The geochemistry of radiogenic strontium.

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Introduction.

The important influence that the radioactive decay of the radioelements uranium and thorium has had on the geochemical distribution of lead is now well appreciated, but little is known about the possible geochemical implications of the radioactivity of rubidium.

Like its alkali metal homologue potassium, rubidium is β radioactive and slowly disintegrates into a stable isotope of strontium: small quantities of strontium have therefore been accumulating in various minerals since the earth's crust first consolidated. This paper will describe firstly, the abundance of radiogenic strontium in the earth's crust as a whole, and secondly, the distribution of radiogenic strontium in various minerals and rocks, in particular those which are of igneous origin.

Calculation of the amount of radiogenic strontium generated by rubidium.

The disintegration equation for any radioactive element may be written as follows:

$$N = N_o e^{-\lambda t}, \qquad (1)$$

where N_o is the number of atoms originally present and N the number left after a time t has elapsed; λ is the disintegration constant for the particular radioactive element in question. According to Hahn and Walling (1) and Hemmendinger and Smythe (2), in rubidium only the isotope Rb^{87} is radioactive and by losing an electron (β particle) a strontium isotope of equivalent mass number, Sr^{87}, is obtained: in the above equation N_o and N refer therefore to atoms of Rb^{87} and λ to the disintegration constant for Rb^{87}.

The radioactive decay of Rb^{87} is extremely slow and during earth periods only a very small proportion of the total rubidium will have decayed, and for calculations on the radiogenesis of strontium one may regard the quantity of rubidium in any mineral or rock as having remained constant throughout its lifetime. Equation (1) can then be simplified and written as follows:

$$N = N_o - \lambda t N \text{ or } N_o - N = \lambda t N. \quad (2)$$

But $N_o - N$ is the number of atoms of Rb^{87} that have decayed and this number is equal to the number of Sr^{87} atoms generated. Furthermore, Rb^{87} and Sr^{87} have the same mass and equation (2) can be expressed as,

$$\% \text{ radiogenic Sr} = \% \text{ Rb}^{87} \times t \times \lambda. \quad (3)$$

If λ, t, and % Rb^{87} are known, it is then possible to calculate the quantity of radiogenic strontium present in any material.

The value of λ.—Hahn (3) gives $1{\cdot}1 \times 10^{-11}$ year^{-1} as the value for λ. This value is probably more accurate than previous direct measurements of λ and is based on Pb/U age determinations of certain ancient uranium minerals which were believed to be of the same age as a rubidium-rich mica which was found to contain only radiogenic strontium. A quantitative analysis was made of the rubidium and also of the strontium in the mica, and because the age of the mineral was known, the period of half-life and the disintegration constant for Rb^{87} could be determined.

The value of t.—The absolute geologic ages of some of the specimens described in this paper are believed to be relatively accurate, since the ages of the formations in which the specimens occur have been determined. The ages of other specimens can only be regarded as approximate and have been obtained by interpolating or extrapolating whatever age data are available. However, for this discussion on the radiogenesis of strontium in minerals very accurate age data are not necessary.

Percentage of Rb^{87}.—Rubidium consists of two isotopes only, Rb^{85} and Rb^{87}, the relative abundance of the latter isotope being 27 %. Total rubidium is estimated spectrochemically (ordinary chemical methods are sometimes used) and 27 % of this quantity = % Rb^{87}.

THE ABUNDANCE OF RADIOGENIC STRONTIUM IN THE EARTH'S CRUST.

To calculate the amount of radiogenic strontium in the earth's crust, t will be equivalent to the time that has elapsed since the formation of the earth. Unfortunately there still is a considerable degree of uncertainty as to the exact age of the earth, although there appears to be little doubt about the order of magnitude: a value of $t = 3{\cdot}0 \times 10^9$ years will be used here.

Much research is still required before it will be possible to estimate accurately the abundance of rubidium in the earth's crust. Goldschmidt (4) places the concentration at about 0·03 % Rb, whereas I. and W. Noddack (5) give a much lower value. With an increased amount of data at one's disposal, it would seem that the former value is more nearly correct and will be used for calculating the amount of radiogenic strontium in the earth's crust. The approximate abundance of Rb^{87} is thus about 0·0075 %, whence by substituting for t, % Rb^{87}, and λ in equation (3) the abundance of radiogenic strontium in the earth's crust is calculated as 0·00025 %.

THE PROPORTION OF RADIOGENIC STRONTIUM IN THE TOTAL STRONTIUM IN THE EARTH'S CRUST.

According to Goldschmidt (4) the concentration of strontium in the earth's crust is about 0·04 %, whereas I. and W. Noddack give about one-half of this value. In general there seems to be considerable disagreement on the amounts of strontium found in different rock types, and in this paper the mean of the above two values (0·03 %) will be used. The proportion of radiogenic strontium in the total strontium in the earth's crust is thus approximately $0{\cdot}00025/0{\cdot}03 \times 100 = 0{\cdot}8$ %.

