^{39}Ar and ^{37}Ar in deep groundwater: Evaluation regarding young components, cross-formation flow and in-situ production

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Introduction

- Isotopes of ^{37}Ar ($t_{1/2}$ = 34.9d) and ^{39}Ar ($t^{1/2}$ = 269 yr) are used to date young groundwater up to ~1200 years and/or investigate mixing between different groundwater components[1].
- Infiltrating surface water has cosmogenic ^{39}Ar of 100%$_{modern}$ and is (essentially) free of ^{37}Ar.
- Along the flow path in the subsurface, ^{39}Ar and ^{37}Ar activities change[2] due to:
  - the decay of cosmogenic ^{39}Ar
  - the addition of ^{37}Ar and ^{39}Ar by formation specific subsurface production (secular equilibrium).
- In layered aquifer systems ("Stockwerk-Bau") ^{39}Ar and ^{37}Ar provide important constraints on the identification of inflow of external fluids or of cross-formation flow between different aquifers, which are important aspects in the context of radwaste repository research in rock formations of sedimentary basins such as the Swiss TBO investigation programme.

Evolution of ^{37}Ar, ^{39}Ar along a deep aquifer flow path

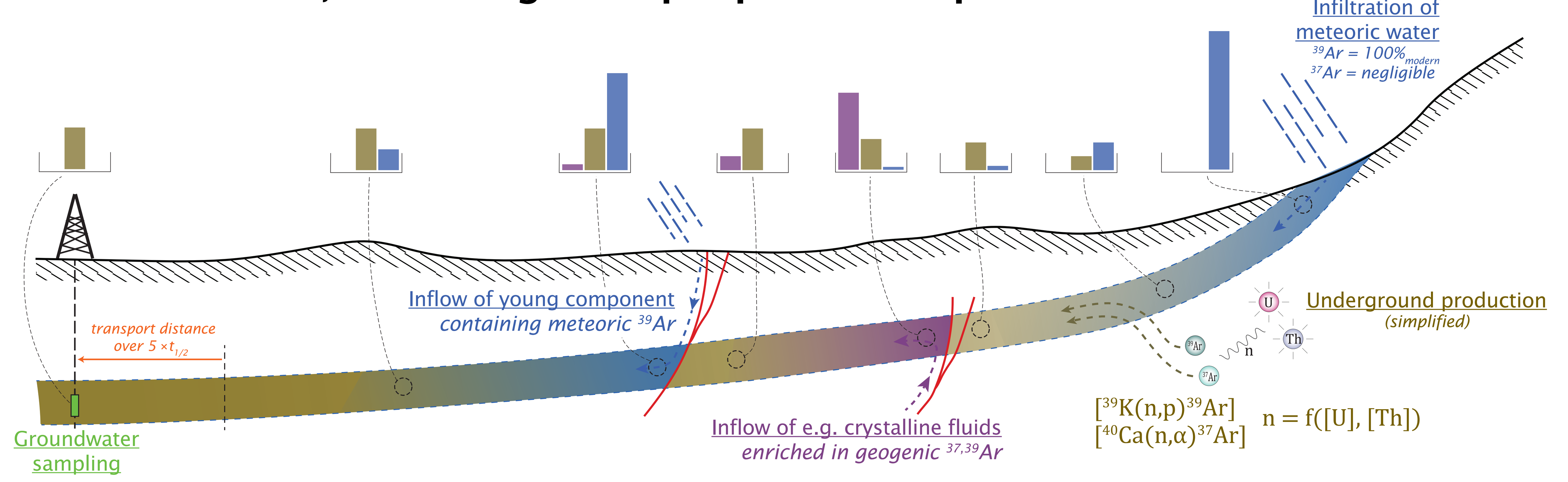


Fig. 1: *Schematic evolution of $^{37,39}Ar$ in a groundwater with a groundwater age of >5 ka at the sampling well. Compositional diagrams on top show ^{39}Ar only.*

The $^{37,39}Ar$ activity of infiltrating meteoric water is modified along the groundwater flowpath by different sources ⊕ and sinks ⊖. These are:

