

THE DISTRIBUTION OF LEAD BETWEEN COEXISTING K-FELDSPAR AND PLAGIOCLASE¹

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ABSTRACT

The concentrations of lead in coexisting K-feldspar and plagioclase were determined by isotope dilution for plutonic, volcanic, and metamorphic rocks covering the geologic age range 25–2500 m.y. The isotope dilution values available for the concentration of lead in K-feldspar ranges from 9.5 [reported by Tilton *et al.*, 1955] to 114 ppm in a pegmatite [this paper]. The most common value of the distribution ratio favors K-feldspar within 10 percent of a factor of 2.4 and no example was found where lead is enriched in plagioclase relative to K-feldspar. A single value of the distribution ratio could mean that the ratio is not a function of temperature, that all the feldspar pairs investigated formed at close to the same temperature, or that lead in minerals exchanges rapidly with their surroundings upon cooling to some minimum temperature.

The distribution ratio of lead for coexisting sanidine and glass from two vitrophyres and one rhyolite obsidian is within 15 percent of 1.

INTRODUCTION

In previous investigations of the distribution of trace elements between coexisting K-feldspar and plagioclase (Heier, 1960, Taylor *et al.*, 1960, Howie, 1955), the lack of systematic trends in the lead distribution factor ($Pb_{K\text{-feldspar}}/Pb_{\text{plagioclase}}$) has posed a vexing problem. Citing the data of Heier (1960) and Howie (1955), Barth (1961, p. 7) suggests that plagioclase, rather than K-feldspar, is the more favorable host for Pb, as evidenced by the fact that, with minor exceptions, plagioclase is richer in Pb than the coexisting K-feldspar. Similarly, Heier (1962, p. 441) also concludes that the ratio $Pb_{K\text{-feldspar}}/Pb_{\text{plagioclase}}$ is $\bar{< 1$ but that reversals in this ratio may occur "in the very late stage of pegmatite formation." These conclusions are contrary to theoretical consideration because: (1) the Pb content of igneous rocks increases with increasing silicity (that is, the relatively K-feldspar-rich rocks, Wedepohl, 1956), from which one might guess that lead would be enriched in K-feldspar; and (2) Ba and Pb minerals are, in general, isotypic and the Ba-feldspar, has a complete solid solution series with K-feldspar so that again one might surmise that Pb would be preferentially enriched in K-feldspar. Indeed, synthesis of lead feldspar reported recently by Sorrell (1962) prompted him to state (p. 308): "The structural and thermal similarities between the lead feldspar and potash feldspars also suggests that a complete isomorphous series might exist between these phases." Heier (1962), however, felt that the tendency of lead to form covalent bonds supported a small fractiona-

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tion factor which might favor plagioclase over K-feldspar. However, most data on lead distribution are by the emission spectrographic method, and confirmation of previous workers' conclusions by an independent, more precise method seemed warranted, especially in view of the apparent contradiction between the existing data and deductions based on crystal chemical reasoning. The present status of this problem is perhaps best stated by Heier (1962, p. 441): "On the whole the available data on Pb is confusing."

A precise determination by isotope dilution of the lead distribution factor between perthite and plagioclase (2.5) from a billion-year-old granite, Ontario, has been available for some time (Tilton *et al.*, 1955) (Sample 7, Table 1). A determination of the distribution factor between microcline and oligoclase (2.2) from a billion-year-old granite, Texas, has become available more recently (Zartman, 1965) (sample 8, Table 1). Even though the lead concentration levels differed by a factor of four, the distribution ratio in these two sample pairs from rocks of comparable Precambrian age were similar. This similarity prompted us to determine the Pb distribution between K-feldspar and plagioclase from other feldspar pairs. This paper includes lead concentration data for additional feldspar pairs from widely separated geographic localities in North America representing a wide span of geologic time (25–2500 m.y.), and pairs from volcanic, plutonic, and metamorphic rocks.

Potassium feldspars with above average and below average Pb contents are included in this study (Table 2). The best estimate to date of the average lead content of K-feldspar (50 ppm) for North America has been determined by Patterson and Tatsumoto (1964) on K-feldspar separated from five beach and river sand composites (West Coast, East Coast, Gulf Coast, and two from the continental interior). Each of the composites contains lead within 10 percent of 50 ppm.

