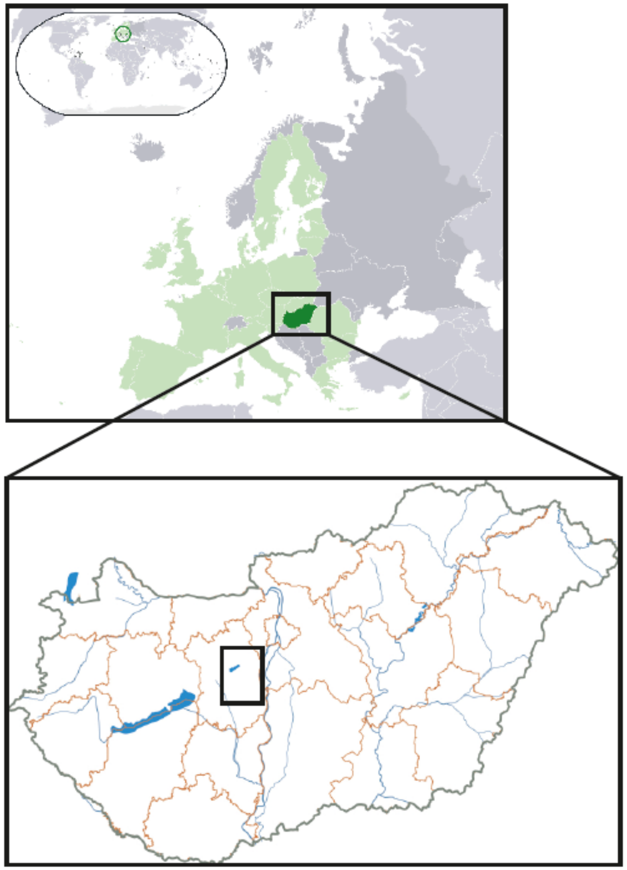
Erőss et al. (2014) modified after Gainon (2008)

New methodology:

Joined application of nuclidespecific measurements and hydrogeological evaluation to understand the spatial distribution of high activity values

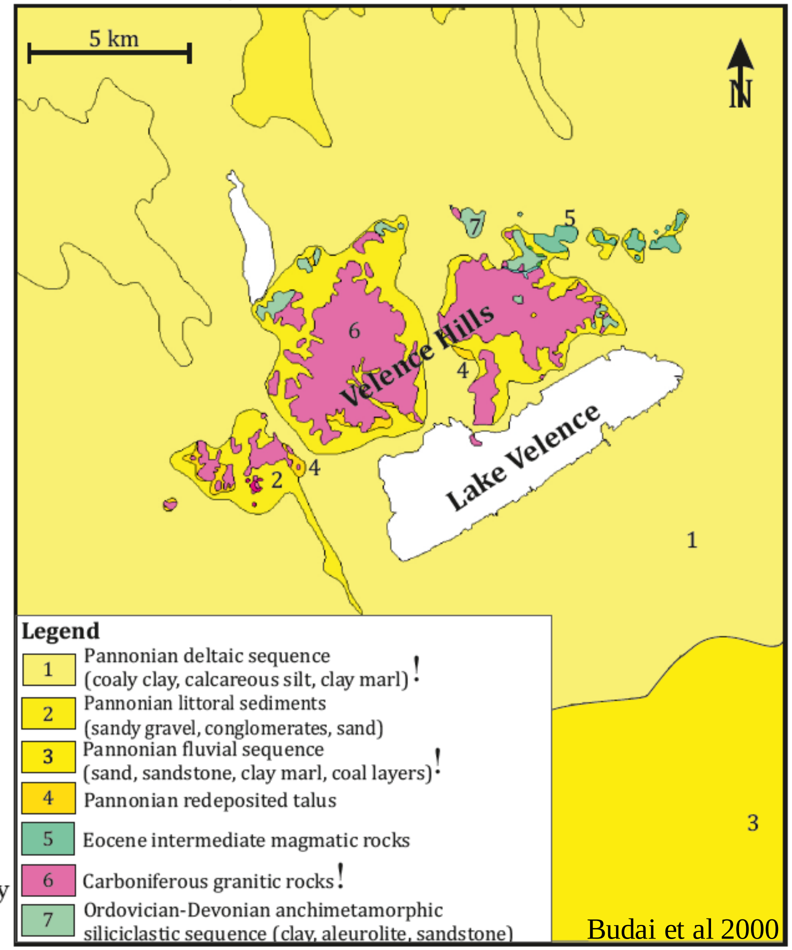
Study area

- Location: Velence Hills and the southern foreland of Lake Velence



https://lad.wikipedia.org/wiki/Dosya:Location_Hungary_EU_Europe.png
https://i.stack.imgur.com/8oNso.jpg

