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Abundance and Distribution of Uranium and Thorium in the Syenite of Piz Giuv (Aar-Massif, Switzerland)

von

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Vorwort der Schweizerischen Geotechnischen Kommission und des Arbeitsausschusses für die Untersuchung schweizerischer Mineralien und Gesteine auf Atombrennstoffe und seltene Elemente

Die vorliegende Arbeit von T. P. Labhart und L. Rybach „Abundance and Distribution of Uranium and Thorium in the Syenite of Piz Giuv (Aar-Massif, Switzerland)" befaßt sich mit dem bekannten Giuvsyenit im östlichen Aarmassiv, einem der radioaktivsten Gesteine der Schweiz. Speziell studiert werden die Thorium- und Urangehalte der verschiedenen Strukturtypen und ihre zonare Verteilung im langgestreckten Gesteinskörper. Die Untersuchung wurde im Rahmen des Programmes des Arbeitsausschusses durchgeführt aus Mitteln des Schweizerischen Nationalfonds, der Schweizerischen Geotechnischen Kommission und einem direkten Bundeskredit. Den Autoren sei für ihren interessanten Beitrag an die wissenschaftliche Erforschung schweizerischer Gesteine auf ihre Gehalte an radioaktiven Elementen vielmals gedankt.

Für den Inhalt des Textes und der Figuren sind die Verfasser allein verantwortlich.

Zürich, den 5. Juli 1971.

Der Präsident der
Schweiz. Geotechnischen Kommission
und des Arbeitsausschusses

Prof. F. de Quervain

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ABUNDANCE AND DISTRIBUTION OF URANIUM AND THORIUM IN THE SYENITE OF PIZ GIUV (AAR-MASSIF, SWITZERLAND)

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ABSTRACT

Labhart, T.P. and Rybach, L., 1971. Abundance and distribution of uranium and thorium in the syenite of Piz Giuv (Aar-Massif, Switzerland). Chem. Geol., 7: 237–251.

The elongated Giuv Syenite (Aar-Massif, Switzerland) contains remarkably high amounts of thorium and uranium. Mean values of 44 samples: 66.0 p.p.m. Th, 22.1 p.p.m. U, Th/U = 3.19. U and Th have, despite the relatively small dimensions (6 × 0.5 km) of the syenite body, a zonal distribution: uranium concentrations increase from a core area with 15.0 p.p.m. towards rim zones with 28.1 p.p.m., whereas the Th/U ratio decreases from 3.68 to 2.75. This zonal arrangement corresponds roughly to the distribution of rock types in the syenite (fine-grained rim facies, porphyritic central facies). The finer-grained variety has higher values (means of 11 samples: 78.7 p.p.m. Th, 30.8 p.p.m. U, Th/U = 2.74) than the porphyritic facies (means of 33 samples: 61.8 p.p.m. Th, 19.2 p.p.m. U, Th/U = 3.35). Potassium is more or less homogeneously distributed within the syenite mass (mean of 44 determinations: 4.9% K, width of variation: 3.5–5.7% K). Anomalously high U concentrations (up to 270 p.p.m.) at shear zones are signs of postintrusive redistribution processes.

INTRODUCTION

The remarkably high radioactivity of the Giuv Syenite was first described by Hirschi (1920), the pioneer of nuclear geology in Switzerland. Detailed radiometric mapping and extensive sampling of this interesting rock unit was carried out in course of the prospecting campaigns of the Arbeitsausschuss Atombrennstoffe 1963–1966. The results of the field measurements and the distribution of uranium and thorium, based on laboratory determinations, are given in this paper. Further studies, especially of the distribution of U and Th in and around single mineral components, are in progress.

The *Aar Massif* forms a more or less autochthonous basement unit in the central Swiss Alps. Its surface of approx. 2,000 km^2, displays a complex framework of pregranitic crystalline rocks like gneisses, schists, amphibolites and migmatites, discordantly interwoven with younger granitic masses (Fig.1), the main units of which (central Aare Granite, Gastern Granite) give Hercynian radiometric ages (270–300 m.y., see Pasteels, 1964; Wüthrich, 1965). Several smaller, not yet dated, eruptive stocks like the Giuv Syenite, break through the older basement rocks. The upper age limit of the Giuv Syenite is determined by the observation of Huber (1948) that the syenite forms xenoliths in the central Aare Granite.

