# A radiological study of pulverised fuel ash (PFA) from UK coal-fired power stations

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#### **Abstract**

In common with all natural fossil fuels and their derivatives, pulverised fuel ash (PFA) contains small amounts of naturally occurring radionuclides. The uranium and thorium decay series both contain an isotope of radon which, being a noble gas, can escape from PFA and any materials into which it is made, such as building blocks for the construction industry.

The EU Products Directive requires that building materials and their constituents do not cause a hazard to health. In addition, several other pieces of UK legislation and EU directives impose certain restrictions on the radionuclide content of materials or the exposure of people working with sources of radiation.

A study was conducted on behalf of the United Kingdom Quality Ash Association (UKQAA) to update the radiological measurements conducted by the National Radiological Protection Board (NRPB) in 1986. Since the majority of coal being burnt in UK coal-fired power stations is no longer from British deep mines, but sourced worldwide, the radionuclide content has changed. The implications of these changes needed to be assessed in the light of current legislation.

Samples of PFA from five UK coal-fired power stations were analysed for the naturally occurring radionuclides in the uranium-238 and thorium-232 series and potassium-40 using a high purity germanium detector. The exhalation rates of radon-222, and various derived quantities, were also determined experimentally.

The radium-226 activities per unit mass of the PFA samples ranged from 70 to 150 Bq kg<sup>-1</sup>. The radon-222 exhalation rates were between 4.2 10<sup>-4</sup> and 4.6 10<sup>-5</sup> Bq m<sup>-2</sup> s<sup>-1</sup>, with exhalation coefficients from 0.003 to 0.026. These are consistent with literature values. On the basis of these analyses, it is considered unlikely that there is a significant increase in the radiological hazard from the use of PFA in building materials. It is also considered unlikely that any of the trigger levels under current UK legislation or EU directives will be exceeded.

#### Introduction

Radon is a naturally occurring radioactive gas that is formed from the decay of uranium in all rocks and soils. On average, it accounts for 50% of the 2.7 mSv (millisievert) annual effective dose\* received by a UK resident¹. Although the main source of radon in a building is from the ground below the foundations, the building materials can make an important contribution. A further 13% of the dose is from gamma rays emitted by the ground and building materials. Although the sources of radioactivity in building materials are natural, the concentrations are controlled, and are subject to legislation. To date, the radioactive content of building materials in the UK has not been a cause of concern. However, building materials in a number of European countries have been shown to contain radioactivity in sufficient quantities³ to warrant intervention and restrictions on their use. A familiar building material can become problematic if the origin of one of the constituents is changed, and increases the concentration of natural radionuclides.

#### **Pulverised Fuel Ash**

The construction industry utilises recycled waste products from power generation, in particular pulverised fuel ash (PFA). This constitutes the residue from burning coal and is used in the manufacture of lightweight building blocks. As PFA is derived from a natural fossil fuel, it contains small amounts of naturally occurring radionuclides such as uranium-238 and thorium-232, their daughter products, and potassium-40. The ashing process concentrates the activity per unit mass six-fold compared with coal from the same source. Radon is the only naturally occurring radioactive gas. Although an isotope of radon occurs in the thorium-232 decay chain, radon-220, it is of limited interest because of its short half-life. However, radon-222 from the uranium-238 decay chain has a half-life of 3.8 days, which means that a significant amount can escape from PFA and any materials into which it is manufactured.

In the UK, the promotion of applications for PFA is carried out by the United Kingdom Quality Ash Association (UKQAA). Its predecessor contracted the National Radiological Protection Board (NRPB, now the Health Protection Agency Radiation Protection Division) to undertake a study in 1986<sup>2</sup> of the radionuclide content and radon-222 exhalation of PFA. The conclusion at that time was that the radiological impact on workers and members of the public from the use of PFA in building materials was negligible. However, the majority of coal burnt in UK power stations is no longer from British deep mines, but sourced worldwide. The significantly higher levels of natural radionuclides that are found in some non-UK coals will affect the resulting PFA<sup>3</sup>. As the EU Construction Products Directive<sup>4</sup> requires that building materials and their constituents do not cause a hazard to health, the current study was conducted to update the available information on PFA given the revised circumstances.

