Enhancement of absorbed dose from natural background gamma radiation due to photoelectron induction in Uranium Particles.
With some comments on Pattison et al 2009
and the Royal Society

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Part I Questionable Science

1. The Background

The health effect of depleted uranium exposure is an area of scientific and political controversy. Despite a great deal of evidence that uranium particles from weapons show anomalous genotoxicity there has been no persuasive explanation of the biological mechanisms involved (ECRR2010). The issue had been considered by a committee set up by the Royal Society (RS) in 2000; the Chair of this committee was Prof. Brian Spratt. The outcome of its deliberations was published in 2001 (Royal Society 2001). It was concluded that it was impossible for the exposures to Depleted Uranium (DU) weapons to be the cause of any illness since the absorbed doses were too low. These conclusions were not accepted by all (e.g Busby 2000, 2003, Bertell 2006). The Ministry of Defence set up an independent board to oversee measurements of uranium in Gulf War veterans and to discuss the science: the Depleted Uranium Oversight Board (DUOB). Around the same time, evidence that internal radiation exposures (including DU) might be significantly more hazardous than current risk models predicted led the then Environment Minister, Michael Meacher, to set up the Committee Examining Radiation Risks from Internal Emitters, (CERRIE) chaired by Prof Dudley Goodhead. Goodhead had been the chief advisor to the RS on radiological issues of DU. Both the CERRIE and DUOB resulted in failures to agree among committee members and separate oppositional reports (www.duob.org; CERRIE 2004a , CERRIE 2004b).

The suggestion that internalised uranium particles and molecular uranium might represent a radiological hazard due to a hitherto overlooked photoelectron enhancement of natural background gamma radiation was first made by Busby in 2003 at the CERRIE international conference in St Catherines College Oxford in 2003 (Busby 2003) although this never was reported by those who assembled the CERRIE majority report. A presentation was also made in 2004 to the DUOB (Busby 2004); Goodhead and Spratt were both present at this presentation. The idea was then published in two papers in 2005 (Busby 2005, Busby 2005a).

The idea was then supported by work by Hainfeld in the USA on the photoelectron enhancement of X-rays by gold particles (Hainfeld 2004), which resulted in a patent for cancer therapy. Two papers on the issue were sent by Busby in 2007 to the Royal Society journal *Philosophical Transactions* B. The papers were immediately *unsubmitted* by the editor. Following some discussion, the papers were sent to a new RS journal, *Interface* edited by Prof. William Bonfield. The papers were set to three referees. These people all recommended publication, with slight amendments. Bonfield rejected the papers against the advice of his referees. His explanation was that there was no space in the journal. Despite complaints he refused to publish the papers or to discuss the issue.

The idea was next presented to an international conference in Braunschweig, Germany and appeared in the proceedings of that conference in early 2008 (Busby and Schnug 2008). It was picked up by the *New Scientist* and became the main news story in September 2008.

The wide publicity that this gave to a potential explanation for the health effects of uranium, and the political arguments that ensued, forced the regulators to respond. The International Commission on Radiological Protection (ICRP) whose risk model was being questioned, agreed to put several scientists on the issue. To date, no response has appeared from the ICRP, except that their Scientific Secretary Dr Jack

Valentin resigned in early 2009. Shortly after his resignation he agreed (on video camera at a meeting in Sweden) that there were significant uncertainties in the ICRP model. He admitted that the model could not be used to predict health effects in exposed populations as the uncertainties for some internal exposures were up to two orders of magnitude (see [http://www.llrc.org/health/subtopic/icrpabdicates.htm]). The UK Health Protection Agency (HPA), formerly the National Radiological Protection Board, made an attempt to downplay the size of the effect in a paper written in response to questions from the Low Level Radiation Campaign as part of a stakeholder response to the proposal to implement to 2007 ICRP model in the UK. Unfortunately, this response contained both mathematical and conceptual errors which were easy to demonstrate (Busby 2009). At this point the HPA refused to continue with the dialogue, stating that they would publish a response in the peer-review literature. To date no paper from HPA or HPA personnel has appeared.

In 2008/9 Monte Carlo mathematical modelling of photoelectron enhancements of gamma radiation was carried out at the University of Ulster. Preliminary results were presented in Madrid (Elsaesser et al 2007), at the 2009 conference of the European Committee on Radiation Risk in Lesvos in 2009 (Elsaesser et al 2009a) and at the Royal Society of Chemistry conference on nanoparticles in Liverpool in 2009 (Elsaesser et al 2009b). The models which compared water, gold and uranium nanoparticles largely supported what had been predicted, namely that due to self absorption, the enhancement was dependent on particle size with a cut off of about 1micron diameter for Uranium. The magnitude of the enhancement was also roughly that predicted by Busby 2005 from theory, in proportion to the fourth power of the atomic number of the absorber.

However, in September 2009 a paper was published in the *Journal of the Royal Society Interface,* editor William Bonfield. This paper, main author an Australian, John Pattison (Pattison et al 2009), claimed to address the issue of the health effects of DU particles and photoelectron enhancements. It presented results of Monte Carlo mathematical simulations of photoelectron enhancement of natural background radiation on cylindrical particles of 10micron diameter and 2 micron diameter. The conclusions of the paper ended with the statement:

We found that although the dose enhancement is significant, of the order of 1-10, it is considerably less than that suggested previously.

But the authors clearly had a political agenda. In an interview with the media shortly after publication Pattison stated:

Our aim in this study has been to help by continuing the process of elimination and, in doing so; we believe that we can in fact rule out DU as a cause of the [Gulf War] *Syndrome from a radiation perspective. Our research found that the enhancement factor is actually of the order of 1 to 10 which, although significant, is at least 50 times smaller than has been suggested in the past.*

The issue is one with major economic, military and political implications.