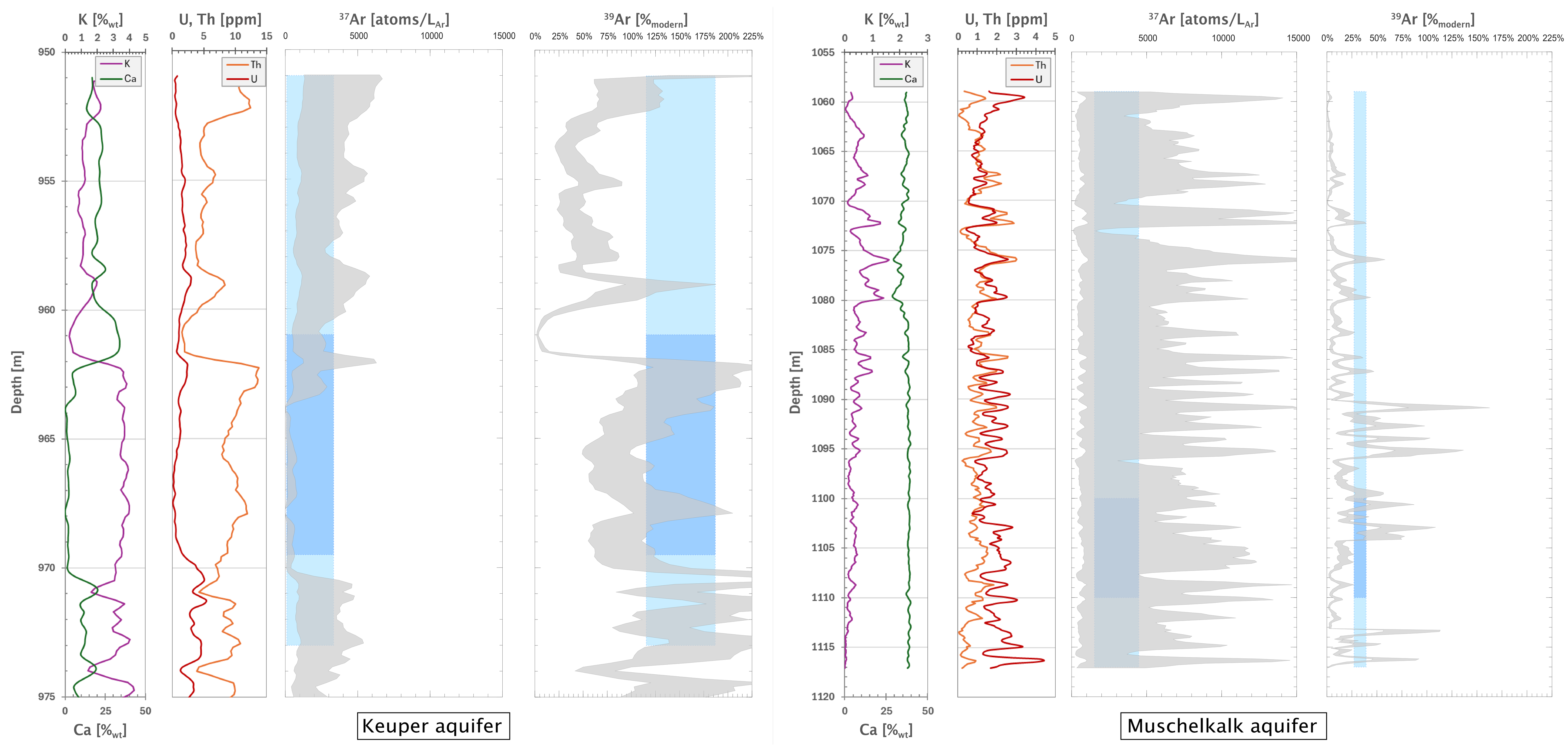
⊕ local production: muon induced[3] (shallow, not depicted) and neutron capture processes[1,4] (deep) as function of geochemistry (mainly U, Th, K, Ca) and material specific emanation factor λ.

