Draft 01/11/2008Final Rev1

UNIVERSITY OF BIRMINGHAM

Skin dose from Ra-226 contamination: Dose estimation & comments

M W Charles

School of Physics & Astronomy
University of Birmingham
Edgbaston
Birmingham B15 2TT

Tel 01214143483
Fax 01214144725
M.W.Charles@bham.ac.uk
Mob 079689 40199

2 November 2008

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EXECUTIVE SUMMARY

An initial evaluation was made in 2006 of the possible skin doses from small areas of Ra-226 contamination. The work presented here takes advantage of recent measurements of the activity levels in 39 environmental samples and now enables a more definitive evaluation of skin dose to be made**.** For the purposes of skin dose evaluation a bench mark Ra-226 point source is considered with an activity of 1 MBq. The Ra-226 is assumed to have been chemically extracted many decades ago so that all daughter products have grown in to an equilibrium level – i.e. a secular equilibrium has been achieved so that each of the daughter/progeny radionuclides also have an activity of 1 MBq. The recently measured levels of Ra-226, Pb-214 and Bi-214 indicate progeny activities are about 90% of the equilibrium value – due presumably to some loss of radon gas from the decay chain.

An understanding of skin dosimetry requires a basic knowledge of skin anatomy. The skin consists of an outer epidermis and a thicker underlying dermis. The epidermal layer has an outer dead layer (keratin) below which there are usually several layers of viable cells. The deepest layer of cells in the epidermis is called the basal layer which contains stem cells that continually renew the epidermis as the outer layers are shed. The epidermal stem cells are widely considered to be the most likely target cells for cancer induction but this remains as yet unproven. Considerable evidence implicates the additional involvement of cells deeper in the skin. The target cells for the most important deterministic effects such as ulceration have been identified and are at various levels in the underlying dermis.

The International Commission on Radiological Protection (ICRP) recommends that for radiological protection purposes the skin dose should be evaluated to the cells of the basal layer. The depth of these cells is often referred to in radiation protection as the skin thickness. The ICRP[1-3] recommend a value of 70 μm for routine skin dose assessment. For non-uniform exposures the ICRP recommend that this dose should be averaged over the most highly exposed area of 1 cm^2. Where the term skin dose is used in this report it will indicate such a measurement or calculation and may be indicated by (70 μm, 1 cm^2).

Skin absorbed doses for beta and photon emitters have been calculated for the bench mark Ra-226 source using the code VARSKIN 3[4]. Alpha absorbed doses have been calculated using the code ALDOSE[5,6]. Skin absorbed doses for individual environmental sources can be assessed by applying pro-rata corrections to the bench mark calculations on the basis of measured activity levels and the extent to which equilibrium is achieved. Point source geometry has been used to evaluate the skin dose, averaged over an area of 1 cm^2 at various tissue depths representative of the skin thickness at the majority of body sites. The dose estimates are also appropriate for small sources with other geometries, providing that self absorption is negligible. The calculated bench mark doses will thus be overestimates of actual doses from environmental samples, particularly for alpha radiation.

Ra-226 alone is primarily an alpha emitter (energy ~ 4.8 MeV) with a few low intensity and low energy beta and gamma emissions. The range of these alpha particles is ~ 30 μm, much less than the nominal skin thickness of 70 μm. The hazard posed by skin exposure from Ra-226 is therefore due primarily to its daughter products which include a number of high energy alpha, beta and gamma emissions. The major contribution to skin dose at the nominal skin depth of 70 μm from Ra-226 and its progeny is from beta radiation. The alpha dose to the skin at this depth is zero and gamma♥ dose is negligible. Using VARSKIN 3 the total skin dose rate (70 μm, 1 cm^2) from a Ra-

♥ This should strictly be referred to as photon dose since x-rays as well as gamma rays are involved.

226 point source with an activity of 1 MBq (in equilibrium with all progeny) is ~ 5.5 Gy/hour. The photon absorbed dose rate to skin is ~ 72 mGy/h. The main contributors to skin dose rate are:

Pb-214: 2.01 Gy/h Bi-214: 1.81 Gy/h Bi-210: 1.63 Gy/h

The basal layer of the epidermis of the skin on some body sites for a proportion of the population may be less than the nominal value of 70 μm recommended by the ICRP. A range of depths of 20-100 μm covers the actual range of skin thicknesses found in a population over the majority of body sites. The basal layer on some body sites may thus be subject to exposure from the higher energy alpha particles from Ra-226 progeny – primarily Po-214 (7.7, MeV, range in skin ~ 64 μm). In the case of 1 MBq Ra-226, in equilibrium with daughter products, an average skin absorbed dose rate at a depth of 20-100 microns from alpha radiations is about 65 Gy/hour (weighted by the skin thickness distribution in a population). It is necessary to be extremely careful in interpreting the significance of such high numerical values of dose rate. Actual doses may be very much less due to self absorption within the sample. The fact that equilibrium in actual measured samples is high indicates that the source is bound and sealed effectively against radon loss. This implies significant self absorption, particularly for alpha particles. This conjecture could be tested directly by contact dose measurements using radiochromic dye film if required. These doses should not be compared directly with recommended dose limits which are based primarily on experience with photon irradiations which irradiate the full skin thickness. A literature search indicates that such large alpha doses have been observed to produce reddening (erythema) and pigmentation in human skin, but evidence is lacking to convincingly link such superficial alpha exposures with more severe detrimental deterministic effects such as ulceration, or with subsequent skin cancer. At such high local skin absorbed doses from alpha radiation it is more likely that cells will be killed, thus preventing any induce mutations from proceeding to malignancy.

Since the relevant contributions to skin dose from Ra-226 for radiation protection purposes are from beta and gamma radiations it is possible to categorise Ra-226 environmental samples in a similar way to that used by the Dounreay Particle Advisory Group (DPAG) for Dounreay fuel fragments (DFFs). Classification for DFFs was based on a consideration of skin dose rate (70 microns, 1 cm^2) likely residence time, and the probability of producing skin ulceration. ‘Significant’ radioactive sources were considered to pose a realistic potential of causing harm. This was equated with sources which produced skin dose rates greater than ~3 Gy/hour. ‘Relevant’ sources are those which produce an order of magnitude lower dose rate of ~ 0.3 - 3 Gy/hour but still warrant monitoring and removal from the environment. ‘Minor’ sources are those with lower skin dose rates. ‘Significant’ Ra-226 sources are those with activity of $\gtrsim$ 0.6 MBq, assuming all progeny are in equilibrium. On this basis the 39 recently evaluated Ra-226 samples would be classified as:

2 significant 11 relevant 26 minor

Appendix 1 discusses the evaluation of the contribution to effective dose from a localized skin exposure from Ra-226, taking into account contributions from beta and alpha exposures and thin skin regions of the body. A ‘Significant’ Ra-226 source of 0.6 MBq activity can give a contribution to effective dose in about 1 hour of ~ 0.4 mSv. This is comparable with the calculated annual contribution to effective dose from radon progeny deposited on the skin from radon in air at average UK radon levels. These evaluations of contribution to effective dose need to be treated with care because of all the provisos regarding the role of alpha radiation in skin cancer induction.