Although the proportion of radiogenic strontium in the total strontium is thus not very considerable, it cannot be regarded as negligible and it will be shown later that in several minerals, particularly in those which have formed from residual pegmatitic emanations, the radioactive decay of rubidium is the chief source of strontium: this follows from the rather different ways in which strontium and rubidium behave during the selective crystallization of magmatic melts and their emanations.

THE ABUNDANCE OF RADIOGENIC STRONTIUM IN MINERALS OF IGNEOUS ORIGIN.

From equation (3) it is readily seen that most radiogenic strontium will be found in ancient minerals relatively rich in rubidium. Rubidium does not form minerals of its own, but is always present in potassium-rich minerals and also the caesium mineral pollucite (Ahrens, 6); in these minerals Rb^+ (radius 1·49 Å.) is able to replace K^+ (radius 1·33 Å.) and Cs^+ (radius 1·65 Å.) within the lattice.

In potassium minerals the rubidium content varies very considerably and the ratio K/Rb is by no means even nearly constant. Since potassium and rubidium are almost identical chemically, the inconstancy of the K/Rb ratio is undoubtedly due to the difference in the radii of their respective ions. Goldschmidt (7) has pointed out that because the radius of Rb^+ is appreciably greater than that of K^+, its electrostatic field of attraction is weaker than that of K^+, and hence rubidium tends to be concentrated relative to potassium in the residual crystals of potassium minerals, for example, in those of late formation in pegmatite. Furthermore, the lattices of certain potassium minerals appear to act as more suitable hosts for accommodating the large rubidium ion; available evidence seems to indicate, for example, that Rb^+ can enter the mica lattice with greater facility than it can the felspar lattice.

The ages of several minerals that have been analysed for rubidium are known and the abundance of radiogenic strontium may be calculated: as mentioned before, the ages of some of the specimens can only be regarded as approximate. Calculations on all known analysed (for rubidium) specimens, the ages of which are known, have not been made; and in the following discussion on the abundance of radiogenic strontium in different minerals of known age, in nearly all cases only specimens that have been analysed for total strontium as well as rubidium are described. Without a knowledge of the total strontium content it is not possible to estimate the proportion of radiogenic strontium unless isotope analyses have been made. (See discussion on the proportion of radiogenic strontium in different minerals.)

Lepidolite.—Lepidolite is the mineral richest in rubidium (and hence in radiogenic strontium) and contains on an average 1·5 % Rb, the latitude of variation being about 0·5–3·0 % Rb. Some analyses and calculations on lepidolites of various ages are given in table 1.

TABLE 1. Amounts of rubidium and radiogenic strontium in lepidolite.

No.	Locality.	Approx. age* (years).	% Rb.	% Rb^{87}.	Calc. % radiogenic Sr.
1.	Pala, California	120×10^6	2·0	0·54	0·0007
2.	,, ,, ...	170×10^6	2·0	0·54	0·001
3.	Newry, Maine, U.S.A. ...	300×10^6	1·0	0·27	0·001
4.	Norway, ,, ,, ...	300×10^6	1·5	0·40	0·0015
5.	Karibib, South-West Africa	800×10^6	1·8	0·49	0·004
6.	,, ,, ,, ,,	900×10^6	0·9	0·24	0·0024
7.	Omaruru, ,, ,, ,,	1250×10^6	2·4	0·65	0·009
8.	Warmbad, ,, ,,	1050×10^6	1·6	0·43	0·005
9.	Black Hills, South Dakota	1000×10^6	1·3	0·35	0·0037
10.	Kubuta, Swaziland ...	2000×10^6	2·5	0·68	0·015
11.	Manitoba	1900×10^6	2·5	0·68	0·015

* According to Sr/Rb age measurements, with the exception of no. 11.

With the exception of no. 11, all above analyses for rubidium are from Ahrens (8); no. 11 is from Strassmann and Walling (9).

Only in the most ancient of the specimens of lepidolite (nos. 10 and 11) does the concentration of radiogenic strontium exceed 0·01 %; these ancient lepidolites contain about the maximum quantity of radiogenic strontium likely to be found in any mineral.[1]

Amazonite (green microcline).—Amazonites appear to be characteristically richer in rubidium than most other potash-felspars and usually contain 0·1–1·5 % Rb: an average concentration for rubidium in amazonite is about 0·5 %. The calculated concentrations of radiogenic strontium in some analysed specimens of known age are given in table 2.

TABLE 2. Amounts of rubidium and radiogenic strontium in amazonite.

No.	Locality.	Approx. age (years).	% Rb.	% Rb^{87}.	Calc. % radiogenic Sr.
1.	Ilmen Mts., Russia ...	200×10^6	1·3	0·35	0·0008
2.	„ „	200×10^6	2·0	0·54	0·0012
3.	Madagascar	$1000 ? \times 10^6$	1·3	0·35	0·0038
4.	Pike's Peak, Colorado ...	1050×10^6	1·3	0·35	0·0038

Rubidium analyses nos. 1 and 2 are from Tolmachev and Filippov (10) and nos. 3 and 4 are from Ahrens (11).

