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Anomalous uranium and thorium associated with a granitic facies of the Bottle Lake Quartz Monzonite, Tomah Mountain area, eastern Maine

by

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Abstract

Rocks and stream sediments from the Tomah Mountain area in eastern Maine contain anomalous concentrations of uranium and thorium that appear to be related to the Topsfield granitic facies, an informally named part of the Bottle Lake Quartz Monzonite. The Topsfield is a muscovite-bearing, biotite-hornblende granite of Devonian age. It has intruded Silurian metasedimentary rocks in the study area. Pyrite is present and molybdenite has been observed in altered granite near shear zones.

Uranium and thorium data for rock samples show that uranium is relatively independent of thorium and that Th/U ranges from 1.8 to 4.8. Altered molybdenite-bearing granite shows a substantial loss of uranium and thorium. The uranium and thorium contents of stream sediments, which are closely related to the underlying rock type, are low over metasedimentary rocks and high over granite. Uranium does not appear to migrate far in the surficial environment, an inference supported by the unusually low uranium content of stream waters.

Although the Topsfield does not fit into the uraniferous two-mica granite group known regionally, the evidence for labile uranium, molybdenum enrichment, and hydrothermal alteration suggests that the granite is favorable for U-Mo vein or contact-metasomatic deposits in the granite or its aureole. The petrologic and geochemical data suggest an affinity to uranium- and

molybdenum-enriched calc-alkalic granites, such as the Devonian Catheart pluton, Katahdin Quartz Monzonite, and Debouillie pluton elsewhere in Maine.

Introduction

The granitic rocks of Maine, because of their rather common enrichment in various metals, have long attracted the attention of explorationists. Copper-molybdenum, molybdenum, molybdenum-uranium, and uranium enrichment has been noted in many plutons throughout Maine and much work has been directed toward understanding the age and geochemistry of these granites (E. Boudette, oral commun., 1980). In a regional stream-sediment geochemistry study of eastern Maine, Post and others (1967) noted anomalous amounts of molybdenum, tungsten, arsenic, bismuth, and antimony in the Tomah Mountain area. Later, Nowlan and Hessin (1972) confirmed the presence of anomalous molybdenum in three drainages and in soils in the Tomah Mountain area. They also reported the occurrence of molybdenite in some granite outcrops.

Because of the common association of uranium with molybdenum and tungsten and of the interest in uranium in the granitic terranes of New England, an investigation was made of the distribution of uranium, thorium, and radon in the rocks, stream sediments, and waters of that area.

Geologic setting

In the Tomah Mountain area (fig. 1), Silurian metamorphic rocks, mostly metasiltstone, impure quartzite, and slate, have been intruded by the Devonian Topsfield granitic facies, an informally named part of the Bottle Lake Quartz Monzonite (Larrabee and others, 1965). The Topsfield is a medium- to coarse-grained, pinkish-red granite composed of microcline, quartz, and plagioclase with minor muscovite, biotite, and hornblende. Sphene and pyrite are present locally. A contact-metamorphic aureole generally less than 1.2 km wide surrounds the pluton. A rusty quartz or quartz-pyrite stockwork is common in

