INTERNAL RADIOACTIVITY IN QUARTZ AND FELDSPAR GRAINS

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INTRODUCTION

When Fleming (1970) proposed the quartz inclusion dating technique for pottery, he assumed that quartz grains contained no internal radionuclides and thus could be regarded as non-radioactive dosemeters in a radioactive clay matrix. Later Sutton and Zimmerman (1976) found that quartz contains trace amounts of uranium and thorium and they estimated that the alpha dose contribution from internal sources could amount to 5% of the total dose.

When the alkali feldspar technique was proposed (Mejdahl and Winther-Nielsen, 1982), it was obvious that the beta dose contribution from inherent potassium had to be included and methods for estimating it were developed. We have now found that the dose contributions from inherent U, Th and Rb must also be taken into account and procedures for estimating these are discussed in the following.

POTASSIUM AND RUBIDIUM

Our mineral separation technique (Mejdahl 1985a) yields two fractions of alkali feldspars: K-feldspar (10-12% K) and Na-feldspar (5-6% K). The K content is determined by beta counting using a multicounter system (Bøtter-Jensen and Mejdahl, 1985) and the beta dose contribution from K-40 is calculated from absorption and converson data given by Mejdahl (1979) and Nambi and Aitken (1986).

The assessment of the beta dose contribution from Rb-87 in a clay matrix was considered by Warren (1978). On the basis of the close geochemical association of Rb with K he suggested the use of a K:Rb ratio of 200:1 when the Rb content has not been measured. In inclusion dating the beta dose contribution from external Rb is negligible because of the low beta energy (E_{av} = 0.104 MeV), but even a 0.1 mm grain will receive the full dose from internal Rb. The dose rate from 1% of Rb is 4.68 Gy/ka (Nambi and Aitken 1986).

We have measured the K and Rb content of 27 samples of feldspar extracted from pottery, burnt stones and sediments; Fig. 1 shows the Rb content as a function of the K content. A rather strong correlation of Rb with K is apparent with a K:Rb ratio of about 270:1. The regression line has the equation.

Rb (ppm) = -9.17 + 38.13 K (%)

For grains in the range 0.1 - 0.3 mm the above content ratio corresponds to a dose ratio of 3.4:1, that is, the contribution from Rb is about 30% of that from K.

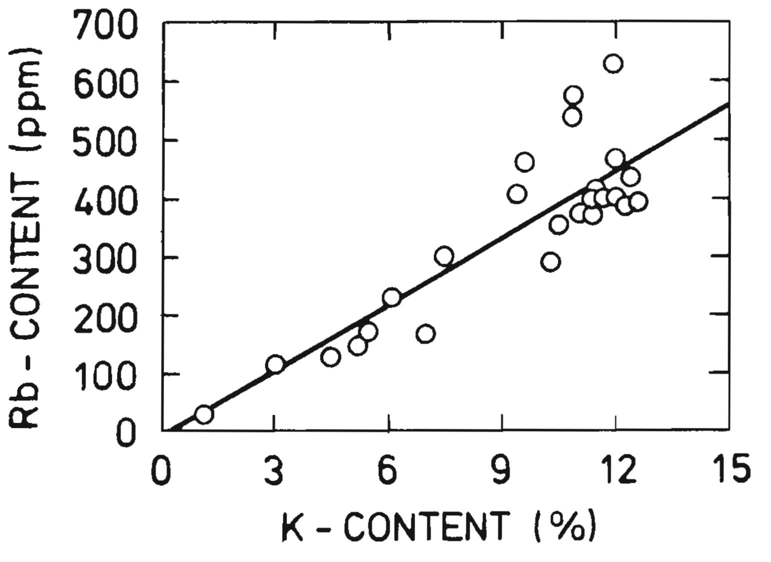

Figure 1. *Rb content as a function of K content for alkali feldspars from archaeological materials and sediments.*

Table 1: Measured (NAA) and estimated (eq. 1) Rb dose rates (D-r) for those points in Fig. 1 for which the deviation from the regression line gives rise to a total dose rate error exceeding 0.5%.