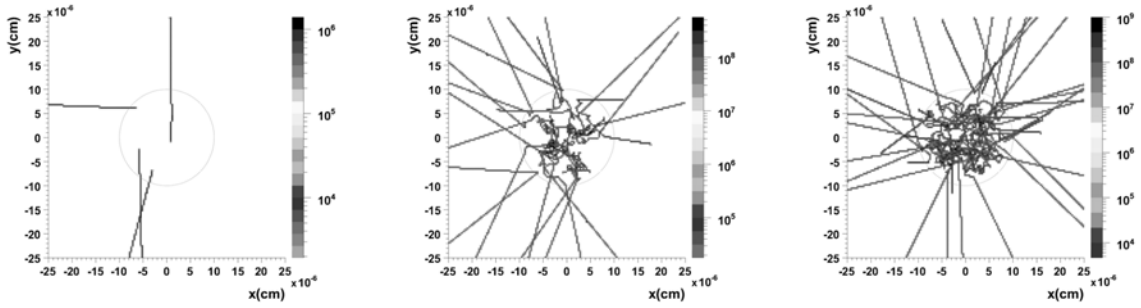
2. The idea

The idea is simple and based upon well-accepted physics. Gamma radiation and X-rays are absorbed by matter in proportion to the fourth or fifth power of the atomic number Z. It follows that high Z elements like gold (Z=79), lead (Z=82) and uranium (Z=92) stop gamma rays from natural background radiation (or gamma or X-rays from any source) more effectively than water, the main component of the human body. The energy of more than 90% of the gamma ray photons in Natural Background

Radiation is converted into energetic electrons, photoelectrons. These are indistinguishable from beta radiation and cause the same effects, namely ionisation and genetic damage. The effect is important for particles of uranium, which has the highest atomic number of any naturally occurring element. However, because uranium also absorbs photoelectrons very strongly, it is only the smaller particles below about 1 micrometer diameter which generate photoelectrons that can escape into the surrounding tissue. Larger particles and pieces of shrapnel have much less effect since all the photoelectrons induced within the body of the larger particle can not escape. In addition, for Uranium there is another serious hazard. Uranium binds chemically to DNA since the UO_2^{++} ion, the soluble form of uranium, has enormous chemical affinity for the DNA phosphate. Whilst bound to DNA, photoelectrons from the uranium attack the DNA directly. Thus there is a biologically plausible mechanism which explains the embarrassingly large number of observations of anomalous genotoxicity from uranium exposures (see Busby and Schnug 2008 for references).

The induction of photoelectrons in water, gold and uranium particles is illustrated by the result of the Monte Carlo simulation shown in Fig 1 (from Elsaesser et al 2009). These FLUKA code results show that the generation of photoelectrons at 100keV is approximately in proportion to a fourth power atomic number law as predicted by conventional physics (Krane 1988).

Fig 1. Photoelectron production and energy deposition by 1000 100keV photons in 10nm nanoparticles of (left to right) water, gold and uranium. Monte Carlo simulation using the FLUKA code. Note that the water particle has 100,000 incident photons (Elsaesser et al 2009). These are projections on a flat xy plane.



3. Pattison et al's paper

Pattison et al 2009 wrote a paper which was clearly aimed at marginalising the issue of the health effects of DU as its primary intention. It included statements which are plainly incorrect, for example, stating that 'hot particles' can be obtained by the weathering of igneous rocks: this is clearly impossible. More important, the authors obtained results which were in error for a number of different reasons. These included:

- The natural background radiation spectrum used as a basis for the model appeared to exclude low energy photons below 50keV: these contribute to short range photoelectrons which dominate the enhancement effect.
- The particles modelled were 10μ and 2μ diameter and were thus significantly larger than those produced by DU impact.

- The particles were modelled as cylinders and not spheres, thus introducing a self absorption error.
- The larger particles have lower photoelectron enhancement than smaller ones below 1μ since photoelectrons induced in the bulk metal do not emerge. This fact was denied as a result of the following:
- The dose enhancements were compared for both sizes of particle through the energy absorbed in tissue within a fixed distance from the particle, a 5μ radial distance from the particle surface. This approach introduced a major conceptual error in the result and led to a conclusion that was plainly silly, namely that the larger particle showed the larger enhancement.
- There are large differences between the results of the Monte Carlo analysis and prior reports of photoelectron enhancements involving measurements rather than mathematical models.

3.1 Natural Background radiation dispersion

If the uranium particles are too small for self-absorption (below 1μ diameter) the magnitude of the photoelectron enhancement is simply a function of the number of photoelectrons generated and their range i.e. the volume into which their energy is dissipated. The number of photoelectrons generated at any energy E is purely a linear function of the number of photons of energy E absorbed by the uranium (less binding energies and assuming minimal self absorption). The number of photoelectrons (and thus absorbed dose) in any volume element at distance D from the particle is a function of the photoelectron energy (inverse square law of electron range) i.e. the more energetic electrons have large ranges but are relatively few. The more frequent low energy photoelectrons have shorter range and are absorbed into less volume, giving a higher dose. It is clear that the enhancement will increase rapidly close to the metal particle and fall off rapidly with distance. This is (a) because there are more low energy photons in natural background radiation and (b) because the further away from the metal particle, the larger the volume into which the electrons deposit their energy. In addition to this, for particles larger than 500nm diameter, higher energy photons which penetrate the particle produce photoelectrons which lose energy on their emission path through the highly absorbing uranium as discussed in Busby 2005. Thus pivotal to an analysis of photoelectron amplification is starting any calculation with the correct number of photons of different energy contained in NBR.
Pattison et al 2009 base their calculations on the photon energy dispersion given in the gamma spectrum of NBR which is copied in Fig 2 below from their paper.

Fig 2. Spectra of Natural Background radiation employed by Pattison et al as photon dispersion input for Monte Carlo simulation. Copy is stretched laterally.

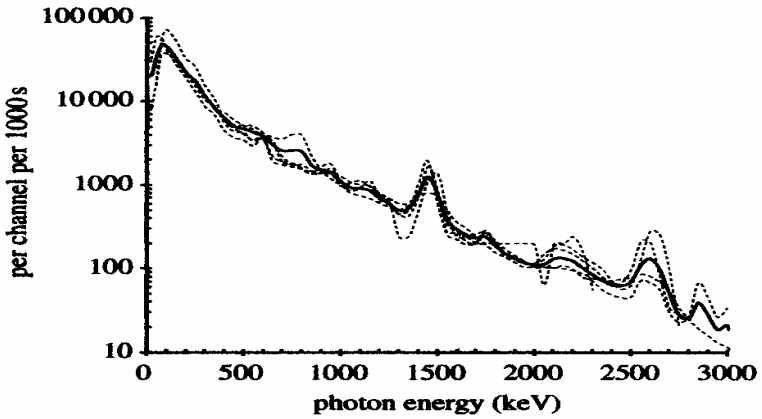


Figure 2. Measured pulse-height spectra using 76 mm × 76 mm cylindrical Na(Tl) crystal with minimum shielding, 256 (12 keV) channels, and 1000 s counting time for five different locations in North America and Europe shown as faint dashed line curves. The weighted-average curve is shown as a bold continuous line curve.