Pollucite.—The mean rubidium content of pollucite appears to be about the same as is commonly found in amazonite (Ahrens, 6). Calculated concentrations of radiogenic strontium in some analysed specimens of pollucite of known age are given below (table 3).

TABLE 3. Amounts of rubidium and radiogenic strontium in pollucite.

No.	Locality.	Approx. age (years).	% Rb.	% Rb^{87}.	Calc. % radiogenic Sr.
1.	Norway, Maine, U.S.A. ...	300×10^6	0·23	0·062	0·0002
2.	Greenwood, „ „ ...	300×10^6	0·68	0·18	0·0006
3.	Varuträsk, Sweden ...	$650 ? \times 10^6$	0·37	0·10	0·0007
4.	Karibib, South-West Africa	1000×10^6	0·54	0·15	0·0017
5.	Black Hills, South Dakota	1350×10^6	0·25	0·067	0·001

The rubidium contents of the above analyses are from Ahrens (6).

In general, the three mineral types discussed above (lepidolite,

[1] The four lepidolite specimens from South-West Africa and the one from Swaziland are from pegmatites associated with the 'Old Granite' of Southern Africa. Little is known about the ages of this 'Old Granite' from different areas, but the available data indicate that 1000×10^6 years may be regarded as an approximate mean age of the 'Old Granite', although it is possible that the Swaziland specimen (No. 10) may be very much older ($\times 2$) than the other 'Old Granite' specimens.

amazonite, pollucite) and probably also hydrothermal microcline other than amazonite, contain higher concentrations of radiogenic strontium than any other mineral.

Muscovite and biotite.—The rubidium content of muscovite and biotite varies very considerably and is in general usually higher than is commonly found in potash-felspar. Pegmatitic muscovites enriched in lithia appear to be enriched in rubidium. For minerals of equivalent age, muscovite and biotite appear to contain as an approximate average 1/5–1/10 of the amount of radiogenic strontium usually found in lepidolite. (See tables 5 and 6.)

Potash-felspar (microcline, orthoclase, and perthite).—Most specimens of potash-felspar appear to contain 0·01–0·3 % Rb, although from some areas felspar may be unusually poor in rubidia (for example, specimens of pegmatitic felspar from the 'Old Granite' near Mid-Illovo, Natal, South Africa, were found to contain usually less than 0·01% Rb), or unusually high (for example, the Varuträsk area in Sweden contains microcline which frequently contains 0·5–2·0 % Rb (12)): in general pegmatitic felspar appears to be richer in rubidium than parent-rock felspar.

Perhaps it is not justified to make a comparison of 'rubidium-rich' and 'rubidium-poor' areas as indicated above, until more data are available, because one important factor, that of phase of mineralization, has to be taken into account. Thus, for example, most microcline found in pegmatite has apparently been formed during the primary consolidation of the pegmatitic base material; sometimes, however, microcline has formed during a later hydrothermal phase, which may be associated with lithianization in the pegmatite, as, for example, at Varuträsk; and from general geochemical principles one would expect a considerable enrichment of rubidium in late hydrothermal microcline of this type. This factor may be an important reason for the relative richness of rubidium in microcline from the Varuträsk pegmatite.

In most cases the amount of radiogenic strontium will be small, and if we assume an average felspar to contain about 0·05 % Rb, then for a relatively young specimen (100×10^6 years) 0·000015 % Sr will be generated; in a medium-aged specimen (500×10^6 years) about 0·00006 % Sr will be generated; whereas about 0·00020 % Sr would be generated in an ancient specimen (1500×10^6 years). These amounts serve to indicate what quantities of radiogenic strontium are likely to be found in potash-felspars of different ages. Reference to the discussion on the proportion of radiogenic strontium in felspar on page 287 will show quantities of radiogenic strontium found in some specimens of felspar discussed in this paper.

The proportion of radiogenic strontium in the total strontium in different minerals and rocks.

Methods for determining the relative abundance of Sr isotopes.—Of more interest than the absolute abundance of radiogenic strontium is its proportion of the total strontium in different minerals and rocks. Common strontium consists of a mixture of isotopes Sr^{84} (0·6 %), Sr^{86} (9·9 %), Sr^{87} (7·0 %), and Sr^{88} (82·6 %), whereas radiogenic strontium consists only of Sr^{87}. In minerals and rocks the proportion of radiogenic strontium may be determined either indirectly or directly.

In the indirect method the abundance of radiogenic strontium is calculated as before and if the total strontium has been estimated experimentally, the proportion of radiogenic strontium can be calculated. This indirect method can only be regarded as rather approximate because the accuracy with which all factors (λ, t, % Rb^{87}, and % total Sr) are known in any instance is not very great. Nearly all the proportions of radiogenic strontium discussed in the next few pages are based on these indirect determinations.