The study consisted of three main parts: the measurement of radionuclide content and radon-222 exhalation rate of PFA samples; a literature review covering other analyses, EU Directives and health risks; and the preparation of a report.

<sup>\*</sup> Usually abbreviated to dose.

# Radionuclide content analysis

Samples of PFA were provided from five unidentified coal-fired UK power stations. Two additional samples were provided which had been 'conditioned', i.e. contained 10-15% water at the time of delivery. In the absence of a detailed sampling programme, these were taken as being representative of PFA being produced in the UK at that time. All the samples were analysed for radionuclide content and the results compared with literature values.

#### **Gamma spectrometry**

Gamma spectrometry is a well-established method of determining the radionuclide content of materials. The gamma rays emitted when many radionuclides decay can be used to identify which radionuclides are present, from the energy of the rays detected, and the content by mass within the sample from the strength of the signal. A number of different radionuclides may produce gamma rays of similar energies. In order to determine which radionuclides are present in the sample, the presence of other members in the relevant decay chains can be investigated. Strict procedural controls are required to give reliable results.

In this study, approximately one litre of each of the samples was re-packed into Marinelli beakers to give the correct geometry for the gamma spectrometry. The net mass of each sample was determined and found to be approximately one kilogram.

The operation of the detectors, which are in a low background laboratory, is UKAS accredited to ISO9001<sup>5</sup>. The high purity germanium (HPGe) detectors have a relative efficiency of 40% and measure gamma emissions in the energy range from 60 keV to approximately 2000 keV, which covers the range of gamma ray energies from the radionuclides of interest. The detectors are shielded on all sides to minimise spurious signals. Each sample was counted for 60,000 seconds and the resulting spectra analysed with the Canberra Genie software.

The presence and content of the naturally occurring radionuclides in the uranium-238 series, thorium-232 series and potassium-40 was determined. As the first two series contain isotopes of radon gas, the relative contents of gamma-emitting radionuclides before and after radon in each series would show whether radon losses had been significant.

#### Results and discussion

The results are given in Table 1. The radium-226 contents by mass (from the uranium-238 decay chain) were at the higher end of literature values of PFA from UK power stations<sup>3</sup>. However, they were all below the typical EU value<sup>3</sup> of 180 Bq kg<sup>-1</sup>, the range of which goes up to an order of magnitude higher. The lead-214 and bismuth-214 values in each case were the same, within experimental uncertainties, as the radium-226 values, which showed that the radon-222 had been retained.

The radionuclides in the thorium-232 chain showed similar behaviour, with lead-212 and bismuth-212 contents similar to actinium-228; radon-220 has a very short half-life and was therefore retained within the sample.

The widest variation was seen within the content of potassium-40. All the values fell within the EU range of 170-1450 Bq kg<sup>-1</sup>; previous UK values have been reported to be around 900 Bq kg<sup>-1</sup>. For the two conditioned samples, no firm conclusions could be drawn on the effect of radionuclide content from this process.

			Sample activities, Bq kg <sup>-1</sup>							
Decay series	Nuclide	Energy, keV	Station A	Station B	Station C	Station C *	Station D	Station D *	Station E	
U-238	Ra-226	186	130 ± 50	150 ± 50	140 ± 50	100 ± 40	100 ± 40	70 ± 30	110 ± 40	
	Pb-214	352	120 ± 20	160 ± 30	120 ± 10	120 ± 20	100 ± 20	80 ± 20	100 ± 20	
	Bi-214	609	110 ± 20	140 ± 20	110 ± 20	100 ± 20	90 ± 20	70 ± 10	90 ± 20	
	Pa-234m	1001	130 ± 40	200 ± 50	120 ± 40	110 ± 40	90 ± 40	110 ± 40	110 ± 30	
Th-232	Ac-228	911	100 ± 40	140 ± 60	100 ± 40	100 ± 40	80 ± 40	70 ± 30	70 ± 30	
	Pb-212	300	70 ± 10	130 ± 20	80 ± 20	130 ± 30	80 ± 10	100 ± 20	70 ± 10	
	Bi-212	727	100 ± 20	150 ± 20	90 ± 20	150 ± 30	90 ± 10	100 ± 20	80 ± 10	
	K-40	1460	650 ± 100	280 ± 40	790 ± 120	540 ± 80	1100 ± 160	880 ± 130	480 ± 70	