The surface extension of the *Giuv Syenite* (< 4 km^2) is quite modest. It forms a narrow band, oriented SW–NE, which reaches a width of 500–1,000 m in the region of the Piz Giuv. The syenite mass consists of two main rock types: the central and western part is occupied by a coarse-grained, porphyritic variety with microclines about 1–2 cm in diameter; at the rim (especially at the southern border) one finds a finer-grained facies with feldspars < 5 mm in diameter . In both rock types the mineral components vary within the following ranges:

Potassium feldspar	40–50 vol. %
Plagioclase (albite–oligoclase)	25–30 vol. %
Quartz	10–15 vol. %
Biotite and hornblende	10–18 vol. %
Sphene	~ 1 vol. %
Accessory minerals	~ 1 vol. %

The porphyritic variety contains definitely more dark constituents than the

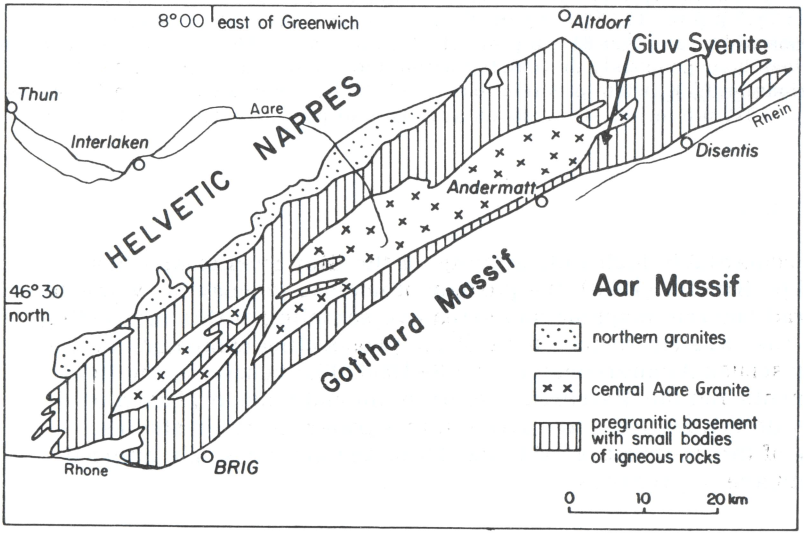


Fig.1. Generalized geologic map of the Aar Massif.

fine-grained variety. According to the nomenclature of Streckeisen (1967), the rock lies in the syenite/monzonite border region, the definition *syenite* (Weber, 1904) thus being acceptable.

Two whole-rock analyses indicate the bulk chemistry of the two main rock types (Table I). The porphyritic variety is markedly higher in FeO, Fe_2O_3, CaO and MgO and lower in SiO_2 than the fine-grained variety. Calculating the differentiation index 1/3 Si + K − (Ca + Mg) after Nockolds and Allen (1953), one obtains + 8.4 for the porphyritic sample (GS 33) and + 11.4 for the fine-grained variety (GS 1).

RADIOMETRIC FIELD MEASUREMENTS

The study of the radioactivity distribution within the outcropping Giuv Syenite began with a detailed radiometric mapping, using portable Landis and Gyr counters (Elbel et al., 1962). The area studied is relatively small and mostly easily accessible. This permitted a tight network of measurement points from which a radiometric field map (Fig.2) was constructed. Because of geometry differences and other factors, this map reflects the radioactivity distribution only in a generalized way. Furthermore, it can not show all the local variations observed at veins, xenoliths.etc. On the other hand, areas of similar radioactivity are more easily outlined by this type of representation than from a limited number of laboratory

TABLE I

Whole-rock analyses of the main types of the Giuv Syenite[1]

	Weight (%)	
	porphyritic variety (sample GS 33)	fine-grained variety (sample GS 1)
SiO_2	59.2	62.80
Al_2O_3	15.85	15.05
Fe_2O_3	1.51	1.26
FeO	3.42	2.38
CaO	5.88	3.87
MgO	3.42	2.37
Na_2O	3.0	3.30
K_2O	6.6	7.00
MnO	0.13	0.07
TiO_2	0.82	0.85
P_2O_5	0.39	0.63
H_2O^+	0.28	0.43
CO_2	–	0.33
Total	100.50	100.34