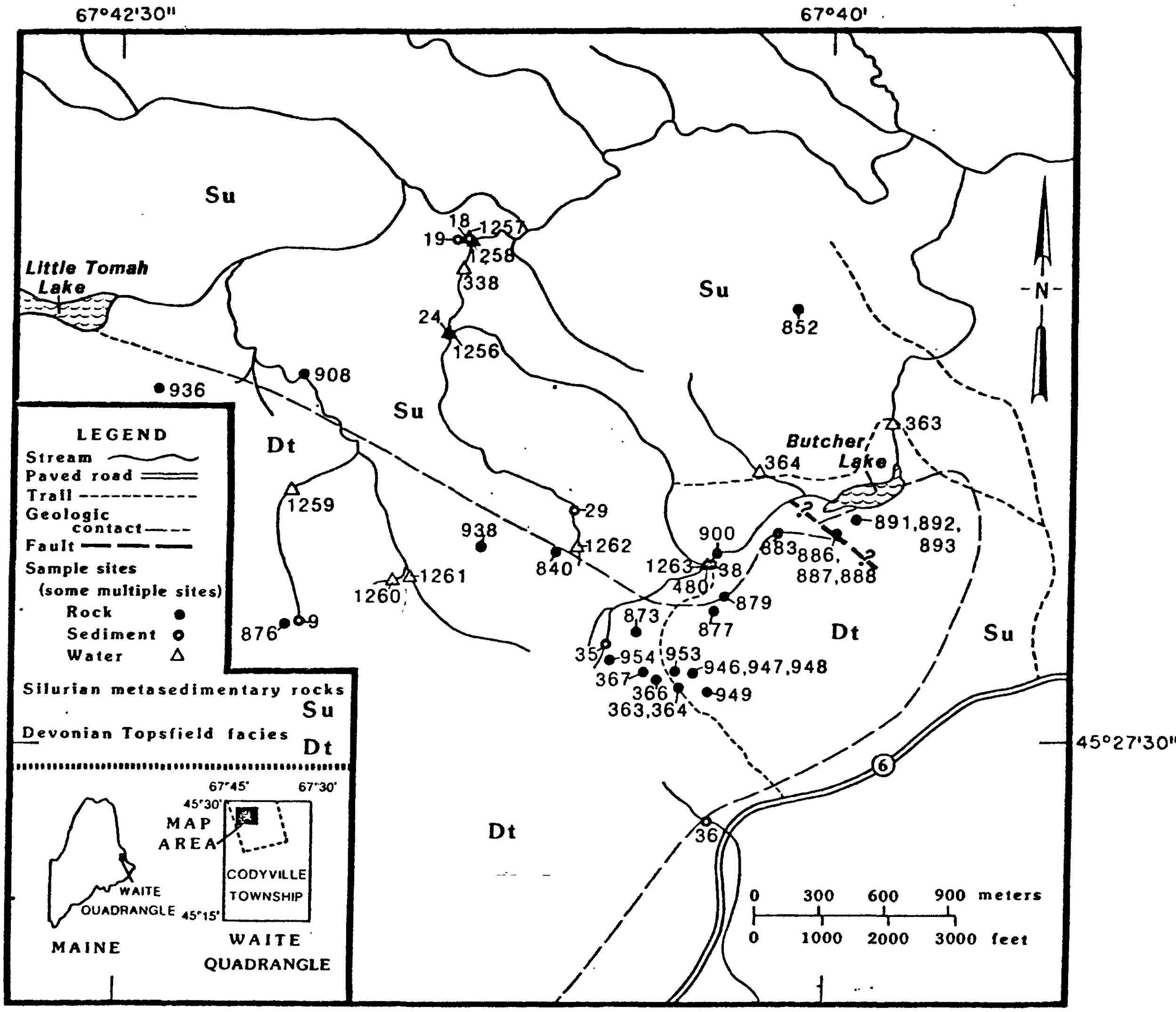

Figure 1.--Sample location map with generalized geology.

outcrops of the metasedimentary rocks. Near shear zones in the metasedimentary rocks, silicification, pyritization, iron-oxide films on cleavage surfaces, and bleaching are common. Sparse molybdenite has been observed in some granitic rocks (Nowlan and Hessin, 1972).

The presence of biotite and hornblende, sphene, and a contact-metamorphic aureole suggests that the Topsfield is a calc-alkalic granite. The 372-m.y. K-Ar age reported by Faul and others (1963) suggests that it is of Acadian age. Calc-alkalic granites in Maine often have copper, molybdenum, and(or) uranium associated with them (E. Boudette, oral commun., 1980).

