



The international fertiliser society

Aspects of remediation of land on sites used for fertiliser manufacture and storage

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1. SUMMARY.

Significant changes have occurred in European fertiliser manufacturing in recent years as results of changes in market conditions, feedstocks used and necessary rationalisation. As a result many of the old phosphate-based fertiliser plants are redundant and are being dismantled. The sites of these plants are now being redeveloped. Some of these sites have areas of low-level radioactive contamination as a result of the enhanced levels of naturally occurring radionuclides present in many phosphate ores. Much of this contamination is associated with radionuclides in the U decay series and particularly ²²⁶Ra and its daughters. The sites are also often contaminated with heavy metals from the ores and various acids used in the manufacturing processes. However, it is the radioactive components of the contamination, which tend to dominate the remediation and waste management strategies.

The rehabilitation of radioactively contaminated land is a small but growing field of activity worldwide. To date most of the sites in or associated with the UK have been small in scale and have generally involved natural radionuclides. This paper addresses certain of the key considerations in remediating fertiliser manufacturing and storage sites contaminated with naturally occurring radionuclides. It considers all of the key aspects in any remediation programme, including the regulatory aspects, characterisation of contaminated areas, assessment of the risks posed by any contamination and potential dean-up criteria. It then considers remediation approaches with the technologies used in their application and also waste management. Finally, it illustrates the key aspects of one remediation approach through an example from a UK site. This relates to a former waste storage and processing area.

2. INTRODUCTION.

As fertiliser manufacturing changes with much of the phosphate concentration and phosphoric acid production now undertaken in the countries of the ores' origin, many old fertiliser plants in the United Kingdom and Europe are redundant and being dismantled. The sites of these plants are being redeveloped either for new plant or completely new applications, including light industry, retailing and even housing.

Some of these sites are contaminated in areas with enhanced levels of naturally occurring radionuclides, which derive originally from the phosphate ores used as feedstocks. Key amongst these contaminants are radionuclides of the ^{238}U series and particularly ^{226}Ra . The contamination had many causes ranging from past waste management and process practices, leaks, accidents, coupled with a lack of appreciation of the radiological significance of the naturally occurring radionuclides in minerals, process plant and waste streams. The rehabilitation of such radioactively contaminated land is a small but growing field in the UK and worldwide. This paper addresses the key aspects of the management and remediation of areas of radioactive contamination within fertiliser manufacturing plant and storage areas.

The largest areas affected by radioactive contamination in the fertiliser manufacturing cycle are those associated with phosphate mining and beneficiation operations. These are significant for all of the major phosphate producers, such as Morocco, the USA and Russia. They are also significant for several European countries, where in the past ore beneficiation and phosphoric acid production were undertaken. For these sites the radioactive contamination is characterised by large volumes and areas but generally low- levels of contamination.

The problems of land contamination by naturally occurring radionuclides and particularly ^{226}Ra are not unique to the fertiliser industry. They have caused low-level contamination at many sites, including several in inner city areas. Radium was widely used for its luminising properties around the world, especially during World War II, when it was extensively used by the military for luminous instrument dials in aircraft, ships, vehicles, etc. Thorium was widely used in the era of gas lighting for incandescent gas mantles and is still used in electrical filaments and as a hardener for magnesium alloys. Rare earths occur naturally in certain phosphate-bearing ores and some sites used to produce and refine them have become contaminated with uranium, radium and thorium. A number of these sites have now been remediated and hence provide relevant experience for fertiliser plants.

2.1. Nature of the radioactive contaminants at fertiliser plants.

Many radionuclides occur naturally in the environment by primordial nature since the formation of the Earth or as a result of cosmic processes. Key amongst the former are those of the uranium, thorium and actinium series. These are three decay chains of radionuclides headed by ^{238}U , ^{232}Th and ^{235}U , respectively. Successive members formed in each chain primarily by α or β decay until the stable isotopes ^{206}Pb , ^{208}Pb and ^{207}Pb , respectively, are ultimately formed. These radionuclides are distributed in low concentrations in many ores, which form feedstocks for the chemical, metallurgical and power industries. Amongst the ores, where such natural radionuclides are significant, are many of the phosphate ores, which have and are used in the manufacture of various phosphate-based fertilisers. The average concentrations of uranium and thorium as elements in the Earth's crust are 4.2 and 12.5 mg/kg with associated specific radioactivity levels of each of the parent radionuclides: ^{238}U and ^{232}Th of $\sim 50\text{Bq/kg}$. Many phosphate ores, including those from Morocco, USA (Florida, Idaho, Wyoming), Israel and Jordan contain much higher levels for the uranium series elements, e.g. ^{238}U and ^{226}Ra each in the range

