

# Dalgety Bay – Hazard Assessment – Skin

*Forward by SEPA*

In 2009, SEPA published its preliminary assessment of the radioactive contamination at Dalgety Bay ([www.sepa.org.uk/radioactive\\_substances/publications/dalgety\\_bay\\_reports.aspx](http://www.sepa.org.uk/radioactive_substances/publications/dalgety_bay_reports.aspx)). This report includes an assessment of the potential hazards posed by the radioactive contamination present. The report indicates potentially widespread low level contamination at Dalgety Bay, which does not pose a significant risk to the public, together with high activity point sources of radium which do pose a significant hazard to members of the public using the beach at Dalgety Bay.

The 2009 report includes direct measurements of the potential doses should one of the point sources be ingested and reports that these doses could be of concern. However, the report does not include direct measurements of the potential hazard to the skin as no data was available at the time and thus SEPA was forced to use modelling to estimate the potential range of external doses.

Following the publication of the report, SEPA has sought to obtain real measured data on the potential skin doses and engaged a contractor to determine this in a similar way as SEPA assessed the potential external doses from contact with fragments of irradiated nuclear fuel at Dounreay. ([www.sepa.org.uk/radioactive\\_substances/publications/dounreay\\_reports.aspx](http://www.sepa.org.uk/radioactive_substances/publications/dounreay_reports.aspx)). The following report details the work undertaken and the conclusions from that work. When considering the findings of this work, it is important to note that the contamination at Dalgety Bay is considered in the radiation protection framework as an existing exposure situation. As such, the annual limit for a current work activity for skin exposure of 50 mSv does not apply. The relevant criteria for radioactive contaminated land are 10 Gray per hour. The report's findings should be considered in that context.

Following the 2009 report by SEPA, the Defence Estates has undertaken a programme of monitoring and recovery of radium sources at Dalgety Bay. Although that work has shown the continuing presence of high activity point sources at Dalgety Bay, SEPA been able to obtain a sample of those sources recovered which will provide further information on the potential hazards arising from them. The primary objective of this new work is to determine the potential consequences of ingesting a source. However, as recommended in this report SEPA has undertaken an initial evaluation of the alpha, beta and gamma emission for the skin dose rates, and will provide the results in a further report.

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## **Skin dose from Dalgety Bay Ra-226 contamination: Dose rate measurements for ten selected samples**

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## EXECUTIVE SUMMARY

Skin dose rates have been measured for 10 Dalgety Bay Ra-226 samples using radiochromic dye film (RDF) dosimetry. Specially constructed RDF stacks have provided estimates of absorbed dose rates in tissue at depths from 15 to 350  $\mu\text{m}$ , including depths of 15, 51 and 77  $\mu\text{m}$  which are relevant to concerns regarding a possible contribution from alpha particles to skin dose. In accord with ICRP recommendations skin doses have been averaged over the highest exposed area of  $1\text{ cm}^2$ .

The ten measured Dalgety Bay samples designated I-X have Ra-226 activities between 0.22 and 0.019 MBq. The measured skin dose rates are at least several times lower than calculated values based on Ra-226 activities for the individual particles, assuming point sources and 100% equilibrium of Ra-226 and progeny. For the 3 largest particles with dimensions of  $\sim 10\text{ mm}$  the measured dose rates at a depth of 77 $\mu\text{m}$  are between 35 -100 times less than calculated values. For smaller sources with dimensions up to  $\sim 3\text{ mm}$  the measured dose rates are between 3-7 times less than calculated values. These differences are in accord with evaluations of self absorption based on calculations for spherical samples with dimensions up to 10 mm and the range of observed densities 0.75-3.5  $\text{g/cm}^3$ .

The lack of any dramatic measured increase in skin dose rate at shallow depths ( $< 70\text{ }\mu\text{m}$ ) is indicative of a negligible contribution from alpha radiation with the full alpha energies expected from Ra-226 and its progeny (maximum alpha energy  $\sim 7.7\text{ MeV}$ , range  $\sim 65\text{ microns}$  in tissue). There is however an indication of a small contribution to skin dose from degraded alpha particles for the shallowest measurements at  $15 \pm 4\text{ }\mu\text{m}$ . For some of the samples the dose rate measured at the shallowest depth appears to be higher by perhaps a factor of up to  $\sim 5$  than the extrapolated underlying beta dose rate. This is compatible with a directly measured, though small, contribution at shallow depths from alpha particles to the total particle count rate from the samples. Crude alpha particle measurements made using a count rate meter for sample III gave an alpha particle range in tissue of  $\sim 25\text{ }\mu\text{m}$ , indicative of a severely degraded alpha particle spectrum for the small proportion of alpha particles emerging from the surface layer of the samples.

Of the 10 Dalgety Bay samples measured here the two with the highest Ra-226 activities (samples III & V; 217 and 93.4 kBq respectively) gave the highest dose rates at all depths. The highest dose rates, from sample III, were:

15 $\pm 4\text{ }\mu\text{m}$	1031 mGy/h
51 $\pm 4\text{ }\mu\text{m}$	214 mGy/h
77 $\pm 4\text{ }\mu\text{m}$	210 mGy/h
289 $\pm 4\text{ }\mu\text{m}$	121 mGy/h

Uncertainties in depth of  $\pm 4\text{ }\mu\text{m}$  represent the thickness of the radiochromic dye layer. Dose rates from sample V were 5-10% less than sample III. All other samples had significantly lower dose rates.

On the basis of these 10 Dalgety Bay samples the evaluation of skin dose for radiological protection purposes is pessimistic if based on calculated beta/gamma dose, using Ra-226 activities, assuming full radioactive equilibrium and neglecting self absorption. Alpha particles from these samples do not contribute significantly to skin doses of relevance to radiological protection.

It is recommended that the alpha particle emission rate is evaluated in future environmental surveys. The use of an alpha count rate meter in conjunction with conventional beta/gamma count and dose rate measurements should enable a rapid qualitative evaluation of the relative contribution of alpha radiation to skin dose rate.

## 1. INTRODUCTION

At the request of Paul Dale (SEPA) in 2006 one of us (MWC)<sup>1</sup> in the absence of any measured information on dose rates made an estimation using a model of the potential skin dose rates that would be delivered by small samples of 1 MBq Ra-226 in equilibrium with all progeny. No information on actual environmental activity levels or the extent of radioactive equilibrium was available at that time. These idealised bench-mark calculations were extended in 2008 using actual measurements of the activity levels in 39 environmental samples (see table 1) to provide a more definitive evaluation of potential skin dose rates<sup>2</sup>. Skin absorbed dose rates for beta and photon emitters were calculated using the code VARSKIN 3<sup>3</sup>. Alpha absorbed doses were calculated using the code ALDOSE<sup>4,5</sup>. Measured levels of Ra-226, Pb-214 and Bi-214 indicated progeny activities are about 90% of the equilibrium value. In the absence of information on particle size, composition, density etc., calculations were made which did not include potential self absorption within the particles (point source geometry was assumed). The skin dose rate was evaluated, averaged over an area of 1 cm<sup>2</sup> at various tissue depths representative of the skin thickness<sup>6</sup> at various body sites. These dose rate 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 of tissue equivalent material will very significantly reduce the alpha radiations considered in these preliminary pessimistic calculations.

The potential effects of self absorption on dose rate to the skin leads to undesirable uncertainties in the determination of the hazard posed by Dalgety Bay radium particles. Improved calculations require details of the size, shape, density and detailed composition and radionuclide content of Ra-226 samples. An alternative approach is to directly measure skin dose in contact with the samples. A similar task was undertaken in the context of radioactive particles in the environs of Dounreay, using radiochromic dye film<sup>6,7</sup>. The Dalgety Bay samples pose rather different problems to the Dounreay particles since they may give a significant contribution to skin dose from alpha radiations which are difficult to detect because of their short range. To provide a more realistic evaluation of the skin dose rate from the Dalgety Bay samples SEPA requested the direct measurement of skin dose rate using radiochromic dye film (RDF). The results of these measurements are presented in this report. A preliminary report was produced in November 2009 before completion of the dose rate and activity measurements<sup>8</sup>. The conclusions of that preliminary report remain unchanged.