The usual direct method for determining the isotopic composition of an element, and which may be used for strontium, is analysis by means of the mass spectrograph. Two other methods which warrant investigation in this respect will be mentioned. One method is based on relative intensity measurements on the hyperfine components of suitable strontium lines in line emission spectra of strontium: Heyden and Kopfermann (13) have been able to demonstrate clearly that for the lines they examined, the hyperfine structure patterns for ordinary strontium were quite different from those emitted by pure Sr^{87}, and it follows that in a mixture of ordinary strontium and radiogenic strontium, relative intensity measurements on the hyperfine components of a suitable line should indicate isotopic composition. A third possible method depends on frequency shifts in certain SrF band spectra shown by $Sr^{88}F$, $Sr^{87}F$, and $Sr^{86}F$. Although it may be possible to make isotope analyses of strontium by means of relative intensity measurements on certain SrF band spectra, the procedure will quite likely be excessively tedious.

The above methods for estimating the relative abundance of strontium isotopes depend in each case on slight variations in nuclear mass.

Recently Professor Robley D. Evans of the Massachusetts Institute of Technology suggested (private communication) another possible method for determining Sr^{87} which is based on nuclear configuration and not mass variations. His suggestion will not be discussed here in detail, but briefly it is as follows: Sr^{87} may be excited by non-capture collision to a metastable state of 2·7 hour half-period, whereas other strontium isotopes are not affected; it may therefore be possible to estimate Sr^{87} by making relative intensity measurements of the radiations emitted after bombardment with suitable missiles, for example, with the aid of a cyclotron or a van der Graaff generator.

Relative proportion of radiogenic strontium in lepidolite.[1]—According to Hahn and Walling (1), a mass spectrographic examination of strontium in an ancient lepidolite from Canada showed the strontium to be 99·7 % radiogenic. Further direct corroborative evidence that strontium in lepidolite is essentially radiogenic has been furnished by Brewer (14) who examined strontium in five different specimens and found no evidence of the presence of non-radiogenic strontium.

Data based on indirect evidence indicate that in the specimens of lepidolite referred to in table 1 the proportion of radiogenic strontium is probably between 80–100 %, with the probable exception of the specimen from Swaziland which may contain about 50 %. These results together with the mass spectrographic data show conclusively that in lepidolite radiogenic strontium is invariably predominant, and only minute traces, if any at all, of magmatic strontium are likely to be present.

Relative proportion of radiogenic strontium in amazonite.—No direct isotope analyses have been made on the strontium that is always found in amazonite. Total strontium has been determined on a few specimens of amazonite of known age, and data on the proportions of radiogenic strontium in four specimens of amazonite are given in table 4. The specimen numbers correspond to the specimen numbers in table 2.

In all these specimens the proportion of radiogenic strontium is high and sometimes predominates. An approximate analysis by Oftedal (15) of some pre-Cambrian amazonite from Telemark, Norway, substantiates these findings: about 0·5 % Rb and about 0·01 % Sr were found in the

[1] Since submitting this paper, new information has been published on the proportion of radiogenic strontium in certain minerals. Using the mass spectrograph, J. Mattauch (Angew. Chem., 1947, A, no. 2, p. 37) finds 97·4–>99·7 % radiogenic Sr in lepidolite (four specimens) and 46·3–84·3 % in microcline (four specimens). Although not stated so by the author, it is very probable that all the specimens of microcline he analysed, one of which was the amazonite variety, are *hydrothermal*.

specimen of amazonite, and because it is ancient, a relatively large proportion of the strontium must be radiogenic.

TABLE 4. Relative proportion of radiogenic strontium in amazonite.

No.		% radiogenic Sr.	% total Sr.	Approx. prop. radiogenic Sr.
1.	Ilmen Mts., Russia ...	0·0008	0·0015	50 %
2.	" " ...	0·0012	0·0020	60
3.	Madagascar	0·0038	0·01	40
4.	Pike's Peak, Colorado ...	0·0038	0·0046	80

Total strontium in nos. 1, 2, and 3 is from Tolmachev and Filippov (10), and in no. 4 from Ahrens (8).

Pollucite.—According to Hahn (3), a mass spectrographic examination of strontium in pollucite from Varuträsk, Sweden, showed it to be largely radiogenic. Indirect evidence on other specimens of pollucite referred to in table 3 indicate strongly that the bulk of the strontium in each of these minerals that has been examined is radiogenic. (See Ahrens, 6.)

Muscovite and biotite.—Recently Nockolds and Mitchell (16) published results of the analyses of several separated micas (all biotites with one exception, no. 12 in table 5) from Caledonian igneous rocks from Scotland. Rubidium and strontium are included in their analytical data, and assuming an age of about 350×10^6 years for these rocks, the approximate proportion of radiogenic strontium in each analysed mineral has been calculated (table 5).

TABLE 5. Amounts of Rb and Sr in Scottish micas.