ASSUMPTIONS CONCERNING MINERAL IMPURITIES

Lead concentrations of each separate were corrected for impurities, if any, based on the assumption that the impurities had the same lead content as in their corresponding concentrates and that quartz contains no appreciable lead. Completely "clean" mineral separates are difficult to obtain from some volcanic rocks because of adhered glass on feldspar grains (e.g., Sample 1) and from some granitic rocks in which the plagioclase is oligoclase and has a specific gravity similar to that of quartz. For each specimen the kind and amount of admixed impurities were estimated by one or more of the following methods: 1) optical counting of oil immersions; 2) modal analysis (by Chayes-type click stage) of stained grain mounts (cobaltinitrite method for K-feldspar, amaranth method of

TABLE 1. MODES OF MINERAL SEPARATES AND DESCRIPTIONS OF SAMPLES

Sample no.	Mineral separate	Volume percent		Glass	Reference	Description
		K-feldspar	Quartz			
0.	Sanidine (Or ₉₁) Plagioclase (An <20) Glass	84.3	5.1	—	This paper	(Field No. 15-65-K) Rhyolite obsidian from Mono Craters, California (119°02.2'W, 37°56.6'N). Age from Evernden and Curtis, 1965.
		1.2	45.2	53.6		
		—	≤3	>97		
1.	Sanidine Plagioclase (avg. An 40) Glass	76.7	22.7	0.6	This paper	(Field No. DS-10) Basal vitrophyre of the Fisher quartz latite at Fisher Mountain, San Juan Mtns., Colo. (106°56'W, 37°41'N). Age from Harald Mehnert (oral communication, 1963).
		—	>99	—		
		—	—	>99		
2.	Sanidine (Or ₇₈) Plagioclase (>An 40) Glass	98.5	0.75	0.75	This paper	(Field No. 75111) Rhyolite vitrophyre of the Lowland Creek Volcanics, southwestern Montana, (46°02'44"N, 112°37'15"W). Age from R. Marvin and H. Thomas (oral communication, 1963).
		—	>95	>5		
		—	—	>5		
3.	Orthoclase microperthite (Or ₉₀) Oligoclase (An ₀₋₃₅) Quartz	>99	—	—	This paper	(Field No. 6K445) Alaskite from the Butte Quartz Monzonite, Boulder batholith, Montana (~46°00'30"N, ~112°16'W). Age from R. Marvin and H. Thomas (oral communication, 1963).
		2.6	95.7	1.7		
		—	1.8	98.2		
4.	Orthoclase microperthite (Or ₈₈) Plagioclase (An ₀₋₁₈) Quartz	>99	—	—	This paper	(Field No. 2T1056) Pegmatite in the leucogranodioritic Donald pluton, Boulder batholith, Montana (48°50'16"N, 112°25'39"W). Age from R. Marvin and H. Thomas (oral communication, 1963).
		—	>99.9	—		
		—	—	>99.9		
5.	Orthoclase microperthite megacrysts (Or ₈₈) Orthoclase microperthite groundmass + megacrysts (Or ₈₈) Plagioclase <An20 An20-An40 >An40	>99.9	—	—	This paper	(Field No. 2T275) Leucogranodiorite, Donald pluton of Boulder batholith, Montana (45°46'28"N, 112°21'46"W).
		>99.9	—	—		
		—	>95	<5		
—	—	53.4	46.6	—	—	
—	—	>95	<5	—	—	

TABLE 1—(continued)

