EGU General Assembly 2022

Natural radioactivity and rock-water interactions in the springs of Sopron Mountains (Hungary)

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25/05/2022

József & Erzsébet Tóth Endowed Hydrogeology Chair Fundation

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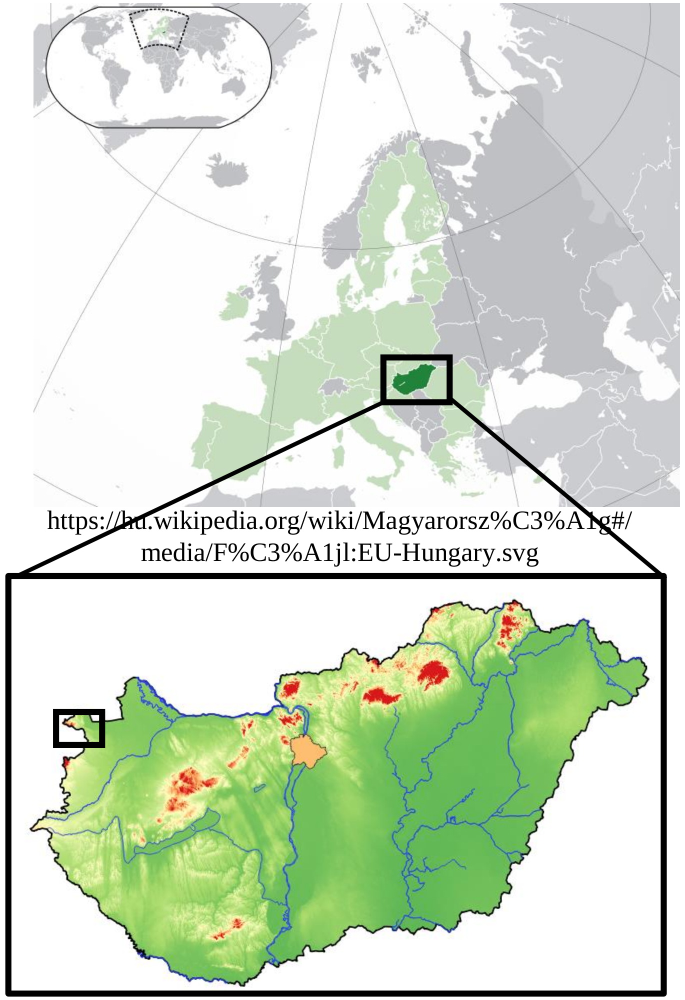
Study area

Location: **Sopron Mountains** in Hungary



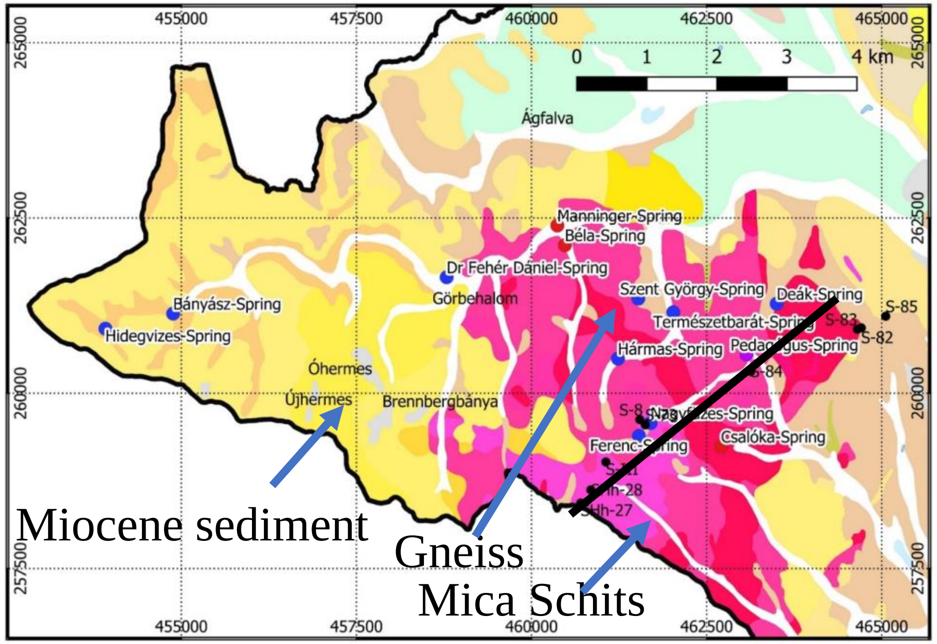
https://hu.wikipedia.org/wiki/Magyarorsz%C3%A1g#/media/F%C3%A1jl:EU-Hungary.svg

Why are the Sopron Mountains intresting?