Two things are immediately apparent. First, and of general interest, it is clear that most of the energy is below 150keV, the extreme left of the graph. This is the area of interest: the higher energy photon spectra are second order and could be usefully ignored. Second, note the rapid fall-off of energy as the graph turns over at about 120keV. In reality, there is no such fall off of energy at the low energy end. What is displayed is an artefact of the measurement technique. Low energy photons are excluded from the detector by shielding and other technical problems with excitation and absorption. Even so, the peak at low energy shown in Pattison et al's Fig 2 is to the right of where it normally is found in a gamma spectrum of natural background radiation. Fig 3 shows a spectrum, obtained with a 2-inch (Scionix) Sodium Iodide detector on the beach at Burnham on Sea. Note the position of the low energy peak which is at 70keV compared with a measured 120kev in the Pattison spectrum in Fig 2. However both spectra are artefacts of the detector and cannot be used to model NBR photons in the low energy range because these low energy photons are absorbed by the metal shielding which packages and protects the NaI crystal. It can be estimated that in the region below 100kev Pattison will have excluded from his model a significant number of the photons which comprise natural background. About 60% of all photons in NBR are below 150keV. It is difficult to assess the extent of this problem. Fig 4 shows the approximate dispersion of energy of photoelectrons produced by Natural Background radiation (NBR) gamma photons and is based upon calculations which allow for the absorption by shielding and for absorption by the human body modelled as water. On passage though the body (modelled as water) the low energy photons increase in number relative to the high energy photons. This is due to photon energy loss, scattering and other processes. Fig 5 shows the significant enhancement factor by energy in the region below 300keV.

Fig 3. Gamma ray spectrum obtained on beach at Burnham on Sea using a 2-inch NaI (Tl) Scionix detector (Busby 2005). Note rollover at about 60keV.

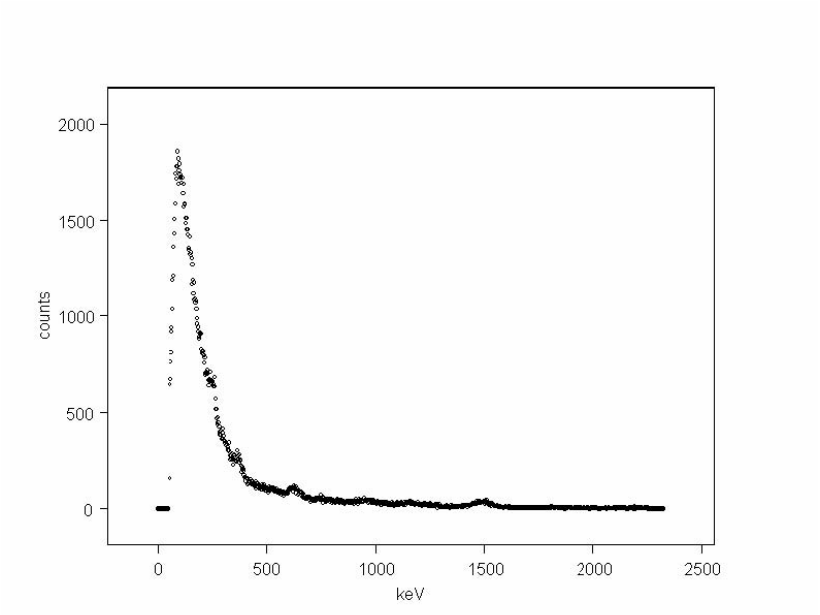


Fig 4 Energy dispersion in the low energy region 0-500keV of the natural background gamma photons at 15cm depth inside a human body. Based on Pattison et al 2009 Fig 3 and unpublished work (Busby 2008) using a gamma probe packed with bags of water. Shielding effects on the primary in-air dispersion below 100keV are uncertain and the energy dispersion of photons inside the body is very uncertain.

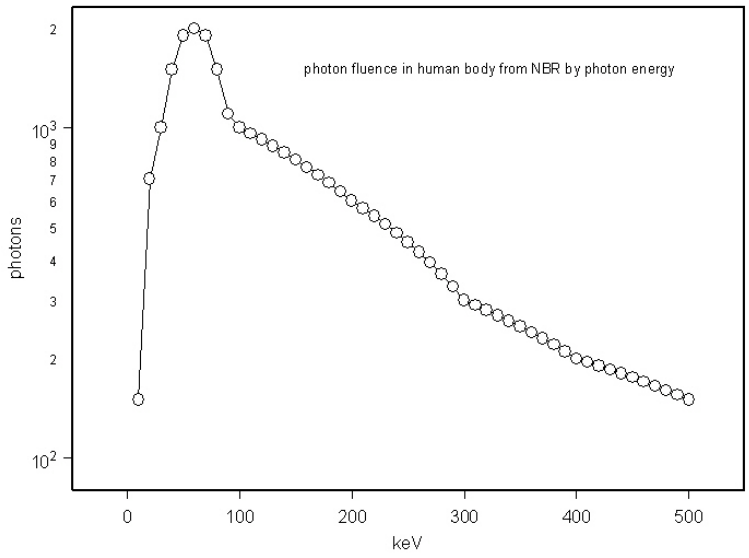
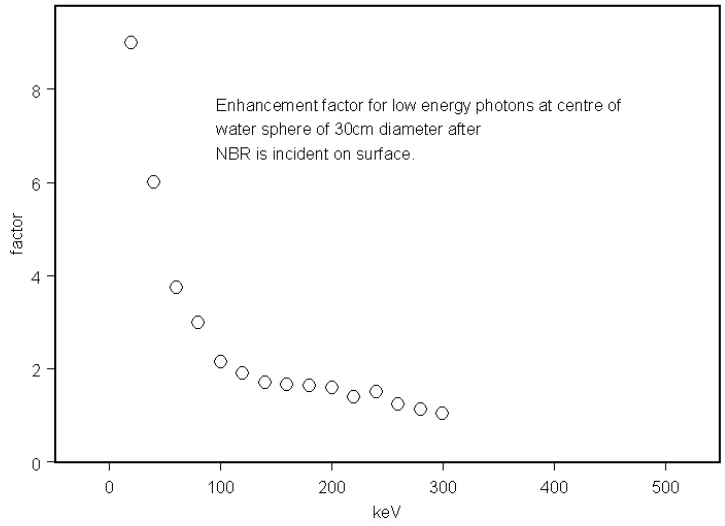


Fig 5 Enhancement of photon energy at different energies on passage through 15cm water. Internal photon fluence divided by external photon fluence. Unpublished measurements.