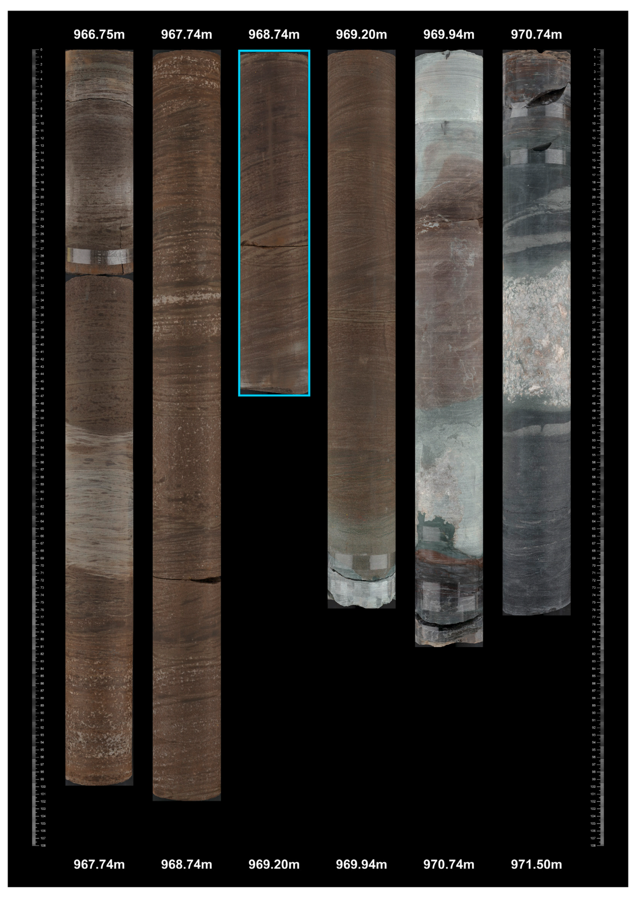
⊕ inflow of external fluids enriched in $^{37,39}Ar$, e.g. young meteoric water with cosmogenic ^{39}Ar or $^{37,39}Ar$ enriched crustal fluids from U, Th rich source regions with high neutron flux.

⊖ radioactive decay with 97% of $^{37,39}Ar$ decaying over $5 \times t_{1/2}$ (≈ 0.5 and 1400 years, respectively).

⟶ Any significant $^{37,39}Ar$ activity in the sample must either be in-situ produced or have entered the groundwater within a distance less than its transport length over $5 \times t_{1/2}$, determined by its flow velocity (based on the aquifers porosity, hydraulic gradient and conductivity).

Profiles of in-situ produced ^{37}Ar, ^{39}Ar equilibrium activity





Keuper aquifer (core photo above):
- Dolomitic/evaporitic strata; likely inflow zone: marls to sandstones
- Estimated flow distance ($5 \times t_{1/2}$): ^{37}Ar = <3 cm; ^{39}Ar = < 70 m

Muschelkalk aquifer:
- Dolo-/limestones (below 1080 m)
- Estimated flow distance ($5 \times t_{1/2}$): ^{37}Ar = <20 cm; ^{39}Ar = < 600 m

Fig. 2: *Accumulation of in-situ produced $^{37,39}Ar$ in groundwater across the Keuper and Muschelkalk aquifer (example: STA2-1 borehole)*
Grey shaded area: calculated $^{37,39}Ar$ equilibrium activity in the groundwater assuming λ of 3-6% (^{39}Ar), 0.4-2% (^{37}Ar) in the Keuper and 0.5-1% (^{39}Ar), 0.07-1% (^{37}Ar) in the Muschelkalk).
Blue shaded area: groundwater sampling interval (light blue) and likely inflow zone (dark blue); the width indicates the range of measured $^{37,39}Ar$ activity in the groundwater.

Calculated ranges of $^{37,39}Ar$ secular equilibrium activities overlap with measured groundwater concentrations in the depth intervals where groundwater flow is most likely. Based on ^{81}Kr model ages and ^{14}C data, the STA-2 Keuper groundwater infiltrated during the middle Pleistocene warm period, while the Muschelkalk groundwater infiltrated during the late Pleistocene cold period[5] and both are therefore free of cosmogenic ^{39}Ar. Based on flow velocities, the distances from the sampling location within which a disturbance of the in-situ production/decay equilibrium activity would still be detectable are in the range of tens of cm (^{37}Ar) to <1 km (^{39}Ar). These distances refer to crustal or meteoric fluid inflow as well as to cross-formation flow between different aquifers. As such, the measured $^{37,39}Ar$ activities can entirely be attributed to subsurface production within the Keuper and Muschelkalk aquifers and no external contribution via cross-formation flow is indicated within less than 1 km from the borehole.

Conclusions and outlook

- Measured $^{37,39}Ar$ activities in Keuper and Muschelkalk groundwater can be explained entirely by in-situ production in the aquifer rocks at specific depths within the expected groundwater flow depth interval.
- Addition of $^{37,39}Ar$ to the groundwater via young meteoric water or fluids evolving in lithologies with higher neutron flux (e.g. crystalline basement) is an implausible explanation for the measured activities because of the short distances (particularly for ^{37}Ar) over which such signatures would be maintained due to the short half-lives of $^{37,39}Ar$.
- Combining modern calculation methods for $^{37,39}Ar$ in-situ production with highly depth resolved geochemical and petrophysical data allows to identify zones with high in-situ production in such heterogeneous strata.
- Further research to better constrain the important lithology specific emanation factor λ is needed.

References

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