The requirements of a direct skin dose measurement are best considered on the basis of an understanding of the possible contributions to skin dose from alpha, beta and gamma radiations. Some aspects of the previous calculated doses will therefore be discussed next.

## 2. ALPHA DOSE TO SKIN - THE NEED FOR MEASUREMENTS

In this and previous assessments of doses from Dalgety Bay radioactive samples calculations have been carried out of the absorbed dose rate (averaged over 1 cm<sup>2</sup>) 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). Beta dose rates were evaluated using VARSKIN 3<sup>3</sup>. Alpha dose rates were calculated using ALDOSE<sup>4,5</sup>. A skin tissue density of 1,100 kg/m<sup>3</sup> was assumed<sup>9</sup>. Such calculations are however subject to considerable uncertainty for a number of reasons.

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<sup>6</sup> Skin is subject to an equivalent dose limit based on exposure of the basal layer cells which are at the base of the epidermis of the skin. The depth of these basal cells is often loosely referred to in radiological protection as the skin thickness.

The skin hazard arising from Ra-226 alone is low since its radioactive emissions are 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 in tissue is ~ 30  $\mu\text{m}$ , much less than the nominal skin thickness of 70  $\mu\text{m}$ <sup>10</sup> recommended by the ICRP for general radiological protection purposes. However, the progeny of Ra-226 (see figure 1) include radionuclides with a number of high energy alpha particle emissions which extend the range of alpha exposure to about 64  $\mu\text{m}$ . they also include a number of high energy beta and gamma<sup>♥</sup> emissions which make the major contribution to skin dose at greater depths (see figures 2 and 3).

Using VARSKIN 3 the calculated total skin dose rate\* from a Ra-226 point source with an activity of 1 MBq (in equilibrium with all progeny) is ~ 5.5 Gy/hour at a depth of 70  $\mu\text{m}$ , averaged over an area of 1  $\text{cm}^2$ . The photon absorbed dose rate to skin is ~ 72 mGy/h. The main contributors to skin dose rate at this depth are:

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

Actual doses from extended sources will be reduced by self absorption of radiation within the source - which depends on the size, shape and density of the source ó factors which are rarely known with any certainty. The physical displacement of a proportion of radioactivity away from the skin for large samples also leads to reduced dose rates compared to small sources.

Alpha dose to the skin may not be negligible in some situations because the thickness of the skin on some body sites for a proportion of the population may be less than the nominal value of 70  $\mu\text{m}$  recommended by the ICRP. A range of 20-100  $\mu\text{m}$  covers the actual range of skin thicknesses found in a population over the majority of body sites<sup>5-9</sup>(see figure 4). ICRP publication 89<sup>9</sup> also includes tabulated reference values of epidermal skin thickness which are age dependent:

#### Reference values for thickness of the epidermis in males and females (ICRP publication 89)

Age	Thickness ( $\mu\text{m}$ )
Newborn	45
1 year	45
5 years	45
10 years	50
15 years	60
Adult	70

These values have however not been referred to in subsequent ICRP publications and their status/utility is unclear. The values fall within the recognized range of values in a mixed age/sex population and the veracity of an actual age dependence is questionable. It is clear however that in a general population, for some people, on some body sites, the skin may be thin enough to allow exposure of the basal layer from the higher energy alpha particles from Ra-226 progeny ó primarily Po-214 (7.7, MeV, range in skin ~ 64  $\mu\text{m}$ ).

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

\* For routine radiation protection the ICRP recommend the use of absorbed dose averaged over an area of 1 $\text{cm}^2$  in the region of the most exposed skin, measured at a depth of 70  $\mu\text{m}$ . Since epidermal skin density is ~ 1,100  $\text{kg}/\text{m}^3$  a linear skin thickness of 70  $\mu\text{m}$  corresponds to a mass thickness of ~ 7.7  $\text{mg}/\text{cm}^2$ . Mass thickness is a useful parameter since equal mass thicknesses of different absorbing materials have similar attenuation for beta and alpha radiations, particularly when the materials' compositions are similar.

In the case of 1 MBq Ra-226 point source, in full equilibrium with daughter products, a calculated 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)<sup>2</sup>. It is necessary to be extremely careful in interpreting the significance of such high numerical values of alpha dose rate. Actual values may be very much less due to self absorption within the sample. The fact that equilibrium in actual measured samples from Dalgety Bay is high indicates that the source is bound and sealed effectively against radon loss. This implies significant self absorption, particularly for alpha particles. Actual doses are very dependent on shape, size and density of the samples and the spatial distribution of radioactivity.

A literature search indicates that exposure to large alpha dose rates, such as those given above, for an hour or more have been observed to produce reddening (erythema) and pigmentation in human skin. However, 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 induced mutations from proceeding to malignancy<sup>11,12</sup>.

On the basis of the above considerations it is clear that the potential contribution to skin dose from alpha particles in the case of samples from Dalgety Bay should be assessed in order to circumvent the uncertainties associated with calculations. Any evaluation of health detriment from alpha doses to superficial tissues should be done with care. Following discussions with SEPA it was decided that direct skin dose measurements in the case of Dalgety Bay contamination should include evaluations of absorbed dose over an area of 1cm<sup>2</sup> at depths which are:

1. relevant to routine radiation protection  $\delta$  70  $\mu$ m
2. relevant to thinner skin  $\delta$  down to  $\sim$  45  $\mu$ m, in accord with ICRP publication 89
3. relevant to the thinnest recorded human skin values of  $\sim$  20  $\mu$ m
4. relevant to deep dermal damage<sup>11</sup> and alimentary tract damage<sup>11,13</sup>  $\sim$  300  $\mu$ m

### 3. SKIN DOSE RATE MEASUREMENTS

#### 3.1 Choice of Ra-226 samples for RDF dosimetry

Ten Dalgety Bay samples were chosen for RDF measurements, in conjunction with SEPA, from the 39 Ra-226 contamination samples previously discussed in an earlier report<sup>2</sup>. They include a range of activities, excluding the most active samples since these were considered to be a potential hazard from external whole body gamma radiation exposure. They are designated for this study by Roman numerals I-X, in order of decreasing activity. The chosen samples are indicated in table 2. The sample with HPA Ref 08-6316, was separated into 3 pieces but one of these was of very low activity and was discarded. The two remaining parts became samples II and III. Some of the other separated samples were also subject to minor break-up and this necessitated the re-measurement of the sample activities using gamma spectrometry (see table 2, column 10).

#### 3.2 Alpha/beta/gamma emissions. Initial evaluation using rate-meter measurements

A Thermo Fisher Scientific DP6AD dual phosphor alpha/beta probe, with an Electra 1A rate-meter, was used to measure alpha and beta count rates from the samples. The samples were sequentially placed in a 14 mm deep plastic sample tray and the count rate was evaluated by placing the 100 cm<sup>2</sup> probe area in contact with the top of the tray. According to the manufacturers specification the beta and alpha counting efficiencies are between 10-20%. Alpha and beta count rates are given in table 2,

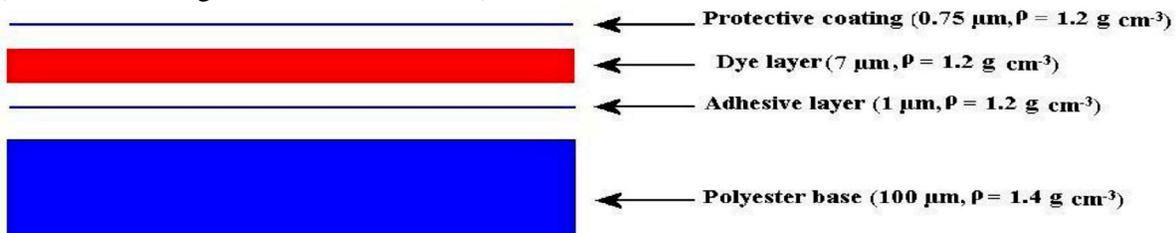
columns 6 & 7. The alpha count rate is at most a few % (range ~ 0.5 ó 4.5%) of the total (alpha + beta) count rate. In the case of the source with the highest alpha count rate (sample III) measurements were repeated using a sequential series of interposed plastic foils (Mylar film, 3.6  $\mu\text{m}$  thick). The alpha count rate reduced monotonically to background levels with the addition of ~ 5 films. This represents a range in tissue of about 25  $\text{m}$  ó rather less than the 65  $\text{m}$  range expected from Ra-226 and progeny. These crude semi-quantitative measurements indicate that the radiation emitted from the samples has a large beta component and only a small contribution from alpha radiation, and that the emitted alpha energy spectrum is degraded, presumably due to self absorption).