INTRODUCTION

At the request of Paul Dale, on behalf of SEPA, I have made an estimation of the potential skin dose rates that would be delivered by small samples of Ra-226. An initial evaluation[7] was made in 2006 based on a nominal 1 MBq activity for Ra-226. No information on environmental activity levels or the extent of radioactive equilibrium was available at that time. The work presented here takes advantage of recent measurements of the activity levels in 39 environmental samples (see table 1) which enables a more definitive evaluation of potential skin doses to be made**.** For the purposes of skin dose calculations a 1 MBq bench mark point source of Ra-226 activity, and equilibrium with progeny/daughter products, has been assumed.

Skin absorbed doses for beta and photon emitters have been calculated using the code VARSKIN 3[4]. Alpha absorbed doses have been calculated using the code ALDOSE[5]. Skin doses for individual sources can be assessed by applying pro-rata corrections to the bench mark calculations - on the basis of activity levels and the extent to which equilibrium has been achieved. Point source geometry has been used to evaluate the skin dose, averaged over an area of 1 cm^2 at various tissue depths representative of the skin depth at various body sites. The dose estimates are also appropriate for small sources with other geometries which are thin enough so that self absorption is negligible. Self absorption is particularly important for considerations of alpha dose to the skin. Sample thicknesses in excess of a few tens of microns will totally absorb the alpha radiations considered here.

I have assumed that Ra-226 was chemically extracted many decades ago so that there is the potential that all daughter products could have grown-in up to an equilibrium level – i.e. a secular equilibrium[♣] could have been achieved with the parent Ra-226, so that each of the daughter radionuclides have an activity of 1 MBq. A major problem with the evaluation of skin dose rate from contact with Ra-226 is the extent to which progeny have remained in the sample and depending on the age of the sample, the extent to which radiation equilibrium has been achieved.

RADIOACTIVE EQUILIBRIUM

Radium 226 is a product of the decay of U-238. Figure 1 shows the decay scheme. The main decay route - which produces the most energetic emitted radiations - is indicated by the emphasized arrows. The decay of Ra-226 produces the radioactive gas radon Rn-222. Unless this escapes the radon activity will build up to its equilibrium value within a few half lives – i.e. 10-15 days. During this time the subsequent 4 radionuclides (Po-218, Pb-214, Bi-214 & Po-214) in the decay chain, which are all short lived, reach secular equilibrium within a few hours. These radionuclides are the main contributors to skin dose from environmental radon[8,9]. The next member of the decay chain is Pb-210 with a half life of 22 years. Secular equilibrium of Pb-210 and subsequent progeny is achieved only over several subsequent decades. A major problem with the evaluation of skin dose rate from contact with Ra-226 is the extent to which progeny have been retained within the radium sample. This will depend upon the construction of the radium sample and the extent to which this has allowed the gas radon 222, and its progeny to escape from the sample.

The extent of equilibrium can be assessed by comparing the recently measured levels of Ra-226, Pb-214 and Bi-214. Figure 2 shows a regression of Pb-214 activity against Ra-226 activity which indicates that over the whole range of the 39 environmental samples the Pb-214 activity is at about 90% of the equilibrium value. Lack of total equilibrium is presumably due to some loss of radon gas from the decay chain. The regression of Pb-214 and Bi-214 indicates that these two progeny are

[♣] Secular equilibrium is the situation where all progeny have the same effective decay rate as the parent radionuclide.

in close equilibrium with each other. The assumption for the purpose of calculations that the Ra-226 decay series are in equilibrium will thus produce an over-estimate of dose by about 10% since progeny in the chain following Rn-22 will be in deficit. The fact that equilibrium has been almost achieved and maintained over several decades is indicative that all radionuclides are well bound and sealed from the external environment. This implies the presence of binding and sealing material within the samples. The relevance of this is that binding materials and external sealants are likely to significantly reduce the alpha dose estimates that I have made – which assume no absorption within the sample. This could be checked if necessary by direct alpha dose measurement using, for example, Radiochromic dye film and thin absorber foils.

SKIN DOSE RATE FOR Ra-226 IN THE PRESENCE OF ALL PROGENY (SKIN THICKNESS ASSUMED TO BE 70 μm)

Ra-226 alone is primarily an alpha emitter (energy ~ 4.8 MeV) with a few low intensity and low energy beta and gamma emissions. The range of the alpha particles is ~ 30 μm, much less than the nominal skin thickness of 70 μm assumed by the ICRU and ICRP. The hazard regarding skin exposure from Ra-226 is therefore due primarily to its daughter products.

If Ra-226 is in equilibrium with all its progeny then all of the radionuclides shown in Figure 1 will contribute to skin dose. The major contribution to skin dose at the nominal skin depth of 70 microns is from beta radiation. On the assumption of a skin thickness of ~ 70 μm the alpha dose to the skin is zero. Gamma[♥] dose is negligible. Using VARSKIN 3 the total skin dose rate (at a depth of 70 μm over an area of 1 cm^2) from a Ra-226 point source activity of 1 MBq (in equilibrium with all progeny) is ~ 5.5 Gy/hour. Photon dose to skin is ~ 72 mGy/h. The main contributors to skin dose are:

Pb-214: 2.01 Gy/h Bi-214: 1.81 Gy/h Bi-210: 1.63 Gy/h

Po-214 and Po-218 are the major alpha emitters but their energies (7.7 & 6.0 MeV) and hence the range/depth of penetration just fails to make a contribution to skin dose at a depth of 70 μm (figures 3 & 4).

SKIN DOSE RATE TO THE SUPERFICIAL BASAL LAYER OF THE SKIN (SKIN THICKNESS < 70 μm)

The basal layer of the epidermis of the skin on some body sites for a proportion of the population may be les than the nominal value of 70 μm recommended by the ICRP (see figure 5). The basal layer on some body sites may thus be capable of alpha exposure from the higher energy alpha particles from Ra-226 progeny – primarily Po-214 (7.7, MeV, range in skin ~ 64 μm).

In the case of a 1 MBq Ra-226 source, in equilibrium with daughter products, an average skin absorbed dose rate at a depth of 0-100 microns (weighted by the skin thickness distribution in a population – figure 5) is about 65 Gy/hour. The surface dose is ~ 6 kGy/hour (figure 4).

It is necessary to be extremely careful in interpreting the significance of such high numerical values of dose rate. Actual doses may be very much less due to self absorption within the sample. The fact that equilibrium in actual measured samples is high indicates that the source is bound and sealed effectively against radon loss. This implies significant self absorption, particularly for alpha

[♥] This should strictly be referred to as photon dose since x-rays as well as gamma rays are involved.

particles. This conjecture could be tested directly by contact dose measurements using Radiochromic dye film if required. The very high surface doses are delivered to the dead keratinized outer layer and are not relevant to induced biological effects. These doses should not be compared directly with recommended dose limits which are based primarily on experience with photon irradiations which irradiate the full skin thickness. A literature search indicates that such large alpha doses have been observed to produce reddening (erythema) and pigmentation in human skin, but evidence is lacking to convincingly link such superficial alpha exposures with more severe detrimental deterministic effects such as ulceration, or with subsequent skin cancer.

