Distribution of Thorium and Uranium in Three Early Paleozoic Plutonic Series of New Hampshire

By JOHN B. LYONS

CONTRIBUTIONS TO GEOCHEMISTRY

GEOLOGICAL SURVEY BULLETIN 1144-F

Prepared partly on behalf of the U.S. Atomic Energy Commission

UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON : 1964

UNITED STATES DEPARTMENT OF THE INTERIOR

STEWART L. UDALL, *Secretary*

GEOLOGICAL SURVEY

Thomas B. Nolan, *Director*

The U.S. Geological Survey Library has cataloged this publication as follows:

Lyons, John Bartholomew, 1916–

Distribution of thorium and uranium in three early Paleozoic Plutonic Series of New Hampshire. Washington, U.S. Govt. Print. Off., 1964.

iv, 43 p. diagrs., tables. 24 cm. (U.S. Geological Survey. Bulletin 1144–F)

Contributions to geochemistry.

Prepared partly on behalf of the U.S. Atomic Energy Commission.

Bibliography: p. 41–43.

1. Thorium—New Hampshire. 2. Uranium—New Hampshire. 3. Geology, New Hampshire. 4. Geology, Stratigraphic—Paleozoic. I. Title. (Series)

For sale by the Superintendent of Documents, U.S. Government Printing Office
Washington, D.C. 20402 - Price 25 cents (paper cover)

CONTENTS

ILLUSTRATIONS

TABLES

CONTRIBUTIONS TO GEOCHEMISTRY

DISTRIBUTION OF THORIUM AND URANIUM IN THREE EARLY PALEOZOIC PLUTONIC SERIES OF NEW HAMPSHIRE

By JOHN B. LYONS

ABSTRACT

Chemical uranium and thorium data were determined for the calc-alkaline Highlandcroft (Late Ordovician), Oliverian (Middle or Late Devonian(?)) and New Hampshire (Late Devonian(?)) Plutonic Series. Ten rock specimens were chemically and modally analyzed, and their constituent minerals analyzed chemically for uranium and thorium and spectrochemically for minor elements.

All series show normal radioactivities, progressive increases in uranium and thorium with increased content of felsic minerals, and no marked trends in thorium to uranium ratios. Highlandcroft rocks show a range of 2.6–5.0 ppm (parts per million) uranium, and 10.6–14.8 ppm thorium; Oliverian rocks 0.6–15.9 ppm uranium and 2.9–57 ppm thorium; New Hampshire rocks 1.5–39 ppm uranium and 2.8–28.5 ppm thorium. Pegmatites have abnormally low thorium to uranium ratios (but abnormally high uranium content), and aplites have high but variable uranium and thorium contents.

A linear relationship exists between K_2O and thorium, but not between K_2O and uranium, supporting the inference that uranium may be relatively volatile. An inverse nonlinear (but irregular) relation also exists between CaO and uranium (or thorium). High uranium and thorium accompany high SiO_2 and K_2O and low CaO.

In rocks of the Highlandcroft and Oliverian Series, both of which are regionally metamorphosed, as much as 67 percent of the uranium and 79 percent of the thorium may be held by sphene and epidote. Both minerals are absent or rare in the higher temperature unmetamorphosed New Hampshire Series, in which monazite and xenotime are characteristic accessories, and in which most of the uranium and thorium are dispersed among the major silicates. Scavenging of dispersed actinides by sphene and epidote is evidently one of the major side effects of regional metamorphism. Despite these differences in the distribution patterns of uranium and thorium, statistical analysis of the chemical and spectrochemical data demonstrates close associations, within the minerals of all the series, between uranium and thorium on the one hand, and rare earth or transition group metals on the other. These results, in turn, imply that there was no extensive addition or subtraction of uranium and thorium subsequent to the formation of the rocks.

Enrichment of the plutonic residuum in elements rejected by early crystallized silicates appears to be the simplest and best explanation of the uranium and thorium distribution pattern.

INTRODUCTION

On the basis of field relations and petrographic characteristics, Billings (1937) distinguished four groups of plutonic rocks in New Hampshire. The three older series—Highlandcroft (Late Ordovician), Oliverian (Middle or Late Devonian(?)), and New Hampshire (Late Devonian(?))—were typically calc-alkaline. The youngest series—the White Mountain Plutonic-Volcanic Series of Late Triassic or Early Jurassic age—had marked alkalic affinities. Isotopic determinations (Tilton and others, 1957; Hurley and others, 1960) demonstrate an age of 180 million years for this series.

A survey of the radioactivities of these four series was first made by Billings and Keevil (1946) using alpha counting techniques. They demonstrated: (a) that radioactivity generally increased with increasingly felsic character in all the series and (b) that, whereas the older series had normal radioactivities, the White Mountain Series was two to three times as radioactive as the older series; they suggested that this abnormality might be due to high concentrations of allanite.

A field reconnaissance survey of radioactivity in New Hampshire has been reported by McKeown (1951). Subsequently Whitfield and others (1959), Rogers and Ragland (1961), Butler (1962), Lyons (1962), and Adams and others (1962) published additional radiometric or chemical data. This paper reports in some detail the distribution of uranium and thorium in the Highlandcroft, Oliverian, and New Hampshire Series, and represents part of the U.S. Geological Survey's continuing study of the distribution of uranium and thorium in plutonic complexes.

METHODS OF STUDY

In addition to a Geiger counter survey of numerous outcrops and roadcuts, which proved unrewarding, a collection was made of approximately 135 fresh representative specimens of the Highlandcroft, Oliverian, and New Hampshire Series. All these samples were chemically analyzed for uranium, and 65 were also analyzed for thorium. In addition, one large (30–35 lb) sample of the Highlandcroft, five large samples of the Oliverian, and four large samples of the New Hampshire Series were chemically and modally analyzed (table 1) and were separated by standard techniques into their constituent minerals. These minerals, if recovered in sufficient quantity, were analyzed for uranium and thorium; the major and minor elements of the minerals were determined by semiquantitative spectrographic methods.

Techniques used by Geological Survey chemists for determining

TABLE 1.—*Chemical analyses, norms, and modes of Highlandcroft, Oliverian, and New Hampshire Plutonic Series rocks*

[Analyses by P. L. D. Elmore, K. E. White, and S. D. Botts. Sample number in parentheses]

Constituent	Highland-croft	Oliverian					New Hampshire			
	Highland-croft granodiorite (NH/L 1-52)	Lebanon granodiorite (NH/H 6-52)	Lebanon Granite (NH/H 1-52)	Lebanon aplite (NH/H 2-52)	Mascoma Group of Chapman and others (1938) (quartz monzonite) (NH/M 50-52)	Oliverian quartz monzonite (NH/MW 4-52)	Bethlehem Gneiss (NH/S 4-52)	Kinsman Quartz Monzonite (NH/LM 1-52)	Concord Granite (NH/C 4-52)	Concord pegmatite (NH/C 5-52)
Analyses (weight percent)										
SiO_2	60.5	58.3	73.3	77.2	72.2	74.5	69.0	68.6	70.2	76.2
Al_2O_3	15.5	16.1	14.3	12.7	14.9	14.2	15.1	15.2	16.0	14.7
Fe_2O_3	3.3	2.9	1.2	.78	.7	1.0	.6	1.1	1.1	.55
FeO	2.7	4.1	.57	.12	1.0	.26	3.6	3.4	1.0	.26
MgO	3.7	4.0	.42	.06	.74	.25	1.2	1.4	.66	.25
CaO	5.1	5.6	1.2	.56	1.7	1.1	2.7	2.6	1.6	.29
Na_2O	2.5	2.7	3.3	2.6	3.5	3.8	3.2	2.8	4.0	5.0
K_2O	4.0	3.5	5.3	6.1	4.4	4.9	2.9	3.2	4.6	2.3
TiO_2	.46	.56	.14	.06	.19	.16	.68	.90	.44	.02
P_2O_5	.22	.24	.04	.00	.07	.02	.22	.28	.23	.08
MnO	.12	.14	.02	.01	.05	.02	.06	.04	.02	.30
H_2O	1.4	1.1	.38	.18	.42	.28	.80	.62	.43	.62
CO_2	.05	.05	.12	.19	.05	.05	.16	.16	.14	.06
Total	99.6	99.3	100.3	100.6	99.9	100.5	100.2	100.3	100.4	100.6

TABLE 1.—*Chemical analyses, norms, and modes of Highlandcroft, Oliverian, and New Hampshire Plutonic Series rocks*—Continued

Constituent	Highland-croft	Oliverian					New Hampshire			
	Highland-croft granodiorite	Lebanon granodiorite	Lebanon Granite	Lebanon aplite	Mascoma Group of Chapman and others (1938) (quartz monzonite)	Oliverian quartz monzonite	Bethlehem Gneiss	Kinsman Quartz Monzonite	Concord Granite	Concord pegmatite
	(NH/L 1-52)	(NH/H 6-52)	(NH/H 1-52)	(NH/H 2-52)	(NH/M 50-52)	(NH/MW 4-52)	(NH/S 4-52)	(NH/LM 1-52)	(NH/C 4-52)	(NH/C 5-52)
Norms (weight percent)										
Quartz	15.0	10.52	31.32	37.91	31.80	31.08	30.12	31.98	25.92	37.68
Orthoclase	23.9	20.57	31.14	36.14	26.13	28.91	17.24	18.90	27.24	13.34
Albite	21.0	23.06	27.77	22.01	29.34	31.96	27.25	23.58	34.06	42.44
Anorthite	19.2	21.41	5.00	1.39	7.51	5.56	11.40	10.01	6.39	.28
Corundum				1.33	1.63	.61	2.45	3.47	2.04	3.88
Diopside:										
Wollastonite	4.0	2.32								
Enstatite		1.50								
Ferrosilite		.66								
Hypersthene:										
Enstatite	8.9	8.50	1.00	.15	1.80	.60	3.00	3.50	1.60	.60
Ferrosilite		3.83			.92		5.02	3.83	.26	.53
Magnetite	4.9	4.18	1.39		.93	.46	.93	1.62	1.62	.93
Hematite			.32	.80		.64				
Ilmenite	.91	1.06	.30	.15	.46	.30	1.37	1.67	.76	
Apatite	.34	.34			.34		.34	.67	.34	.34
Calcite	.10		.30	.50			.40	.40	.30	.10
H_2O	1.4	1.10	.38	.18	.42	.28	.16	.16	.14	.06
Total	99.7	99.1	99.0	100.6	101.3	100.4	99.7	99.8	100.7	100.2

Modes (volume percent)

Quartz	20.2	17.5	26.1	36.5	29.9	29.8	36.8	33.2	30.2	27
Potassium feldspar	17.1	7.8	35.7	34.9	17.8	32.9	9.1	1.5	26.7	23
Plagioclase	30.0	30.3	20.2	23.2	45.3	34.3	39.8	35.5	31.8	47
Hornblende	20.5	4.6								
Biotite and chlorite	5.6	25.3	3.9		4.5	.8	12.2	13.2	6.7	
Muscovite			8.3	3.6	.5	1.2	1.3	15.7	3.3	3
Garnet					Tr.			.20		.90
Epidote and allanite	4.3	11.9	4.9	.9	2.4	.1	Tr.		.002	
Sphene	.1	.2	.3	.4	.11	.3				
Apatite	.35	.3	.03	Tr.	.07	.03	.20	.30	.30	.003
Zircon	.002	.014	.01	.01	.015	.01	.01	.01	Tr.	Tr.
Monazite							.01	.007	.01	
Xenotime							.002		Tr.	.03
Magnetite	3.3	.3	.25	.1	.33	1.20	.30	1.20	.20	.003
Ilmenite		Tr.	Tr.	Tr.	Tr.	Tr.	.10	.50	.10	Tr.
Hematite				Tr.						Tr.
Pyrite		.01	.2	Tr.		.003	.02		.01	
Molybdenite	Tr.	Tr.	Tr.	Tr.		Tr.	Tr.	Tr.		
Graphite								.20		
Rutile								Tr.	Tr.	Tr.
Fluorite				Tr.						
Calcite	Tr.									
Tourmaline		Tr.				Tr.				
Anorthite content of plagioclase (weight percent)	8	23	11	10	20	18	33	30	22	6

730-708—64—2

the amount of uranium (Grimaldi and others, 1952) and thorium (Levine and Grimaldi, 1958) yield results of high precision. Consequently, where discrepancies have arisen (for example, tables 3, 7, and 11) in comparing uranium and thorium data derived by different sampling procedures, the anomalies are considered to be due to factors such as sample inhomogeneity or failure to identify or isolate all the mineral phases (only the 60–200 mesh size range was analyzed), that is, due to errors in determining absolute amounts of minerals.

ACKNOWLEDGMENTS

The writer is grateful to Professor E. S. Larsen, Jr., who initiated and directed these studies, and to George Phair, David Gottfried, and E. S. Larsen, 3d, all of the U.S. Geological Survey, for their assistance and guidance. He is particularly indebted to the chemists of the U.S. Geological Survey upon whose analytical work this report is based. Rock analyses were made by P. L. D. Elmore, K. E. White, and S. D. Botts; uranium analyses by Marian Schnepfe; thorium analyses by Esma Campbell; semiquantitative spectrographic analyses by Joseph Haffty; and radiometric analyses by F. J. Flanagan. Most of the work was done on behalf of the Division of Research of the U.S. Atomic Energy Commission.

HIGHLANDCROFT PLUTONIC SERIES

The Highlandcroft Plutonic Series occurs only as a few stocks and dikes in westernmost New Hampshire and easternmost Vermont. It was emplaced during the Taconic orogeny, and has been regionally metamorphosed to the green-schist facies during the Acadian orogeny.

