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Uranium-series disequilibrium studies of drillcore Km3 from the Kamlunge test-site, northern Sweden

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ABSTRACT. Studies of the U decay series (^{238}U–^{234}U–^{230}Th) have been carried out on samples from unaltered bedrock and highly altered fracture/crush zones from drillcore Km3 (Kamlunge test-site). The fracture zones are characterized by abundant iron oxide coatings (hematite and hydroxy iron oxides) resulting from the passage of hydrothermal solutions coeval with the Lina granite intrusion. Enrichments of U and Th, thought to be due to co-precipitation (or preferential sorption) processes together with the iron-oxides, are also present.

The isotopic results show that out of a total of twelve rock samples measured, six indicate isotopic disequilibrium mostly due to unequal depletions of ^{234}U and ^{238}U; one near-surface sample indicated some minor assimilation of U. The major fracture zones generally indicate removal of total U. This has resulted from interaction with groundwaters which are still marginally oxidizing, even at depths of 375 m.

Isotopic disequilibrium has occurred within recent geological times, i.e. during the last 0.5 Ma as imposed by the half-lives of ^{234}U and ^{230}Th. In terms of radioactive disposal considerations, the results are important in that: (1) the investigated bedrock environment (100–500 m) is generally reducing; however (2) there is some evidence to indicate that rock/water interactions, leading to the removal of total U, have resulted from the presence of less reducing groundwaters within those large-scale fracture/crush zones which intersect the bedrock surface.

KEYWORDS: uranium, disequilibrium, radioactive waste, Kamlunge, Sweden.

THE Kamlunge test-site is situated in northern Sweden about 35 km from the Baltic Sea coast (fig. 1). It represents one of several sites in Sweden which have recently been investigated as potential repositories for radioactive waste disposal (Ahlbom *et al.*, 1980; Tirèn *et al.*, 1981; Albino *et al.*, 1982; Carlsten *et al.*, 1982). In common with the other areas, Kamlunge represents a crystalline rock environment which is Precambrian in age. It has been investigated in detail, both geologically (Albino *et al.*, 1983) and hydrologically (Danielson, 1983), this being facilitated by the diamond drilling of 16 drillholes (down to a maximum depth of 600 m) complemented by twenty-two percussion drillholes (down to a maximum depth of 150 m). Groundwater sampling was carried out at various levels from four holes; surface-water sampling from lakes, streams, and springs was carried out to evaluate the recharge environment.

This paper describes part of a larger programme of study which involves U-series disequilibrium studies of both surface and groundwaters, together with the geological bedrock through which the waters permeate. Presented here are some preliminary results from drillcore Km3.

U-series disequilibrium studies have now become a fairly standard investigative method within the field of geochemistry. Pioneered by Cherdynstev (e.g. 1955, 1971) the methods have been subsequently improved and applied by many workers to a wide range of geological problems; see, for example, Rosholt (1959); Thurber (1962); Rosholt *et al.* (1963); Koide and Goldberg (1965); Rosholt *et al.* (1966); Kigoshi (1971); Kronfeld (1974); Osmond and Cowart (1976); Fleischer and Raabe (1978); Ivanovich and Harmon (1982); and Rosholt (1983).

For crystalline rocks, such as those described in this paper, the U-series disequilibrium system has been used as a sensitive indicator of recent U mobility during the last 0.5 Ma (Zielinski *et al.*, 1981; Schwarcz *et al.*, 1982; Smellie, 1982; Rosholt, 1983; Smellie and Rosholt, 1984). To the extent that the U-series disequilibrium is a product of rock/water interaction, these measurements may help to evaluate the effective permeability of unfractured and macroscopically fresh rocks, and they can be used to assess geologically recent U mobility in 'sealed' fracture zones.

Geological setting

The regional geology of the area (fig. 1) has been described generally by Ödman (1957) and Lundqvist (1979), and in more detail by Albino *et al.* (1983). The region is characterized by metamorphic and migmatitic rocks which are Precambrian in age; the region is regarded as having been relatively stable for long periods of geological time. Briefly, the oldest rocks in the area are basement Archaean granite gneisses. In Finland and further north in Sweden, similar basement rocks reveal ages ranging from 2600-2800 Ma (Welin *et al.*, 1971; Perttunen, 1980). Unconformably overlying the Archaean basement are the younger Svecokarelian rock groups. The Svecokarelian is initially represented by metasediments and metavolcanics. These are followed by the Haparanda series of deep intrusives comprising granodiorite and gabbro which have been dated by Rb-Sr to around 1840 Ma (Welin *et al.*, 1970). The earlier metasediments and metavolcanics are thus considered to be within the range of 1900-2500 Ma. Metamorphism in the region culminated during the Svecokarelian epoch at approximately 1800 Ma resulting in widespread migmatitization. Coeval with metamorphism, and forming large-scale intrusions in the region, is the Lina granite which indicates a range of Rb-Sr ages from 1565-1800 Ma (Welin *et al.*, 1970).