BIOLOGICAL EFFECTS OF Ra-226 CONTAMINATION OF THE SKIN

Comparisons with Dounreay fuel fragments (DFFs)

A consideration of the potential biological effects in the skin of small sources of Ra-226 is similar to that described in recent HPA evaluations of the effect of small fuel fragments found in the vicinity of the Dounreay nuclear site[10]. A major factor in all such adventitious exposures is the duration of the exposure. A discussion of this issue has been given in the context of fuel fragments at Dounreay[11]. Skin cancer risks from DFFs are negligible due to the small area of skin exposed and the low tissue weighting factor for the skin[ℵ]. On the basis of considerations of international animal experiment data it was concluded that the threshold skin dose (measured at 70 microns, over an area of 1 cm^2) for transient small area superficial skin ulceration is ~ 2 Gy and the dose for probability of 50% incidence was 10 Gy. On this basis the Dounreay Particle Advisory Group (DPAG)[12] placed Dounreay fuel fragments into 3 categories depending on their hazard, which can be related to skin dose rate:

Significant	> 10^6 Bq Cs-137	Skin dose rate (70μm, 1 cm^2)	> 3 Gy/hour
Relevant	10^5 - 10^6 Bq Cs-137	Skin dose rate (70μm, 1 cm^2)	0.3 – 3 Gy/hour
Minor	< 10^5 Bq Cs-137	Skin dose rate (70μm, 1 cm^2)	< 0.3 Gy/hour

The minor category was considered to be low risk and did not require monitoring or removal.

On this basis Ra-226 environmental samples with activity > 0.6 MBq (skin dose rate 5.5 Gy/MBq x 0.6 MBq x 0.9 = 3.0 Gy/hour, taking into account incomplete equilibrium) would be categorised as ‘significant’. Ra-226 environmental samples with activity between 0.06 - 0.60 MBq would fall into the ‘relevant’category. Ra-226 environmental samples with activity less than 0.06 MBq would be considered ‘minor’.

Of the 39 Ra-226 samples in table 1, the DPAG classification would give:
2 significant 11 relevant 26 minor

The main difference between exposures from Dounreay fuel fragments and Radium-226 contamination is the presence in Ra-226 of significant quantities of alpha emitters. The biological effects in the skin from alpha emitters has been the subject of recent reviews in the context of skin exposure form radon in air[8,9] and from depleted uranium[13]. A brief summary of some of the main points discussed in these reviews is given below.

[ℵ] The tissue weighting factor for the skin is 0.01. The whole body skin area is about 2 x 10^4 cm^2. Skin cancer risk is proportional to the area of skin exposed - so for the exposure of 1 cm^2 a further weighting factor of ~ 0.00005 must be used.

Deterministic effects of alpha radiation

For deterministic effects there is a paucity of information on the dose response relationship for alpha exposures, particularly at high energies ($\gtrsim$ 6 MeV) which are capable of penetrating the outer layers of skin on the majority of body sites. For lower energy radiations which can only irradiate the basal layer of the epidermis on thinner skin areas, threshold absorbed doses for erythema (skin reddening) in human skin are ~ 1 kGy. Total doses up to several tens of kGy (surface absorbed dose) have been administered over a period of minutes without more severe effects than erythema or pigmentation being observed. These alpha doses are at least 1000 times higher than beta doses required to produce similar effects. Dose limits for the skin are based on the results of clinical and animal data for photons and high energy beta radiations. It is clear therefore that the high skin doses that can be calculated from alpha exposure to the superficial layers of the skin should not be compared directly with the skin dose limit[♠]. High LET radiations such as alpha particles are invariably more effective per unit absorbed dose than low LET radiations. The relative ineffectiveness of alpha radiations to produce deterministic damage in the skin is thus most likely to be due to their superficial depth doses and their ineffectiveness at damaging important target cells that presumably reside at deeper depths.

Figure 6 shows that Pm-147 has a similar depth dose to Po-214 (alpha 7.7 MeV). Pm-147 has a high penetration 'tail' and is capable of irradiating deeper cells than Po-214. Pm -147 can induce erythema and acute epidermal necrosis (AEN)[∞] – which in simple terms is a transient superficial moist surface lesion, but cannot produce more severe responses even at very high doses. Higher energy beta radiations such as Sr/Y-90 (beta E max 2.28 MeV) can penetrate the skin into the deep dermis and can produce deep dermal ulceration – which is a clinically significant lesion at high doses that may be difficult to treat and may leave residual small scars. The threshold surface dose for a small Pm-147 source to produce AEN is about 500 Gy[3]. A surface absorbed dose of about 720 Gy has a 50% probability of producing AEN. Po-214, like Pm-147, will not be capable of producing more severe skin responses. Higher threshold doses would be expected for Po-214 on the basis that it is less penetrating than Pm-147.

A 1 MBq Ra-226 benchmark source will produce a surface absorbed dose in a 1 hour exposure of ~ 6 kGy over an area of 1 cm^2. This is very likely to produce transient erythema and pigmentation and possibly superficial transient AEN. The same source will also deliver ~ 5.5 Gy over 1 cm^2 at a depth of 70 μm, largely from beta radiation. This will produce a transient ulceration of the skin in about 10-50% of those exposed. The tissue damage resulting from the Ra-226 radiation exposure will thus be dominated by beta radiation, even taking into account alpha dose to thin skin regions of the body.

Cancer risks from alpha radiation

In my opinion the balance of evidence is against any causal link between alpha exposure and skin cancer[8,9]. The possible risk of skin cancer from alpha exposure of the skin is however subject to ongoing controversy. There is for example no proven link between skin cancer risk and alpha

[♠] The ICRP recommended ooccupational annual skin dose limit is 500 mSv, for the general public it is 50 mSv[2].

[∞] AEN is a term introduced especially to describe the unique superficial transient lesion produced by small area exposures from low energy beta sources such as Pm-147[3]. This type of minor lesion is not observed for higher energy beta sources which produce more severe responses due to dermal damage.

exposure from radon progeny in the environment. There is however a reported excess of skin cancer in Czech uranium miners subject to skin contamination with uranium dust. This has not been confirmed in any other miner study and there are possible confounding factors that cast doubt on the findings of this study – such as arsenic exposure in the mines (arsenic causes skin cancer) and preferential medical care that might have led to a reporting bias. Animal studies are distinctly lacking in support of alpha exposure of the epidermis being linked to skin cancer induction. Unfortunately there has not yet been any clear identification of the target cells for skin cancer. It is commonly thought that the basal cells of the epidermis are the target cells but in fact this remains unproven. Damage to the underlying deeper dermis of the skin appears to be an important factor in skin cancer induction in animals. The root of the hair follicle has been suggested as a target but this also remains as yet unresolved. Target cells in the hair follicles or superficial dermis would be beyond the range of naturally occurring alpha particles.

There are other factors which mitigate against alpha induced skin cancer. At high alpha doses it is likely that cells will be killed and thus incapable of subsequent expression of malignancy. Any calculation of skin cancer risk from small area contamination should also take account of the fact that cancer risk is proportional to the area exposed. ICRP skin cancer risk figures are for the exposure of the skin of the whole body, area ~ 20,000 cm^2. Cancer risk should be reduced according to the area exposed[14].

Appendix 1 discusses the evaluation of effective dose associated with a localized skin exposure from Ra-226, taking into account contributions from beta and alpha exposures and thin skin regions. 'Significant' Ra-226 sources can give an effective dose in an hour or less which is comparable with the calculated annual contribution to effective dose from radon progeny deposited on the skin from radon in air at elevated UK levels. Such evaluations of effective dose need to be treated with care. Effective dose is not strictly defined outside ICRP dose limits and at such high local absorbed skin doses from alpha radiation it is more likely that cells will be killed, thus preventing any induce mutations from proceeding to malignancy.

