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The radium legacy: Contaminated land and the committed effective dose from the ingestion of radium contaminated materials

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ABSTRACT

The manufacture and use of radium in the early to mid-20th century within industrial, medicinal and recreational products have resulted in a large number of contaminated sites across a number of countries with notable examples in the USA and Europe. These sites, represent a significant number of unregulated sources of potential radiological exposure that have collectively and hitherto not been well characterised. In 2007, the Radioactive Contaminated Land (RCL) Regulations came into force in the UK, providing the statutory guidance for regulators to classify and deal with RCL. Here we report on results derived from digestion experiments to estimate committed effective dose, a key aspect of the RCL Regulations, from the ingestion of radium contaminated sources that can be found in the environment. This case study includes particles, clinker and artefacts that arise from past military activities on a site that was once an airfield at Dalgety Bay on the Firth of Forth, UK. Since 2011 the number of radium contaminated finds has increased by one order of magnitude on the foreshore areas of Dalgety Bay. The increase in finds may in large part be attributed to a change in monitoring practice. A subsample of sixty sources was selected, on the basis of their activity and dimensions, and subjected to digestion in simulated stomach and lower intestine solutions. The study demonstrated that more radium-226 (^{226}Ra) and lead-210 (^{210}Pb; driven by Polonium solubility) are dissolved from sources in artificial 'stomach' solutions compared with 'lower intestine' solutions. The combined 'gut' solubility for ^{226}Ra and apparent ^{210}Pb varied from less than 1% to up to 35% ICRP 72 conversion factors were used to convert the activities measured in solution to committed effective dose. A little over 10% of the sources tested dissolved sufficient radioactivity to result in 100 mSv committed effective dose to an infant. Using the solubility of 35% as a worst case, minimum source activities necessary to deliver 100 mSv to the full age range of users of the foreshore were estimated. All the estimated activities have been detected and recovered through routine monitoring.

1. Introduction

Radium, which is an alpha particle emitter, was frequently used in the early to mid-20th century as a luminescent paint (Harvie, 1999; Pratt, 1993). Research into the health effects of radium contamination in the human body started in the US in the mid 1920s, following the significant increase in morbidity and mortality amongst radium dial workers in New Jersey (Stebbings, 2001). The history of the radium dial painters, primarily women, has been well documented along with the follow-up health studies (e.g. Fry, 1998; Stebbings, 2001) and Rowland (1994) provides a classic review of the impact and health related research undertaken in the US. In the UK, radium production ended by the mid 1930s although imported radium salts continued to be used for medicine, industry and military purposes into the 1960s (Harvie, 1999).

In the UK, a number of possible radium contaminated sites were recognised by the Government in 1990 (House of Commons, 1990). A recent UK Government report (DECC, 2012)[1] estimated that there are between 150 and 250 contaminated sites across the UK associated with Ministry of Defence (MOD) activities and acknowledged that there may be as many as 1000 sites. These sites represent an important source of potential radiological exposure situations compared with the limited number of conventional nuclear sites, although there is uncertainty as to the number of sites and the consequential hazards they pose.

In 2007 the RCL[2] Regulations (Scottish Statutory Instruments, 2009) came into force in the UK, providing the environmental regulatory agencies with the statutory instrument to classify radioactively contaminated land and the duty to implement appropriate enforcement strategies. The Scottish Environment Protection Agency

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[2] Radioactively Contaminated Land.

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(SEPA) has used the criteria in the statutory guidance (Statutory Guidance to support the Radioactive Contaminated Land (Scotland) Regulations, 2007) to describe the amount of contamination that may give rise to significant harm to humans (Dale et al., 2009, 2011, 2012). For *homogeneous contamination*, significant harm is defined when lasting exposure gives rise to an individual dose exceeding one or more of the following criteria:

(a) An effective dose of 3 mSv per annum;
(b) An equivalent dose to the lens of the eye of 15 mSv per annum;
(c) An equivalent dose to the skin of 50 mSv per annum.

Contamination in the environment that is *heterogeneous* and associated with particles or artefacts complicates the assessment of contaminated land. Dale et al. (2008) proposed that the nature of the hazard along with the probability of encounter should also be considered within a risk assessment. For heterogeneous contamination the statutory guidance (Statutory Guidance to support the Radioactive Contaminated Land (Scotland) Regulations, 2007) provides thresholds above which significant harm is being caused irrespective of the probability of encounter. These thresholds are:

(d) The potential total effective dose is greater than 100 mSv; or
(e) The contact with contamination would result in a dose to the skin greater than 10 Gy hr^{-1}.

Should conditions (a) to (e) not be met then the probability of encounter should be taken into consideration and the possibility of harm is significant when the product of the dose and probability exceeds the thresholds described in (a) to (c) above. Current site management arrangements are also considered prior to any designation of contaminated land.