Indicated in fig. 1 is the location of the Kamlunge area. Topographically, the area forms a plateau with its highest point some 175.2 m above sea-level; ice-movement has imposed a dominant NNW-SSE orientation on the local topographic features. The plateau may be described as a horst-type feature, bounded on three sides by large-scale regional lineaments which are presumed to be near vertical in orientation. Outcrop exposure is generally good, otherwise a thin to moderate moraine cover is usual (approx. 5-10 m).

Hydrologically, the investigated plateau region represents part of the groundwater recharge area; groundwater discharge outlets occur mostly along the regional lineament features, forming lakes and marsh areas.

The oldest rocks exposed at the Kamlunge test-site area are the metasediments and metavolcanics. These are present as quartzitic gneiss, biotite gneiss, and amphibolite which are later intruded by granodiorite and gabbro of the Haparanda series. The Lina granite, with widespread pegmatitic activity, intrudes all older rock types.

Large-scale lineaments measured in an area of approximately 600 km^2 around Kamlunge revealed three main groups characterized by orientation. The most important group was orientated parallel to the dominant ice-movement direction (N. 40° W.-S. 40° E.); the remaining more minor trends were N. 10° E.-S. 10° W. and N. 30° E.-S. 30° W.

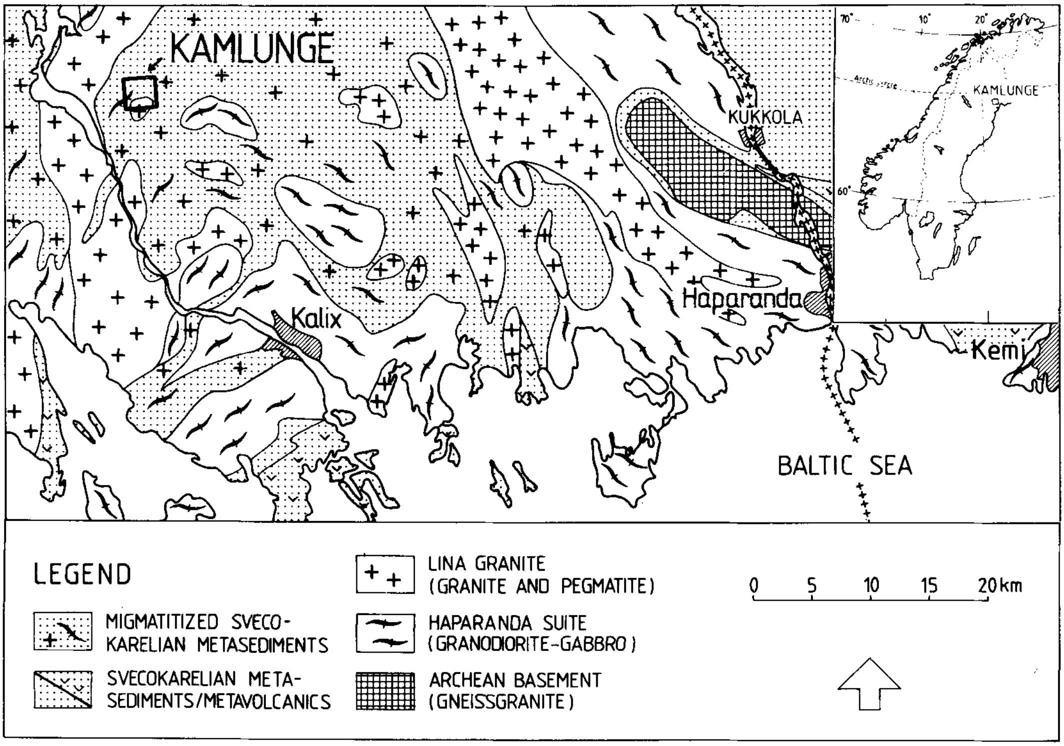

FIG. 1. Regional geological setting of the Kamlunge test-site area, northern Sweden (modified after Albino *et al.*, 1983).

respectively. Detailed fault and joint patterns have been measured from the exposed bedrock at the Kamlunge test-site. Major crush zones have been indicated by different geophysical methods, and have suggested the presence of seven zones ranging in width from less than 5 to 15 m which traverse the test-site area. Several of these have subsequently been confirmed from the drilling programme.

