The LLWR Environmental Safety Case

Assessment of Individual Radioactive Particles and WAC for Active Particles

T J Sumerling

LLWR/ESC/R(13)10056
August 2013

<table>
<thead>
<tr>
<th>Role</th>
<th>Name</th>
<th>Signature</th>
<th>Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Author</td>
<td>Trevor Sumerling</td>
<td>[Signature]</td>
<td>09.08.13</td>
</tr>
<tr>
<td>Checked by</td>
<td>Andy Baker</td>
<td>[Signature]</td>
<td>09.08.13</td>
</tr>
<tr>
<td>Approved by</td>
<td>Richard Cummings</td>
<td>[Signature]</td>
<td>09.08.13</td>
</tr>
</tbody>
</table>

© Copyright in this document belongs to the Nuclear Decommissioning Authority
This page is left blank intentionally.
Executive Summary

This report examines the biophysical and material constraints on radioactive particles that are present, or could realistically be present, in low-level waste (LLW). This provides the basis for assessments of committed effective doses from encounter with such particles in the environment, especially during the erosion of the LLWR. This is to support the development and application of waste acceptance criteria (WAC) and guidance to LLWR staff and consigners to identify wastes that could be a source of radioactive particles that could be of concern.

Detailed assessments of the potential radiological impacts of radioactive particles that might credibly be present in the LLWR have been made in a series of ESC memos. These assessments include the calculation of absorbed doses, effective doses and radiological risk from credible particles and small objects. We affirm that these assessments provide a complete and cautious assessment of the impacts from radioactive particles that may be present in past and future disposals at the LLWR, and show that assessed impacts are consistent with the environment agencies’ Guidance on Requirements for Authorisation (GRA), both during the erosion of the facility by coastal recession and in the event of human intrusion.

Having considered our assessments of radioactive particles and related issues of heterogeneity the Environment Agency has developed supplementary ‘Advice to Environment Agency Assessors on the Disposal of Discrete Items, Specific to the Low Level Waste Repository near Drigg’. The advice has been brought forward because the Environment Agency considers that the expected future evolution of the LLWR site, which is destruction by coastal erosion, raises issues that were not fully considered in developing the GRA.

The advice suggests, amongst other things, that for future disposals, an optimised approach should seek to prevent items individually carrying ‘a significant burden of radioactivity’ from being disposed of in the LLWR. This includes radioactive particles.

This report analyses the Environment Agency advice and proposes that an assessed effective dose from encounter in the range 3 mSv to 20 mSv provides a reasonable test to define what constitutes a significant burden of radioactivity. It is also shown that, based on results from the previous assessment and from biophysical principles, inadvertent ingestion of particles is the limiting consideration.

The report then revisits the evidence concerning particles that could, or in some cases are, present in wastes disposed in the LLWR. This provides the basis for realistically cautious assessments of the committed effective dose from ingestion of such particles. It is determined that only particles of high specific activity material bear sufficient activity to deliver a committed effective dose in the range 3 mSv to 20 mSv, and assessed realistically none of the particles deliver a committed effective dose above 20 mSv.

On this basis, WAC are proposed by which we will limit, as far as practicable, disposal of particles that individually carry an activity sufficient to give rise to a significant assessed effective dose. This includes a definition of ‘Active Particle’ and a requirement that waste consigners must consider the possibility that a waste may contain such particles. Waste containing or that may contain Active Particles may be accepted for disposal but only on approval of a Waste Consignment Variation Form.
This page is left blank intentionally.
Contents

1 Introduction .................................................................................................................. 5

2 Background .................................................................................................................. 7
   2.1 Previous assessments .............................................................................................. 7
   2.2 Environment Agency ‘Advice to Assessors’ .......................................................... 8
   2.3 Our approach to WAC ......................................................................................... 9

3 Biophysical constraints ............................................................................................... 11
   3.1 Inadvertent and deliberate ingestion ................................................................. 11
   3.2 Inhalation ............................................................................................................ 12
   3.3 Skin contact ....................................................................................................... 13
   3.4 Implications for assessment in support of WAC ................................................. 14

4 Material constraints ................................................................................................... 15
   4.1 Thorium-bearing mineral sands ........................................................................ 15
   4.2 Dalgety Bay radium finds .................................................................................. 16
   4.3 Sellafield active particle finds .......................................................................... 17
   4.4 Dounreay active particle finds .......................................................................... 18
   4.5 Uranium particles .............................................................................................. 19
   4.6 Spent fuel particles .......................................................................................... 20
   4.7 Higher activity materials ................................................................................. 22

5 Assessment .................................................................................................................. 23
   5.1 Particle cases .................................................................................................... 23
   5.2 Dose per unit ingestion and f1 values .............................................................. 25
   5.3 Committed effective doses from ingestion of the selected particles .......... 27
   5.4 Effect of particle size ...................................................................................... 30

6 Summary and WAC for Active Particles ................................................................. 31
   6.1 Summary of technical position ........................................................................ 31
   6.2 WAC for Active Particles .............................................................................. 32
   6.3 Assessment of variations .................................................................................. 33

References ..................................................................................................................... 35
This page is left blank intentionally.
1 Introduction

This report examines the biophysical and material constraints on radioactive particles that are present, or could realistically be present, in low-level waste (LLW). This provides the basis for assessments of committed effective doses from encounter with such particles in the environment, especially during the erosion of the LLWR. This is to support the development and application of waste acceptance criteria (WAC) and guidance to LLWR staff and consigners to identify wastes that could be a source of radioactive particles that could be of concern.

Detailed assessments of the potential radiological impacts of radioactive particles that might credibly be present in the LLWR have been made in a series of ESC memos [1,2,3]. These assessments include the calculation of absorbed doses, effective doses and radiological risk from credible particles and small objects. We affirm that these assessments provide a complete and cautious assessment of the impacts from radioactive particles that may be present in past and future disposals at the LLWR, and show that assessed impacts are consistent with the environment agencies’ Guidance on Requirements for Authorisation (GRA) [4], both during the erosion of the facility by coastal recession and in the event of human intrusion.

Having considered our assessments of radioactive particles and related issues of heterogeneity the Environment Agency has developed supplementary ‘Advice to Environment Agency Assessors on the Disposal of Discrete Items, Specific to the Low Level Waste Repository near Drigg’ [5]. The advice has been brought forward because the Environment Agency considers that the expected future evolution of the LLWR site, which is destruction by coastal erosion, raises issues that were not fully considered in developing the GRA.

The advice suggests, amongst other things, that for future disposals, an optimised approach should seek to prevent items individually carrying ‘a significant burden of radioactivity’ from being disposed of in the LLWR. This includes radioactive particles.

The structure of this report is as follows:

- Section 2 summarises our previous assessments of radioactive particles, analyses the Environment Agency’s ‘Advice to Assessors’ and sets out our proposed approach to WAC.

- Section 3 examines the biophysical constraints on exposure from radioactive particles, including consideration of inadvertent and deliberate ingestion, inhalation, skin contact. The implications for assessment of particles in support of WAC are summarised.

- Section 4 examines the material constraints. This includes revisiting the evidence on particles found on beaches in the vicinity of Sellafield and Dounreay and at Dalgety Bay, and considering particles that could be present in LLW, including irradiated fuel fragments.

- Section 5 assesses the committed effective dose from a range of credible particles employing both cautious uptake factors as used in previous assessments, plus more realistic values supported by the evidence reviewed in Section 4.
• Section 6 summarises the technical position, sets out proposed WAC and our application of the WAC. This includes a definition of 'Active Particle', requirements on waste consigners and the way in which we will assess variations.
2 Background

2.1 Previous assessments

Detailed assessments of the potential radiological impacts of radioactive particles that might credibly be present in the LLWR have been made in a series of ESC memos [1,2,3]. These assessments include the calculation of absorbed doses, effective doses and radiological risk from credible particles and small objects. The characteristics of the particles and objects were based on the characteristics of particles and objects actually found on beaches at Dounreay, Sellafield and Dalgety Bay, and items and particles known to be present in the LLWR (e.g. low activity sources and mineral sands). Potential radiological impacts were assessed using the methodology and habit data developed by the Health Protection Agency (HPA) for their assessment of items found on beaches in the vicinity of Sellafield [6,7] and also applied to assessment of items found at Dounreay and Dalgety Bay.

Our application of the methodology has been reviewed by the HPA [8] on behalf of the Environment Agency. The HPA review abstract states:

‘The main conclusion of the HPA review is that the assessments generally follow the approach used by the HPA in recent work on assessing radiological consequences from the use of beaches contaminated by radioactive objects. The reviewers found that the methodologies used in the assessment generally employ a suitable level of caution and include the most significant exposure scenarios, pathways and groups of people likely to be the most exposed. The reviewers suggested that more detailed information on some aspects of the methodology should be provided, including a discussion of the likely impact of exposure scenarios not considered in detail.’

Our previous assessment of radioactive particles and items during coastal erosion [1] concluded that, making cautious assumptions concerning possible radioactive particles or items and encounter with those particles or items:

- Calculated absorbed dose levels are such that no deterministic effects are expected as a result of likely contact times with skin and residence times in the gastrointestinal tract for any of the postulated radioactive particles. There is no possibility of absorbed doses exceeding any threshold for severe deterministic injury as defined in paragraph 6.3.40 of the Guidance on Requirements for Authorisation (GRA) [4].