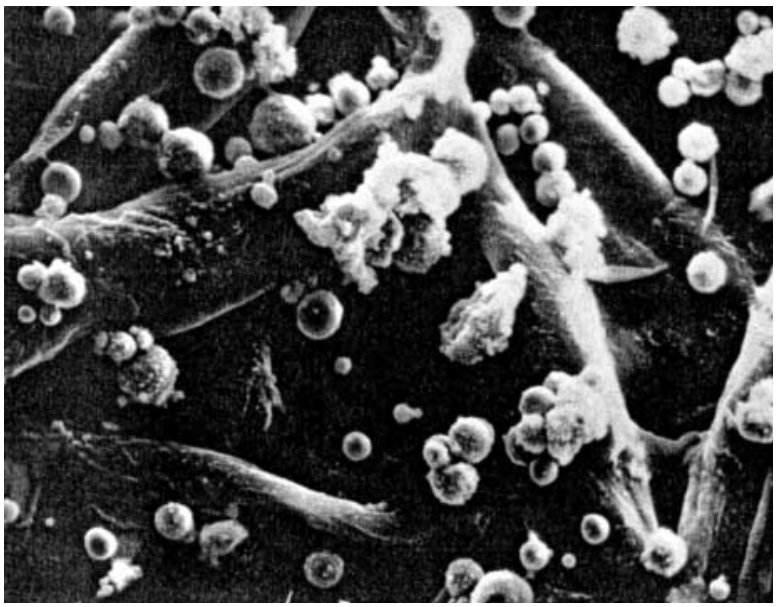


It turns out that 60% of in-air NBR photons have energy below 150keV. Photoelectrons of this latter energy have a mean (CSDA) range in tissue below 20μ (ICRU35, 1984). From Pattison et al's own logarithmically displayed relation (Fig2) it is clear that the dispersion trend below 100keV has been moved from a linear intercept at zero energy of about 60,000 photons to 15,000 photons at the y axis.

3.2 Particle size and shape

Pattison et al 2009 modelled cylindrical particles of 2 and 10μ diameter and height They stated that these were the particles found in the battlefield and in support of this choice they cited the Royal Society report and authors who apparently examined particles from battlefields. Their citation of the Royal Society report is not however supported by reference itself (Royal Society 2001) where a mean diameter of 1μ is employed (RS p44). Indeed particles larger than this are unlikely to pass from the lung into the blood. Measurements on particles collected from battlefields long after the battle are unlikely to show all but the largest diameters since the sub-micron particles will have dispersed. The instaneous dispersion of particle size from DU impacts was obtained using special cascade impactor collectors at the US Aberdeen proving grounds by Glissmeyer et al. (1979). Fig 6 shows spherical DU particles trapped in a cascade impactor. The smaller particles were trapped in a high volume cascade impactor placed behind a frame. The mean geometric diameter for these particles were given by Glissmeyer et al as 0.8μ.

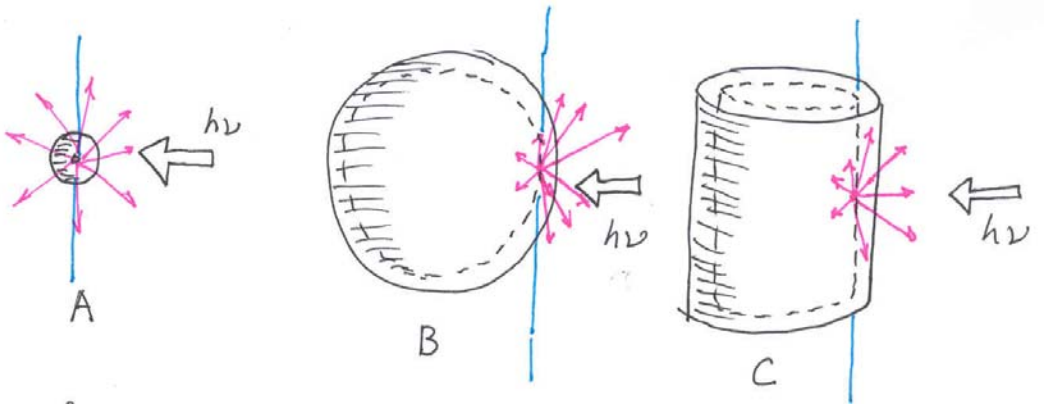
Fig 6 DU particles from a test firing. Geometric mean diameter of all particles collected in all impactors was 2.5μ with very wide standard deviation. Particles were generally spherical.



The curious choice of Pattison et al 2009 to model the larger particles was not as unusual as their choice to model them as cylinders. Photoelectron enhancement is greater the smaller the particle for an obvious reason: there is less internal absorption by the bulk uranium of photoelectrons induced inside the particle. But the choice of a cylinder suggests an agenda since a cylinder absorbs a greater proportion of internally induced photoelectrons than a sphere. Photoelectrons induced by photons striking the surface of the cylinder along a radius may emerge if scattered radially but will be absorbed by the bulk uranium if scattered in the direction of the cylinder axis. This will significantly reduce the photoelectron emissions to tissue, especially in the larger particle and the lower photon energies. The argument is illustrated in Fig 7. This is in addition to the reduction in the photoelectron enhancement resulting from absorption inside the bulk uranium which occurs in the larger particles.

Finally, it is worth pointing out that we are not really concerned with modelling the true sizes of DU particles, we are primarily concerned with health, and therefore even if only 20% of the DU particles are below 200nm, it is perhaps these which are biologically sufficiently mobile to deliver enhanced radiation doses to critical organs or structures. It is not an answer to those who are concerned about this possibility to deal with their fears by modelling particles which are too large to enter the body.

Fig 7. Illustrating the absorption of photoelectrons which scatter along the axis of a cylinder compared with a sphere.



3.3 Curious choice of target volume

This error is really the most extraordinary of all. Pattison et al decided to define their photoelectron enhancement factor based on a arbitrary fixed volume of tissue 5μ deep surrounding the particle being irradiated. Since the particles themselves were of different sizes, the *volumes involved were different* in the two cases; nevertheless Pattison et al concluded from their results that the idea that smaller particles gave greater elative photoelectron enhancements (which is clearly obvious) was actually wrong. The volumes of uranium metal and tissue into which the photoelectrons are absorbed are given for each case in Table 1.

Table 1 Ratios between uranium volume and tissue volume for 2 and 10μ particles modelled by Pattison et al 2009.