- Lithology:
  Recharge area: Carboniferous granitic rocks
  Aquifer: Pannonian sandy aquifer with carbonatic and organic-rich beds



Methods

- 53 water samples were taken from groundwater, surface water and springs
  - Physico-chemical properties (T, DO, EC, pH, ORP)

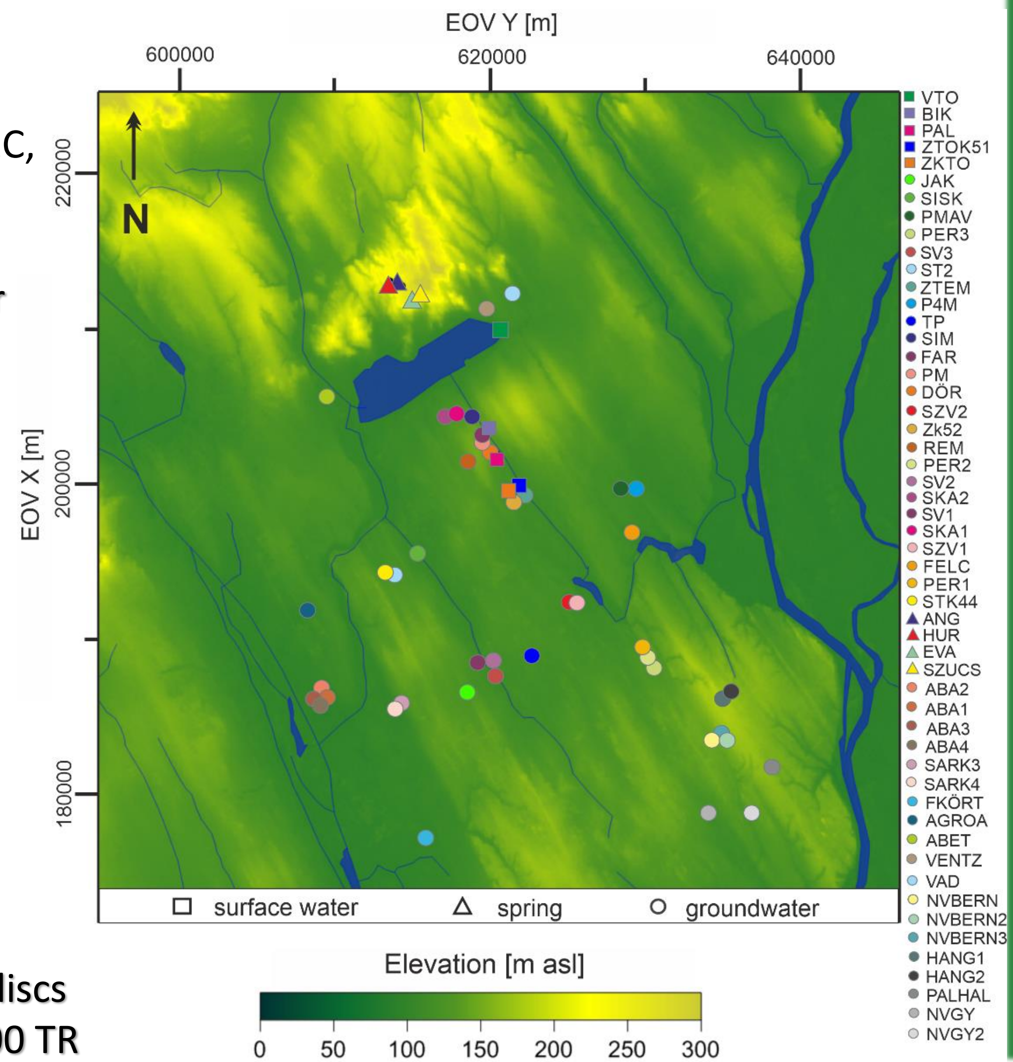
    YSI Pro Plus multiparameter water quality instrument

  - Water chemistry (Na^+, K^+, Ca^{2+}, Mg^{2+}, HCO_3^-, SO_4^{2-}, Cl^-)