Sampling and analysis

Stream sediments collected in the study area were generally rich in silt, usually from beneath or beside boulders. Samples were collected at 43 sites, and multiple samples were taken at eight of those sites. The -0.25-mm fraction was analyzed for Mo, As, Mn, Fe, and Cu by Nowlan and Hessin (1972). The unused part of the samples was retrieved for the present study. Rock samples were taken from outcrops in the study area and were prepared by crushing and grinding. For this study, splits of nine of the original stream-sediment samples and all of the rock samples were submitted to the Branch of Analytical Laboratories for analysis by delayed-neutron activation. Loss on ignition was also determined for the sediment samples.

Water samples were collected by Gary Nowlan in the late summer of 1978 during a period of low water flow. Most samples were taken from a flowing stream, but one was taken from a spring in bedrock. At each site, one sample was filtered and acidified for trace-metal analyses; a second, untreated sample was used for major species, pH, and specific conductance analyses; and a third sample was placed in glass bottles with tight caps for radon determinations.

Analytical techniques are summarized in table 4.

Uranium in stream sediments

Sample data are shown in table 1, and locations are given on figure 1. The uranium content of the nine stream-sediment samples from the Tomah Mountain area ranges from 11.4 to 57.5 ppm. The uranium content of samples taken from streams where they flowed over the granite was two to five times higher than that of the streams over the metasedimentary rocks. The correlations between uranium and manganese (+0.29), uranium and iron (-0.18), and uranium and organic matter (-0.06), estimated by loss on ignition, are not statistically significant at the 95-percent level on the basis of nine samples.

Owing to limitations inherent in the radiometrics, thorium analyses by delayed-neutron techniques are not considered reliable where U/Th >1. The data are reported in table 1, but no further comment can be made regarding their significance.

Uranium and thorium in rocks

Uranium content of granite samples from the area ranged from 4.4 to 12.0 ppm and averaged 8.1 ppm (table 2). Two samples of altered molybdenite-bearing granite contained 3.5 and 2.7 ppm uranium. A sample of aplite contained 5.4 ppm uranium. Four samples of metamorphic rocks ranged from 2.6 to 3.5 ppm uranium.

Thorium content for the granite samples ranged from 12.2 to 82.9 ppm, but most samples were between 20 and 30 ppm. The altered granite samples contained 4.9 and 5.6 ppm thorium; aplite, 8.7 ppm; and the metamorphic rocks, 6.0 to 10.7 ppm. The thorium-to-uranium ratio averaged about 3.5.

Table 1.--Uranium, thorium, loss on ignition in stream sediments

Sample site	ppm[1] U	ppm[1] Th	%[2] LOI	Underlying rock type
9	47.8	20.2	31	Granite
18	11.4	9.3	45	Metamorphic
19	12.3	14.1	40	Metamorphic
24a	12.1	10.6	25	Metamorphic
24b	16.8	3.1	35	Metamorphic
29	15.8	6.4	27	Metamorphic
35	54.1	26.1	45	Granite
36	34.3	15.4	19	Granite
38	57.5	20.8	28	Granite-metamorphic contact

[1]Analyses by H. Millard, R. Knight, A. Bartel, J. Hemming, R. White, R. Vinnola, and E. Brandt. Thorium data cannot be considered reliable where U/Th >1.

[2]Analyses by W. Ficklin.

Table 2.--Uranium and thorium in rocks

[All data in ppm. Analyses by H. T. Millard, Jr., C. M. Ellis, and V. C. Smith]

Sample no.	U	Th	Sample no.	U	Th
Granite					
363	8.2	33.4	949	10.7	31.5
364	5.5	21.6	953	8.2	26.0
366	9.4	22.7	954	10.3	30.0
873	7.9	82.9	879	7.1	28.8
876	7.8	28.2	887	4.9	23.9
877	8.9	22.2	892	8.9	27.7
883	12.0	21.8	893	7.3	25.6
886	4.4	12.2	946	6.9	23.4
891	7.0	27.3	947	7.7	29.8
936	9.7	29.5	948	9.9	24.6
Altered granite with molybdenite			Aplite		
840	3.5	4.9	367	5.4	8.7
938	2.7	5.6			

Metamorphic rock

Sample no.	Rock type	U	Th
480	Quartzite	3.2	7.8
852	Metasiltstone	3.2	6.0
900	Metasiltstone	2.6	8.0
908	Metasiltstone	3.5	10.7

A plot of thorium versus uranium in rocks (fig. 2) suggests that uranium content is largely independent of thorium content. The Th/U ranged from 1.8 to 4.8 in the group of samples with thorium content between 21 and 33 ppm. Only two samples differ from this trend. Uranium and thorium probably are in different sites within the granite and uranium may possibly be in the more labile sites. The low thorium and uranium content of two samples of altered granite with molybdenite suggests that both uranium and thorium may have moved during hydrothermal alteration.