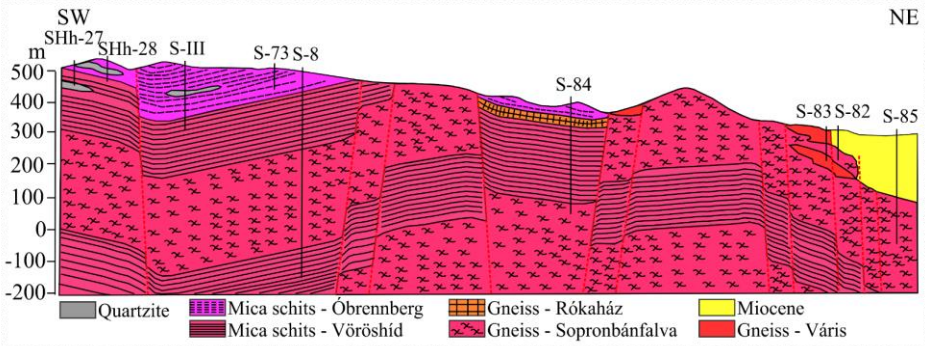
- Metamorphic rocks → the protolith of the gneiss is granite. U conc.:
  Metagranite, gneiss → **2.2 – 3.4 ppm**
  Leucophyllite → **1.6 – 3.4 ppm** (Török, 2001)

U conc. in the Earth's crust ~2.5 ppm
(Wedephol, 1995**)**

- Ra-226 conc. in the soil **88 Bq/kg**, (avarage in Hungary 33 Bq/kg) (Freiler et al. 2016)
- The area was previously the site of **fissile material research**
- Significant **radon anomaly** is known at the geophysical observatory **(500-1000 kBq m^{-3})**
- The Rn activity concentration in some springs **exceeds the recommended level of 100 Bq L^{-1}.** For example Csalóka Spring **220-230 Bq L^{-1}** (Aros, 2003; Freiler, 2009; Freiler, 2016)



Lithology: metamorfic rocks (Gneiss, Mica Schits)



Geological section from the Sopron Mountains modified after Fülöp (1990)

Radionuclides in the gravity driven groundwater flow system

Most common in groundwater ^{40}K and uranium series members (^{238}U, ^{234}U, ^{226}Ra, ^{222}Rn) are present

Radionuclides are **natural tracers** – their behaviour in rock-water systems are well known.

Uranium mobile mainly in oxidizing environments: recharge area, local flow system

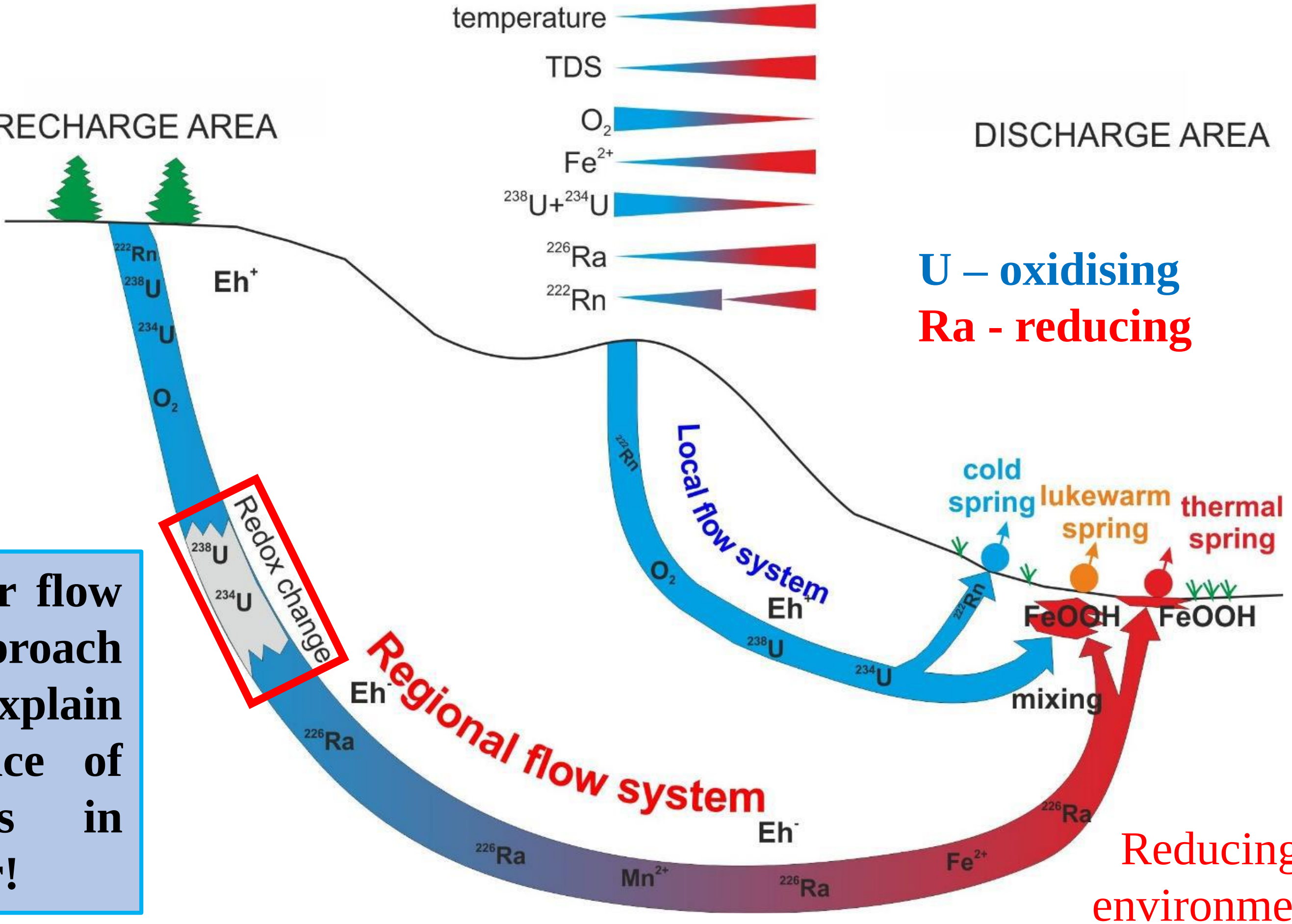
Radium mobile mainly in reducing environments: discharge area, regional flow system