A Mini SmartIon dose rate meter (an air ion-chamber device) was used to evaluate the beta and gamma dose rates from the samples in the same way as for the alpha dose rate described above. The results are given in columns 8 & 9 of table 2. The dose rate ratio  $(\beta+\gamma)/\gamma$  was between 10-45.

In some cases the dose rate was dependent on particle orientation. For the subsequent RDF measurements the most active face of the sample was presented to the RDF films to maximize the recorded skin dose.

### 3.3 Depth-dose measurements using RDF film stacks

Depth-dose and spatial dose distribution measurements for 10 Dalgety Bay samples were carried out using the radiochromic dye film (RDF) technique<sup>7,14</sup>. The GAFChromic<sup>TM</sup> radiochromic dye film type HD-810 used in the present study has 4 layers: a 100  $\mu\text{m}$  thick polyester base and a surface 7  $\mu\text{m}$  radiation sensitive dye layer with 1  $\mu\text{m}$  and 0.75  $\mu\text{m}$  adhesive and protective coatings (see schematic figure below, not to scale).



The total mass thickness of one RDF sample is approximately  $15 \text{ mg cm}^{-2}$ . Radiochromic dye film develops a distinctive and characteristic blue colour upon exposure to ionising radiation. For HD-810 films the threshold dose is ~ 10 Gy. Above this threshold the optical density can be related to absorbed dose over a wide dose range up to several kGy. The dose range is extended by using red, green and blue image components. For doses from 5 Gy to 800 Gy films were calibrated against a standard ion chamber using a <sup>60</sup>Co teletherapy source and LINAC photons. For higher doses (800-5500 Gy) further sets of films from the same batch were exposed using a large area <sup>90</sup>Sr/<sup>90</sup>Y beta source (~ 5 GBq) cross calibrated in the lower dose range with photons.

Three types of RDF stacks were used to facilitate the measurement of doses in the superficial epidermis.

#### Type A Stacks

Depth dose measurements were carried out using stacks of 3 RDF films. These were mounted on Perspex blocks ( $5 \times 5 \times 1 \text{ cm}^3$ ) and covered with a thin plastic protective layer (4  $\mu\text{m}$  Mylar, density  $1.4 \text{ g cm}^{-3}$ ). The Ra-226 samples were mounted onto similar Perspex blocks using a thin layer of self adhesive tape and covered with a thin protective plastic film (4  $\mu\text{m}$  Mylar, density  $1.4$

g/cm<sup>3</sup>). The RDF stacks were inverted and placed onto the radioactive samples to facilitate exposure. In the case of large samples (I, IV & VIII, see figure 5) the two plastic blocks were separated at their edges by thin strips of polystyrene foam to maintain the blocks approximately parallel. Each RDF film was mounted with its sensitive layer uppermost, towards the radioactive sample.

The mass thickness between the radioactive sample and the middle of the first RDF dye layer, as described above, is:

$$(4.25 \text{ m} \times 1.2 \text{ gcm}^{-3}) + (8 \text{ m} \times 1.4 \text{ gcm}^{-3}) = 0.51 + 1.12 \text{ mgcm}^{-2} = 1.62 \text{ mgcm}^{-2}$$

The dose is thus measured within a 7 micron dye layer at a depth (in terms of mass thickness) of  $1.62 \pm 0.42 \text{ mgcm}^{-2}$

Assuming a density of  $1.1 \text{ gcm}^{-3}$  for the epidermis<sup>9</sup>, the dose is measured at an equivalent skin depth of  $(1.62 \pm 0.42 \text{ mgcm}^{-2}) / 1.1 \text{ gcm}^{-3} = 15 \pm 4 \mu\text{m}$ .

The two underlying RDF films in the stack will measure the dose at greater depths given by the equivalent thickness of the RDF film, i.e.

$$(8.75 \text{ m} \times 1.2 \text{ gcm}^{-3}) + (100 \text{ m} \times 1.4 \text{ gcm}^{-3}) = 1.05 + 14 \text{ mgcm}^{-2} = 15.05 \text{ mgcm}^{-2}$$

This is equivalent to a thickness of skin of 137 microns

The design of this stack thus measures the dose at depths of

$15 \pm 4 \mu\text{m}$

$152 \pm 4 \mu\text{m}$

$289 \pm 4 \mu\text{m}$

### **Type B stacks**

Type B stacks instead of having a  $4 \mu\text{m}$  layer of Mylar covering the 3 RDF films have a layer of  $50 \mu\text{m}$  of low density polyethylene (density =  $0.92 \text{ g cm}^{-3}$ ). This layer has an equivalent skin thickness of  $42 \mu\text{m}$ . Type B stacks thus measures dose at depths of:

$51 \pm 4 \mu\text{m}$

$188 \pm 4 \mu\text{m}$

$325 \pm 4 \mu\text{m}$

### **Type C stacks**

Type C stacks instead of having a  $4 \mu\text{m}$  layer of Mylar covering the 3 RDF films have a layer of  $80 \mu\text{m}$  of low density polyethylene (density =  $0.92 \text{ g cm}^{-3}$ ). This layer has an equivalent skin thickness of  $67 \mu\text{m}$ . Type C stacks thus measures dose at depths of:

$77 \pm 4 \mu\text{m}$

$214 \pm 4 \mu\text{m}$

$351 \pm 4 \mu\text{m}$

In order to provide skin dose estimates at the required superficial depths for each Ra-226 sample it was necessary to make 3 separate measurements using the 3 different stack types (A, B & C).

A limitation for the low activity Ra-226 samples is the high threshold dose of the RDF technique, which necessitated long exposure times of up to 18 weeks for the lowest activity sources. Samples were stored in the dark during these extended exposures to minimize errors due to the small light sensitivity of RDF. The RDF film stacks were periodically photographed *in-situ* (Figure 5) in order to decide when the exposure was adequate for evaluation. Figure 6 shows an *in-situ* photograph of an RDF stack type C on sample V and Figure 7 shows the RDF false colour image corresponding to figure 6 produced using a Reflecta 7200 film scanner to measure the optical density of the exposed film. The radial dose distribution software (RADODS)<sup>14</sup> was used to determine the distance of each optical density data point from the centroid of the optical density pattern for each irradiated film using a resolution of 300 dpi. RADODS uses calibration curves for red, green and blue images, to convert the measured optical density to absorbed dose  $\dot{D}$  on a pixel by pixel basis. This gives dose rate as a function of radial distance which enables the dose to be evaluated over any area of choice  $\dot{D}$  in this case 1 cm<sup>2</sup> for radiological protection purposes. The skin depth dose results are given in figure 8.

#### 4. DISCUSSION, CONCLUSIONS & RECOMMENDATION

Figure 8 gives calculated depth doses in the skin for point source Ra-226 samples (in equilibrium with progeny). Calculated doses are shown for samples with Ra-226 activities of 0.1 and 0.01 MBq. Below skin depths of ~ 70  $\mu\text{m}$  the predicted depth dose curves show a sharp increase due to alpha particle dose. For depths between 10-30  $\mu\text{m}$  the calculated alpha absorbed dose is more than 2 orders of magnitude greater than the beta absorbed dose. The dashed extrapolated lines represent calculations which exclude alpha absorbed dose. At depths greater than 70  $\mu\text{m}$  the calculated dose rates are dominated by beta radiation

Measured skin doses are also shown in figure 8 for the ten Dalgety Bay samples I-X which have Ra-226 activities between 0.217 and 0.019 MBq (see tables 2 & 3). The measured doses fall between and below the two calculated depth doses (dashed lines) for 0.1-0.01 MBq Ra-226, which include contributions only from beta and gamma radiations, excluding alpha dose. The measured doses are thus in general terms several times lower than calculated values based only on beta/gamma dose, assuming radioactive equilibrium and neglecting self absorption.