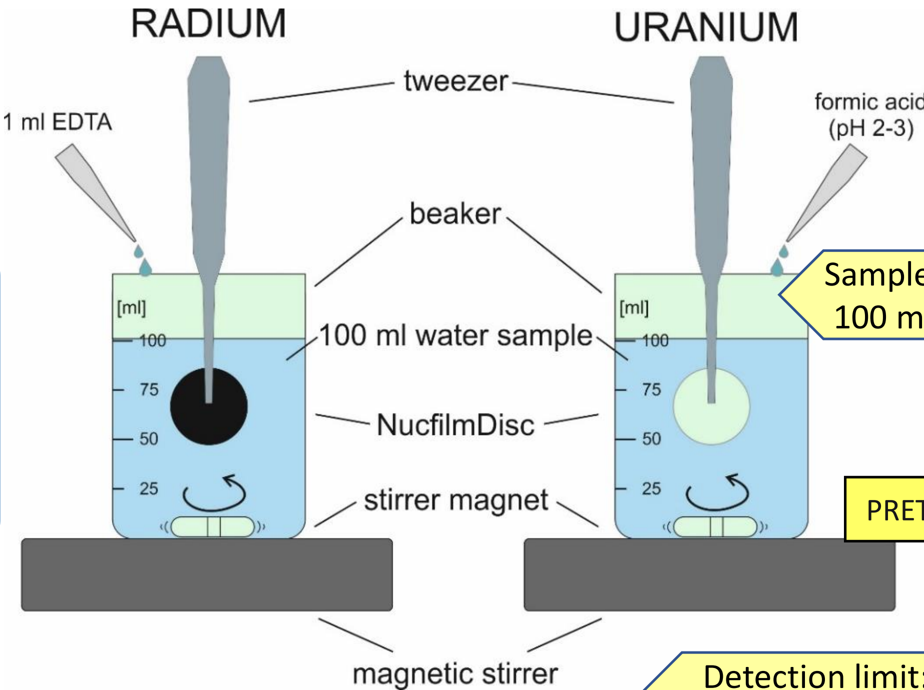
    ICP-MS
    ion chromatography
    UV-VIS photometer
    titrimetry

- Radionuclides (^{234}U+^{238}U, ^{226}Ra, ^{222}Rn)

  Alpha-spectroscopy using Nucfilm discs
  Liquid scintillation using Tricarb 1000 TR



Nuclidespecific measurements



- $^{234}U+^{238}U$ and ^{226}Ra activity concentrations were measured by solid-state alpha-spectroscopy using selectively adsorbing Nucfilm discs

Surbeck (2000)

PRETREATMENT

Sample: 100 ml

Sample: 10 ml

MEASUREMENT

Detection limit: 5 mBq/l

Detection limit: 2 Bq/l

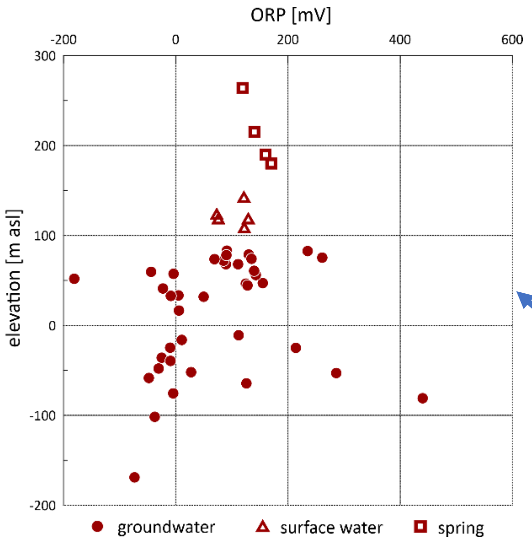
Duration: 32-44 h

Duration: 20 min

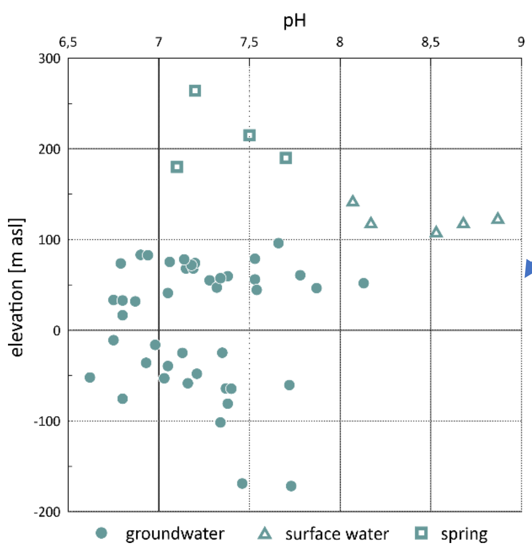
- ^{222}Rn activity concentrations were measured by liquid scintillation using Tricarb 1000 TR