Volumes (cm^3)	**Small particle; cylinder 2μ diameter and height**	**Large particle; cylinder 10μ diameter and height**
Volume of particle **v**	6.3E-12	7.85E-10
Volume of cylinder 5μ radius from surface of particle **V**	1.357E-9	5.45E-9
Dose volume **V-v**	1.351E-9	5.45E-9
Ratio of dose volume to particle volume **(V-v)/v**	214	6.9
Enhancements reported	1.0 to 1.4	1.3 to 16.7

Clearly since the large uranium particle is more than 120 times more massive than the small one, there will be roughly more than 120 times the photoelectrons (ignoring self absorption). But since Pattison et al have chosen to define their dose volume as largely the same in both cases, it is clear that the greater apparent enhancement will be found for the larger particle i.e. relative to the background dose from NBR in the same tissue volume. But this does not mean that the effect is greater, since there are 124 times more small particles than large particles for the same mass of uranium.

Pattison et al argued that 5μ represented a reasonable 'dose scoring region' since this was roughly the dimension of a cell. However, 1 particle of 10μ diameter is equivalent in terms of uranium to 1.2 x 10^5 particles of 200nm diameter.

3.4 Differences between the Pattison et al results and literature reported measurements; questions about the accuracy of Monte Carlo programs.

A number of attempts have been made to estimate or to measure the dose enhancements due to photoelectrons near high atomic number elements. In general it seems that Monte Carlo modelling analysis produces results which are different from empirical measurements. For example, Pattison et al have found enhancements of between 1 and 10 in the 5μ region close to uranium particles exposed to natural background radiation. The effective energies involved are below 150keV with a peak relative absorption between tissue and uranium at about 50kev.

Cho 2005 carried out a Monte Carlo study of enhancements due to gold (Z=79) nanoparticles. Dose enhancements were found to be maximally a factor of 2 for 140kVp X-rays (about 120keV photons). On a Z^4 basis, we should expect maximal enhancement of 4 times for uranium in such a system. This largely agrees with Pattison et al. However this result is at odds with the measurements: Regulla et al 1998 carried out a very simple but sophisticated experimental determination of the true enhancements in the vicinity of a thin gold foil exposed to various energies of X-ray photons. Results are given in Table 2. These gave very different and much greater enhancements to the predictions of the modelling. From the Regulla et al study and on the basis of the Z^4 relation we should expect enhancements of about 200-fold for Uranium at the peak energy of 50KeV. Regulla et al were looking at a 100μ depth. But for 60keV photons, 90% of the emitted photoelectrons had ranges below 10μ and so the enhancements to the 10μ region outside the gold foil would have been as high as 500-fold, 1000-fold for uranium and increasing as the photon energy diminished, as predicted.

Table 2. Photoelectron enhancement of dose within 100μ tissue equivalent material touching a 150μ gold foil irradiated with photons of different energy (Regulla et al 1998). Also shown is CSDA approximate enhancement into 10μ tissue from Gold and Uranium

Mean energy keV	CSDA range in tissue (ICRU35) μ	Enhancement of dose in 100μ	Enhancement of dose in 10μ Gold (Uranium[a])
33	18	98	544 (980)
48	44	114	260 (470)
65	60	62	103 (185)
85	99	73	74 (133)
100	145	55	55 (99)

[a] *Calculated as Z^4 ratio Au and U.*

4. Discussion of the Pattison et al 2009 paper.

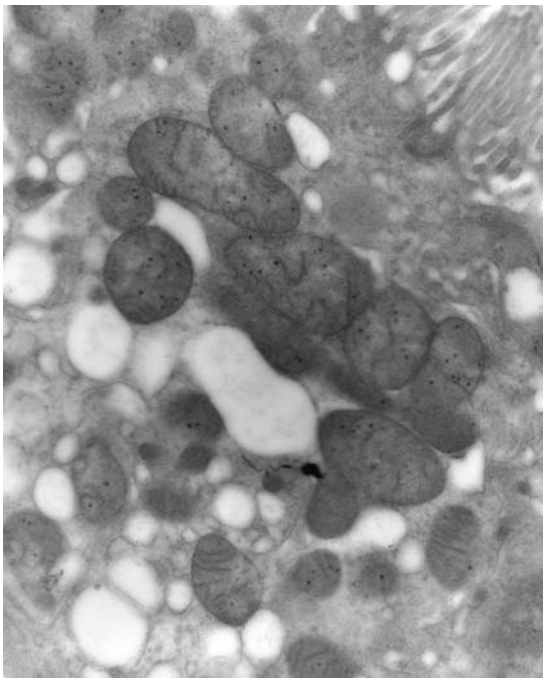
The paper by Pattison et al 2009 has a number of problems and for the purpose for which they are reported its results are misleading. This is a pity, since it would have

been relatively easy to examine the true levels of enhancement close to realistic spherical DU particles of less than 1μ diameter. It will be argued that the paper is biased and, by failure to adequately address the whole issue, dishonest. This author approached the editor of the *Journal of the Royal Society Interface* with the same points which have been addressed here, and asked for space in the journal to raise these issues. The editor refused. This is also a pity, and supports the view that this is a political issue and that the study was carried out not as a scientific exercise but as part of a political project to exonerate the Royal Society from an oversight in their 2001 report and to exonerate the editor himself from an earlier decision.

But the relevant question here is not, what is the dose enhancement in some pre-defined volume, but what is the dose close to the particle and how does it fall off with distance? Why did Pattison et al choose a 5μ volume to absorb the induced photoelectrons? Why not a 10 μ distance or even a 100μ radius volume? This would have shown no enhancement at all as most of the photoelectrons deposit their energy close to the particle. This is what the HPA effectively did in their first response to this issue: they modelled a kidney into which a few milligrams of uranium had been uniformly diluted; naturally this was enough to show an enhancement of gamma NBR absorption to the whole kidney that was extremely low. From this they concluded there was no problem. However, in the real world, there are plenty of problems; this exact situation was studied in a very sick Balkans veteran who was investigated at Manchester Royal Infirmary. The uranium was found in the kidney (Ballardie et al 2008). The calculated dose was low: the man was ill. The uranium was clearly visible in the scanning electron microscope images published by the authors. I reproduce one in Fig 8.

There is a further concern. Pattison in his press interview states that his results show that the concerns about exposure to DU are unsubstantiated. This would not be true even if his results were correct. This is because, as has been argued elsewhere, uranium binds strongly to DNA and the photoelectrons from the uranium on the DNA phosphate leads to effect on the DNA. The existence in a cell of particles of uranium will increase the general concentration of UO_2^{++} ions in the cell and hence the concentration of uranium on the DNA. This mechanism has not been addressed by the Pattison et al study.

Fig 8. Electron micrograph of uranium particles in kidney cells from a Balkans veteran suffering from Gulf War syndrome (from Ballardie et al 2008).