CONCLUSIONS

Skin absorbed doses for beta and photon emitters have been calculated for a bench mark 1 MBq point Ra-226 source using the code VARSKIN 3[4]. Alpha absorbed doses have been calculated using the code ALDOSE[5,6]. Skin absorbed doses for individual environmental sources can be assessed by applying pro-rata corrections to the bench mark calculations on the basis of measured activity levels and the extent to which equilibrium is achieved. Skin dose, averaged over an area of 1 cm^2, have been calculated at various tissue depths representative of the skin thickness at the majority of body sites. The dose estimates are also appropriate for small sources with other geometries, providing that self absorption is negligible. The calculated bench mark doses will thus be overestimates of actual doses from environmental samples, particularly for alpha radiation.

Ra-226 alone is primarily an alpha emitter (energy ~ 4.8 MeV) with a few low intensity and low energy beta and gamma emissions. The range of these alpha particles is ~ 30 μm, much less than the nominal skin thickness of 70 μm. The hazard posed by skin exposure from Ra-226 is therefore due primarily to its daughter products which include a number of high energy alpha, beta and gamma emissions. The major contribution to skin dose at the nominal skin depth of 70 μm from Ra-226 and its progeny is from beta radiation. The alpha dose to the skin at this depth is zero and gamma dose is negligible. Using VARSKIN 3 the total skin dose rate (70 μm, 1 cm^2) from a Ra-226 point source with an activity of 1 MBq (in equilibrium with all progeny) is ~ 5.5 Gy/hour. The photon absorbed dose rate to skin is ~ 72 mGy/h. The main contributors to skin dose rate are:

Pb-214: 2.01 Gy/h Bi-214: 1.81 Gy/h Bi-210: 1.63 Gy/h

The basal layer of the epidermis of the skin on some body sites for a proportion of the population may be less than the nominal value of 70 μm recommended by the ICRP. A range of depths of 20-100 μm covers the actual range of skin thicknesses found in a population over the majority of body sites. The basal layer on some body sites may thus be subject to exposure from the higher energy alpha particles from Ra-226 progeny – primarily Po-214 (7.7, MeV, range in skin ~ 64 μm). In the case of a 1 MBq Ra-226 source, in equilibrium with daughter products, an average skin absorbed dose rate at a depth of 20-100 microns from alpha radiations is about 65 Gy/hour (weighted by the skin thickness distribution in a population). It is necessary to be extremely careful in interpreting the significance of such high numerical values of dose rate. Actual doses may be very much less due to self absorption within the sample. The fact that equilibrium in actual measured samples is high indicates that the source is bound and sealed effectively against radon loss. This implies significant self absorption, particularly for alpha particles. This conjecture could be tested directly by contact dose measurements using Radiochromic dye film if required. These doses should not be compared directly with recommended dose limits which are based primarily on experience with photon irradiations which irradiate the full skin thickness. A literature search indicates that such large alpha doses have been observed to produce reddening (erythema) and pigmentation in human skin, but evidence is lacking to convincingly link such superficial alpha exposures with more severe detrimental deterministic effects such as ulceration, or with subsequent skin cancer.

Since the relevant contributions to skin dose from Ra-226 for radiation protection purposes are from beta and gamma radiations it is possible to categorise Ra-226 environmental samples in a similar way to that used by the Dounreay Particle Advisory Group (DPAG) for Dounreay fuel fragments (DFFs). Classification for DFFs was based on a consideration of skin dose rate (70 microns, 1 cm^2) likely residence time, and the probability of producing skin ulceration. ‘Significant’ radioactive sources were considered to pose a realistic potential of causing harm. This was equated with sources which produced skin dose rates greater than ~3 Gy/hour. ‘Relevant’ sources are those which produce an order of magnitude lower dose rate of ~ 0.3 - 3 Gy/hour but still warrant monitoring and removal from the environment. ‘Minor’ sources are those with lower skin dose rates. On this basis the 39 recently evaluated Ra-226 samples would be classified as:

2 significant 11 relevant 26 minor

Appendix 1 discusses the evaluation of effective dose associated with a localized skin exposure from Ra-226, taking into account contributions from beta and alpha exposures and thin skin regions. ‘Significant’ Ra-226 sources can give an effective dose in an hour or less which is comparable with the calculated annual contribution to effective dose from radon progeny deposited on the skin from radon in air at elevated UK levels. Such evaluations of effective dose need to be treated with care. Effective dose is not strictly defined outside ICRP dose limits and at such high local absorbed skin doses from alpha radiation it is more likely that cells will be killed, thus preventing any induce mutations from proceeding to malignancy.

APPENDIX 1

CONSIDERATION OF EFFECTIVE DOSE FROM DFFs AND Ra-226 SOURCES

DFFs

DPAG[12] have used a skin absorbed dose rate (70μm, 1 cm^2) of > 3 Gy/hour as the criteria for a 'significant' source in the case of DFFs. This is at a level near the threshold for just perceptible transient and superficial visible skin damage if the duration of exposure is about 1 hour. Exposure in this case is dominated by beta radiation. The contribution to effective dose from such an exposure is small since the proportion of the whole body skin that is exposed is small and the tissue weighting factor for skin is only 0.01.

The contribution to effective dose E from an absorbed dose D to skin of 3 Gy, delivered primarily by beta radiation, measured over 1 cm^2 at a depth of 70 μm, is given by:

E_{skin} = D x W_R x W_T x proportion of whole body skin exposed

Proportion of skin exposed ~ $\frac{1\,cm^2}{2x10^4\,cm^2}$

The effective dose associated with the DPAG category of 'significant' source is thus:

$$E_{skin} \approx \frac{3Gy\ x\ 1\ x0.01}{2\ x\ 10^4} = 1.5\ x\ 10^{-6}\ Sv \approx 1.5\mu Sv$$

Ra-226

In the case of 'significant' Ra-226 sources the same evaluation of a contribution to effective dose of 1.5 μSv can be made as for DFFs, neglecting any alpha radiation contribution since the alpha radiation does not irradiate the basal layer at the nominal depth of 70 μm recommended by the ICRP. A bench mark Ra-226 source with an activity of ~ 1 MBq will deliver this dose in 0.5 - 1 hours.

It is possible however to go beyond the general ICRP recommendations to explore the implications of non-standard assumptions such as thinner skin than the nominal 70 μm value. I have previously argued on the basis of an overall evaluation of the evidence that alpha radiation does not produce skin cancer[8,9] and should therefore not be included in an evaluation of effective dose. Even so, I will evaluate an effective dose for the purposes of comparison purposes. For a 0.6 MBq Ra-226 source, which in 1 hour delivers a beta absorbed dose to skin (70μm, 1 cm^2) of 3 Gy, the alpha absorbed dose to skin over 1 cm^2 at depths of 20-100 μm is ~ 40 Gy (weighted by the skin thickness distribution in a population). In this case the contribution to effective dose E is given by

$$E_{skin} \approx \frac{43\,Gy\ x\ 20\ x0.01}{2\ x\ 10^4} \approx 4\ x\ 10^{-4}\ Sv \approx 0.4mSv$$

