

Moisture correction for annual gamma dose

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Introduction

The annual gamma dose in a moist medium is commonly taken as

$$D_w = \frac{D_d}{(1 + xWF)} \quad (1)$$

where,

D_d = dose in the same burial medium when dry,

W = saturation water content expressed as (wt. water / dry wt.),

F = fraction of pore space occupied by water, and x is the ratio of effective mass absorption coefficients for water and burial medium.

Equation (1) is essentially what was proposed by Zimmerman (1971); he used $x = 1.14$ on the basis that for gamma rays of around 1 MeV this was the ratio of the coefficients for water and aluminium, the latter having absorption properties fairly close to those for soil. Subsequently Bowman (1976) suggested that a value for x of 1.0 ± 0.1 should be used; this was on the basis that the ratio is less than unity below about 0.2 MeV (see fig.1) and on an assumption that 30% of the energy is carried by gamma rays below 0.2 MeV.

With the availability (Jain et al., 1979) of spectra (primary plus secondary) for wet and dry soil-like media a further assessment of the effect can be attempted. Whereas Zimmerman and Bowman obtained values for x from the ratio of mass energy-absorption coefficients, it is now possible to base evaluation on the lowered flux and modified spectrum of gamma rays that is present in the moist medium (compared to the same medium when dry). The basic computations performed are evaluations, for various spectra, of

$$\frac{D_w}{D_d} = \frac{\sum(\mu/\rho)E(\Delta n)_w}{\sum(\mu/\rho)E(\Delta n)_d} \quad (2)$$

where

(μ/ρ) = mass energy-absorption coefficient of quartz for gamma rays of energy E ,

$(\Delta n)_w$ and $(\Delta n)_d$ = numbers of gamma photons in 10 keV energy intervals centred on E for the wet and dry cases respectively,

and where the summations are carried out from 15 keV to the maximum energy of the spectra concerned (and multiplied by $(1 - P)$ - see later).

For soil having 'typical' relative concentrations of K, Th and U the mean value of x derived from equations (1) and (2) is 1.065. However, as will be discussed, this is not the whole story, neither for sediment and pottery, nor for compact samples having dimensions greater than a few millimetres such as flint, calcite and tooth enamel.

In the case of sediment and pottery it is necessary to consider the intermediate stage that intervenes before traps are filled. This stage consists of an effectively uniform flux of electrons (from Compton and photoelectric interactions) steadily degrading in energy; it is uniform because most of the primary electrons have ranges greater than the particle size and the size of the water-filled pores. Hence rather than the relative values of gamma absorption coefficients it is the relative values of electron stopping powers that determine the partition of energy between solid and water. Consideration of stopping powers indicates an effective value for x that is not sufficiently different to the value of 1.14 mentioned above for a change to be suggested. It is noted incidentally that the value of 1.25 proposed by Zimmerman in respect of annual beta dose appears to be about 5% too high, though more comprehensive calculations are required before it is justifiable to recommend a change.

The uniform electron flux argument does not apply in the case of the compact samples because the sample size is comparable with, or greater than, the electron range. On the other hand because the gamma spectra of Jain et al. will be distorted by the sample further investigations are needed in this case too before accepting 1.065 as the final answer.

Hence the net message of the paper is 'no change' for the moment. The justification for submitting for publication is that it may save others in luminescence and ESR dating the effort of going through the same steps and possibly interest someone in the more comprehensive calculations indicated. Of course even if the value of 1.065 was substituted for 1.14, it is only in exceptional circumstances (high water content combined with dominance of gamma dose) that there would be any significant effect on date.

An incidental point from the Jain et al spectra that may be mentioned is that the 0.2 MeV limit corresponds to substantially less than 30% of the energy carried - see Murray (1981) or p.330 of Aitken (1985). Figure 2 shows an example of a flux spectrum; in terms of energy carried the region below 0.5 MeV becomes much less important.

The spectra

A photon deposits its energy in matter through several possible interactions, the major processes in the energy range of interest for dating being the Compton effect and the photoelectric effect. It is the equilibrium spectra that are relevant and Jain et al. have calculated these by numerical solution of transport equations for sandstone and shale, for various degrees of wetness. For shale the chemical composition (see table 1) is similar to that for a typical soil (Murray, 1981).