Description of drillcore Km3

Drillcore Km3 was chosen for examination because it intersects two of the major fracture/crush zones and one minor crush zone revealed at the bedrock surface by geophysical investigations. In addition, the intersection points were sampled for groundwaters thus providing a suitable opportunity to directly relate the water chemistry with the fracture mineral chemistry within the bedrock.

Drillcore Km3 (fig. 2) is 700.15 m long and represents a vertical depth of 580 m. In terms of rock composition, 42.9% of the intersected bedrock is composed of granodiorite, 25.3% granite/pegmatite, 22.2% amphibolite, and 9.6% biotite gneiss. With regard to fracture density, granodiorite recorded 4.94 fractures/m, granite 4.76, amphibolite 4.32, and biotite gneiss 3.54. In general, pegmatite horizons are most fractured. Effective porosity measurements of individual rock units using methods modified after Collett (1959) (S. Sehlstedt, 1983, pers. comm.), show a generally low porosity (< 0.3%). However, significant differences do exist between the different rock units, for example, amphibolite (mean 0.26%) records slightly higher values than biotite gneiss (mean 0.23%) and granite/pegmatite (mean 0.22%) but significantly higher than granodiorite (mean 0.13%).

Rock composition. The upper 400 m length of the drillcore (fig. 2) is dominated by massive, often foliated, granodiorite. In hand specimen it is generally porphyritic in texture, grey in colour and fine- to medium-grained, consisting of large plagioclase grains (2–5 mm) set in a biotite-rich matrix. Microscopically, the rock is composed of 20–25% mafic constituents (mainly biotite and hornblende) together with quartz, plagioclase, and potassium feldspar (mostly microcline). Subordinate amounts of epidote (clinozoisite), sphene, apatite, magnetite, and pyrite are also present with accessory zircon. The unaltered rock shows little microscopic evidence of alteration apart from some minor sericitization of the feldspars and some chlorite from biotite breakdown. Below 400 m and down to 660 m, the main rock type is amphibolite (metavolcanic) which is mainly fine-grained, often foliated, and dark green in colour. Microscopically, the rock is equigranular with up to 50% mafic phases. The main mineral phases are quartz, plagioclase, hornblende, and biotite with subsidiary amounts of sphene and magnetite; accessory apatite and epidote (clinozoisite) are also present. Epidote is of local importance occurring as bands and vein infillings. Within the amphibolite are horizons or schlieren of coarse-grained material rich in quartz, plagioclase, potassium feldspar (microcline now partly albitized), and biotite with accessory amounts of zircon and apatite. Below 660 m strongly foliated biotite gneiss occurs which is dark-grey in colour and fine- to medium-grained in texture. Microscopically, the main constituents are quartz, plagioclase, potassium feldspar, and biotite ± amphibole (hornblende) with subordinate epidote (clinozoisite) and magnetite; apatite occurs in accessory amounts. Localized areas are rich in epidote and/or quartz and feldspar.

Occurring within all rock-types are intrusive horizons of medium- to coarse-grained pegmatite/granite. Granite horizons with pegmatite veins can be up to 10 m in thickness; the pegmatites are generally 1–2 m in thickness. The granite is commonly red in colour characterized by quartz, potassium feldspar (microcline), plagioclase with subordinate biotite, and accessory amounts of zircon, apatite, sphene, magnetite, and allanite. The pegmatite, both red and grey varieties, tends to be typically coarse-grained and composed dominantly of potassium feldspar (mostly microcline) and quartz with subsidiary plagioclase, muscovite and biotite, and minor amounts of magnetite. Uranothorite has also been observed.

Alteration features of the fracture/crush zones. Drillhole Km3 intersects several open fracture/crush zones; of these, Zones 2, 3, and 4 (fig. 2) can be correlated to the bedrock surface. These zones are observed as irregular thicknesses of crushed rock together with moderately to highly fractured rock. Losses of drillcore material within the crush zones are quite common. These zones exhibit some common features of alteration which can be generally summarized as follows:

(*a*) Moderate to strong alteration of feldspar to sericite; complete pseudomorphing is not uncommon.

(*b*) Breakdown of biotite ± hornblende to chlorite; pseudomorphing partly by calcite is also observed.

(*c*) Martitization of magnetite to hematite.

(*d*) Strong Fe^{3+} (hematite + hydrous iron oxides) penetration along fractures and within the adjacent rock matrix forming impregnations and coatings.

(*e*) Stringers and small concentrations of anatase/leucoxene as end-products of magnetite and biotite breakdown.