(atomfizika.elte.hu)

Physicochemical properties & water chemistry



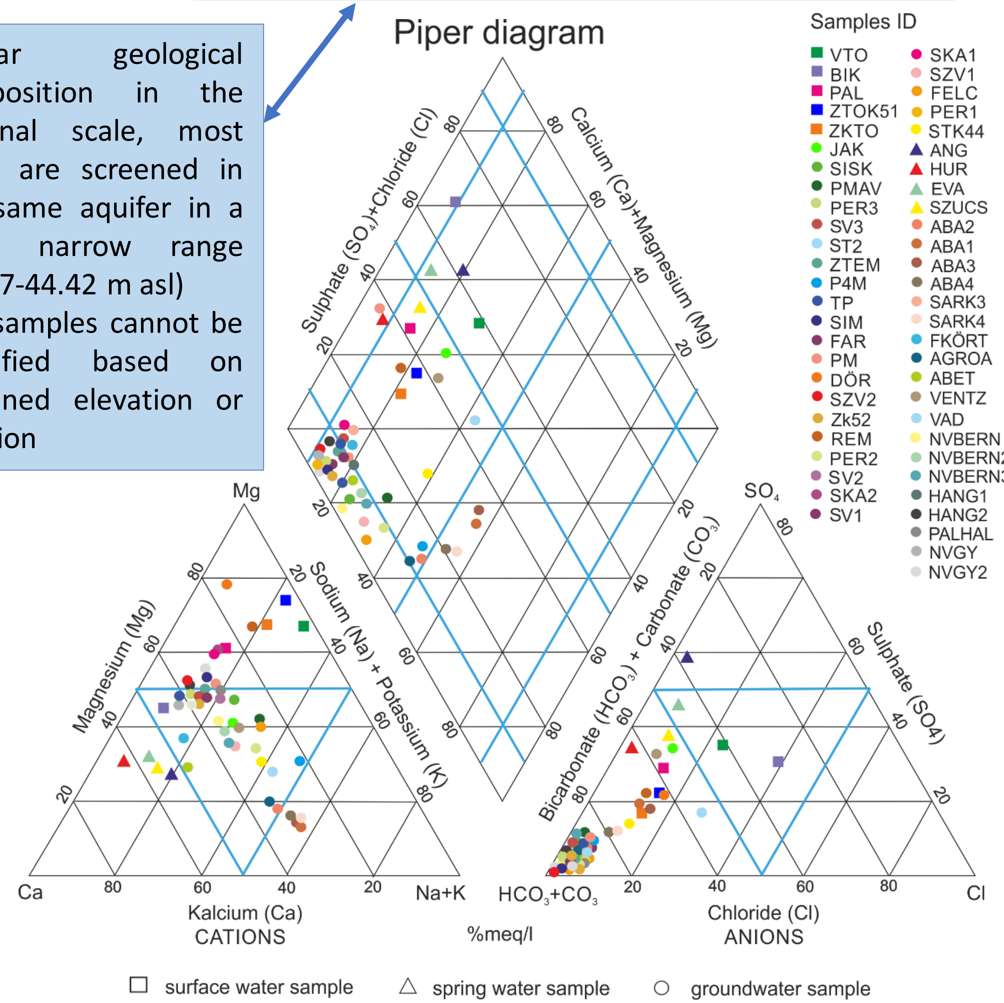
Oxidising environment in most cases



Circumneutral pH

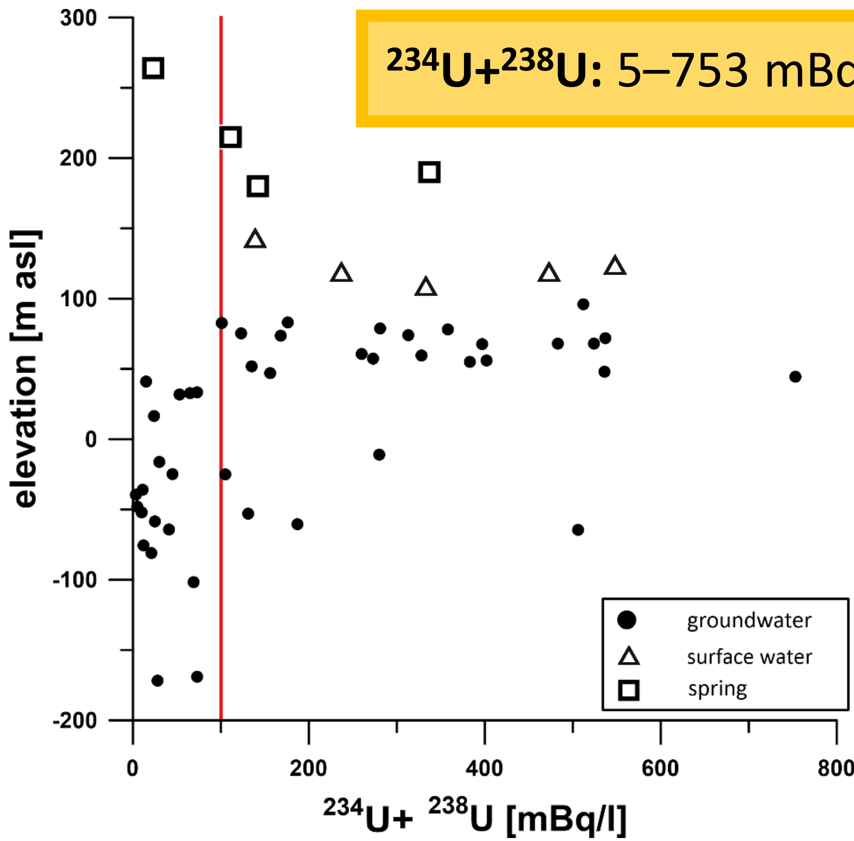
- Similar geological composition in the regional scale, most wells are screened in the same aquifer in a quite narrow range (78.77-44.42 m asl)
- The samples cannot be classified based on screened elevation or location

calcium-sodium-bicarbonate (*23 samples*), calcium-sodium-bicarbonate-chloride-sulphate (*20 samples*) → **two of the most hydrochemical facies**