Reported uncertainties are for the 95% level of confidence.

Table 1 - The activities of gamma-emitting radionuclides in the PFA samples

#### Radon-222 exhalation

The same samples were analysed for their radon-222 exhalation characteristics. During the process of radioactive decay, the newly formed radon atom is left with kinetic (recoil) energy, which allows it to move away from its original location. Through linked pores or air spaces, the random movement of the radon-222 atoms results in a net migration from a higher to lower concentration: this is diffusion. Thus radon-222 can be exhaled from the surface of a PFA sample.

#### Diffusion curve analysis

The exhalation characteristics of radon-222 from a sample can be determined by placing it in a sealed chamber containing low radon air. Radon-222 will diffuse from the open surfaces along the concentration gradient causing the radon-222 level in the chamber air to rise. After a period of time, the exhalation of radon-222 from the sample will be balanced by the radon-222 in the air of the chamber undergoing radioactive decay. Repeated or continuous measurements of the radon-222 concentrations in the chamber air will show an initial increase followed by a plateau<sup>6</sup>. Analysis of the shape of the curve allows quantities such as the free exhalation rate and exhalation coefficient to be determined, which can be compared with literature values.

<sup>\*</sup> Conditioned sample

A PFA sample of approximately one kilogram was placed in a steel and Perspex exhalation chamber that was filled with low radon air. The chamber was connected to a small pump and a Lucas Alpha-scintillation Flask by a closed loop of rubber hoses and self-sealing Swagelok connectors. Care was taken to ensure that all joints were airtight. At intervals, the air in the Flask was refreshed with that from the exhalation chamber and the Flask removed for counting. The counts per hour from the Pylon AB-5 scintillation monitor were converted to a radon-222 gas concentration. The same Flask was re-connected to the system and the procedure repeated until the concentration values reached a plateau.

#### Results and discussion

Previous studies have reported radon-222 exhalation data in a variety of ways depending upon the chosen experimental technique. This meant that values needed to be obtained for a number of derived quantities to allow a full comparison with the published data.

For each sample, the exhalation rates (and derived quantities) were estimated using both the slope of the initial increase and the radon-222 concentration in chamber once the plateau had been reached<sup>6</sup>. Measurements were made of the size, shape and mass of the sample and estimates of the net internal volume of the chamber and Flask. The curve for PFA from Station A shown in Figure 1 is nearly textbook in form and estimates of the exhalation rates by the two methods were in close agreement. In other samples, a plateau was reached within a few hours, which made the initial increase difficult to assess given the limitations of the measurement technique. Conversely, with one sample a plateau had not been reached after more than 400 hours in the exhalation chamber. For each sample, therefore, a judgement was made on which method gave the best estimate of the exhalation rates and these values are shown in Table 2.

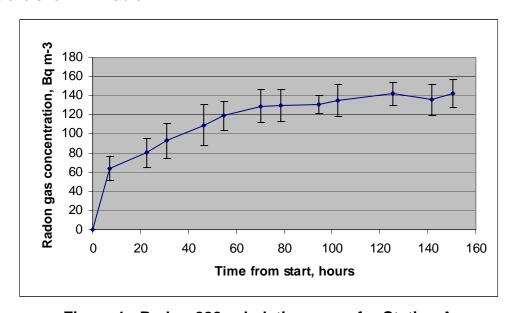


Figure 1 - Radon-222 exhalation curve for Station A

The earlier gamma spectrometry results had shown that the radon-222 was largely being retained within the samples, which can be seen from the small exhalation coefficients. The exhalation values obtained in this study were consistent with

literature values, although with a wide range. Once again, no firm conclusions could be drawn on the effect of conditioning of the samples.