Sample no.	Mineral separate	Volume percent			Reference	Description
		K-feldspar	Plagioclase	Quartz		
6.	Orthoclase Plagioclase	95.7	4.3	—	This paper	(Field No. FD13) From a pluton of the Sierra Nevada batholith, Calif. (106°56'W, 37°40'N). Age from Kistler and Dodge (1966).
		—	91.6	8.4		
7.	Perthite Plagioclase	>99	—	—	Tilton <i>et al</i> 1955	Granite exposed between Tory Hill and Essonville, Ontario (approximately 78°30'W, 45°N).
		—	>99	—		
8.	Microcline Oligoclase	99.8	0.1	0.1	Zartman (1964)	(Field No. 3gr) Town Mountain Granite of the Lone Grove pluton, Llano Uplift, Texas (30°44.5'N, 98°27.2'W).
		—	99.2	0.8		
9.	Orthoclase Plagioclase	>99	—	—	This paper	(Field No. 8-4-131) Biotite muscovite granite from Big Thompson Canyon, Front Range, Colorado, 40°25'N, 105°25'W (description and age are from Z. E. Peterman, oral communication).
		4.0	86.5	9.5		
10.	Microcline	74.3	25.7	—	This paper ¹ Z. E. Peterman	(Field No. KA354) Granite from Ben Island, Ontario (48°40'N, 93°08'W). The age is given by S. S. Goldich (personal communication) as K-Ar on biotite.
		76.2	23.8	24.8		
	Plagioclase	20.0	80.0	—	This paper ¹ Z. E. Peterman	
		19.8	79.0	1.2		
11.	Microcline Plagioclase	98.1	1.9	—	This paper	(Field No. BH-14(B)2) Granitic gneiss along Little Elk Creek, Black Hills, So. Dakota (44°14.3'N, 103°29.6'W). Age from Zartman, Norton, and Stern (1964).
		—	47.6	52.4		

¹ Written communication.

Or contents are for the potassic phase estimated from (201) reflection, hence, in the case of micropertithites, the bulk Or content would be slightly less. An contents are estimated from specific gravity and/or refractive indices and extinction angles.

TABLE 2. CONCENTRATION OF LEAD IN SELECTED FELDSPARS AND WHOLE ROCKS

Sample no.	Rock name	State or province	Age ¹ (m.y.)	Pb concentration raw values				Pb concentration corrected for impurities				Pb distribution	
				K-feldspar	Plag.	Glass or whole rock	Qtz.	K-feldspar	Plag.	Glass or whole rock	Qtz.		
Volcanic Rocks													
0.	Rhyolite obsidian	Calif.	<0.06	26.2	6.9	28.8	—	30.2	14.4	28.8	—	—	—
1.	Quartz latite vitrophyre	Colo.	25	27.6	13.1	27.5	—	31.8	13.1	27.5	—	—	—
2.	Rhyolite vitrophyre	Mont.	50	36.6	14.4	41.6	—	36.8	14.4	41.6	—	—	—
Plutonic rocks													
3.	Alaskite ²	Mont.	75	45.1	19.1	—	≤1.8	45.1	18.7	—	—	—	—
4.	Pegmatite	Mont.	75	114	46	—	≤0.18	114	46	—	—	—	—
5.	Leucogranite ³	Mont.	75	41.2	—	—	—	41.2	—	—	—	—	—
	megacrysts			43.2	—	—	—	43.2	—	—	—	—	—
	megacrysts + groundmass												
	(<An20)				18.8	—	—	18.8	—	—	—	—	—
	(An20-An40)				9.73	—	—	18.2	—	—	—	—	—
	(>An40)				17.7	—	—	17.7	—	—	—	—	—
6.	Granodiorite	Calif.	90	51	35±5 ⁴	—	—	52	38	—	—	—	—
7.	Granite	Ontario	1000	9.5	3.8	9.3	—	9.5	3.8	9.3	—	—	—
8.	Granite	Texas	1000	36.2	16.2	21.4	—	36.2	16.2	21.4	—	—	—
9.	Granite	Colo.	1400	79.5	18.6	—	—	79.5	17.8	—	—	—	—
10.	Granite	Ontario	2500	32.9	17.6	—	—	39.7	12.2	—	—	—	—
Metamorphic rocks													
11.	Gneiss ⁵	So. Dakota	2500	34.5	7.4	—	—	34.9	15.5	—	—	—	—

Mineral separates for 2-5, were donated by R. I. Tilling; those for 0 and 1 were made by G. T. Coburn; 6, by Frank Dodge; 9, by W. A. Braddock; 10, by Z. E. Peterman; and 11, by R. E. Zartman.

