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Radioelement abundance data for some Dalradian rocks from Co. Donegal, Ireland

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ABSTRACT. Data for U, Th, and K, determined by epithermal neutron activation methods, are reported for Lower and Middle Dalradian lithologies from Co. Donegal, Ireland. Abundances are identical to rocks with stratigraphical correlation in the Scottish Dalradian and appear not to have been affected by regional metamorphism to biotite and possibly even garnet grade during the Grampian orogenic phase, or later contact metamorphism in the aureole of the Main Donegal pluton. Uraniferous Dalradian lithologies have not been encountered in Donegal. Incorporation of Dalradian rocks, either by assimilation or as extensive raft trains within the Main Donegal granite, is therefore unlikely to have contributed to the development of known pegmatite-hosted U mineralization associated with this late Caledonian pluton.

KEYWORDS: radioactive elements, Dalradian, Co. Donegal, Ireland.

BOTH pegmatite-hosted and vein-type U mineralization are associated with the Main Donegal granite, northwestern Ireland (O'Connor *et al.*, 1984; O'Connor, in press). A low Th uraninite is the principal ore mineral in the mineralized biotite-pegmatites and its textural relations are consistent with late magmatic formation. The uraninite yields a $^{207}Pb/^{206}Pb$ age of 407 ± 4 Ma, concordant with the age of emplacement of the Main Donegal pluton dated by whole-rock Rb-Sr methods at 407 ± 23 Ma (O'Connor *et al.*, 1982); the vein-type mineralization consists of later pitchblende with a $^{207}Pb/^{206}Pb$ age of 295 ± 4 Ma.

The distribution and NE-SW linearity of radiometric anomalies in the Main Donegal granite, and their main development at the southwestern end of the pluton, suggest a dominant structural control on uranium mineralization. The linear uraniferous zones may represent annealed shears within the pluton. The occasional association of the mineralized zones with strike-parallel raft trains of Dalradian metasediments within the pluton gave rise to the present study in which the various Dalradian lithologies in the envelope of the pluton (represented in the raft trains) were sampled to establish their radioelement contents and possible contribution to mineralization. The sampling programme was supplemented by a gamma spectrometric survey over the region (using a portable Geometrics GR410 spectrometer).

Methods. Rock samples, each weighing in excess of 3 kg, were collected wherever possible from working quarries, railway and/or road cuts. The freshest possible material was selected in each case and care was taken to obtain sample sets which reflected observed lithological variations in the Dalradian metasediments (fig. 1). Gross-count gamma ratemeter readings indicated that none of the whole-rock sampling sites is radiometrically anomalous; none of the samples analysed is mineralized. Samples were disaggregated and passed through a jaw crusher and further reduced to < 100 mesh powders in a Tema agate mill.

U and Th analyses were carried out at the Liverpool and Manchester Universities research reactor (Risley) by epithermal neutron activation by techniques described in O'Connor *et al.* (1981). K was determined by instrumental neutron activation analysis for all samples. The precision of each analysis, which respresents the weighted mean of results obtained by comparing the few most intense lines of the ^{239}Np and ^{233}Pa spectra in the 200-400 KeV region for samples and standards, was found to be $\pm 10\%$ with a lower limit of detection of 1 ppm for U and Th.

A suite of samples was chosen for a more detailed U distribution study within rock thin sections by fission-track registration in Lexan plastic overlays (Basham *et al.*, 1982). Thin sections were irradiated in the AWRE Aldermaston Herald reactor, an integrated flux of 5×10^{15} thermal neutrons/cm^2 being given, and afterwards the Lexan detector was etched in NaOH solution to reveal the induced fission fragment tracks of ^{235}U. Standard U glasses were included for comparison to obtain quantitative measurements, and a detection level of less than 1 ppm was achieved.

Results. Radioelement abundances of representative Dalradian lithologies which outcrop in the study area are given in Table I. Comparative radioelement data for similar lithologies elsewhere are given in Table II, while Table III compares mean data for Dalradian lithologies obtained by both neutron activation and gamma spectrometric analysis.