Sample No.	Mat'l	K	Rb Meas.	Rb Est.	Rb Diff.	Rb D- r	Tot. D- r	Tot. Error
R-840708, FN35	B	10.3	288	384	-96	0.045	5.31	0.8
R-831804,FN58	B	7.0	171	258	-86	0.040	5.76	0.7
R-832009,FK1-2	B	10.9	74	406	168	0.078	8.53	0.9
R-832105, FK13	S	11.4	372	426	-54	0.025	1.94	1.3
R-842201, FK35	C	9.6	459	357	102	0.047	5.34	0.9
R-854301, FK13	S	12.6	394	471	77	0.036	4.50	0.8
R-854409, FK13	B	11.9	628	445	183	0.085	9.34	0.9
UNITS		%	ppm	ppm	ppm	Gy/ka	Gy/ka	%

Notes.
Diff. is the difference between measured and estimated values.
Material (Mat'l); B = burnt stone, C = ceramics, S = sediment.

Because routine measurements of Rb would be difficult, it is desirable to utilize the correlation of Rb with K and it is therefore of interest to estimate the error ensuing from deviations from the regression line in Fig. 1.

Measured and estimated Rb values are given in Table 1 for those samples for which the deviation gives rise to a total dose rate error exceeding 0.5%. It can be seen that the total dose rate error is smaller than 1% in all cases except that of sample R-832105 which consists of sand from a frost wedge cast (Kolstrup and Mejdahl 1986). The error that would have resulted from neglecting the Rb contribution varies from 1 to 4% for all samples except R-832105 for which it is 9%. A total dose rate as low as 1.33 Gy/ka (1.48 Gy/ka including the Rb contribution) was encountered for cover sand from Jutland (Mejdahl 1985b).

The largest difference in Table 1 (168 ppm) together with this dose rate would result in a total error of 5% while the error resulting from neglecting the Rb contribution would be 20%.

The conclusion from the results presented here is that the contribution from internal Rb in alkali feldspar grains must be taken into account and can be calculated with sufficient accuracy from equation (1).

URANIUM AND THORIUM

The U content of all grain fractions dated in our laboratory is measured routinely by means of the delayed neutron counting technique (Kunzendorf et al., 1980). Results for a number of quartz and feldspar samples are given in Fig.2. The U content was below 0.5 ppm for most of the samples, but in a few of them values above 1 ppm were found.

The Th content of about 30 samples was measured by means of neutron activation analysis. Because it is not feasible to measure Th routinely, the relation of Th to U was studied. A marked difference between sediments and heated archaeological samples was seen. The results for sediments are discussed first.

The Th measurements included ten sediment samples, one quartz and nine K-feldspar. The U and Th contents for these were less than 0.4 and 1 ppm respectively. Fig. 3 shows a plot of Th vs U. A linear regression line with the equation.

Th (ppm) = 0.004 + 2.36 U (ppm).

was fitted to the points. From the equation, the Th:U ratio is about 2.36:1, corresponding to an alpha dose-rate ratio of 0.63:1 using conversion factors from Nambi and Aitken (1986). The beta dose rates are small compared with those from alpha particles; they are considered in a later section.

Because the points in Fig. 2 scatter considerably, it is important to estimate the errors resulting from deviations from the regression line. Resulting dose-rate errors for those points that deviate most are given in Table 2. In calculating the effective alpha dose to feldspar an a-value of 0.2 was assumed, based on results for three sediment samples measured by Ann Wintle (personal communication).

The total error resulting from the scatter of the points in Fig. 2 is smaller than 1% except for fsample no. R-832105 for which it is 11.2%. The error that would have resulted from neglecting the alpha dose contributions from U and Th is about 10%. It is obvious, therefore, that these contributions cannot be neglected for coarse-grained feldspars from sediments with a low external dose rate.

The U and Th contents in grains from archaeological samples are considerably higher than those in sediment samples. A plot of Th vs. U for ten samples is shown in Fig. 3. Linear regression for these points yields the equation:

Th (ppm) = 0.826 + 0.783 U (ppm)