No.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
1.	0·10	0·027	0·01	0·0001	1·0 %
2.	0·15	0·041	0·01	0·00016	1·6
3.	0·15	0·041	0·008	0·00016	2·0
4.	0·20	0·054	0·007	0·00021	3·0
5.	0·08	0·022	0·007	0·000085	1·2
6.	0·10	0·027	0·008	0·0001	1·3
7.	0·15	0·041	0·01	0·00016	1·6
8.	0·25	0·068	0·005	0·00026	5·2
9.	0·15	0·041	0·003	0·00016	5·3
10.	0·15	0·041	0·02	0·00016	0·8
11.	0·10	0·027	0·007	0·0001	1·4
12.	0·015	0·0041	0·003	0·00001	0·33
13.	0·04	0·011	0·003	0·000042	1·4
14.	0·08	0·022	0·008	0·000085	1·1

In most of the above specimens the proportion of radiogenic strontium is 1–5 %: although not very appreciable, these proportions are nevertheless very significant. Had these Caledonian rocks been ancient e.g. 1500×10^6 years), then the proportion of radiogenic strontium would have been as much as 20–30 % in some of the specimens.

In pegmatites complete data (% Rb, % total Sr, and age) on muscovite and biotite are very meagre, but one may infer that because pegmatitic muscovites invariably contain only small traces of strontium (frequently 0·001–0·01 %) and often contain appreciable traces of rubidium, the proportion of radiogenic strontium will frequently be considerable. In two ancient specimens of pegmatitic muscovite (Usakos, South-West Africa, and Game Reserve, eastern Transvaal) which were found to contain respectively 0·14 % Rb, 0·0005 % Sr, and 0·28 % Rb, 0·0012 % Sr, the proportions of radiogenic strontium have been calculated as 90 % and 70 % respectively, on the assumption that the ages of these minerals are about 1000×10^6 years. These high proportions of radiogenic strontium can by no means be regarded as being representative, and undoubtedly in many pegmatitic muscovites, in particular young ones poor in rubidia, the proportion of radiogenic strontium must be very much smaller. Thus Bray (17) analysed three specimens of pegmatitic muscovite from pre-Cambrian rocks from Colorado (see table 6) and found the mean strontium content to be 0·007 %. If we assume about 0·1–0·2 % Rb to be present (an amount frequently found in muscovite) and the ages of these pegmatites to be roughly 1000×10^6 years (Holmes, 18), about 0·0003–0·0006 % radiogenic strontium will be present and the proportion of radiogenic strontium will be approximately 5–10 %.

Noll (19) has also analysed strontium from pegmatitic muscovite and found only 0·001 % SrO: because this amount of strontium is very small one must conclude that much of it is radiogenic. A very small quantity of strontium (0·0035 %) was also found by Bray (17) in biotite from pegmatite associated with the Silver Plume granite of Colorado. Here again the proportion of radiogenic strontium must be very appreciable. In some other pegmatitic biotite specimens analysed by Bray much more strontium was found and very probably the proportion of radiogenic strontium is consequently much less.

Phlogopite.—Work carried out by the author (unpublished results) on a specimen of phlogopite from the Palabora complex, Transvaal, containing about 0·09 % Rb, showed that a very small quantity of strontium (0·0005–0·00008 %) was present and that a large proportion of the strontium is probably radiogenic.

Noll (19) analysed phlogopite from Bamle, Norway, and found less than 0·001 % Sr: since all specimens of phlogopite appear to contain appreciable traces of rubidium, one may conclude that in the Bamle specimen an appreciable proportion of the strontium must be radiogenic.

Potash-felspar other than amazonite.—Several authors (e.g. Noll, 19) have shown that strontium tends to be considerably more abundant in felspar than in mica. This may be shown conclusively by separating both felspar and mica from the same rock. Thus Bray (17) separated potash-felspar and muscovite from granites and associated pegmatites, and found the following strontium contents (table 6).

TABLE 6. Content of Sr in felspar and mica from granites, Colorado.

No.	Description.		% SrO Potash-felspar.	% SrO Muscovite.	Ratio: % SrO (mica). % SrO (felspar).
1.	Boulder Creek granite	Parent-rock ...	0·087	0·063	0·72
		Pegmatite ...	0·044	0·0055	0·12
2.	Overland Mt. granite	Parent-rock ...	0·13	0·0098	0·075
		Pegmatite ...	0·13	0·009	0·07
3.	Silver Plume granite	Parent-rock ...	0·081	0·015	0·18
		Pegmatite ...	0·061	0·0066	0·11

With the exception of the parent-rock felspar and muscovite from the Boulder Creek granite where the decrease of strontium in the muscovite relative to the felspar is not very marked, all the above muscovite specimens contain very much less strontium than the associated felspar. (Approximate ratio of % SrO in muscovite : % SrO in felspar = 1/10.) There is very little doubt that in the above specimens of felspar the proportion of radiogenic strontium is very low and usually less than 1 % of the total strontium.

Nockolds and Mitchell (16) also found an enrichment of strontium in potash-felspar relative to mica (biotite) in the Caledonian rocks they examined. The strontium contents of potash-felspars, together with other data on rubidium and radiogenic strontium, are given in table 7, where the specimen numbers correspond to the numbers assigned to the associated micas in table 5.