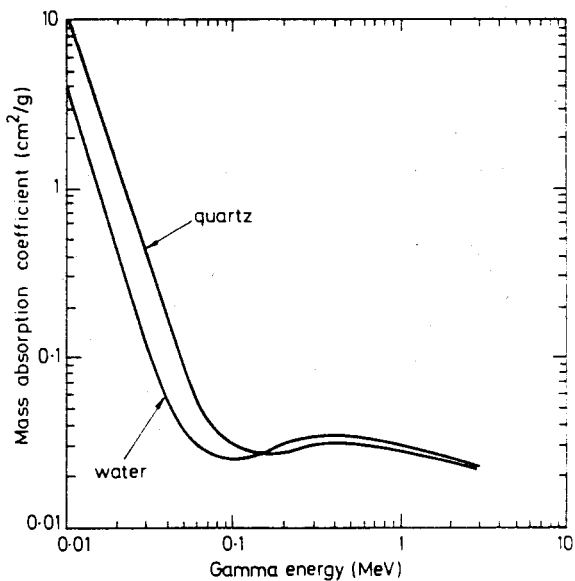


Figure 1. Mass energy-absorption coefficients (μ/p) for water and quartz (derived from Storm and Israel, 1970).

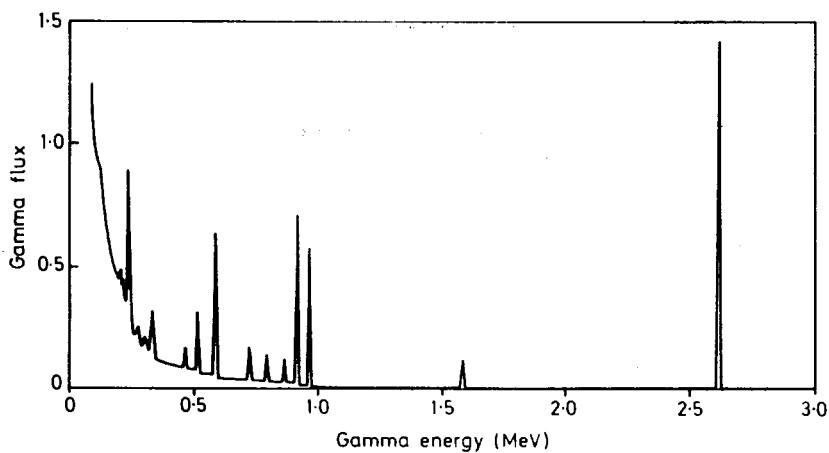


Figure 2. Gamma flux spectrum for 0.094 ppm of Th in shale of zero porosity. Units of vertical scale: photons/cm²/s/0.01 MeV interval.

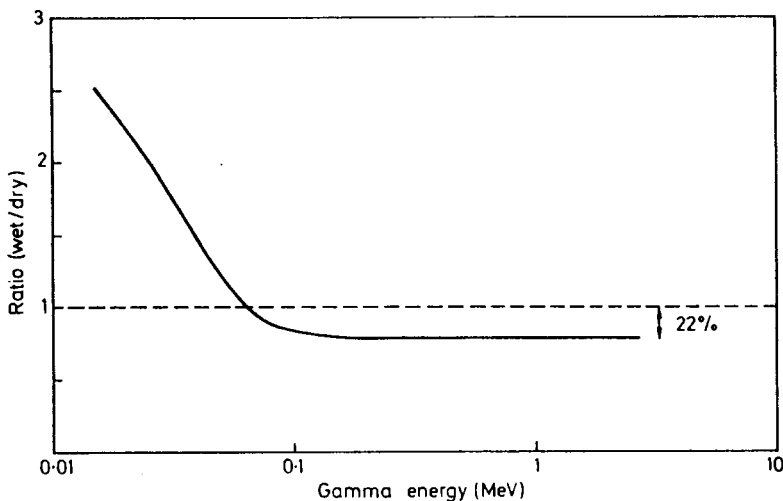


Figure 3. Ratio of gamma flux from Th in wet shale to that in dry shale. Water content: 0.255 g of water per 1 g of shale ($P = 0.4, S = 1.0$).