	Station	Α	В	С	C *	D	D *	E	Average
Exhalation	Unit								
Free	Bq s <sup>-1</sup>	2.0E-06	1.1E-06	1.2E-06	5.1E-06	2.4E-06	1.4E-06	5.6E-06	2.0E-06
Per unit area	Bq m <sup>-2</sup> s <sup>-1</sup>	2.8E-04	9.1E-05	9.8E-05	4.2E-04	3.6E-04	9.1E-05	4.6E-05	2.0E-04
Per unit volume	Bq m <sup>-3</sup> s <sup>-1</sup>	2.1E-03	1.2E-03	1.4E-03	5.6E-03	2.6E-03	1.5E-03	6.2E-04	2.1E-03
Per unit mass	Bq kq <sup>-1</sup> s <sup>-1</sup>	1.9E-06	1.3E-06	1.2E-06	5.5E-06	2.1E-06	1.5E-06	6.7E-07	2.0E-06
Coefficient		0.007	0.004	0.004	0.026	0.010	0.010	0.003	0.009
Derived rate	Bq m <sup>-2</sup> h <sup>-1</sup>	0.99	0.33	0.36	1.50	1.31	0.41	0.17	0.72
Normalised rate	kg m <sup>-2</sup> h <sup>-1</sup>	0.008	0.002	0.003	0.015	0.013	0.006	0.002	0.007

<sup>\*</sup> Conditioned sample

Table 2 - Estimated and derived radon-222 exhalation rates

# Relevant legislation and health risks

Ultimately, the experimental results must be put into context. Radiation exposure is a recognised health risk and a raft of UK legislation and EU Directives exists to protect employees and members of the public. Extensive literature reviews were therefore conducted to identify the relevant regulations, both current and expected, and an assessment made of their impact on the use of PFA.

#### **EU Directives and Guidance and Application in the UK**

#### **Basic Safety Standards Directive (BSS)**

The European Union has its own set of regulations relating to radioactive materials, the Basic Safety Standards Directive (BSS)<sup>8</sup>. Although its general scope does not cover exposures from natural sources other than when part of the nuclear fuel cycle, it also applies to: 'work activities which involve the presence of natural radiation sources and lead to a significant increase in the exposure of workers or members of the public which cannot be disregarded from the radiation protection point of view'. Individual Member States have to identify relevant work and apply the requirements of the Directive. In the UK, the BSS has been implemented through the production or amendments of regulations and Directions to the applicable Environment Agencies. The two relevant pieces of primary legislation are as follows:

#### Radioactive Substances Act 1993 (RSA93)

The RSA93<sup>9</sup> covers work involving materials containing naturally occurring radionuclides. However, Schedule 1 contains exclusion levels for solid, liquid and gaseous materials containing naturally occurring radionuclides (other than those

involved in the nuclear fuel cycle). If a material has radionuclide concentrations below those values listed it is not considered to be radioactive and therefore excluded from the provisions of the Act.

The radionuclide concentrations measured during this study are substantially below the exclusion threshold values specified. This is consistent with other studies in this area<sup>3</sup> and shows that PFA is not subject to regulation under RSA93.

### **Ionising Radiations Regulations 1999 (IRR99)**

The application of the IRR99<sup>10</sup> to work with substances containing naturally occurring radionuclides is given in Regulation 3. In work other than a practice (including the production, processing, handling, use, holding, storage, transport or disposal of radioactive substances), e.g. coal-fired power stations, the IRR99 only apply if 'their use is likely to lead to employees receiving an effective dose of ionising radiation in excess of 1 millisievert in a year'.

The results of studies<sup>3</sup> of the radiological consequences of the use of ash in building materials indicate that doses to workers and members of the public from the ash are significantly less than 1 mSv y<sup>-1</sup>, and thus the IRR99 do not apply.