In each of these specimens of potash-felspar (with the exception of the aplite specimen) the concentration of total strontium is much greater than in the associated micas (table 5); the ratio % SrO in mica : % SrO in felspar varies from 1/10 to 1/50 (average 1/25).

Because of this marked increase in the amount of strontium in felspar compared with associated mica, the proportion of radiogenic strontium becomes extremely small and more or less negligible. Even if these rocks had been ancient, the proportion of radiogenic strontium would still be

TABLE 7. Amount of Rb and Sr in Scottish potash-felspars.

No.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
1.	0·04	0·011	0·2	0·000042	0·021 %
2.	0·04	0·011	0·1	0·000042	0·042
3.	0·06	0·016	0·2	0·000064	0·03
4.	0·04	0·011	0·2	0·000042	0·021
5.	0·2	0·054	0·005	0·00021	4·0*
6.	0·006	0·0016	0·2	0·0000064	0·003
7.	0·012	0·0032	0·2	0·000012	0·006
8.	0·06	0·016	0·1	0·000064	0·064
9.	0·06	0·016	0·1	0·000064	0·064
10.	0·04	0·011	0·2	0·000042	0·021
11.	0·06	0·016	0·08	0·000064	0·08
12.	0·02	0·0054	0·15	0·000021	0·021
13.	0·02	0·0054	0·1	0·000021	0·021
14.	0·08	0·021	0·2	0·000085	0·042

* Aplite.

insignificant. As already stated, the proportion of radiogenic strontium in the biotite specimens from these Caledonian rocks varies approximately from 1 to 5 %, whereas in the associated felspars (with the exception of the specimen of aplite) the proportion varies from 0·003 to 0·08 %, and thus the proportion of radiogenic strontium in the micas is about one hundred times greater than in felspar.

Mention has been made of the relative richness of rubidium in *hydrothermal* microcline from Varuträsk, Sweden, and since the phase of formation of this mineral is late, one should expect the amount of magmatic strontium to be very small, and hence the proportion of radiogenic strontium to be high. Hahn (3) mentions a mass spectrographic examination of strontium in a specimen of Varuträsk microcline which showed a very high proportion of radiogenic strontium. In this respect it is likely that ordinary hydrothermal microcline and amazonite are very similar; both are late hydrothermal and both contain appreciable amounts of rubidium and, probably in most cases, a high proportion of radiogenic strontium.

In potash-felspars other than the above two types the proportion of radiogenic strontium will probably almost always be very low. In plagioclases the proportion of radiogenic strontium will invariably be negligible.

RELATIVE PROPORTION OF RADIOGENIC STRONTIUM IN ROCKS.

Amongst rocks, only in granites and similar types relatively rich in potash and hence also in rubidium, is the proportion of radiogenic strontium in the total strontium sometimes likely to be appreciable. In more basic types of rocks magmatic strontium may increase considerably, particularly where much calcium is present, whereas the abundance of rubidium decreases sharply, and one may conclude therefore that even in the most ancient of the more basic rock varieties the proportion of radiogenic strontium will be negligibly small and of no significance.

Since the amount of total strontium varies very considerably in granites from different localities (about 0·001–0·3 % Sr) and because the amount of rubidium also tends to vary quite considerably (about 0·01–0·2 % Rb), the proportions of radiogenic strontium in various granites of like age will be very erratic. Hence it is more or less meaningless to assign an average strontium and rubidium value for granites and then calculate the proportion of radiogenic strontium in 'average granite' of different ages: more instructive information can be obtained by making calculations on the available data (although still meagre) for rubidium and strontium in granites and similar types, the ages of which are known approximately.

Caledonian rocks from Britain.—Reference has already been made to the rubidium and strontium contents of felspar and mica from Caledonian igneous rocks from Great Britain. In the rocks as a whole the proportion of radiogenic strontium is very small and more or less negligible.

Rapakivi-granite from Finland.—The analyses of several rapakivi-granites have been given by Sahama (20). For the calculation on the amounts of radiogenic strontium formed, an age of about 900×10^6 years has been used, and in this respect I was fortunate in being able to consult Dr. Kalervo Rankama, of the University of Helsinki, about the ages of several Finnish formations referred to in the text.

The data given in table 8 refer to Sahama's standard mixture of rapakivi-granites from eastern Fennoscandia and also to three specimens from the Salmi area.

TABLE 8. Amounts of Rb and Sr in granites, Finland.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radio-genic Sr.	Approx. prop. radio-genic Sr.
Standard mixture	0·15	0·04	0·009	0·0004	4·5%
Granite-porphyry boulder, Uomaa	0·10	0·027	0·0045	0·00027	6·0
Even-grained rapakivi-granite, 2 km. NW. of Koirinoja ...	0·09	0·025	$<0·003$	0·00025	$<9·0$
Ovoidic type, Pensanoja ...	0·11	0·03	0·007	0·0003	4·0

Sahama has also analysed some rapakivi-granites and mica-schist from Ihovaara hill, Suistamo, where these two formations are in contact. Analytical data for rapakivi-granite, mica-gneiss and also a mica-gneiss fragment in rapakivi-granite, and a contaminated specimen of rapakivi-granite, are given below (table 9). The age of the mica-gneiss has been taken to be about 1500×10^6 years.