Figure 9 shows the dependence of calculated skin dose on sample size and density. It indicates the expected reduction in skin dose for actual radioactive samples compared with idealized point source calculations due to self absorption of beta radiation. Estimates of mass and size have been recorded by SEPA<sup>15</sup> for radium contamination samples at Dalgety Bay. This indicates that most of the samples used in this study have densities between 0.75-3.5 g/cm<sup>3</sup>. Since the samples measured here had dimensions of several mm, reductions in skin dose rate by factors of 5-10 or more due to self absorption would thus be expected in comparison with calculations for point sources. Table 3 compares measured and calculated (beta+gamma) dose rates at a tissue depth of 77 microns for all 10 samples. As expected, measured values are significantly less than the calculated values based on point source geometry, particularly for the larger sources. Similar differences are also obviously seen for the shallowest depths where alpha dose is significant. For the 3 largest particles with dimensions of ~ 10 mm the measured dose rates at a depth of 77 $\mu\text{m}$  are between 35 -100 times less than calculated values. For smaller sources with dimensions up to ~ 3mm the measured dose rates are between 3-7 times less than calculated values.

More detailed comparisons between measured and calculated dose rates have not been carried out since this would be an inappropriate over-interpretation of the data. Dose rates depend on particle shape, density, and spatial distribution of activity within the sources  $\dot{D}$  none of which are known in detail.

An obvious question is ó are there any indications of an alpha component to skin dose from the samples I-X? For most of the samples the dose rate measured at the shallowest depth ( $15 \pm 4 \mu\text{m}$ ) appears to be higher by perhaps a factor of up to  $\sim 5$  than the extrapolated underlying beta dose (dashed lines, figure 8). This is indicative of an alpha dose component for superficial depths. This is compatible with the directly measured, though small, observed alpha contribution to particle count rate from the samples (table 2). Since RDF slightly under-responds<sup>16</sup> to alpha particles of this energy ó by 0.8 compared to low LET radiations (beta & gamma) ó the alpha dose enhancement may be somewhat higher. The measured doses at a depth of  $51 \pm 4 \mu\text{m}$  show little sign of a significant alpha dose enhancement. This is in accord with the crude alpha particle range measurement made for sample III\* of  $\sim 25 \mu\text{m}$ .

The RDF measurements reported here were time consuming - partly due to the low inherent sensitivity of RDF and also in order to provide definitive alpha dose estimates for all samples over a range of depths. A higher sensitivity alpha dose measurement system for the skin is desirable but does not currently exist. A crude but rapid evaluation of the rate of alpha particle emission and alpha range could be carried out using a rate meter as described here. This would quickly determine whether there was a significant radiological skin hazard. No alpha count rate assessments appear to have been carried out in early surveys. Since this is a relatively easy procedure it is recommended that this be done in future surveys.

Of the 10 Dalgety Bay samples measured here the two with the highest Ra-226 activities (samples III & V; 217 and 93.4 kBq respectively) gave the highest doses at all depths. The highest dose rates, from sample III, were:

15 $\pm 4 \mu\text{m}$	1031 mGy/h
51 $\pm 4 \mu\text{m}$	214 mGy/h
77 $\pm 4 \mu\text{m}$	210 mGy/h
289 $\pm 4 \mu\text{m}$	121 mGy/h

Dose rates from sample V were 5-10% less than sample III. All other samples had significantly lower dose rates (figure 8).

On the basis of these 10 Dalgety Bay samples the evaluation of skin dose for radiological protection purposes is pessimistic if based on calculated beta/gamma dose, using Ra-226 activities, assuming full radioactive equilibrium and neglecting self absorption. Alpha particles from these samples do not contribute significantly to skin doses of relevance to radiological protection.

It is recommended that the alpha particle emission rate is evaluated in future environmental surveys. The use of an alpha count rate meter in conjunction with conventional beta/gamma count and dose rate measurements should enable a rapid qualitative evaluation of the relative contribution of alpha radiation to skin dose rate.

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\* See earlier section - Alpha/beta/gamma emissions ó an initial evaluation using rate-meter measurements

## Tables and figures

**Table 1: Sampling data. Gamma ray spectrometry evaluation of total activity in 39 samples for the radionuclides Ra-226, Pb-214 and Bi-214. These measurements were made prior to RDF dose assessment – which required selection, separation and mounting. In the majority of cases this led to reductions in activity. The revised activities for the selected samples are given in column 10 of table 2.**

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

**Table 2. Dalgety Bay: 39 Samples arranged in order of increasing Ra-226 activity.**

The samples selected for dose measurements are given in columns 4 & 5. Alpha and beta count rates (columns 6 & 7) were measured with a Thermo Electra DP6AD (S/N 2488) dual phosphor alpha/beta probe. Beta and gamma dose rates (columns 8 & 9) were measured with a SmartIon dose rate meter (REMS GLA 34368, S/N 002264). Re-measured Ra-226 activities are given in column 10.

HPA Ref	NUVIA Ref	Original Ra-226 activity /Bq	Selection priority	Source ID for this work	$\alpha+\beta$ kcps	$\alpha^{\#}$ cps	$\beta+\gamma^{\$}$ $\mu\text{Sv/h}$	$\gamma$ $\mu\text{Sv/h}$	Revised Ra-226 activity/Bq
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	10	X	3.15	16	47	3.5	18800
08-6328	DB/08/019	24000							
08-6301	DB/08/002	27000							
08-6343	DB/08/034	28500	9	IX	1.0	15	20	1.7	Not measurable
08-6302	DB/08/003	33100	8	VIII	2.4	25	42	4.2	21500
08-6333	DB/08/024	36000							
08-6329	DB/08/020	42000							
08-6347	DB/08/038	44000	7	VII	4.9	61	71	4.2	29600
08-6318	DB/08/009	90000	6	VI	18.2	255	225	10	79000
08-6325	DB/08/016	105000							
08-6341	DB/08/032	109000							
08-6330	DB/08/021	116000							
08-6300	DB/08/001	122000	5	V	28.3	650	700	15.3	93400
08-6321	DB/08/012	147000	4	IV	11.2	145	215	14	114000
08-6324	DB/08/015	150000							
08-6317	DB/08/008	187000							
08-6316	DB/08/007	313000	2 & 3	II, III	10.7 41.3	450 1480	245 960	9 38	4820 217000
08-6304	DB/08/005	315000	1	I	13.0	50	255	28	92500
08-6323	DB/08/014	420000							
08-6305	DB/08/006	624000							
08-6322	DB/08/013	870000							