# EC Recommendation on radon dwellings

The BSS does not apply to radon in dwellings. However, the EC recommendation<sup>11</sup> introduces a design level for radon exposure to assist the relevant national authorities setting regulations, standards or codes of practice. In the UK the Government endorsed the NRPB recommendation of 1990 that an action level<sup>12</sup> be set at an average annual radon gas concentration of 200 Bq m<sup>-3</sup>.

# Radiation protection 112 - Radiological protection principles concerning the natural radioactivity of building materials

The EC has produced and adopted guidance (which is not legally binding on Member States) to limit radiation exposure due to materials with enhanced levels of natural radionuclides<sup>13</sup>. Whilst the general principle is that doses to members of the public should be kept as low as reasonably achievable (ALARA), it is recognised that low doses from building materials are ubiquitous, so controls should be based on exposure levels that are higher than normal. The guidance given is that:

- The amount of radium (the precursor of radon) in building materials should be restricted to at least a level where it is unlikely that it could be a major cause of exceeding the design level for indoor radon of 200 Bq m<sup>-3</sup>
- Exceptionally high individual doses should be restricted. Gamma doses exceeding 1 mSv y<sup>-1</sup> due to the building materials are very exceptional. When gamma doses are limited, the radium-226 concentrations are limited, in practice, to levels that are unlikely to cause indoor radon concentrations to exceed 200 Bq m<sup>-3</sup>
- Controls on building materials should be based on doses in the range 0.3 1 mSv y<sup>-1</sup> (in excess of the gamma dose received outdoors), although some higher doses could be accepted in exceptional cases where materials are used locally.

In addition, building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them does not increase the effective dose to a member of the public by more than 0.3 mSv y<sup>-1</sup> (in excess of the gamma dose received outdoors).

The report<sup>13</sup> concluded that it was possible for doses to exceed 0.3 mSv y<sup>-1</sup> with almost any use of concrete in bulk amounts. Moreover, 1 mSv y<sup>-1</sup> was possible if the concrete contained large amounts of fly ash, blast furnace slag, or aggregates rich in natural radionuclides. This EC guidance therefore indicated that some restrictions on concretes and other building materials containing ash might be required. However, the later publication of Radiation Protection 122 has produced some inconsistencies.

# Radiation Protection 122 - Practical use of the concepts of clearance and exemption: Part II Application of the concepts of exemption and clearance to natural radiation sources

This later guidance<sup>14</sup> relates to different ways of avoiding wastage of regulatory resources where there would be little or no benefit. It concludes that it is appropriate to lay down a single set of levels for both clearance and exemption, i.e. levels of radionuclide concentrations in materials below which no regulatory controls are required.

After considering various set scenarios concerning the use of by products in building materials, such as ash and slag, the exemption and clearance levels were calculated. For both uranium-238 and thorium-232 in secular equilibrium the derived clearance-exemption level is 0.5 Bq g<sup>-1</sup>. Clearance-exemption levels were also derived for segments of the decay chains, which are shown in Table 3.

<sup>\*</sup> Secular equilibrium is achieved where the half-life of the parent radionuclide is much longer than the half-life of the daughter. If the radionuclides are not separated, the daughter decays at the rate at which it is produced, meaning that effectively the parent and daughter radionuclides decay at the same rate.

Nuclides	Clearance levels for all materials (except wet sludges from oil and gas industry), kBq kg <sup>-1</sup>				
Uranium-238 sec incl Uranium-235 sec*	0.5				
Uranium (natural)	5				
Radium-226+	0.5				
Lead-210+	5				
Polonium-210	5				
Thorium-232 sec*	0.5				
Thorium 232	5				
Radium-228+	1				
Thorium-228+	0.5				
Potassium-40	5				

<sup>\*</sup> Secular equilibrium

Table 3 - Selected rounded general clearance levels

Comparing these exemption-clearance levels with the concentrations of the radionuclides in ash from UK power stations measured as part of this study (and those from an earlier study³), it can be seen that there should be no restriction on the use of ash in building materials within the UK if these levels were to be adopted. However, these levels have not been adopted by the EC and their future remains uncertain. As mentioned above, there is some concern that the conclusions of Radiation Protection 122 apply as their impact on building materials are not entirely consistent with those of Radiation Protection 112. Some further work might be needed to resolve these issues.