TABLE 9. Amounts of Rb and Sr in granite and gneiss, Finland.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radio-genic Sr.	Approx. prop. radio-genic Sr.
Mica-gneiss, 1 km. E. of Ihovaara hill	0·07	0·018	0·035	0·00028	0·8%
Mica-gneiss from immediate vicinity of contact	0·06	0·016	0·027	0·00026	1·0
Mica-gneiss fragment in rapakivi-granite from contact ...	0·12	0·032	0·015	0·00052	3·4
Contaminated rapakivi-granite	0·16	0·044	0·006	0·00044	7·5
'Normal' rapakivi-granite ...	0·11	0·03	0·0035	0·0003	9·0

From these available analyses it seems that the proportion of radiogenic strontium in the rapakivi-granite is roughly constant (averages about 6–7 %) and the granite appears to contain a characteristically higher proportion of radiogenic strontium than the more ancient gneiss (average of two analyses 0·9 %).

Rocks from southern Finnish-Lapland.—Sahama (21) has published some interesting quantitative data on the abundance of the minor constituents in rocks from south Finnish-Lapland. All his specimens, several of which were granite types, were analysed for rubidium and strontium and since the approximate ages of several of the different formations may be assessed, the proportions of radiogenic strontium may be calculated in some instances. All the analyses for rubidium and strontium (total) in the Finnish rocks referred to in the next four tables have been taken from Sahama's paper.

(1) The ancient sedimentary gneisses of the Tuntsa-Savukoski-formation.—This formation is one of the most ancient in Scandinavia and although its age is not known accurately, it is probably about $1500\text{–}2000 \times 10^6$ years. Analytical data are given in table 10.

In each of these three specimens of ancient gneiss the proportion of radiogenic strontium is quite considerable (about 20 %).

(2) Ancient gneissic granites.—Analytical data on these rocks are given in table 11: an age of about 1500×10^6 years has been used for calculating the amounts of radiogenic strontium generated.

TABLE 10.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
Kyanite-mica-gneiss: Karhutunturi	0·08	0·022	0·0027	0·00048	20 %
Coarse mica-gneiss: Lyöpäköngäs	0·04	0·011	0·0009	0·00024	27
Gneiss: Ulmapaljakko	0·06	0·016	0·0027	0·00035	13

TABLE 11.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
Granite-gneiss: Pomovaaran ...	0·10	0·027	0·0027	0·00046	17 %
Granite-gneiss: Sarakkavaara	0·19	0·052	0·0027	0·00089	33
Granite-gneiss: western Sotatunturi ...	0·19	0·052	0·0054	0 ·00089	17
Granodiorite-gneiss: Kemijoen	0·03	0·008	0·027	0·00014	0·5
Granodiorite-gneiss: Värriöjoki	0·06	0·016	0·027	0·00027	1·0

Although a large number of specimens have not been analysed, it seems very probable that the granite types contain an appreciably higher proportion of radiogenic strontium (17–33 %) than the granodioritic types (0·5–1·0 %). This is only to be expected when one examines the abundance of some of the major constituents in the granite and granodiorite: the granite contains about three times more potash than the granodiorite, whereas the latter rock contains about three times as much calcium as the former. As is now well known, rubidium tends to be associated with potassium, whereas strontium tends to follow calcium in minerals.

(3) Hetta type of granite.—An age of about 1200×10^6 years for this granite has been assumed for calculating the data on radiogenic strontium given below in table 12.

TABLE 12.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
Granodiorite: Suasjärvi ...	0·05	0·014	0·09	0·00018	0·2 %
Gneissic granite: Piernakkaselkä	0·06	0·016	0·09	0·00021	0·2
Granite: Urakkaselkä	0·19	0·051	0·018	0·00065	3·5

In the granodiorite and gneissic granite a small proportion of the total strontium is radiogenic, whereas in the specimen of granite, the proportion of radiogenic strontium is about twenty times higher.

(4) Younger granites.—For calculating the data on radiogenic

strontium, given below in table 13, an age of about 900×10^6 years has been assumed for these granites.

TABLE 13.

Description.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
Granite: Untosenjärvi lake ...	0·19	0·051	0·018	0·0005	2·8 %
Granite: Korkeamaa	0·19	0·051	0·0027	0·0005	19
Granite: Eskelinen	0·10	0·027	0·018	0·00027	1·5
Coarse granite: Polvirova ...	0·10	0·027	0·0027	0·00027	10

The proportion of radiogenic strontium in these specimens varies considerably: in two of them the proportion is high, whereas in the other two specimens the proportion is appreciably lower, although by no means unusually low for granites.