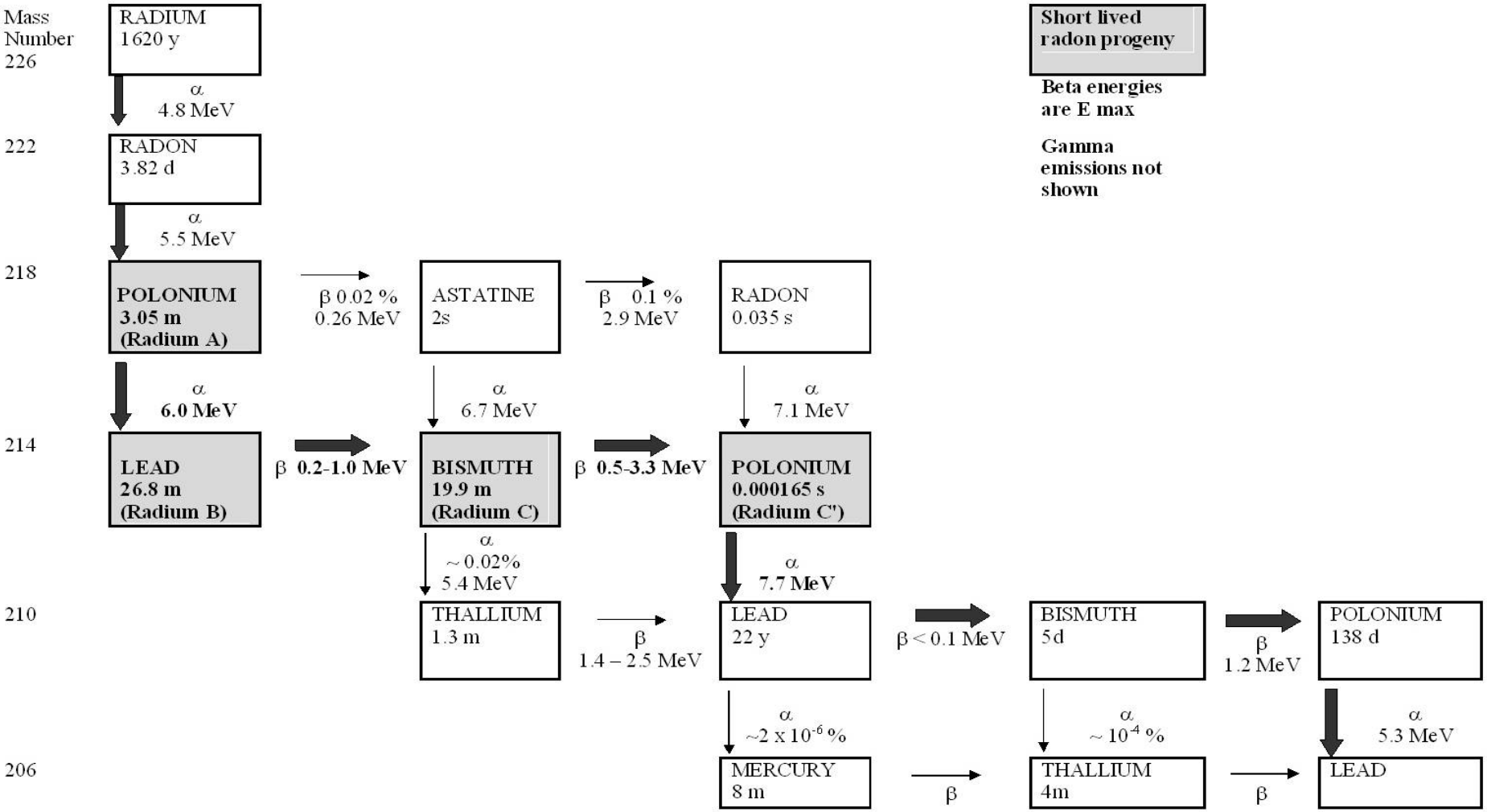
The radiation weighting factor W_R for alpha radiation for routine radiological protection situations is 20. A lower value of the RBE of 10 is considered more appropriate[9] but has not been used here.

For comparison purposes it is possible to consider skin dose from the decay products of radon in air. Radon progeny are deposited continuously onto the skin surface to maintain the decay chain from Po-218 onwards. The estimate of skin equivalent dose based on direct measurement of deposited radon progeny on people in the UK using TASTRACK detectors[15,16] is 22 (9-35) mSv, using $W_R = 20$[9]. This evaluation took into account the range of skin thickness across the body as shown in figure 5. Assuming that this exposure is to the whole body the contribution to effective dose can be derived by using the tissue weighting factor W_T for the skin of 0.01 and is ~ 0.1-0.4 mSv. Indoor and outside radon concentrations in this study were 20 and 7 Bq m^{-3} – similar to the average UK values. Exposure values several times this would have presumably been obtained in high radon areas in the UK.

It can thus be seen that the contribution to effective dose due to skin exposure from the highest activity Ra-226 sources found in the environment, delivered in about 1 hour, taking into account the exposure of thin skin areas to alpha radiation, is comparable to the contribution to annual effective dose due to skin exposure from natural radon in air at average UK levels (evaluated on the same basis). Several hours exposure from these high activity Ra-226 sources would give an effective dose similar to the annual effective dose due to skin exposure from natural radon in regions of elevated radon levels in the UK. The effective dose contribution from skin exposure is of course a small component of the total effective dose from all background radiation exposures - which on average in the UK is ~ 2.5 mSv/year. These evaluations of contributions to effective dose need to be treated with care because of all the provisos regarding the role of alpha radiation in skin cancer induction.

Table 1: Sampling data. Gamma ray spectrometry evaluation of total activity in 39 samples for the radionuclides Ra-226, Pb-214 and Bi-214

		Ra-226		Pb-214		Bi-214	
HPA Ref	NUVIA Ref	Bq/sample	± 2σ	Bq/sample	± 2σ	Bq/sample	± 2σ
08-6300	DB/08/001	122000	± 37000	109000	± 33000	102000	± 31000
08-6301	DB/08/002	27000	± 8100	22800	± 6900	24400	± 7400
08-6302	DB/08/003	33100	± 10000	28400	± 8600	28900	± 8700
08-6303	DB/08/004	5500	± 1700	4490	± 1400	4370	± 1400
08-6304	DB/08/005	315000	± 95000	278000	± 84000	268000	± 81000
08-6305	DB/08/006	624000	± 190000	592000	± 180000	619000	± 190000
08-6316	DB/08/007	313000	± 94000	257000	± 78000	252000	± 76000
08-6317	DB/08/008	187000	± 57000	159000	± 48000	152000	± 46000
08-6318	DB/08/009	90000	± 27000	81600	± 25000	78200	± 24000
08-6319	DB/08/010	3400	± 1100	3550	± 1100	4050	± 1300
08-6320	DB/08/011	13200	± 4000	8030	± 2500	3010	± 910
08-6321	DB/08/012	147000	± 45000	132000	± 40000	128000	± 39000
08-6322	DB/08/013	870000	± 270000	749000	± 230000	752000	± 230000
08-6323	DB/08/014	420000	± 130000	385000	± 120000	418000	± 130000
08-6324	DB/08/015	150000	± 45000	119000	± 36000	149000	± 45000
08-6325	DB/08/016	105000	± 32000	95000	± 29000	99000	± 30000
08-6326	DB/08/017	5300	± 1600	4300	± 1300	4900	± 1500
08-6327	DB/08/018	5000	± 1500	3040	± 920	1000	± 300
08-6328	DB/08/019	24000	± 7200	17900	± 5400	4200	± 1300
08-6329	DB/08/020	42000	± 13000	34400	± 11000	32000	± 9600
08-6330	DB/08/021	116000	± 35000	94000	± 29000	110000	± 33000
08-6331	DB/08/022	10000	± 3000	5600	± 1700	2070	± 630
08-6332	DB/08/023	4200	± 1300	3490	± 1100	3550	± 1100
08-6333	DB/08/024	36000	± 11000	30700	± 9300	32500	± 9800
08-6334	DB/08/025	8200	± 2500	6950	± 2100	7100	± 2200
08-6335	DB/08/026	2240	± 680	1420	± 430	510	± 160
08-6336	DB/08/027	2330	± 700	1480	± 450	440	± 140
08-6337	DB/08/028	920	± 280	580	± 180	210	± 63
08-6338	DB/08/029	75	± 23	48	± 15	17	± 6
08-6339	DB/08/030	201	± 61	120	± 36	45	± 14
08-6340	DB/08/031	97	± 30	61	± 19	22	± 7
08-6341	DB/08/032	109000	± 33000	90500	± 27000	97800	± 30000
08-6342	DB/08/033	11800	± 360	7060	± 2200	6950	± 2100
08-6343	DB/08/034	28500	± 8600	13400	± 4100	7460	± 2300
08-6344	DB/08/035	7300	± 2200	4540	± 1400	2010	± 610
08-6345	DB/08/036	3220	± 1000	2000	± 600	940	± 290
08-6346	DB/08/037	480	± 150	310	± 93	150	± 45
08-6347	DB/08/038	44000	± 14000	35700	± 11000	34800	± 11000
08-6348	DB/08/039	15000	± 4500	8680	± 2700	3000	± 900