[1]Analyst: Miss L. Schopfer

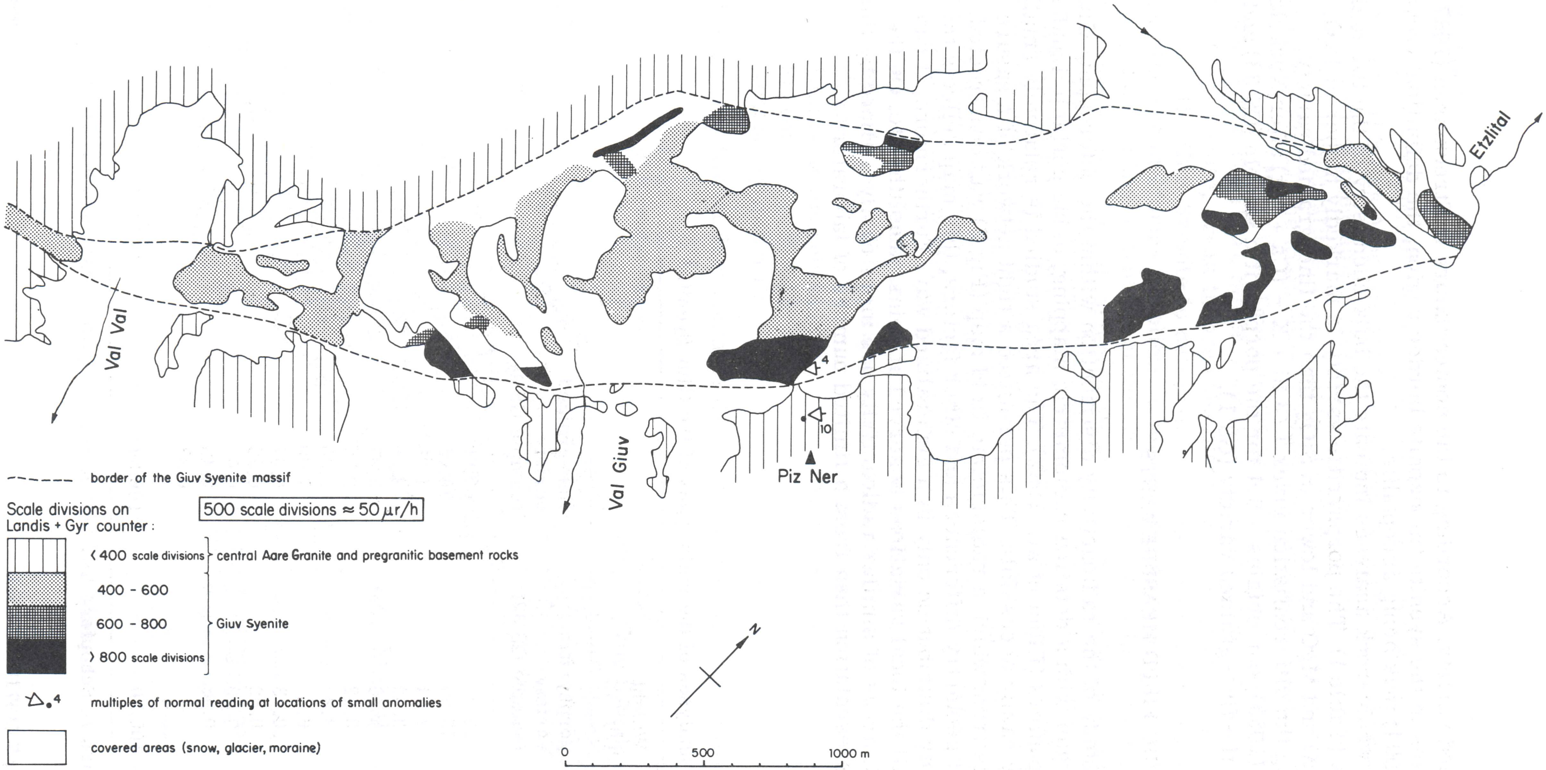


Fig.2. Map of radiometric field measurements showing zonal distribution of radioactivity in the Giuv Syenite body. 500 scale divisions on the Landis and Gyr counter used, correspond approximately to 50 μr/h. Cosmic radiation intensity at a medium altitude of 2,500 m above sea level: 13 μr/h (Herbst, 1964).

determinations. The dashed lines in Fig.2 mark the contact of the syenite mass with the Central Aare Granite. The syenite has a remarkably higher radioactivity than its surroundings.

Within the syenite mass, a rim zone of higher radioactivity, corresponding (except in an area in the NE part) to the fine-grained variety of the syenite, can be distinguished.

During the radiometric field measurements, a number of spots (extending from a few dm^2 to 1 m^2) have been found, where the readings exceeded even the high radiation level of the syenite. In Fig.2 the two strongest anomalies (4 to 10 times the average syenite reading) are marked by arrows. Both anomalies are located near the SE contact of the syenite, one in a shear-zone within the syenite, the other a few hundred meters southeast of the syenite in the gneisses of the old crystalline basement unit of the Aar Massif.

LABORATORY DETERMINATIONS

During the field measurements fresh, representative rock material was sampled. Uranium, thorium and potassium concentrations have been determined quantitatively in the collected samples, using gamma ray spectrometry as the main technique. About 350 g rock powder was filled into standardized aluminum sample cans. Special care was taken to fill these containers completely. Incomplete filling causes erroneously high concentrations [$c(\Delta h)$]. The experimentally determined correction curve (Fig.3) enables the calculation of the exact concentrations (c_{true}). The sample cans were placed on a top of a 3 inch × 3 inch NaI(Tl) crystal surrounded by

