

Investigations on water regimes and transport processes of substances in waste and mining dumps and natural aquifers by environmental isotopes – applying ^{85}Kr as an alternative isotope in hydrogeology.

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Introduction

Environmental isotopes in hydrogeology become more and more essential to investigate, estimate and determine water regimes and transport processes of substances below and on top of the Earth's surface. Knowledge about this is important if human intervention in such natural systems is already or will not be negligible anymore. These questions become increasingly relevant as we continue to predict the limits of groundwater resource availability in future. As well as for natural regimes that applies to waste and mining dumps, mine cavity systems, and others.

Isotope analysis therefore complements geochemistry and physical hydrogeology. The stable isotope composition of water can be modified, for instance, by meteoric processes. In a particular recharge area this results in a characteristic isotopic signature of the groundwater, what in turn serves as an indicator for its provenance. On the other hand radioactive decay provides the possibility to put up the 4th, i.e. the time dimension on groundwater regimes, and thus groundwater renewability. Furthermore we can get informations about groundwater quality, geochemical evolution, recharge processes, origin of salinity and contaminant processes.

This work focuses on radioactive isotopes, i.e. on ^{85}Kr in comparative investigation with Tritium, and others. Aim is to develop and provide the theoretical and practical frame to apply ^{85}Kr analysis in commercial projects, where geo-engineering companies, public authorities, or other deal with various hydrogeological problems.

Basics about Krypton and its isotope ^{85}Kr

The percentage of Krypton in the atmosphere is ca. $1,1 \times 10^{-4}$ Vol% [1]. Estimations for the steady-state natural inventory of ^{85}Kr result in $5,18 - 8,8 \times 10^{11}$ Bq [2/3], a value about 6 orders of magnitude lower than the actual activity, and thus the natural background is extremely low (1950: 18 ± 9 dpm/mmol Kr [4]). Natural ^{85}Kr production is caused by (n, γ)-reactions of cosmic ray neutrons with the stable ^{84}Kr isotope in the atmosphere as well as spontaneous fission of uranium and thorium in the lithosphere.

It can be assumed, that the atmosphere is the main reservoir for ^{85}Kr , where its level is primarily controlled by decay and anthropogenic emission rates associated with fission based power generation, especially from nuclear reprocessing plants.

In the Northern hemisphere a meridional trend is apparent as the $^{85}\text{Kr}/\text{Kr}$ value decreases slightly from north to south. At the intertropical convergence zone it suddenly drops [5]. For the Southern hemisphere a more homogenous distribution is observed [6].

[1] GRAEDEL & CRUTZEN (1994); [2] DIETHORN & STOCKHO (1972); [3] SUZUKI & INOUE (1972); [4] ROZANSKI & OSTROWSKI (1977); [5] WEISS et al. (1982); [6] FRITZ & FONTES (1986)

A slight altitude dependence has been measured by TELEGADAS & FERBER (1975) [7] as well as WEISS et al. (1982) [5]. This is a simple effect of: Gravitation, because compared to other air components Kr is more heavier; Kr is permanently washed out by precipitation (to a small amount) [8]. Furthermore the density of air, and thus the number of Kr atoms decreases with altitude. So the total activity falls slightly.

Seasonal variations has been detected in a range of 40 to 80 dpm/mmol [5]. Most likely it is connected with the seasonal variations in the vertical mixing of the atmosphere [5].

Krypton is inert and does not engage in chemical reactions under normal conditions. Existing isotope fractionation processes are below detection limit and thus are out of importance. Diffusion of Krypton into the soil is less effective [9]. Therefore the $^{85}\text{Kr}/\text{Kr}$ input function for the atmosphere also applies to groundwater and does not depend on sample quantities (equilibrium). The solubility of krypton in water amounts to 8 Vol% of the atmospheric Krypton.

As the input function rises monotonous and global atmospheric mixing is relatively quick (1,0 – 1,7 a [5/10]) the ^{85}Kr -atmosphere-hydrosphere system seems to be more or less simple. So unlike with Tritium and ^{14}C interpretation of ^{85}Kr values is never ambiguously. The half life time of ^{85}Kr is 10,76 a, and thus it can exchange Tritium (12,43 a) from its major position in futur. But it can also serve as a complement in multitracing studies.

NITZSCHE & HEBERT (1993) [11] have applied the following input function for Germany on measurement values between 1958 - 1992:

$$C_{\text{in}}(t) = A_1(t) - A_2(t^2) + A_3(t^3) - A_4(t^4); [t = 0 \text{ for January 1958}];$$

$$A_1 = 5,775; A_2 = 0,2057; A_3 = 0,003943; A_4 = 2,5 \times 10^{-5}$$

Disadvantages:

Unfortunately ^{85}Kr analysis requires large sample quantities (at least 1 m³). The expenditure for measuring devices and other equipment is high, and thus the costs. One goal therefore is to reduce the costs.

There is some more... in work!

[7] TELEGADAS & FERBER (1975); [8] ROZANSKI (1979a); [9] NCRP (1975); [10] GUDKOV et al. 1976; [11] NITZSCHE & HEBERT (1993)

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