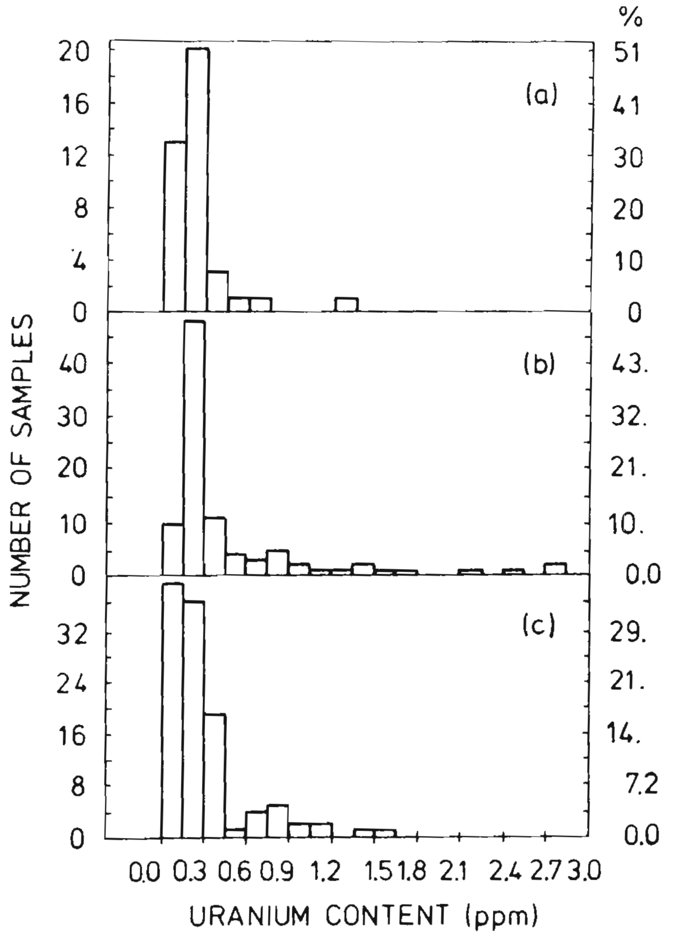

Figure 2 Uranium content in quartz and feldspar samples. a) quartz, 39 samples, b) sodium feldspar, 93 samples, and c) potassium feldspar, 110 samples.

Compared with sediments the Th content relative to U is smaller in the archaeological samples and there is no proportionality. The reason might be that the grains have been enriched preferentially in U during firing of the materials. Not included in Fig. 3 are three samples that gave somewhat abnormal results; these are listed in Table 3. The samples in table 3 are burnt stones from an area in Sweden just north of Uppsala.

The errors that would result from using the regression line in Fig. 3 are listed in Table 4 for the three samples in Table 3 and those in Fig. 3 that show the largest deviation from the regression line. It can be seen that the errors are negligible except for the three samples from Table 3. Two of these give a total error of 3..5% which is just acceptable within the general uncertainty of about 6%. The errors that would result from neglecting the Th contribution are 5 and 7% in the two cases and about 2% for the other samples.
The question of zoning remains to be considered. Because U and Th tend to accumulate in cracks in the grains, the disdtribution is likely to be inhomogeneous. We have not yet carried out any zoning studies, but Sutton and Zimmerman (1978) found in their study of 1 mm quartz grains from a granite fireplace rock that most of the radioactivity has agglomerated in large areas (~50 µm) that had low luminescence efficiencies. In such cases the alpha dose rate will be overestimated. Another effect resembling that of zoning in individual grains arises from the occurrence of red-coloured grains appear to contain more U and Th and have less

luminescence efficiency than lighter grains and, consequently, the alpha dose rate will be over-estimated in a sample containing a mixture of light and red grains.

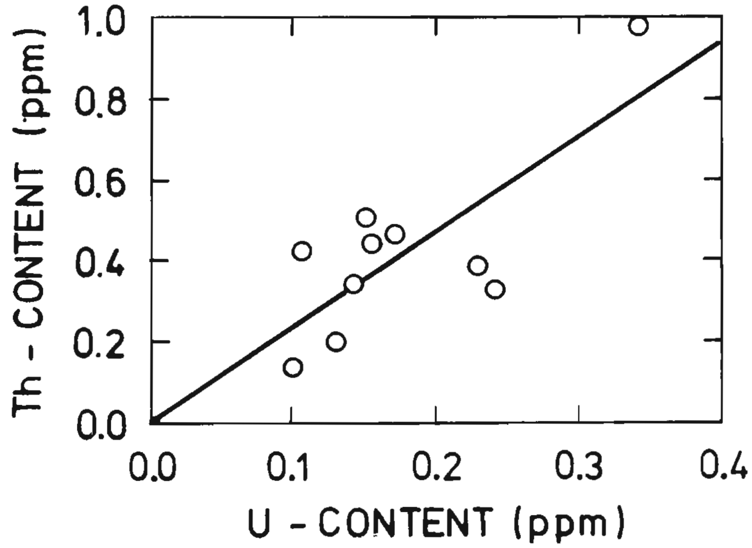

Figure 3. Th content as a function of U content for quartz and feldspar from sediments.