Table 2 lists the uranium and thorium analytical data for rocks of the Highlandcroft Plutonic Series. The mean uranium and thorium contents of the quartz monzonite are 3.7 and 12.6 ppm, respectively.

TABLE 2.—*Uranium and thorium contents of some Highlandcroft Plutonic Series rocks*

Description	Sample	Location	Uranium (ppm)	Thorium (ppm)	Thorium: uranium
Highlandcroft granodiorite.	NH/L 1-52	Littleton quadrangle.	3.0 2.9	11.8	3.9
Fairlee Quartz Monzonite.	NH/MC 1-52	Mount Cube quadrangle.	5.0 4.7		
	NH/MC 2-52	do	3.5 3.1		
	NH/MC 3-52	do	4.4 4.4	14.6 14.8	3.3
	NH/MC 4-52	do	3.9 4.3 4.6 4.9	12.7	2.9
	NH/MC 5-52	do	2.7 2.6		
	NH/MC 6-52	do	2.7 2.6	10.6	3.9
Sodaclase tonalite	NH/H 5-52	Hanover quadrangle.	3.1 3.0	11.3	3.6

The granodiorite and sodaclase-tonalite each contain 3 ppm (parts per million) uranium and about 11.5 ppm thorium. The mean thorium to uranium ratios for the quartz monzonite, the granodiorite, and the sodaclase tonalite are 3.4, 3.9, and 3.6 respectively. The absolute amounts and the ranges in the thorium and uranium values are in excellent agreement with published data for rocks of similar petrographic character (Larsen and Gottfried, 1960; Larsen and Gottfried, 1961; Adams and others, 1959).

Uranium and thorium abundances in the Highlandcroft granodiorite minerals are shown in table 3. In this and subsequent similar tabulations, the weight percent of the major mineral fractions is

TABLE 3.—*Uranium and thorium contents of minerals of the granodiorite in the Highlandcroft Plutonic Series (sample NH/L 1–52)*

[Marian Schnepfe and Esma Campbell, analysts. Mwp, mineral weight percent of sample]

Mineral	Mineral weight percent of sample	Uranium		Thorium		Thorium: uranium ratio
		Parts per million	$\frac{\text{Mwp}\times\text{U(ppm)}}{100}$	Parts per million	$\frac{\text{Mwp}\times\text{Th(ppm)}}{100}$	
Quartz	19.3	0.7	0.13	2.6	0.50	3.7
Microcline	15.7	.4	.06	3.5	.55	8.7
Plagioclase	28.7	.8	.23	2.6	.75	3.2
Hornblende	23.1	1.6	.37	7.9	1.83	4.9
Biotite and chlorite	5.6	1.8	.10			
Epidote	4.3	5.4	.23	130	5.20	24
Sphene	.1	586	.59	360	.36	0.6
Apatite	.35	30	.10			
Zircon	.002	342	.01			
Magnetite	3.3	.28	.01			
Pyrite	Tr.					
Molybdenite	Tr.					
Calcite	Tr.					
Total uranium (by sum of minerals)			1.83			
Total uranium in bulk rock (measured)			3.0			
Total thorium (by sum of available analyses)					9.2	
Total thorium in bulk rock (measured)					11.8	
Thorium: Uranium for bulk rock (measured)						3.9

determined by adjusting the mode for mineral densities. Minor minerals are determined by weighing the separated mineral fractions. The quantity of accessories recovered by heavy-mineral separations is generally lower than is indicated by the norm, because of inaccuracies of the modal determination of trace constituents, inefficiencies of mineral recovery, and probable loss of some accessories in the minus 200-mesh fraction.

An estimate of the efficiency of recovery of the accesssory minerals is provided by the modes of table 1 and the by spectrochemical analyses of tables 4, 8, and 12. Mass balance calculations for the five analyzed rocks of the Oliverian Series yield an average zirconium content of 90 ppm (range 74–116 ppm), which is appreciably less than the 120 ppm

average for five Oliverian granitic rocks reported by Billings and Rabbitt (1947). The data are not precise, but indicate that a third or more of the zircon is probably not recovered. Presumably monazite, sphene, and other minerals have comparably poor percentage recoveries.

Two minerals, sphene and epidote, contain 45 percent of the total uranium and 60 percent of the total thorium in Highlandcroft rocks. These figures may be contrasted with data from several Mesozoic batholiths in the Western United States (Larsen, Jr., and Gottfried, 1961) in which only 10 to 30 percent of the total uranium is found in the accessory minerals. Comparable data for thorium are not available.

Epidote in the Highlandcroft rocks forms metamorphically by the breakdown of plagioclase. Sphene occurs in two habits: in large crystals as an apparent primary accessory mineral and in very fine-grained granular aggregates associated with chlorite. In the latter occurrence, the sphene is apparently of metamorphic origin and has formed as a side reaction product in the breakdown of hornblende and biotite. Metamorphism is therefore indicated to have had a

TABLE 4.—*Semiquantitive spectrographic analyses of minerals of the granodiorite of the Highlandcroft Plutonic Series*

[Joseph Haffty, analyst]

Weight Percent	Element(s) in mineral indicated				
	Quartz	Plagioclase	Microcline	Hornblende	Epidote
>10	Si	Si	Si	Si	Si, Al
5–10			K		Ca
1–5		Al, K, Na	Al	Mg, Ca, Al, Fe	
0.5–1	Al		Na		Fe
0.1–0.5	Na, Ca, Fe	Ba, Ca	Ba, Ca	K, Na, Ti	Mg, Ti, Sr, Na
0.05–0.1	Mg	Fe, Sr, Mg	Fe, Sr	Sr	
0.01–0.05	Ba, Ti, B		Mg	Mn, Ba, Ni, Cr, Zr, V	Mn, Cr, Ba, Ga, B, V
0.005–0.01	Sr, Mn		B	La, Cu	
0.001–0.005	Cr, V, Zr, Cu	Ti, Cr, Mn, Ga, Zr	Ti, Cr, Ga, Mn	Co, Ga, Sc, Y, Pb	Pb, La, Cu, Zr, Y, Ni, Sc
0.0005–0.001					
0.0001–0.0005		Cu	Cu	Be	

Weight Percent	Element(s) in mineral indicated				
	Sphene	Chlorite	Magnetite	Apatite	Zircon
>10	Si, Ca, Ti	Si	Fe	Ca, P	Zr, Si
5–10		Al			
1–5		Mg, Fe	Si		
0.5–1	Al	K			
0.1–0.5	Ce, Fe, Mg Nd, Th	Ca, Na, Ti	Al, Mg	Si, Ce, Al, Mg, La	Hf, Ti, Al, Ca, Y
0.05–0.1	La, Y, Zr		Ca, Cu	Na, Fe, Sr	
0.01–0.05	V, Na, Sr, Mn, Sn, Ba, Cr, Gd	Ba, Mn, Ni, Si, Cr	Ni, Mn, Cr, Na, Ba, Sn, Ti	Zr, Nd, Ti, Mn, Y	Fe, Mg, Er, Yb, Lu
0.005–0.01	Dy	Cu, Ga, Co	Co, V, Sr	Ba, Er	B, Mn
0.001–0.005	Er, Pb, Lu, Yb, Mo, Cu	V, Y, Pb, Zr, Sc	Zr	Ni, Cr, Pb, Cu, V, Sc	Sc, Pb, Cr, Cu, Sr, V
0.0005–0.001				Yb	
0.0001–0.0005					

strong effect in redistributing thorium and uranium (see table 3) in rocks of this series.

To examine the relationship of minor elements to uranium and thorium distributions, the minerals of table 3 were analyzed by semiquantitative spectrochemical methods (table 4). Results were investigated statistically by using the rank correlation coefficient method as outlined by Flanagan (1957). Significant (at the 95 percent level) correlations are indicated between uranium and zirconium, yttrium, cerium, neodymium, lanthanum, thorium, lutetium, dysprosium, ytterbium, lead, molybdenum, and gadolinium. Thorium shows significant associations with calcium, yttrium, lanthanum, lead, and chromium. The results are interpreted in pages 35 to 38.

OLIVERIAN PLUTONIC SERIES

The Oliverian Plutonic Series has been emplaced in a series of elongate en echelon domes, approximately parallel to the course of the Connecticut River throughout western New Hampshire. The estimated areal abundances of the various petrographic types are given by Billings and Keevil (1946, p. 816) as quartz diorite 10 percent, granodiorite 30 percent, granite 17 percent, and syenite 13 percent. Field relations are consistent with the Middle and Late Devonian(?) age for the series.

The individual results for the Oliverian Series are listed in table 5 and are summarized in table 6. The analyses show the expected progression in amounts of both uranium and thorium as the rocks become increasingly felsic, but no significant trend in thorium to uranium ratios. Wallrocks (quartz-mica schists and amphibolites) have radioactivities approximating those of quartz diorites. For the series as a whole, the ranges and abundances of uranium and thorium agree with those found in other calc-alkaline complexes (Whitfield and others, 1959; Larsen and Gottfried, 1960). Analyses reported here for rocks from the Mascoma and Lebanon domes also agree reasonably well with those reported from these two plutons by Whitfield and others (1959) and Rogers and Ragland (1961).

Although the Oliverian Series has normal thorium and uranium contents, there is seemingly marked variability from pluton to pluton. Granite of the Lebanon dome (8.3 ppm uranium; 26 ppm thorium), for example, is far more radioactive than that of the Smarts Mountain pluton (2.4 ppm uranium, 10 ppm thorium). These results have no apparent explanation and require statistical confirmation. The most uraniferous Oliverian rock (Lebanon Granite with 11.7 ppm uranium) is heavily pyritized, but there is no other indication that hydrothermal conditions are involved in concentrating uranium in rocks of this series.

TABLE 5.—*Uranium and thorium contents of some Oliverian Plutonic Series rocks*

[Marion Schnepfe, Marjorie Malloy, Esma Campbell, Roosevelt Moore, and Alice Caemmerer, analysts]

Description	Sample	Quadrangle	Uranium (ppm)	Thorium (ppm)	Thorium: uranium ratio
Oliverian quartz diorite	NH/K 1-52	Keene	1.3 1.5	8.1 8.3	5.9
Oliverian quartz diorite, hydrothermally altered.	NH/K 2-52	do	1.4 1.6		
Oliverian quartz monzonite	NH/K 3-52	do	2.9 3.3	6.2	2.0
Oliverian quartz diorite	NH/K 4-52	do	2.5 2.6 3.9 4.5	9.2	2.7
	NH/K 5-52	do	1.4 1.5		
	NH/K 6-52	do	1.8 1.9		
	NH/K 8-52	do	1.1 1.1		
	NH/K 9-52	do	.80 .80 .88 .90	3.1	3.5
Amphibolite dike(?) in Oliverian gneiss.	NH/K 10-52	do	.40 .50 .80		
Oliverian gneiss (granite to quartz monzonite).	NH/BF 5-52	Bellows Falls	1.5 1.6 2.5		
Oliverian gneiss (quartz monzonite).	NH/BF 6-52	do	1.1 1.1	4.1	3.7
Oliverian gneiss (granite)	NH/BF 7-52	do	1.4 1.8 2.7	7.5	3.9
Croyden Group:[1]					
Pegmatitic lens	NH/S 1-52	Sunapee	1.3 1.4	3.6	2.8
Granodiorite	NH/S 2-52	do	4.1 4.3		
Quartz diorite	NH/S 3-52	do	2.7 2.9		
Granodiorite	NH/M 23-52	Mascoma	2.5 2.5		
Pegmatite near amphibolite septum.	NH/M 23-52	do	11.0 11.6 14.6	4.7	.38
Aplite	NH/M 26-52	do	2.0 2.2	5.5 5.4	2.6
Oliverian gneiss (quartz diorite(?)).	NH/CL 1-52	Claremont	1.8 1.8		
Ammonoosuc Volcanics	NH/CL 2-52	Claremont	1.3 1.3		
	NH/CL 3-52	do	1.2 1.4	4.8 4.6	3.6
	NH/CL 4-52	do	.6 .6		
Lebanon Granite	NH/H 1-52	Hanover	8.0 9.0	32	3.8
Aplite in Lebanon Granite	NH/H 2-52	do	9.4 9.6 11.0 7.5 7.6	55	6.2
Lebanon Granite, pyrite rich	NH/H 3-52	do	12.9 13.1 10.5 10.4	22	1.85
Lebanon Granite	NH/H 4-52	do	4.8 4.8		
Lebanon granodiorite	NH/H 6-52	do	2.1 2.1 2.5 2.5	10.0 10.8	4.5
Mascoma Group:[2]					
Granodiorite	NH/M 1-52	Mascoma	.8 .8		
Granite	NH/M 3-52	do	7.6 8.0	12.1	1.55

See footnotes at end of table.