Radon noble gas → significantly mobile, but short half life and/ or fast travel time

The distribution of radionuclides varies along the flow path!
Redox sensitive parameters

Oxidising environment

U – oxidising
Ra - reducing



Reducing environment

Groundwater flow system approach helps to explain the occurence of radionuclides in groundwater!

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

Objective

Understand the rock-water interactions!

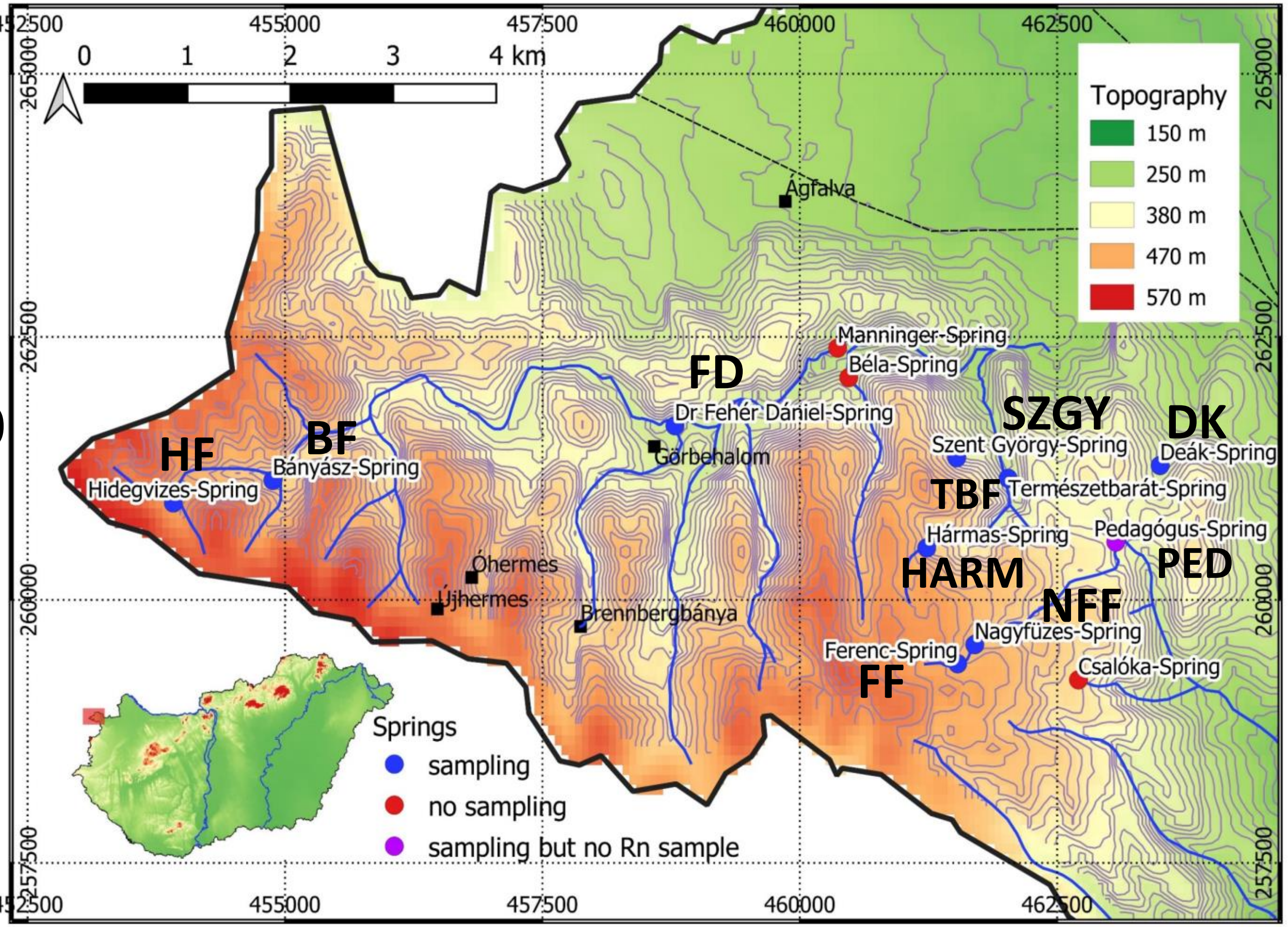
- Investigation of the uranium, radium, radon activity concentration in spring waters from groundwater flow system approach