Table 2.: Errors in Th alpha dose rate and total dose rate for those points in Fig. 3 that deviate most from the regression line

Sample No.	Meas. (NAA)	Est. (eq.2)	Diff. error	Th D-r	Tot. D-r	Tot. error
R-841604	0.38	0.55	-0.16	0.024	3.98	0.6
R-841605	0.98	0.81	0.17	0.025	3.92	0.6
R-841608	0.42	0.25	0.17	0.025	4.63	0.5
R-832105	0.51	0.36	0.15	0.022	1.90	1.2
R-832401	0.33	0.57	-0.24	0.035	5.04	0.7
UNITS	ppm	ppm	ppm	Gy/ka	Gy/ka	%

Notes.
The samples listed are potassium feldspar from sediments. An alpha efficiency factor of 0.2 was assumed. Conversion factor from Nambi and Aitken (1986). Diff. is the difference between measured and estimated value

Table 3: U and Th concentration for three samples of burnt stones not included in Fig. 4.

Sample No	U (ppm)	Th (ppm)
R-853604, K1-2	1.13	0.98
R-853612,FK1-2	0.83	4.77
R-853632, FN58	0.40	2.41

Table 4.: Errors in Th alpha dose rate and total dose rate for the samples in Table 3 and those points in Fig. 3 that deviate most from the regression line.

Sample No.	Th Content Meas. (NAA)	Th Content Est. (eq.3)	Diff. error	Th D-r	Tot. D- r	Tot. Error
R-840713, FN58	0.74	0.98	-0.24	0.04	5.33	0.7
R-831804, FN58	0.98	1.27	-0.29	0.04	5.76	0.7
R-832009, FK1-2	1.30	1.06	0.23	0.03	8.53	0.4
R-853604, FK1-2	0.98	1.71	-0.73	0.11	8.38	1.3
R-853612, FK1-2	4.77	1.48	3.29	0.49	13.94	3.5
R-853622, FN58	2.41	1.14	1.27	0.19	5.30	3.5
R-834410, FN35	0.86	1.09	-0.24	0.04	6.11	0.6
R-854406, FK1-2	1.33	1.04	0.29	0.04	8.76	0.5
UNITS	ppm	ppm	Gy/ka	Gy/ka	%	%

Notes.
All samples are alkali feldspar from burnt stones. An alpha efficiency factor of 0.2 was assumed and the conversion factor was taken from Nambi and Aitken (1986). Diff. is the difference between measured and estimated values.

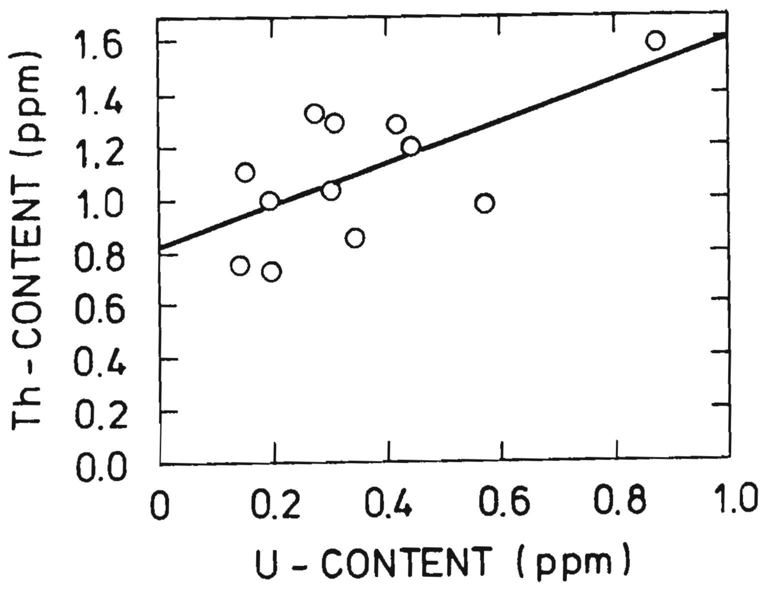

Figure 4. The content as a function of U content for feldspars from archaeological materials.