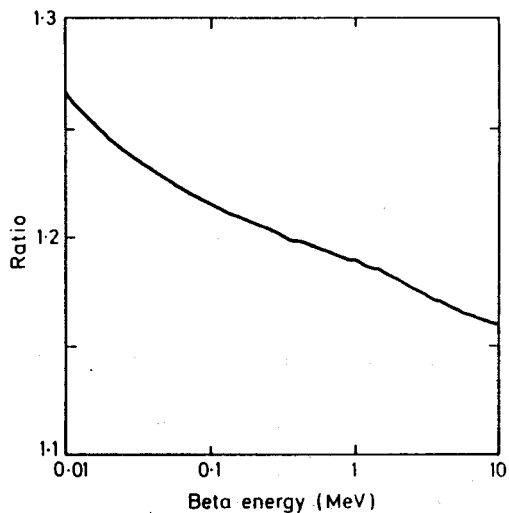


Figure 4. Electron collision stopping power ratio between water and quartz derived from Berger and Seltzer, 1982).

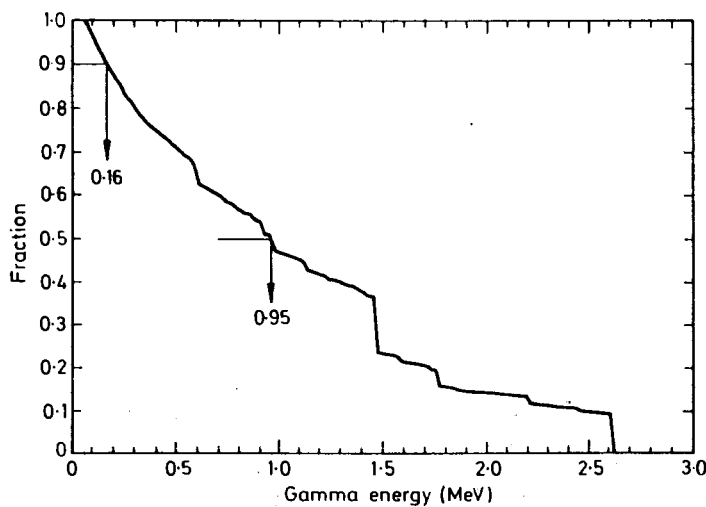


Figure 5. Fraction of total absorbed energy (in quartz) obtained from gamma rays above a given energy, for a composite spectrum from 1% of K, 10 ppm of Th and 3 ppm of U, in dry shale.

site spectrum from 1% of K, 10 ppM of Th and 3 ppM of U, in dry shale. The data provided by Jain et al. are in the form of tables giving the gamma flux as gamma rays/cm²/s/10-keV interval. The spectra are for 'unit sources', namely 1 photon/cm³/s, which according to Jain et al. corresponds to 302.1, 0.2463 and 0.0813 mg/cm³ for K, natural Th and natural U respectively; the primary gamma rays and intensities used by Jain et al. are listed on p.329 of Aitken (1985). Spectra are given for various values of porosity, P, and pore space saturation, S, ranging from P = 0 to P = 0.4 and from S = 0 to S = 1. Both P and S are 'volume fractions' and whereas F in equation (1) equals S, the saturation wetness that has been defined above is given by

$$W = \frac{P}{\rho(1 - P)} \quad (3)$$

where ρ equals 2.6117 g/cm³ for shale and 2.6263 for sandstone, and the factor, x, is given by

$$x = \frac{\{(D_d/D_w) - 1\}}{WF} \quad (4)$$

In evaluating the ratio (D_d/D_w) it is necessary to normalize to the same concentrations (of K, Th and U) per unit dry weight; this is done by multiplying values obtained from the spectra by (1 - P).

Table 1. Composition (as % by weight) of sandstone and shale (Jain et al., 1979) and 'typical soil' (Murray, 1981).

Element	Sandstone	Shale	Soil
H	-	1.340	1.0
C	1.394	1.551	-
O	52.165	49.776	52.6
Mg	0.716	1.501}	
Al	2.551	8.290}	8.3
Si	37.086	27.760	28.2
K	1.112	2.738	1.5
Ca	3.979	2.259	2.5
Fe	0.998	4.786	4.6
Na	-	-	1.3

As illustrated in fig. 3, relative to a dry medium the gamma flux in a wet medium is depressed in the high energy region but enhanced in the low energy region. Presumably the enhancement is associated with the low photoelectric crosssection of water relative to medium.