Methods

13 springs were visited (24.08.2021-25.08.2021)

10 spring water + 1 surface **water sample**

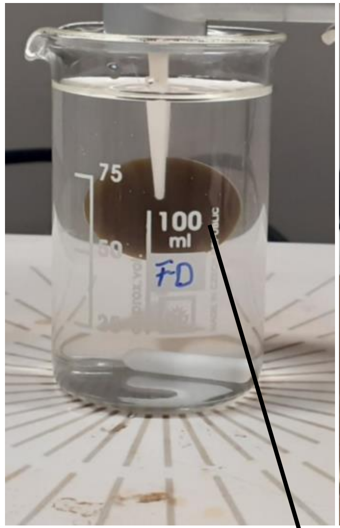
- Physico chemical properties (T, DO, EC, pH, ORP)
  YSI Pro Plus multiparameter water quality instrument
- Water chemistry (Na^+, K^++, Ca^{2+}, Mg^{2+}, HCO_3^-, SO_4^{2-}, Cl^-)
- Radionuclides (sum of U isotopes, ^{226}Ra, ^{222}Rn):
  Alpha-spectroscopy based on Nucfilm discs and liquid scintillation detection

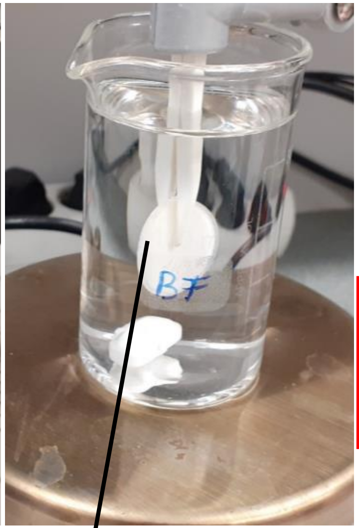


(HF, BF … name of sample)

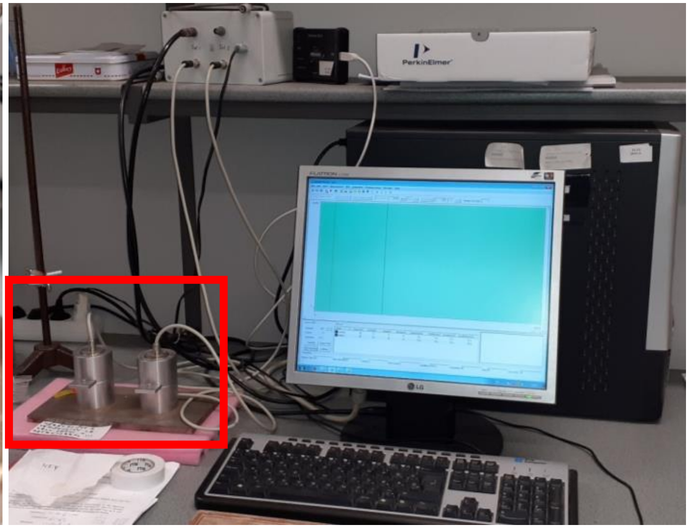
Alpha spectroscopy

sample preparation





measurement



Radium

Nucfilm disc

Uranium

single channel alpha spectroscop

Duration:
preparation 8h (radium) + 20h (uranium) - measurement 24 h

Detection limit: 5 mBq L^{-1}

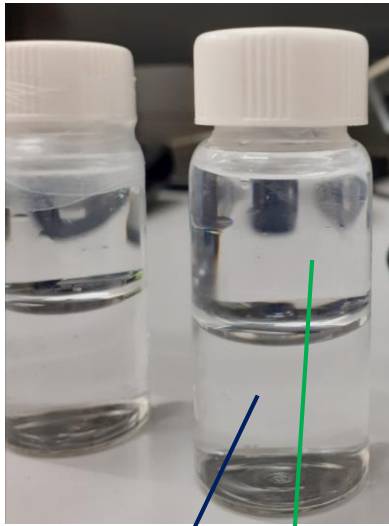
Measurement of ^{234}**U** + ^{238}**U** and ^{226}**Ra** activity concentrations by alpha spectroscopy **based on Nucfilm discs preparation -** Surbeck (2000)

