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**ATMOSPHERIC WASHOUT OF RADON SHORT-LIVED DAUGHTERS:
IMPLICATIONS FOR THE RADIATION DOSE TO THE SKIN**

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Summary

Plastic alpha-particle track detectors have been used to measure the surface deposition (plateout) of the ^{222}Rn radioactive short-lived decay product aerosols ^{218}Po and ^{214}Po outdoors. Unusually high average levels of activity are recorded, with highest values up to 120 Bq m^{-2} . This is partly explained by the higher deposition rates outdoors compared with indoors but the main factor relates to wet deposition from the atmosphere. Paatero (2000) measured up to 10^5 Bq l^{-1} of radon decay products in rain water in Finland. Our results have implications for the radiation dose to the skin basal layer from deposition of radon decay products on the human body. For continuous outdoor exposure, skin dose rates exceeding the ICRP recommended public exposure limit of 50 mSv y^{-1} are estimated, compared with only 2.5 mSv y^{-1} indoors.

Introduction

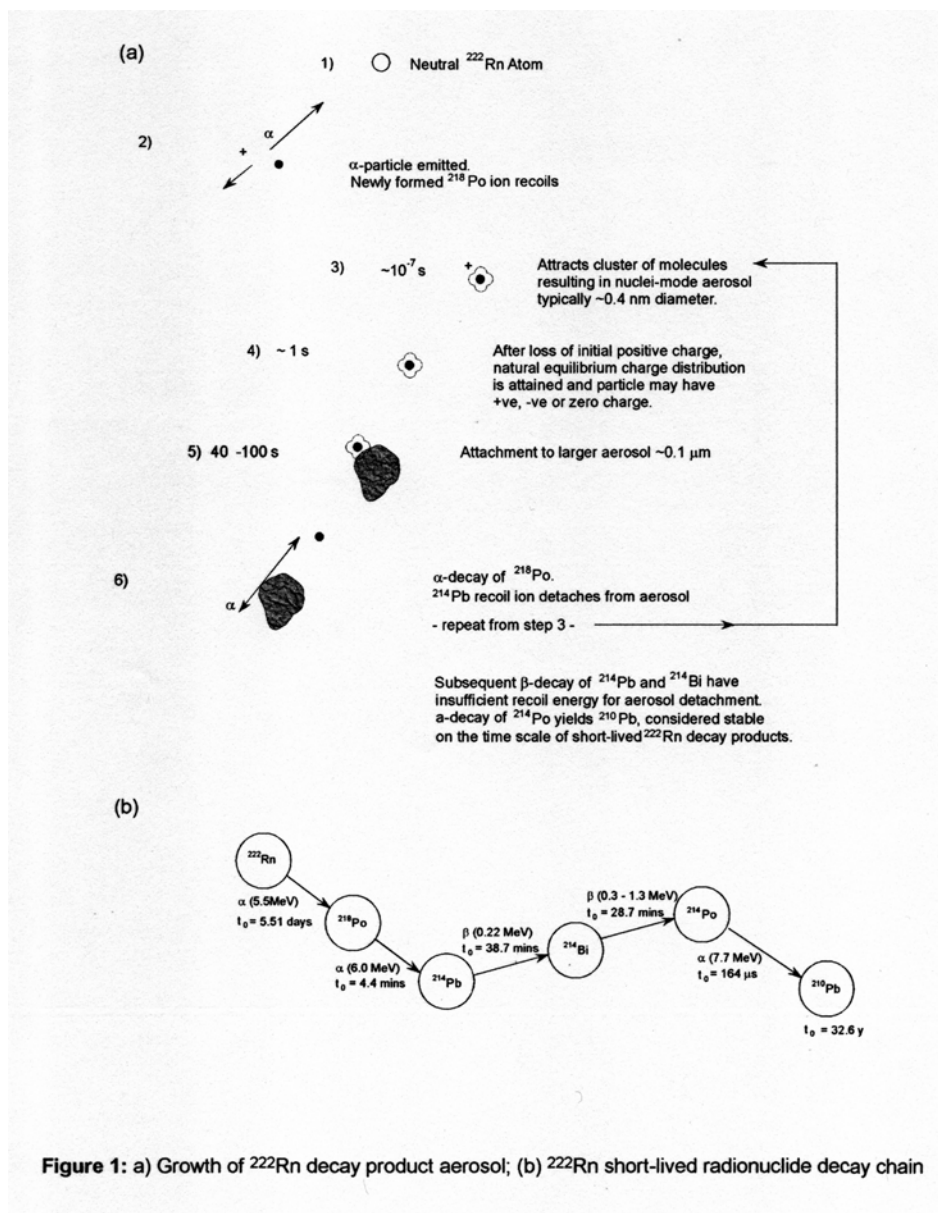
In the field of radiation protection, the radiation dose to the basal layer of the skin arising out of radioactive aerosols landing on the skin has been evaluated. The International Commission on Radiological Protection, ICRP has recommended that for the general public the annual dose rate to the skin basal layer should not exceed 50 mSv y^{-1} (NRPB 1997). Plateout on exposed areas of the human body of the naturally occurring radon (^{222}Rn) decay product aerosols ^{218}Po and ^{214}Po results in an alpha-radiation dose to the skin basal layer. Eatough and Henshaw (1992) have estimated that at the average UK indoor radon exposure of 20 Bq m^{-3} the dose rate is 2.5 (range 1.7 to 17) mSv y^{-1} . The range in estimate reflects the wide variation in published plateout data. Using radiation risk factors, Eatough and Henshaw (1995) estimated that 2% (range 1% to 10%) of non-melanoma (common) skin cancer could be linked to the plateout of radon decay products on the body. The analysis of Eatough and Henshaw however was concerned solely with indoor radon exposure. This is a common practice in radon dosimetry where it is assumed that radon levels outdoors are very low and can be ignored and in any case on average, people spend only 10% of their time outdoors.