In Sahama's paper the strontium and rubidium contents of various other rock types from south Finnish-Lapland are also given: in some of the rocks the proportion of radiogenic strontium is undoubtedly negligible (<0·001 % of total strontium), for example, in various basic rocks and also limestones, whereas in some formations the proportion of radiogenic strontium is very considerable. For example, strontium could not be detected in certain specimens of quartzite and should therefore be present in a concentration of less than 0·001 %; these quartzites contain on an average 0·05 % rubidium and assuming an approximate age of 1000×10^6 years, at least 15 % of the strontium is radiogenic.

Grängesberg iron-ore deposits, Sweden.—Landergren (22) has analysed several materials associated with the Grängesberg iron ores, and since his analyses include strontium and rubidium it is possible to calculate data on radiogenic strontium: an approximate age of 2000×10^6 years has been used for these calculations. Analytical data are given in table 14; the specimen numbers refer to Landergren's specimen numbers given in table 3 of his paper. Reference should be made to his paper for a more detailed account of the types of materials he analysed.

TABLE 14.

No.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
2	—	—	0·02	—	—
3	0·3	0·08	0·004	0·0018	45 %
4	<0·0003	<0·00008	0·01	<0·0000018	<0·018
5	0·02	0·0054	0·03	0·00012	0·4
6	0·09	0·024	0·02	0·00053	2·1
7	0·3	0·08	0·01	0·0018	18
8	0·1	0·027	0·001	0·0006	60

(*Table 14 continued*)

No.	% Rb.	% Rb^{87}.	% total Sr.	% radiogenic Sr.	Approx. prop. radiogenic Sr.
9	—	—	0·002	—	—
10	0·04	0·011	0·002	0·00024	12 %
11	—	—	0·005	—	—
12	0·0003	0·00008	0·002	0·0000018	0·1
13	0·0003	0·00008	0·004	0·0000018	0·05
14	0·1	0·027	0·004	0·0006	15
15	0·4	0·11	0·007	0·0024	35
16	0·2	0·054	0·01	0·0012	12
17	—	—	0·02	—	—
18	—	—	0·02	—	—
19	0·0009	0·00024	0·02	0·0000053	0·025
20	0·5	0·14	0·006	0·0031	52
21	0·3	0·08	0·01	0·0018	18
22	0·0009	0·00024	0·004	0·0000053	0·13
23	—	—	0·01	—	—
24	0·2	0·054	0·01	0·0012	12
25	0·009	0·0024	0·003	0·000053	1·8
26	<0·0003	<0·00008	0·0003	<0·0000018	<0·5
28	0·08	0·022	0·001	0·00048	48
29	0·009	0·0024	<0·0003	0·000053	>18
30	0·0009	0·00024	0·008	0·0000053	0·6
31	<0·0003	<0·00008	<0·0003	<0·0000018	—
32	—	—	<0·0003	—	—
33	<0·0003	<0·00008	<0·0003	<0·0000018	—
34	0·005	0·0014	0·001	0·00006	6·0
35	—	—	<0·002	—	—
36	—	—	0·003	—	—
39	0·006	0·0016	0·009	0·000035	0·6
40	—	—	0·0003	—	—
41	—	—	0·0007	—	—
42	0·14	0·038	0·001	0·00084	84
43	0·0009	0·00024	<0·0003	0·0000053	>1·8
44	—	—	<0·0003	—	—
45	—	—	<0·0003	—	—
46	—	—	0·0008	—	—
47	0·08	0·022	0·005	0·00048	9·6
48	0·004	0·0011	0·003	0·000024	0·8
49	0·2	0·054	0·001	0·0012	100
50	<0·0003	<0·00008	0·003	<0·0000018	<0·06
51	0·003	0·0008	0·002	0·000018	0·9
52	0·03	0·008	0·02	0·00018	0·9
53	0·009	0·0024	0·006	0·000053	0·9
54	0·04	0·011	0·001	0·00024	24 %
55	<0·0003	<0·00008	0·01	<0·0000018	<0·018
56	0·0009	0·00024	0·02	0·00000053	0·025
57	<0·0003	<0·00008	0·02	<0·0000018	<0·009
58	0·005	0·0014	0·03	0·00006	0·2
59	0·0009	0·00024	<0·0003	0·0000053	>1·8
60	0·25	0·068	<0·0003	0·0015	>100

Reference to the above table shows that the proportion of radiogenic strontium in the total strontium is extremely erratic and varies from negligible proportions to about 100 % radiogenic strontium. In this respect the above collection of minerals is indeed interesting and provides one with a good indication of how the proportion of radiogenic strontium varies from specimen to specimen: unfortunately for our purpose the author has not provided detailed descriptions of the minerals analysed.

THE Ca–Sr–Ba ASSOCIATION IN MINERALS.

The association of these three alkaline earths in minerals and rocks is frequently discussed by geologists; in particular Ca/Sr and Ba/Sr ratios in different minerals and rocks has elicited considerable interest. In these discussions all strontium is assumed to be magmatic, but because the proportion of radiogenic strontium may on occasion be appreciable and in a few instances may predominate, cognizance should therefore be taken of the rubidium content and age of each specimen in these discussions.

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