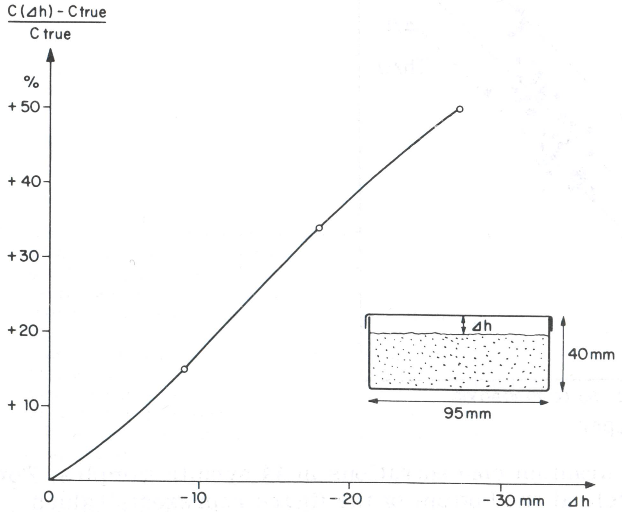


Fig.3. Influence of incomplete filling of the sample container on the calculated uranium concentration (see text).

5 inch of lead to reduce the background. The gamma ray spectra were obtained from a Nuclear Data 128-channel analyzer on punched paper tape. These data were then transferred to a magnetic tape which served as the input for the calculation of uranium, thorium and potassium concentrations by a least-squares program using standard spectra (Rybach et al., 1966). For each sample a card containing sample number, sample weight and counting time is punched. All calculations were performed on a CDC-1604 computer. Counting times were of the order of 10 min per sample, the precision is about 5–10% of the amounts present. U and Th have been calculated with the assumption that the parent elements ^{238}U and ^{232}Th are in secular equilibrium with their decay products. Similarly, one assumes that the ratio $^{40}K/K$ is constant in nature (0.0119 atom %). A number of uranium results have been checked by fluorimetric determinations, the agreement found was satisfactory.

In Table II the uranium, thorium and potassium concentrations of 54 samples are given. 10 samples are xenolithic or sheared varieties, the remaining 44 are typical syenites having the following mean values:

	U (p.p.m.)	Th (p.p.m.)	K (%)	Th/U
Mean[1] of all syenite samples	22.1	66.0	4.9	3.19
Mean of the fine-grained variety (11 samples)	30.8	78.7	5.0	2.74
Mean of the porphyritic variety (33 samples)	19.2	61.8	4.9	3.35

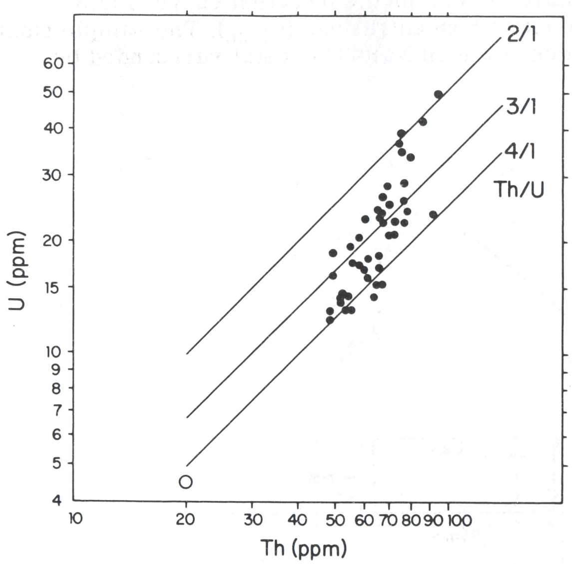


Fig.4. Thorium vs. uranium concentrations in 44 syenite samples. For comparison: the open circle at the bottom of the figure represents values for an average granite (after Clark et al., 1966).

[1]The arithmetic mean values have been calculated before rounding the results (for Table II) to two significant figures.

TABLE II

Uranium, thorium and potassium concentrations, Th/U ratios of samples from the Giuv Syenite, determined by gamma ray spectrometry