Radon, uranium, and other species in water

Radon values showed wide variability in the waters sampled; however, waters from sites underlain by granite or just downstream from the granite-metamorphic rock contact showed higher to much higher radon content than waters from sites distant from the contact (table 3, fig. 1). A spring sample from the granitic terrane in the western part of the area contains 13,000 picocuries/liter. A stream sample taken a few hundred meters downstream from Butcher Lake contained very little radon (15.6 picocuries/liter). The radon content of waters in the area all appears high when compared to the data of Dyck and others (1971) for a uranium area in Saskatchewan, Canada. That area in Saskatchewan, however, was largely a sedimentary terrane with low levels of background uranium and radon. In that area the uranium deposits contributed significantly to radon levels in waters. In Maine, most granitic terranes appear to have very high radon levels in ground waters. Average ground waters in granitic terranes contain 22,100 picocuries/liter. The nearby Meddybemps granite pluton averages almost 40,000 picocuries/liter in six water wells (Hess and others, 1979). Use of radon as an exploration tool for uranium in granitic terranes in Maine is likely to be hampered by the very high radon contents (as much as 300,000 picocuries/liter), but radon perhaps could be

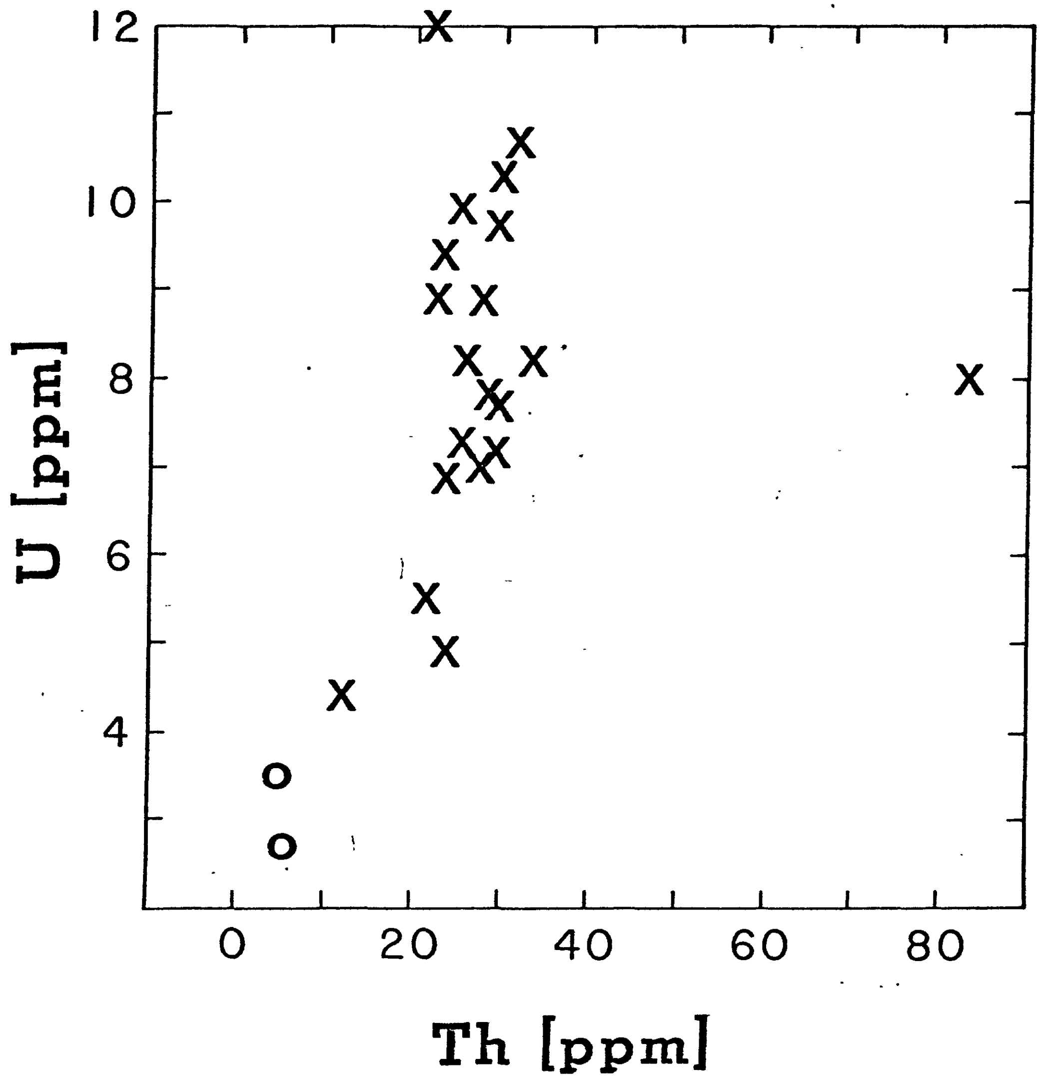

Figure 2.--Uranium versus thorium in samples of the Topsfield facies. X = Unaltered granite; O = Altered granite with molybdenite.