Dominant anion: bicarbonate → **uranium mobilisation)**

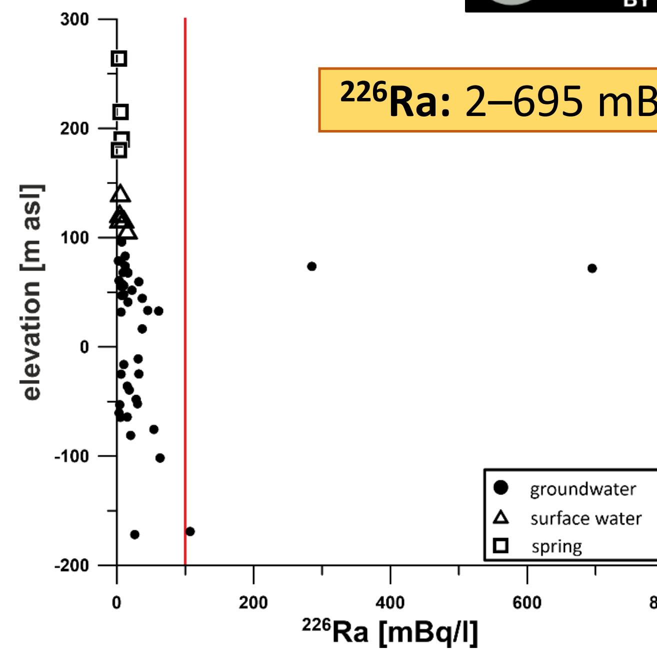


Radionuclide mobilisation is affected mainly by pH and redox contitions

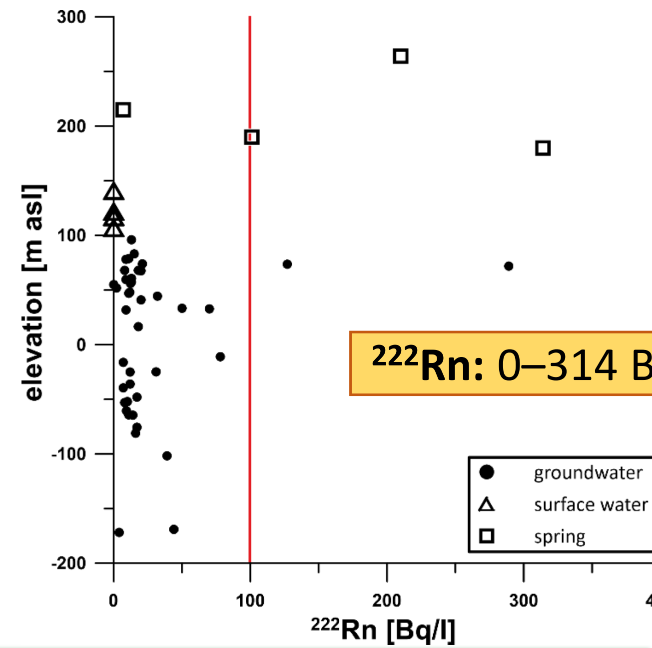
Radionuclides



$^{234}U+^{238}U$: 5–753 mBq/l



^{226}Ra: 2–695 mBq/l



^{222}Rn: 0–314 Bq/l

- drinking water quality measurements showed gross alpha activity values exceeded the 0.1 mBq/l threshold
- necessity of nuclidespecific measurements to indentify the exact radionuclide which is dissolved in drinking water
- uranium is responsible for the high gross alpha activity