1300-2300 Bq/kg (Baetsle, 1991; Schmidt, 1995; Hofmann, 2000). Their thorium series contents, by comparison, are much less enhanced with ^{232}Th specific activities in the range 10-110 Bq/kg (Baetsle, 1991).

The basic fertiliser manufacturing processes do not significantly concentrate the radioactivity over the levels, which are present in the ores. Where concentration occurs, concentrations are raised typically by up to a factor of 2. Much greater concentrations can occur on the internal surfaces of plant and around areas of leakage from pipework, valves, traps, etc, as results of scale formation and evaporation. These processes have been observed to increase concentrations by factors of 10-100.

The main sources of residual radioactivity related to phosphate fertiliser plants are associated with the phosphogypsum storage/disposal areas from sulphuric acid processing and slag disposal areas from thermal processing. About 14% of the ^{238}U , 80% of the ^{226}Ra and 30% of the ^{232}Th in the original ore remains in the phosphogypsum. Average radionuclide contents in phosphogypsum are -500 Bq/kg ^{238}U , 3000 Bq/kg ^{226}Ra , 1300 ^{210}Pb , 900 Bq/kg, ^{210}Po and 10 Bq/kg ^{232}Th (Baetsle, 1991).

Before any redevelopment of redundant fertiliser plants can take place, it is necessary to determine the levels of any residual contamination, i.e. radioactive, chemical or otherwise hazardous. Assessments can next be made of the risks posed by that contamination with reference to the proposed future uses. Any necessary remedial action can then be undertaken to reduce the risks to acceptable levels.

3. REGULATORY FRAMEWORK

The United Kingdom operates separate regulatory regimes for radioactive and non-radioactive substances and wastes, although in future a unified approach is promised. Its primary act of legislation defining which materials and wastes are radioactive under law is the Radioactive Substances Act of 1993 (RSA 93). This law states the necessary requirements for the registration, storage and disposal of radioactive materials and waste. The act is supported by a series of exemption orders, which exclude some items, substances and materials from some or all of its provisions. These exemption orders were introduced to aid practical implementation of the act by removing a large number of judged low risk substances, etc, from control under the act. RSA 93 specifies that a material or waste is radioactive, if it contains:

a) A substance containing an element specified in Schedule 1 of RSA 93, which is present at specific activity levels greater than those given in the adjoining columns of that schedule. All of the elements specified in Schedule 1 (U, Th, Ac, Pa, Ra, Rn, Po and Pb) are members of the natural uranium, thorium and actinium-series chains. For fertiliser site remediation the key elements in the Schedule 1 list with their limits are uranium, thorium and radium with respective specific activities of 11.1, 2.59 and 0.37 Bq/g. If these elements are present in any substance at lower activity levels than those given in Schedule 1, then that substance is not classed as radioactive, i.e. it is de minimus. There is currently no clear regulatory guidance as to how the Schedule 1

limits should be interpreted. They can be interpreted as applying to the heads of the natural decay chains and excluding short-lived daughter products. This interpretation has been proposed by Hill and Wakerley in a recent DETR/DEFRA report (Hill, 2000). Thus the Schedule 1 limit of 11.1 Bq/g would only apply for separated uranium, which will not be present in fertiliser plants. For natural uranium with its daughters below U in some degree of secular equilibrium, which is present in such plants, the limiting activity of any radionuclide in its chain would be controlled by that for radium, i.e. ^{226}Ra , at 0.37 Bq/g.

b) A substance, whose radioactivity is wholly or partly due to a nuclear fission, neutron or ionising irradiation process, not occurring in nature.

