

Review comments on 'Attenuation of beta radiation in granular matrices: implications for trapped-charge dating' by Cunningham et al.

General comments

This is a very interesting manuscript, undoubtedly worth publishing. I really enjoyed reading it (although I had to scratch my head a few times...). Having myself worked on the issue at stake here, I can only congratulate the authors for such a great work. Whereas the issues at stake are very similar to those investigated by Guérin et al. (2012), this manuscript makes a great step forward – especially with sections 2 and 6.

Sections 2 (formalising the issues at stake with equations) and 6 (on the relationship between grain size and radioelement concentration) indeed are, in my view, especially instructive.

I have more issues with part of section 4 (in particular Fig. 3, geometry C, comparison between BEM and Φ approach) and with section 5, on the effect of moisture. In the first case, there are unexplained differences between the present work and published evidence (Guérin et al., 2012); in the latter case, not only are there discrepancies (with the same reference), but I also believe I have good arguments against the authors' (and their results).

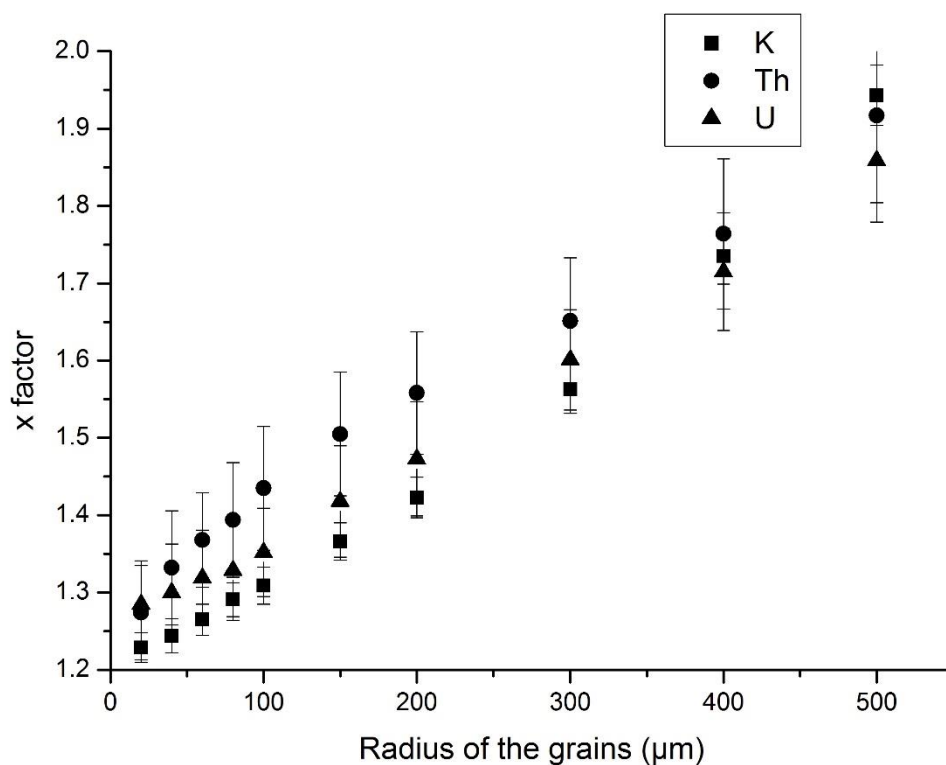
On the first issue, let us first look at Fig. 3, Geometry C, surface emission: there seems to be a disagreement between the results presented here and those obtained by Guérin et al. (2012: Fig. 3) when radioelements are located on the surface. Indeed Guérin et al., in the caption of their Fig. 3, wrote: 'In the 180-250 μm range, the classical approach, based on attenuation factors, underestimates dose rates to quartz by: 15% for K (a), 25-30% for U (b) and 40-45% for Th (c).' The grain size distribution of the intercomparison sample (Murray et al., 2015, modelled by Guérin et al., 2012) is quite similar, at face value, to the grain size distribution of Geometry C, used to generate Fig. 3 of the present paper. I am quite surprised that the obtained results differ by such an amount – I cannot find a good explanation. While Guérin et al. defined the 'surface' (emission volume) as the outer 2 μm , the authors use only 1 μm – but I would be surprised that this issue matters much. The beta spectra are very similar (and even identical for U and Th) in both studies. Perhaps the authors could at least mention the comparison – and at best (try to) explain where the discrepancy comes from? Since both models (the 2012 using Geant4 and the present one using MCNP) agree for source grains, and since both also agree with the BEM developed here – also for the source grains – then where does the problem come from?