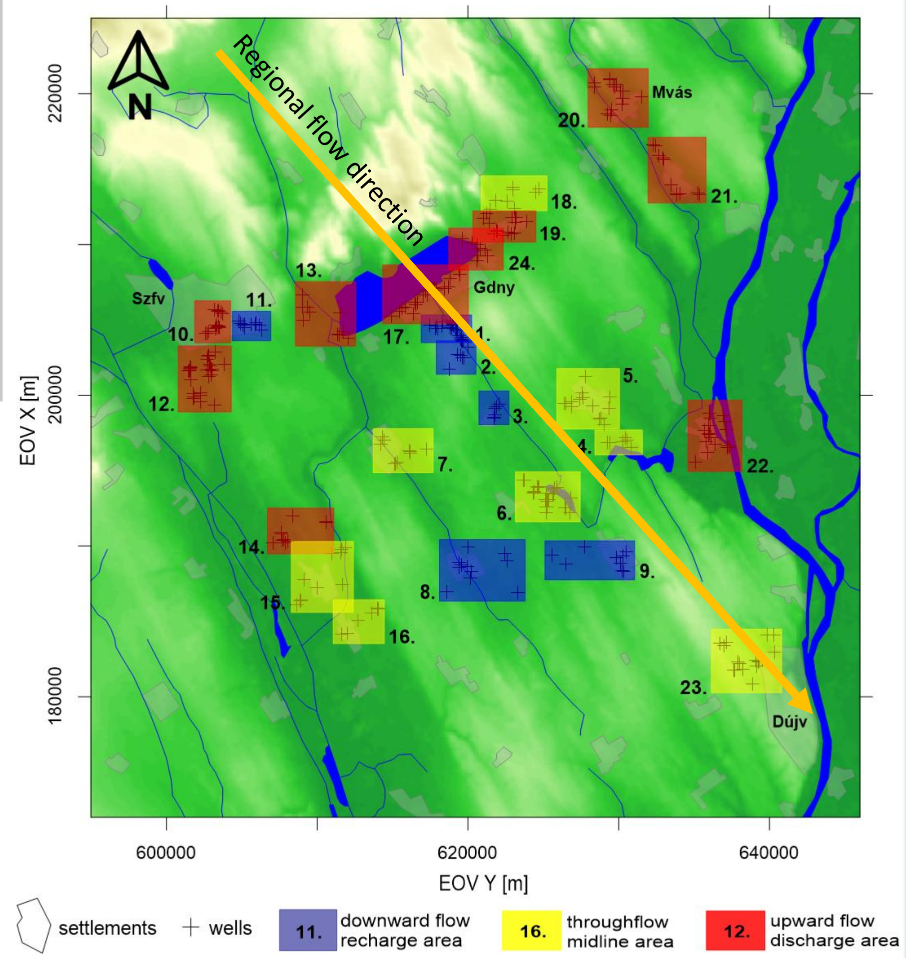
Hydrogeological evaluation

Physico-chemical properties (e.g. temperature, pH, ORP, dissolved solid content etc.) are changing along the flow paths and according to the flow regime (recharge, midline, discharge area)

→ In general: local flow system, recharge/midline area can be oxidising whereas regional flow system, discharge area can be reducing environment

→ **U** mobilisation: oxidising environment
Ra mobilisation: reducing environment

- Based on archive well data (coordinates, elevation, screened interval, depth of static water level) both the vertical and horizontal direction of the groundwater flow can be determined
- The southern foreland of Lake Velence is dominated by recharge and midline area; groundwater flow discharges mainly at the lake and the River Danube