- The highest effective doses are calculated for ingestion of small radioactive items or particles. Effective doses up to a few tens of mSv are calculated in some cases, and for very pessimistic assumptions exceeding 100 mSv, notably for the assumed ingestion by an infant of the highest activity alpha-rich particle found on beaches in the vicinity of Sellafield. Even assuming that the total inventory of relevant radionuclides in the LLWR is entirely in the form of such particles, the probability of encountering high-activity particles is very low. Realistically, although it is possible that a few such particles are present in the LLWR, they can only represent a small fraction of the inventory since the forms of disposals are known and relate mainly to surface contamination and dispersed radionuclides.
• The highest effective absorbed doses and effective doses due to inhalation of individual particles were shown to be much lower than can be delivered by particles in contact with skin or ingested particles. This is a function of the maximum size of particles that can be retained in contact with skin, ingested or inhaled; only very small particles can be inhaled and, given the same source material, these must necessarily bear very much lower levels of radioactivity.

• In all cases, the calculated radiological risks are consistent with, or well below, the risk guidance value of $10^{-6}$ per year set by the GRA.

2.2 Environment Agency ‘Advice to Assessors’

Having considered our assessments of radioactive particles and issues of heterogeneity, the Environment Agency came to the conclusion that the case of a repository, the expected future evolution for which was destruction coastal erosion, raised issues that were not fully considered in developing the GRA. The Environment Agency has therefore developed ‘Advice to Environment Agency Assessors on the Disposal of Discrete Items, Specific to the Low Level Waste Repository near Drigg’ [5]. The Environment Agency emphasise that the advice is supplementary to the GRA and that none of the requirements or guidance provided in the GRA is superseded by the advice. In particular, GRA Requirement R6, Risk guidance level after the period of authorisation, continues to apply to the coastal erosion scenario for both past and future disposals.

Reference [5] includes the following advice relevant to the control of radioactive particles in the future waste disposals:

‘For future disposals, an optimised approach is likely to entail preventing items individually carrying a significant burden of radioactivity from being disposed of in the LLWR. Equally, an optimised approach is likely to entail preventing any processes within the LLWR or as a result of coastal erosion that might lead to the production of high dose particles. These considerations may require improvements to waste characterisation and segregation, but we consider such improvements themselves to be part of the optimised approach. They are judged to be desirable in view of the expected scenario of coastal erosion.’

The advice states it is acceptable to use the GRA risk guidance level as a criterion for random encounters with particles, but also sets a ‘Test of Significance’ applicable to casual curiosity and deliberate searches, which includes the case of ‘a particle or multiple particles collected as a result of a deliberate search’:

‘As the test of significance, it is proposed that the assessed effective dose to any person during and after coastal erosion of the LLWR should not exceed a dose guidance level in the range of around 3 mSv/year to around 20 mSv/year.’

This is consistent with the guidance for human intrusion as set out under Requirement R7 in the GRA. The Advice further notes:

‘LLWR Ltd may wish to adjust its waste acceptance criteria for future disposals to the LLWR to make them consistent with the proposed test of significance.’
2.3 Our approach to WAC

Our approach to developing waste acceptance criteria (WAC) for individual characterisable items (i.e. larger items that are known to be present in the waste) is set out elsewhere. A basis for WAC for low-activity sources has been set out in the reference [9]. Our approach to the development of WAC for other characterisable, durable items that could be eroded onto the beach (e.g. hand tools, cemented waste drums, large metal items) is set out in reference [10].

This report is concerned with waste streams that might contain, or give rise to, high activity particles that will persist and provide a potential source of exposure if eroded onto the beach.

The assessments presented in the previous memos [1,2,3] were deliberately cautious and aimed at bounding the maximum credible doses and risks that might result from encounter with a range of realistic and hypothetical particles. The assessments included some very cautious assumptions, especially concerning the bioavailability of radionuclides present on particles. Here, consistent with the aim to support the development of WAC and guidance to consigners, we focus on particles that might realistically be present in significant numbers in a waste stream and move towards realistically cautious assessments of the maximum effective doses from encounter with such particles.

As a basis for WAC for active particles, we are concerned with inadvertent exposure to persons using the beach area as surveyed and assessed by HPA. We are not concerned with exposure, from unusual habits or very unlikely events, e.g. deliberate ingestion of small objects that also happen to be radioactive, or from finding and inspection of items that are visually identifiable as out of the ordinary, e.g. a low activity source disc. (The latter case is dealt with in reference [9].)

Following the Environment Agency ‘Advice’ that ‘an optimised approach is likely to entail preventing items individually carrying a significant burden of radioactivity from being disposed of in the LLWR’, we need to establish what constitutes ‘a significant burden’. Although it was not directly proposed for this purpose, the ‘test of significance’ of an assessed effective dose in the range of around 3 mSv/year to around 20 mSv/year seems a reasonable test to apply.

As shown in reference [1], the number of particles present in the repository that have the potential to individually give rise to a significant dose must be very low. Hence, it is unlikely that an individual will randomly encounter more than one such particle in a year, or even over a number of years. This indicates adopting 20 mSv as the criterion by which to decide what constitutes ‘a significant burden’. In considering WAC, however, and bearing in mind the uncertainties in estimating the activity on a particle and the assessed effective dose from encounter, we think the range 3 mSv to 20 mSv provides a reasonable ‘test of significance’ and is cautious. Thus, we should first identify any radioactive particles with the potential to lead to an assessed effective dose in the range of 3 mSv to 20 mSv or above, and then consider whether it is possible to identify waste streams that have the potential to include significant numbers of such particles.

Ultimately, the impact from random encounters with active particles will be assessed against the GRA risk guidance level, so that both the effective dose from encounter and annual probability of encounter are relevant. The Environment Agency, however, have suggested via their ‘Advice’ [5] that an optimised approach should
entail limiting, as far as practicable, disposal of particles that individually carry an activity sufficient to give rise to a significant assessed effective dose. We propose to do this by limiting or placing conditions on wastes that have potential to include significant numbers of particles that individually could give rise to assessed effective dose within or above the range of 3 mSv to 20 mSv.

It is not practical to prevent every single particle entering the LLWR. There is always the possibility of a single or few active particles in a waste consignment, but it would not be optimum to limit the disposal of wastes in which the frequency of such particles must be very low to zero.
3 Biophysical constraints

The radiation dose that can arise from single or multiple particles is limited by biophysical and material constraints. Biophysical constraints (discussed in this section) limit the dimensions of a particle that can reasonably be ingested, inhaled or retained in close contact to skin so that a radiation dose is received. Material constraints (discussed in Section 4) limit the specific activity of the material from which particles are composed and hence, combined with its dimensions, the maximum activity that may be present in a particle of such material.

The HPA give upper size limits for objects to be ingested, inhaled and to adhere to the skin in Table 27 of reference [7], reproduced here as Table 1; the values are supported by detailed evidence presented in Appendices of F, G and H of reference [7].

Table 1: Summary of the upper size limits for objects to be ingested, inhaled and to adhere to the skin (Table 27 from HPA reference [7])

<table>
<thead>
<tr>
<th>Route of exposure</th>
<th>Upper size limit (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inadvertent ingestion without detection in the mouth⁹</td>
<td>0.1</td>
</tr>
<tr>
<td>Inadvertent ingestion with detection in the mouth⁹</td>
<td>1</td>
</tr>
<tr>
<td>Deliberate ingestion, adult⁶</td>
<td>70</td>
</tr>
<tr>
<td>Deliberate ingestion, child⁶</td>
<td>40</td>
</tr>
<tr>
<td>Deliberate ingestion, young child⁶</td>
<td>20</td>
</tr>
<tr>
<td>Respirable size for inhalation (alveolar-interstitial region of the lungs)⁸</td>
<td>0.01</td>
</tr>
<tr>
<td>Respirable size for inhalation (extra thoracic airways)⁸</td>
<td>0.1</td>
</tr>
<tr>
<td>Lodging under fingernail⁹</td>
<td>1</td>
</tr>
<tr>
<td>Adhesion to skin⁹</td>
<td>1</td>
</tr>
</tbody>
</table>

(a) These object sizes were not found to be significantly dependent on the individual's age.
(b) Not considered for the general population of beach users (see Section 6.2).

3.1 Inadvertent and deliberate ingestion

Inadvertent ingestion of discrete objects might occur via the consumption of sand, for example, on food eaten on the beach, by hand-to-mouth transfer, or swallowing of windblown particles. The HPA [7] note that for the general population it is only appropriate to consider inadvertent ingestion of particles. Small, stone-sized objects might be deliberately ingested, for example by very young children or individuals with pica, but we do not consider this a useful test in developing WAC for radioactive particles.
On this basis, we adopt an upper size limit of 1 mm for particles that might be inadvertently ingested, as suggested by HPA. As noted in Table 1 and discussed in reference [7], a particle of this size is liable to be detected in the mouth, but may be swallowed especially if present with food.

### 3.2 Inhalation

If a particle is small enough then it may become airborne, for example, due to wind action or human activity, and thence may be inhaled or deposited in the mouth or throat if mouth breathing. As quoted in reference [7], particles with aerodynamic diameter larger than 30 µm deposit almost exclusively in the extra-thoracic region, which comprises the nasal passages, larynx, pharynx and mouth. In the ICRP Human Respiratory Tract Model (HRTM), a cut-off of 100 µm (0.1 mm) is suggested as an upper limit. Particles with aerodynamic diameters less than about 30 µm may deposit in the airways of the lung (i.e. the trachea, bronchi and bronchioles), but only particles with aerodynamic diameter smaller than 10 µm (0.01 mm) are likely to reach the alveolar region, see Figure 1.