Although it does not affect the value obtained for x it may be noted that the concentration values quoted above from Jain et al. correspond to 1 chain disintegration /cm³/s in the cases of Th and U, rather than 1 photon/cubic cm³/s.

Evaluation of x

Table 2 gives values of x that have been computed for various cases. The mass energy-absorption coefficients for quartz have been derived from Storm and Israel (1970) and those for other minerals of dating interest are similar. For 'typical soil' containing 1% of K, 10 ppM of Th and 3 ppM of U the weighted average for x

of U, in dry shale.

is 1.065, with the individual values (for shale) lying between 1.06 and 1.08.

In the first two lines of table 2 results are given for two cases of zero wetness but, whereas the first is for P = 0 the second is for P = 0.3. The agreement confirms that for a given radioisotope concentration per unit weight the annual dose is independent of bulk density; the comparison also provides a check of computational accuracy.

Table 2. Evaluation of x

P	S	WF	Sum	D _d /D _w	x
<i>Potassium (in shale)</i>					
0	0	0	19.91	-	-
0	0	0	19.91	-	-
0.3	0.5	0.0821	18.29	1.088	1.074
0.4	1.0	0.2553	15.60	1.276	1.081
<i>Potassium (in sandstone)</i>					
0	0	0	20.12	-	-
0.3	1.0	0.1632	17.04	1.181	1.109
<i>Thorium (in shale)</i>					
0	0	0	11.53	-	-
0.4	1.0	0.2553	9.07	1.271	1.061
<i>Uranium (in shale)</i>					
0	0	0	10.38	-	-
0.4	1.0	0.2553	8.17	1.271	1.061

Notes

1. WF and x are calculated according to equations (3) and (4).
2. The values under Sum are computed as indicated in equation (2). Units are MeV/g/s per unit source.

Discussion

While the value for x just derived would seem to be better based than from the coefficient ratio approach there are two further aspects to be considered. In the cases of flint, calcite and tooth enamel there is the question of distortion of the gamma spectra due to the finite sizes of these intrusive samples. Probably the effect is not serious but before accepting the value of 1.065 as final, further consideration is needed.

In the cases of pottery and sediment there is a more serious bar to acceptance of the new value. The energy absorbed from the gamma flux is carried by electrons with an energy spectrum extending from zero up to the maximum energy of 2.6 MeV (from Tl-208 in the Th chain) and hence with a spread of ranges up to several millimetres. The water-filled volumes in pottery and sediment are of sub-millimetre dimensions and consequently for a substantial portion of the spectrum 'charged particle equilibrium (CPE)' obtains, i.e. the electron flux is uniform throughout water and solid. Thus in considering the partition of ionization between water and solid, for this part of the spectrum it is the ratio of electron stopping powers that are relevant. For the annual beta dose Zimmerman (1970, 1971) proposed the use of x = 1.25, this being the average ratio of the stopping powers of water and aluminium from 0.1 - 3.0 MeV, based on the tables of Berger and Seltzer (1964). Because the stopping powers of quartz and feldspar are a few percent higher than that of aluminium this is an

overestimate; a direct computation of the water/quartz ratio (see fig.4), using Berger and Seltzer (1982), gives an average value of 1.19. Only the collision component has been taken into account (i.e. radiation loss has been excluded); the slight effect on the ratio of the density correction has been ignored.

Some indication of the fraction of the secondary electrons for which there is CPE can be obtained from fig.5. This gives, for a composite gamma spectrum from K, Th and U, the energy absorbed from the gamma flux expressed as a fraction of the total energy absorbed; thus we see that 90% of the absorbed energy is obtained from gamma photons having an energy in excess of 0.16 MeV. When absorption is by the photoelectric effect the electron has the same energy as the photon, but in the case of Compton interaction there is a spectrum of electron energies from zero up to near the energy of the photon (within 30% for 0.5 MeV photons and within a lesser percentage for higher energy photons). Inspection of fig. 5.1 of Evans (1955, p.693) indicates that this spectrum does not vary strongly with energy and hence as far as annual dose is concerned it will be weighted towards the high energy end.

Hence the major part of the annual gamma dose is delivered by electrons having an energy in excess of 0.16 MeV; the range corresponding to this energy is about 0.1 mm and hence there is CPE for these electrons -- the median grain size in loess is around 0.04 mm and the pore size in pottery somewhat less. For this part of the gamma dose the appropriate value of x is 1.19 rather than 1.065.