Liquid scintillation detector (LSC)

measurement

TriCarb 1000 TR instrument

cuvette



Water sample

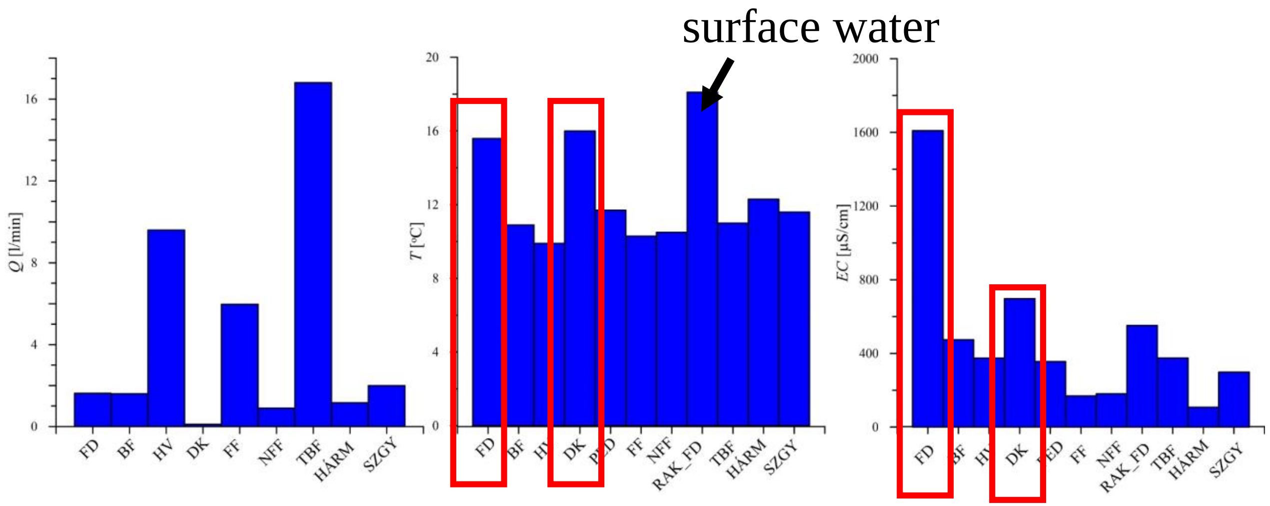
Liquid scintillation cocktail

Duration:
measurement 20 min

Detection limit: 2 Bq L^{-1}

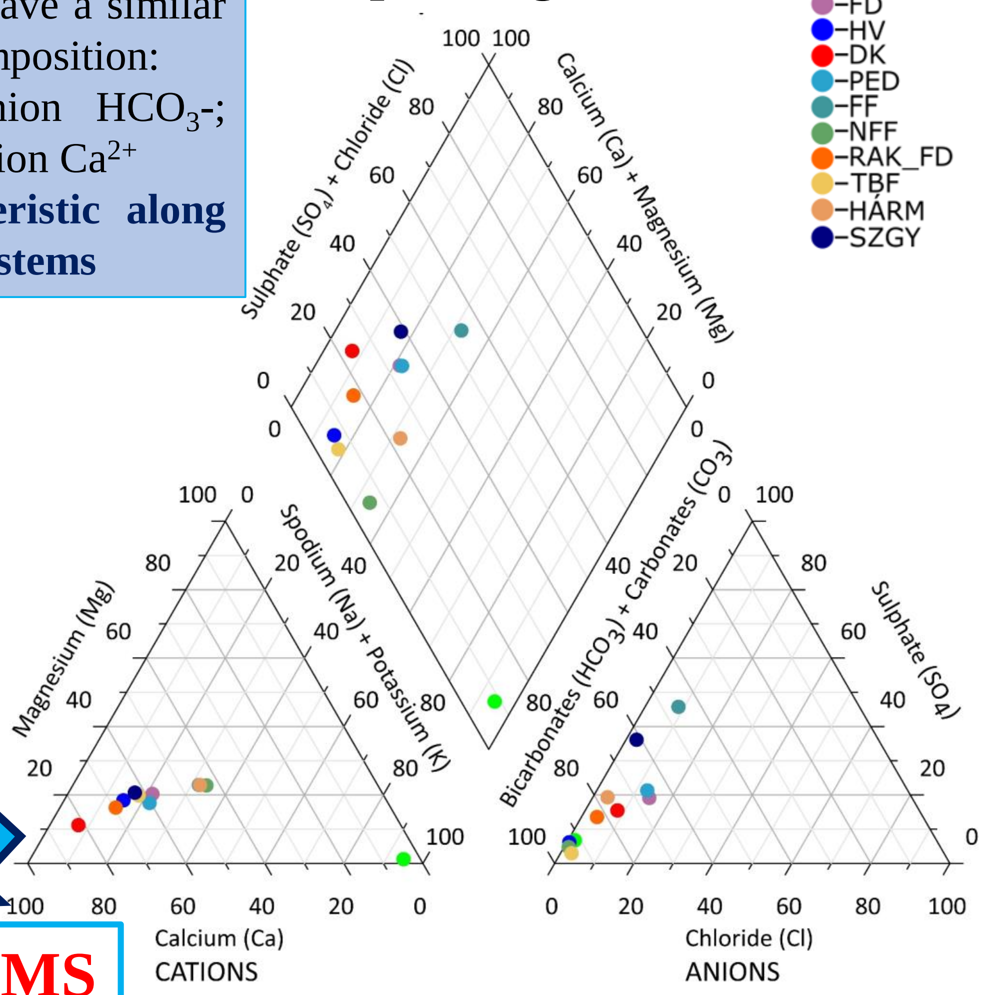
^{222}**Rn** activity concentrations were measured by liquid scintillation using Tricarb 1000 TR