However, in outdoor experiments designed to study the excess deposition of aerosols on the human body under high voltage powerlines (Fews *et al* 1999), unusually high average levels of plateout activity have been recorded, with highest values up to 120 Bq m^{-2} . These high values are partly explained by the higher aerosol deposition rates outdoors compared with indoors, but the main factor relates to wet deposition from the atmosphere. Paatero (2000) measured up to 10^5 Bq l^{-1} of radon decay products in rain water in Finland. The author found evidence that even the smallest amounts of precipitation deposit high levels of radon decay products and continued rainfall merely dilutes the activity in a larger water volume. There was also evidence that atmospheric washout was associated with air movements on a scale length of several hundred kilometres.

Here we present further evaluation of existing Bristol data and the results of further experimental measurements of wet deposition of radon decay products. This subject is of fundamental interest in aerosol and atmospheric science, but the consequences for the natural radiation dose to the skin may also be of fundamental importance to the aetiology of skin cancer in the general population.

Radon and its decay products in air

Figure 1 shows the production of radon decay product aerosols in air. The detailed behaviour is complex owing to the short mean lifetimes of individual decay product nuclei and the attachment and detachment of individual decay products to and from larger aerosols. In indoor air Reineking *et al* (1994) found that between 5 and 18% of radon decay products are in nuclei mode aerosols <100 nm, that is, they are unattached to larger aerosols.



It is assumed that most of this unattached fraction exists in small ion molecular cluster form, ~ 1 nm in size. There are few measurements for outdoor air but some suggest a similar fraction to that indoors exists in nuclei mode (Reineking *et al* 1988 & 1992).

In the present application we are interested in the surface deposition (plateout) of these radioactive aerosols. Indoors this occurs if, during the mean lifetime of the decay products atom, diffusion and air movements generally bring the aerosol into contact with a surface. Outdoors we are also interested in wet deposition or direct washout in rain as studied by Paatero (2000).

Materials & Methods

The surface deposition of the short-lived radon decay products was measured using alpha-particle sensitive TASTRAK plastic track detectors (Fews 1992a, b, Henshaw *et al.* 1994, 1995, 1996).

In the present application 7 x 5 cm sheets of TASTRAK were held horizontally in air outdoors, sampling ^{222}Rn and its decay product aerosols in air and the deposition (plateout) of the latter on the detector surface.

^{222}Rn decay product aerosols deposited on the surface of TASTRAK detectors emit α -particles of the characteristic energy of the decay products ^{218}Po and ^{214}Po . Those α -emissions in the direction of the plastic will be recorded at their characteristic full energy of respectively 6.0 and 7.7 MeV and ranges in TASTRAK of 40.2 and 60.5 μm .