¹ References to ages are given in Table 1.

² Uranium and thorium concentrations are in ppm respectively, 0.29 and 0.31 for K-feldspar, 0.17 and 0.20 for plagioclase, and 0.08 and 0.25 for quartz by isotope dilution.

³ Has "hybrid" isotopic compositions suggestive of metasomatic contamination (see Table 3 and text).

⁴ Poor mass spectrometer analysis.

⁵ Uranium and thorium concentrations are, respectively, 0.10 and 0.19 for K-feldspar, 0.23 and 0.21 for plagioclase by isotope dilution.

Laniz *et al.*, 1964, for plagioclase); and 3) X-ray diffraction intensities of powder patterns.

Plagioclase separates are probably biased toward higher or lower anorthite contents than the average value in the rock, depending on the nature of compositional zoning and of the separatory techniques used to obtain the separate with the least quartz. The possible effect of this mineral purification on Pb content was tested on Sample 5, in which the plagioclase was split into three ranges of An content ($< \text{An } 20$, $\text{An } 20\text{-An } 40$, and $> \text{An } 40$) by specific gravity differences. The lead content of these three fractions (given in Table 2) varied no more than 10 percent, indicating that Pb data for plagioclases probably are not biased to any great extent in the mineral separation procedure.

The assumption that quartz contains insignificant lead is probably valid. Murthy and Patterson (1961) determined the lead contents of two quartz samples (separated by the fluorboric acid method) from the Boulder batholith as 0.20 and 0.13 ppm. We have augmented their analyses with two additional quartz samples (concentrated by specific gravity separations only). Sample 4 was a good separate and contained a lead content comparable to the values of Murthy and Patterson. The quartz from sample 3 contained ≤ 1.5 ppm, but the slightly cloudy, grayish appearance of the quartz perhaps reflects the presence of submicroscopic impurities that may contain Pb in addition to the observed plagioclase impurities. These data support the conclusion that quartz contains much less lead than feldspars, probably less than 0.5 ppm. This conclusion is corroborated by the analyses of the plagioclase in Sample 5, in which the fraction of intermediate An content also contained an intermediate lead content when corrected for admixed quartz assuming the quartz contained no lead.

The final step in the mineral purification procedure is a leach with hot 6N HCl just prior to decomposition of the sample. Plagioclase is more reactive to HCl than is K-feldspar, and on Sample 3 a test was made of the amount of lead removed by the leach. The amount of lead leached was equivalent to 0.3 ppm on the K-feldspar and 0.4 ppm on the plagioclase, values that amount to 1–3 percent of the lead remaining in the feldspar. Thus, the lower lead content of the plagioclase (relative to the coexisting K-feldspar) cannot be ascribed to the leaching procedure. The weight loss of the sample was measured, and the leachate was analyzed for Pb content. It was found that the leachate contained an extremely high concentration of lead, > 300 ppm, which probably was derived from adsorbed bromoform used in the heavy liquid separation even though the samples were thoroughly washed with acetone.

The lead contents determined for the mineral separates have analytical

uncertainties of about 2 percent. Additional uncertainties introduced in the mineral purification procedure and in correcting for mineral impurities increased the analytical uncertainty to about 5 percent for each mineral or about 10 percent for the distribution ratio.