Physicochemical properties and water chemistry



The waters have a similar chemical composition: dominant anion HCO_3^-; dominant cation Ca^{2+} → **Characteristic along local flow systems**

Piper diagram



Discharge rate → **Low** - exceeds only 5 l/min in 3 cases (protection)
TBF (16.8 l min^{-1}) ; HF (9.6 l min^{-1}); FF (5.97 l min^{-1})
Temperature → **Low** (9.9-12.3°C) – **EXCEPTION** FD (15.6°C), DK (16°C)
Specific Electric Conductivity → **Low** (107.5-474.2 μS cm^{-1}) – **EXCEPTION** FD (1609 μS cm^{-1}), DK (697 μS cm^{-1})
Redox potential, dissolved oxygen content → **Oxidative conditions**

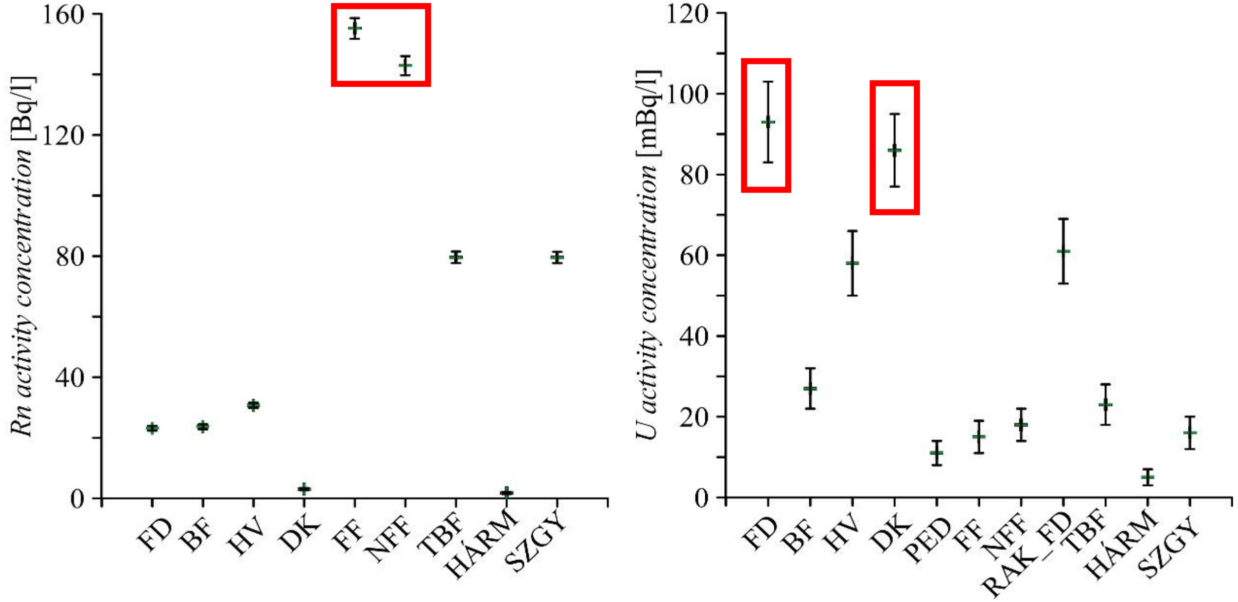
BUT: Possible explanation for higher T and conductivity of **FD** and **DK:** greater penetration depth, longer flow path → Longer travel time → more time for rock-water interactions

LOCAL FLOW SYSTEMS

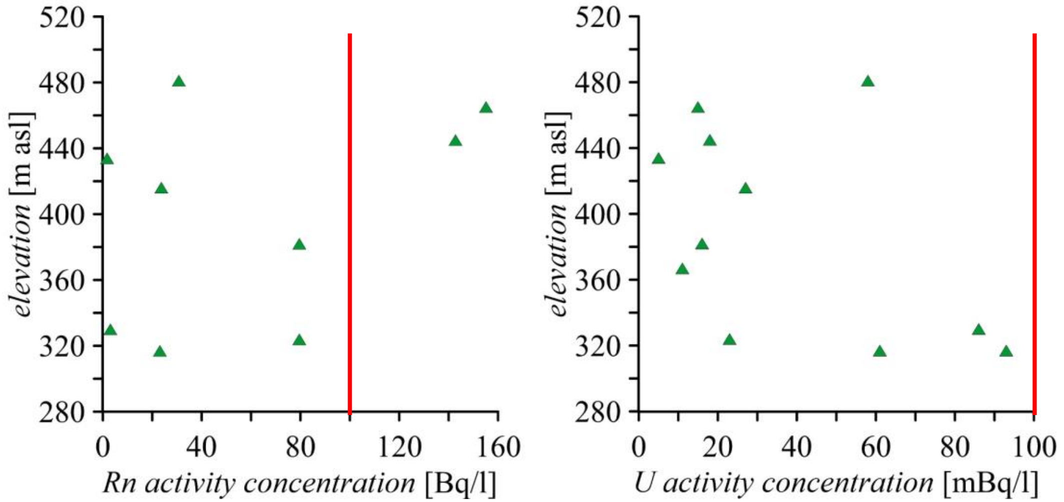
Short travel time!