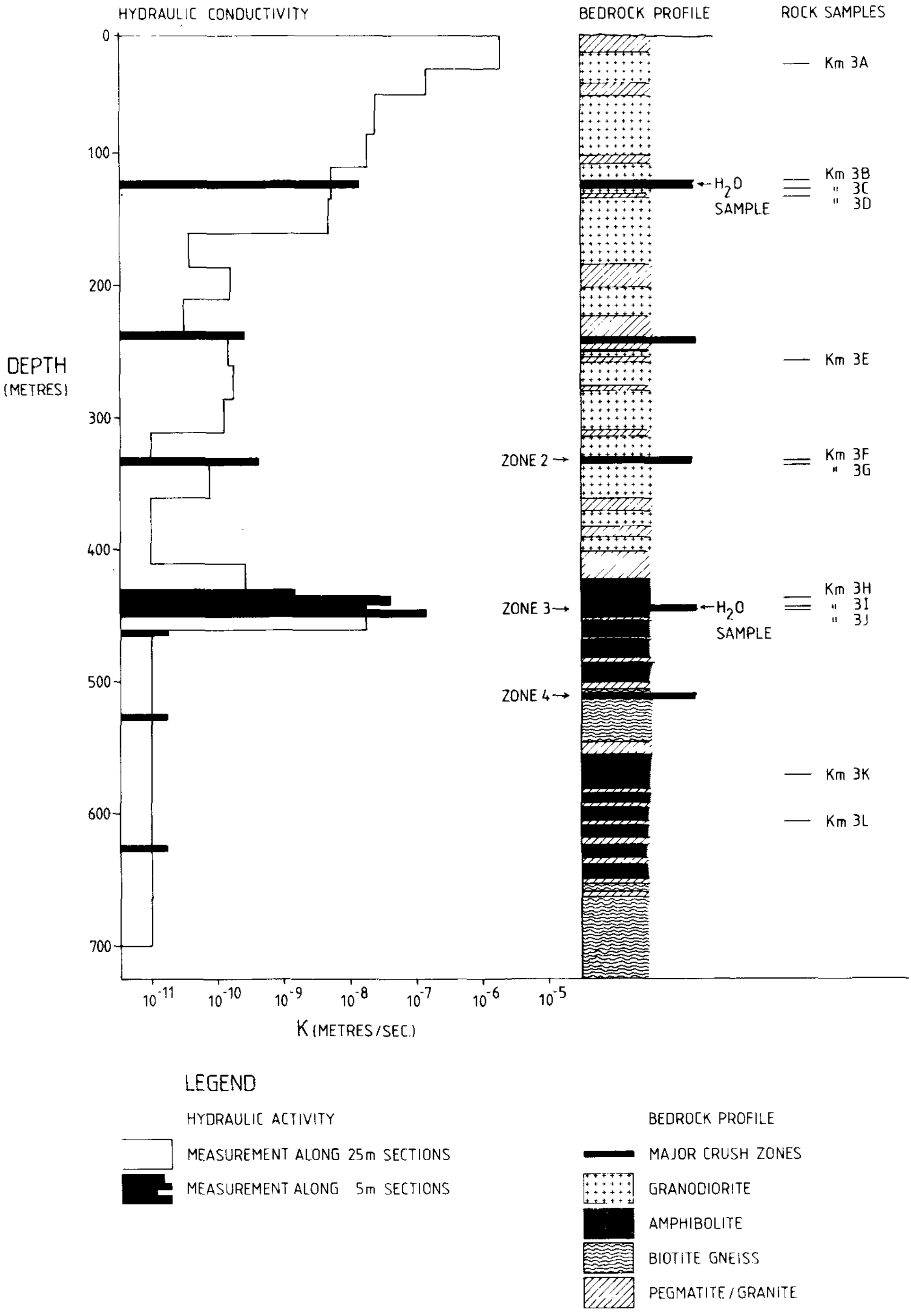

FIG. 2. Some hydrogeological features of drillcore Km3 (Kamlunge test-site) showing specimen locations for rock and groundwater samples.

Common to all rock-types is the presence of fractures containing iron-oxide in the form of hematite and hydrous iron oxides. According to Tullborg and Larson (pers. comm., 1983) the greatest percentage of iron-oxide fractures occur in the granite/pegmatite (70%), followed by quartz/biotite gneiss (25%), granodiorite (3%), and amphibolite (2%). The iron oxides are present as coatings along the fracture margins and around matrix minerals and their pseudomorphs. Hematite also occurs as a fracture/vein infilling constituent and as an impregnation within the rock matrix peripheral to the fractures. Especially in the amphibolite, hematite-rich fractures occur in association with hydrothermal phases such as epidote. Some of these iron-oxide concentrations are considered by the author to be the result of hydrothermal activity probably in association with the intrusion of the Lina granite which dominates the region (fig. 1).