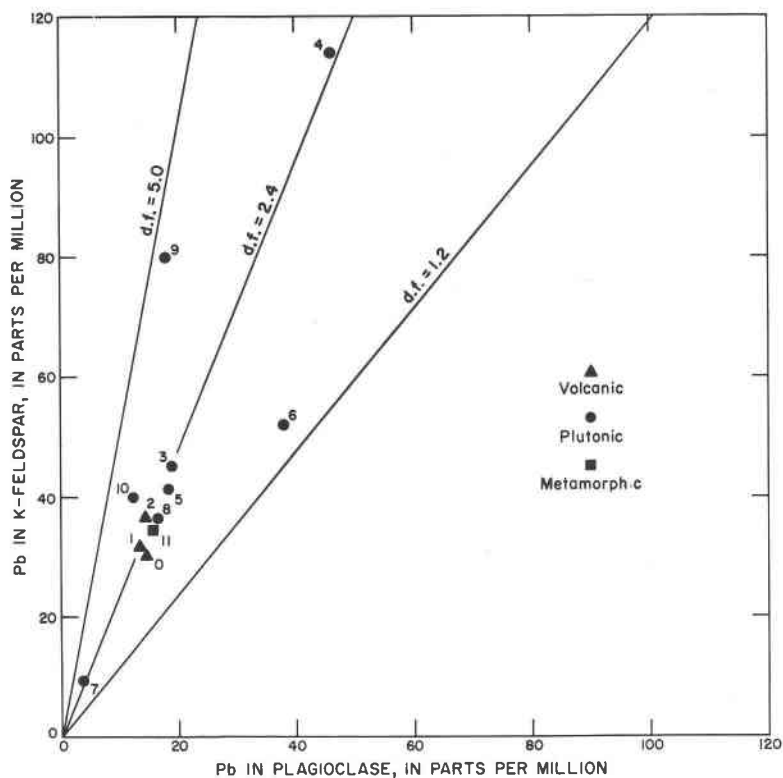


FIG. 1. Distribution of lead between coexisting K-feldspar and plagioclase. Distribution factors (d.f.) of 5.0, 2.4, and 1.2 are shown for reference. (Numbers refer to samples in Tables 1 and 2.)

DISCUSSION OF DATA

The lead concentration data are given in Table 2 and shown on Figure 1, the modes of the mineral separates are given in Table 2, and some lead isotopic compositions in Table 3. The data (Table 2) show that for all the sample pairs analyzed by isotope dilution, in contrast to earlier spectrographic data, Pb is enriched in the K-feldspar relative to plagioclase. Furthermore, 9 of the 12 samples have Pb distribution factors within 20 percent of a factor of 2.4 after the mineral separates are cor-

rected for mineral impurities. This nearly uniform distribution factor is particularly remarkable in view of the fact that the feldspars analyzed range from 25 to 2500 m.y.; they include volcanic, plutonic, and metamorphic rocks; and they represent rocks which have K-feldspars with unusually high and unusually low lead contents relative to the average lead content of K-feldspar (50 ppm) of Patterson and Tatsumoto (1964).

TABLE 3. ISOTROPIC COMPOSITION OF LEAD IN SELECTED FELDSPARS
(Normalized to Ta lot 1 filament material values by constants given in Doe, Tilton, and Hopson, 1965)

Feldspars from:	Pb ²⁰⁶ /Pb ²⁰⁴	Pb ²⁰⁷ /Pb ²⁰⁴	Pb ²⁰⁸ /Pb ²⁰⁴	Pb ²⁰⁶ /Pb ²⁰⁷	Pb ²⁰⁶ /Pb ²⁰⁸
Analytical uncertainties ¹ :	0.29	0.37	0.47	0.16	0.29
Mono Craters, California Rhyolite obsidian (Sample 0)	19.123	15.66	38.92	1.2207	0.4914
Boulder batholith, Montana Donald pluton (leucogranodiorite) (average of two analyses)	17.309	15.53	38.22	1.1143	0.4529
Donald pluton (leucogranodiorite with "hybrid" isotopic compositions (see text) (sample 5) Megacrysts	17.190	15.51	38.13	1.1084	0.4508
Megacrysts + groundmass K-feldspars	16.994	15.45	37.97	1.0997	0.4476
plagioclase (An ₂₀ -An ₄₀)	17.013	15.45	38.01	1.1013	0.4476
Granodiorite of Rader Creek (average of 4 samples)	16.909	15.41	37.68	1.0970	0.4487
Black Hills, South Dakota BH-14 (2) K-feldspar	19.082	16.44	38.14	1.1613	0.5006
plagioclase	19.847	16.50	38.53	1.2026	0.5153

¹ Standard deviation as percent of a ratio.