Table 3--Analyses of waters

[Radon analyses by W. H. Ficklin, D. J. Preston, A. R. Stanley, and J. B. McHugh. Analyses for radon, SiO_2^{--}, SO_4^{--}, Cl^-, HCO_3^-, F^-, pH, and specific conductance were done on untreated waters. Other analyses were performed on water filtered through a 0.45 m filter and then acidified to about pH2 with concentrated HNO_3. ---not determined]

Sample site	Sample type	Radon picocuries/ℓ	SiO_2 mg/ℓ	SO_4^{--} mg/ℓ	Cl^- mg/ℓ	HCO_3^- mg/ℓ	F^- mg/ℓ	pH^2	Spec. conduct. umhos/cm	Cu μg/ℓ	Zn μg/ℓ	Mo μg/ℓ	As μg/ℓ	U μg/ℓ	Na μg/ℓ	Co μg/ℓ	Ni μg/ℓ
1256	Stream	84	8	2.7	0.29	10	0.12	6.7	30	<1.0	2.1	1.9	5.6	<0.2	1.9	<1.0	<1.0
1257	Stream	58	8	2.0	0.12	12	0.05	6.6	30	<1.0	3.7	1.1	10.	<0.2	1.6	<1.0	<1.0
338	Seep	177	---	---	----	---	----	---	---	---	---	---	---	---	---	---	---
1258	Stream	---	8	2.9	0.41	9	0.07	6.9	27	<1.0	1.9	<1.0	4.1	<0.2	1.7	<1.0	<1.0
1259	Spring	13,000	8	4.4	0.90	4	0.25	6.1	24	<1.0	4.2	<1.0	<1.0	<0.2	2.0	<1.0	<1.0
1260	Stream	97	8	4.1	0.77	2	0.15	6.4	20	<1.0	1.2	<1.0	<1.0	<0.2	1.6	<1.0	<1.0
1261	Stream	---	8	3.7	0.49	5	0.12	6.5	25	<1.0	2.8	<1.0	<1.0	<0.2	1.7	<1.0	<1.0
1262	Stream	356	8	2.5	0.29	5	0.13	6.3	25	<1.0	1.4	<1.0	1.5	<0.2	1.8	<1.0	<1.0
1263	Stream	445	6	1.8	0.34	8	0.13	6.5	24	<1.0	1.7	<1.0	<1.0	<0.2	1.6	<1.0	<1.0
364	Stream	487	---	---	----	---	----	---	---	---	---	---	---	---	---	---	---
363	Stream	15.6	---	---	----	---	----	---	---	---	---	---	---	---	---	---	---

used to assess relative favorability for uranium of the various granites.