Radionuclides

Radon and uranium activity concentrations in springs



Radon and uranium activity concentration vs. elevation



Radon → low, but in 2 cases the concentration exceeded the recommended level of 100 Bq L^{-1}: NFF (142 Bq L^{-1}); FF (155 Bq L^{-1})

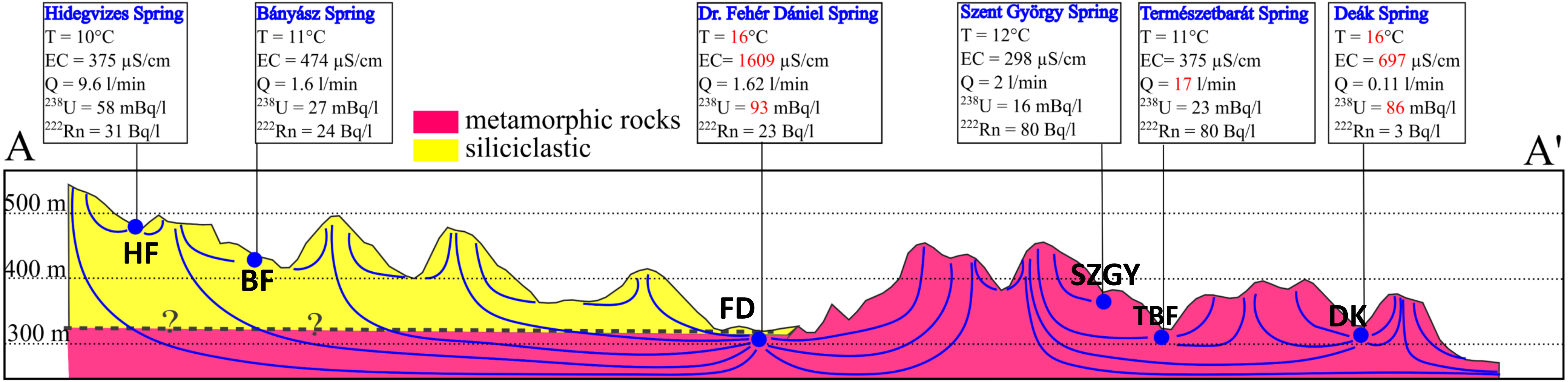
Radium → around the **detection limit** (5 mBq L^{-1})
→ **OXIDISING** conditions (Ra is mobile in reducing environment)

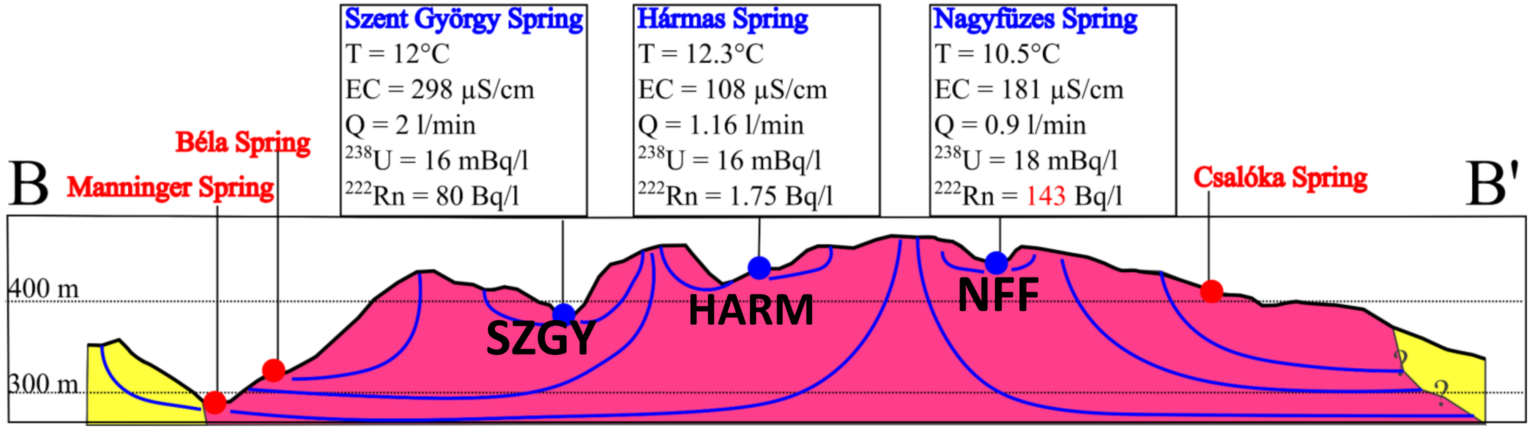
Uranium → 5-93 mBq L^{-1} FD (93 mBq L^{-1}); DK (86 mBq L^{-1})