The metamorphic pair from a granitic gneiss (Sample 11) from Black Hills, South Dakota, comes from rocks about 2500 m.y. old metamorphosed at 1800 m.y. as determined by radiometric dating (Zartman, Norton, and Stern, 1964). Lead isotope data (Table 3) support such a conclusion and suggest that lead at least was added to the feldspars during the subsequent metamorphism (Figure 2). The lead isotope data for Sample 7, another rock of granitic composition, suggest that it too was metamorphosed and probably should be counted as a metamorphosed pair because the values of Pb²⁰⁶/Pb²⁰⁴ and Pb²⁰⁸/Pb²⁰⁴ in the feldspars (Tilton et al., 1955) are about 7 to 10 percent greater than expected for

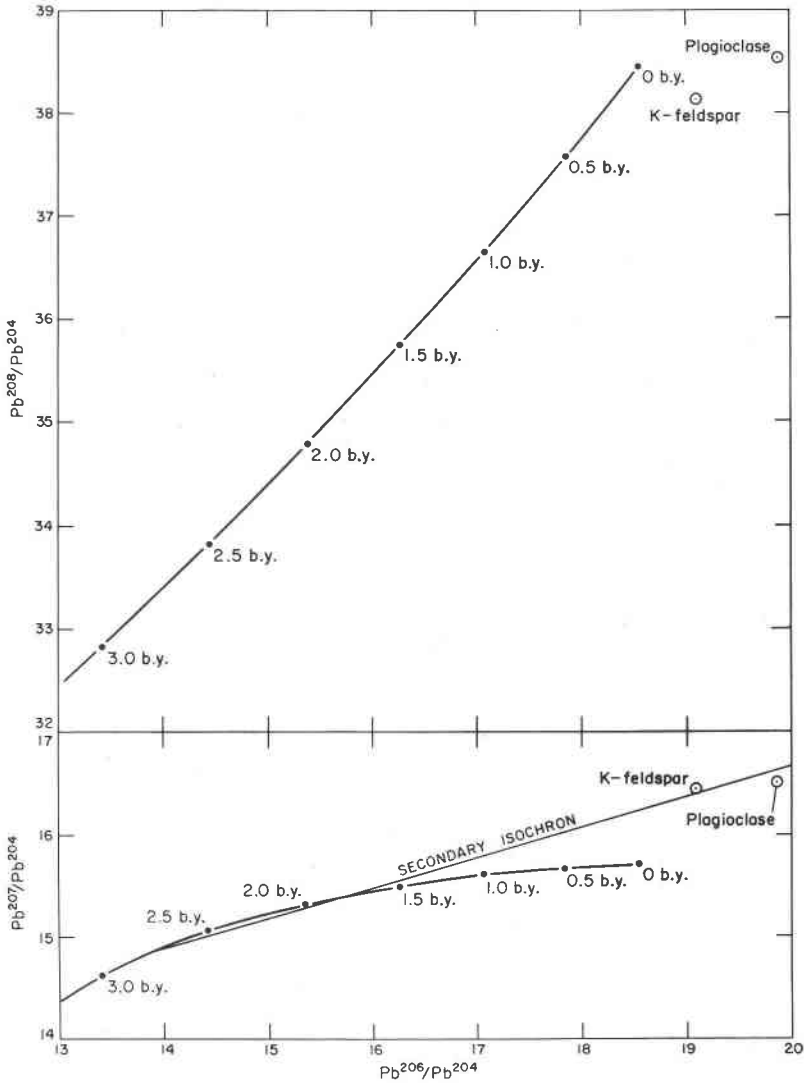


FIG. 2. Pb^{207}/Pb^{204} and Pb^{208}/Pb^{204} against Pb^{206}/Pb^{204} for a feldspar pair formed at 2.75 b.y. and metamorphosed at 1.75 b.y. Heavy lines are the *normal growth curves* for lead, indexed for various geologic times. Light straight line is a *secondary isochron* along which all leads must lie if feldspars with normal leads in a rock with any value of U^{238}/Pb^{204} , formed by 2.75 b.y. ago, had radiogenic lead added to them by metamorphism 1.75 b.y. ago.

feldspars of that age (Doe, 1962; Doe, Tilton, and Hopson, 1965; Zartman, 1965).