The ^{218}Po and ^{214}Po α -particles have respective ranges in air of 5.1 and 7.6 cm. Therefore, a proportion of the ^{222}Rn decay product aerosols in the air in front of the detector as well as ^{222}Rn itself may also be recorded if their α -emissions occur both within range and in the direction of the detector surface. (Note that TASTRAK is not sensitive to the β -particles emitted from the decay products ^{214}Pb and ^{214}Bi , so that the detection of the extremely short half-life ^{214}Po is a signature of these preceding β -emitting decay products).

In the present work cone-like etch tracks were revealed by etching in 6.25 M NaOH at 75° C for 4.5 h. The detectors were analysed using an image analysis system in which up to 14 parameters of the shape and size of each recorded α -particle track were measured (Fews 1992a and b). These measurements, combined with track geometry data from calibration exposures, enable the α -particle energies to be determined.

Figure 2 illustrates how recorded track size varies with α -particle energy and angle of incidence. The projected track length (the major axis of the etch track opening mouth on steep tracks, and the track length in projection on shallow tracks) is plotted versus the minor axis diameter. The data fall within an envelope characteristic of the α -particle track response for the etch conditions employed. Bands are seen which originate from α -emissions from ^{218}Po and ^{214}Po deposited on the plastic surface. These arise from the deposited α -radionuclides observed at constant energy over a range of incident angles. Airborne emission will give a variation of both energy and angle, and discrete bands would not be observed. Therefore, a separate measurement of both the deposited and airborne components is made from the same TASTRAK detector.

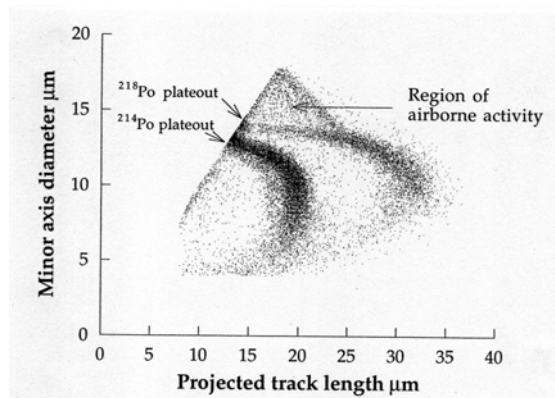
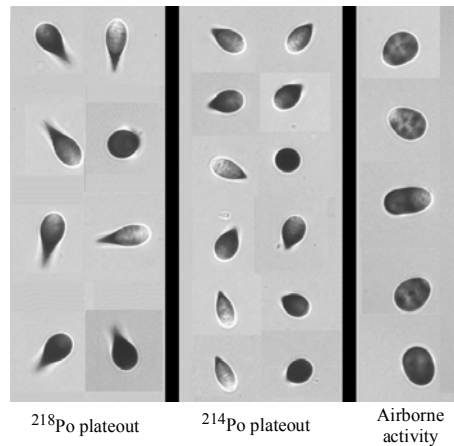


Figure 2: Photomontage of etched tracks and scatter plot of track minor axis versus projected track length, Showing ^{218}Po and ^{214}Po plateout and the region of airborne activity

Use of TASTRAK outdoors requires comparatively long exposure periods to acquire enough counting statistics, although exposures of several days have the advantage of integrating over short-term variations in aerosol concentration. In practice TASTRAK exposed outdoors suffers solar UV damage which limits its exposure period to around six days. Under these conditions typical count densities range from a few hundred tracks per cm^2 from deposited activity, to < 30 per cm^2 from the air activity.

Fews *et al* (1999) present measurements using alpha-particle sensitive TASTRAK plastic track detectors to evaluate the deposition of radon decay product aerosols on models of the human head situated under high voltage powerlines. A subset of control detectors was exposed horizontally outdoors typically for six days. Weather conditions were recorded throughout the exposure. These detectors form the first set of data on wet deposition presented here.

A further set of TASTRAK detectors was later exposed horizontally for periods from 15 to 240 minutes. These exposures commenced with the onset of rain. The frequency, duration and amount of rain both before and during each exposure were noted. After exposure the detectors were shielded for three hours prior to processing to allow decay of accumulated radon decay products.