Conclusion

To establish a precise value would require Monte Carlo type computations and these are hardly justified bearing in mind the relative unimportance of the gamma dose as far as pottery and sediment are concerned. It is therefore recommended that the original Zimmerman value of 1.14, which is midway between the two values just mentioned, should be retained for pottery and sediment. On the other hand for compact samples having dimensions greater than a few millimetres the appropriate value is likely to be towards 1.065.

As far as the annual beta dose itself is concerned it would be logical to change to 1.19. However since averaging should take account of the beta spectra involved, as well as CPE effects, any definitive change from the Zimmerman value of 1.25 should await comprehensive treatment. A more important uncertainty needing attention is the question of whether or not it is appropriate to make any correction at all in the case of alpha dose (see Fleming and Stoneham, 1973; also, p.76 of Aitken, 1985). However the present authors do not have intentions of attempting either task, nor that posed by compact samples.

Acknowledgements

We are appreciative of the Reviewer's thoroughness and thank him for various suggestions and corrections which we have incorporated, as well as for the useful comments which follow. We thank Mohan Francis of UK NRPB for kindly providing extracts from Berger and Seltzer (1982). JX would like to thank the Xian Laboratory for Loess and Quaternary Geology, Academia Sinica, for partial support.

References

- Aitken, M. J. (1985) *Thermoluminescence dating*. Academic Press, London.
- Berger, M. J. and Seltzer, S. M. (1982) Stopping power of electrons and positrons (2nd edition), *NBSIR 82-2550-A*, National Bureau of Standards, Washington DC 20234.
- Bowman, S. G. E. (1976) *Thermoluminescence dating: the evaluation of radiation dosage*. Unpublished D.Phil. thesis, Oxford University.
- Evans, R. D. (1955) *The Atomic Nucleus*. McGraw-Hill, New York, Toronto and London.
- Fleming, S. J. and Stoneham, D. (1973) The subtraction technique of thermoluminescent dating. *Archaeometry* 15, 229-238.
- Jain, M., Evans, M. L., Close, D.A. (1979) Non-destructive assay technology for uranium source evaluation: infinite medium calculations, *Los Alamos report LA-7713-MS/UC-51*, University of California. Available from: NTIS, US Dept. of Commerce, 5285 Port Royal Road, Springfield, VA 22161.
- Murray, A. S. (1981) *Environmental radioactivity studies relevant to thermoluminescence dating*. Unpublished D.Phil. thesis, Oxford University.
- Storm, E. and Israel, H. I. (1970) Photon cross sections from 1 keV to 100 MeV for elements 1 through 100, *Nuclear Data Tables A7*, 565-681.
- Zimmerman, D. W. (1971) Thermoluminescent dating using fine grains from pottery, *Archaeometry* 13, 29-52.

PI Reviewers Comments (John P. Prescott)

It is good that someone has got round to carrying out the non-trivial task reported in this paper. As the authors point out, the analysis is incomplete and there are still issues to be resolved. There is an element of judgement in the recommendations. On the whole I agree with them and my comments are primarily directed to supplementary points.

Presumably flint, calcite and tooth enamel have internal radioactivity which is low with respect to typical soil; the gamma contribution may well dominate in such cases and the effect of the improved moisture correction will be significant. However, one could argue that the dose to a compact sample having the dimensions greater than a few millimetres would not include contributions from low energy photons from outside the sample and that a higher figure (say 1.14 for the sake of argument) would be better after all. Perhaps I have missed something. In the case of pottery and sediment, where the distribution of radioelements is more homogeneous, the gamma ray calculations are certainly valid even when the electron stopping power argument is subsequently accepted.

Although I do not believe it would make very much difference to the final conclusions, I believe that the discussion section understates the contribution of low energy electrons. As the paper points out, the Compton electrons carry away only a fraction of the photon energy; about 50% at 250 keV, 40% at 160 keV (the figure mentioned in the discussion) and a decreasing fraction at lower energies. Similarly, although the photoelectric effect eventually results in the release of the full photon energy, the energy of the photoelectron itself is smaller than the photon energy by the binding energy of the shell from which it comes. The remaining energy is subsequently deposited by secondary photoelectrons and Auger electrons. In short, the energy from photons is actually deposited by a number of electrons of significantly smaller energy.