The spatial extent of radium contamination can be characterised by in-situ and mobile gamma spectrometric methods (e.g. Tyler, 2008) and techniques have been developed to detect and isolate spatially *heterogeneous* radioactive contamination including particles (Tyler et al., 2010). However, until the sites are identified, characterised and where necessary remediated, the risk of exposure to society remains. Whilst many routes of exposure are possible, ingestion of contaminated artefacts, materials and food products remains one of the primary areas of concern. The literature on the gastro-absorption of radium across the gut is limited (Rundo, 1999), with early studies based on soluble radium salts indicating the fractional absorption of gastro-absorption of between 0.25 and 0.35 (Seil et al., 1915). However, the availability of radium on contaminated sites is likely to be highly dependent on the physical and chemical forms of the radium, which may be controlled by the site history and subsequent environmental influences on the contaminated material.

Dalgety Bay (Fig. 1), on the north side of the Firth of Forth, is linked with ^{226}Ra contamination as the result of the wartime and post-war activities of the airfield operated by the RNAS Donibristle and HMS Merlin. The airfield closed in 1959 and the new town of Dalgety Bay was developed across the entire area in the 1960s and 1970s. The first ^{226}Ra contaminated particle was discovered and recovered from the foreshore at Dalgety Bay in 1990 as part of the baseline monitoring campaign by Babcock Engineering Services for the Rosyth Naval Base. Once it was identified as ^{226}Ra, its origin was attributed to the historical operations on the Donnibristle airfield. The National Radiological Protection Board (NRPB) undertook two surveys of the Bay in 1990 and 1991 and recovered 220 and 354 sources. Subsequent surveys have been undertaken by various contractors for the MOD during which sources are removed from the foreshore to provide some protection to beach users. Fig. 2 charts the annual finds of sources until September 2011. SEPA then initiated their own survey work at Dalgety Bay and in the period October 2011 to October 2012 and recovered over 800 sources from 36 surveys. The activity of sources recovered ranged from around 1 kBq to 76 MBq and Fig. 3 provides a Log_{10} distribution of the ^{226}Ra source activities. Over the same period since, several hundred additional sources were recovered by the MOD contractors over the 800 m coastline.

The physical origin of the radium sources is part of an on-going site investigation. Fig. 1 shows the locations where the majority of the sources have been recovered and are centred around the site of the sailing club. Contaminated materials range from sources (typically <3 mm in dimension) to clinker (up to a few 10s mm in dimension) from incineration which is occasionally associated with ash deposits. Wilson and Tyler (2012), describe SEM–EDS analysis on a small subsample of particles that shows some sources to have been incinerated whilst others appear to be intact and all sources have high concentrations of Zn and S, which can be attributed to radium paint.

Site contamination and any subsequent designation require an assessment of the likely doses that could be received by individuals, children or adults, through the accidental ingestion of contaminated material on the foreshore areas. An in vitro methodology is described to assess the solubility of sources. Results are presented from three separate batches of digestion experiments to assess the possible committed effective dose to an individual. The experiment is designed to mimic the acidic, enzymatic and temperature conditions of the stomach and small intestine. Whilst the approach to estimate solubility is not specifically novel, the complexity of the sources raises a number of measurement challenges and the results provide an important approach to enable the RCL Regulations to be interpreted.

2. Materials and methods

2.1. Source selection

The radium sources selected for digestion were initially selected on the basis of their ingestible size (Litovitz et al., 2010) and activity. In 2011, a total of 45 samples were collected from the MOD's contractor at Dalgety Bay, isolated from the surrounding sedimentary matrix and radium content estimated via gamma spectrometry. The size was measured by placing the sources on 1 mm graph paper and estimated via digital photography. From the 45 samples, a subsample of ten sources was selected for digestion within simulated stomach solutions. The selection covered a range in ^{226}Ra activities with source dimensions less than 5 mm to allow for ingestion. One exceptional item was a copper aircraft dial (Fig. 4), with 166 kBq of ^{226}Ra, to estimate the general availability of radium from such artefacts irrespective of size. Following the large number of finds made by SEPA later in 2011, a second batch of 30 physically smaller sources was selected. These were all less than 5.5 mm in dimension in the longest axis with ^{226}Ra activities ranging from 2 kBq to 50 kBq. Following the analysis from the first two batches, a third batch of 18 sources was selected in early 2012, focused around the estimated threshold activity likely to deliver a 100 mSv dose to an infant, all with dimensions less than 5 mm in the longest axis. To this set of 18, additional two sources were added: a 150 kBq of ^{226}Ra (2×2 mm) particle, resembling a centre pin head of a dial (Fig. 5); and a 1.26 MBq ^{226}Ra (10×7 mm) piece of clinker (Fig. 6). A total of 60 radium sources were digested and each batch of digestions included a blank sample.

2.2. Source digestion

The committed dose is considered from in vitro digestion of sources using simulated solutions for the stomach solution and small intestine solution. To ensure consistency with previous investigations on Dounreay hot particles (DPAG, 2006), the method for digestion was based on Harrison et al. (2005). The composition of the 'stomach' solution and 'small intestine' solution is presented in Tables 1 and 2 respectively. The chemicals listed in Table 1 were dissolved in 0.5 l of distilled water and adjusted to a pH 2 using hydrochloric acid for a final volume of 1 l. For the small intestinal solution, the chemicals listed in Table 2 were dissolved in 0.5 l of distilled water, adjusted to pH 7 with 5 M