Sample no.	Rock	U(p.p.m.)	Th(p.p.m.)	Th/U[1]	K(%)
GS 1	Giuv Syenite, fine-grained variety	33	80	2.40	5.7
GS 10		26	76	3.00	4.3
GS 11		39	75	1.93	4.9
GS 27		18	66	3.63	5.1
GS 28		35	75	2.15	4.9
GS 29		28	69	2.49	4.6
GS 30		23	90	3.91	5.4
GS 31		23	78	3.39	4.6
GS 32		49	93	1.89	5.1
GS 34		42	85	2.05	5.2
GS 52		23	78	3.29	5.1
GS 2	Giuv Syenite, porphyritic variety	14	54	3.77	5.1
GS 3		23	67	2.92	5.0
GS 4		17	59	3.50	5.1
GS 5		22	71	3.17	3.5
GS 6		23	76	3.36	5.7
GS 8		29	75	2.64	5.2
GS 9		37	73	2.00	4.9
GS 12		13	49	3.69	4.8
GS 13		14	52	3.61	5.0
GS 14		13	54	4.15	4.4
GS 15		12	49	4.04	5.1
GS 16		13	55	4.24	5.0
GS 17		20	58	2.84	5.1
GS 18		19	49	2.60	5.0
GS 19		16	61	3.79	4.3
GS 21		26	66	2.52	5.5
GS 22		18	62	3.42	5.1
GS 24		15	67	4.32	5.1
GS 26		24	67	2.84	5.0
GS 33		24	67	2.86	5.5
GS 35		21	72	2.92	5.2
GS 36		21	69	3.30	5.1
GS 37		15	66	4.27	5.5
GS 38		17	66	3.89	4.8
GS 39		14	64	4.49	4.8
GS 40		14	53	3.88	3.6
GS 41		23	67	2.95	5.5
GS 42		17	59	3.48	4.7
GS 43		19	55	2.84	4.6
GS 44		23	59	2.59	5.4
GS 45		25	69	2.79	3.5
GS 46		18	55	3.01	5.2
GS 48		14	52	3.79	4.8
GS 7A	unusually light syenite with scattered amphibole phenocrysts	16	29	1.82	6.0
GS 7B	dark, amphibole-rich syenite	29	53	1.81	1.4
GS 16R	weathered part of sample GS 16	15	57	3.73	5.5
GS 21R	weathered part of sample GS 21	12	57	4.64	5.4
GS 49	syenite, completely transformed into schist during the Alpine event	19	59	3.08	4.4
GS 50	syenite, completely transformed into schist during the Alpine event	11	37	3.40	4.1
GS 25	dark, biotite-rich xenolith	18	61	3.33	6.1
GS 23	dark, biotite-rich xenolith	34	52	1.55	4.1
GS 51	schistose syenite with anomalously high radioactivity	274	109	0.40	3.5
	(2 samples)	273	102	0.37	2.0
Mean[1] of the fine-grained variety		30.8	78.7	2.74	5.0
Mean[1] of the porphyritic variety		19.2	61.8	3.35	4.9
Mean[1] of all Giuv Syenite samples		22.1	66.0	3.19	4.9

[1]Calculated before rounding data to two significant figures.

The uranium vs. thorium plot (Fig.4) displays a trend of decreasing Th/U ratio with increasing uranium concentrations. The average values of Th and U in granitic rocks lie considerably lower (open circle in Fig.4, after Clark et al., 1966).

Fig.5 shows the frequency distribution of U and Th in the 44 syenite samples. A tendency towards a lognormal distribution of uranium in both syenite types seems to be present, despite the relatively small number of determinations. For thorium this tendency is less clear.

The highest uranium contents were found in samples from the above-mentioned shear-zone at the southeast contact of the syenite ($c_{U\ max}$ = 274 p.p.m.). Uranium is here enriched by a factor of 8 with respect to the normal country rock, a fine-grained syenite. Whereas the Th content is normal, the Th/U ratio is as low as 0.4. Uranium seems to originate from the syenite itself. Whether this mobilisation took place during the latest phases of the solidification of the syenite mass, or during younger events, cannot be decided. Uranium mineralisations in the old gneisses further to the SE may indicate the wandering of U over longer distances. The uranium occurrences in the vicinity of Trun (Upper Rhine valley, ca. 22 km E of the Giuv Syenite) have been known for a few years (Hügi et al., 1962). The Giuv