![Diagram of respiratory system regions](Figure 1)

**Figure 1:** Percent regional deposition in each HRTM region for particles ranging in size from 1 to 100 µm aerodynamic diameter (note logarithmic scale) in wind speeds from 1 to 9 m s\(^{-1}\) (Figure G3 from HPA reference [7])

Figure 1 shows calculated percent regional deposition (probability of deposition) in each HRTM region for particles ranging in size from 1 to 100 µm aerodynamic diameter in wind speeds from 1 to 9 m s\(^{-1}\), for an adult undertaking light exercise, mouth breathing and facing into the wind; the spread of deposition in the extra-
thoracic (ET) regions shows the effect of variation in wind speed, which only affects larger particles.

Particles that deposit in anterior nasal passages (region ET1) are liable to be expelled (nose blowing). Particles that deposit in the posterior nasal passages, larynx, pharynx and mouth (ET2) are liable to be swallowed, i.e. ingested. Particles that deposit in the trachea and main bronchial passages (BB) will be cleared by action of the mucociliary escalator towards the laryngopharynx and thence swallowed. Only particles that reach and deposit in the bronchioles (bb) and alveolar region of the lungs (Al) can be regard as inhaled and give rise to dose to bronchiolar or alveolar tissues, or to other organs if absorbed into the body.

On this basis, HPA suggest an upper limit of 10 µm (0.01 mm) for a respirable particle to enter the alveolar-interstitial region of the lungs (see Table 1). Larger particles may be deposited in the extra-thoracic airways, but will be then be cleared and swallowed.

A further correction that is important for dense (heavy) particles is that the values given above refer to the aerodynamic diameter of the particle. The aerodynamic diameter of a particle is the diameter of a unit density spherical particle that has the same settling velocity in air. A denser (heavier) particle will tend to settle more rapidly in still air and, correspondingly, be more likely to impact on airway surfaces when breathed in due its higher momentum.

Calculations show that, a 10 µm uranium oxide particle, density about 10 g cm$^{-3}$, will behave aerodynamically as a 30 µm particle of unit density. An upper limit of 10 µm aerodynamic diameter for a respirable particle implies the largest uranium oxide particle that can be inhaled is about 3 µm. This is important because, for constant specific activity, the activity of a particle goes as the mass, which goes as the cube of the diameter. Thus the 3 µm uranium oxide particle bears only 1/30$^{1/3}$ of the activity of the 10 µm particle.

This reinforces the conclusion in our assessment of exposure to possible high activity particles on the beach [1], that inhalation can never be the limiting pathway when assessing absorbed or effective doses from individual radioactive particles. The effective dose due to inhalation can exceed the effective dose due to inadvertent ingestion in some circumstances, but this is for conditions in which large numbers of very small particles are inhaled. This is the situation modelled in our standard ‘continuum’ assessment models, not related to the existence or not of high activity particles.

### 3.3 Skin contact

From data reviewed in Appendix H of reference [7], HPA conclude that 1 mm is an appropriate upper size limit for particles to adhere to, or be retained in contact with, the skin (see Table 1).

As shown in Table 15 of reference [1], only the particle assumed to contain the entire activity of a typical Ra-226 source (0.2 MBq) gives rise to a calculated absorbed dose rate to skin such that the threshold for tissue effects of 2 Gy might be approached. (The particles the absorbed dose from which arises from Sr-90 and C-137 can be ruled out, because these radionuclides will have decayed to trivial levels by 300 years after disposal.)
As described in reference [1], this is a very cautious and bounding calculation; absorption in the particle is ignored and it is most unlikely that the entire source would be present on a particle 1 mm in diameter. A more realistic estimate of the activity that could be present on a 1 mm particle is that on particle S084 found on Dalgety Bay or on a particle of radium-zinc sulphide paint. Table 15 in reference [1] indicates that the particle of radium paint and the Dalgety Bay particle S084 would have to be in contact with the same patch of skin for 30 hours and 7 hours, respectively, to reach the threshold for tissue effects. SEPA measured a contact dose rate of 100 mGy/h for a particle of 80 kBq [14] (nearest equivalent in activity to the S084 particle), and this would give a contact time of 20 hours to reach the threshold for ulceration. The measured dose rates were a factor of three lower than calculated dose rates; this is due to self-absorption. Residency times on the skin and under nails for small items (1 mm) are likely to be up to a few hours and are unlikely to extend to 20 hours. Hence, no deterministic effects are likely to occur from a Ra-226 bearing particle contacting skin.

Reference [1] concludes that absorbed doses are such that no deterministic effects (tissue effects) are expected as a result of likely contact times with skin and residence times in the gastrointestinal tract for any of the postulated radioactive particles. It also shows that the effective doses from skin contact are very low on account of the very small area of skin that is irradiated.

### 3.4 Implications for assessment in support of WAC

We conclude that it is reasonable focus on assessing ingestion of active particles, since this mode of exposure will yield the highest effective doses. The calculated committed effective dose from ingestion can be compared with a ‘test of significance’, that the assessed effective dose from any individual particle should not exceed a level in the range of around 3 mSv to 20 mSv (see Subsection 2.3).

We adopt a 1 mm diameter particle as the reference assumption, since the HPA identify this as the largest size particle that is liable to be inadvertently ingested. We also comment on the sensitivity of effective doses immediately around this value. Smaller particles can be ingested, and much smaller particles, can be inhaled, but in both cases the committed effective doses will be lower.

The assessment of inadvertent ingestion of 1 mm particles gives an appropriate basis to inform WAC.

Committed effective doses from ingestion depend on the fraction of activity that is absorbed as the particle passes through the gastro-intestinal (GI) tract. If the particle dissolves completely in the GI tract then the uptake is characterised by the fractional absorption value, $f_1$, which is the fraction that is directly absorbed from the GI tract into the body fluids when it is in biologically available form. If the particle does not dissolve completely in the GI tract then only the dissolved component is available for uptake. If a particle present on the beach is readily soluble, then it will not remain on the beach for an extended period. Hence, the particles we are considering are not expected to dissolve completely in the GI tract. Information on particle solubility and potential uptake is given for the particle types considered in the following section.
4 Material constraints

The activity that can be present on any individual particle depends on the material from which it composed, or activity that could be present on its surface.

LLW includes low activity radioactive residues and generally lightly contaminated materials and items not arising from areas where highly radioactive materials are present. Some wastes in the LLWR are known to be in particle form, notably the thorium mineral sands disposed in Trench 2, but radioactive particles that individually might deliver a significant radiation dose are infrequent. As assessed in reference [1], based on the pessimistic assumption that the whole inventory of key radionuclides is present entirely in the form of 1 mm particles, it is calculated that during erosion of the LLWR there would only be one such particle in every tonne of sand.

The potential for higher activity particles is reviewed in Section 4 of reference [1] to determine the highest activity of any radionuclide that could be present on any particle. Here, we revisit and expand this evidence, focussing on the material form of the possible particles. This includes thorium mineral sands disposed in the trenches, particles such as found on the beaches at Sellafield, Dounreay and Dalgety Bay, and particles based on high activity nuclear material, fragments of which might be present in future waste. Hypothetical particles related to low activity sources or source fragments are not revisited, since the disposal of sources is controlled by WAC based on the characteristics of the sources [9].

4.1 Thorium-bearing mineral sands

The LLWR inventory reports [11] and [12] state that the monazite and thorite sands were disposed of in Trench 2 in 1960 and 1972-73 respectively as surplus to requirements.

Monazite is a phosphate mineral containing rare earths and is an important ore for thorium, lanthanum and cerium. It occurs usually as small isolated crystals as a placer deposit (formed by differential gravitational settling). It is relatively dense, about 4.6 to 5.7 g/cm$^3$. Grain size is as for other sands, typically 0.06 to 2 mm.

Reference [12] states that previous inventory estimates assumed the material is 5% thorium by weight and there seems there seems no reason to alter this as a best estimate to calculate the total inventory of thorium. The thorium content of monazite is variable, however. Commercial monazite sands typically contain between 6 and 12% thorium, although ores up to 20 to 30% are known whereas some monazite sands are essentially thorium-free.

Thorite is a rare nesosilicate of thorium and uranium that forms tetragonal crystal systems. Owing to differences in composition, the specific gravity varies from 4.4 to 6.6 g/cm$^3$. A variety of thorite, ‘uranothorite’, is particularly rich in uranium and has been a viable uranium ore, but the radioactivity of thorite arises mainly from the thorium content due to the more abundant and higher energy alpha and photon emissions of thorium.
Reference [12] states that previous inventory estimates assumed the material is 4.4% thorium by weight and there seems no reason to alter this as a best estimate to calculate the total inventory of thorium. The thorium content of thorite is variable, however, with concentrations up to 37% and 46% being reported.

### 4.2 Dalgety Bay radium finds

Radioactive material was first detected on a part of the foreshore at Dalgety Bay in 1990. Monitoring has been undertaken by SEPA and by the Ministry of Defence and periodically radioactive items have been removed. The contamination originates from the residue of radium-luminised instrument panels and other items from military aircraft incinerated and land-filled in the area at end of World War II.