TABLE 5.—*Uranium and thorium contents of some Oliverian Plutonic Series rocks*—Continued

Description	Sample	Quadrangle	Uranium (ppm)	Thorium (ppm)	Thorium: uranium ratio
Mascoma Group—Continued					
Granodiorite	NH/M 4-52	Mascoma	4.1 4.2 4.8		
Granodiorite	NH/M 5-52	do	1.0 1.1 1.6 1.3	6.2 6.9	5.3
Quartz diorite	NH/M 5-52	do	1.7 1.8		
Quartz diorite	NH/M 9-52	do	1.1 1.4 1.6 1.6	2.9	2.1
Granodiorite	NH/M 12-52	do	1.8 1.8	11.0	6.1
Quartz diorite	NH/M 13-52	do	2.0 2.1		
Granodiorite	NH/M 14-52	do	1.3 1.3	6.9	5.3
Granodiorite	NH/M 15-52	do	2.8 2.9		
Granite	NH/M 16-52	do	3.3 3.4 3.1 3.1	25	7.6
Quartz monzonite	NH/M 17-52	do	5.3 5.3	17.5	3.3
Granodiorite	NH/M 18-52	do	1.0 1.1		
Quartz monzonite	NH/M 50-52	do	3.8	15.3 15.8	4.1
Granodiorite	NH/MC 9-52	Mount Cube	2.0 2.2	38	18.0
	NH/MC 10-52	do	1.5 1.6		
	NH/MC 11-52	do	1.5 1.6		
Quartz diorite	NH/MC 12-52	do	1.3 1.3		
Mica schist inclusion in Mascoma Group quartz diorite.	NH/M 7-52	Mascoma	1.1 1.3		
	NH/M 10-52	do	1.6 1.9		
Ammonoosuc Volcanics (amphibolite schist) at contact with Mascoma Group granodiorite.	NH/M 2-52	do	1.7 1.9		
Smarts Mountain Granite [3]	NH/M 19-52	do	1.7 1.8 1.8 2.0	10.4	5.8
	NH/M 20-52	do	1.6 2.4 2.1	9.8	4.9
Granodiorite	NH/MC 14-52	Mount Cube	.8 .9		
	NH/MC 15-52	do	3.6 4.5 5.4		
Baker Pond Gneiss: [4] Quartz diorite	NH/MC 18-52	do	3.6 3.7		
Granite	NH/MC 19-52	do	5.8 6.1	26	4.3
Quartz diorite	NH/MC 20-52	do	1.7 1.8	4.6	2.6
Granite	NH/R 1-52	Rumney	4.7 5.2 6.8		
	NH/R 2-52	do	8.9 9.6		
Quartz diorite	NH/R 3-52	do	4.9 5.1	14.8	3.0

See footnotes at end of table.

TABLE 5.—*Uranium and thorium contents of some Oliverian Plutonic Series rocks*—Continued

Description	Sample	Quadrangle	Uranium (ppm)	Thorium (ppm)	Thorium: uranium ratio
Owls Head Granite [5]	NH/R 13-52	Rumney	5.8 6.5	21	3.4
Aplite dike in Owls Head Granite.	NH/R 14-52	do	13.8 14.8 19.2	58 56	3.6
Fine-grained quartz monzonite.	NH/MW 2-52	Mount Washington	3.4 3.6	21	6.0
Biotite quartz monzonite	NH/MW 3-52	do	2.2 2.4		
Coarse granite	NH/MW 4-52	do	5.4 5.2 5.0 5.4 5.7 5.6	27	4.9

[1] Croyden Group of Chapman and others (1938).
[2] Mascoma Group of Chapman and others (1939).
[3] Smarts Mountain Granite of Fowler-Lunn and Kingsley (1937).
[4] Baker Pond Gneiss of Hadley and others (1938).
[5] Owls Head Granite of Billings (1935).

TABLE 6.—*Mean uranium and thorium contents of the major rock types of the Oliverian Plutonic Series*

Rock type	Uranium			Thorium			Mean thorium: uranium ratio [1]
	Samples analyzed	Parts per million		Samples analyzed	Parts per million		
		Mean	Range		Mean	Range	
Granite	13	5.7	1.8–11.7	10	19.3	7.5–32	4.2
Quartz monzonite	8	3.1	1.1–5.3	6	12.7	4.1–21	4.1
Granodiorite	15	2.2	.8–4.6	5	14.6	6.6–38	7.8
Quartz diorite	16	2.0	.8–5.0	5	7.6	2.9–14.8	3.4
Gabbro (amphibolite)	1	.6	.6				
Pegmatite	2	6.4	1.3–12.4	2	4.2	3.6–4.7	1.6
Aplite	3	9.0	2.1–15.9	3	39.2	5.5–57	4.1
Wallrocks and inclusions	6	1.3	.6–1.8	1	4.7		3.6

[1] Averages of the individual thorium to uranium ratios, not the ratios of the average thorium to the average uranium contents of all specimens.

The pegmatite samples merit discussion. One (NH/S 1–52) is a segregation in the gneiss of the Croyden Group of Chapman and others (1938), and has uranium and thorium contents and a thorium to uranium ratio typical of the quartz diorite enclosing it. The other (NH/S 23–52) is a pegmatite vein and has an abnormally low (0.4) thorium to uranium ratio such as characterizes pegmatites of the New Hampshire Series. Note that the aplites, by contrast, have high uranium and thorium contents, but a normal thorium to uranium ratio.

Minerals of Oliverian Series rocks were purified (in the 60–200 mesh size range) and analyzed chemically for uranium and (if available in sufficient amount) for thorium. Results of these analyses and a calculation of the contribution made by each mineral to the

total uranium and thorium contents of the rock are shown in table 7. Comparisons, in table 7, of uranium and thorium contents of bulk rock samples to uranium and thorium contents computed from the mineral data of table 7 are disappointingly inconsistent. The major difficulty is probably the fact that mineral recoveries (particularly for the accessory minerals) are not accurately representative of the rock. It is notable in the Oliverian Series rocks that between 45 and 67 percent of the uranium and 49 to 79 percent of the thorium shown by the mineral analyses are in the epidote and sphene.

Both field and petrographic evidence show that the Oliverian Series has been metamorphosed: the texture is commonly granoblastic, amphibolites (that is, metagabbros) occur within or peripheral to the main units of the series, and the plagioclase is crowded with epidote and sericite. A small amount of allanite in some rocks of the Oliverian Series is possibly a primary relict accessory, but the bulk of the epidote is undoubtedly metamorphic. Sphene in these rocks is also, in part, a primary relict accessory, but about 50 percent occurs as typical fine-granular metamorphic aggregates associated with epidote and recrystallized (?) green-brown biotite. As in the Highlandcroft rocks, it is apparent that metamorphism has effected a major redistribution in the uranium and thorium of the Oliverian Series.

The group of analyses of the Lebanon granodiorite, granite, and aplite show a parallel relation between the total uranium in each rock and the amounts of uranium in the rocks' respective minerals. This relation is true, evidently, for silicates in which uranium is structurally bonded (for example, zircon), as well as for those in which it is not (for example, quartz and feldspar).

Relationships in Oliverian Series rock-forming minerals of uranium and thorium to minor elements were investigated by computing rank correlation coefficients for the semiquantitative spectrographic analyses which are listed in table 8.

Elements that show consistently strong positive correlation (at the 95-percent confidence level) with uranium are zirconium, yttrium, lutetium, ytterbium, and erbium. Less consistent positive correlations are indicated by titanium, scandium, lead, thorium, hafnium, neodymium, lanthanum, samarium, beryllium, and copper. A negative correlation (95-percent confidence level) is established for sodium in two cases and is very nearly proven in the other three. With the exception of titanium, there is no suggestion of a correlation between uranium and any of the major rock-forming elements.

Because of fewer analyses, the statistical results for thorium are somewhat less satisfactory. No correlation can be established at the 95-percent level of significance; at the 90-percent level, zirconium,

TABLE 7.—*Uranium and thorium contents of minerals of the Oliverian Plutonic Series rocks*

[Mwp, mineral weight percent of sample. Analyses by Marian Schnepfe and Esma Campbell]

Mineral	Lebanon granodiorite (NH/H 6-52)						Lebanon Granite (NH/H 1-52)					
	Mineral weight percent of sample	Uranium		Thorium		Th:U	Mineral weight percent of sample	Uranium		Thorium		Th:U
		Parts per million	Mwp×U (ppm)/100	Parts per million	Mwp×Th (ppm)/100			Parts per million	Mwp×U (ppm)/100	Parts per million	Mwp×Th (ppm)/100	
Quartz	16.4	0.25	0.041	4.9	0.81	19.6	24.8	0.85	0.21	3.7	0.92	4.4
Potassium feldspar	7.3	.25	.018				34.9	1.1	.38	4.6	1.6	4.2
Plagioclase	32.2	.25	.081	4.9	1.6	19.6	20.4	.85	.17	3.7	.76	4.4
Hornblende	5.1	.84	.043									
Biotite Chlorite	26.8	1.11	.31	3.4	.92	3.1	4.3	3.4	.15	5.6	.24	1.6
Muscovite							9.1	[1] 3.4	.31			
Epidote	11.9	11.7	1.39	110	13.1	9.4	6.1	63.0	3.84	200	12.2	3.2
Sphene	.2	220	.44				.3	285	.86	88	.26	.31
Apatite	.3	11.1	.33	17	.03	1.5	.03	24.4	.01			
Zircon	.014	466	.053				.01	1,385	.14			
Magnetite	.3	2.34	.007				.25	5.2	.01			
Pyrite	.01						.02	178	.04			
Allanite							Tr.					
Ilmenite	Tr.						Tr.					
Tourmaline	Tr.											
Molybdenite	Tr.						Tr.					
Fluorite												
Hematite												
Garnet												
Total uranium (by sum of minerals)			2.73						6.0			
Total uranium (chemical)			2.5						8.5			
Total thorium (by sum of available analyses)					16.5						15.9	
Total thorium in bulk rock (measured)					10.4						32	
Total thorium: uranium in bulk rock (measured)						4.5						3.8

[1] Estimated.

TABLE 7.—*Uranium and thorium contents of minerals of the Oliverian Plutonic Series rocks*—Continued

Mineral	Lebanon aplite (NH/H 2-52)			Mascoma group of Chapman and others (1938) (quartz monzonite; NH/M 50-52)						Oliverian quartz monzonite (NH/MW 4-52; Mount Washington)		
	Mineral weight percent of sample	Uranium		Mineral weight percent of sample	Uranium		Thorium		Th:U	Mineral weight percent of sample	Uranium	
		Parts per million	MwpXU (ppm) / 100		Parts per million	MwpXU (ppm) / 100	Parts per million	MwpXTh (ppm) / 100			Parts per million	MwpXU (ppm) / 100
Quartz	36.7	1.45	0.53	30.0	0.82	.25	2.6	.78	3.2	28.8	0.49	0.14
Potassium feldspar	34.2	1.45	.48	17.4	.45	.08	2.9	.51	6.4	30.9	.34	.11
Plagioclase	23.6	2.3	.54	45.2	.82	.37	2.6	1.2	3.2	33.0	.63	.21
Hornblende				n.d.						n.d.		
Biotite	}			5.2	5.8	.30	44	2.3	7.6	.8	24.6	.20
Chlorite	}									.1	14.9	.02
Muscovite	4.1			.3						1.3		
Epidote	1.1	204	.22	2.4	55.7	1.34	150	3.6	2.7	.1		
Sphene	.4	322	1.29	.11	300	.33	900	1.0	3.0	.3	411	1.23
Apatite	Tr.			.07	14.6	.01	16	.01	1.1	.03	26.3	.01
Zircon	.01	2,770	.28	.015	1,080	.16				.01	885	.09
Magnetite	.1	25.7	.03	.33	1.0	.00	7.8	.03	7.8	1.2	2.4	.03
Pyrite	Tr.									.003		.05
Allanite	.004	266	.01									
Ilmenite	Tr.			Tr.						Tr.		
Tourmaline												
Molybdenite	Tr.									Tr.		
Fluorite	Tr.									Tr.		
Hematite	Tr.									Tr.		
Garnet				Tr.								
Total uranium (by sum of minerals)			3.38			2.83						2.29
Total uranium (chemical)			10.0			3.8						5.7
Total thorium (by sum of available analyses)								9.4				
Total thorium in bulk rock (measured)								15.5				
Total thorium: uranium in bulk rock (measured)									4.1			

TABLE 8.—*Semiquantitative spectrographic analyses of minerals of Oliverian Plutonic Series rocks*

[Joseph Haffty, analyst]

Weight percent	Element(s) in mineral indicated							
	Granodiorite of the Lebanon Granite (NH/H 6-52)							
	Quartz and plagioclase	Horneblende	Biotite	Sphene	Epidote	Magnetite	Apatite	Zircon
>10	Si	Si	Si	Ca, Ti, Si	Al, Si	Fe	Ca, P	Zr, Si
5–10			K, Al		Ca			
1–5	Al, K, Na	Mg, Fe, Ca, Al	Fe, Mg	Al	Fe	Si		Ti
0.5–1						Al		
0.1–0.5	Ba, Ca	K, Na	Ti, Ca	Fe, Zr, Mg	Ti, Mg	Mg	Si, Al, Ce	Hf, Ca, Al, Y
0.05–0.1	Fe, Sr		Na, Ba		Sr	Ca, Na, Cu	Fe, La, Sr, Mg, Ti	Mg, Fe
0.01–0.05	Mg	Mn, Ti, Zr, Sr, B	Mn, Cr, B	Hf, Na, Sr, Mn, Nd, Ba, Cr, Y, V	Mn, V, Cr, Ga, Na, Pb, Nd, La	Mn, Ti, Ni, Cr, Sn	Nd, Y, Na, Mn, Zr	Lu, B
0.005–0.01	B	Cr, Ba	Ni, Co, Sr, Ga		Sc, Y, Ba	Co, V, Ba, Pb	Er	Mn, Er, Yb
0.001–0.005	Cr, Mn, Pb, Ga Ti	V, Ga, Ni, Y, Sc, Co, Cu, Pb	V, Cu, Y, Zr, Pb	Cu, Er, Sn, Pb, Lu, Yb, Ga, Sc	Zr, Sn, Cu	Sr, Zr	Ba, Cu, Pb, Cr, Yb	Pb, Sc
0.0005–0.001								Cr
0.0001–0.0005	Cu					Ag		
0.00005–0.0001								
0.00001–0.00005								Cu

Lebanon Granite (NH/H 1-52)