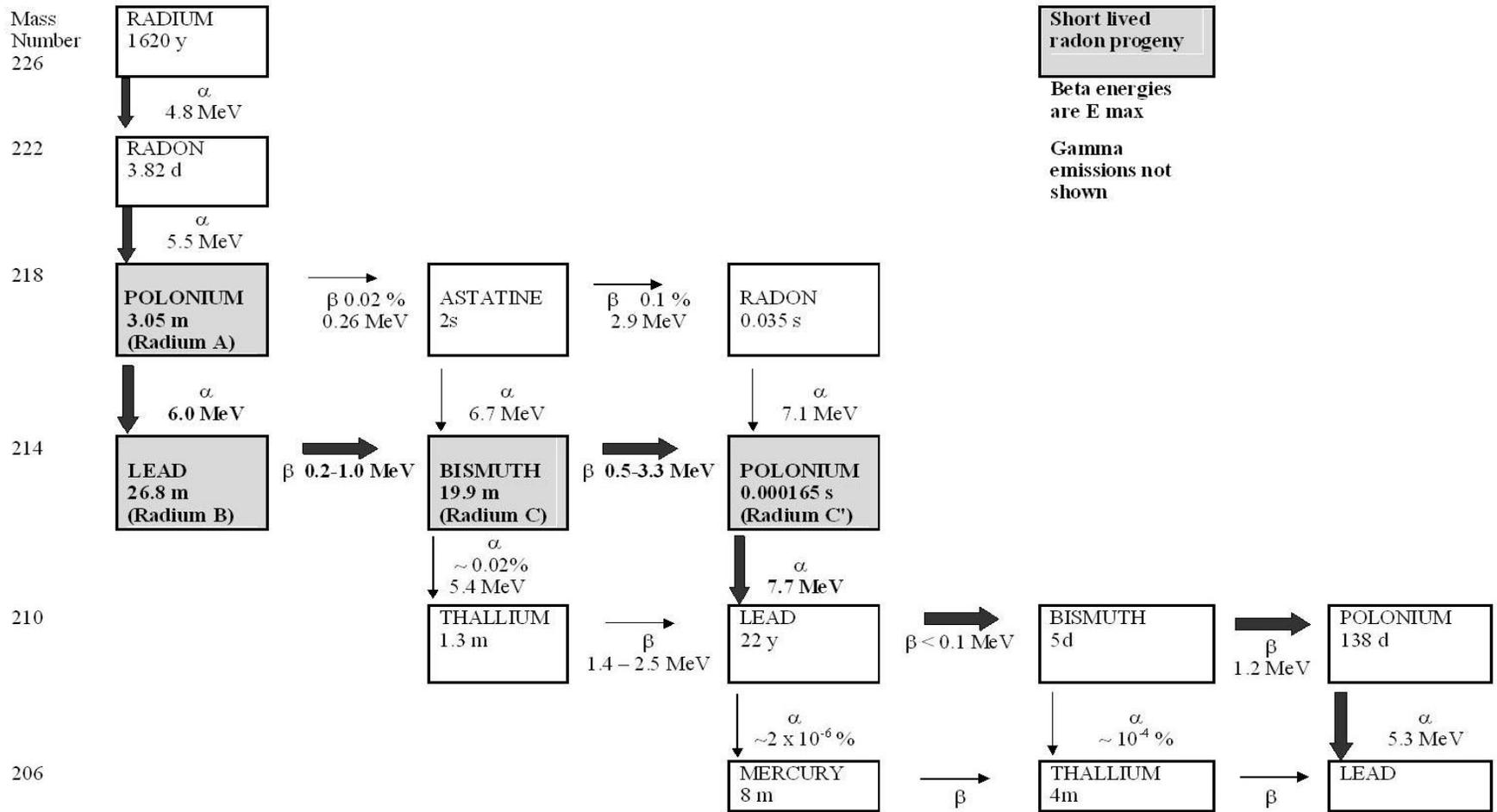
N.B. # alpha background ~ 8 cps. \$ Beta + gamma dose rate background ~ 1  $\mu\text{Sv/h}$

**Table 3. Comparison of calculated and measured (beta +gamma) dose rates\* at a depth of 77 microns. Absorbed doses were measured with RDF 'C' stacks. Calculated values are for point sources. The size of Dalgety Bay samples was crudely categorised into large (L), medium (M) and small (S) on the basis of linear dimensions of about 10, 3 and 1 mm respectively.**

Sample	I	II	III	IV	V	VI	VII	VIII	IX	X
Ra-226/Bq (Re-measured after dose measurements)	92500	48200	217000	114000	93400	79000	29600	21500	NA#	18800
1 sigma uncertainty on Ra-226 activity/Bq	14000	7500	33000	17500	14500	12000	4450	3250	NA	2850
Calculated/Measured dose rate at 77 microns %	35	5	6	41	3	4	7	100	NA	6
Size (Large/Medium/Small)	L	M	M	L	M	M	M	L	M	S