Part II. The dose near a uranium nanoparticle

5 The dose distribution close to high Z particles

It is possible to employ the results of the FLUKA Monte Carlo modelling carried out by Elsaesser et al 2007, 2009 to make an independent estimate the doses to tissue from NBR induced photoelectrons at varying distances from particles of uranium and gold since Elsaesser modelled a water particle of the same dimensions. For particles which are smaller than the mean electron range in uranium all that is needed is to compare the number of photoelectrons of different ranges which emerge into the tissue and to compare the absorbed doses in the cases of presence and absence of the uranium particle. The CSDA electron ranges in tissue and in uranium can be obtained for different energies from tables published by the ICRU (1984). The number of photoelectrons emitted following the interaction with 100keV photons and 10nm particles of water, gold and uranium is given in Table 2 where it compares well with the fourth power Z law prediction although it is larger than would be expected from comparison of the NIST linear photon energy absorption coefficients.
The particle employed here has a diameter of 400nm. This is approximately the diameter of the particles found by Ballardie et al in 2008 in the kidney of a Balkans veteran (Fig 8)
Then the following steps were carried out.

1. The absorption of the 400nm uranium particle is assumed to result from a photon flux which would produce a dose of 1mGy in the equivalent particle of water. Thus the absorption is enhanced by a factor which varies with the photon energy but which is obtained from a comparison of the absorption coefficients at different energies of uranium and muscle tissue published by

NIST. The spectrum of photon energies is that given in Fig 5 and (ignoring electron absorption and binding energies) Table 3. The spectrum is normalised to 1mGy total dose and the enhanced absorption by the particle is normalised to an enhancement factor of 25000 at 100keV obtained by Elsaesser at al and in agreement with the Z^4 power relationship. The result of this step is to calculate a total energy absorption in Joules per uranium particle **E**.

2. This energy is then converted into photoelectrons of different energies and ranges given by a Table 3 modified by internal absorption effects which shift the spectrum slightly to the low energy end by a small amount. This shift is calculated by establishing the absorption of electrons in the bulk of the uranium and the resulting lowering of the energy of the low energy photoelectrons.
3. The energy of these photoelectrons is then diluted into sequential spherical shells outside the particle according to the proportion of their CSDA range which traverses the shell. Dose from the photoelectrons is calculated as Joules per kg. The shells have depth 100nm. Each shell is assumed to be made of ICRU muscle tissue and to have a background dose of 1mGy

Consider the energy deposited into **n** spherical shells of depth **x** distance **d** from the surface of the uranium particle radius **r**.

Employing the CSDA (continuous slowing down) approximation the dose into each volume shell $\mathbf{V_n}$ is made up from the photoelectrons which have **1/n** fraction of the energy from photoelectrons with range **d**. Thus photoelectrons which only have energy to reach shell **n=1** will just deposit all their energy into that shell. Photoelectrons which have sufficient energy to reach shell **n= 2** are assumed to deposit half (1/n) their energy in shell 1 and so on.

$$\text{Then } \mathbf{V_n = 4/3.\ \pi.\ (r + nx)^3 - V_{n-1}}$$

And the photoelectron dose in shell **n**, distance $\mathbf{d = nx}$ is simply:

$$\mathbf{D_d = (1/n\ .\ \Sigma\ E_{nx})/V_n}$$

Table 2 Number of photoelectrons emitted following exposure of a 10nm particle of water, gold and uranium to 100keV photons (normalised to water). Comparing FLUKA results from Elsaesser et al 2008/2009 with Z^4 predictions. See Fig 1.

	Water (Z=7.5)	**Gold (Z=79)**	**Uranium (Z=92)**
Elsaesser et al	1	12,900	29,200
Z^4	1	12,300	22,600

The distribution of photoelectron energy by 10keV bins in the natural background spectrum is given in Table 3 where the CSDA ranges of these photoelectrons (from ICRU 35, 1984) are also tabulated. Also given is the fraction of all energy in NBR associated with the PEs of the relevant energy range. It should be emphasised that the flux of low energy photons from NBR and secondary effect inside tissue is uncertain.

Table 3 Percentage of photoelectrons of energy equal to the photon energy distribution of external NBR together with their ranges in μ. (from Pattison et al, unpublished work and ICRU35, 1984)

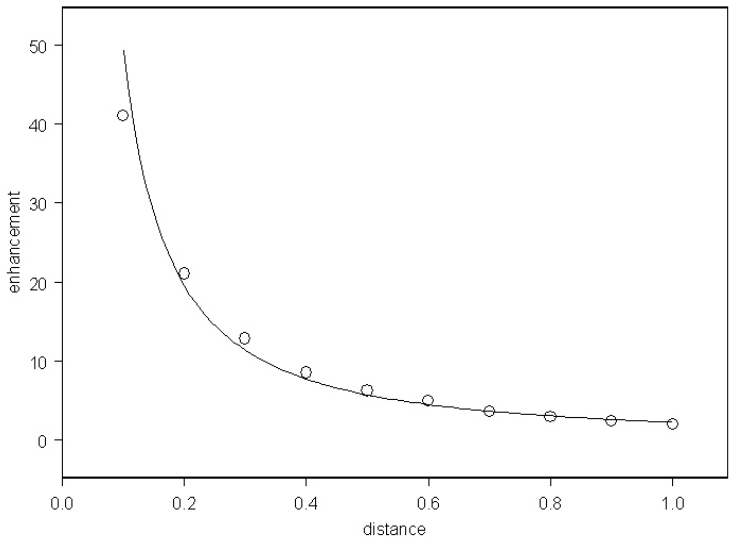
keV bin	% of all photons	Range μ.
5	0.5	1.2
10	2.35	2.5
20	3.36	8.6
30	5.04	17.5
40	6.39	29
50	6.72	44
60	6.39	60
70	5.04	99
80	3.7	113
90	3.36	126
100	3.22	144
110	3.09	170
120	2.96	200
130	2.82	230
140	2.69	258
150	2.56	280
160	2.42	314
170	2.29	348
180	2.15	382
190	2.01	416
200	1.9	450
210	1.8	486
220	1.7	522
230	1.61	558
240	1.51	594
250	1.41	630
260	1.31	666
270	1.21	702
280	1.11	738
290	1.01	774
300	1.01	810
310	0.94	864
320	0.90	918
330	0.87	972
340	0.84	1026
350	0.81	1080
360	0.77	1134
370	0.74	1188
380	0.71	1242
390	0.67	1296
400	0.65	1350
410	0.64	1394
420	0.622	1438
430	0.606	1482
440	0.588	1526
450	0.571	1570
460	0.555	1614
470	0.538	1658
480	0.521	1702
490	0.505	1746
500	0.490	1790
500-2500	2.2	

The results of the calculation for a 400nm diameter uranium particle show dose enhancements which fall off rapidly with distance in 100nm shells of tissue. These results are given in Table 4 and are displayed in Fig 9.