The Uranium – 238 decay chain, from Ra-226 to Pb-206 (gamma emissions are not included)

Figure 1

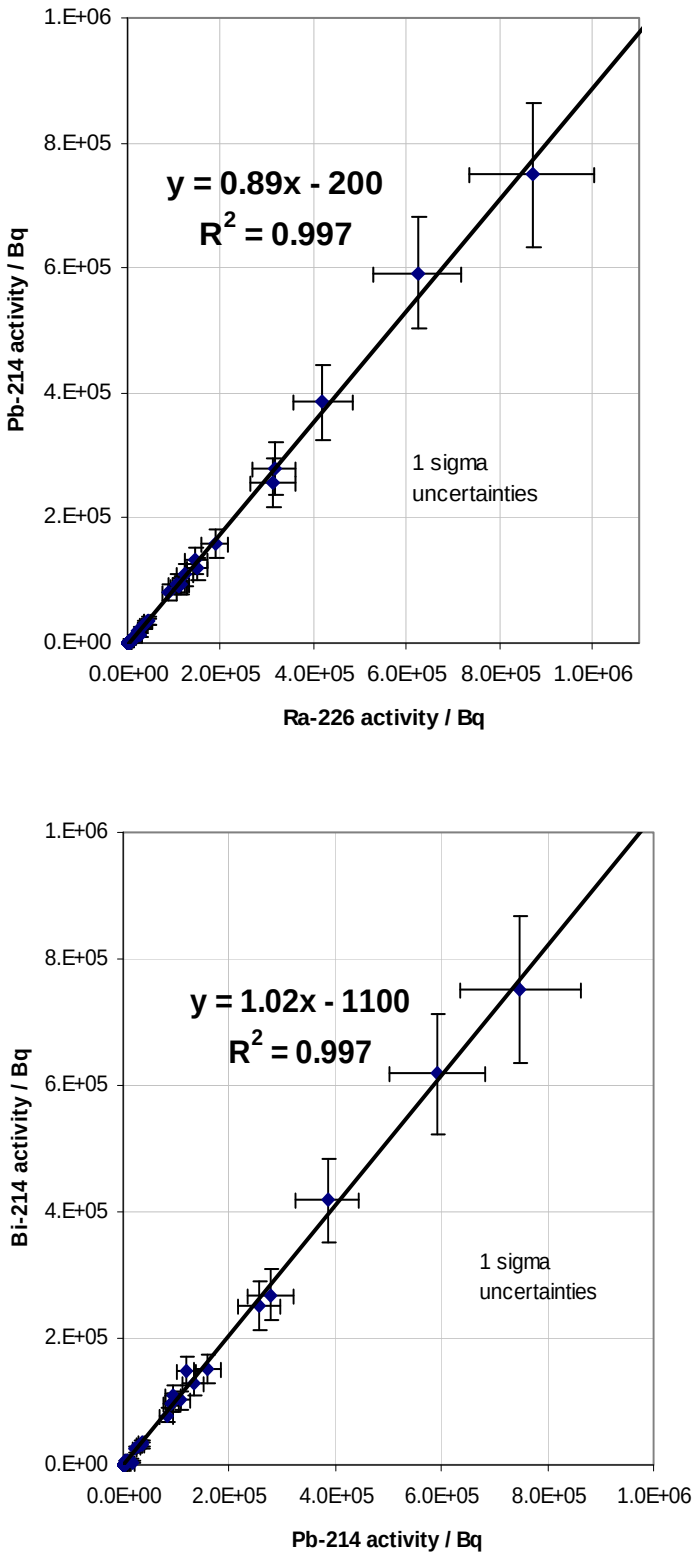


Figure 2. Correlation of the activity levels found in environmental samples. Top figure; Pb-214 is present in samples at a level of about 90% of the parent Ra-226. This is statistically significantly less that 100%. Bottom figure: The activity levels of Pb-214 and its daughter Bi-214 are closely correlated at levels which are not statistically significantly different.