Thus any substance containing man-made radionuclides will be classified as radioactive and subject to the provisions of RSA 93.

RSA93 specifies that a waste is a material, which if not a waste, would be a radioactive material or has become contaminated with such material during the production, keeping or disposal of radioactive material.

3.1. Applicable exemption orders

Exemption orders specify the conditions under which materials or wastes defined as radioactive under RSA 93 can be made "Exempt", i.e. excluded, from some or all of the provisions of the Radioactive Substances Act. There are two exemption orders, which are particularly relevant to fertiliser sites' remedial works. These are:

- a) Statutory Instrument 1986, No. 1002 and amended 1992, No. 647. The Radioactive Substances (Substances of Low Activity) Exemption Order.
- b) Statutory Instrument 1962, No. 2648. The Radioactive Substances (Phosphatic Substances, Rare Earths, etc.) Exemption Order.

3.2. The Substances of Low Activity (SoLA) Exemption Order.

The SoLA Exemption Order specifies that solid radioactive waste is excluded, i.e. it is "Exempt", from the provisions of Section 6(1) and (3) of RSA, provided that it is substantially insoluble in water and has an activity, which does not exceed 0.4Bq/g. These provisions include the requirement to have an authorisation from the Environment Agency to dispose of the waste.

The 0.4 Bq/g may be interpreted as applying to the sum of any anthropogenic radionuclides present above their background level plus any Schedule 1 elements above their Schedule 1 limits. In the de-licensing of some nuclear facilities the "background" level of each radionuclide is currently being taken as its 95-percentile background level. The same approach may be applied to radionuclides on fertiliser sites.

3.3. The significance of "natural" background

The Radioactive Substances Act 1993 and its Exemption Orders do not specify whether the activity levels are to be judged additional to natural background levels. Indeed, there is no reference in any of these documents to background levels of

radioactivity. It has generally been practice with implicit acceptance by the Environment Agency, that this is the case in respect of application of the SoLA EO. This has not been the case with the Phosphatic Substances, etc, where natural background levels have been included in the totals. This apparent ambiguity effectively disappears with application of Schedule 1. This is because the natural background in most places is almost entirely derived from Schedule 1 elements, which are below their de minimus levels. Hence they are not to be classed as radioactive under the terms of RSA93 by definition. In addition, the anthropogenic contribution is usually so small at -0.03 Bq/g as to be within measurement errors and effectively negligible. Thus it may be seen that with the appropriate interpretation and application of Schedule 1 for uranium, thorium and their daughters, the need to account for background does not arise.

3.4. The Phosphatic Substances, etc, Exemption Order.

The Phosphatic Substances, etc, Exemption Order states that material, which is radioactive solely because of the presence of one or more of the Schedule 1 elements, is unconditionally exempted from the provisions of RSA, provided that the specific activity of *each* of the Schedule 1 elements present does not exceed 14.8 Bq/g. This exemption includes waste disposal. It only applies to materials, which do not contain man-made radionuclides, as is normally the case in the fertiliser industry.

The Phosphatic Substances Exemption Order relates to elements. Thus for uranium and radium the limit would normally apply to the sums of the specific activities of (^{238}U , ^{235}U and ^{234}U) and (^{228}Ra , ^{226}Ra , ^{224}Ra and ^{223}Ra), respectively.

Different interpretations of the Phosphatic Substances Exemption Order do exist (Williams, 1999; McHugh, 1999). These have come about in order to accommodate the greater level of risk now attributable to unit doses of radiation compared to the values, when the EO was first prepared. In particular, the "element" limits have been applied to each member of the uranium and thorium series chains. Such an interpretation leads to anomalies with the Schedule 1 limits, as is acknowledged by the Environment Agency. Indeed, the Schedule 1 interpretation discounts the short-lived daughters, whilst the Phosphatic Substances EO uses them.