Lead distributions were determined for feldspar pairs from a sample in which the isotopic composition of lead is complex (Sample 5, Table 3), presumably reflecting metasomatic action. The isotopic compositions of lead in what is essentially groundmass feldspar and plagioclase are similar, and the lead distribution factor of 2.4 is similar to that found for feldspar pairs from other samples. Megacrysts up to 4 cm in length in Sample 5 grew late and have a distinctly different lead isotopic composition from the groundmass feldspars. The lead distribution factor between megacryst K-feldspar and groundmass plagioclase is 2.3, again similar to other samples in spite of the isotopic difference between the two minerals.

This isotopically "hybrid" rock was unequivocally mapped as part of the leucogranodioritic Donald pluton (SiO_2 —70.1%, biotite > hornblende) on the basis of its distinctly leucocratic aspect and its sharp contact with the adjacent older granodiorite of Rader Creek (average $\text{SiO}_2 \approx 64.0\%$, hornblende > biotite) which is entirely devoid of K-feldspar megacrysts. The isotopic composition of lead in the megacrysts of the hybrid rock (Sample 5) is more similar to that of the Donald pluton (Table 3); however, the groundmass feldspars contain leads with isotopic compositions more similar to that of feldspars in the granodiorite of Rader Creek. Within analytical uncertainties, the isotopic data suggest the source material of the megacrysts was derived (apparently metasomatically) from the younger leucogranodiorite of the Donald pluton which added K-feldspar megacrysts to the adjacent older granodiorite of Rader Creek with little or no effect on the groundmass feldspars, thus resulting in the "hybrid" array of lead isotopic compositions. The "hybrid" (Sample 5) leucogranodiorite of the Donald pluton bears no other imprint of metasomatic action (neither in bulk chemical composition nor in petrographic characteristics). The lead content of the megacryst K-feldspars and what is essentially groundmass K-feldspars are similar to each other within 5 percent. This similarity in lead contents could be considered a coincidence as there is no demonstrable isotopic interchange between the megacrysts and groundmass feldspars; however, the lead content and physico-chemical parameters of the K-feldspar depositing fluid may not have been completely independent of the rock unit that was penetrated. An example more clearly including isotopic interchange is desired for this case (metasomatic-hydrothermal) but is not available.

As stated previously, in no sample studied is the distribution ratio less than 1.0 and in 9 of the 12 samples the distribution ratios are clustered about 2.4. The causes of the apparent departure of the three samples from the 2.4 factor are not understood. Two of the samples depart from the

most common ratio by about 35 percent (Samples 6 and 10); the other departs by about 90 percent (Sample 9). Mass spectrometric data on Sample 6 are not as good as on the other samples because of the lack of the usually accepted ion intensity and of problems with lead contamination in the laboratory. Additional material was not available for re-determination of Sample 6. Plagioclases in the Precambrian samples analyzed tend to have a greater opacity (due to alteration and submicroscopic impurities?) than plagioclases in the other samples. Perhaps these possible alteration effects may account for some depletion of lead in the Precambrian plagioclase of Samples 9 and 10. We therefore have no conclusive evidence of primary distribution factors different from 2.4.

Theoretically, the apparently uniform distribution factor (2.4) of the majority of the samples could reflect that lead exchanges rapidly during cooling of a magma to some minimum temperature, that all mineral pairs formed at nearly the same temperature, or that the distribution factor is relatively insensitive to temperature. Another possibility is simply that the observed relations are purely coincidental owing to small sample size and the survey nature of this study. However, the sample pairs represent widely separated geographic localities and a wide range of geologic time and contain lead concentration levels that differ by a factor of more than 10, including very low levels as well as high levels. Therefore, we do not believe that the rather uniform lead distribution factor is a mere coincidence. More data, particularly from sample pairs from comagmatic igneous suites and on metamorphic rocks of different bulk compositions with additional thermal and chemical information (e.g., K, Na, Ca, Ba contents) are needed before we can evaluate the petrologic and geochemical parameters which govern the partitioning of Pb between coexisting K-feldspar and plagioclase.

Three distribution factors of Pb (K-feldspar)/Pb (glass) were determined in the course of this study and were found to be within about 15 percent of 1.0. Apparently the distribution factor between sanidine and glass is nearly one, but more data are needed to arrive at any conclusions from such a distribution factor.

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