\* Doses averaged over an area of 1 cm<sup>2</sup>

# Not available - activity too low for measurement



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

Figure 1

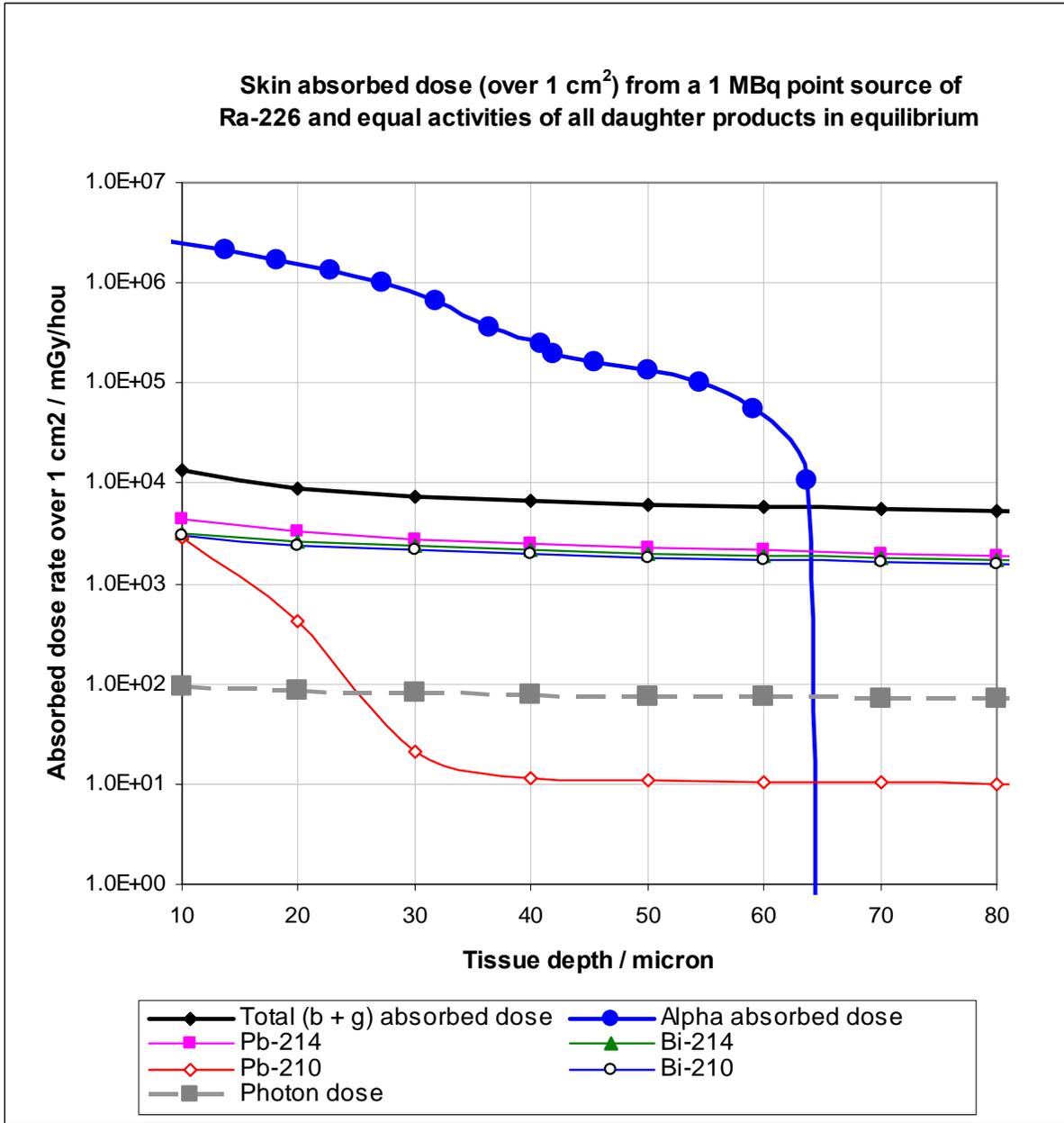
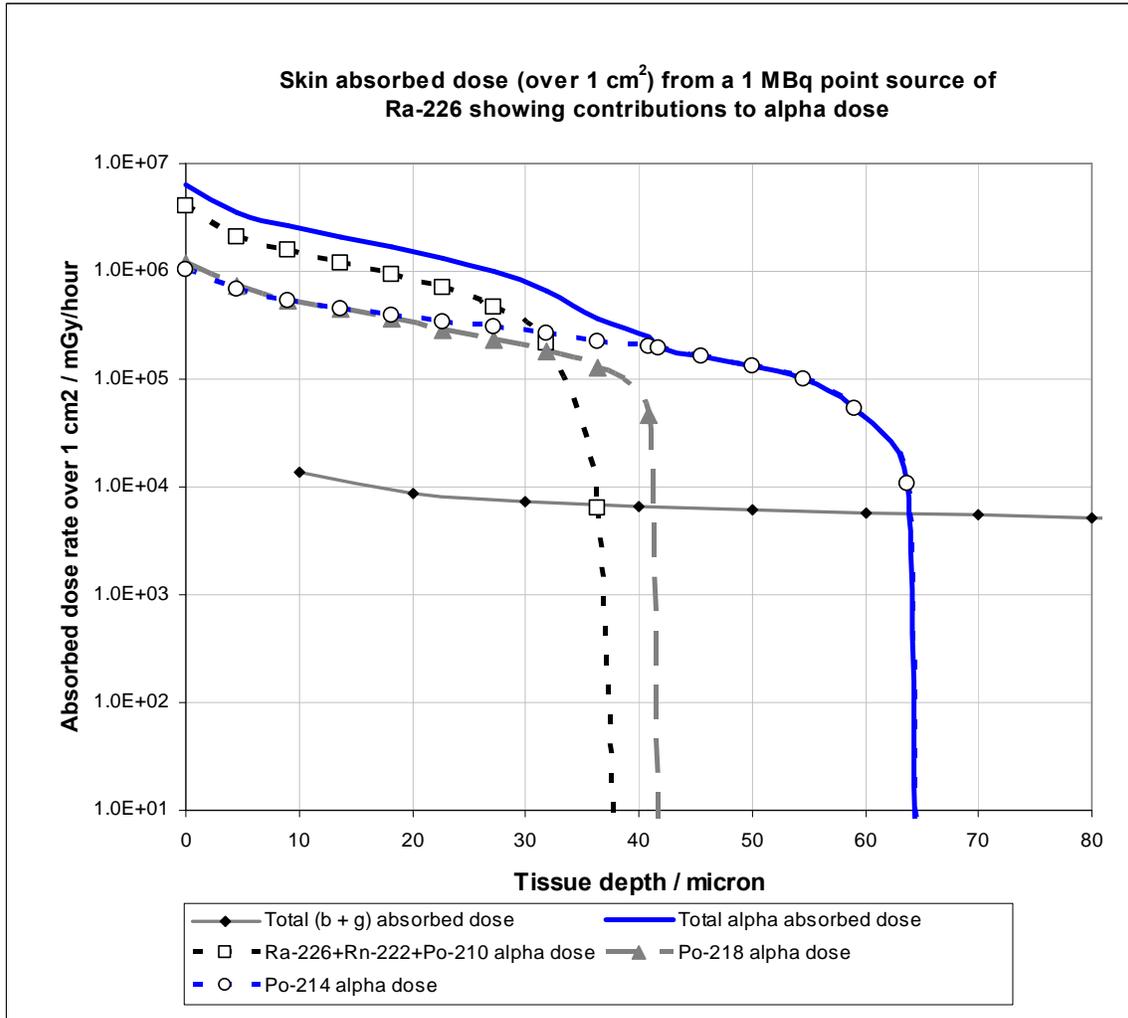
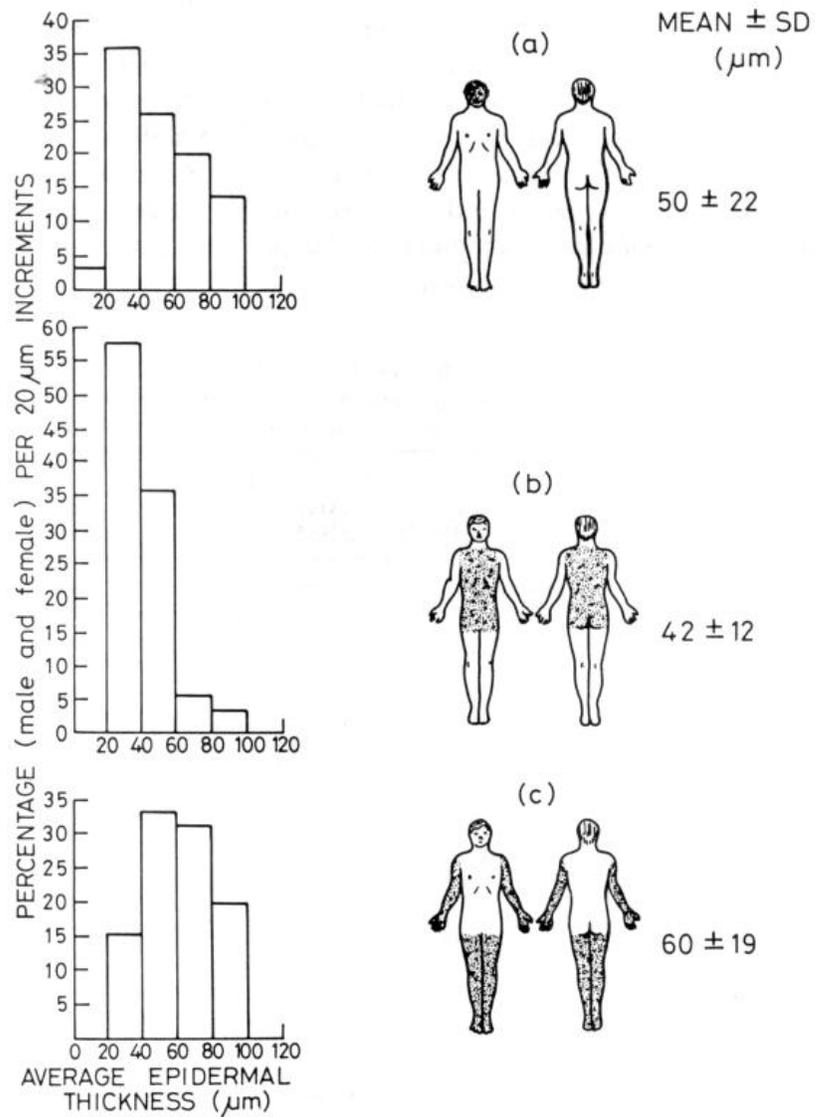


Figure 2. Calculated absorbed dose rates (averaged over 1 cm<sup>2</sup>) to skin at various depths from a 1 MBq point source of radium-226 in equilibrium with all its progeny (i.e. all products have activity of 1 MBq). Beta dose rates were evaluated using VARSKIN 3<sup>3</sup>. Alpha dose rates were calculated using ALDOSE<sup>4,5</sup>. A skin tissue density of 1,100 kg/m<sup>3</sup> was assumed. The various contributions to beta dose from Pb-210/214 and Bi-210/214 are indicated.