The guidance currently recommended by the EA to its inspectors (McHugh, 1999) is that the specific activity of any radionuclide in the ^{238}U chain is restricted by the limit for polonium, which has three isotopes present, i.e. (^{218}Po , ^{214}Po and ^{210}Po). Thus with secular equilibrium, the specific activity of each polonium isotope and each radioisotope of any other element present in the chain would be restricted to 14.8/3 Bq/g. If secular equilibrium does not prevail or only partially so, higher limits could prevail, provided $Z (^{218}\text{Po} + ^{214}\text{Po} + ^{210}\text{Po}) \leq 14.8 \text{ Bq/g}$ For the ^{232}Th chain there are three pairs of radionuclides: ($^{232}\text{Th} + ^{228}\text{Th}$), ($^{228}\text{Ra} + ^{224}\text{Ra}$) and ($^{216}\text{Po} + ^{212}\text{Po}$) limiting the radioelements. Likewise, the radioelement limit for the actinium series headed by ^{235}U is restricted by the thorium pair ($^{231}\text{Th} + ^{227}\text{Th}$) Thus for each of these latter two chains the limit for any radioisotope present would be 14.8/2, i.e. 7.4 Bq/g. Again, if any dis-equilibrium prevails, the limits could be higher, provided that no Schedule 1 element exceeds a total of 14.8 Bq/g. These limits are very important,

as higher activity material requires registration under RSA 93 and disposal as low-level waste with major cost implications.

4. CHARACTERISATION.

The characterisation of contaminated sites is a very important component of any remediation programme and best practice guidance is available (BSI, 2001; CIRIA, 2000). It can also be a significant part of the overall cost, typically being of the order of 10%. Characterisation can have several functions. It is important to recognise these from the outset and to plan and optimise the characterisation programme to satisfy their various requirements. The purposes of characterisation include:

- a) Determining the current level of hazard posed by the site as a precursor to regulatory/decision making on any future action, including emergency responses;
- b) Assessment of the long-term nature of the hazard posed through risk assessments/pathway analyses;
- c) Input to planning the remediation approach and clearance criteria; and
- d) Input to categorisation of resulting wastes with a view to minimising volumes and costs and utilising available disposal routes.

The first stage of any characterisation programme should include as extensive a review as possible of the past history of the site in terms of activities carried out there:

- a) Likely radionuclides present, concentrations and distribution.
- b) Other contaminative processes and industries present.
- c) Local and regional backgrounds of potential contaminants.
- d) Geology and hydrogeology of the site and region.
- e) Soil types present.
- f) Vegetation.

The scale of the contaminated area is also a major consideration in determining the characterisation approach. Sites can vary very considerably in size ranging from a few tens of square metres for an area of scale deposit or a leak through several thousand square metres as at phosphogypsum and waste storage areas. With small scale sites foot-based radiation surveys can be used very cost effectively. This approach has been used for the majority of sites in the United Kingdom.

As the scale increases, so the practicability of this approach decreases. Provided a significant gamma emitting radionuclide is present amongst the contaminants, vehicle- or even aerial-based surveys can become cost effective. Vehicle-based surveys with GPS position location are now capable of considerable precision, but are still limited to detection levels - 10 kBq/m^2 , dependent on the natural background. Where a significant gamma source is not present, then discrete surface and core sampling have to be undertaken to obtain an estimate of the contamination distribution. The same approach is necessary for potential chemical contaminants, including heavy metals, toxic anions and organics. The limitations of this approach become significant when the contamination is a result of discrete particles or spots rather than being homogeneously distributed. For this case the use of a statistical sampling approach is very important (BSI, 2001; Ferguson, 1993; USEPA, 1997). However, it may still require very large numbers of samples, dependent on contaminant spot frequency and size and the degree of certainty required in

detecting all significant areas of contamination.

5. WASTE MANAGEMENT.

Remediation of the contaminated sites can result in both radioactive and chemically contaminated wastes. The following categories of waste, which bear radionuclides in concentrations above their background levels, can arise during the remedial works:

- a) De minimus waste;
- b) Exempt waste; and
- c) Low-level waste.