BETA DOSE FROM U AND TH IN QUARTZ AND FELDSPAR GRAINS

The beta dose rate from U and Th in quartz and feldspar grains can be estimated from the absorbed fraction values listed by Mejdahl (1979). The dose rates that would result from 1 ppm of U and Th for various grain sizes are listed in Table 5. It is clear from Fig. 2 that the U content is less than 1 ppm for most samples. From eqs. (2) and (3) the Th content corresponding to 1 ppm U is 2.4 ppm or less. Since the total dose rate is usually at least 1 Gy/ka, the contribution from beta radiation emitted by internal U and Th will be be negligible in nearly all cases. In those cases where higher internal contents of U and Th were found, the total dose rate was also correspondingly higher.

Table 5: Beta dose rates for 1 ppm internal concentration of U and Th in quartz and feldspar grains.

Grain diameter (mm)	Dose rate (Gy/ka) U	Th
0.1	0.014	0.004
0.3	0.028	0.007
0.5	0.039	0.009
1.0	0.059	0.013
5.0	0.114	0.023

Notes.
Absorbed fractions taken from Mejdahl (1979).

The beta particles from U and Th will be counted along with those from K in our technique for estimating the K content of feldspar grains (Bøtter-Jensen and Mejdahl 1985). In order to investigate the interference of the beta particles from U and Th we measured the apparent K content of the U and Th standards NBL 105 (10 ± 1 ppm U) and NBL 109 (104 ± 3 ppm Th + 3.7 ppm U). We found that 1 ppm U and 1 ppm Th would correspond to apparent K contents of 0.15% and 0.04%, respectively. The corresponding infinite-matrix dose rates calculated from these apparent K-contents are 0.122 Gy/ka and 0.033 Gy/ka which correspond rather well to the values 0.147 Gy/ka and 0.029 listed in Nambi and Aitken (1986). However, because the absorbed fractions for K are considerably smaller than those for U and Th, one would not estimate the beta dose from U and Th to smaller grains correctly by counting the beta particles along with those from K.

The result of the counting of beta particles from U and Th with those from K in the K determination will be the inclusion of 25-80% of the beta dose from U and Th, depending on the grain size. This can be regarded as an advantage but one of little practical eonsequence because, as mentioned above, the beta dose contribution from internal U and Th is negligible.

CONCLUSION

Our studies of the Rb content of feldspar grains and U and Th contents of quartz and feldspar grains have shown that the dose rate contribution from these sources is significant and must be taken into account.
Rb appears to be rather well corelated with K, and the contribution can be included with sufficient accuracy by assuming a K:Rb ratio of 270.1 corresponding to a dose rate ratio of 3.4:1.
The U content of all our samples is measured routinely by delayed neutron

counting, but routine measurements of the Th content is not feasible. We therefore studied the relation of Th to U. For sediments approximate proportionality was found with a Th:U ratio of 2.36:1. For heated archaeological samples there was a linear relation, but not proportionality. The conclusion is that these relations allow determination of Th with sufficient accuracy from the known content of U. In all cases studied the beta dose contribution from U and Th was negligible.

It should be emphasized that the results are preliminary. NAA studies of a larger number of samples are planned in order to provide a broader base for the conclusions.

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PI . Reviewer's Comments(S R Sutton)

These important measurements on internal radioactivity in quartz and feldspar represent primarily good news with regard to practical dating. The Rb contribution in alkali feldspars appears to be the greatest contribution, up to 20% of the total dose-rate, however, the geochemical coherence of K and Rb allows the internal Rb dose-rate to be predicted with sufficient accuracy from the K content alone. Alpha doses from internal U and Th tend to be lesser contributions, up to 10%. Consequently, the approach of using a typical Th/U ratio to estimate the Th content of these grains from their U content, as suggested by Mejdahl, is adequate. U and Th beta doses were negligible in the samples studied. These results suggest that internal dose-rates in quartz and feldspar can be estimated with sufficient accuracy from their K and U contents.

It is difficult to predict the universal nature of these conclusions, since the samples studied were presumably restricted to Scandanavian materials. Individual samples from other geologic provenances may have greater internal dose-rate contributions, and, therefore, greater total dose-rate errors, than those reported here. Ceramics may be highly variable in this regard since the inclusions and the clay typically derive from different geologic sources, ie quartz and feldspar with relatively high radioactivity can be incorporated in low radioactivity clay and vice versa.

Author's Response:

It is true that all samples were from Scandanavia, and, therefore, the results are not directly applicable in other parts of the world without further studies. However, I should think that the mineral proerties governing the uptake of uranium and thorium would be more or less the same everywhere.