#### The Construction Products Directive

The Construction Products Directive (CPD)<sup>4</sup> was introduced as one of the 'New Approach' Directives, which seek to remove technical barriers to trade within the European Economic Area (EEA). The intention of CPD is to replace existing national standards and technical approvals with a single set of Europe-wide technical specifications for construction products. This allows manufacturers to use the familiar CE mark, which means that products cannot be refused entry to the EEA on technical grounds.

European technical specifications are of two types - harmonised European standards and European Technical Approvals (ETAs). Manufacturers wishing to take advantage of the Single Market should ensure that their products meet the requirements of one or the other. The 'harmonised' parts of European standards address regulations across the Member States and must be met in order to affix the CE marking. Where harmonised standards cannot be written, for instance where recognised national standards do not exist, manufacturers may apply for an ETA to

be issued against their product, i.e. an assessment of the product's likely performance.

Standards are written by the European Committee for Standardization (CEN). Standards and ETAs detail the performance characteristics required of products to allow works to meet the essential requirements of the CPD. Standards and guidelines also set out the systems of attestation of conformity. In the UK, CPD came into force in 1991<sup>15</sup>, with a subsequent amendment in 1994<sup>16,17</sup>.

The authors are not aware of any current European-wide technical specifications under the CPD that contain standards for the exhalation of radon from building materials, or their radionuclide content. However, the CPD makes reference to these potential hazards and in the future such specifications, including possible threshold concentrations for radionuclides, might be developed.

#### Annex 1 of the CPD requires that:

'The construction works must be designed and built in such a way that it will not be a threat to the hygiene or health of the occupants or neighbours, in particular as a result of the following:

... - the emission of dangerous radiation...'

Interpretative Document No 3: 'Hygiene, Health and the Environment' expands and develops upon the requirement above in relation to five specific aspects: indoor environment, water supply, wastewater disposal, solid waste disposal, and outdoor environment.

The 'indoor environment' aspect is relevant to radon and external gamma irradiation from building materials, and in particular the air quality requirement, which 'is concerned with the elimination or control of pollutants in the indoor environment. In the following text 'pollutant' includes gamma radiation (although strictly it is not an airborne pollutant).

The construction works must provide a healthy indoor environment for occupants and building users, taking account of pollutants including:

... - radon and radioactive substances emitting gamma radiation...'

The requirements for building materials may be expressed in a number of ways, including:

- Acceptable average and peak concentrations of specific pollutants in indoor air.
- The prohibition or limitation of use of named substances generally or for specific purposes.
- Limitations on the rates of release and nature of pollutants from materials or products.

In addition to the above, Guidance Paper H (A Harmonised Approach Relating to Dangerous Substances under the Construction Products Directive)<sup>19</sup> is intended to address the problem of dangerous substances and radiation when relating to

products falling under the CPD. It is aimed at those involved in writing technical standards, and advises that threshold levels could be set where compliance is demonstrated by being below this value, or declared if exceeded:

'The threshold level could be set at what is considered to be effectively zero, which in this case could be the level of natural radioactivity that is unavoidable and causes no danger to the user of the works.'

Guidance Paper H also gives radioactivity in construction products as an example. Following the principles cited above:

'The concentration of different radionuclides should be measured (Bq/kg) and the "activity concentration index" calculated. The calculated value should be declared with the CE marking only when the value is above the threshold given in the specification. The threshold could for example be the same as the gamma dose present in the earth's crust, which would mean "effectively zero"...The declared value will make it possible for regulators/designers to estimate the annual effective dose of radiation (mSv) and this see if the regulatory requirements of the works can be met.'