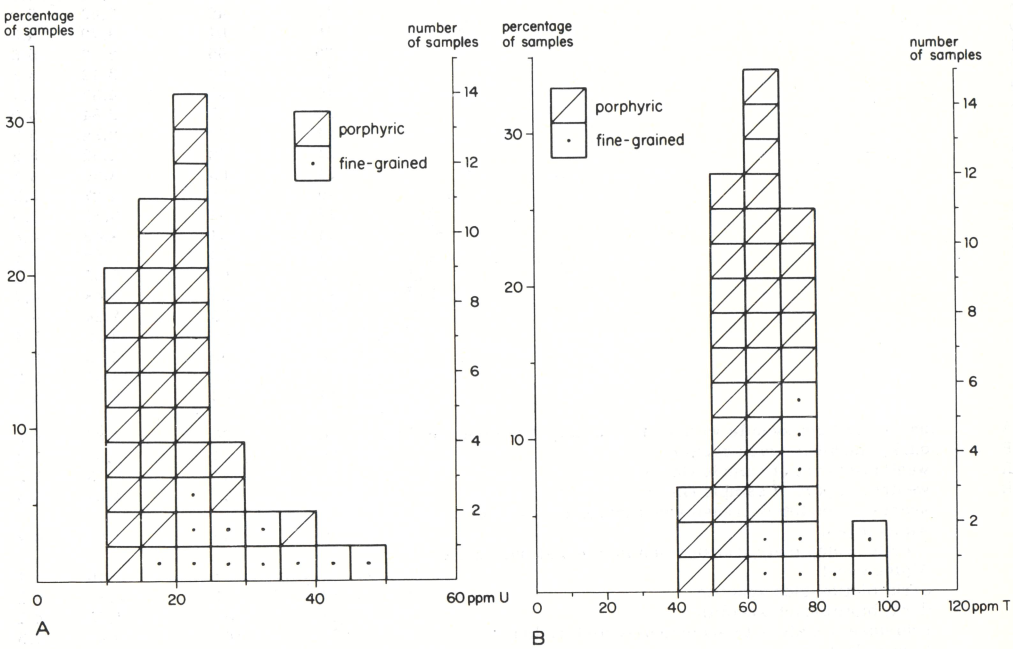


Fig.5A. Frequency distribution of uranium concentrations in 44 Giuv Syenite samples. B. Frequency distribution of thorium concentrations in 44 Giuv Syenite samples.

Syenite (or similar continuation[1] towards the east) may be a possible source for these mineralisations.

THE DISTRIBUTION OF U AND Th WITHIN THE SYENITE BODY

Fig.6 shows the locations of the samples and their U and Th content. The larger gaps between samples are due to debris (talus), and glacier and snow cover of the outcrops. The U and Th distributions clearly show zonal patterns. The dotted area at the rim of the syenite in Fig.6A contains all samples with > 21 p.p.m. U. The mean of these 23 samples is 28.1 p.p.m., whereas the mean of the 21 samples from the core zone is only 15 p.p.m. Thorium has a similar distribution (Fig.6B). All samples with > 60 p.p.m. Th are located in rim zones (29 samples here have a mean of 72.1 p.p.m. Th, the 15 samples in the core area have a mean of only 54.2 p.p.m. Th). The mean Th values in the zones of Fig.6A (uranium map) are 57.3 p.p.m. (core) and 73.9 p.p.m. (rim).

As uranium is relatively more enriched in the rim than thorium, the mean Th/U ratios decrease from 3.68 (core) to 2.75 (rim).

The rim zones of higher radioactivity consist mainly of the fine-grained syenite. They also contain, however, areas of the porphyritic variety. The factor determining the distribution of the radioelements thus seems to have been the position within the syenite body rather than the petrologic development of the rocks.

The agreement between the radiometric field map (Fig.2) and the radioelement distribution maps (Fig.6) is remarkable. The distribution of *potassium* is, however, random throughout the whole syenite mass, no significant difference between core and rim is present.

Some indication of the distribution between mineral phases was furnished by gamma ray spectrometric determinations on accessory-mineral concentrates from sample GS 1 (Rybach et al., 1970):