Results – possible scenarios to explain elevated activity

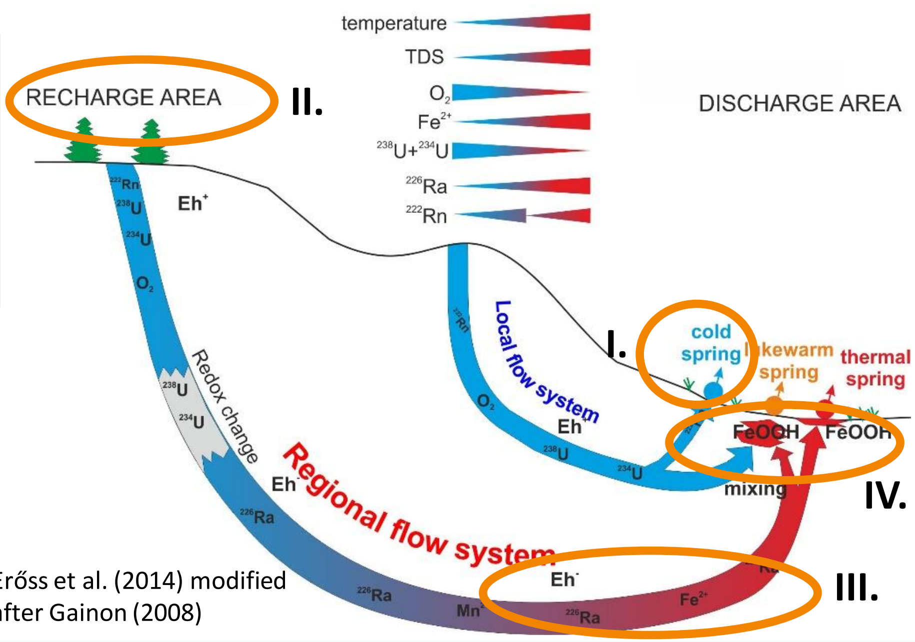
I. Springs: local flow system, radon emitted from the soil **101–314 Bq/l**, uranium originated from the bedrock (granite) **111–337 mBq/l**

II. Local flow system (different flow regimes: recharge, throughflow, discharge)

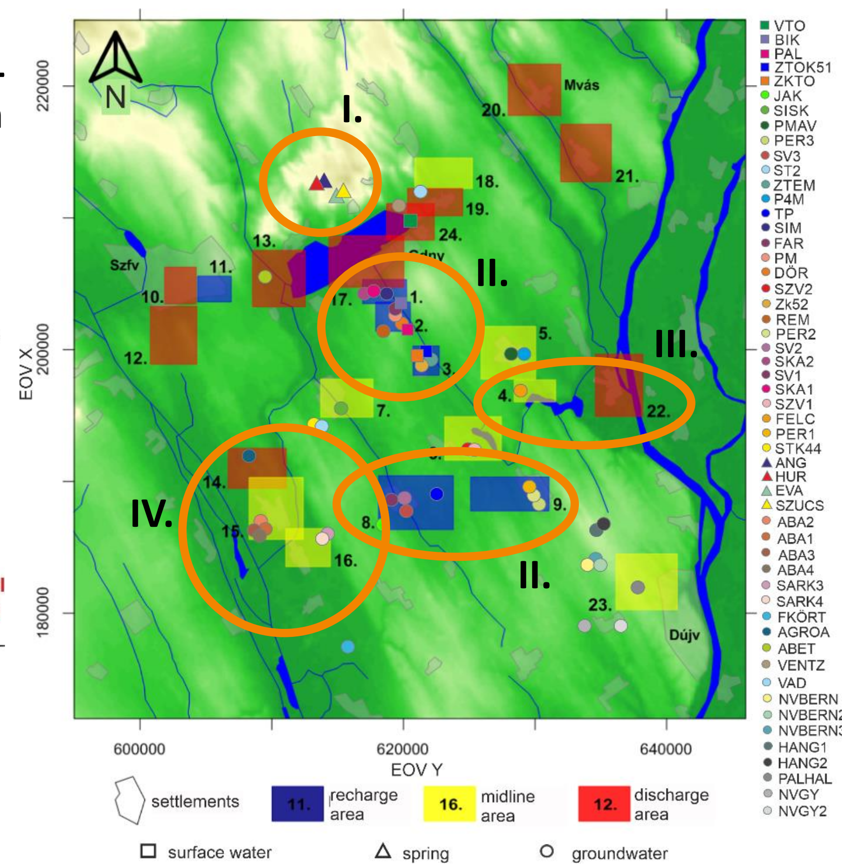
→ oxidising environment (uranium can be mobilized by groundwater) **3–358 mBq/l**

III. Regional flow system toward the regional discharge area (River Danube) with reducing environment (radium can be mobilized by groundwater) **107 mBq/l**

IV. Mixing of different flow systems (local and regional) - dissolved uranium (**168–537 mBq/l**) because of the oxidising environment, radium (**285–695 mBq/l**) and radon (**127–289 Bq/l**) accumulation through the forming of FeOOH and MnOOH minerals