In addition, the works can also give rise to:

- Special waste. This is waste, which contains elevated levels of hazardous chemical contaminants, as defined in the Special Waste Regulations of 1996,
- Controlled wastes. These include any material, which is surplus to requirements with levels of contamination from zero up to the trigger levels for Special waste.
-

The applicable definitions for the categories of waste, where the radionuclide content needs to be considered, are given in the Radioactive Substances Act of 1993 (RSA 93) and its associated exemption orders.

5.1. Activity limits for the different categories of waste.

The limits to be applied for the various categories of wastes, containing radionuclides in concentrations above those occurring in the natural background around UK fertiliser plants, are summarised in the Table 1.

Table1 *Specific activity limits to be applied in determining categories of wastes from consideration of their radionuclide content.*

| Waste Characteristics | Specific Activity, Bq/g | Waste Category |
|--|---|----------------|
| 1. Wastes containing man-made radionuclides | ≤ 0.4 (<i>excluding</i> contributions from all Schedule 1 elements and their daughters, <i>provided</i> none exceed their Schedule 1 Column 2 limits) | Exempt |
| | > 0.4 (<i>excluding</i> contributions from all Schedule 1 elements, which do not exceed their Schedule 1 Column 2 limits, and their daughters) | LLW |
| 2. Wastes containing mixtures of Schedule 1 elements | \leq (Schedule 1, Column 2 limit for each element) | De minimus |
| | $>$ (Schedule 1, Column 2 limits) and ≤ 14.8 for each of Schedule 1 elements | Exempt |
| | > 14.8 for any of the Schedule 1 elements | LLW |

6. REMEDIATION APPROACHES.

As with inorganic and unlike organic contamination, radioactive contamination cannot be destroyed. Hence there are only two basic options for its "remediation". These are containment of the radioactivity on the affected site or its removal to a more suitable site for storage or disposal. The containment options include partial or complete encasement by the use of surface caps, sub-surface barriers and even purpose-built vaults. They also include immobilisation, where the radioactive contamination is bound by encapsulation as with cement grouts or intimately fixed in a chemical or glass matrix. The removal option also requires the containment stage at the new disposal site. However, there the conditions and facilities should be purpose developed to contain the radionuclides for the long-term by use of natural and man-made barriers.

The removal approaches can involve bulk separation of all of the contaminated soil, etc, including much uncontaminated material. This usually involves conventional earth-moving technologies. The alternative approach is selective removal of the contamination. This can be performed either in-situ or ex-situ. The in-situ methods, which are generally less well developed and hence have much less experience of application, include electro-remediation, phyto-remediation and in-situ soil washing. The ex-situ technologies include physical separation methods, often of the types used in ore beneficiation, such as flotation and gravity settling. They also include others, which use radiation monitors to specifically detect elevated levels of radioactivity and separate such materials by gates, etc, in conveyor systems. Other techniques use chemical extraction and soil washing. Appendix 2 provides a review of available remediation approaches. It includes all of the main existing technologies and some, which are being developed, but have yet to achieve wide application.

Dependent primarily on the areal scale of contamination, sites may be cleaned up to background levels or to a predetermined level of risk. By far the most common approach employed in the former case is contaminant removal by excavation often with some form of segregation. Dependent on the availability of disposal routes and the costs of storage and disposal for the different waste categories, the latter can be used to minimise the volumes of higher activity waste or to concentrate the activity into a small volume with the bulk of the soil being released as essentially free of contamination. With very large contaminated areas full clean-up is invariably impractical and it is only cost effective to reduce the level of risk to the exposed population. The key consideration then becomes the level of cost, which it is worth spending to achieve a given risk and hence radiation dose reduction. The latter will vary from site to site, dependent on the economic priorities. In this case clean-up may only be a small part of the total risk reduction approach.

To determine the best risk reduction strategy, it is very important to determine the various exposure pathways and their relative importance. This is best achieved using a focused, staged quantitative risk assessment. The latter would examine the potential contributions made both on- and off-site by the direct irradiation, inhalation and ingestion pathways to members of the critical group, workers and the general public. Such studies invariably require additional data as they proceed. Hence, there

is much merit in using a staged approach with an initial scoping study being used to identify the key exposure pathways associated with any site. The results of this study can then be used to focus additional characterisation work on those areas, which will yield the key data required for a more comprehensive risk assessment. Once the baseline level of risk posed by the current state of contamination of any site is established, the risk assessment methodology can be applied to determine both clean-up levels required to achieve a given level of residual risk and the level of risk posed by the site after different rehabilitation strategies have been implemented.