Deposition of ^{218}Po and ^{214}Po on the surface of TASTRAK detectors has been used as the basis of a skin dosimeter. Here the recorded plateout alpha-activity per unit area of detector has been used to estimate the corresponding dose to the basal layer of the skin (Eatough 1997, Eatough *et al* 1999).

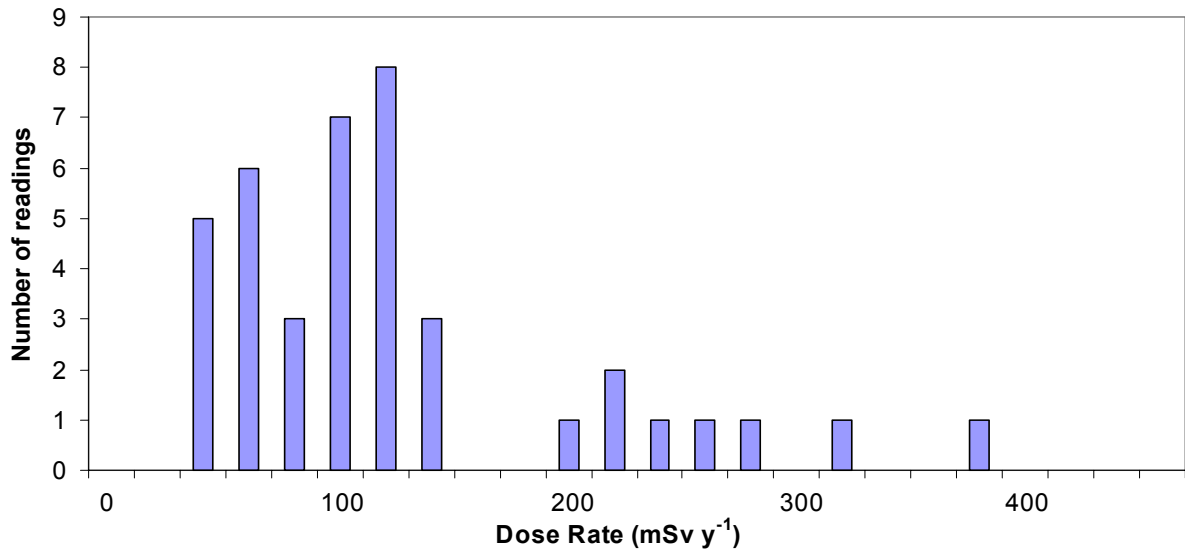


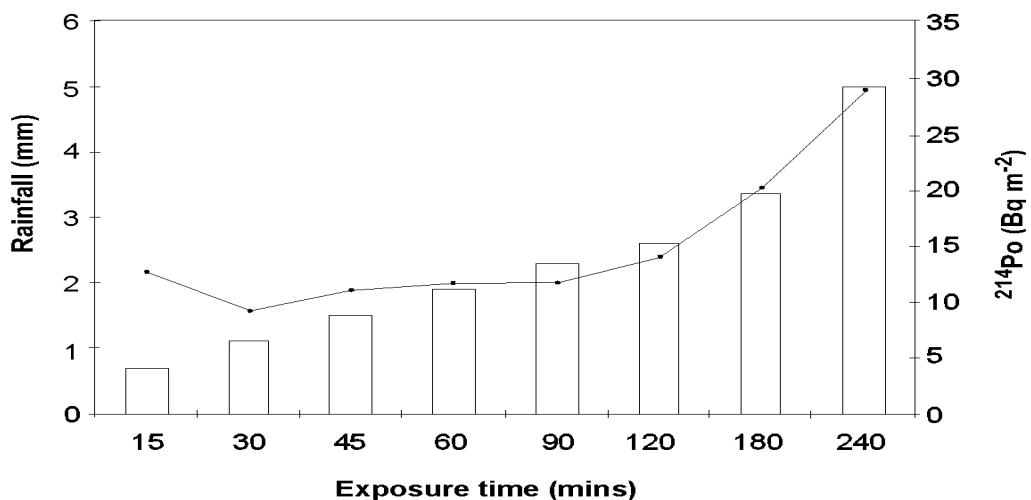
Figure 3. Distribution of dose rate to the skin basal layer from plateout of radon decay products

Results

For the detectors used as controls in the experiments of Fews (1999), the average recorded activity was $42.2 \pm 3.8 \text{ Bq m}^{-2}$. This was dominated by ^{214}Po with only a small signal present for ^{218}Po . The weather was dry for 0.86 of the exposure. Rainfall averaged over the whole 6-day exposure was 3.6 mm per day.