On the second problem (effect of moisture), first I would like to bring the attention to an article not considered by the authors, although I think it is relevant. I. 283-284: 'Guérin et al. (2012) have questioned the accuracy of x for granular sediment, noting that variations in pore size and moisture content may lead to a different value for x' '. Please see also, on the effect of water on gamma (not beta) dose rates: Guérin, G., Mercier, N., 2012. Preliminary insight into dose deposition processes in sedimentary media on a grain scale: Monte Carlo modelling of the effect of water on gamma dose-rates. *Radiation Measurements*, 47, 541-547. (more on that below)

I. 292-294: 'Consider a sediment that is mostly water, in which grains lie further apart than the maximum range of beta radiation. In this case, water and quartz have the same effect on the dose rate to the grains, because all radiation is absorbed by the pore medium, and hence $x = 1$ '. I disagree with this statement: if water absorbs all the energy, then the dose rate to quartz is 0, we can't conclude anything on the value of x . (I would even argue that x probably tends to the infinite – water absorbs it all, not only 'more than its fair share'). It seems clear to me that, since radiation first interacts with water and then quartz (because quartz is inert, so radiation must cross water in pore space before entering quartz, at least in the grain source case. NB: the spherical geometry, in the surface emission

case, implies that >50% of the radiation will first cross water and then quartz), it follows that water will absorb more dose rate when grains and pores are of comparable range to the range of beta particles, than when the medium is homogeneous at the beta scale. Let us consider a mono-energetic spectrum of low E electrons (e.g. 10 keV): most, if not all of these electrons will never enter quartz, water will absorb everything. Since x represents ‘how much more dose rate water receives compared to quartz’, it must be greater than 1.2. In other words, 1.2 has to be the minimum (rather than maximum) value. In the light of this reasoning, I am very surprised by the results shown in Fig. 4.

Note: here is an unpublished graph dating back to Geant4 simulations that I performed when working on the effect of water on gamma dose rates (Guérin and Mercier 2012). The geometry was much simpler (only one grain size, all grains arranged in a crystallographic lattice) but the x factor increased with increasing grain size (as I expected following the arguments given above).



This figure is – on top of the arguments spelled out above – why I get really surprised by Fig. 4 of the present manuscript. It seems to me that one of our teams must have made a calculation error somewhere. It can of course be me, but if so I would like to understand why... and have convincing arguments.

Specific comments

I. 36-37: ‘This formulation implicitly assumes that the contribution to the IM dose rate from the internal activity of any one dosimeter grains is negligible, which is usually true.’ Well, for K-feldspar the large

number of grains in a given size class leads to double counting (perhaps a word could be phrased here?).

I. 38-39: 'Calculation of c_{atten} derives from the self-dose values of the dosimeter grains (Bell, 1979; Mejdahl, 1979; Aitken, 1985)'. I would suggest adding Guérin et al. (2012) in the list, as this is where latest self-dose values were published.

I. 44: 'The key assumption in the use of $1 - \varphi$ is that the matrix surrounding the grain is homogenous'. I would suggest adding 'at the scale of natural beta radiation, and that it consequently absorbs more than the IM dose rate'. Note that Guérin et al. (2012) precisely discussed this phenomenon (see their Fig. 3 and their section 2.2): if part of the sediment absorbs less than the IM dose rate, then another part of the sediment must absorb more than the IM dose rate (at least when the mass distribution is approximately even).