Radium based luminescent paint was typically made by mixing a radium salt, zinc sulphide and a carrier material (typically varnish or lacquer). The specific activity of radium paint used in the UK is known from Admiralty specification and luminising practice. As described in reference [13]: "zinc sulfide was mixed with a radium salt to yield products containing about 25 to 300 µg [one µg is essentially the same as one µCi] of radium element per gram. The more concentrated preparations (generally containing 215 µg of radium per gram to meet British Admiralty standards) were used on aircraft and ship instrument dials, while lower-grade materials containing about 50 to 100 µg of radium per gram of mixture were used on watches, switch markings, and other devices requiring less critical reading."

Particle and small stone-sized radium bearing objects, plus larger items, including identifiable objects such as radium-luminised dials and levers have been recovered from the beach and shore areas of the bay.

Up to 2010, the highest specific activity of the measured particles recovered from Dalgety Bay is particle S084, which is about 1 mm x 2 mm in size and contains about 50 kBq Ra-226 [14]. The mass of S084 is not given in reference [14], but based on dimensions, activity and assumed density the specific activity is in the range 4 to 7 MBq/g (depending on assumed density), which converts to 100 to 200 µCi/g, i.e. 100 to 200 µg of radium per gram. This is consistent with particle being a fleck of incinerated radium paint.

Since 2010 (in 2011 and 2012) a larger number of higher activity items has been recovered [15]; SEPA advances several reasons as to why this is the case. SEPA comments that items recovered from Dalgety Bay are highly heterogeneous in nature, both in terms of physical properties (size, mass, solubility) and radioactive content, with no apparent direct relationship between these variables.

Experimental measurements have been made on the dissolution of radium on recovered items in hydrochloric acid and in a representative gut solution, developed by the HPA. A range between zero and 36 % has been found. Analysis of the data assuming a log-normal distribution yields a mean of 8% and standard deviation of 2.54. The upper 95th percentile of the distribution is about 20%, i.e. 95% of the items have a solubility of less than 20% [15].

The bulk of radium in the LLWR is associated with process wastes disposed in the trenches.
In the 2011 ESC inventory, based on the 2007 UKRWI, the principal source of Ra-226 in future wastes was Stream 7S01 (contaminated soil, ash and rubble from Defence estates), which contributed 1.93 TBq of Ra-226. A much lower estimate of the activity of this stream is given in the 2010 UKRWI [16] although this may at least partly relate to change in land ownership and hence liability. In any case, in future, other waste streams containing Ra-226 may be identified for which disposal at the LLWR could be an option. Some of these wastes could contain radium luminised items or debris from ex-radium luminising sites.

4.3 Sellafield active particle finds

As described in reference [17], investigations have been undertaken into radioactive particles and stones (collectively referred to as ‘finds’) discovered on beaches close to Sellafield since routine strandline monitoring since the mid-1980s. Following the discovery of a relatively high activity particle on the beach near to Sellafield in July 2003, a more intensive study was launched and an improved large area beach monitoring technique introduced.

Using this improved technique, a total of 609 finds have been recovered for analysis to the end of March 2009. These finds comprise 356 stones (defined as ≥2mm in any dimension) and 253 particles (<2mm). Finds with an Am-241 activity greater than Cs-137 activity are classified as “alpha-rich”. Of the total finds, 56 have been designated as alpha-rich (2 stones and 54 particles).

The most likely source of the finds is from historical liquid effluent discharges made before more effective abatement techniques were introduced to minimise free solids. Alpha-rich finds that contained sufficient actinide concentrations (notably Pu-238, Pu-239, Pu-241 as well as Am-241) were matched to the most likely type and average burn-up of fuel and reprocessing operations at Sellafield. Based on the results, it is concluded that all alpha-rich finds studied are associated with reprocessing operations between 25 and 45 years ago.

At this time, finely dispersed ferric hydroxide flocs were produced in the liquid effluent treatment facilities through the neutralisation by ammonia and aluminium hydroxide. These flocs would have trapped actinides in their structure. Prior to 1975 these neutralised effluents were routinely discharged to sea (via the pipelines) without an effective means of removing the floc. After 1975 settling tanks were used to separate sludge, although this process is believed to have been inefficient. A suitable method (e.g. ultrafiltration) was not commercially available until the mid to late 1980s.

The collected evidence from characterisation of the finds and process knowledge is that the alpha-rich particles are most likely combinations of naturally occurring beach material (e.g. silt, sand) and alpha-rich ferric hydroxide flocs produced through neutralisation of aqueous raffinates. These solids were discharged mainly prior to 1975. In addition, some particles trapped with corrosion products in the old sea discharge lines might have been released into the environment during the removal of the lines between the early 1990s and 2006.

There is no reason to think particles of this type are present in future wastes coming to the LLWR. However, we retain the Sellafield alpha-rich particle in the assessment in order to provide a link with our earlier particle assessment in reference [1]. The maximum activity in an alpha-rich particle found on beaches in the vicinity of Sellafield is 84 kBq Pu-238, 309 kBq Pu-239/240 and 634 kBq Am-241 (a total of...
approximately 1 MBq) [7]. Minimum detectable activities (MDAs) are given in Appendix A of reference [7] for objects:

- on the beach surface, 11 kBq Cs-137 and 37 kBq Am-241;
- at greater depth, 1.3 MBq Cs-137 (at 0.3 m) and 31 MBq Am-241 (at 0.15 m).

This indicates that there could be a significant population of alpha-rich (Am-241 > Cs-137) particles with lower activity that are not detected. That is, the 1 MBq is probably not representative, although it is a reasonable upper value to carry forward to assessment in this report.

Studies have been carried out to evaluate the solubility of fragments associated radionuclides in the gut by both in vitro methods (using solutions representing GI tract fluids) and in vivo methods (determining uptake in rats). Reviewing these, the HPA recommended an uptake fraction, \( f_1 \), of \( 3 \times 10^{-5} \) should be used for both americium and plutonium from the beach particles [6], which is still considered cautious, this being the highest value obtained from in vitro studies [7].

### 4.4 Dounreay active particle finds

Active particles have been found on beaches in the vicinity of Dounreay since the 1980s. As summarised on the Dounreay Site website [18], the discovery of much higher activity particles on the seabed in the late 1990s led to an extensive programme of investigation and consultation to determine the best practicable environmental option; this was determined as recovering the particles from the area of seabed where the highest population of active particles has been found. The most hazardous fragments are located close to an old discharge point on the seabed. Their disintegration is believed to be the source of the smaller, less hazardous particles detected on local beaches.

The particles are fragments of irradiated cladding and nuclear fuel discharged to sea as a result of reprocessing practices during the 1960s and 70s. The used fuel from the Materials Test Reactor (MTR) was dismantled underwater in ponds. The underwater dismantling techniques generated fragments, known as swarf, consisting of metal cladding and occasionally some of the underlying fuel, which accumulated in the bottom of the ponds. The ponds were periodically drained to allow the pond to be cleaned out, and the bulk of the accumulated swarf was recovered. However, some swarf was discharged from the pond with the drained water. This was routed to settling tanks that were designed for removal of solids before discharging liquid radioactive effluent to sea. Hundreds of thousands of particles probably entered the drains connected to this system but the number of particles that was released to the sea from the settling tanks is not known.

The isotopic ratios on the particles points to the release of particles occurring primarily from fuel dismantling in the late 1960s, but further releases into the active drainage system before filtration was fitted at the source facilities in 1984 cannot be ruled out. Particles could have been held up in the effluent system and may have been released after this.

The majority of the fragments retrieved from the local beach at Sandside Bay initially came from the processing of fuel from the Materials Test Reactor (MTR). As the research and monitoring programme developed, there was an increase in the proportion of the number of fragments found that originated from the Dounreay Fast
Reactor (DFR). Fragments that are of DFR origin may be characterised by the presence of Nb-94, although the absence of this radionuclide does not necessarily mean that the fragment came from the MTR. Both types of fragment contain Cs-137 together with Sr-90 and its decay product Y-90; if such fragments came into contact with tissue most of the assessed dose came from Sr-90 and Y-90 [19].

Studies have been carried out to evaluate the potential solubility of spent fuel fragments and associated radionuclides in the gut by both in vitro and in vivo methods. Results as summarised in Table 6 of reference [19] are reproduced in Table 2; their basis is set out in reference [20].

Table 2: Results as summarised in Table 6 of reference [19]; best single estimate and ranges

<table>
<thead>
<tr>
<th>Fragment type</th>
<th>% of ingested activity absorbed to blood</th>
<th>137Cs</th>
<th>90Sr</th>
<th>239Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTR</td>
<td>1 (0.02 - 2)</td>
<td>0.01</td>
<td>0.001 (0.001 - 0.03)</td>
<td>0.001 (5x10^-4 - 5x10^-3)</td>
</tr>
<tr>
<td>DFR</td>
<td>0.03 (0.001 - 0.05)</td>
<td>0.003</td>
<td>0.003 (0.001 - 0.02)</td>
<td>0.001 (5x10^-5 - 5x10^-4)</td>
</tr>
</tbody>
</table>

As discussed in reference [1], the maximum activity of Cs-137 in a fuel fragment found on beaches in the vicinity of Dounreay is 100 MBq (3 mm diameter), and a more representative value is 100 kBq (300 micron diameter). Specific activities for Cs fuel fragments found on the Dounreay beaches are about 2 GBq/g [20]. This specific activity indicates that the activity on a 1 mm diameter particle from a fuel fragment would be about 3 MBq. However, after 300 years the Cs-137 activity would have decayed to 0.001 of its initial value, i.e. about 3 kBq, a rather low activity.