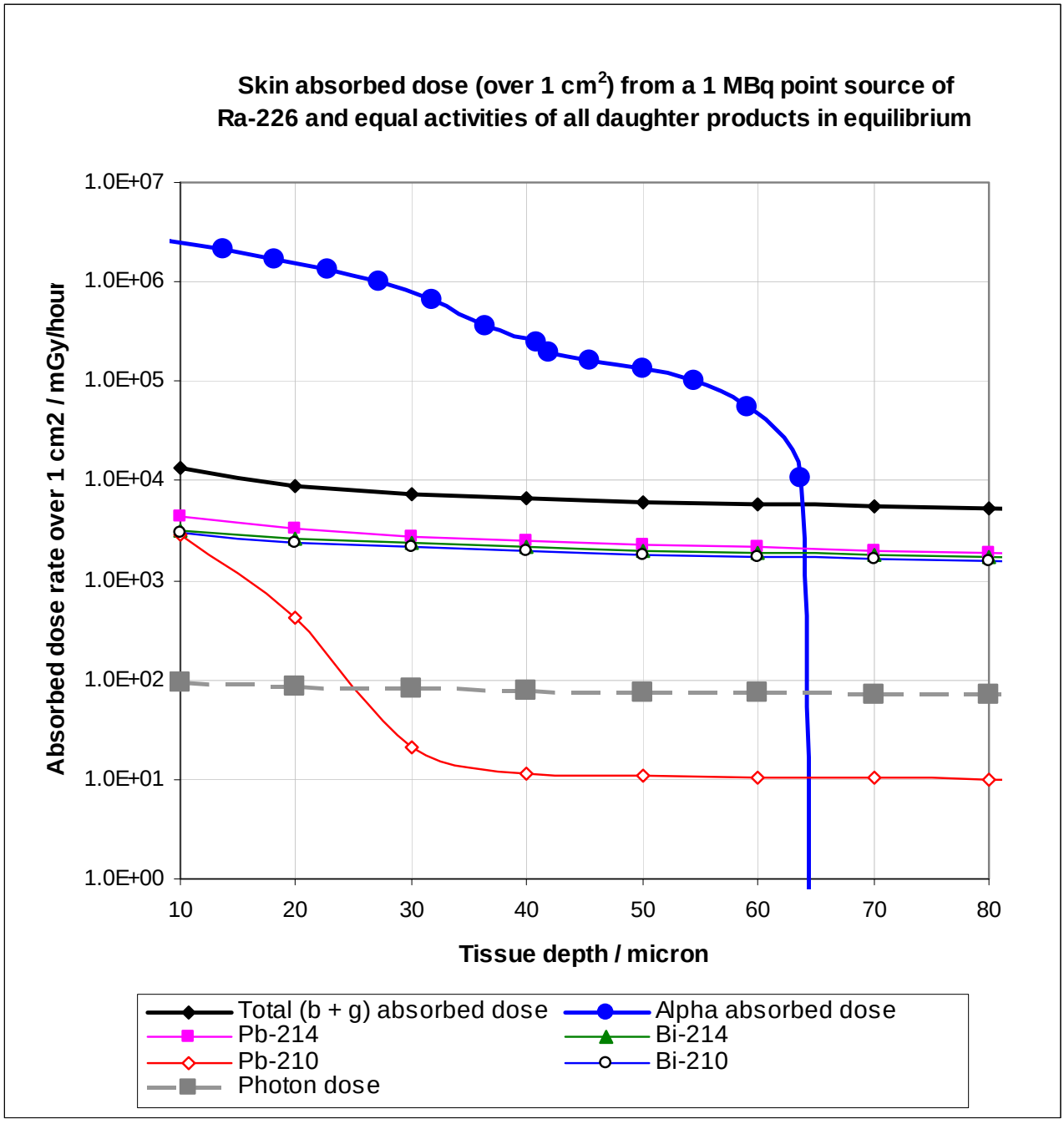


Figure 3. Absorbed dose (averaged over 1 cm^2) to skin at various depths from a 1 MBq point source of radium-226 in equilibrium with all its progeny (i.e. all products have activity of 1 MBq) Absorbed dose calculations assume a thin source at a semi-infinite tissue/air interface. Beta dose rates were evaluated using VARSKIN 3[4]. Alpha dose rates were calculated using ALDOSE[5,6]. A skin tissue density of 1,100 kg/m^3 was assumed

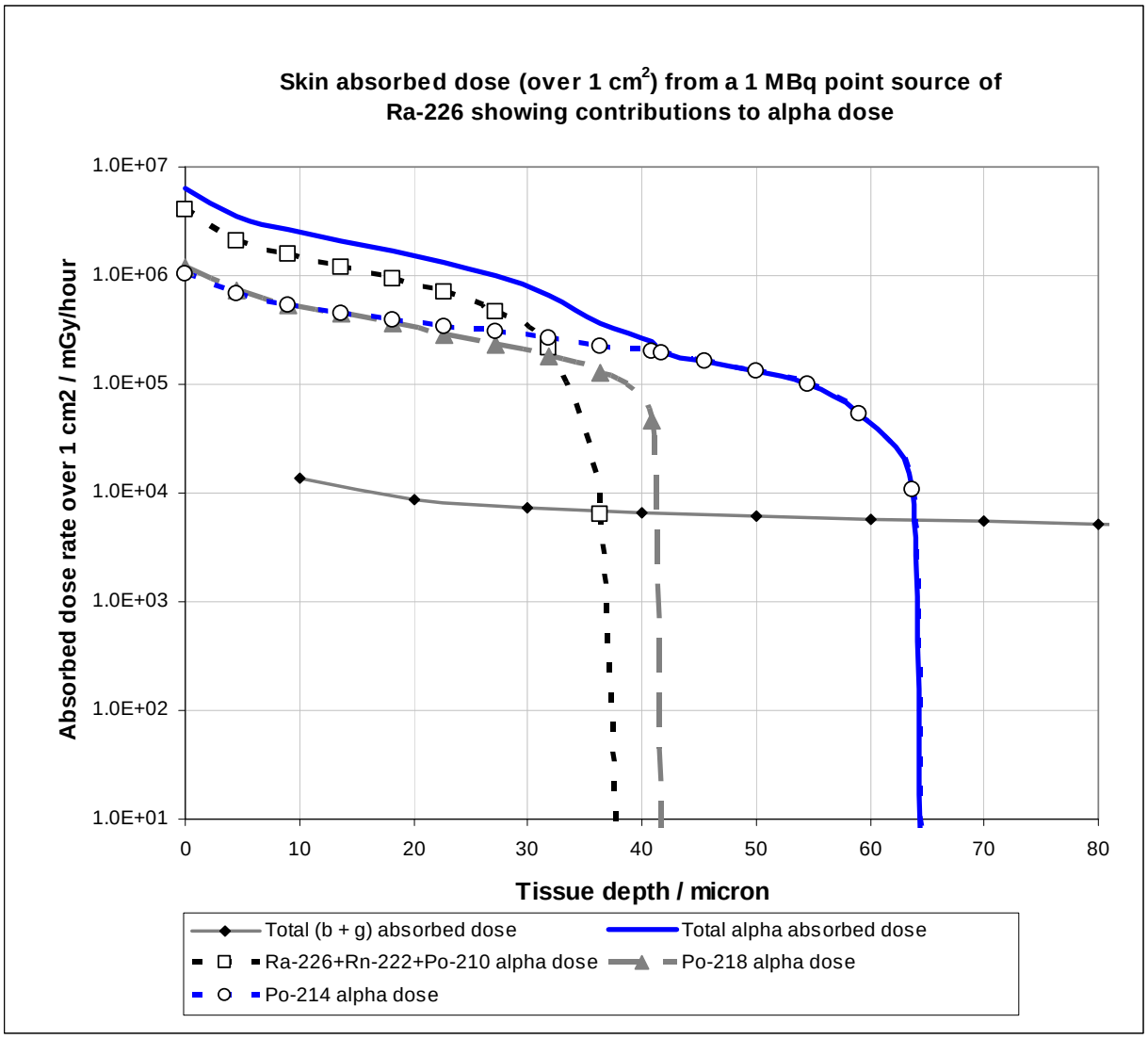


Figure 4. Absorbed dose (averaged over 1 cm^2) to skin at various depths from a 1 MBq point source of radium-226 in equilibrium with all its progeny (i.e. all products have activity of 1 MBq). Contributions to alpha dose to the skin are dominated by Po-214 (7.7 MeV). Alpha dose rates were calculated using ALDOSE[5,6]. A skin tissue density of 1,100 kg/m^3 was assumed.