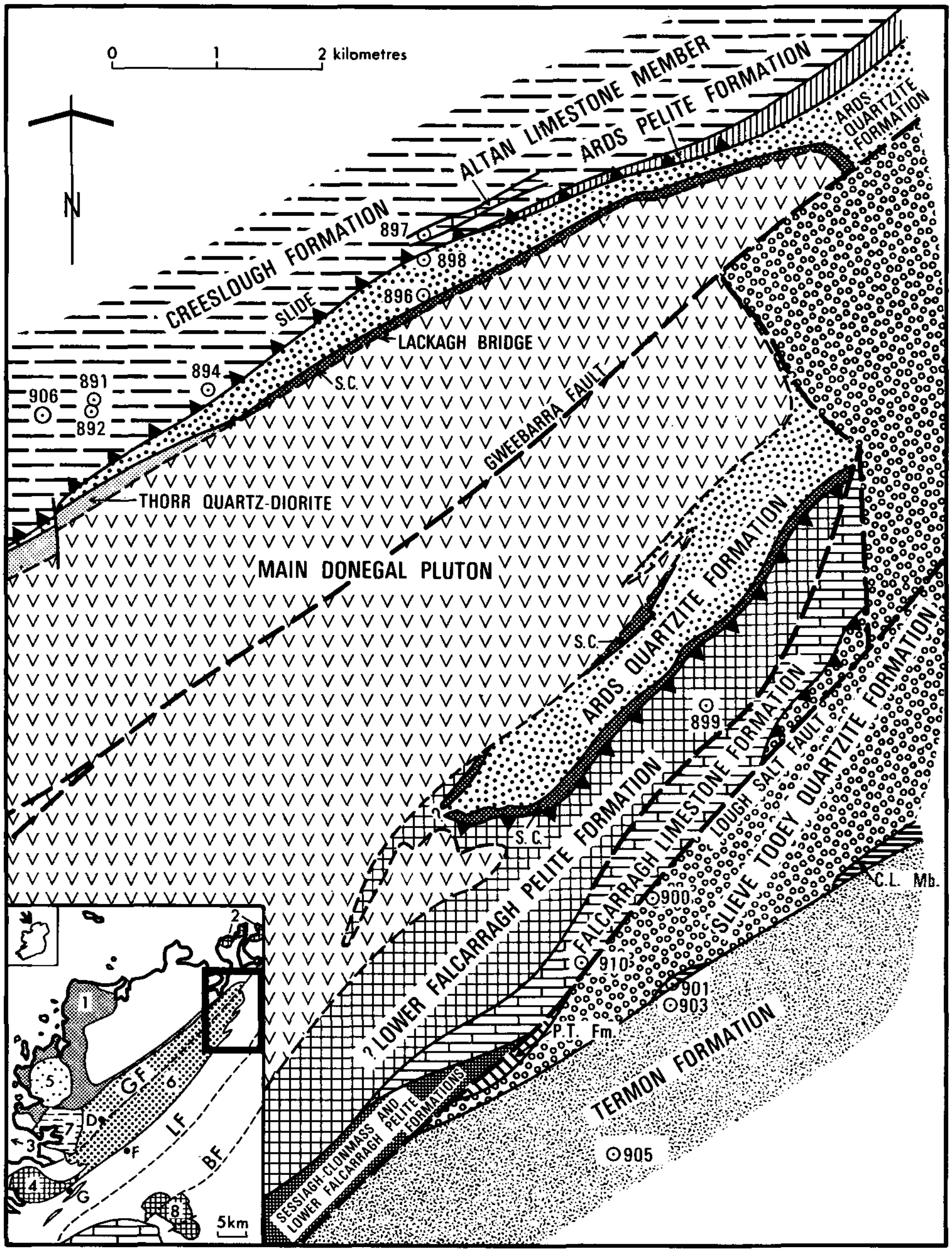

FIG. 1. Geological sketch-map of the northeastern end of the Main Donegal granite showing whole-rock sample localities in the Dalradian metasediments. Abbreviations are C.L. Mb.—Cranford Limestone Member (Termon Formation); P.T. Fm.—Port Askaig Tillite Formation; S.C.—Sessiagh Clonmass Formation. On inset map plutons numbered are (1) Thorr, (2) Fanad, (3) Toories, (4) Ardara, (5) Rosses, (6) Main Donegal, (7) Trawenagh Bay, (8) Barnesmore; BF—Belshade Fault; GF—Gweebarra Fault; LF—Leannan Fault; D—Doochary; F—Fintown; G—Glenties.