	Quartz and plagioclase	Potassium feldspar	Biotite	Epidote	Magnetite	Sphene	Apatite	Pyrite	Zircon
>10	Si	Si, Al, K	Fe	Si, Al	Fe	Ca, Ti, Si	Ca, P	Fe	Zr, Si
5-10			Al, Si	Ca					
1-5	Al	Na	K, Mg	Fe		Al			
0.5-1	Na		Ti						
0.1-0.5	Ca	Ca, Ba	Ca, Na, Mn	Sr, Ti, Mg	Al, Si	Fe, Zr, Mg, Y	Ce, Al, Si, Y	Al	Hf, Al, Ca, Y
0.05-0.1				Mn, Na	Mg, Mn	Nd	Nd, La, Sr	Co	Th
0.01-0.05	Fe, Mg, B	Sr, Fe, B	Li, Ba, Zn, V	Ce, La, V, Nd, Zr, Y, Pb	Ca, Na, Ni, V, Cr, Ti	Mn, Na, Ce, Sr, La, Sn, V, B, Lu, Dy	Fe, Mn, Zr, Mg, Ti, V, Na, Dy, Sm	Si, Ca, Cu, Ti	Fe, Mg, Lu, Yb, Er, B
0.005-0.01	Ba, Sr, Ti	Mg, V, Ti	Ga, Ni, Sr	Ga, Sc	Cu	Pb, Yb, Er	Lu, Er, Yb	Ni, Pb	Mn, Sc, Pb
0.001-0.005	Ga, V, Cr, Mn	Pb, Ga, Cr	Co, Y, Pb, Cr, Cu	Cr, Ba, Ni, Cu, Yb, Sn	Co, Ba, Pb	Sc, Ba, Ga, Cu, Ge, Cr, Mo	Pb, Ni, Cu, Cr, Ba, Sn, Sc	V, Y, Mn, Cr, Ba, Sr, Mg	V, Cu
0.0005-0.001	Cu	Mn							
0.0001-0.0005	Be	Cu	Be	Be		Be			Be
0.00005-0.0001									
0.00001-0.00005			Ag						

TABLE 8.—*Semiquantitative spectrographic analyses of minerals of Oliverian Plutonic Series rocks*—Continued

[Joseph Haffty, analyst]

Weight percent	Elements in mineral indicated							
	Lebanon aplite (NH/H 2-52)							
	Plagioclase	Quartz	Potassium feldspar	Epidote	Magnetite	Sphene	Allanite	Zircon
>10	Si, Al	Si	Si, Al, K	Si, Al	Fe	Ca, Ti	Si	Zr, Si
5-10	K			Ca		Si	Al, Fe, Ce	
1-5	Na		Na	Fe		Al, Fe, Y	La, Ca, Nd, Pr	
0.5-1		Al					Mg	Hf, Fe
0.1-0.5	Ca	Na, Ca	Ca	Na, Y, Mn	Al, Si	Nd, Ce, Mg, Yb, Mn	Th, Ti, Mn, Y	Th, Al, Y, Ca
0.05-0.1	Fe			Nd, Ce, Mg, Sr, Zr	Ca, Mn	Dy, Na, Nb, Sm, Pr, La, Zr	Gd, Sm	
0.01-0.05	Ba, B	Fe, Mg, B	Ba, Fe, B	La, Ti, Ga, Pb, V, Dy, B	Ti, Mg, V	Er, Sn, Lu, Sr, Tm, B, Sc, Ho	Sr, Pb, Zr, Eu, Er, Dy, Ga, B, V, Sc	Yb, Nd, Pb, Mg, Lu, Mn, Er, Ba, B
0.005-0.01	Mg, Ti, Sr	Ti, V	Mg, Sr, V	Cr, Sm, Ni, Ba	Ni, Cr, Y	Ga, V	Lu, Yb	La, Sm, Cu
0.001-0.005	Cr, Ga, Pb, Mn	Cr, Mn, Ba, Zr	Ti, Ga, Pb, Cr	Yb, Sc, Sn, Cu, Mo, Nb	Cu, Sr, Pb, Co, Ba	Cr, Pb, Ba, Cu, Ge, Mo	Sn, Cu, Ba, Cr	Sc, V, Cr, Sr, Be
0.0005-0.001			Mn			Be		
0.0001-0.0005	Be, Cu	Cu	Be, Cu	Be, Ag			Be	
0.00005-0.0001								
0.00001-0.00005							Ag	

Granodiorite (NH/M 50-52) of the Mascoma Group of Chapman and others (1938)

	Quartz and plagioclase	Potassium feldspar	Biotite	Epidote	Magnetite	Sphene	Apatite	Zircon
>10	Si	Si, K	Si	Si, Al	Fe	Ca, Ti, Si	Ca, P	Zr, Si
5–10	Al		Al, Fe, K	Ca				
1–5	Na, Ca	Al, Na	Mg, Ti	Fe		Al		
0.5–1								
0.1–0.5	K	Ba, Ca	Ca, Na	Mg	Al, Si	Fe, Th, Y, Nd, Ce, Mg, Zr	Al, Si	Hf, Y, Al, Ca
0.05–0.1	Mg, Sr		Ba	Sr	Mg, Ca		Y, Sr	Th
0.01–0.05	Fe, Ba, B	Sr, Fe, B	Mn, Zr, V, Co	Mn, Ti, V, Ce, Na, La, Ga	Ti, Mn, Ni, Cr, Cu, V	La, Gd, Yb, V, Mn, Sr, Nb, Dy, Er, Lu, Pr, Sm, Sn	Mg, Fe, Ti, Na, Mn, Er	Fe, Mg, Ti, Yb, Er, Lu, Pb
0.005–0.01	Ti, V, Mn	Mg	Ga, Sr, Ni	Zr, Cr, Pb, Sc, Y	Co	Cu	Zr, Dy	Mn
0.001–0.005	Ga, Cr, Pb, Zr	Pb, Ga, V, Cr, Ti	Cr, Sc, Y, Pb	Yb, Ba	Zr, Sr, Ba	Pb, Ho, Cr, Ga, Ba, Sc, Mo	La, Yb, Ba, V, Pb, Cr	Sc, V, Sr, Cr
0.0005–0.001		Mn	Cu					
0.0001–0.0005	Cu			Cu			Cu	Cu

TABLE 8.—*Semiquantitative spectrographic analyses of minerals of Oliverian Plutonic Series rocks*—Continued

Weight percent	Elements in mineral indicated								
	Quartz monzonite of Mount Washington quadrangle (NH/MW 4-52)								
	Plagioclase	Quartz	Potassium feldspar	Biotite	Chlorite	Magnetite	Sphene	Apatite	Zircon
>10	Si	Si	Si, K	Si	Al	Fe	Ca, Ti, Si	Ca, P	Zr, Si
5–10	Al		Al	Al	Si				
1–5	Na	Al	Na	K, Fe, Mg, Ti	Mg, Fe, K		Al		
0.5–1	K			Ca	Ti		Fe		
0.1–0.5	Ca	Na, Ca	Ba, Ca	Ce, La	Ca, Ce	Al, Si	Nd, Th, Ce, Mg, Y, Zr, La	Al, Si, Ce, Zr, Y, Th	Hf, Al, Ca, Y
0.05–0.1	Sr			Li	La	Ca, Mg	Mn	Mg, Nd, Sr	
0.01–0.05	Fe, Mg, Ba, B	Fe, Mg, B	Sr, Fe, B	Mn, Zr, Nd, Ba, Sr, Pr, Co, V	Mn, Li, Ba, Ga, Zr, Nd, V	Cu, Mn, Cr, Ni, Ti, Zr, V	Gd, Nb, Er, V, Sr, Sn, Dy, Yb, B, Pr, Sm	Fe, Ti, Mn, La, Er, B, Sm, V, Dy	Ti, Yb, Fe, Mg, Er, B, Ba, Lu
0.005–0.01	Ga, Ti, Mn	Sr	Mg, Ca	Ga, Ni, Y	Sr, Co, Ni	Co	Lu, Pb		V, Pb, Mn, Sc
0.001–0.005	V, Cr, Zr, Pb	V, Ti, Cr, Ba, Zr, Mn, Ga	V, Pb, Ti, Cr, Mn	Pb, Sc, Cr, Cu	Pb, Y, Sc, Cu, Cr	Ba, Sr	Cr, Ho, Ba, Sc, Ga, Mo	Yb, Bi, Pb, Ba, Sc, Cr	Cr
0.0005–0.001									
0.0001–0.0005	Cu, Be	Cu, Be	Cu	Ag, Be	Ag, Be		Be	Cu	Cu, Be
0.00005–0.0001									
0.00001–0.00005									

yttrium, lanthanum, uranium, scandium, ytterbium, titanium, cerium, manganese, and lead do show a correlation. As with uranium, however, there is no suggestion of a positive association between thorium and any of the major rock-forming elements.

NEW HAMPSHIRE PLUTONIC SERIES

Plutons of this series consist of a group of stocks and batholiths, many of them strongly foliated, which intrude formations as young as the Lower Devonian Littleton Formation. The series is of Acadian age and consists (Billings and Keevil, 1946, p. 812) of the following rock types: diorite 1 percent, amphibolite 1 percent, Remick Tonalite (quartz diorite) 2 percent, Winnipesaukee Quartz Diorite to granodiorite 23 percent, Bethlehem Gneiss (granodiorite to quartz monzonite) 14 percent, Kinsman Quartz Monzonite 26 percent, and Concord (and other) Granite 33 percent.

Thorium and uranium analyses of the common petrographic types of the New Hampshire Plutonic Series are listed in table 9, and summarized in table 10.

The ranges of thorium and uranium in New Hampshire Series rocks are less than for the Oliverian Series, probably because of a more limited range in petrographic types. Although the absolute amounts of thorium and uranium in the series are within the ranges expected for calc-alkaline rocks (Whitfield and others, 1959; Larsen 3d and Gottfried, 1960), the thorium to uranium ratio is somewhat higher than normal (see also fig. 1). Uranium and thorium analyses listed by Whitfield and others (1959, p. 252) for two New Hampshire Series rocks are consistent with those of table 10.

Within individual plutons of the New Hampshire Plutonic Series there is remarkably small variation in uranium and thorium contents. For example, the Mount Clough pluton of the Bethlehem Gneiss is an almost concordant sheet, 78 miles long, and ½ to 3 miles thick (Billings, 1956, p. 125). Twenty-nine samples from this body showed a range of only 2.5 to 5.2 ppm uranium and 11.7 to 18.7 ppm thorium.

Generalizations true for the Oliverian Series are also valid for the New Hampshire Series. Wallrocks and inclusions have mean thorium and uranium values close to those of quartz diorite. There are also striking differences between the aplites, whose thorium to uranium ratios agree with those of other members of the rock series, and the pegmatites, which have abnormally low thorium. The most uraniferous (39 ppm uranium) of the 135 analyzed samples was a pegmatite of the New Hampshire Series. This sample was the only one in which uraninite (25 mg) was identified. Uranium and thorium

TABLE 9.—*Uranium and thorium contents of some New Hampshire Plutonic Series rocks*

Description	Sample	Quadrangle	Uranium (ppm)	Thorium (ppm)	Thorium: uranium ratio
Bethlehem Gneiss	NH/BF 1-52	Bellows Falls	3.7 3.7		
	NH/BF 3-52	do	3.7 4.2		
	NH/BF 4-52	do	4.9 5.3		
	NH/LM 4-52	Lovewell Mountain	3.1 3.3 3.6		
	NH/S 4-52	Sunapee	4.3 4.3	18.7 18.4	4.3
	NH/S 6-52	do	2.9 2.7	13.8	4.9
	NH/S 7-52	do	3.4 3.7	14.0	4.0
	NH/S 8-52	do	4.5 4.6		
	NH/M 11-52	Mascoma	3.7 4.1 4.5	15.4	3.8
	NH/M 21-52	do	2.9 3.2 3.3 3.2	16.8	5.4
	NH/M 22-52	do	3.1 3.2		
	NH/M 25-52	do	4.9 5.3 5.4		
	NH/C 1-52	Mount Cardigan	2.2 2.2	18.2 18.0	8.3
	NH/C 8-52	do	3.8 4.0		
	NH/MC 7-52	Mount Cube	3.1 3.2		
	NH/MC 8-52	do	4.0 4.2		
	NH/MC 13-52	do	3.6 3.6		
	NH/MC 16-52	do	3.1 3.2		
	NH/MC 17-52	do	2.9 2.9		
	NH/MC 21-52	do	3.7 3.8		
	NH/MC 22-52	do	2.5 2.5	12.1	4.8
	NH/MC 23-52	do	2.9 3.2	11.7	3.8
	NH/R 6-52	Rumney	2.3 2.2	13.2	5.9
	NH/R 8-52	do	2.9 2.9		
	NH/R 9-52	do	3.0 3.2		
	NH/R 10-52	do	2.5 2.7		
	NH/Mo 2-52	Moosilauke	3.5 3.6 3.7		
	NH/Mo 3-52	do	3.8 2.9 3.0	15.6	5.3
Aplite in Bethlehem Gneiss	NH/R 5-52	Rumney	4.0 4.3 3.2 3.3	52	14
	NH/S 4-52	Sunapee	2.3 2.4 5.2 3.3	18.7 18.4	4.1
Micaceous schlieren in Bethlehem Gneiss.	NH/R 7-52	Rumney	3.5 3.7		
Mica schist at contact with Bethlehem Gneiss.	NH/S 9-52	Sunapee	4.4 4.9 5.0 5.0	16.0	3.3
Sillimanite schist at contact with Bethlehem Gneiss.	NH/BF 2-52	Bellows Falls	4.6 4.6		
Trap dike cutting Bethlehem Gneiss.	NH/R 11-52	Rumney	2.0 2.1		