Figure 3 shows a histogram of recorded values in terms of annual dose rate in mSv y^{-1} to the skin basal layer. The dose is assumed to arise solely from ^{214}Po , using a dose conversion of $1 \mu\text{Sv/decay/cm}^2$ (Eatough 1997). The average dose rate is $132 \pm 12 \text{ mSv per year}$ or $15.2 \pm 1.4 \mu\text{Sv per hour}$.

For the set of further detectors, figure 4 shows the exposure time to rainfall and the corresponding plateout activity of ^{214}Po . Negligible ^{218}Po plateout was detected. As the total rainfall increases the plateout increases but in lower proportion. Figure 5 shows the corresponding data per unit rainfall for a set of detectors exposed up to 60 minutes. Figure 6 shows similar data for a second set exposed for up to 240 minutes (4 hours). In both figures 5 and 6 there is evidence that most ^{214}Po plateout occurs in the first few minutes of rain.



Figures 4. Exposure time to rainfall and the corresponding plateout activity of ^{214}Po
 Bars = rainfall, dots = ^{214}Po

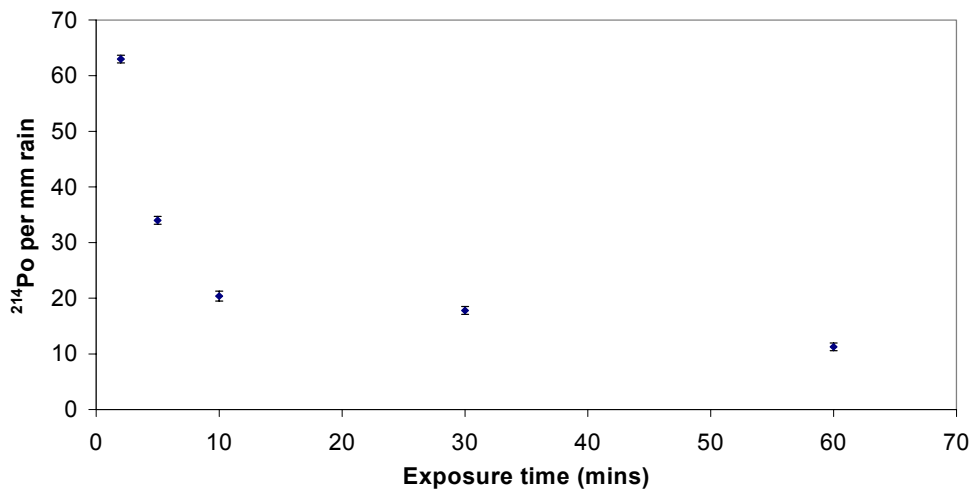


Figure 5. ^{214}Po plateout per unit rainfall as a function of exposure time – set 1.

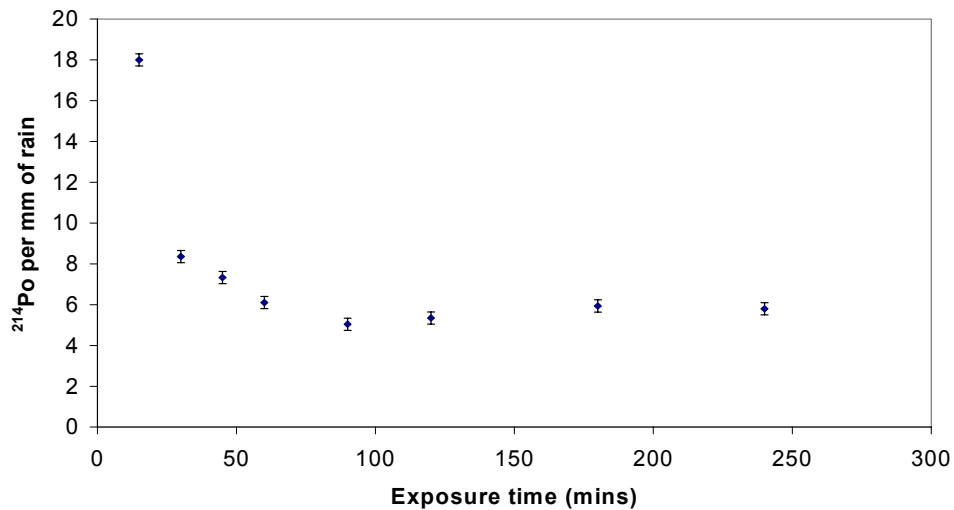


Figure 6. ^{214}Po plateout per unit rainfall as a function of exposure time – set 2.