Table 4 Dose enhancements (multiplier) for photoelectrons induced by natural background radiation within sequential spherical shells of depth 100nm from a 400nm diameter uranium particle embedded in tissue (ICRU muscle).

Shell (μ)	**Dose enhancement (-fold)**
Particle surface-0.1	41
0.1-0.2	21
0.2-0.3	12.8
0.3-0.4	8.5
0.4-0.5	6.2
0.5-0.6	4.9
0.6-0.7	3.6
0.7-0.8	2.9
0.8-0.9	2.4
0.9-1.0	2
Particle surface – 1.0	**4.6**

Fig 9 Enhancement of dose in sequential tissue shells by distance in microns from a 400nm diameter uranium particle exposed to Natural Background Radiation



What emerges from the calculation considerations is the extreme dependency of the result on the distribution of low energy photons inside the body at the position of the particle. Note that the dose falls off with distance, as expected, and if a dose scoring

region of 5μ had been chosen in the present calculation, it is unlikely that the enhancement factor would have been significant. This calculation showed that the enhancements found depended critically on the photon flux in the region between 0 and 100keV. Since this is not really accessible from gamma spectra of natural background radiation all kinds of assumptions have to be made about its true nature inside the human body where all kinds of secondary photon processes occur. It seems questionable whether the Monte Carlo programs employed by Pattison et al can accurately predict the low energy photon fluxes; it is noteworthy that the measurements of Regulla et al who embedded a thin detector in the experiment next to the irradiated gold foil showed significant enhancements of ionisation density, more than found here.

As with Pattison et al, the enhancements found by Cho 2005 who Monte Carlo modelled gold nanoparticles were very modest (a factor of 2) and seem highly unlikely to be able to explain the profound effects on X-ray enhancements to tumour irradiation shown by Hainfeld et al. 2004.

It is suggested that the only accurate way to determine the enhancements due to photoelectron induction at particles of high Z is by experiments involving those particles in which the energy is measured in some way directly, physically with embedded dosimeters of some kind or biologically, and not by mathematical modelling which seems to depend on inputs of parameters which are not accessible directly and may therefore be incorrect.

6. Dose or track density? Mobility of the uranium particle, multiple scattering. Inability of physical models to deal with the complexity of interactions in biological media on the nanometer scale

In the calculations made in Section 5, what is calculated is the absorbed dose, energy per unit mass, in spherical shells close to the uranium particle. However, dose itself is not necessarily the most important parameter. As the photoelectron energy falls, the number of tracks per unit dose increases proportionately: there are more photoelectrons. This means that for DNA or chromatin which intercepts or is intercepted by such a particle, the density of tracks is very great, which allows the target to receive more than one hit in a shorter space of time. This results in a second order kinetic regime where two or more hits in space or in time are much more likely to occur than for the longer range photoelectrons induced by higher energy photons. Thus Second Event processes and dose squared processes are more likely to occur (see CERRIE 2004b).

It should also be borne in mind that these particles are mobile, and move in time, therefore the probability of interception with a chromosome or with DNA is much higher than might be believed on the basis of simple distance volume arguments.

Finally, the processes involved in radioactive decay and photoelectron scattering also produce secondary photons; the decay of U-238 produces a 48keV photon; there are atomic fluorescence processes, there are gamma photon emissions when the beta daughters (Th234, Pa234m) decay; the atomic and molecular processes in the complex atomic media which make up living tissue result in complex multiple inelastic scattering of photons which will all interact with the uranium particles. In addition, these particles will be dissolving slowly, resulting in high concentrations in the cell of uranyl ions, which will bind to DNA. The particles themselves are likely to be powerful chemisorption foci for DNA; another reason why they will present a

hazard which is not considered by simple physics based vacuum or water interface calculation.

Endnote for Sociologists and Historians :
Bias in Science and Policy: Monty Charlo codes

Pattison and Hugtenburg, two of the authors of Pattison et al 2009 were *fully aware* of the measurements made by Regulla et al 1998 which showed high levels of enhancement of radiation near thin gold foils due to photoelectrons induced by gamma radiation (Table 2). Yet *no mention is made in their paper* of these important and relevant results and Regulla et al 1998 *is not cited*. Why? If it had been, there would immediately have been a question about how their own Monte Carlo results did not show enhancements similar to those measured by Regulla et al within 150μ of a thin film of gold, a material with a lower Z than uranium. Regulla et al 1998 *is* however cited in another earlier paper by Hugtenberg, Chaoui and Pattison published in 2007. This paper (Hugtenburg 2007) addresses the difficulty of employing Monte Carlo modelling for the purposes of determining doses in microdosimetric volumes of the order of less than a few microns. It states: *the radiobiological characteristics of the photoelectron and Auger electrons generated in the photoionisation process are not well know, in addition, the physics required to transport low energy (below 10keV) electrons at cellular dimensions has not generally been incorporated into general purpose Monte Carlo codes.* We do not hear anything about these difficulties in Pattison et al (2009). Earlier in the 2007 paper we read: *although* [Monte Carlo studies do not show significant enhancements with Gold and Platinum] *studies show that the degree of dose enhancements could be of the order of 100 for cells in close proximity to a metal surface [Regulla et al 1998]*. Scientific reports, if they are to be of value and honest should address all sides of any issue; it is unacceptable for Pattison et al 2009 not to have drawn attention to the evidence from Regulla et al 1998 that their own modelling might have been in error, nor to raise this question which two of the authors had already raised in a separate paper published two years earlier.

The question of Science advice to Policy was discussed in 2001-2005 among some 50 eminent doctors, epidemiologists and researchers in the EU- PINCHE advisory group, the Policy Information Network for Child Health and Environment. It was concluded that bias regularly existed in scientific publications which addressed environmental health (van den Hazel et al 2007). It was concluded that some degree of bias was inevitable in scientific publications from authors who were working for various organisations which had a political or economic agenda. It was recommended that the only way for policymakers to make best decisions about environmental agents which were suspected of causing health problems was to commission oppositional reports. This is an early response to the psychological concept of *groupthink* (Janis and Mann 1977) whereby groups under pressure over previous mistakes affirm their mistake by ignoring evidence and excluding dissent: the US Pentagon now are aware of such errors and have set up *Red-teaming* groups to ensure such groupthink errors are detected before a disaster occurs (like the BSE Mad Cow disease disaster, a perfect example of groupthink). In internal low dose radiation issues the CERRIE committee was set up as such a group, but in the event, the Minister who set up the committee was removed from office and legal threats were made to suppress dissenting views in the final report. A minority report was published (CERRIE 2004b).