TABLE 9.—*Uranium and thorium contents of some New Hampshire Plutonic Series rocks*—Continued

Description	Sample	Quadrangle	Uranium (ppm)	Thorium (ppm)	Thorium: uranium ratio
Kinsman Quartz Monzonite	NH/K 7-52	Keene	2.9 3.1 3.5		
	NH/MD 6-52	Monadnock	2.1 2.2		
	NH/LM 1-52	Lovewell Mountain	1.6 1.8 1.4 1.4	19.2	12.4
	NH/LM 2-52	do	2.9 3.6 4.0	14.5	4.1
	NH/S 11-52	Sunapee	1.5 1.6	18.1	11.7
	NH/S 12-52	do	3.1 3.3		
	NH/S 16-52	do	1.9 2.0		
	NH/C 2-52	Cardigan	3.3 3.6		
	NH/C 3-52	do	1.9 2.0	19.4	10.0
	NH/C 6-52	do	2.5 2.7	14.1	5.4
	NH/R 12-52	Rumney	3.0 3.0		
	NH/F 1-52	Franconia	5.5 6.2 6.5	9.1	1.5
	NH/F 2-52	do	3.6 4.2 5.0 5.7		
	NH/Mo 4-52	Moosilauke	3.3 3.4 3.5 4.2 4.3	11.2 10.7	3.0
	NH/Mo 5-52	do	6.2 6.5		
Aplite in Kinsman Quartz Monzonite.	NH/S 15-52	Sunapee	12.5 12.4 14.2 14.3	4.3	.32
Micaceous inclusion in Kinsman Quartz Monzonite.	NH/S 13-52	do	.8 .9		
	NH/C 7-52	Cardigan	2.5 2.7		
Fitzwilliam Granite	NH/Md 1-52	Monadnock	4.7 4.9	18.3 19.6	3.9
	NH/Md 3-52	do	4.5 4.4		
Pegmatite of Fitzwilliam Granite.	NH/Md 2-52	do	34.2 35.6 38.8 48.0	5.0	.13
Concord Granite	NH/C 4-52	Cardigan	2.9 3.5 3.9	22	6.5
	NH/S 10-52	Sunapee	2.6 3.0		
	NH/S 14-52	do	5.4 6.0 6.1	11.6 11.5	2.0
Pegmatite in Concord Granite (edge).	NH/C 5a-52	Cardigan	3.2 4.6 5.5 6.5	3.9	.8
Pegmatite in Concord Granite (center).	NH/C 5b-52	do	6.0 6.6	3.0 2.6	.44
Bickford Granite	NH/MW 1-52	Mount Washington	3.6 4.1 5.5	10.7	2.4
French Pond Granite	NH/Mo 1-52	Moosilauke	3.9 4.4 4.7	27 30	6.6
Sugar Hill Quartz Monzonite	NH/Mo 6-52	do	2.5 2.5		
Spaulding Quartz Diorite	NH/Md 4-52	Monadnock	2.8 3.0	14.6	5.0
	NH/Md 5-52	do	3.3 3.5 3.6		

TABLE 10.—*Mean uranium and thorium contents of the major rock types of the New Hampshire Plutonic Series*

[Ratios are the averages of the individual thorium:uranium ratios, not the ratios of the average thorium and average uranium contents of all specimens]

Rock types	Uranium			Thorium			Mean thorium: uranium ratio
	Samples analyzed	Parts per million		Samples analyzed	Parts per million		
		Mean	Range		Mean	Range	
Granite	7	4.3	2.8–5.8	5	18.2	10.7–28.5	4.3
Quartz monzonite (chiefly Kinsman Quartz Monzonite)	15	3.3	1.5–6.4	7	15.1	9.1–19.4	6.9
Quartz monzonite to granodiorite (Bethlehem Gneiss)	28	3.4	2.2–5.2	10	14.9	11.7–18.5	5.0
Quartz diorite	2	3.2	2.9–3.4	1	14.6	----------	5.0
Pegmatite	3	16.8	4.7–39	3	3.9	2.8– 5.0	.46
Aplite	3	7.2	3.3–14.2	3	25.0	4.3–52	6.1
Wallrocks and inclusions	5	3.3	0.8– 5.0	1	16.0	----------	3.3

analyses of the minerals of the New Hampshire Series rocks and the contributions made by each mineral to the total uranium and thorium contents of the rocks are shown in table 11.

Mass balances for uranium and thorium, as computed from amounts of these elements in the rock minerals, generally show unsatisfactory agreement with assays of the bulk rocks. These disagreements, in part, reflect inaccuracies in determining absolute amounts of the mineral phases, but also suggest variabilities in the amounts of uranium and thorium carried by the major rock-forming silicates. Strong fluctuations in the thorium to uranium ratios in these minerals also point toward this conclusion and indicate a partial lack of crystal-chemical control in the distribution of the two elements. The fact that the somewhat sketchy mineral data indicates that between 53 and 77 percent of the uranium and a large (but indeterminate) amount of thorium is carried by the major rock silicates likewise argues for this conclusion.

Table 12 lists semiquantitative spectrographic analyses for minerals of the New Hampshire Series rocks. Rank-correlation coefficient calculations show that uranium has a strong positive correlation (at the 95-percent confidence level) with thorium, gadolinium, samarium, zirconium, yttrium, cerium, neodymium, lanthanum, and praseodymium; the elements phosphorus, ytterbium, erbium, scandium, lutetium, dysprosium, europium, niobium, thulium, holmium, nickel, and beryllium show a somewhat less regular relation. Sodium shows negative correlation (at the 95-percent confidence level) in three cases out of four, and silicon and aluminium in one case out of four.

The small number of thorium analyses makes statistical inference difficult. However, many of the elements which show an association

TABLE 11.—*Uranium and thorium contents of minerals of the New Hampshire Plutonic Series rocks*

[Analyst, Marian Schnepfe. Mwp, mineral weight percent of sample]

Mineral	Bethlehem Gneiss NH/S 4-52						Kinsman Quartz Monzonite		
		Uranium		Thorium		Th:U		Uranium	
	Mineral weight percent of sample	Parts per million	$\frac{\text{Mwp}\times\text{U (ppm)}}{100}$	Parts per million	$\frac{\text{Mwp}\times\text{Th (ppm)}}{100}$		Mineral weight percent of sample	Parts per million	$\frac{\text{Mwp}\times\text{U (ppm)}}{100}$
Quartz	36.8	0.94	0.348	2.9	1.06	3.1	33.2	0.10	0.033
Potassium feldspar	9.1	.43	.039	3.0	.27	7.0	1.5	.10	.002
Plagioclase	39.8	6.6	2.63	2.9	1.16	.43	35.5	.30	.107
Biotite	12.2	2.0	.24	11.0	1.34	5.5	13.2	.53	.070
Muscovite	1.3	3.1	.04				15.7	[1] 3.0	.047
Garnet							.2	.88	.002
Magnetite	.3	20.7	.62				1.2	2.0	.024
Hematite									
Ilmenite	.1	19.3	.019				.5	2.0	.010
Pyrite	.02	16.9	.034						
Apatite	.2	16.4	.033				.3	13.8	.041
Monazite	.01	2,570	.257				.007	1,355	.095
Xenotime	.002	798	.016						
Allanite	Tr.								
Zircon	.01	500	.05				.01	620	.062
Fluorite									
Rutile							Tr.		
Graphite							.1		
Molybdenite	Tr.						Tr.		
Total uranium (by sum of minerals)			4.27						0.49
Total uranium (chemical)			3.3						1.7
Total thorium (by sum of available mineral analyses)					3.83				
Total thorium in bulk rock (measured)					18.5				
Thorium: uranium in bulk rock (measured)						4.3			

[1] Estimated.

TABLE 11.—*Uranium and thorium contents of minerals of the New Hampshire Plutonic Series rocks*—Continued

Mineral	Concord Granite NH/C 4-52						Concord pegmatite NH/C 5-52					
	Mineral weight percent of sample	Uranium		Thorium		Th:U	Mineral weight percent of sample	Uranium		Thorium		Th:U
		Parts per million	Mwp×U (ppm)/100	Parts per million	Mwp×Th (ppm)/100			Parts per million	Mwp×U (ppm)/100	Parts per million	Mwp×Th (ppm)/100	
Quartz	30.2	0.36	0.109	5.7	1.72	15.8	27	2.5 2.0	0.61	0.9	0.24	0.4
Potassium feldspar	26.7	.36	.096	4.0	1.07	11.1	23	1.8 1.8	.41	1.6	.37	.9
Plagioclase	31.8	.36	.115	5.7	1.81	15.8	47	1.8 2.4	.99	3.9	1.83	1.9
Biotite	6.7	2.6	.174	80	5.3	30.7						
Muscovite	3.3	1.4	.046	17	.56	12.1	3	2.4 1.9	.06			
Garnet							.9	5.2 5.5	.05	1.8	.02	.34
Magnetite	.20	39.5	.079				.03	30 31	.01			
Hematite							Tr.					
Ilmenite	.1	32.7	.033	230	.23		.003					
Pyrite	.01	373	.037				Tr.					
Apatite	.3	1.3	.004	290	.87		.2	22 25	.04			
Monazite	.01	624	.062				Tr.					
Xenotime	Tr.											
Allanite	.002	329	.007									
Zircon	Tr.						.003	18,000 20,000	.57			
Fluorite	Tr.											
Rutile	Tr.						Tr.					
Graphite												
Molybdenite												
Total uranium (by sum of minerals)			0.76						2.74			
Total uranium (chemical)			3.4						5.4			
Total thorium (by sum of available mineral analyses)					11.6						2.5	
Total thorium in bulk rock (measured)					22						3.9	
Thorium: uranium in bulk rock (measured)						6.5						0.8

TABLE 12.—*Semiquantitative spectrographic analyses of minerals of New Hampshire Plutonic Series rocks*

[Joseph Haffty, analyst]

Weight percent	Element(s) in mineral indicated											
Bethlehem Gneiss (NH/S 4-52)												
	Plagioclase	Microcline	Quartz	Muscovite	Biotite	Apatite	Magnetite	Ilmenite	Pyrite	Xenotime	Allanite	Monazite	Zircon
>10	Si, Al	Si, Al, K	Si	Si, Al	Si, Fe	Ca, P	Fe	Fe, Ti	Fe	Y, P	Si	Ce, P	Zr, Si
5-10				K	Al, K, Mg						Al, Ce	Nd	
1-5	Ca, Na	Na	Al	Na, Fe	Ti		Si			Er, Th	Nd, Fe, La, Th, P, Ca	Th, La, Ca	
0.5-1				Ti, Mg				Si	Si	Yb, Dy, U, Ca, Si	Y	Pr, Y, Sm, Si	Hf
0.1-0.5	K	Ca, Ba	Ca, Na	Ca	Na, Ca	Si, Fe, Al Y	Mn, Al, Ni, Ca	Al, Mn, Mg, Ca	Al, As, Ca, Co, Na	Ce, Nd, Gd, Al, Tm, Lu, La, Sm	Mn, Pr, Dy, Mg, Sm, Gd	Al, Gd, Fe	Y, Al
0.05-0.1				Ga	Mn				Ni	Pr, Ho, Zr, Fe	Ti, Er, Yb, Zr	Dy, Mg	
0.01-0.05	Sr, Fe, Mg	Sr, Pb, Fe	B, Fe	Ba, B, Zr V, Mn	Li, Ba, Co, Ni, Ga	Mn, Na, Mg, Sr, Zr	Co, Cu, Ti, Mg, Cr, La, Y, Zr	Ni, Co, Cu, Zr	Pb, Cu, Ti, Mg, Mn	Pb, Mg	Pb, Lu, Ho	Pb, Er, Mn, Zr, Yb, Ho, Lu, Ti	Fe, Yb, Sc, Dy, B, Lu, Er, Mg, Ti
0.005-0.01	Ga	Mg	Mg	Cr	Cr	La, Er, Dy	Sn, V	Mo, Ba, Nb	Y, Ba	Mn	Ba, Tm, Sc	Tm, Sc	
0.001-0.005	Mn, Zr, Pb, Ba	Ga	Sr, Ba, Zr	Sn, Sc, Sr	Zr, Sc, Sn, Sr	Yb, Ho	Yb, Sr	Sc, Cr, Sr	Zr, Yb, Sr	Sc, Cu, Ba, Sr	Cu, Cr	Cu, Ba	Ba, Mn
0.0005-0.001	Cu		Cu		Cu	Cr, Cu							
0.0001-0.0005	Be										Be		
0.00005-0.0001			Be									Be	

TABLE 12.—*Semiquantitative spectrographic analyses of minerals of New Hampshire Plutonic Series rocks*—Continued

Weight percent	Element(s) in mineral indicated									
Kinsman Quartz Monzonite (NH/LM 1-52)										
	Quartz	Plagioclase	Microcline	Biotite	Garnet	Magnetite	Ilmenite	Apatite	Monazite	Graphite
>10	Si	Si, Al	Si, K	K, Fe, Si	Si, Fe	Fe	Fe, Ti	Ca, P	Ce, P	
5–10			Al	Al, Mg	Al				La, Nd	
1–5	Al	Na, Ca	Na	Ti	Mg	Si			Ca, Th, Pr	
0.5–1	Na								Sm	Fe
0.1–0.5	K, Ca	K	Ba, Ca	Na, Ca	Mn	Ti, Mn, Ni, Al, Mg, Cu	Si, Mg, Al	Fe, Y, Mg, Si, Al	Si, Y, Fe, Al, Gd, Mg	Si, Al, Ca, Mg
0.05–0.1				Ba, Li	Ca		Mn	Ce	Pb	
0.01–0.05	Ba, Fe, Mg, V, B	Sr, Fe, Mg, Ti, V, B	Fe, Sr, Mg, B, Pb	Ni, Cr, Zn, Mn, V, B	Ni, Y, Ti, B	Ca, Co, Zr, Sn, Cr, B, V	Ca, Zr, Cu, B	Na, Nd, Sr, Zr, La, Mn, Sm, Ti, Gd	Zr, Er, Ti, Eu, V, B	Na, Ti, Zn, Cu, V, B
0.005–0.01	Ti	Ga		Co, Ga	Cr		Nb, V			
0.001–0.005	Sr, Pb, Cr	Ba, Pb, Cr, Zr	Cr, Ti, V, Ga	Nb, Zr, Sc, Cu	V, Sc, Ga, Ba, Zr, Co Ni	Pb, Ba	Cr, Co, Ni Ba	V, Cr, Yb, Ba	Yb, Mn, Cu, Ba	Mn, Co, Ba, Ni, Be
0.0005–0.001		Mn			Cu, Yb					Cr
0.0001–0.0005	Cu, Mn	Be, Cu	Cu					Cu	Be	Ag