The maximum Pu-239/240 activity found on a particle on beaches in the vicinity of Dounreay is about 100 kBq, and a more representative value is 100 Bq. In reference [1], it is estimated that 1 mm diameter particle of this material would contain about 9 kBq Pu-238, 3 kBq Pu-239 and 3 kBq Am-241.

4.5 Uranium particles

We have no example of uranium bearing particles being identified as a problem in the environment. The LLWR, however, contains a significant total inventory of uranium, and uranium is present in many future waste streams. Thus, it is useful to examine whether uranium particles, individually, have the potential to deliver a significant effective dose if encountered in the environment.

Table 3 shows the isotopic composition of natural uranium and uranium enriched to 3.5% U-235, which is typical of enrichment of PWR fuels. Composition by mass is as given in reference [21] and composition by activity is calculated.
Table 3: Isotopic composition of natural uranium and 3.5% enriched uranium by mass and by activity

<table>
<thead>
<tr>
<th>Isotope</th>
<th>% by mass from reference [21]</th>
<th>Specific activity Bq per g U</th>
<th>% by total U isotope activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-234</td>
<td>0.0054%</td>
<td>1.24E+04</td>
<td>49.03%</td>
</tr>
<tr>
<td>U-235</td>
<td>0.7204%</td>
<td>5.76E+02</td>
<td>2.27%</td>
</tr>
<tr>
<td>U-238</td>
<td>99.2742%</td>
<td>1.23E+04</td>
<td>48.70%</td>
</tr>
<tr>
<td></td>
<td>100.0000%</td>
<td>2.54E+04</td>
<td>100.00%</td>
</tr>
</tbody>
</table>

3.5% U-235 enriched uranium

<table>
<thead>
<tr>
<th>Isotope</th>
<th>% by mass from reference [21]</th>
<th>Specific activity Bq per g U</th>
<th>% by total U isotope activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-234</td>
<td>0.0290%</td>
<td>U-234</td>
<td>6.68E+04</td>
</tr>
<tr>
<td>U-235</td>
<td>3.5000%</td>
<td>U-235</td>
<td>2.80E+03</td>
</tr>
<tr>
<td>U-238</td>
<td>96.4710%</td>
<td>U-238</td>
<td>1.20E+04</td>
</tr>
<tr>
<td></td>
<td>100.0000%</td>
<td></td>
<td>8.16E+04</td>
</tr>
</tbody>
</table>

Uranium could be present in a number of forms. Uranium ore minerals are uraninite (UO$_2$) or pitchblende (U$_3$O$_8$). Yellowcake is typically 70 to 90 percent U$_3$O$_8$ by weight with other compounds formed during the precipitation of the ore leach solutions.

Uranium metal (19 g/cm$^3$) is the most dense form possible, i.e. would give rise to the highest activity on a 1 mm particle. However, the metal corrodes in air to form UO$_2$, which spalls from the surface. In damp conditions, a corrosion rate of about 1 $10^{-3}$ mg/cm$^2$/h is indicated, which will lead to complete disintegration of a 1 mm metal particle in less than a year. A reasonable choice, therefore, is to assume UO$_2$ in the form of a uraninite particle or fuel fragment, either of which has a density of about 11 g/cm$^3$.

4.6 Spent fuel particles

Spent (irradiated) nuclear fuel has been identified as the source of radioactive particles in the environment at Dounreay (Subsection 4.4). The activity of used fuel particles, and the potential hazard they present, depends on the source of the fuel, especially the fuel type and irradiation history.

Used fuel contains a range of radionuclides. At early times, on a scale of decades, the radiological hazard from the fuel is dominated by fission products, principally Sr-90 and Cs-137. At later times, beyond about 100 years (although the time depends on fuel type and burn up), the radiological hazard is dominated by the major alpha-emitting actinides. This is illustrated in Table 4, which shows the activity of selected radionuclides in a medium burn up AGR fuel at 10 and 310 years after discharge from the reactor (g/t U at 10 years from reference [22], GBq/t U at 10 and 310 years calculated).
Table 4: The relative activity of selected radionuclides in a medium burn up AGR fuel at 10 and 310 years after discharge from the reactor

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>10 years after discharge from the reactor</th>
<th>310 years after discharge</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>g/t U initial</td>
<td>% of mass</td>
</tr>
<tr>
<td>Sr-90</td>
<td>2.28E+02</td>
<td>0.023%</td>
</tr>
<tr>
<td>Tc-99</td>
<td>4.25E+02</td>
<td>0.043%</td>
</tr>
<tr>
<td>Ag-108m</td>
<td>1.83E-03</td>
<td>0.000%</td>
</tr>
<tr>
<td>I-129</td>
<td>8.63E+01</td>
<td>0.009%</td>
</tr>
<tr>
<td>Cs-137</td>
<td>5.11E+02</td>
<td>0.051%</td>
</tr>
<tr>
<td>U-234</td>
<td>1.17E+02</td>
<td>0.012%</td>
</tr>
<tr>
<td>U-235</td>
<td>5.64E+03</td>
<td>0.564%</td>
</tr>
<tr>
<td>U-238</td>
<td>9.68E+05</td>
<td>96.800%</td>
</tr>
<tr>
<td>Pu-238</td>
<td>3.25E+01</td>
<td>0.003%</td>
</tr>
<tr>
<td>Pu-239</td>
<td>2.70E+03</td>
<td>0.270%</td>
</tr>
<tr>
<td>Pu-240</td>
<td>1.54E+03</td>
<td>0.154%</td>
</tr>
<tr>
<td>Am-241</td>
<td>2.82E+02</td>
<td>0.028%</td>
</tr>
</tbody>
</table>

* Radionuclide contributing more than 1% to the activity of the listed radionuclides in blue font.

The results in Table 4 indicate that at about 300 years or more after disposal, the total activity of the used fuel is dominated by the actinides (Pu-238, Pu-239, Pu-240, Am-241). Since the ICRP dose per unit ingestion factors of the actinides are typically an order of magnitude greater than from any of the fission products, the radiological hazard from ingestion of a particle of used fuel will also be dominated by the actinides.

A more detailed examination of actinide activities in spent fuel has been made based on data given in reference [23]. The reference gives calculated isotopic composition spent fuel, including all significant uranium and actinide isotopes by % mass, for a range of spent fuels. Selecting three fuel types that are representative of common UK fuel types, the data in reference [23] have been used to calculate isotopic composition by activity of each of the fuels at 300 year after discharge, including taking account of relevant decay chains, e.g. Pu-241 -> Am-241 -> Np-237, Am-242m -> Pu-238 -> U-238 etc. This leads to the expected conclusion that, at 300 years, the total actinide activity is dominated by Pu-238, Pu-239, Pu-240 and Am-241; the uranium isotope activity is more than three to four orders of magnitude lower, depending on fuel type.

Table 5 shows the actinide activity in three representative UK fuel types at 300 years after discharge from the reactor.

---

Table 5: Activity of the major actinides in three representative UK fuel types at 300 years after discharge from the reactor

<table>
<thead>
<tr>
<th>Reactor type: % U-235; burn up</th>
<th>Activity: GBq per initial t U at 300y</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pu-238</td>
</tr>
<tr>
<td>PWR: 3.40%; 35 GW.d/t</td>
<td>1.58E+04</td>
</tr>
<tr>
<td>AGR: 2.40%; 20 GW.d/t</td>
<td>4.31E+03</td>
</tr>
<tr>
<td>Magnox: 0.71%; 6 GW.d/t</td>
<td>1.93E+03</td>
</tr>
</tbody>
</table>

PWR and AGR fuel pellets are composed of uranium dioxide (UO$_2$) powder compacted to cylindrical pellets and sintered at high temperatures to produce ceramic pellets with a high density and well defined properties. Magnox fuel is uranium metal, but as discussed above this is not stable in the presence of oxygen or water and corrodes to UO$_2$, but the corrosion products will be less compact than UO$_2$ fuel pellets and of uncertain properties.

The sum of actinide activities for each fuel type in Table 5 corresponds to 160, 64 and 36 MBq/g respectively. The volume of a 0.5 mm radius spherical particle is $5.2 \times 10^{-4}$ cm$^3$. Hence, for the PWR and AGR fuel pellets, density 11 g/cm$^3$, this gives the activity of a 1 mm diameter spherical particle as 0.90 and 0.37 MBq respectively.

4.7 Higher activity materials

Other, higher activity materials could be considered, notably MOX fuels, irradiated highly-enriched uranium from materials test reactors and from naval reactors, plus weapons grade enriched uranium and plutonium. The range of materials is quite wide and assessment of such particles is outside the scope of this report.

It can be stated that all these materials, if able to form stable durable particles, and mechanisms for creating such particles exist during material and waste management processes, could be a source of active particles that are more hazardous than the PWR fuel particle that is assessed in this report.
5 Assessment

5.1 Particle cases

Based on information presented in the previous section, we select the following possible and hypothetical particles for assessment.