Erőss et al. (2014) modified after Gainon (2008)



Summary

Measured gross alpha activity above the limit → nuclidespecific measurements → high uranium activity concentrations

GRAVITY DRIVEN GROUNDWATER FLOW AS A GEOLOGIC AGENT → MOBILIZE, TRANSPORT AND ACCUMULATE MATTERS (RADIONUCLIDES AS WELL)

p(z)	flow regime	U [mBq/l]		Ra [mBq/l]		Rn [Bq/l]		ORP [mV]	
		min	max	min	max	min	max	min	max
1	⬇	139	753	2	37	11	32	121	130
2	⬇	237	536	4	10	12	14	114	143
3	⬇	187	548	3	10	9	13	73	140
4	⇨		73		107		44		-74
5	⇨	28	41	15	26	4	11	-12	115
6	⇨	69	273	8	63	13	39	-37.5	4
7	⬇	135	537	22	695	2	289	-181	85
8	⬇	156	524	7	16	8	21	89.4	155
9	⬇	21	280	7	16	8	78	112	440
13	⬆		53		6		9		49.7
14	⬆		168		285		129		69
15	⇨	12	73	32	61	17	70	-10	5
16	⇨	24	176	12	37	15	18	6	91
18	⇨		15		16		20		-23
23	⇨		25		34		23		-48
24	⬆		358		6		9		90

1 p(z) profile ID

⬇ discharge area

⇨ througflow

⬆ recharge area

the measured activity concentration

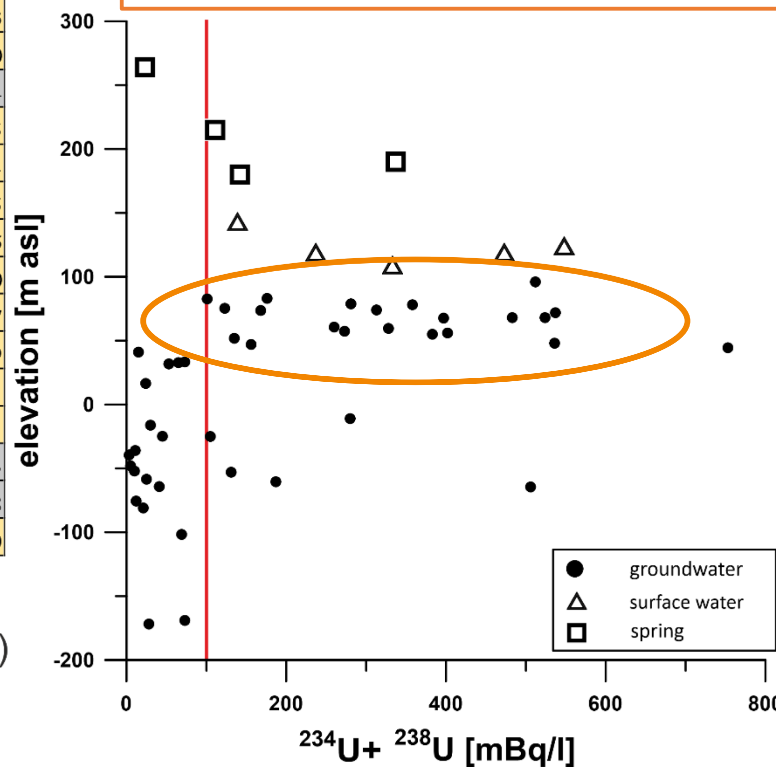
exceeds / doesn't exceed the threshold (gross alpha 0.1 mBq/l radon 100 Bq/l)

based on the measured ORP value, the environment is

oxidising

reducing

! Although the lithology is regionally uniform, there is a wide range of uranium activity values



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Conclusion

- Elevated uranium activity ← elevated gross alpha activity

possible sources exist: granitic rocks, organic-rich lake sediments, carbonatic and organic rich beds in the aquifer – GEOLOGICAL COMPOSITION

spatial distribution of high activity concentration – GRAVITY DRIVEN GROUNDWATER FLOW SYSTEM AS A GEOLOGIC AGENT

recharge areas with oxidising conditions → uranium mobilization → elevated uranium activities

TAKE HOME MESSAGE

- Screening natural radioactivity in drinking water through gross measurements BUT nuclide specific measurements are neccessary to understand the connection between geology, groundwater flow system and the occurrence of natural radionuclides in groundwater
- This complex approach can help to improve the drinking water supply strategy

Acknowledgement

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