One of the major discussions in CERRIE was about the effects of hot particles. Battlefield DU should have been discussed but was excluded by the Chair, Dudley Goodhead, on the basis that the DUOB would be discussing the issue. Nevertheless, the argument that uranium and plutonium particles in the Irish sea were being resuspended and were a potential cause of the excess cancers near the Irish Sea coast were addressed though the Chair's unilateral (and opposed) decision to commission a report by Dr Monty Charles on the University of Birmingham. This report claimed to address the issue and appeared in the *Journal of Radiological Protection*, whose editor was Dr Richard Wakeford of British Nuclear Fuels. Naturally Monty Charles concluded that hot particles were not a problem and cited all the reports which supported this, excluding those which did not. Charles was also at the Oxford international meeting of CERRIE in 2003, when Busby advanced the uranium photoelectron idea. Charles immediately attacked it saying that the *Bragg effect* meant that no enhancement could occur because the particles/ tissue volumes were too small. This was nonsense. In trying to track down the reason for Pattison et al deciding to study the issue it soon became clear that there were links between Pattison, Hugtenburg and Monty Charles, the latter two being both at the University of Birmingham. Charles has links with the National Radiological Protection Board and is a member of the British Nuclear Energy Association. NRPBs John Harrison, another member of CERRIE, wrote the invited editorial in 2003 in Wakeford's *J Radiol Prot* which discussed the Charles and Mill paper exonerating hot particles. Harrison is now the HPAs (and the ICRPs) main man on internal radiation effects.

These connections are teased out to indicate that the paper by Pattison et al 2009 was not just the result of some sudden decision by Pattison or Hugtenburg or Green to wake up one morning and think, *I know, let's look at that DU effect suggested by Busby.* Some questions are valid here. How did they hear about this obscure idea. Who paid for all that work? No organisation is acknowledged. How did it get into the *Journal of the Royal Society Interface*? Why did Bonfield allow it through when he had refused to publish the paper by Busby which was the basis for Pattison et al's calculation? The authors were reduced to citing an internet source for the theory that they were attacking. How extraordinary! Why did Bonfield not allow Busby to respond with the queries raised in this paper, which were all sent to Bonfield? What, if any, was the role of Monty Charles? Was this whole affair driven by the Health Protection Agency and the ICRP (largely the same people)?

In 1998, at the International Conference in Stratford on Avon of the British Nuclear Energy Society, *Health Effects of low dose Radiation: Challenges for the 21st Century* the Green Party carried out a non violent direct action. Richard Bramhall of the Low Level Radiation Campaign dressed and made up as Death (from Bergman's Seventh Seal) chained himself to the platform during the keynote speech of the late Sir Richard Doll (who was denying the link between childhood cancer and radiation from nuclear plants, a link which has now been largely accepted). This was to draw attention to the delegates that *Death* was at all their conferences. That *death* was the consequence of their getting things wrong. In the photograph of this event, taken by Busby at the time and published in his 2006 book *Wolves of Water* sitting next to *Death* was Dr Monty Charles (Busby 2006) (Fig 10).

Fig 10 Richard Bramhall of the Low Level Radiation Campaign dressed as Death and chained to the platform at the British Nuclear Energy Society conference at Stratford on Avon, 1998. Next to him is Dr Monty Charles, advisor to the NRPB on hot particles and their health effects.

The scientific bias fortress which surrounds the health effects of low dose internal radiation and health has been well described recently by Paul Zimmerman, a New York academic (Zimmerman 2009). The desperation of the ICRP, HPA and other members of this magic circle to dismiss or marginalise the increasing evidence that they have got it very wrong and that people have been dying because of this is reminiscent of the tobacco industry arguments, the asbestos industry issue and thalidomide. At some point the wall will come down, and at that point, it has been argued recently in a conference held at the Royal Society in 2008, the individuals who participated in the biased science that supported the obsolete ICRP risk model should be individually prosecuted. If this happens, as it should, then the machinery behind the creation of papers like Pattison et al 2009 will become clear in a court of law and the innocence or guilt of the parties established by an independent jury.

These matters, as they relate to the overall assessment of radiation and health and the groups which underpin the incorrect model of the ICRP are discussed in some detail in the new ECRR2010 report (ECRR2010, 2010)

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Abstract

The health effects of exposure to uranium weapons is an area of scientific dispute. The arguments pivot on the admissibility of the ICRP radiation risk model for predicting the effects of internal exposures from uranium micro- and nano- particles produced when uranium weapons strike their targets. One recent explanation for the failure of the ICRP radiation risk model has involved the enhancement of dose near particles of uranium due to its high atomic number. This results in the conversion of natural background gamma radiation into local photoelectrons which increase radiation dose to local tissues. It now seems that this idea has been accepted but arguments remain about its magnitude. A recent mathematical study by Pattison et al 2009, published in the prestigious *Journal of the Royal Society Interface* finds that the effect is significant but modest with enhancements of between 1 and 10 for particles modelled as 2 and 10 micron diameter cylinders. The present paper criticises the Pattison et al findings on five grounds:

- The particles are cylindrical and not spherical thus reducing the enhancement dose
- The particles are far larger than those found in the battlefield which are spherical and smaller than 1 micron in diameter.
- The choice of the same 5 micron target volume for large and small particles results in incorrect and misleading conclusions about the effect of particle size
- The input data removed a significant proportion of low energy photons from the natural background radiation spectrum, thus reducing the enhancement, since it is the low energy photons that contribute to the short range photoelectrons
- The results were not supported by real measurements and biological effects reported by different groups.

The present paper goes on to employ results from Monte Carlo mathematical modelling by Elsaesser et al 2007 to develop semi-empirical calculations of dose enhancement near a 400nm uranium particle embedded in ICRU tissue. Results show increasing enhancement close to the particle surface with a maximum value of 50-fold within 100nm of the surface. However it is found that there is a critical dependence of modelling results on the low energy photon spectra in tissue and it is suggested that mathematical modelling of these small particle, low energy photon interactions is unsafe, and that results should be obtained from experiment and not modelling. Those experiments which have been done e.g. by Regulla et al 1998 and Hainfeld et al 2004

suggest enhancements of greater than 2000-fold would exist close to uranium particles.
The role of the Royal Society and the *Journal of the Royal Society Interface* in this important political issue is criticised, as is the paper by Pattison et al, 2009.