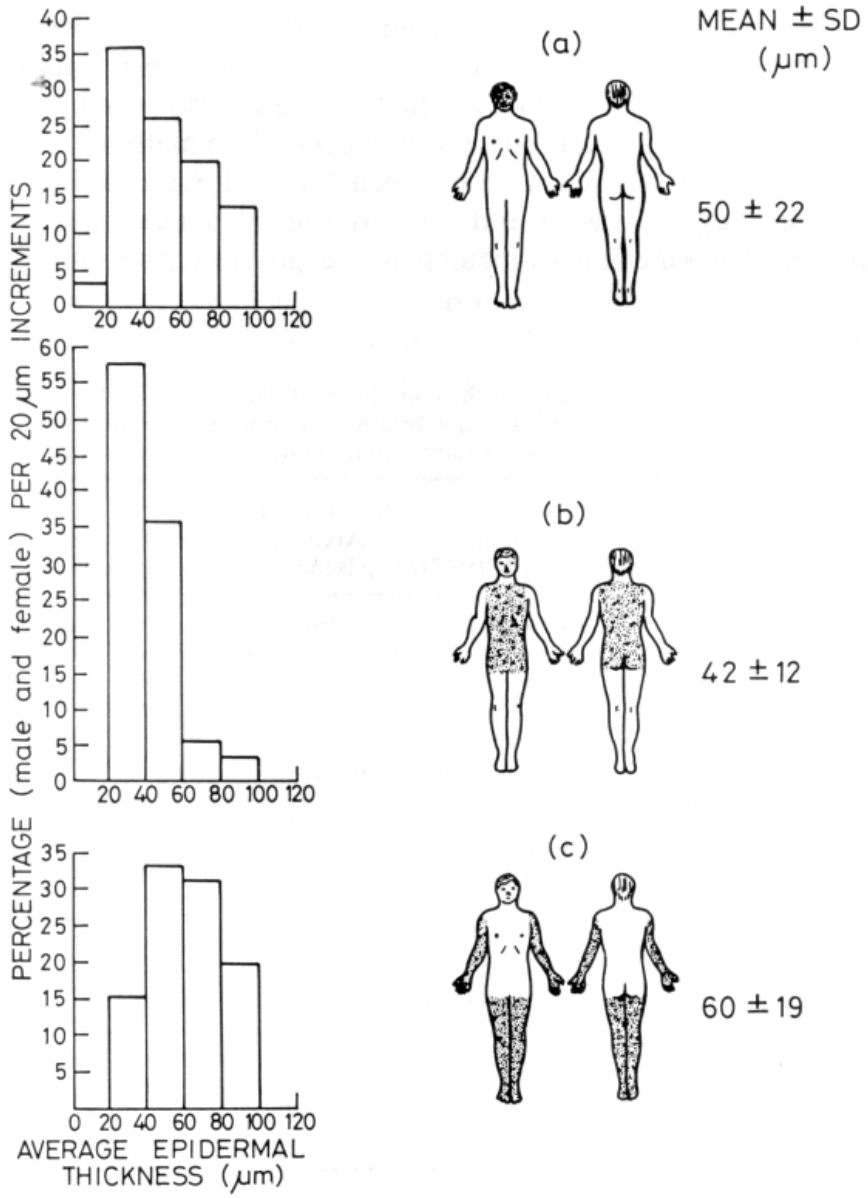


Figure 5. Variation in the distribution of the average epidermal thickness in man for three body regions (a) face (b) trunk (c) arms and legs (Whitton and Everall[17], ICRP[18]).

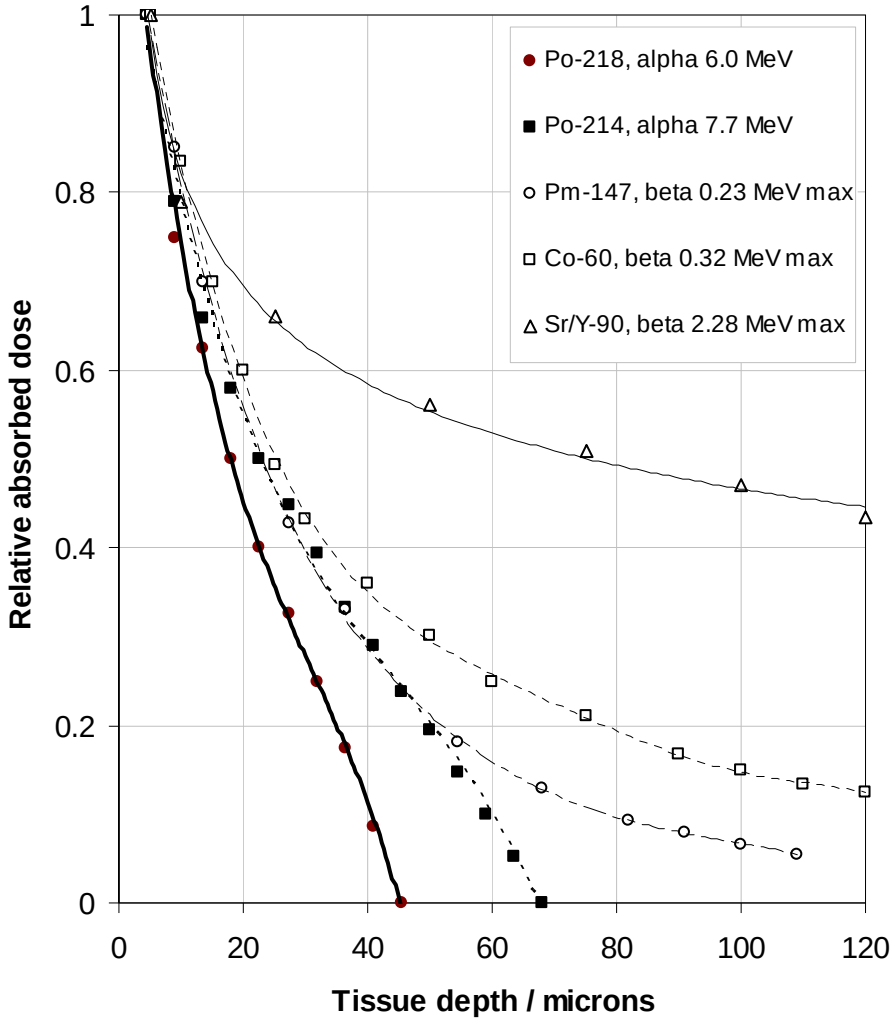


Figure 6. Relative depth doses from plane alpha and beta sources. Absorbed dose calculations assume a thin source at a semi-infinite tissue/air interface (relative to values at a depth of 5 microns). Alpha radiation depth-doses have been calculated using the code ALDOSE, and beta radiation depth-doses using the code VARSKIN. Near the skin surface the absolute absorbed dose rate (per unit activity per unit area) is a factor of ~ 100-500 less for beta compared with alpha sources, in accord with differences in LET.

Table 2. Sampling data. Samples are arranged in order of increasing Ra-226 activity. Categories of Minor, Relevant and Significant have been given on the basis of considerations of skin dose rate – as in the case of Dounreay fuel fragments[12].

HPA Ref	NUVIA Ref	Ra-226 Bq/sample
08-6338	DB/08/029	75
08-6340	DB/08/031	97
08-6339	DB/08/030	201
08-6346	DB/08/037	480
08-6337	DB/08/028	920
08-6335	DB/08/026	2240
08-6336	DB/08/027	2330
08-6345	DB/08/036	3220
08-6319	DB/08/010	3400
08-6332	DB/08/023	4200
08-6327	DB/08/018	5000
08-6326	DB/08/017	5300
08-6303	DB/08/004	5500
08-6344	DB/08/035	7300
08-6334	DB/08/025	8200
08-6331	DB/08/022	10000
08-6342	DB/08/033	11800
08-6320	DB/08/011	13200
08-6348	DB/08/039	15000
08-6328	DB/08/019	24000
08-6301	DB/08/002	27000
08-6343	DB/08/034	28500
08-6302	DB/08/003	33100
08-6333	DB/08/024	36000
08-6329	DB/08/020	42000
08-6347	DB/08/038	44000
08-6318	DB/08/009	90000
08-6325	DB/08/016	105000
08-6341	DB/08/032	109000
08-6330	DB/08/021	116000
08-6300	DB/08/001	122000
08-6321	DB/08/012	147000
08-6324	DB/08/015	150000
08-6317	DB/08/008	187000
08-6316	DB/08/007	313000
08-6304	DB/08/005	315000
08-6323	DB/08/014	420000
08-6305	DB/08/006	624000
08-6322	DB/08/013	870000

Minor

Relevant

Significant

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