Concord Granite (NH/C 4-52)

	Quartz and plagioclase	Microcline	Biotite	Muscovite	Magnetite	Ilmenite	Apatite	Allanite	Pyrite	Monazite
>10	Al, Si	K, Si	Si, Fe	Al, Si	Fe	Fe, Ti	Ca, P	Si	Fe	Ce, P
5–10	Na	Al	Al, K	K	Ti			Al, Fe, Ca, Ce		La, Nd
1–5	Ca	Na	Mg, Ti	Fe				Ti, Nd, La		Th, Ca, Si, Pr
0.5–1	K			Na, Ti, Mg, Ca	Si	Si		Mg, Th, Pr, P	Al	Ti, Al
0.1–0.5		Ca, Ba	Ca, Na	Ba	Al, Ce, Mn	Al, Ce, Ca, Zr	Ce, Al, Si, Y	Mn	Si, Ce	Gd, Sm, Fe, Y
0.05–0.1	Sr, Fe, Ba	Pb, Sr	Ba, Li	Li	Ca, Mg, Zr, Ni, Gd	Gd, Mg, Mn, La	Sr, Fe, Na, Mn	Sm, Zr, Y, Pb, Ba, Gd, Sr	Ca, Ti, Pb, Zr, Ba, Co, Cu, Mg	Ba, Mg, Pb, Zr
0.01–0.05	Cr, Mg	Fe	Mn, Ce, V, Zn	V, Ce, Ga, La, Mn, Sr	La, Na, Cr, Cu, Pb, Dy, Er	Na, Cr, Co, Eu, Dy, Er, Pr	V, La, Mg, Zr, Ti	Eu, Sc, Nb	Ni, La, Gd, Na	Eu, Tb, Nb, Sc, Sr
0.005–0.01	V, Ni, B, Mn, Ga	B, Cr, Mg, V, Ga	B, Cr, Ga, La, Zr, Ni, Sr, Dy, Er	Pb, Sn, Zr, B	B, Nb, Co, Eu, Sn	B, Nb, Pb, Ni, Co, Sn	B, Be, Gd, Yb, Ni, Eu	B, Cr, Ni, Ga, V, Dy, Er, Cu	B, Mn, Zn, V	B, Cr, Mn, Lu, Dy, Er, V, Ni
0.001–0.005	Ti, Zr, Pb, Nb	Ti, Mn, Zr	Gd, Co, Sc, Nb, Pb, Mo Cu	Cr, Cu	V, Y, Ba, Sc, Sr, Mo	V, Y, Sc, Ba, Cu, Sr, Mo	Dy, Er, Sc, Pb, Cr	Yb	Cr, Sr, Mo, Y	Yb
0.0005–0.001										
0.0001–0.0005	Be, Cu		Be	Be	Yb, Be	Yb, Be	Cu	Be	Yb, Be	Cu, Be
0.00005–0.0001		Cu								

TABLE 12—*Semiquantitative spectrographic analyses of minerals of New Hampshire Plutonic Series rocks*—Continued

Weight percent	Element(s) in mineral indicated							
	Concord pegmatite (NH/C 5-52)							
	Albite	Quartz	Perthite	Muscovite	Garnet	Magnetite	Apatite	Zircon
>10	Si, Al	Si	Si	Al, Si	Si, Mn, Al	Fe	Ca, P	Zr, Si
5–10	Na		Al, K	K	Fe			
1–5			Na	Fe				Hf
0.5–1	K			Na		Mn, Si		U, Fe
0.1–0.5	Ca	Al		Mg	Ca, Mg	Al, Ni	Si, Fe, Y, Al	Y, Al, Ca
0.05–0.1				Ti	Ce	Na	Mn, Ce, Na, Sr	Ce
0.01–0.05	Fe, Ba, Sr	Na, Fe	Fe, Ba	Ga, Mn	La, Nd, Y, Ti	Cr, Co, Cu, Ti	Ba, Ti, La	Pb, Sr, Ba, Ti, Dy, Mn, Mg, Nd, La, Er
0.005–0.01	Mg, Mn		Sr, Pb	Cr		V	Zr	Yb, Lu
0.001–0.005	Ga, Pb, Zr, Be	Mg, Mn	Ga, Mg	V, Ba, Sn, Be, Sr	Yb	Mg	Mg, Cu, Pb	Be, Cu, Sc
0.0005–0.001	Cr, Cu	Cr	Mn, Cr				Yb, Cr	
0.0001–0.0005			Be	Cu				

with uranium (such as cerium, zirconium, scandium, gadolinium, nickel) also show an association with thorium. In the one series of statistical tests possible, sodium and silicon show a negative correlation with thorium.

PETROLOGY AND DISTRIBUTION OF URANIUM AND THORIUM

Data for all paired uranium and thorium analyses are plotted in figure 1. The following relationships become evident: (a) wide dispersion in the range for both elements within any given petrographic type and rock series, (b) a very limited high uranium to low thorium

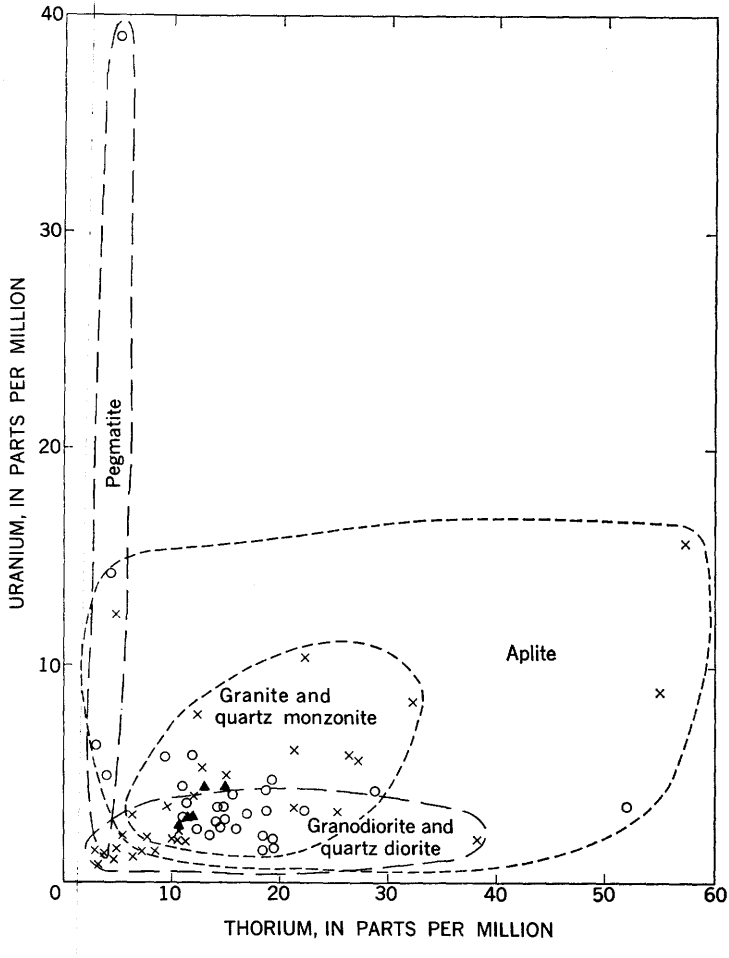

FIGURE 1.—Thorium and uranium analyses of rocks of the Highlandcroft (▲), Oliverian (×), and New Hampshire (○) Plutonic Series.

ratio in the pegmatites but wide variation of this ratio among the aplites, and (c) generally higher thorium to uranium ratios for the New Hampshire Series, as contrasted with the Oliverian Series. For the latter rocks (27 analyses), the plot does not appear to confirm the suggestions by Rogers and Ragland (1961; 7 analyses) that the thorium to uranium ratio increases with igneous evolution (that is, toward the granitic end of the series). This ratio is possibly characteristic of the different petrographic provinces, as suggested by Turovsky (1957) and Larsen 3d and Gottfried (1960).

Because of the limited number and range of rock analyses (table 1), it is impossible to draw rigorous comparisons concerning uranium and thorium distributions and amounts of most of the major oxides for the different related series. For K_2O, thorium, and uranium, however, the data are sufficiently plentiful (Rogers and Ragland, 1961; Whitfield and others, 1959; table 1) to permit statistical treatment, particularly if the rocks are treated as a whole and considered to be representative calc-alkaline types. The Th:K_2O analyses (fig. 2) display a close linear relationship which may be fitted to a least squares curve at a confidence level of 99.9 percent (product moment-correlation coefficient $r=0.806$). On the other hand, the

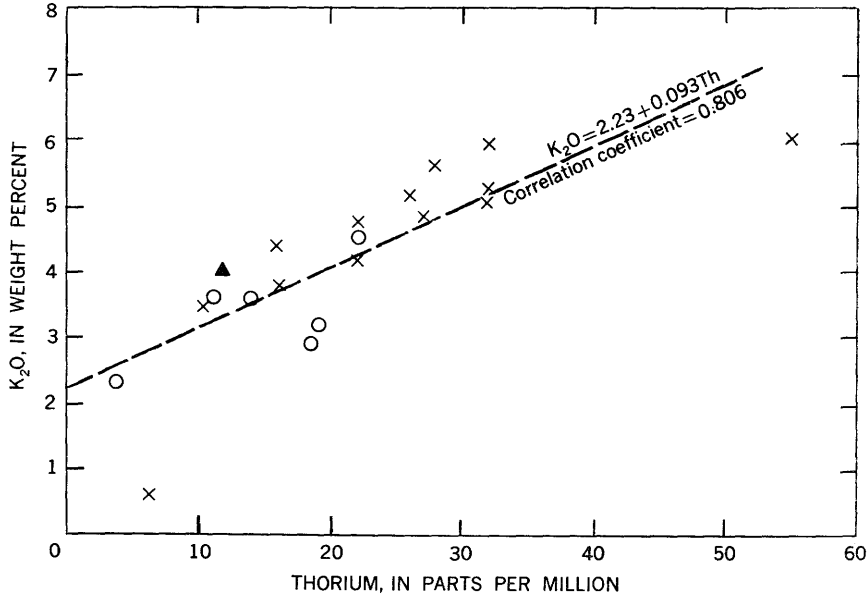

FIGURE 2.—K_2O plotted against thorium. Data from tables 1, 5, and 9, from Whitfield and others (1959), and Rogers and Ragland (1961). Highlandcroft (▲), New Hampshire (○), and Oliverian (×) Plutonic Series.

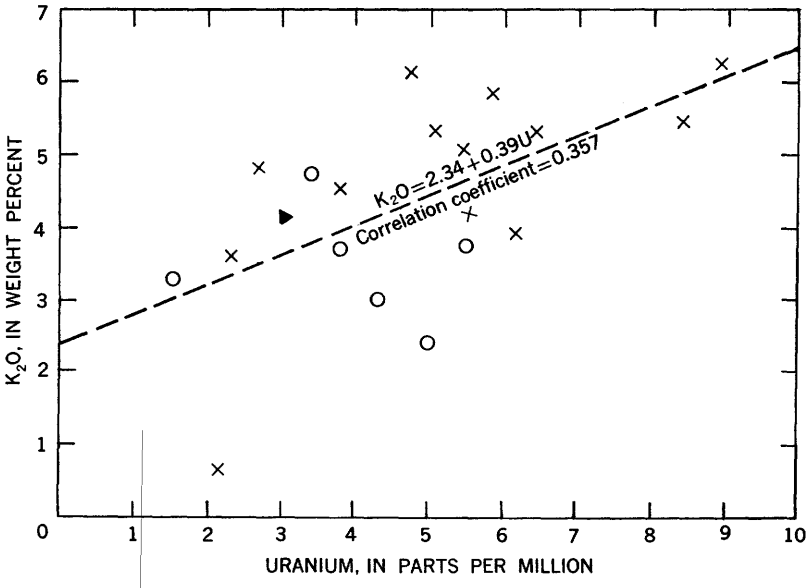

FIGURE 3.—K_2O plotted against uranium. Data from tables 1, 5, and 9, from Whitfield and others (1959), and Rogers and Ragland (1961). Highlandcroft (▲), New Hampshire (○), and Oliverian (×) Plutonic Series.

least squares curve for the K_2O : U data (fig. 3) is not significant (r=0.35) even at the 90-percent level. Stated another way, these data imply that a determination of K_2O in any of these rocks provides a reasonably accurate estimate of thorium (and vice versa), but the variation in uranium is much more random. If K_2O content is regarded as a measure of the degree of petrogenetic evolution, the inference is clear that uranium is not as closely related to the magmatic residuum as is thorium, and that it may therefore be more volatile. Suggestions have been made by several writers (for example, Phair, 1952; Larsen and Phair, 1954; McKelvey and others, 1955; Adams and others, 1959) that the difference in behavior of thorium and uranium in igneous rocks is due to the formation of soluble $(UO_2)^{+2}$ which may escape from the magma. As pointed out by Page (1960), it seems geologically probable that uranium may be expelled at almost any stage of magmatic evolution.

