Thick source alpha counting: the measurement of thorium

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Introduction

From time to time, in papers from our laboratory, we have discussed the technique of thick source alpha particle counting (TSAC) for determining uranium and thorium concentrations and hence dose rates, more particularly as it relates to its use in luminescence and ESR dating (e.g. Jensen and Prescott 1983, Akber et al 1983, Woithe and Prescott 1995 (hereinafter, Woithe and Prescott without the date).

In the last-mentioned of these references we discussed the question of the efficiency of the zinc sulphide screens used in the method. In summary, we presented evidence that the effective efficiency is not only less than 100% but varies a percent or two from batch to batch of screens. A typical efficiency was shown to be between 85% and 90% for the screens supplied commercially by W.B. Johnson. This work was undertaken because of a widely expressed belief that the efficiency is 100%.

At the time of publication of Woithe and Prescott, the reviewer, Huntley, commented that the paper made a case that, "all is not well" and made a number of suggestions about methodology, all of which had already been incorporated into the project. Huntley also remarked that, in his own use of TSAC (Huntley et al, 1986) he finds efficiencies of close to 100% for standards, although thorium seems to be underestimated, and he applies a correction for this. As mentioned in Woithe and Prescott, the late John Hutton had also derived an empirical correction for underestimation of thorium.

TSAC : the method

It will be useful briefly to recall the process of calibrating TSAC with a zinc sulphide screen and photomultiplier, as originally set out by Aitken (1985 Appendix J). Using a certified U or Th standard in known geometry, the count rate is measured as a function of discriminator setting. The discriminator setting for subsequent practical use is set at 85% of the extrapolated counting rate for Th and 82% for U. With this setting, the count rate for the standard should correspond to that predicted by Aitken's calculations or corresponding calculations by others. If the observed count rate is less than that predicted, it is concluded that the efficiency of the zinc sulphide screen is less than 100% and its value is determined. This was the procedure in Woithe and Prescott.

Our contention remains that, when calibration is carried out with standard sources according to the original prescription of Aitken (1985) but using updated conversion factors, with finely ground samples (Jensen and Prescott, 1983) an efficiency of less than 100% is found. Woithe and Prescott used the conversion factors of Huntley et al (1986) and found efficiencies between 85% and 90%.

The most recent calculation relating concentrations of U and Th with count rates for a particular geometry is that of Adamiec and Aitken (1998). Their factors are smaller than those of Huntley et al (1986) by about 6%. Since these conversion factors are used in calibrating the counting system and finding the efficiency of the screens, the effect of applying the Adamiec and Aitken conversion factors would be to increase the measured efficiency of the screens by 6%. In our case, all would now be now better than 90%.

In fact, from the point of view of calculating dose-rates for luminescence dating, the conversion factors are not critical since they are used in both the calibration and the subsequent alpha counting of samples. Provided that a corresponding screen efficiency is used, the measurement is essentially a comparison with the standard. This is not true, of course, if the efficiency is wrongly assumed to be 100%. There is a further caveat for efficiencies less than 100%: The pairs counting technique requires the detection of two alpha particles, which follow each other in the thorium decay chain. Consequently a

correction for both requires the efficiency correction factor to be squared.

Methodology of the measurement of thorium

We have now returned to the problem from a different angle, concentrating on a comparison of two independent measurements of thorium concentration viz, TSAC and neutron activation analysis (NAA). We show that our deduction of an efficiency of less than 100% is sustained. Further, incorporation of delayed neutron activation (DNA) measurements of uranium improves the TSAC estimate of thorium.

In the standard usage of TSAC, the total combined count rate, for thorium with its daughters and uranium with it daughters, is recorded. In addition "slow pairs" record successive alpha decays of ^{220}Rn and its daughter ^{216}Po in the thorium decay chain and hence give a measure of the concentration of thorium. Since the rate of slow pairs is commonly about 3% of the total alpha count rate, the statistical accuracy is necessarily low for counting times of a day or two. Nevertheless, in calculating dose-rates from alpha-counting data, some measure of the ratio of uranium to thorium is better than no estimate at all, or a guess.