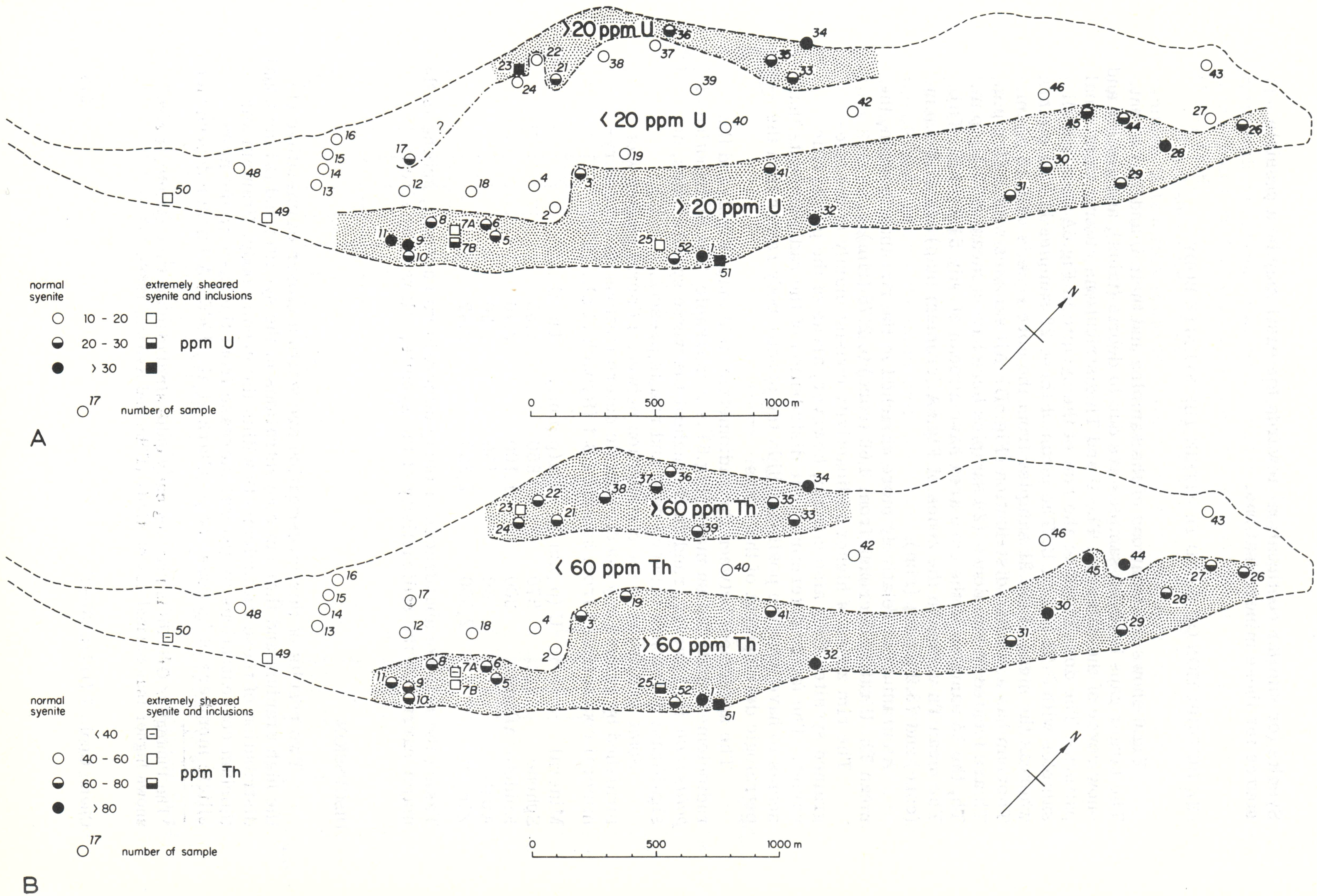
Mineral	U (p.p.m.)	Th (p.p.m.)	Th /U
Sphene	570	850	1.50
Epidote/allanite	300	1,100	3.70
Apatite	60	130	2.20
Zircon	2,800	2,300	0.82

Taking the relative abundances of these minerals into account, sphene is the major carrier of the radioactivity of the syenite, followed by zircon.

DISCUSSION

Two results of this investigation are worth a closer examination: (1) the high uranium and thorium concentrations in the syenite; and (2) the zonal distribution of these radioelements within the syenite body. Uranium and thorium concentrations in general increase from the basic to the more silicic members of igneous rocks (Clark et al., 1966). A further dependence

[1]The Punteglias Granite is petrologically quite similar to the Giuv Syenite, it also shows higher than normal radioactivity (Föhn und Rybach, 1968).


>20 ppm U
<20 ppm U
>20 ppm U
normal syenite
extremely sheared syenite and inclusions
10 - 20
20 - 30
> 30
ppm U
number of sample
0
500
1000 m
N
A
>60 ppm Th
<60 ppm Th
>60 ppm Th
normal syenite
extremely sheared syenite and inclusions
<40
40 - 60
60 - 80
> 80
ppm Th
number of sample
0
500
1000 m
N
B

on bulk chemistry and mineralogical composition has been established in a number of cases, but the dependence does not seem to follow any general rule (Coppens and Jurain, 1966).

Silicic igneous rocks contain on the average 4.9 p.p.m. U and 20 p.p.m. Th (Clark et al., 1966), with a Th/U ratio of about 4. With increasing Th content, this ratio also increases (Rogers and Ragland, 1961). High Th and U concentrations are associated with high potassium concentrations: Heier and Rogers (1963) calculated a mean K/Th ratio of $4.9 \cdot 10^4$ and a mean K/U ratio of $1.29 \cdot 10^4$ for granitic rocks.

When compared with common silicic igneous rocks, *the Giuv Syenite has anomalously high U and Th concentrations*. In Table III the values for the Giuv Syenite are listed together with those of some typical igneous rocks rich in Th and U (the list is not intended to be complete). The uranium contents of the Giuv Syenite belong to the highest values reported in the literature (about 5 times the normal amount in the porphyritic, and 6 times the normal amount in the fine-grained variety). Extremely high U concentrations have been determined in the lujavrites of the Ilímaussaq intrusion in southwest Greenland: average values of 100–200 p.p.m., with extremes up to 1,200 p.p.m. (Bondam and Sorensen, 1958). The thorium concentrations in the Giuv Syenite are 3.5–4 times the normal in rocks of similar bulk chemistry. The fine-grained variety has, however, with a mean Th content of 80 p.p.m., a Th/U ratio of 2.74, which is the lowest value in Table III. The general tendency for increasing Th/U ratio with increasing Th contents, is demonstrated in Table III. The strong correlation between uranium and thorium is also well expressed in Fig.4.