**Figure 3. Calculated absorbed dose rate (averaged over 1 cm<sup>2</sup>) 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<sup>4,5</sup>. A skin tissue density of 1,100 kg/m<sup>3</sup> was assumed.**



**Figure 4. 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<sup>17</sup>, ICRP<sup>9,18</sup>).**

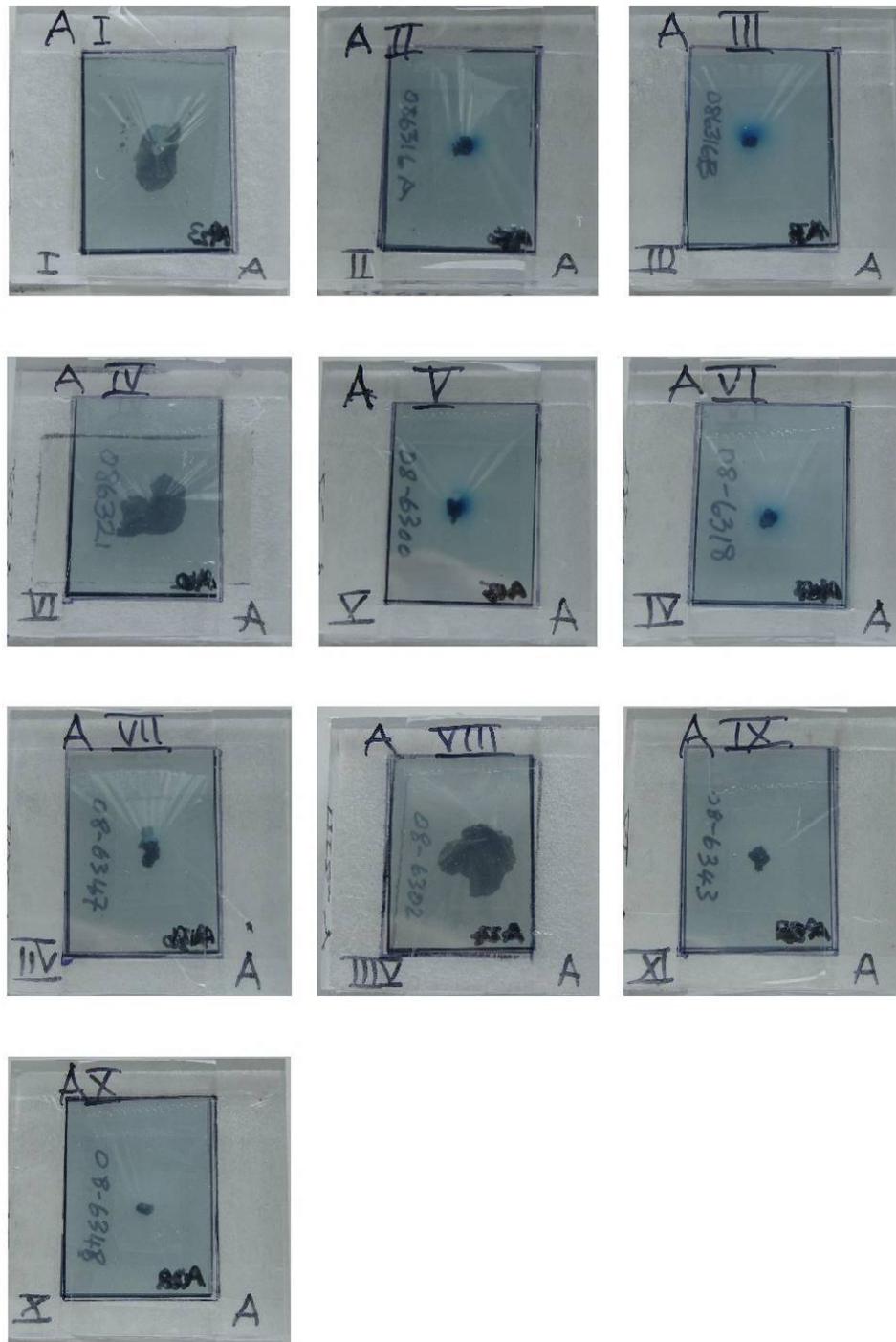
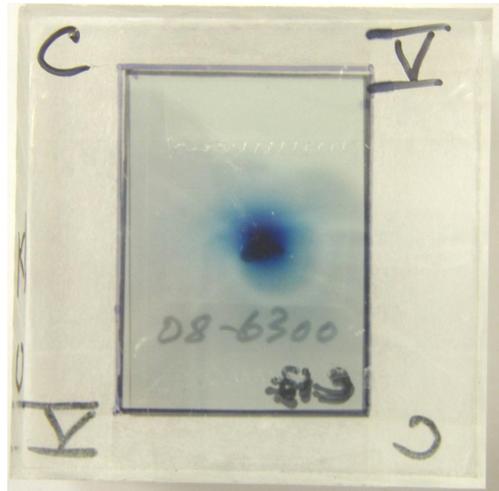
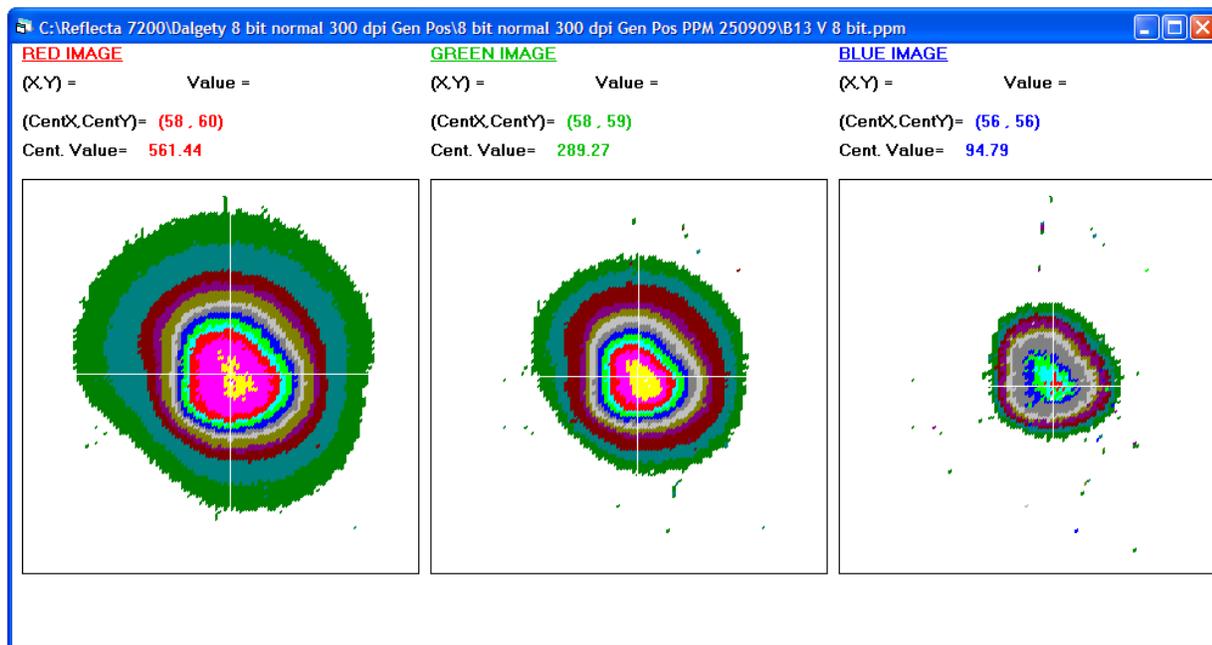


Figure 5. Montage of photographs (taken *in-situ* during exposure) of type A stacks on Perspex blocks in contact with radioactive samples, 48 hours after start of exposure. RDF stacks are uppermost (nearest to camera). Roman numeral designations of the samples are shown at the top of each image. Block dimensions 5 x 5 x 1 cm<sup>3</sup>. RDF films dimensions 36 x 27 x 0.1 mm<sup>3</sup>. Samples I, IV & VIII which had dimensions ~ 1cm required the Perspex blocks to be supported at the edges with polystyrene foam strips to maintain parallel geometry.