Discussion

In our measurements of the alpha-emitting components of the ^{222}Rn short-lived decay products, most of the deposited activity arose from ^{214}Po with little evidence for the presence of ^{218}Po . This is understandable in terms of the relative mean lifetimes of these radionuclides. Thus, the mean lifetime of ^{218}Po is 5.5 minutes. The mean lifetime of ^{214}Po is only 164 μs but it may be deposited in the form of any of its parent radionuclides, including ^{218}Po which together have a mean lifetime of 72 minutes. The apparent absence of deposition of ^{218}Po argues against washout of these decay products on a time scale of a few minutes, but the substantial presence of ^{214}Po suggests that considerable washout is taking place on a time scale up to 72 minutes or more.

The results in figures 5 & 6 are consistent with those of Paatero (2000) that even short, small amounts of precipitation deposit high levels of radon decay products and continued rainfall merely dilutes the activity in a larger water volume. Paatero developed an automatic precipitation gamma analyser which recorded gamma-emission from ^{214}Pb and ^{214}Bi . The

instrument was used to make continuous measurements in northern Finland from August to December 1998. The author found radon decay product concentrations in rainwater of up to 10^5 Bq l⁻¹. Concentrations above ~ 0.2 Bq l⁻¹ were exclusively associated with wind directions between southeast and northeast. These correspond to weather systems over land areas compared with the sea from westerly directions. There was evidence that the high washout of radon decay products was associated with wind movements over several hundred kilometres. However, the mean lifetime of the radon decay products, 72 minutes in total, would suggest a limiting distance from which recorded washout activity can originate. It is clear that local variations in radon emission from the ground, such as in Southwest England are not major factors governing the washout of radon decay products.

The results shown in figure 3 were obtained for exposures times of around 6 days. If this is typical of the deposition rates of radon decay products, then for continuous exposure outdoors considerable radiation doses to the basal layer of the skin can accrue, up to several hundred mSv y⁻¹ compared with the ICRP recommended limit of 50 mSv y⁻¹. On the other hand, the observation that the majority of washout occurs in the first few minutes of rain suggests that in the UK merely being '*caught in a shower*' outdoors could lead to significant skin deposition of radon decay products.

Wet deposition of radon decay products on uncovered areas of the human body has clear implications for the natural radiation dose to the basal layer of the skin. Current estimates by Eatough & Henshaw (1992) of 2.5 (range 1.7 to 17) mSv y⁻¹ for the average UK radon exposure of 20 Bq m⁻³ is based solely on indoor radon exposure. These values of dose rate suggest that 2% (range 1% to 10%) of non-melanoma (common) skin cancer in the general population could be linked to natural plateout of radon decay products.

Clearly, the estimated dose rate values will increase when account is taken of outdoor exposure. In the UK people are thought to spend on average 10% of their time outdoors. In Fews *et al* (1999) we estimated that taking this into account, the annual dose rate would rise to ~ 18 mSv y⁻¹. The estimate would rise further for those in certain occupations such as farming.

On the other hand there are many areas of the world where populations spend lengthy periods of time outdoors, or who live outdoors. In counties experiencing tropical rainstorms for example the washout of radon decay products on the skin could be quite considerable.

For continuous outdoor exposure, the skin dose rates could exceed 50 mSv y⁻¹, the value recommended for public exposure by ICRP. At this limit, radiation risk factors would suggest that around 30% of skin cancer (for cancer initiation) could be attributed to skin plateout of radon decay products.

Some interesting questions arise from our current understanding of radiation risks:

(i) is the proportion of common (non-melanoma) skin cancer induced by natural alpha-particle radiation from radon decay products far higher than currently suggested;

(ii) are deterministic effects such as the ageing of the skin linked to natural alpha-radiation of skin basal cells?

Or, alternatively

(iii) are the standard risk factors used in radiation protection widely in error and natural skin deposition of radon decay products is of little radiological consequence?

Further research on these questions is clearly needed.

Acknowledgements

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