The potassium concentrations in the Giuv Syenite (mean value: 4.9%K) can be considered as normal. The anomalously high Th and U concentrations cause extremely low K/Th and K/U ratios: for the average Giuv syenites these ratios are $0.07 \cdot 10^4$ and $0.22 \cdot 10^4$, contrasting with the above mentioned figures of $4.9 \cdot 10^4$ and $1.29 \cdot 10^4$ in normal granites.

Magmatic differentiation in batholiths generally yields rocks of more mafic types on the margins, and progressively more felsic rocks toward the core. Ragland et al. (1967) describe an interesting deviation from this rule: in the Enchanted Rock batholith (Llano uplift, Texas) the outer zones contain the most felsic rocks while the intermediate and central zones consist of the most mafic ones. A further peculiarity of this rock unit is the fact that U and Th do not follow normal differentiation trends: Th is essentially invariant and U even decreases with increasing fractionation. In the case of the Giuv Syenite the most felsic differentiates are also located at the margins. U and Th follow, however, normal fractionation trends: the porphyritic sample GS 4 with a Nockolds-Allen differentiation index of +8.4 contains 16.9 p.p.m. U and 59.2 p.p.m. Th, whereas the fine-grained variety GS 1 with an index of +11.4 has 33.2 p.p.m. U and 79.8 p.p.m. Th. According to these results, the distribution of thorium and uranium in the Giuv Syenite seems to be controlled by primary processes.

Fig.6A. Distribution of uranium within the Giuv Syenite, determined by laboratory gamma ray spectrometry. Figures: sample numbers (see Table II). B. Distribution of thorium within the Giuv Syenite, determined by laboratory gamma ray spectrometry.

TABLE III

Some Th- and U-rich igneous rocks

Th(p.p.m.) mean	U(p.p.m.) mean	Th/U mean	Rock/locality	Literature
222	50	4.4	trachyte/phonolites (Vico Volcano, Italy)	Locardi and Mittempergher (1967)
185	18.5	10.0	granosyenite, quartz syenite (northern Kirgizia)	Turovskij (1957)
116.2	35.2	3.04	lujavrite of the Ilímaussaq-intrusion (southwestern Greenland)	Hamilton (1964)
98	8.3	11.7	Conway Granite (New Hampshire, U.S.A.)	Rogers and Ragland (1961)
97	9.3	10.4	Tokovskii Granite (middle Dnjepr region)	Filippov and Komlew (1959)
91	9	11.0	leucocratic granite	Smirnov (1962)
78.7	30.8	2.74	fine-grained Giuv Syenite (Switzerland)	this paper
74	14	5.3	syenite (northern Kirgizia)	Turovskij (1957)
70.1	7.2	9.7	Alaskite Granite (northern Kirgizia)	Turovskij (1957)
63	16.2	4.5	leucocratic granite (Rum Jungle Complex, Australia)	Heier and Rhodes (1966)
62.9	13.7	4.5	syenite (Kzyl-Ompul Massif)	Leonova et al. (1961)
61.8	19.2	3.35	porphyritic Giuv Syenite (Switzerland)	this paper

Zonal distribution patterns for radioelements have been described several times. In most cases an increase of the radioactivity towards the contacts is found[1]. In the classical paper of Ingham and Keevil (1951), an increase of α-activity by a factor of 3–6 from the core towards the rim zone is described for four Canadian batholiths. The Rotondo Granite (Gotthard Massif, Switzerland) consists of a core zone with 17 p.p.m. eU (U+Th) and a rim zone with 22.4 p.p.m. eU (Rybach et al., 1962).

In rim zones U is usually more highly enriched than Th (Krylow, 1959; Rybach et al., 1966). The Giuv Syenite demonstrates this tendency clearly: the increase of the uranium contents by a factor of 2 from core to rim is accompanied by a decrease in the Th/U ratio from 3.68 to 2.75. The reason for this behaviour is not yet clear. The higher radioactivity of the fine-grained syenite variety indicates the importance of petrologic factors for the distribution of the radioelements. On the other hand the anomalously high U concentrations at some spots in the southeast part of the area studied, are possibly signs of postintrusive remobilisations. Most likely the distribution pattern found is a superposition of several different processes acting at different times. Further U and Th determinations on the rock-forming minerals and leaching experiments which are in progress, may answer some of these questions.

It must be mentioned here that the described model-like distribution occurs in a syenite body which is, at its broadest exposure, less than 1 km wide. Therefore, the observation of Ingham and Keevil (1951) that a contrast between core and rim occurs in igneous rock bodies of great surface extension only, cannot be taken as a general rule.

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[1] There are some exceptions: Hofmänner (1964) describes the increase of U + Th from the rims toward the center of the Gamsboden granite-gneiss body (Gotthard Massif, Switzerland).

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