We know from the previous assessment of particles [1] that individual monazite or thorite sand particles do not bear sufficient activity to give rise to a significant committed effective dose. We retain them in this assessment because they are present in the LLWR. Three cases are considered:

- LLWR monazite or thorite (approximately 5% Th by weight);
- High thorium monazite (30% Th by weight);
- High thorium thorite (40% Th by weight).

Based on finds at Dalgety Bay, and knowing that wastes from clean up of radium contaminated sites may be considered for disposal at the LLWR, we assess particles of radium paint. Two cases are considered:

- Radium-zinc sulphide paint at 50 µg/gram;
- Radium-zinc sulphide paint to Admiralty specification (215 µg/gram).

There is no reason to think particles of the type found on beaches near to Sellafield could be present in future wastes coming to the LLWR. However, we include the highest activity Sellafield alpha-rich particle in the assessment in order to provide a link with our previous assessment [1]. One case is considered:

- The highest activity alpha-rich particle found near to Sellafield, bearing about 1 MBq Pu and Am alpha.

The Dounreay beach particles consist of fragments of cladding, sometimes with fuel attached. Although it is not intended, it is possible that particles of cladding and spent fuel could be present in some wastes accepted at the LLWR. The specific activity of such a particle will depend on the fuel type, irradiation history and the fraction of particle that is fuel. We include one case:

- The highest Pu and Am activity particle found on a beach near to Dounreay, bearing about 0.015 MBq Pu and Am alpha.

Uranium is present in many future waste streams. It is useful to examine whether uranium particles, individually, have the potential to deliver a significant committed effective dose if ingested. Two cases are considered:

- Uraninite or fuel fragment (UO₂) with isotopic composition of natural uranium;
- Uraninite or fuel fragment (UO₂) with isotopic composition of uranium enriched to 3.5% U-235.

Although it is not intended, it is possible that particles of spent fuel could be present in some wastes accepted at the LLWR. It is shown in Subsection 4.6 that, at 100
years or more after discharge from the reactor, the radiological hazard from ingestion is dominated by the major actinides. The activity present depends on the fuel type and irradiation history. Three cases are considered:

- PWR fuel (UO$_2$) enriched to 3.40% U-235 and burn up to 35 GW.d/t;
- AGR fuel (UO$_2$) enriched to 2.40% U-235 and burn up to 20 GW.d/t;
- Magnox fuel (isotopic composition of natural uranium) with burn up to 6 GW.d/t.

Magnox fuel is uranium metal but, as discussed in Subsection 4.6, this is not stable and will corrode to UO$_2$, although the density and integrity of the corrosion products is uncertain.

Table 6 summarises the selected particles. In each case a 1 mm diameter particle is assumed.

### Table 6: Particles for assessment: specific activity of source material and particle activities ‘at disposal’ and 300 years later

<table>
<thead>
<tr>
<th>Particle materials</th>
<th>Primary radionuclides</th>
<th>Specific activity of material</th>
<th>Activity of 1 mm particle</th>
<th>Activity of particle after 300 years</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLWR monazite/thorite (~5% Th)</td>
<td>Th-232</td>
<td>2.0E-04</td>
<td>5.5E-07</td>
<td>No change (negligible decay)</td>
</tr>
<tr>
<td>High thorium monazite (30% Th)</td>
<td>Th-232</td>
<td>1.2E-03</td>
<td>3.3E-06</td>
<td></td>
</tr>
<tr>
<td>High thorium thorite (40% Th)</td>
<td>Th-232</td>
<td>1.6E-03</td>
<td>4.7E-06</td>
<td></td>
</tr>
<tr>
<td>Radium ZnS paint (50 µg/g)</td>
<td>Ra-226</td>
<td>1.8E+00</td>
<td>3.9E-03</td>
<td>3.4E-03</td>
</tr>
<tr>
<td>Admiralty specification (215 µg/g)</td>
<td>Ra-226</td>
<td>7.9E+00</td>
<td>1.7E-02</td>
<td>1.5E-02</td>
</tr>
<tr>
<td>Sellafield alpha-rich particle</td>
<td>Pu-238</td>
<td>Not known</td>
<td>8.4E-02</td>
<td>7.8E-03</td>
</tr>
<tr>
<td></td>
<td>Pu-239+240</td>
<td></td>
<td>3.1E-01</td>
<td>3.1E-01</td>
</tr>
<tr>
<td></td>
<td>Am-241</td>
<td></td>
<td>6.3E-01</td>
<td>3.9E-01</td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td></td>
<td>1.0E+00</td>
<td>7.1E-01</td>
</tr>
<tr>
<td>Dounreay cladding/fuel particle</td>
<td>Pu-238</td>
<td>Not known</td>
<td>9.0E-03</td>
<td>8.4E-04</td>
</tr>
<tr>
<td></td>
<td>Pu-239+240</td>
<td></td>
<td>3.0E-03</td>
<td>3.0E-03</td>
</tr>
<tr>
<td></td>
<td>Am-241</td>
<td></td>
<td>3.0E-03</td>
<td>1.9E-03</td>
</tr>
<tr>
<td></td>
<td>Total</td>
<td></td>
<td>1.5E-02</td>
<td>5.7E-03</td>
</tr>
<tr>
<td>Natural uranium as UO$_2$</td>
<td>U-234</td>
<td>1.2E-02</td>
<td>6.3E-05</td>
<td>No change (negligible decay)</td>
</tr>
<tr>
<td></td>
<td>U-235</td>
<td>5.8E-04</td>
<td>2.9E-06</td>
<td></td>
</tr>
</tbody>
</table>
### 5.2 Dose per unit ingestion and $f_1$ values

The committed effective dose per unit ingestion depends on the fraction of radionuclide taken up from the GI tract into the blood and thence to other body tissues; this is denoted $f_1$.

For most of our previous assessment calculations for particles, as presented in references [1,2,3], the committed effective dose per unit ingestion coefficients used were taken from the compilation in reference [24]. This compiles the dose per unit ingestion coefficients assuming the most readily biologically assimilated form of radionuclides as given by IRCP for members of the public [25]. This is generally appropriate for ingestion of radionuclides in groundwater and in foodstuffs, but will be cautious in respect of active particles that might be present in the repository, or
dispersed on the beach. In this case, the particle itself provides an enclosing matrix for the radionuclide and the radionuclides are likely to be in oxide and other inorganic forms that are not readily absorbed from the GI tract. That is, the $f_1$ values assumed in previous particle assessment calculations were cautious, in some cases very cautious.

Dose coefficients for intakes of radionuclides by workers are given in reference [26] and consider of a wider range of $f_1$ values, in light of the different forms of radionuclides that may be present in the workplace. Dose coefficients for intakes by workers and members of the public, based on ICRP Publication 60 primary guidance, have recently been collected together in ICRP Publication 119 [27]. In addition, as discussed in Section 4, for particles found at Dalgety Bay, Sellafield and Dounreay, the fraction of particle activity that is dissolved in gut or simulated gut conditions, i.e. an $f_1$ value, has been determined experimentally.

Table 7 compares $f_1$ values for radionuclides of interest as given by ICRP, determined experimentally and adopted here for realistic assessment.

Column (1) shows $f_1$ values as given in ICRP Publication 72 for adult members of the public, which presumes environmentally dispersed radionuclides; these are the values incorporated in the dose per unit ingestion coefficients compiled in reference [24] used in previous particle assessments. Column (2) shows values as given in ICRP Publication 68 for workers in light of the different forms of radionuclides that may be present in the workplace. Column (3) shows values derived from measurements on particles recovered at Dalgety Bay, Sellafield and Dounreay, as discussed in Section 4.

<table>
<thead>
<tr>
<th>Particle</th>
<th>Relevant elements</th>
<th>(1) Reference</th>
<th>(2) ICRP 68 values</th>
<th>(3) Particle measurement</th>
<th>(4) Adopted</th>
</tr>
</thead>
<tbody>
<tr>
<td>Monazite / thorite</td>
<td>Thorium 5E-4</td>
<td>5E-4</td>
<td>2E-4</td>
<td>none</td>
<td>2E-4</td>
</tr>
<tr>
<td></td>
<td>Radium 0.2</td>
<td>0.2</td>
<td></td>
<td></td>
<td>2E-3</td>
</tr>
<tr>
<td>Radium paint</td>
<td>Radium 0.2</td>
<td>0.2</td>
<td></td>
<td>0.08</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>Lead 0.2</td>
<td>0.2</td>
<td></td>
<td></td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>Polonium 0.5</td>
<td>0.1</td>
<td></td>
<td></td>
<td>0.08</td>
</tr>
<tr>
<td>Sellafield alpha rich</td>
<td>Plutonium 5E-4</td>
<td>5E-4</td>
<td>1E-4  1E-5</td>
<td>3E-5</td>
<td>3E-5</td>
</tr>
<tr>
<td></td>
<td>Americium 5E-4</td>
<td>5E-4</td>
<td></td>
<td></td>
<td>3E-5</td>
</tr>
<tr>
<td>Dounreay alpha</td>
<td>Plutonium 5E-4</td>
<td>5E-4</td>
<td>1E-4  1E-5</td>
<td>1E-5</td>
<td>1E-5</td>
</tr>
<tr>
<td></td>
<td>Americium 5E-4</td>
<td>5E-4</td>
<td></td>
<td>none</td>
<td>1E-5</td>
</tr>
<tr>
<td>Uranium</td>
<td>Uranium 0.02</td>
<td>0.02</td>
<td>0.002</td>
<td>none</td>
<td>0.002</td>
</tr>
<tr>
<td>Spent fuel</td>
<td>Plutonium 5E-4</td>
<td>5E-4</td>
<td>1E-4  1E-5</td>
<td>none</td>
<td>1E-5</td>
</tr>
<tr>
<td></td>
<td>Americium 5E-4</td>
<td>5E-4</td>
<td></td>
<td></td>
<td>1E-5</td>
</tr>
</tbody>
</table>