Later veinlets devoid of iron oxide and consisting of low-temperature calcite ± chlorite with minor amounts of quartz (± zeolite), occur parallel and obliquely to the earlier iron-oxide dominated fracture patterns.

Sampling procedures and analysis

Rock specimens for U-series disequilibrium studies were selected as follows: (*a*) representative macroscopically fresh samples from all rock types at varying depths; (*b*) fractured samples, especially from the more major fracture/crush zones which extend to the bedrock surface; and (*c*) fractured samples from those horizons sampled for groundwater.

Samples consisted of approximately 30 cm lengths of drillcore, and for the major fracture zones up to three specimens were selected (centre of fracture/crush zone, margin of zone, and one from just within the adjacent unaltered bedrock). Location of the samples are shown in fig. 2. Points chosen for goundwater sampling were based on detailed drillcore mapping and correlation with hydraulic conductivity measurements carried out every 25 m along the drillhole (fig. 2); lengths of promising potential were subsequently tested every 5 m (Danielson, 1983). From fig. 2 it can be seen that the points of highest hydraulic conductivity not surprisingly coincide with the major fracture/crush zones. The generally high conductivity values for the upper 150 m is fairly typical for the Swedish bedrock. This is due to a high concentration of tensional joints/fractures resulting from adjustments subsequent to the removal of the ice overburden in recent Pleistocene times.

From each rock sample a thin slice was first removed from the length of the drillcore piece, for mineralogical and fission-track studies. The remainder was crushed and portions sent for isotopic analysis using high resolution alpha spectrometry at the Department of Radiation Physics, University of Lund, Sweden, and the USGS in Denver, USA. At Denver, details of sample preparation are according to Rosholt (1980, 1983). The quoted errors (Table I), better than ±2% for the ^{234}U/^{238}U ratio and ±3% for the ^{230}Th/^{234}U and ^{230}Th/^{238}U ratios, are due to counting statistics only. At Lund, general details of sample preparation are outlined by Duniec *et al.* (in prep.). Errors (2σ) for the ratios ^{234}U/^{238}U, ^{230}Th/^{234}U, and ^{230}Th/^{238}U are presented in Table I.

The remaining drillcore portions were analysed for U (delayed neutron activation) and Th (sealed-can gamma spectrometry). Fission-track analyses according to the method of Kleeman and Lovering (1967) were carried out on polished sections from all samples to assess the location and distribution of uranium.

Water samples, normally 5 l in volume, were collected from surface and near-surface localities in the vicinity of borehole Km3; groundwater samples were collected at vertical depths of approximately 106 and 375 m from the borehole (fig. 2). All samples were immediately filtered (0.45 μm pore diameter) and treated with sufficient hydrochloric acid to attain a pH of 1.0. U recovery and analysis were carried out at the University of Lund.

Results and discussion

Radioelement distribution in the Km3 bedrock profile. The results are presented in Table I; sample locations are given in relation to both the drillcore length and the vertical depth. U concentrations within the three fractured zones (ranging from 2.3 to 9.7 ppm U) shows a significant departure from the typical background values of the petrographically unaltered host bedrock (ranging from 0.7 to 1.9 ppm U); this is especially marked within the fracture/crush zones located at 122 and 336 m (fig. 2). Fission-track studies show that the background activity recorded for the unaltered bedrock samples are due to low U concentrations (10–30 ppm) associated mostly with subordinate to accessory amounts of zircon, apatite, epidote, and sometimes magnetite. Within the altered fracture zones, U (< 5–30 ppm) is associated with iron-oxides and very small anatase/leucoxene aggregates. No significant U is associated with later-stage calcite and chlorite (± zeolite) infillings.

Increases of Th within the fractured zones (especially marked in samples Km3F and Km3G which show contents of 83.6 and 312 ppm Th and ^{230}Th/^{232}Th activity ratios of 0.113 ± 0.016 and

Table 1: Uranium and thorium contents and isotopic activity ratios from drillcore material, and uranium contents and activity ratios from associated surface- and ground-waters. Drillcore Km3, Kamlunge test-site, N. Sweden.