Stratigraphically, the Donegal rocks investigated are of Lower and Middle Dalradian age (Pitcher and Berger, 1972) and underwent prograde regional metamorphism to upper greenschist (biotite) facies and possibly to epidote-amphibolite (garnet) facies, followed by retrograde metamorphism prior to granite emplacement. A recent study of radioelement abundance levels in stratigraphically equivalent and lithologically similar Scottish Dalradian rocks (Atherton and Brotherton, 1979) serves as a convenient basis for comparison. Moreover, the abundance data of Atherton and Brotherton (1979) were obtained by similar epithermal NAA methods and at the same research establishment (Risley, UK) as those of this study.

Limestone has a low geochemical affinity for U, Th, and K. In Table II, Dalradian limestones have similar radioelement contents to Russian limestones (Baranov *et al.*, 1956) and North American limestones (Adams and Weaver, 1958; Rogers and Adams, 1969).

Four Donegal Dalradian pelites have a mean U content of 2.8 ppm, which is slightly higher than the overall Scottish Dalradian mean of 2.3 ppm for this lithology (Atherton and Brotherton, 1979). However the mean of six Lower Dalradian samples

TABLE I Radioclement abundance data* for Dalradian rocks, Co. Donegal, Ireland.

GSI Sample Number	Lithology	Grid Reference (Defines 10m square)		U (ppm)	Th (ppm)	K %	Th/U	U/K x 10^4	Th/K x 10^4
790892	Metadolerite (Creeslough Fm.)	C0643	2979	1.3	1.0	0.36	0.77	3.61	2.78
790896	Quartzite (?Ards Quartzite Fm.)	C0955	3106	2.1	8.2	1.54	3.90	1.36	5.32
790900	Quartzite (Slieve Tooey Quartzite Fm.)	C1225	2562	<1.0	2.8	1.06	>2.80	<0.94	2.64
790905	Psammite (Termon Fm.)	C1222	2298	1.6	4.2	0.50	2.63	3.20	8.40
790891	Calc-pelite (Creeslough Fm.)	C0645	2984	3.3	15.3	2.90	4.64	1.14	5.28
790898	Graphite Schist (Ards Pelite Fm.)	C0954	3163	3.2	21.6	1.90	6.75	1.68	11.37
790899	Pelite (L. Falcarragh Pelite Fm.)	C1274	2427	2.8	20.7	1.69	7.39	1.66	12.25
790903	Phyllite (Termon Fm.)	C1252	2463	2.0	11.6	2.40	5.80	0.83	4.83
790894	Calc-silicate Schist (Creeslough Fm.)	C0756	3012	<1.0	2.5	0.56	>2.50	<1.79	4.46
790906	Limestone (Creeslough Fm.)	C0592	2967	1.0	5.3	1.07	5.30	0.93	4.95
790897	Limestone (Altan Lst. Mb., (Creeslough Fm.)	C0949	3181	<1.0	<1.0	-	~1.00	-	-
790910	Limestone (Falcarragh Lst. Fm.)	C1155	2487	<1.0	1.9	0.26	>1.90	<3.85	7.31
790901	Limestone (Cranford Lst. Fm.)	C1253	2487	1.1	<1.0	0.20	<0.91	5.50	<5.00

* U, Th and K determined by epithermal neutron activation at the Liverpool and Manchester Universities Research Reactor, Risley.

TABLE II Comparative radioelement abundance data+ for Dalradian lithologies and other sedimentary rocks

Lithology	Number of Samples	Method	U (ppm)	Th (ppm)	K%	Th/U	U/K x 10^4	Th/K x 10^4	Reference
Dalradian Limestones	5	ENAA	0.8(0.3)	2.2 (1.9)	0.52(0.40)	3.16(2.33)	2.24(2.18)	5.06(1.62)	This Paper
Russian Limestones	128	Emanation	2.1	2.4	-	1.1	-	-	Baranov et al. 1956
N. American Limestones	25	Radiochemical	2.2	1.1	-	0.7	-	-	Adams & Weaver 1958
Dalradian Pelites	4	ENAA	2.8(0.6)	17.3(4.7)	2.22(0.54)	6.15(1.20)	1.33(0.42)	8.43(3.92)	This Paper
Dalradian Pelites/psammites	46(52)*	ENAA	2.3(0.9)	17.2(7.5)	3.3 (1.2)	8.1 (2.8)	0.73(0.28)	5.7(2.3)	Atherton and Brotherton 1979
Dalradian Schists	20	Gamma Spec.	1.8 (0.8)	12.0(6.2)	-	6.5 (1.7)	-	-	Richardson and Powell 1976
Dalradian Quartzite	3	ENAA	1.5(0.7)	5.1(2.8)	1.03(0.52)	3.51(0.76)	1.74(1.31)	5.45(2.88)	This Paper