Of course, for luminescence and ESR dating, the important quantity is the dose-rate, which is derived from the U, Th and K concentrations together with cosmic ray intensity. For a given total alpha particle count, the dose-rate is very insensitive to the relative amounts of Th and U (Sasidharan et al 1978; Aitken 1990).

In the present paper, we compare the thorium concentrations obtained by TSAC with those obtained by NAA. In the first instance we have chosen to stay with our original (Huntley et al 1986) conversion factors.

Results

"Raw" Pairs Counting

Samples were selected, more or less at random, from our data set to cover the range of thorium concentrations from less than 1 $\mu g.g^{-1}$ to about 30 $\mu g.g^{-1}$. Initially, all samples were field samples, collected in the course of dating assignments from a variety of sites in Australia, Europe, Thailand and China. All samples had been analysed by TSAC, DNA and NAA. A few analyses were repeated. With exceptions noted below, samples known or suspected to be in radioactive disequilibrium were excluded. Such samples have been thought to be rare in our experience. However, interestingly, when all the data were assembled, five samples were found to give inconsistent uranium analyses. Reanalysis confirmed previously unsuspected disequilibrium and these samples were excluded. It might be argued that disequilibrium is more common than usually supposed.

Concentrations of thorium larger than 15 $\mu g.g^{-1}$ are rare in our sample collection. However, three widely-separated sites in Western Australia gave unusually large Th concentrations. We were initially doubtful of using these because the associated uranium analyses showed disequilibrium. However, they were considered satisfactory for the present purpose of finding the thorium concentration from pairs in the thorium chain, since these are quite independent of the U-chain and disequilibrium in the Th chain is unlikely because the lifetimes of the daughters are short on a geochemical time scale necessary to transport them in the environment. However, to fill in the upper range of concentrations, we made up artificial substandards of 15, 20, 25 and 30 $\mu g.g^{-1}$ by diluting aliquots of the New Brunswick thorium Standard NBL108 (520 $\mu g.g^{-1}$ Th) with a sediment sample measured to have 0.32 $\mu g.g^{-1}$ Th). Since NBL108 also contains a small amount of uranium, these substandards contain between 0.7 and 1.2 $\mu g.g^{-1}$ U; this is of no consequence.

Figure 1 shows a comparison of Th analyses by NAA and TSAC, the former being taken as the independent variable. In this comparison, the TSAC Th concentration is determined only by the rate of slow pairs.

The continuous straight line in the figure is an unweighted least squares fit; the equation of the fitted line and the correlation coefficient are shown. The screen efficiency used was that appropriate to each individual sample and ranged between 85 and 89%. The fitted slope is 1.03 ± 0.04 which is not significantly different from one. We consider that the agreement between NAA and TSAC is good. The dashed line corresponds to the TSAC values that would be obtained if the efficiency were assumed to be 100%. To obtain this line, the original data were reprocessed on the assumption of 100% efficiency and the data refitted. (To avoid a cluttered diagram, the data points have been omitted). The slope is 0.76 ± 0.03, which corresponds to an average efficiency of 87%. We claim that fig. 1 supports our contention that the screen efficiency is less than 100%.

"Adjusted" thorium concentrations

The estimate of thorium concentration from TSAC can be improved by making an independent measurement of uranium, say by DNA. The expected U count rate is then calculated from this value, subtracted from the total count rate and the Th concentration is then calculated from the remainder. For the reasons given earlier, this is not very important for dosimetry but it is an improvement, and is useful for comparison of TSAC with Th concentrations measured in other ways, e.g. NAA or XRS.

Figure 2 shows such a comparison of Th by TSAC/DNA with Th by NAA. The figure was constructed in the same way as fig.1. The fitted slope is 1.05 ± 0.03. This, too, is not statistically different from 1. Comparison of figs 1 and 2 shows that the correlation coefficient is increased, from 0.95 to 0.97, although both of these show that the fits are excellent. The uncertainty in the slope is reduced for the data in fig. 2.