**Figure 6. Photograph of an RDF film stack used for depth dose measurements for Ra-226 sample V. The stack design is in this case type C. The RDF films on one Perspex block and the Ra-226 sample on the other Perspex block are both covered by a thin plastic film. For type C stacks the thicknesses of the plastic film was chosen so that the dose measured in the sensitive layer of the first RDF film is at an equivalent tissue depth of  $77 \pm 4 \mu\text{m}$ . For stack types A and B the plastic film thicknesses was chosen so that the doses measured in the first RDF film are at equivalent tissue depths of  $15 \pm 4$  and  $51 \pm 4 \mu\text{m}$  respectively. If there is a significant alpha dose from Ra-226 this should be seen as a very marked increase in the surface dose for stack type A.**



**Figure 7. False colour images produced in the dosimetry/image analysis program RADODS for the RDF film shown in figure 6. The centroid position is given by the cross wires. Red, green and blue images provide coverage of different dose ranges.**

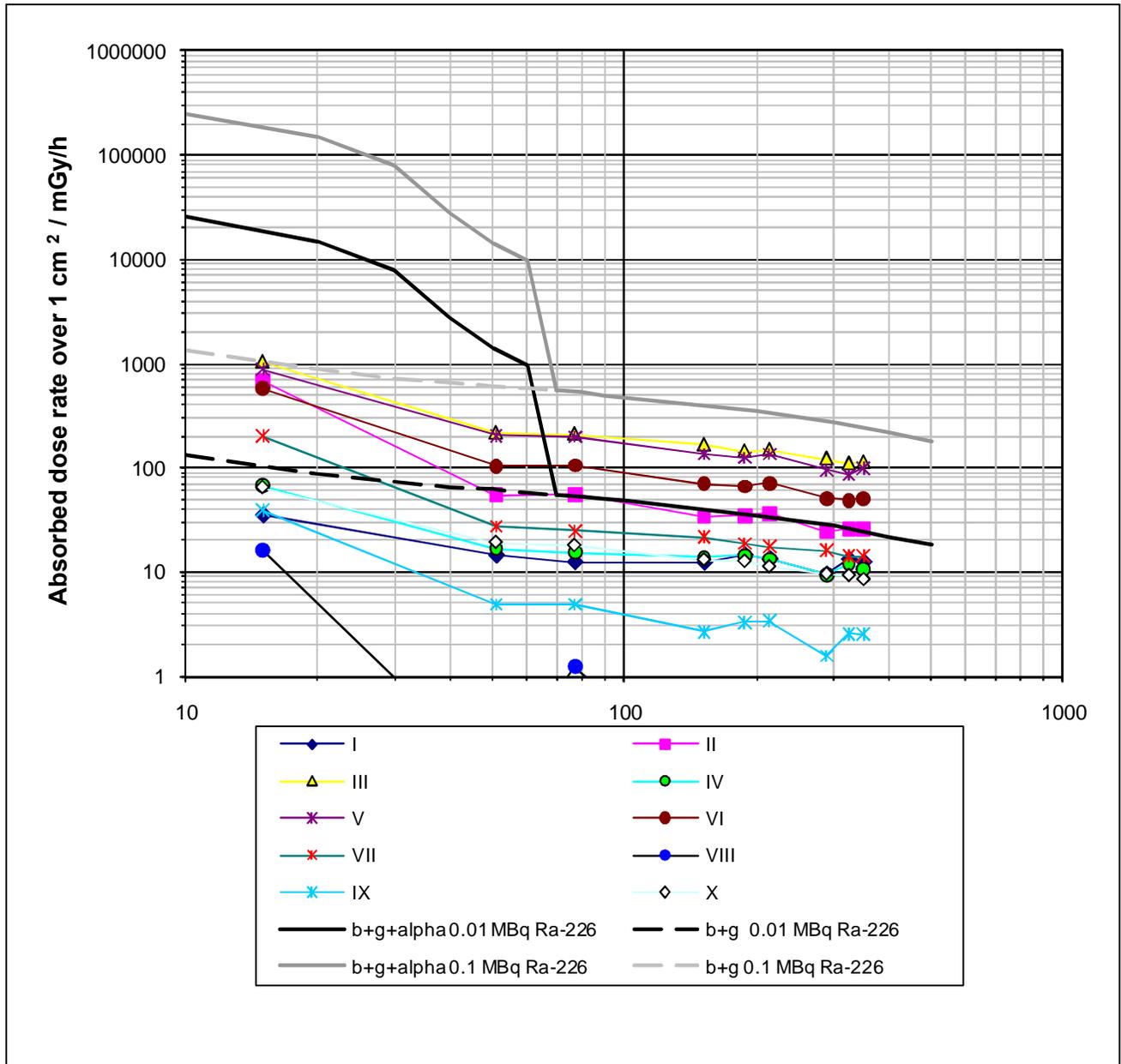
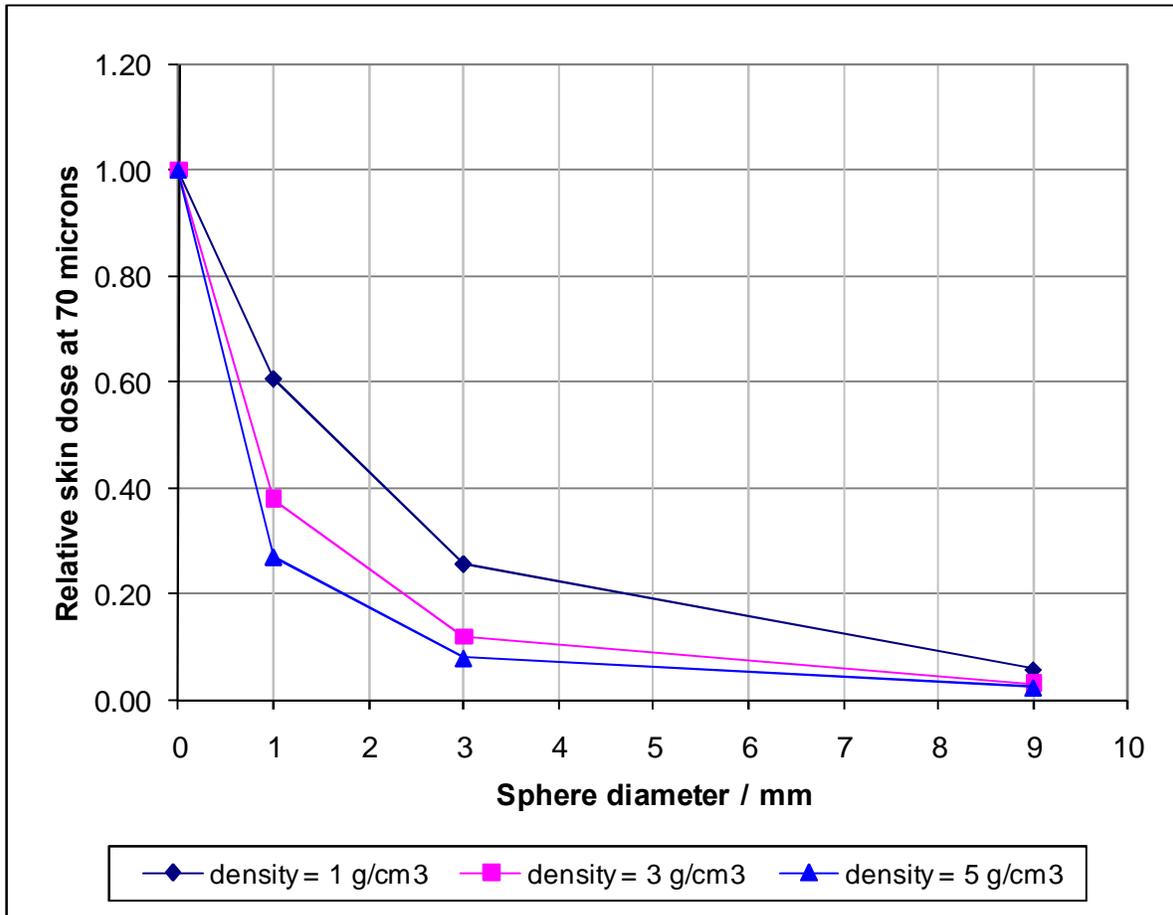


Figure 8. Measured depth dose rates are shown for the 10 Dalgety Bay samples I-X. Calculated values are also indicated for point sources of Ra-226 of activity 0.1 and 0.01 MBq. Calculated doses are given for beta (b) + gamma (g) and b + g + alpha radiations. Alpha doses have been calculated using the code ALDOSE, and beta/gamma radiation doses using the code VARSKIN 3. Skin doses are average values over an area of 1 cm<sup>2</sup>. A skin tissue density of 1,100 kg/m<sup>3</sup> was assumed. Measured dose rates of a few mGy/hour were on the borderline of statistical significance for even the most prolonged exposure times used here of ~ 3,000 hours.



**Figure 9. Calculated dependence of skin dose rate (70 μm, 1 cm<sup>2</sup>) on particle size and density. Only beta dose is included. A sphere of uniform density has been assumed. Calculations used VARSKIN 3.**

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