Column (4) shows $f_1$ values that we consider realistic. The rationale for each of the adopted values is as follows.
For the thorium in the monazite / thorite mineral particles we adopt $2 \times 10^{-4}$, representative of oxide and other slowly released forms. For radium (Ra-228) present with the thorium a value of $2 \times 10^{-3}$ is adopted, taking some credit that dissolution of radium will be controlled by its association within a thorium mineral matrix. (According to the relative half-lives of Ra-228 and Th-232 there is only atom of radium for every $4 \times 10^{10}$ atoms of thorium, i.e. radium dissolution will be limited by the surrounding thorium.) No credit has been taken specifically for the nature of the mineral particle, which has been stable over geological timescales, thus the adopted $f_1$ values are still likely to be cautious.

For radium in paint we adopt the mean measured value obtained from particles recovered from Dalgety Bay, see Subsection 4.2. For lead and polonium it is reasonable to assume the same $f_1$ value as for radium. This is based on ICRP 68 $f_1$ values for workplace forms of the radionuclides and that, as discussed for thorium, the mass of lead and polonium present is small compared to that of radium, and all radionuclides are within the same matrix.

For plutonium and americium in the Sellafield alpha-rich particle we adopt the HPA recommended $f_1$ value based on measured values obtained from particles recovered from Sellafield, see Subsection 4.3.

For plutonium and americium in the Dounreay highest alpha particle we adopt the HPA recommended $f_1$ value based on measured values obtained from fuel fragments recovered from Dounreay, see Subsection 4.4.

For uranium, assumed to be in the form $\text{UO}_2$, we adopt an $f_1$ value of 0.002. This is based on the ICRP 68 value that is considered to be most appropriate to oxide forms. This is most probably very cautious because it takes no credit for the particle matrix.

For plutonium in spent fuel we adopt an $f_1$ value of $1 \times 10^{-5}$, which is the ICRP 68 value that is considered to be most appropriate to oxide forms, and also coincides with the HPA recommended $f_1$ value based on measured values on fuel fragments from Dounreay. For americium we adopt the same $f_1$ value as for plutonium since both are present within the same matrix.

Note that the committed effective dose per unit ingestion is not proportional to the $f_1$ value. At higher $f_1$ values, the committed effective dose is dominated by doses due to uptake of the radionuclide from the GI tract and deposition in (and hence irradiation of) various body tissues. At very low $f_1$ values, the dose to these tissues becomes smaller, but the dose to the tissues of GI tract during the passage of the radionuclide through the tract is not much changed. In the case of plutonium isotopes, for example, at $f_1 = 5 \times 10^{-4}$ the contribution of effective dose to the GI tract is small, but at $f_1 = 1 \times 10^{-4}$ about one half the total committed effective dose is from effective dose to the GI tract.

5.3 Committed effective doses from ingestion of the selected particles

Table 8 summarises the particle activity, $f_1$ values and calculated committed effective dose for each of the particles identified in Table 6. Committed effective dose is summed over the radionuclides present as listed in Table 6. Contributions from shorter-lived progeny are included as appropriate.
Table 8: Summary of particle activity, $f_1$ values and calculated committed effective dose (CED)

<table>
<thead>
<tr>
<th>Particles</th>
<th>Primary radio-nuclides</th>
<th>Activity of 1mm particle at 300y, MBq</th>
<th>Reference (cautious) $f_1$ value</th>
<th>CED for reference $f_1$, mSv</th>
<th>Adopted (realistic) $f_1$ value</th>
<th>CED for adopted $f_1$, mSv</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLWR monazite/thorite (~5% Th)</td>
<td>Th-232</td>
<td>1.6E+00</td>
<td>0.0005</td>
<td>5.9E-04</td>
<td>0.0002</td>
<td>7.4E-05</td>
</tr>
<tr>
<td>High thorium monazite (30% Th)</td>
<td>Th-232</td>
<td>3.3E-06</td>
<td>0.0005</td>
<td>3.5E-03</td>
<td>0.0002</td>
<td>4.4E-04</td>
</tr>
<tr>
<td>High thorium thorite (40% Th)</td>
<td>Th-232</td>
<td>4.7E-06</td>
<td>0.0005</td>
<td>5.0E-03</td>
<td>0.0002</td>
<td>6.3E-04</td>
</tr>
<tr>
<td>Radium ZnS paint (50 µg/g)</td>
<td>Ra-226</td>
<td>3.4E-03</td>
<td>0.2</td>
<td>7.5E+00</td>
<td>0.08</td>
<td>2.0E+00</td>
</tr>
<tr>
<td>Admiralty specification (215 µg/g)</td>
<td>Ra-226</td>
<td>1.5E-02</td>
<td>0.2</td>
<td>3.2E+01</td>
<td>0.08</td>
<td>8.6E+00</td>
</tr>
<tr>
<td>Sellafield alpha rich particle</td>
<td>Pu+Am-alpha</td>
<td>7.1E-01</td>
<td>0.0005</td>
<td>1.6E+02</td>
<td>3.0-05</td>
<td>1.3E+01</td>
</tr>
<tr>
<td>Dounreay fuel particle</td>
<td>Pu+Am-alpha</td>
<td>5.7E-03</td>
<td>0.0005</td>
<td>1.3E+00</td>
<td>1.0-05</td>
<td>4.8E-02</td>
</tr>
<tr>
<td>Natural uranium as UO$_2$</td>
<td>U-alpha</td>
<td>1.3E-04</td>
<td>0.02</td>
<td>6.3E-03</td>
<td>0.002</td>
<td>1.0E-03</td>
</tr>
<tr>
<td>Enriched uranium as UO$_2$</td>
<td>U-alpha</td>
<td>4.1E-04</td>
<td>0.02</td>
<td>2.0E-02</td>
<td>0.002</td>
<td>3.4E-03</td>
</tr>
<tr>
<td>PWR spent fuel (3.4% 35 GWd/t)</td>
<td>Pu+Am-alpha</td>
<td>7.9E-01</td>
<td>0.0005</td>
<td>1.7E+02</td>
<td>1.0-05</td>
<td>6.2E+00</td>
</tr>
<tr>
<td>AGR spent fuel (2.4% 20 GWd/t)</td>
<td>Pu+Am-alpha</td>
<td>3.2E-01</td>
<td>0.0005</td>
<td>7.1E+01</td>
<td>1.0-05</td>
<td>2.6E+00</td>
</tr>
<tr>
<td>GCR spent fuel (0.71% 6 GWd/t)</td>
<td>Pu+Am-alpha</td>
<td>1.8E-01</td>
<td>0.0005</td>
<td>4.0E+01</td>
<td>1.0-05</td>
<td>1.4E+00</td>
</tr>
</tbody>
</table>
Figure 2 shows the calculated committed effective dose for reference (cautious) and adopted (realistic) $f_1$ values. The figure also indicates the 3 mSv to 20 mSv range that, based on Environment Agency ‘Advice to Assessors’ (see Subsection 2.2), we consider can be used as a significance test for the assessed effective dose from exposure to a single particle (see Subsection 2.3).

The only particles for which the calculated committed effective dose intercepts, or in most cautious $f_1$ cases exceeds the 3 to 20 mSv range are cases of radium paint particles, the highest alpha-rich Sellafield particle and spent fuel particles. Focusing on the calculated committed effective dose for realistic $f_1$ cases (red bars), only the radium paint to Admiralty specification, the highest alpha-rich Sellafield
particle and PWR spent fuel particles enter the 3 to 20 mSv range, and in no case is 20 mSv exceeded.

5.4 Effect of particle size

All the above calculations assume a 1 mm particle, which the HPA consider is the largest particle that could be inadvertently ingested. Assuming the same specific activity of source material, the activity of a spherical particle is proportional to the cube of the diameter. That is a 2 mm particle would bear eight times the activity of a 1 mm particle, and a 0.5 mm particle would bear one-eighth the activity of a 1 mm particle.

Thus particles much smaller than a millimetre are of low interest because they cannot bear sufficient activity to deliver a committed effective dose in the 3 to 20 mSv range.

Particles much larger than a millimetre are not of interest as an immediate source of internal exposure because they are too large to be ingested. Consideration should be given to larger particles, however, since a larger particle may degrade to smaller fragments within the size range of interest.

To define a practical limitation for WAC it is useful to work from the ISO definition of gravel and sand by grain size, thus:

- fine gravel 2.0 to 6.3 mm;
- coarse sand 0.63 to 2.0 mm;
- medium sand 0.2 to 0.63 mm;
- fine sand 0.063 to 0.2 mm.