Sample	Rock-type	Sample Location	Length along drillcore (metres)	Vertical Depth (metres)	Uranium (ppm)	Thorium (ppm)	Th/U	$^{234}U/^{238}U$	$^{230}Th/^{238}U$	$^{230}Th/^{234}U$	Equilibrium State	Groundwaters	
												U-content (ppb)	$^{234}U/^{238}U$
				Surface Water								0.264 ± .068	1.74 ± .34 *
				Spring Water								0.147 ± .036	1.82 ± .32 *
				Borehole 12 m								27.3 ± 4.1	2.53 ± .06 *
Km3A	Granodiorite	Compact bedrock	33.4	29.20	0.9	3.3	3.7	1.140 ± .220	0.922 ± .155	0.810 ± .130 *	-		
Km3B	Granodiorite	Centre of crush zone	121.50	105.50	9.7	4.4	0.5	0.992 ± .080 0.995 ± .020	0.999 ± .097 1.009 ± .030	1.007 ± .098 * 1.014 ± .030 §	Equilibrium Equilibrium	18.91	2.75 *
Km3C	Granodiorite	80 cm below crush zone	122.80		4.1	3.3	0.8	1.080 ± .128 1.007 ± .020	1.171 ± .148 0.990 ± .030	1.150 ± .145 * 0.984 ± .030 §	- Equilibrium		
Km3D	Granodiorite	Compact bedrock	126.00	109.00	1.0	2.9	2.9	1.080 ± .350 1.006 ± .020	0.900 ± .270 0.987 ± .030	0.830 ± .240 * 0.981 ± .030 §	Equilibrium Equilibrium		
Km3E	Pegmatite	Compact bedrock	260.00	222.30	23.1	14.6	0.6	0.933 ± .076	0.969 ± .119	1.039 ± .129 *	Equilibrium		
Km3F	Granodiorite	Margin of crush zone	336.70	286.50 (Zone 2)	3.4	83.6	24.6	0.943 ± .091	1.611 ± .140	1.709 ± .152 *	-		
Km3G	Granodiorite	Centre of crush zone	336.80		9.5	312	32.8	1.033 ± .041	1.510 ± .149	1.462 ± .144 *	-		
Km3H	Amphibolite	Compact bedrock	438.15	369.80	1.9	4.8	2.5	1.050 ± .290	1.040 ± .240	0.990 ± .230 *	Equilibrium		
Km3I	Amphibolite	Centre of crush zone	444.25	375.00 (Zone 3)	2.3	12.6	5.5	0.983 ± .117 1.001 ± .020	1.090 ± .135 1.160 ± .030	1.110 ± .138 * 1.159 ± .030 §	Equilibrium -	3.46	2.73 *
Km3J	Amphibolite	Centre of crush zone	444.55		3.2	13.9	4.3	1.162 ± .149	1.187 ± .179	1.021 ± .150 *	-		
Km3K	Amphibolite	Compact bedrock	582.30	488.00	0.7	2.9	4.1	1.080 ± .260 0.973 ± .020	0.960 ± .220 0.969 ± .030	0.880 ± .200 * 0.996 ± .030 §	Equilibrium Equilibrium		
Km3L	Pegmatite	Compact bedrock	608.00	508.50	26.3	12.8	0.5	1.007 ± .055	1.217 ± .101	1.209 ± .100 *	-		

* Analyses by the Department of Radiation Physics, University of Lund, Lund, Sweden.

§ Analyses by the U.S. Geological Survey, Denver, U.S.A.

Drillcore uranium analyses (delayed neutron activation) were carried out by Studsvik Energiteknik AB, Sweden. Drillcore thorium analyses (sealed-can gamma spectrometry) were carried out by the Danish Atomic Energy Commission Research Establishment, Risø, Denmark.

0.085 ± 0.016 respectively) generally correlate with U, suggesting that both U and Th have accumulated by sorption or co-precipitation with iron oxide. In contrast, Gascoyne (1982) records a preferential sorption or co-precipitation of U (rather than Th) in hematized fractures from the Eye-Dashwa Lakes pluton near Atikokan, NW Ontario, Canada.

If the observed enrichment of Th within the fracture/crush zones (Zones 2 and 3; fig. 2) relative to the host-rock background values is the result of rock/water interaction, this is surprising considering Th's generally assumed immobility in groundwater. It is more likely that the Th is of primary hydrothermal origin (as a co-precipitation with iron oxide and U) which accompanied the intrusion of the Lina granite.