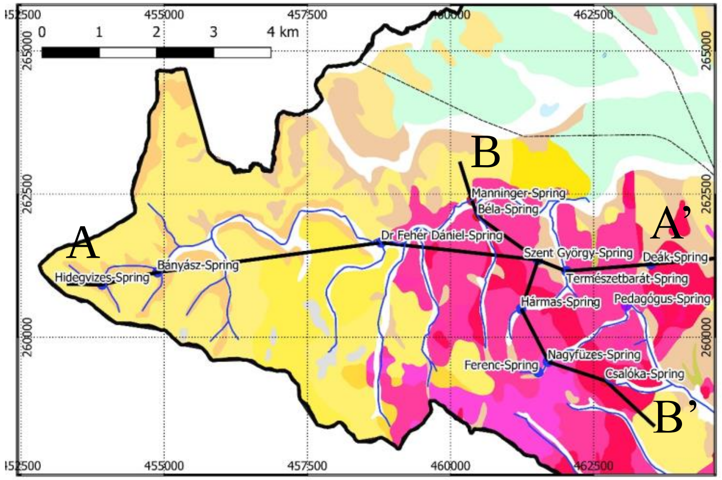
Uranium concentration is higher where the travel time is longer! – FD, DK
Radon concentrations are higher above metamorphic rocks

Schematic sketch of the area

Fehér Dániel Spring - FD, (Deák spring - DK) →„Foothill position" = longer travel time = more time for rock-water interaction







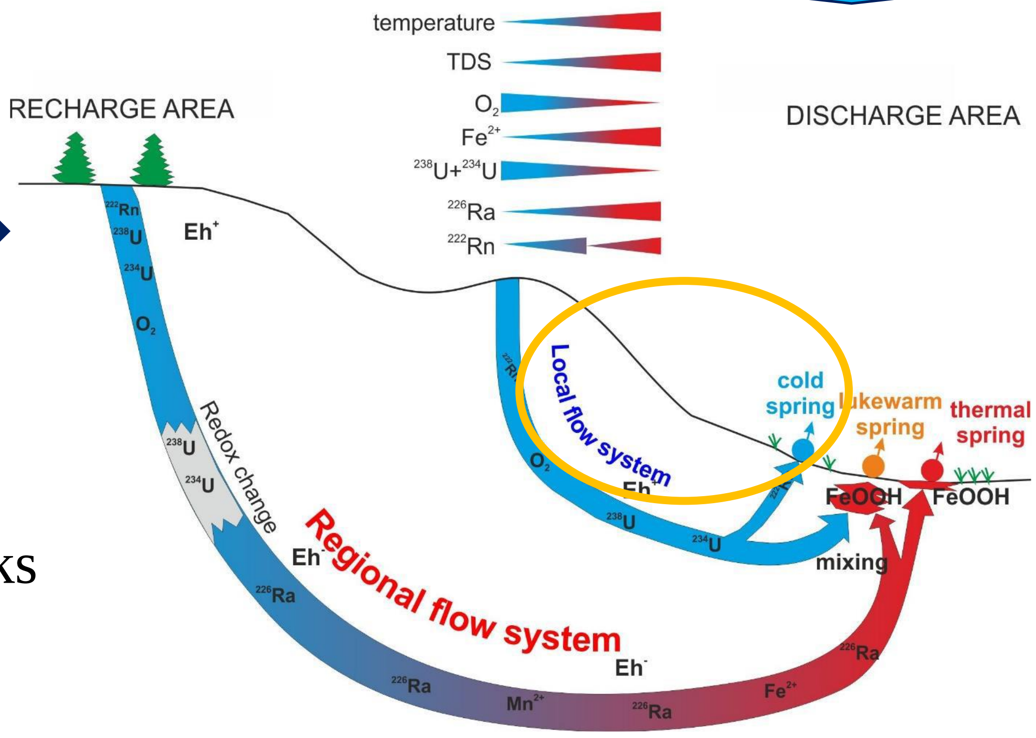
Conclusion

short travel time
short time for rock-water interactions

- There are mainly **local flow systems** in the area

- Uranium is higher in foothill springs (FD, DK): longer-travel time, ie. more time for rock-water interactions

- Radon concentrations are higher above metamorphic rocks



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

There is no health risk **<u>in the springwaters in the recharge area</u>**, but the radon activity concentration needs to be monitored! C_{Rn} **> 100 Bq L^{-1}**

springs serve as a source of drinking water for hikers

TAKE HOME MESSAGE

Groundwater flow system approach helps to explain the occurence of radionuclides in groundwater.

Thank you for your attention!