+ Numbers in brackets are one standard deviation
* 46 values for Th and Th/U; 52 values for U

TABLE III Comparison of neutron activation (NAA) and gamma spectrometric (GS) mean data for different Dalradian lithologies, Donegal*

	U (ppm)		Th (ppm)		K %	
Lithology	GS	NAA	GS	NAA	GS	NAA
Limestone	0.9 (7)	0.8 (5)	1.3 (7)	2.2 (5)	0.84 (7)	0.52 (5)
Quartzite	2.5 (10)	1.5 (3)	3.9 (10)	5.1 (3)	1.98 (10)	1.03 (3)
Psammite	1.55 (2)	-	9.4 (2)	-	2.40 (2)	-
Pelite	2.6 (13)	2.8 (4)	17.3 (13)	17.3 (4)	3.39 (13)	2.22 (4)
Graphitic Pelite	2.8 (13)	3.2 (1)	21.1 (13)	21.6 (1)	4.74 (13)	1.90 (1)

* Numbers in brackets are number of analyses

analysed by these authors is 3.1 ppm U and is in closer agreement with our figure. A lower mean of 1.8 ppm U has been reported by Richardson and Powell (1976) for twenty Lower Dalradian schists (Leven Schists).

Our pelite mean of 17.3 ppm Th is closely similar to the overall Dalradian mean of 17.2 ppm Th quoted by Atherton and Brotherton (1979) and larger than the Dalradian mean of 12.0 ppm Th given by Richardson and Powell (1976). Values in the range 11–13 ppm Th for shales of varying age and provenance are given by Rogers and Adams (1969).

Our mean value of 2.2 % K is lower than 3.3 % K reported by Atherton and Brotherton (1979) for Dalradian pelites and more in keeping with typical shale K values quoted by Heier and Billings (1970).

Mean Th/U ratios in the range 6–8 have been obtained for pelites; similar ratios for Scottish analogues have been presented by Atherton and Brotherton (1979) and by Richardson and Powell (1976). Continental shales have been reported to have lower ratios in the range 3 to 5 (Baranov *et al.*, 1956; Adams and Weaver, 1958; Pliler and Adams, 1962).

The Th versus U plot (fig. 2) shows positive correlation, as does the K versus U plot. The K versus Th plot (fig. 2) suggests that most of the Th is contained in layered clay minerals, with some Th in other minerals, such as zircon. A strong positive K/Th correlation is also evident from gamma spectrometric data (O'Connor and Long, unpublished open file report to EEC 1981, Contract PU/3/79).

Our fission track studies show that the limestone lithologies are characterized by a low density of dispersed tracks; very rare point sources, where identifiable, relate to small Th or zircon crystals. In psammitic lithologies most activity is associated with detrital grains of dark reddish-brown and partly altered Th, strongly coloured monazite and euhedral to sub-rounded zircon; minor levels of activity are associated with biotite breakdown and altered ferruginous material. Among pelites, U is located in pre-metamorphic detrital zircon and subhedral monazite and is also dispersed throughout the rock.

Discussion and conclusions. Our data complement the findings of Atherton and Brotherton (1979) that Dalradian pelites have relatively higher Th and lower U contents compared with sediments discussed in the literature; Th/U ratios are also a little higher. They concluded that the present radioelement signatures of these pelites were probably primary features of the original sediments.

They found no evidence to support the suggestion that, as the content of graphite (of believed organic origin) increased in Dalradian schists, there was a corresponding increase in U levels; our data (Table III) for pelitic lithologies show no U enrichment where these are graphitic. Rocks with the highest U levels tended to plot close to the composition of natural muscovite on the A'KF plot of Atherton and Brotherton (1979), indicating that U is mainly associated with dispersed K-minerals rather than organic matter. These observations are supported by our fission track studies.