Uranium concentrations in the waters sampled were uniformly below detection limits (0.2 g/). Water samples from the granitic terrane appear to be high in SO_4^{--} and F^- content whereas those distant from the granite were slightly higher in specific conductivity, pH, Cl^-, and As. Trace-metal concentrations were generally low.

Discussion

The rock-geochemical data suggest that the Topsfield facies has a somewhat high uranium content that averages at least 8 ppm. The wide variation in Th/U over a restricted thorium content suggests that some uranium loss may have occurred locally. Substantial amounts of uranium appear to be entering the surficial environment from the granite because the uranium content of stream sediments over the granite is unusually high. The low uranium and thorium content in altered, molybdenite-bearing granite suggests that these elements were mobile during hydrothermal alteration of the rock.

The Topsfield facies to be part of the Acadian calc-alkalic suite of granites known throughout much of New England. It does not fit the characteristics of uraniferous two-mica granites (Boudette, 1977), which are a favored uranium exploration target in Maine. The uranium-molybdenum association apparent in the Topsfield suggests that it may be similar to other calc-alkalic plutons that are enriched in Cu, Mo, and(or) U such as the Katahdin Quartz Monzonite, Catheart and Debouillie plutons (E. Boudette, oral commun., 1980). Miller (1980) has noted that highly differentiated members of the biotite-hornblende granite suite in eastern Washington and northern Idaho appear to be the uraniferous granites rather than the two-mica granites.

The presence of muscovite in the Topsfield may suggest differentiation or late-stage deuteric alteration. The presence of molybdenite in altered granite, abundant shear zones, rusty quartz or quartz-pyrite stockwork, and elevated uranium contents in rocks and stream sediments suggests some favorability for Mo-U vein or contact metasomatic mineralization.

The high radon content in stream waters, the high uranium content in stream sediments, and the unusually low uranium content in stream waters suggest a model for the movement of uranium in the environment. Uranium leached from underlying country rock appears to be essentially completely removed from surface waters by some media in the sediment. The most likely mechanism is adsorption by iron or manganese hydroxides. Langmuir (1977, fig. 22) shows that uranium is essentially adsorbed by amorphous ferric oxyhydroxides in the pH range (6 to 7) for waters in these streams. Radon tends to persist in the waters until mixing with air removes it, until other surface or ground waters dilute it, or until it decays to other species. Mixing by wave action and the long residence time in the lake may explain the unusually low radon content in stream waters draining Butcher Lake (Dyck and others, 1971).

Table 4.--Analytical methods used in this study

Constituent	Method	Instrument or reference
Solid		
U, Th	Delayed neutron analysis	Millard, (1976).
Loss on ignition	Muffle furnace at 500°	
Waters		
Radon	Instrumental	Radon detector, model RD-200 E.D.A. Electronics, Ltd.
Silica	Molybdate blue	Brown, Skougstad, and Fishman (1970, p. 138-140).
Sulfate	Ion chromatography turbidimetric	Smee and Hall (1978).
Chloride	Gran's plot titration with silver nitrate	do.
Bicarbonate	Gran's plot titration with sulfuric acid	Orion Research, Inc. (1978, p. 6).
Fluoride	Ion chromatography	Smee and Hall (1978).
pH	Instrumental	
Specific conductance	Instrumental	
Copper	Flameless atomic absorption spectrophotometry	Miller and Ficklin (1976).
Zinc	do.	do.
Molybdenum	do.	do.
Arsenic	do.	Aruscavage (1977).
Uranium	Fluorometric	Ward and Bondar (1977).
Sodium	Flameless atomic absorption spectrophotometry	Brown, Skougstad, and Fishman (1970, p. 143).
Cobalt	do.	W. Ficklin (unpublished).
Nickel	do.	do.

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