Other plots, such as thoruim versus Larsen abscissa ($\frac{1}{3}SiO_2+K_2O$-FeO-MgO-CaO) (fig. 4), uranium versus Larsen abscissa (fig. 5), thorium versus CaO (fig. 6) and uranium versus CaO (fig. 7) have reasonably consistent nonlinear trends, but the comparisons suffer because of a lack of sufficient samples and the necessity of combining data from the different petrographic provinces. In a general way we

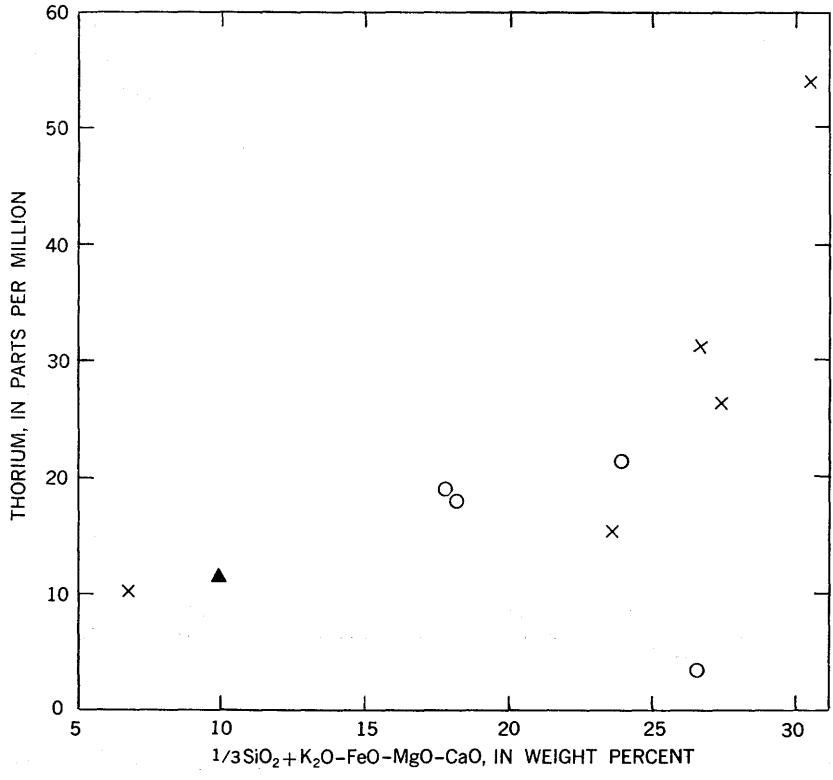

FIGURE 4.—Thorium plotted against the Larsen abscissa (⅓ SiO_2+K_2O-FeO-MgO-CaO) for rocks of the Highlandcroft (▲), New Hampshire (○), and Oliverian (×) Plutonic Series. Low thorium sample is a pegmatite.

may conclude that the most interesting rocks from the standpoint of concentration of uranium and thorium are those in which the Larsen abscissa exceeds 25 percent (or SiO_2 exceeds 73 percent), K_2O exceeds 5 percent, and CaO is 0.6 percent or less. Such rocks are rare and are most closely approximated by aplite.

In the light of presently available information on uranium and thorium abundances in igneous rocks, it is unlikely that any sweeping generalizations will be found to be universally valid; each rock series presents its own pattern. The rule that uranium and thorium increase as rocks evolve toward more felsic differentiates is apparently reversed in the Honolulu Volcanic Series (Larsen and Gottfried, 1960, p. 158). Whereas K_2O and uranium show no consistent pattern in most rock series (Whitfield and others, 1959; Coulomb and others, 1958; this paper, p. F33), they do in the Lassen volcanic rocks (Adams, 1955; Vistelius, 1958). Increase in the thorium to uranium ratio

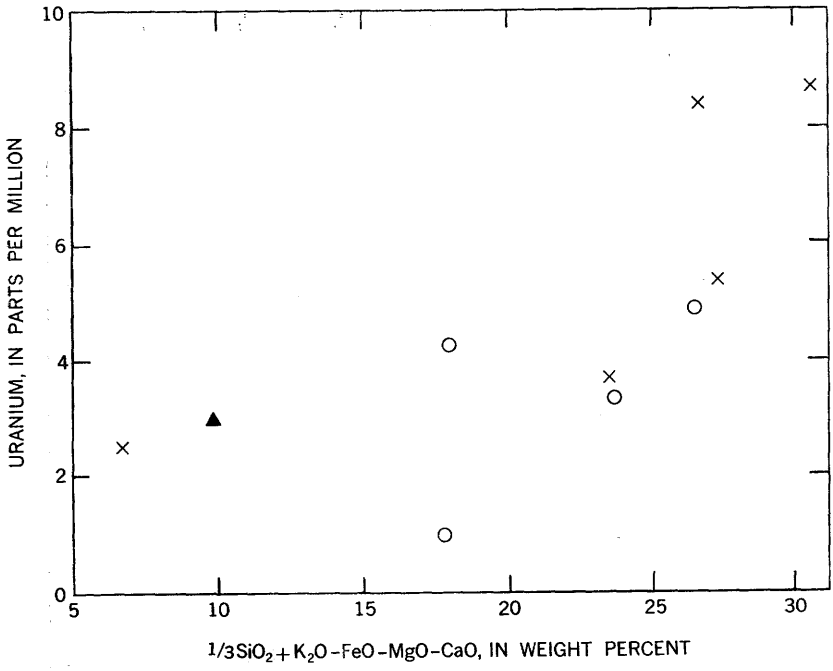

FIGURE 5.—Uranium plotted against the Larsen abscissa (⅓ SiO_2+K_2O-FeO-MgO-CaO) for rocks of the Highlandcroft (▲), New Hampshire (○) and Oliverian (×) Plutonic Series.

with petrogenetic evolution has been reported by Whitfield and others (1959), but was not substantiated by Larsen 3d and Gottfried (1960), nor by data reported here.

MINERALOGY AND DISTRIBUTION OF MINOR ELEMENTS, URANIUM, AND THORIUM

Semiquantitative spectrochemical mineral analyses (tables 4, 8, and 12), and uranium and thorium mineral analyses (tables 3, 7, and 11) have been made to determine whether abundances of one or another of the minor elements might imply similar abundances in uranium or thorium and also to determine to what extent the commonly accepted laws of geochemical distribution have been followed in the rock-forming minerals. Anomalies in the distribution of uranium or thorium might indicate the possibility that one or both of these elements was added to or subtracted from the rocks (see Whitfield and others, 1959, p. 265) subsequent to their crystallization. Moreover, in most plutonic series a very large proportion (40 to 90 percent; see Brown and Silver, 1956, and Larsen and Gottfried, 1961) of the ura-

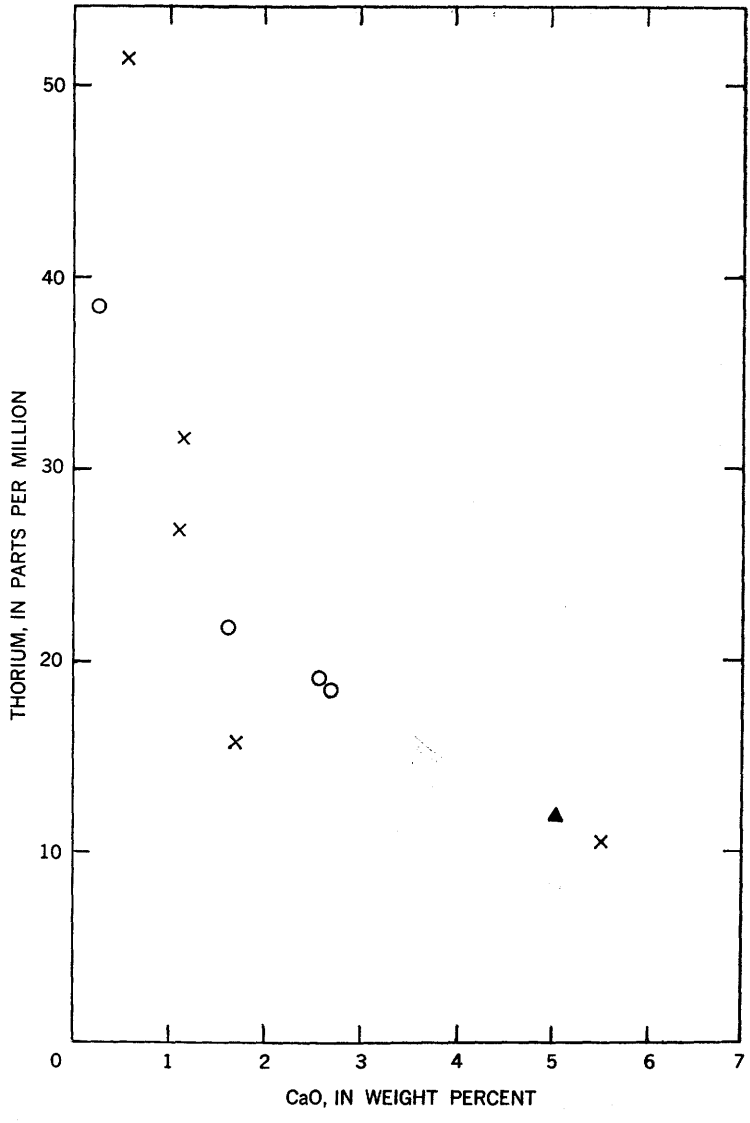

FIGURE 6.—Thorium plotted against CaO for rocks of the Highlandcroft (▲), Oliverian (×), and New Hampshire (○) Plutonic Series.

nium and thorium is never accommodated at structural sites within crystals, but occurs at such places as grain boundaries and defect sites where it is easily leached. This situation presumably occurs within the New Hampshire Series but not within the Oliverian and Highlandcroft, and opens the possibility of recognizing differences in the dispersal patterns of the minor elements.

The spectrographic and chemical data have been analyzed by the

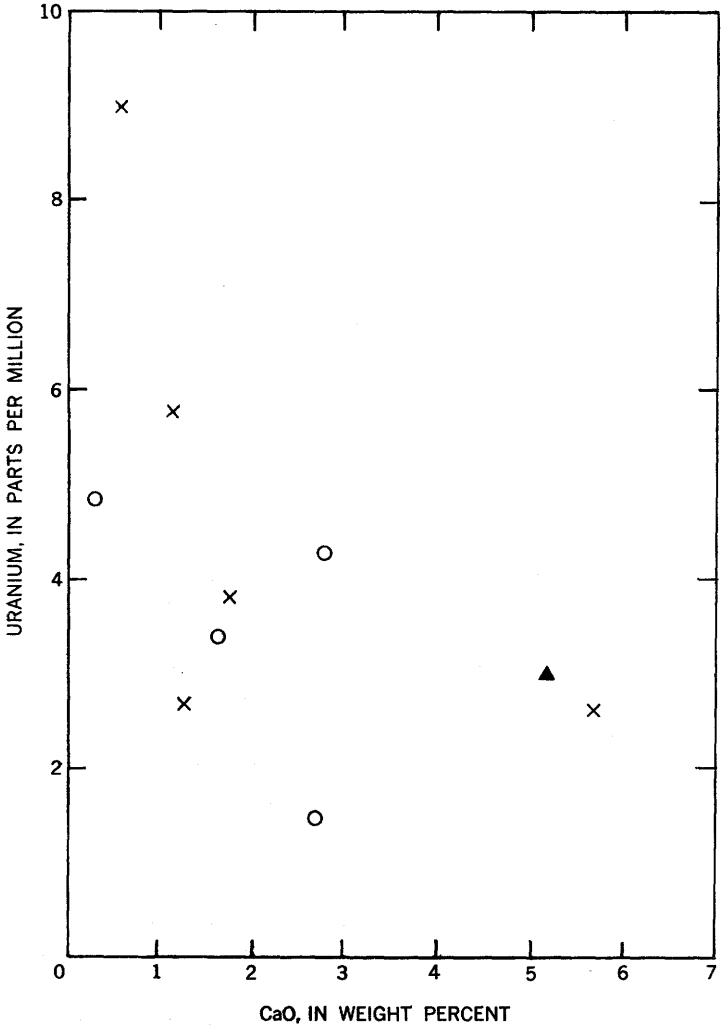

FIGURE 7.—Uranium plotted against CaO for rocks of the Highlandcroft (▲), Oliverian (×), and New Hampshire (○) Plutonic Series.

rank correlation coefficient method using procedures suggested by Flanagan (1957). Variations in the limits of sensitivity for detecting the various elements, tied ranks (that is, more than one element at the same level of abundance), possible sample inhomogeneity or impurity, and other factors to some extent undermine the reliability of the statistical analysis. Flanagan (1957), in one series of tests, found 80 percent agreement between statistical calculations based upon semiquantitative spectrochemical analyses as contrasted with computations based upon gravimetric chemical analyses, but it is not certain that this level of agreement will obtain universally.

The calculations have not been listed in detail. They show no detectable differences in the distribution patterns of uranium, thorium, and major and minor elements within the three rock series, and provide no basis for inferring uranium or thorium loss or gain. Moreover, no single minor element serves as an index of high uranium or thorium abundances. As expected, the statistical treatment establishes generally close positive associations for both uranium and thorium, with the trivalent or quadrivalent transition or rare-earth metals having ionic radii between 0.70 and 1.18A and electronegativities between 1.3 and 1.6 on the Pauling scale. Thus, elements such as yttrium, zirconium, and the rare earths show consistently high positive correlations with uranium; scandium, titanium, and niobium(?) show somewhat less regular associations. Data for thorium follow a somewhat similar pattern, but an insufficient amount of information has been gathered to permit rigorous comparison with the uranium results.