Atherton and Brotherton (1979) concluded that U has not moved out of the Scottish schists regionally metamorphosed to garnet grade, though statistically, there was possibly very slight U mobility in schists that attained staurolite, kyanite, and sillimanite grades judging from possibly higher Th contents in these rocks. With regard to the area studied around the NE end of the Main Donegal granite, it is also reasonable to believe that neither regional nor contact metamorphism altered the U or Th contents of the Dalradian schists to any appreciable extent. Atherton and Brotherton (1979) also found that Lower Dalradian rocks of Scotland

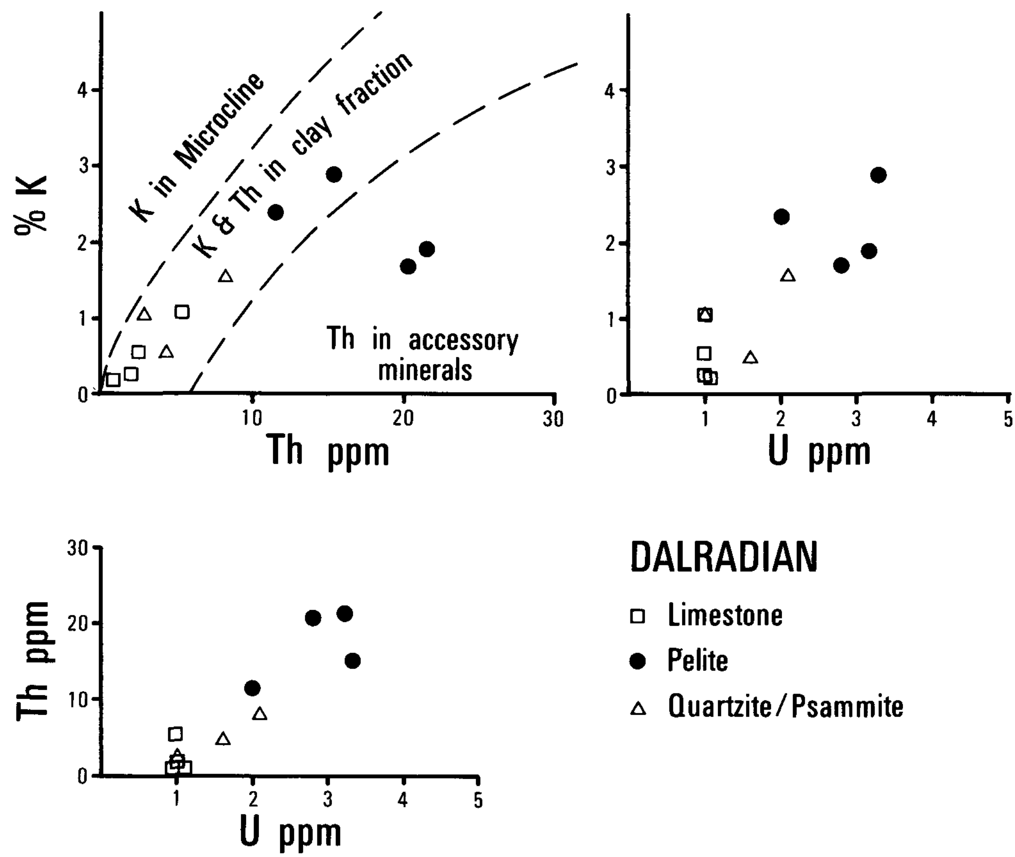

FIG. 2. U–Th–K relations in Dalradian metasediments, Co. Donegal (data in Table I).

had lower U, Th, and Th/U than Middle Dalradian schists, and that different geographical areas had different Th values, most probably indicating differences in Th content with provenance, rather than metamorphic grade. The sample set from Donegal is too small to allow meangingful statistical tests of differences between regions to be carried out.

None of the Dalradian lithologies investigated to date in Donegal (or Scotland) have high radioelement contents, so that the melting of such sediments or their incorporation in granitic magmas would not enhance the radioelement content of the latter substantially. The origin of U mineralization associated with the Main Donegal granite is not attributable to digestion of uraniferous lithologies in the exposed Dalradian rocks of the envelope or internal raft trains.

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