We define the size range of most interest as approximating to coarse sand (0.6 to 2.0 mm) but note that larger particles are relevant as source material for smaller particles.
6 Summary and WAC for Active Particles

6.1 Summary of technical position

Our previous assessment of radioactive particles released into the coastal environment [1], which considered a range of possible particles, showed that is very unlikely that an encounter with any of the particles could give rise to tissue effects; even for prolonged contact times any effect would be minor, e.g. localised erythema or a small lesion at the site of contact. There is no possibility that any of the particles could lead to ‘severe deterministic injury’ as defined in paragraph 6.3.40 of the GRA [4].

The impact from random encounters with radioactive particles will be assessed against the GRA risk guidance level. The Environment Agency, however, have suggested via their ‘Advice to Agency Assessors’ [5] that an optimised approach should entail limiting, as far as practicable, disposal of particles that individually carry an activity sufficient to give rise to a significant assessed effective dose. We propose to do this by limiting or placing conditions on wastes that have potential to include significant numbers of particles that individually could give rise to an assessed effective dose in the range of 3 to 20 mSv or above. This dose range is derived from a ‘test of significance’ suggested in the ‘Advice to Agency Assessors’.

In this report, we have demonstrated that it is reasonable to focus on assessing ingestion of active particles, since this mode of exposure will yield the highest effective doses. We adopt a 1 mm diameter particle as the reference assumption, since the HPA identify this as the largest size particle that is liable to be inadvertently ingested.

We have assessed a range of radioactive particles that might be present, or are similar to particles that might be present, in LLW. We have calculated committed effective dose assuming both cautious and realistic $f_1$ values (fractional uptake from the GI tract to blood).

We find that, assuming realistic $f_1$ values, the only assessed particles that have the capacity to lead to calculated committed effective dose in the range 3 to 20 mSv are radium paint to Admiralty specification, the highest alpha-rich Sellafield particle and PWR spent fuel particles. In no case is 20 mSv exceeded. In all cases, the calculated committed effective dose is due to long or moderately long-lived alpha-emitting radionuclides. (It has been shown that at 300 years after disposal any contribution from other radionuclides will be small.)

Table 9, below, extracts key data from Table 6 on the assessed particles giving the highest committed effective doses from ingestion.
Table 9: Particles giving calculated committed effective dose from ingestion above 3 mSv, assuming realistic $f_1$ values (extracted from Table 6)

<table>
<thead>
<tr>
<th>Particle materials</th>
<th>Primary radionuclides</th>
<th>Specific activity of material</th>
<th>Activity of 1 mm particle</th>
<th>Activity of particle after 300 years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Admiralty specification radium paint (215 µg/gram)</td>
<td>Ra-226</td>
<td>7.9E+00</td>
<td>1.7E-02</td>
<td>1.5E-02</td>
</tr>
<tr>
<td>Sellafield alpha-rich particle</td>
<td>Pu and Am alpha</td>
<td>Not known</td>
<td>1.0E+00</td>
<td>7.1E-01</td>
</tr>
<tr>
<td>PWR spent fuel (3.4% 35 GWd/t) (10 years after reactor discharge)</td>
<td>Pu and Am alpha</td>
<td>2.3E+02</td>
<td>1.2E+00</td>
<td>7.9E-01</td>
</tr>
</tbody>
</table>

The alpha-rich Sellafield particles are somewhat anomalous as they were formed in liquid effluent treatment processes. It is unlikely that such particles could be produced in the LLWR where aqueous concentrations of radionuclides are many orders of magnitude lower. The activity on the highest activity alpha-rich particle (as in the Table above) is similar to that of the PWR spent fuel particle.

The specific activity of the Admiralty specification radium paint and PWR fuel are about 8 MBq (Ra-226) per gram and 230 MBq (Pu+Am) per gram. The radium is relatively more potent than Pu and Am isotopes due to the higher $f_1$ value, see Table 7, and presence of alpha-emitting progeny in equilibrium.

As noted in Subsection 4.7, other, higher activity materials could be considered, notably MOX fuels, irradiated highly-enriched uranium from materials test reactors and from naval reactors, plus weapons grade enriched uranium and plutonium. The data given in Table 6 give an indication of the level of specific activity and of particle activity that is of concern.

### 6.2 WAC for Active Particles

To comply with the Environment Agency ‘Advice’ [5], we will seek to limit, as far as practicable, disposal of particles that individually carry an activity sufficient to give rise to a significant assessed effective dose. The focus is on limiting or placing conditions on wastes that have potential to include significant numbers of particles such as identified in Table 9, or particles of other similarly high specific activity material.

Waste consigners will be obliged to consider the possibility that a waste may contain such particles through the WAC as follows. The WAC will contain the following criterion ²:

² This version of the WAC is modified from the draft version consulted on by LLWR in June and July 2013.
• Waste containing or that may contain Active Particles, or that may breakdown to form Active Particles, may be accepted for disposal but only on approval of a Waste Consignment Variation Form by LLW Repository Ltd.

Active Particles will be defined in the Glossary of the WAC as follows.

• Active Particle means a particle in the size range of 0.6 to 2.0 millimetres\(^3\) of high-specific activity material such that a single particle could bear of the order of a megabecquerel (1 MBq) or more of alpha-emitting radionuclides or 0.01 megabecquerel (0.01 MBq) or more of radium-226. This implies a fragment of a high-activity material, typically more than about 100 megabecquerel per gram (100 MBq/g) of most alpha-emitting radionuclides or 10 megabecquerel per gram (10 MBq/g) of radium-226. Examples of Active Particles include fragments of Admiralty specification radium-sulphide paint, fragments of irradiated nuclear fuel (especially PWR, MOX or highly enriched uranium fuels) or plutonium.

As noted in Subsection 2.3, it is not practical to prevent every single particle entering the LLWR. There is always the possibility of a single or few Active Particles in a waste consignment because it is not possible to characterise and monitor wastes to identify all Active Particles. A consigner will be required to consider the questions:

Is a waste stream known to contain or, due its source or nature, have the potential to contain a significant number (meaning of the order of hundreds or more) Active Particles?

Or possibly contain larger particles that could be the source for Active Particles?

The key concern is the potential for particles bearing alpha-emitting radionuclides but, depending on the source material, the particles may bear other radionuclides that may be more readily detected. For example, a 1 MBq (Pu+Am alpha) particle of irradiated AGR fuel will, at ten years after reactor discharge, bear about 20 MBq of Cs-137, see Table 4. Thus, depending on the waste form and monitoring arrangements, it may be possible to rule out the presence of such particles.

### 6.3 Assessment of variations

Wastes identified by consigners as containing or potentially containing Active Particles will be assessed as follows.

We will assess the potential dose impact from particles of the potential source material by the methods indicated in this report. Consideration will then be given to the practicability of characterising, separating and managing the wastes in other ways. This will be done in consultation with the consigner.

• If the committed effective doses from ingestion of the particles lie below the 3 to 20 mSv range we would generally expect to give approval for acceptance of the wastes; however, consideration would be given to the nature and concentration of the particles in the wastes and the practicability of separation of the particles and management by some other route.

• We would also generally expect to accept wastes containing or that might

\(^3\) Grain size of coarse sand, see Subsection 5.4.
contain particles with the potential to give committed effective doses consistent with the 3 to 20 mSv range where the nature and concentration of particles is such that it is not practical or practicable to characterise and manage the wastes in another way.

- Based on the Environment Agency’s ‘Advice’, we would generally expect to refuse to accept wastes containing significant concentrations of particles that could produce committed effective doses around or above the 3 to 20 mSv range. We would expect such wastes to have a low volume and hence it be practicable to manage by another route.

It is expected that by dialogue between consigners and LLWR, and assessment of consignment variation requests and enquiries, a library of cases will be built up. From this case library, it may in future be possible to give more specific guidance to consigners on identifying waste streams and consignments that have potential to contain Active Particles.
References


8. Oatway WB and Higgins NA, Review of Assessments of Prospective Exposure to Low Activity Sources or Particulate Material following Coastal Erosion of, and Intrusion into, the LLWR, Health Protection Agency, Centre for Radiation, Chemical and Environmental Hazards, CRCE-EA-3-2013.


10. LLWR, Assessment of Discrete Items and Basis for WAC, LLWR/ESC/R(13)10055, August 2013.


12. LLWR, ESC2011: The Disposed and Forward Inventory of LLWR, Serco/E003756/12 Issue 2, April 2011.


15. SEPA, Dalgety Bay Radioactive Contaminated Land Risk Assessment, 29/05/2013

16. Serco, A Forward Inventory for LLWR Based on the 2010 UKRWI, SERCO/E.005766/001 Issue 1, February 2011.

18 Dounreay Site website: http://www.dounreay.com/particle-cleanup
19 Wilkins B, Harrison J et al, Health Implications of Fragments of Irradiated Fuel at the Beach at Sandside Bay: Module 6: Overall Results. HPA RPD-EA-03-2006.
21 http://www.wise-uranium.org/rup.html
22 Electrowatt-Ekono (UK) Ltd, Radionuclide Content for a Range of Irradiated Fuels, Contractor’s Report to Nirex, 17503/74/1 Rev. 2, July 2003.
25 ICRP, Age-dependent Doses to Members of the Public from Intake of Radionuclides (Part 5), Compilation of Ingestion and Inhalation Coefficients, ICRP Publication 72, Ann. ICRP 26(1), 1996.