Two elements show abnormal patterns. Lead is significantly (at the 95-percent confidence level) correlated with uranium in 4 samples out of 10 and with thorium in 2 samples out of 5. The amount of lead in the rocks greatly exceeds that to be expected from radioactive breakdown, and the question remains as to whether the calculations demonstrate real geochemical associations of uranium and lead, or analytical difficulties. Although U^{+4} shows limited isomorphism with Ca^{+2} in apatite (Altschuler and others 1958), and presumably also in sphene and epidote, this effect is apparently overcome by the otherwise random relations of the two elements in these rock series.

Sodium and uranium show inverse correlation coefficients, valid at the 95-percent level of significance in 6 samples out of 10. This relation may be interpreted to mean that lattice positions most favorable for sodium are least favorable for uranium, and that the final localization of uranium is sensitive to both positive and negative structural crystallographic factors. More and better analytic data would doubtless bring out other significant negative associations. It is perhaps worthwhile to emphasize that negative (or positive) geochemical associations in minerals are not necessarily indicative of trends in the magma; elements which may (and do) concentrate residually at a parallel rate (for example, thorium and potassium) may become enriched in different mineral hosts in a fully consolidated rock.

METAMORPHIC AND TEMPERATURE EFFECTS

Among the striking differences between the Highlandcroft and Oliverian Series contrasted to the New Hampshire Series is the makeup of their accessory mineral suites. A comparison of these suites as well as information on accessory mineral suites of some metamorphic

rocks is shown in table 13. These data are based on separation of accessory minerals from samples weighing from 5 to 30 pounds; they are somewhat more detailed than, but in general agreement with, a similar tabulation compiled by Billings (1956, p. 178-180).

The obvious differences in table 13 are the associations of monazite and xenotime with high-grade metamorphic rocks and with the New Hampshire Plutonic Series, and the associations of epidote and sphene with lower grade metamorphic rocks and with the Highlandcroft and Oliverian Series. In mineral concentrates of the last two series, it is also notable that ilmenite is present only in trace amounts (that is, <0.01 percent), but in the New Hampshire Series it ranges from 0.20 to 0.50 percent in abundance.

Another significant discordance is in the amount of titanium carried by the biotites. In the New Hampshire Series (table 12) this is consistently in the semiquantitative spectrographic range of 1 to 5 percent; in the Oliverian Series (table 8) only two of four biotites lie within the range and 2 fall below (0.1 to 0.5 percent, and 0.5 to 1 percent). Several writers (e.g., Ramberg, 1952, p. 73) have pointed out that amounts of titanium in ferromagnesian minerals generally increase with temperature. The textural evidence previously cited concerning the development of metamorphic sphene in Highlandcroft and Oliverian rocks, coupled with the above data, strongly suggests that the source of the titanium for metamorphic sphene comes in large part from biotite (or hornblende) by a reaction such as: calcium-rich plagioclase+titanium-rich biotite $\leftrightarrows$ sphene+titanium-poor biotite+calcium-poor plagioclase.

Although sphene is not unstable in high-temperature rocks, its abundance is apparently inversely related to the abundance of titanium accepted in ferromagnesian rocks, or crystallized as ilmenite. Rarity or absence of sphene (and epidote) thus deprives the rocks of acceptors for the rare-earth elements and induces the development of monazite and xenotime. In the southern Appalachians, Overstreet and others (1956) have noted a coincidence of the sillimanite and monazite isograds—a situation which is reproduced in the regionally metamorphosed rocks of western New Hampshire. Because monazite has been synthesized at temperatures as low as 200°C (Anthony, 1957), its development in high-temperature metamorphic rocks is apparently largely conditioned by the availability of sphene and epidote rather than by the pressure-temperature factor alone.

The mineralogy of metamorphosed plutonic complexes thus exercises major control on the distribution of uranium and thorium. In high-temperature rocks where sphene may be rare and epidote absent, zircon, monazite, xenotime, apatite, and the opaque accessories can accept only a limited amount (10 to 30 percent) of the total available

TABLE 13.—*Accessory minerals in plutonic and metamorphic rocks of western New Hampshire*

Rock series	Samples studied	Samples containing—																
		Pyrite	Pyrrhotite	Magnetite	Ilmenite	Molybdenite	Rutile	Sphene	Allanite	Epidote	Monazite	Xenotime	Zircon	Apatite	Tourmaline	Garnet	Sillimanite	Others (sparingly present)
New Hampshire Plutonic Series	19	13	5	15	14	7	6	0	6	0	16	7	19	18	3	14	1	(1)
Littleton Formation (schist of sillimanite zone)	15	13	4	13	15	6	1	1	5	2	13	5	12	14	1	13	5	(2)
Oliverian Plutonic Series	5	5	0	5	5	3	0	5	2	5	0	0	5	5	1	1	0	(3)
Highlandcroft Plutonic Series	3	3	0	3	3	2	0	3	0	3	0	0	3	3	0	1	0	
Schists of staurolite or lower zones	30	13	4	10	28	0	20	7	2	21	0	0	2	5	3	28	0	(4)

1 Fluorite, hematite, graphite, uraninite.
2 Hematite, uraninite.
3 Fluorite, hematite.
4 Staurolite, kyanite, calcite, ankerite, sideroplesite, cordierite.

uranium and thorium. Because of a lack of suitable hosts, these two elements will therefore be dispersed among the major silicates, occupying such sites as structural defects and cation-exchange positions (Neuerberg, 1956) from which they may easily be leached. As indicated by the data in tables 3 and 7 of this paper, if epidote and sphene are plentiful, they can accept as much as 80 percent of the available U^{+4} and Th^{+4}, probably by isomorphous substitution for Ca^{+2}. The presumption is, therefore, that unmetamorphosed plutonic rocks are much more likely sources of leachable uranium and thorium than their metamorphosed equivalents, but this suggestion as yet lacks experimental confirmation.

LITERATURE CITED

Adams, J. A. S., 1955, The uranium geochemistry of Lassen Volcanic National Park, Calif.: Geochim. et Cosmochim. Acta, v. 8, p. 74–85.

Adams, J. A. S., Osmond, J. K., and Rogers, J. J. W., 1959, The geochemistry of thorium and uranium, *in* Ahrens, L. H., and others, eds., Physics and chemistry of the Earth, v. 3: New York, Pergamon Press, p. 298–348.

Adams, J. A. S., Kline, M. C., Richardson, K. A., and Rogers, J. J. W., 1962, The Conway granite of New Hampshire as a major low-grade thorium resource: Natl. Acad. Sci. Proc., v. 48, p. 1898–1904.

Altschuler, Z. S., Clarke, R. S., and Young, E. J., 1958, Geochemistry of uranium in apatite and phosphorite: U.S. Geol. Survey Prof. Paper 314–D, p. 45–90.

Anthony, J. W., 1957, Hydrothermal synthesis of monazite: Am. Mineralogist, v. 42, p. 904.

Billings, M. P., 1935, Geology of the Littleton and Moosilauke quadrangles, New Hampshire: Concord, N.H., New Hampshire Plan. Devel. Comm., 51 p., 3 pls.

——— 1937, Regional metamorphism of the Littleton-Moosilauke area, New Hampshire: Geol. Soc. America Bull., v. 48, p. 463–566.

——— 1956, Bedrock geology, pt. 2 *of* The geology of New Hampshire: Concord, N.H., New Hampshire Plan. Devel. Comm., 203 p.

Billings, M. P., and Keevil, N. B., 1946, Petrography and radioactivity of four Paleozoic magma series in New Hampshire: Geol. Soc. America Bull., v. 57, p. 797–828.

Billings, M. P., and Rabbitt, J. C., 1947, Chemical analyses and calculated modes of the Oliverian magma series, Mt. Washington quadrangle: Geol. Soc. America Bull., v. 58, p. 573–596.

Brown, H. T., and Silver, L. T., 1956, The possibilities of obtaining long-range supplies of uranium, thorium and other substances from igneous rocks: *in* Page, L. R., Stocking, H. E., and Smith, H. B., eds., Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on peaceful uses of atomic energy, Geneva, Switzerland; U.S. Geol. Survey Prof. Paper 300, p. 91–95.

Butler, A. P., 1962, Ratio of thorium to uranium in some plutonic rocks of the White Mountain plutonic-volcanic series in New Hampshire; *in* Geological Survey Research 1961: U.S. Geol. Survey Prof. Paper 424–B, art. 31, p. B67–B69.

Chapman, C. A., and others, 1938, Geologic map and structure sections of the Mascoma quadrangle, New Hampshire: New Hampshire Highway Dept.

Coulomb, René, Goldsztein, M., and Le Mercier, M., 1958, L'uranium dans quelques granites français: Geochim. et Cosmochim. Acta, v. 15, p. 10–17.

Flanagan, F. J., 1957, Semiquantitative spectrographic analyses and rank correlation in geochemistry: Geochim. et Cosmochim. Acta, v. 12, p. 315–322.

Fowler-Lunn, K. S., and Kingsley, Louise, 1937, Geology of the Cardigan quadrangle, New Hampshire: Geol. Soc. America Bull., v. 48, p. 1363–1386.

Grimaldi, F. S., May, Irving, and Fletcher, M. H., 1952, U.S. Geol. Survey fluorimetric methods of uranium analyses: U.S. Geol. Survey Circ. 199, 20 p.

Hadley, J. B., and others, 1938, Geologic map and structure sections of the New Hampshire portion of the Mt. Cube quadrangle: New Hampshire Highway Dept.

Hurley, P. M., Fairbairn, H. W., Pinson, W. H., and Faure, G., 1960, K–A and Rb–Sr minimum ages for the Pennsylvanian section in the Narragansett Basin: Geochim, et Cosmochim. Acta, v. 18, p. 247–258.

Larsen, E. S., Jr., and Phair, George, 1954, The distribution of uranium and thorium in igneous rocks, *in* Faul, Henry, ed., Nuclear geology: New York, John Wiley & Sons, p. 75–89.

Larsen, E. S., Jr., and Gottfried, David, 1961, Distribution of uranium in rocks and minerals of Mesozoic batholiths in western United States: U.S. Geol. Survey Bull. 1071–C, p. 63–103.

Larsen, E. S., 3d, and Gottfried, David, 1960, Uranium and thorium in selected suites of igneous rocks: Am. Jour. Sci., v. 258–A, p. 151–169.

Levine, Harry, and Grimaldi, F. S., 1958, Determination of thorium in parts per million range in rocks: Geochim. et Cosmochim. Acta, v. 14, p. 93–97.

Lyons, J. B., 1962, Uranium and thorium in the older plutonic rocks of New Hampshire; *in* Geological Survey Research 1961: U.S. Geol. Survey Prof. Paper 424–B, art. 32, p. B69–B71.

McKelvey, V. E., Everhart, A. L., and Garrels, R. M., 1955, Origin of uranium deposits: Econ. Geology (50th Anniversary volume), p. 464–533.

McKeown, F. A., 1951, Reconnaissance of radioactive rocks of Vermont, New Hampshire, Connecticut, Rhode Island, and southeastern New York: U.S. Geol. Survey TEI 67, issued by U.S. Atomic Energy Comm. Tech. Inf. Service, Oak Ridge, Tenn., 46 p.

Neuerburg, G. F., 1956, Uranium in igneous rocks of the United States: *in* Page, L. R., Stocking, H. E., and Smith, H. B., eds., Contributions to the geology of uranium and thorium by the United States Geological Survey and Atomic Energy Commission for the United Nations International Conference on peaceful uses of atomic energy, Geneva, Switzerland; U.S. Geol. Survey Prof. Paper 300, p. 55–64.

Overstreet, W. C., Cuppels, N. C., and White, A. M., 1956, Monazite in southeastern United States: U.S. Geol. Survey Prof. Paper 300, p. 597–604.

Page, L. R., 1960, The source of uranium in ore deposits: Internat. Geol. Cong., 21st, Copenhagen, 1960, Rept., pt. 15, p. 149–164.

Phair, George, 1952, Radioactive Tertiary porphyries in the Central City district, Colorado, and their bearing upon pitchblende deposition: U.S. Geol. Survey TEI 247, issued by U.S. Atomic Energy Tech. Inf. Service, Oak Ridge, Tenn., 53 p.

Ramberg, Hans, 1952, The origin of metamorphic and metasomatic rocks: Chicago, Chicago Univ. Press, 317 p.

Rogers, J. J. W., and Ragland, P. C., 1961, Variation of thorium and uranium in selected granitic rocks: Geochim. et Cosmochim. Acta, v. 25, p. 99–109.

Tilton, G. R., Davis, G. L., Wetherill, G. W., and Aldrich, L. T., 1957, Isotopic ages of zircons from granites and pegmatites: Am. Geophys. Union Trans., v. 38, p. 360–371.

Turovsky, S. D., 1957, On the geochemistry of uranium and thorium—the distribution of radioactive elements in the eruptive rocks of north Kirghizia: Geokhimiya [in translation], 1957, No. 2, p. 161–176 [1960].

Whitfield, J. M., Rogers, J. J. W., and Adams, J. A. S., 1959, The relationship between the petrology and the thorium and uranium contents of some granitic rocks: Geochim. et Cosmochim. Acta, v. 17, p. 248–271.

Vistelius, A. B., 1958, Paragenesis of sodium, potassium, and uranium in volcanic rocks of Lassen Volcanic National Park, Calif.: Geochim. et Cosmochim. Acta, v. 14, p. 29–34.

○