#### **Health Risks**

Ultimately, the measurement and regulation of PFA must be put into context in terms of the health risk. A wide ranging review of the health risks deriving from the manufacture and use of PFA in building materials was reported recently<sup>3</sup>. A summary of the conclusions is given below.

Exposure pathways were considered for three groups of people: workers manufacturing building products containing PFA; construction workers using such products; and members of the public living in houses built from these materials. Four exposure pathways were considered: external radiation and inhalation for all three groups, and skin contamination and inadvertent ingestion of dust for the worker groups. In all cases, the calculations were on the basis that PFA replaces other constituents that also contain naturally occurring radionuclides.

The annual excess doses are the quantities of interest. These were estimated to be 4.5  $\mu$ Sv y<sup>-1</sup> for workers manufacturing building materials, 1.7  $\mu$ Sv y<sup>-1</sup> for construction workers and approximately 200  $\mu$ Sv y<sup>-1</sup> for members of the public living in houses built from PFA-containing materials. In the last group, around one third of the dose was from the inhalation of radon-222 (66  $\mu$ Sv y<sup>-1</sup>) and the remainder due to external radiation from the building materials (135  $\mu$ Sv y<sup>-1</sup>). The values obtained in this study for the radiological content and exhalation characteristics of PFA are consistent with the source data used for the dose estimates above.

These doses must be put into context. The average annual radiation dose to a member of the public in the UK is 2.7 mSv, of which 50% is from radon. The inhalation dose of  $66~\mu\text{Sv}~\text{y}^{-1}$  quoted above corresponds to an increase in the radon level of 1.3 Bq m<sup>-3</sup>, which is less than 10% of the average indoor radon level<sup>20</sup> of 20 Bq m<sup>-3</sup> and less than 1% of the Action Level<sup>12</sup> of 200 Bq m<sup>-3</sup>. Although at 20 Bq m<sup>-3</sup> the dose contributions from the building materials and the ground are similar, the contribution from the latter can increase the indoor radon level by several orders or magnitude; radon concentrations of 2000 Bq m<sup>-3</sup> are not unusual in several parts of the UK <sup>20</sup>. Thus the excess contribution from PFA in the building materials is small.

The average dose to the UK population from external gamma irradiation is 350  $\mu Sv$  y , with a range of about 100-1000  $\mu Sv$  y . The estimate from the review is that the dose from external gamma irradiation is 758  $\mu Sv$  y from materials that do not contain PFA, which is in good agreement with UNSCEAR . The incorporation of PFA into the materials increases the estimate by 135  $\mu Sv$  y , which is an increase of less than 20%. The average gamma dose contributions from the natural radionuclides are not dissimilar, with 39% from potassium-40, 34% from the uranium-238 series and 27% from the thorium-232 series. However, the contribution from potassium-40 can vary greatly given the wide range of observed concentrations by mass in the PFA.

The health risks from the natural radionuclides in PFA are therefore mainly from the external gamma irradiation and inhalation of the radon decay products in buildings made from PFA-containing materials. The doses are sensitive to the potassium-40 concentration, which can vary widely. However, the extra dose contribution from the use of PFA is small and well within the normal building to building variation. Nevertheless, any change in the source of coal will have a significant impact on the

potassium-40 concentration, and only continuing surveillance would give information on the current situation.

One of the limitations of the current study was that PFA was studied but not PFA-containing building materials. Changes in the structure of the PFA that occur during manufacture might affect the exhalation of radon-222, which would impact upon the estimates of inhalation doses.

#### Conclusions

The conclusions are:

- The change of coal source for UK power stations has resulted in PFA with slightly higher radionuclide contents than those measured in the 1980s.
- The radon-222 exhalation characteristics are similar to those measured previously.
- Building materials made from PFA similar to the samples tested would not cause a significantly increased hazard to health.
- It is considered unlikely that any of the trigger levels under current UK legislation or European Union Directives will be exceeded.
- Given the wide geographical range of possible sources of coal, further continuing surveillance is recommended.

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