Within the limits of analytical error, the $^{234}U/^{238}U$, $^{230}Th/^{238}U$, and $^{230}Th/^{234}U$ activity ratios indicate equilibrium or near-equilibrium in six out of a total of twelve analysed samples (Table I). For the two inter-laboratory analysed samples not in agreement, i.e. Km3C and Km3I, the Denver laboratory values are preferred because of their greater counting precision. Isotopic equilibrium indicates that little or no migration of the isotopes has occurred in these six samples over a period of at least 1.25 Ma, as required by the half-lives in the decay sequence ^{238}U-^{234}U-^{230}Th. Of the remaining six samples recording disequilibrium, unequal depletions of ^{234}U and ^{238}U rather than additions of ^{230}Th which is generally assumed to be immobile in natural waters, would mostly account for the observed deviations. Disequilibrium in the $^{230}Th/^{234}U$ system, which is evident in five out of the six samples, indicates a disturbance within the last 0.35 Ma.

Isotopic equilibrium is indicated in three granodiorite samples (Km3B, Km3C, and Km3D), one of which (Km3B) represents the upper fracture zone at 122 m. This contrasts with the uppermost granodiorite sample (Km3A) at 33.40 m which indicates assimilation of both ^{238}U and ^{234}U and the fracture/crush zone at 336 m (Km3F, Km3G; Zone 2) which indicates depletions of ^{234}U and ^{238}U. At deeper levels, within the amphibolite rock-type, the two petrographically fresh samples (Km3H and Km3K) exhibit isotopic equilibrium. Samples Km3I and Km3J, representing the major fracture/crush zone at 444 m (Zone 3), show a loss of total U.

The pegmatite samples (Km3E and Km3L) which are macroscopically fresh but petrographically altered, exhibit both isotopic equilibrium (Km3E) and disequilibrium (Km3L), the latter resulting from depletions of both ^{234}U and ^{238}U. This mobilization of uranium in Km3L is supported by fission-track studies which show significant interstitial U present within the rock matrix. In addition, primary U- and Th-bearing phases (e.g. uranothorite) exhibit marked evidence of alteration and leaching.

Microscopically, the three open fracture/crush zones intersected by drillhole Km3 (fig. 2) show extensive rock/water alteration characteristics. As described above, U-series disequilibrium is indicated from Zones 2 and 3 in contrast to the uppermost zone (122 m) which appears to have remained relatively undisturbed for at least 1.25 Ma. It is known through geological investigations that Zones 2 and 3 extend to the bedrock surface; the orientation and extent of the upper zone is unknown. It is therefore reasonable that Zones 2 and 3, both of which are characterized by a strong positive piezometric pressure (Danielson, 1983), facilitate the downward movement of groundwater. This has resulted in rock/water interaction and hence isotopic disequilibrium along and peripheral to the fractures. The absence of disequilibrium in the 122 m fracture is therefore unexpected and may reflect insensitive sampling. Here, in contrast to Zones 2 and 3, the fracture/alteration areas are more confined to narrow zones. Samples most likely include sufficient adjacent bedrock material at isotopic equilibrium to mask any disequilibrium effects resulting from rock/water interaction along these fine fracture channels. Future studies of this type could be improved by the leaching of mineral coatings from fracture mineral separates in order to achieve a better sensitivity of measurement.

U chemistry of waters collected from the Km3 bedrock profile. Surface, near-surface, and groundwaters have been sampled in the Kamlunge area. Borehole sampling procedures and the equipment used are described by Almén *et al.* (1983) and Wikberg *et al.* (1983). The chemical analyses of the Kamlunge waters are still not complete at this juncture, and will be eventually reported elsewhere. However, available ^{3}H and ^{14}C data indicate that groundwater mixing is widespread, resulting in a high near-surface component. This is reflected in the tritium contents which range from 37 to 56 TU in Zone 3 (vertical depth 375 m) and 35 to 49 U from the uppermost fracture zone (122 m; vertical depth 105.50). These can be compared to surface water samples which record 30 to 41 TU. ^{14}C ages of the groundwaters collected from the two zones ranged from 2985 to 3575 years old and probably also reflect dilution from younger waters.

Dissolved U contents and $^{234}U/^{238}U$ activity ratios for the sampled waters are presented in Table I. The recharge environment is characterized by surface and near-surface waters containing low amounts of dissolved U (0.147–0.264 ppb) and activity ratios which indicate isotopic disequilibrium (1.74–1.82). At a depth of 12 m, in the vicinity of borehole Km3, the groundwater collected from a percussion drillhole contained up to 27.3 ppb U (activity ratio 2.53). Groundwaters sampled over a period of 22 days from the upper fracture zone (vertical depth of 105.50 m) are also rich in dissolved U (mean of 18.91 ppb; activity ratio mean of 2.75), especially when compared to Zone 3 (mean of 3.46 ppb; activity ratio 2.73). The moderately high ^{234}U/^{238}U activity ratios indicated by all the sampled waters thus show widespread disequilibrium within the hydrological environment due to excess ^{234}U. The decrease in dissolved U content between the two sampled levels suggests a greater reducing environment with increasing depth. Such changes in groundwater composition, accompanied by increasing isotopic disequilibrium due to excess ^{234}U, are well documented in the literature (e.g. Osmond and Cowart, 1976). However, the sampled groundwaters at Kamlunge contain significantly high amounts of dissolved U, even at a depth of 375 m. This indicates that true reducing conditions, which are normally reflected by extremely low U contents (usually less than 0.1 ppb), are therefore not present, and that the groundwaters at the deepest level sampled (Zone 3) are still marginally oxidizing. This means that the relatively high ^{234}U/^{238}U activity ratios are still mostly attributable to preferential chemical etching of ^{234}U, rather than solid to solution recoil build-up processes.