This comparison is essentially independent of the previous one and makes no use of the pairs count. The effect of this procedure is not only to give an improved value for an individual thorium concentration but also to reduce its experimental error. This is because the estimate of thorium is now based on two data sets that are independent of the pairs count and of much greater precision. We recall that the Th concentration based on pairs is based on a relatively small number of pairs and the statistical uncertainty is commonly of the order of 15%.

It is necessary to comment on the fact that the number of data points differs in the two figures, viz: in fig. 2, the high thorium samples from Western Australia do not appear. This is because the uranium chains are in disequilibrium for these samples and DNA analysis for U cannot be used.

It may be remarked that the main reason for our introducing DNA measurements of uranium is to provide a simple and rapid check for radioactive disequilibrium. In this way we compare the concentration of the parents with the uranium concentration inferred from the alpha count rate. Since the latter is the sum of alphas from all parts of the decay chain, a discrepancy between DNA and inferred uranium usually indicates loss or gain of members of the decay chain. The thorium chain is much less likely to be in disequilibrium. Nevertheless, an independent measurement of thorium is sometimes useful, as in the present contribution.

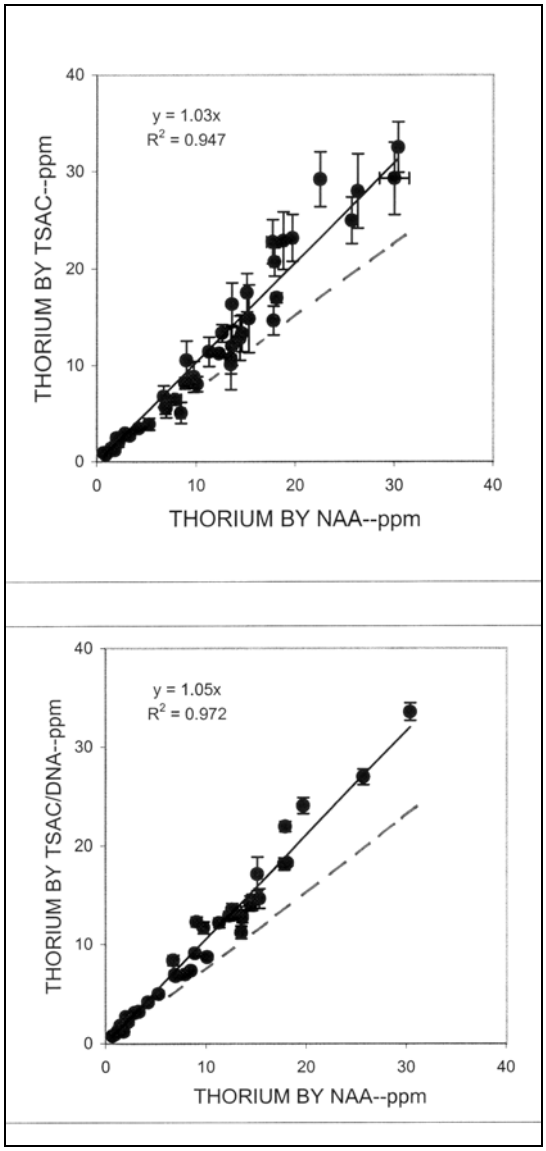

Figure 1. (upper)
Comparison of thorium concentrations found by thick source alpha counting by pairs (TSAC) and neutron activation analysis (NAA). The fitted and dashed lines are discussed in the text.

Figure 2. *(lower)*
Comparison of thorium concentrations found by thick source alpha particle counting combined with delayed neutron analysis (TSAC/DNA) and neutron activation analysis (NAA). The fitted and dashed lines are discussed in the text.

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Reviewer

D.J. Huntley

Comments

It would be much appreciated if someone would calculate new conversion factors from U and Th contents to TSAC rates, using the latest alpha particle ranges of Ziegler. The software for calculating the ranges can be downloaded from Ziegler's web site: http:/www.SRIM.org/