I. 52-53: 'In such cases, it is not clear that the assumption of a homogenous matrix is reasonable (Guérin et al., 2012)'. I would move the 'not' as follows: 'In such cases, it is clear that the assumption of a homogenous matrix is *not* reasonable (Guérin et al., 2012).'

I. 84-85: 'The larger the size of the external grains, the stronger the effect.' I would replace 'external' by 'inert' or a similar adjective (these grains are part of the sediment – see also my comment below on the use of the words 'matrix' and 'sediment').

I. 90: 'Although $1 - \varphi$ has been defined above as the inverse of the self dose'. Unless I am mistaken, in mathematics this is not the inverse, but the complement.

Eqs. (3) and (4): I think I see your point, but (it took me some time and) I find the notation φ_{matrix} rather confusing, especially when you write above: 'normalised to the mass-weighted average $1 - \varphi$ for the bulk matrix'. I guess what you mean is that the sum of all $(1 - \Phi) \cdot m \cdot corr \cdot IM \text{ dose rate} / \text{sum of mass} = IM \text{ dose rate}$. In other words, this equation ensures that no energy is lost. If so, in your reasoning you don't calculate a Phi value that is characteristic of the matrix, but a $(1 - \Phi)$ characteristic of the sediment (see my comment below on your use of the terms 'matrix' and 'sediment'). Perhaps having the subscript after brackets (i.e. writing $(1 - \Phi)_{matrix}$) would be less misleading? An alternative would be to develop the full calculation; even although it is just a normalisation issue, I think it is important to get the message across... Eventually, if I am not mistaken your approach is equivalent to calculating a weighted mean Phi value. In my view, all this needs to be made clearer. Another solution would be to start from the self-dose fraction absorbed by source grains (Eq. 4); at least for me it is easier and more natural to sum absorbed doses than attenuation factors...

I. 101-102: 'Only the portion of energy that leaves the source grains ($= 1 - \varphi_{source}$) is available to dosimeters, and all of that energy must be absorbed by the matrix as a whole'. I think you are using the word 'matrix' to describe different things; sometimes it is all of the sediment surrounding a grain (as is the case here), sometimes it denotes a homogeneous medium surrounding 'large' grains (as in Fig. 1). Please check (to avoid confusion, I would suggest giving your definition/use of these terms somewhere in the article).

I. 117-118: 'With θ written out in full in Eq. 4, it is apparent that if $\varphi_{source} w = \varphi_{sediment} w$ then the equation reduces to $cw = 1 - \varphi_{dosimeter} w$ – i.e. the standard definition for a grain in a homogenous matrix'. This is exactly the conclusion reached by Guérin et al. (2012) – see their Fig. 4 and associated discussion on the case of the intercomparison sample (Murray et al., 2015), for which K-feldspar grains have the same size distribution as the bulk sediment.

Sections 2 and 3: For better readability, I would consider putting section 3 (where the concepts are familiar to luminescence dating practitioners) before section 2, which is more challenging. It would make the difficulty more gradual for the reader.

I. 238-240: 'In the case of whole-grain sources, the attenuation estimated by $1 - \phi_{dosimeter}$ and the BEM are identical, and correspond closely to the Monte Carlo simulation (Fig. 3a and 3b)'. Guérin et al. (2012: Fig. 4) obtained the same result and gave the following interpretation: un-accounting for self-absorption in the source grains, together with a wrong estimation of the attenuation parameter, leads to $1 - \phi_{dosimeter}$ being correct (in another words, the two errors arising from the infinite matrix approach compensate for each other). I think it would be worth telling the reader (and perhaps refer to Fig. 4 of Guérin et al., 2012?).

Caption of Fig. 3: 'The effective source grain-size distribution for surface sources is indicated in the grain-size distribution.' I assume this 'effective source grain-size distribution' is shown as empty bars of the histograms? In any case, this should be spelled out.