It is proposed that near-surface groundwaters, which will be relatively oxidizing in type, are picking up U with preferential chemical etching of ^{234}U dominating over solid-solution recoil build-up processes. As the pegmatite/granite dominates within the Kamlunge area, is usually fractured, and is known to contain labile U, the waters soon achieve high (saturation?) levels of dissolved U and appreciable amounts of excess ^{234}U at fairly shallow depths. At deeper levels, exemplified by Zone 3, the groundwater is less oxidizing and the dissolved U content has correspondingly decreased. With its high mafic content (i.e. high Fe^{2+} availability), the amphibolite (and to a lesser extent the biotite gneiss) present within the Kamlunge area will greatly contribute to the reducibility of the groundwater environment.

The oxidizing characteristics of the groundwaters are generally supported by the isotopic data from the fracture zones. The removal of total U is particularly evident at the Zone 2 intersection (286.50 m) and less apparent at the Zone 3 intersection (375 m). Moreover, even within the deepest pegmatite sample (Km3L; 508.50 m) the groundwaters appear to have been sufficiently oxidizing to remove ^{238}U and ^{234}U.

According to secondary U accumulation processes as discussed by Osmond *et al.* (1983), the present isotopic disequilibrium data from the fracture zones indicate a mixture of groundwater environments. For example, Km3F (Zone 2) suggests up-flow oxidizing conditions whereupon ^{230}Th exceeds ^{238}U and ^{234}U. Similar observations resulting from the leaching and remobilization of pre-existing accumulations of U have been noted by the above authors. In the Kamlunge context, earlier U (and Th) enrichments are a characteristic feature of the fracture zones under investigation, and thus the fractures can be regarded as having been mineralized in a sub-economic sense. In contrast to Km3F, samples Km3G (Zone 2), Km3I and Km3J (Zone 3), and Km3L from the deepest sampled pegmatite, indicate a non-steady state system which, according to Osmond *et al.* (1983), reflects an environment characterized by U mobilization events and relatively abrupt changes from precipitation to leaching processes. In such cases, ^{234}U is in excess of ^{238}U (reducing down-flow environment) and at the same time deficient in relation to ^{230}Th (oxidizing up-flow environment). The non-steady-state processes suggested from the isotopic data are perhaps not so surprising when considering the large-scale extent of these zones (5–15 m wide) and the fact that they extend to the bedrock surface. Groundwater flow is in all probability facilitated by these zones which will intersect different groundwater flow-paths at various levels as they continue down through the bedrock. This will result in the mixing of groundwater masses of differing origins and chemistries, and may also enable more oxidizing groundwater to locally penetrate to greater depths.

Conclusions

Studies of the U decay system (^{238}U–^{234}U–^{230}Th) show that the macroscopically fresh and unfractured bedrock samples generally appear to be either at or near isotopic equilibrium. Notable exceptions include one near-surface granodiorite sample which has experienced some minor assimilation of U, and a pegmatite sample at a vertical depth of 508.50 m which has undergone a depletion of total U. The most representatively sampled fracture zones (Zones 2 and 3), which have been initially mineralized (U and Th enrichments) by hydrothermal activity during the latter stages of the Lina granite intrusion, generally indicate depletions of ^{238}U and ^{234}U. This has resulted from the interaction of groundwaters still marginally oxidizing, even at depths of 375 m (Zone 3).

The rock/water interaction processes giving rise to the isotopic disequilibrium have taken place within geologically recent times, i.e. during the last 0.5 Ma as imposed by the half-lives of ^{234}U and ^{230}Th. In terms of radioactive waste disposal considerations, the results are important in that (1) the investigated bedrock environment (100–500 m) is generally reducing, however (2) there is some evidence to indicate that rock/water interaction processes leading to the removal of total U has resulted from the presence of less reducing groundwaters within those high-risk areas, namely the large-scale fracture/crush zones extending from depth to the bedrock surface.

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