It is important to be realistic in what can be achieved in risk reduction and to plan the rehabilitation to reduce the risks in the order of their magnitude.

7. RISK ASSESSMENT AND CLEAN-UP CRITERIA.

In the past the clean-up criteria were often based on dose rate, e.g. 0.3 uSv/h compared to a typical background level of 0.15-0.2 uSv/h, definitions of radioactive materials and limits for the exemption of materials from radioactive controls and some qualitative consideration of the risks arising from the remaining wastes (Stearn, 1993). However, most current criteria are generally directly or indirectly health risk-based and normally derive from the recommendations of the ICRP. The choice of limit or indeed the applicability of a limit are influenced by whether current exposures are controlled and hence the risk reduction works are classified as in support of a "practice" or an "intervention", as defined under ICRP 60 (ICRP, 1991). For fertiliser plant sites practice criteria are likely to be more appropriate with their maximum dose limits and dose constraints. For remediation around some phosphate mining sites intervention criteria could be appropriate.

Appendix 1 provides a review of the various clean-up criteria used in major European and North American countries for the clean-up of radioactive contamination derived from naturally occurring radionuclides. It also includes the developing guidance provided by the International Atomic Energy Agency and the International Commission on Radiological Protection.

In some cases the criteria are interpreted in terms of an acceptable annual dose limit. For other sites, the clean-up levels for various radionuclides have been set as specific activity levels of selected radionuclides in soil, at fractions of the individual Generalised Derived Limits (GDLs) (NRPB, 1987) or by comparison with the surrounding backgrounds at similar industrial sites. In each of these cases likely current and post remediation doses can be related to contamination levels through a quantitative risk assessment using an analysis of potential exposure pathways. Indeed, GDLs and current proposed exemption limits are now derived on the same basis, differing only in the degree of conservatism built into the analysis and the underlying assumptions.

7.1. The remediation levels.

The remediation levels to which soils, etc, need to be cleaned up can be developed by reference to the findings of the risk assessment and through consultation with the Environment Agency. The risk assessments normally consider the optimum

acceptable level of risk of serious harm or death to the most exposed individual to be 1×10^{-6} per annum. This is the level of risk, which the UK Health and Safety Executive (HSE) considers is acceptable to members of the public. It is also the acceptable level of risk proposed by the International Commission on Radiological Protection (ICRP) and the International Atomic Energy Agency (IAEA, 1988) and which the UK Government with guidance from the National Radiological Protection Board (NRPB) is expected to confirm for the reclamation of contaminated land (DETR, 1998; NRPB, 1992; Barraclough, (1996). For radioactivity this risk level was judged to be equivalent to an additional annual dose of 30 uSv. In accordance with the ALARP Principle, greater levels of risk reduction should always be achieved wherever practicable, but do not warrant the expenditure of further significant resources to achieve them.

In a recent remediation programme at a site extensively contaminated with ²²⁶Ra (Graham, 2001), the Environment Agency approved the following remediation criteria. The minimum risk-based remediation level for the proposed activities and receptor groups on the site was set at 0.15 uSv/h (inclusive of background radiation, which at this site is approximately 0.1 uSv/h). This value is comparable with the proposed remediation value for radioactively contaminated land to be redeveloped for domestic housing being considered by the Environment Agency (permitted excess dose of 0.05 uSv/h). In addition:

Max. permissible concentration of ²²⁶Ra in general areas

0.37 Bq/g Ambient gamma dose level

0.15 uSv/h Within the gardens of houses on the site boundary:

Maximum permissible concentration of ²²⁶Ra

0.10 Bq/g to 0.5 m depth

0.37 Bq/g below 0.5 m Ambient gamma dose level

0.10 uSv/h

The level of 0.37 Bq/g for ²²⁶Ra would correspond to a risk level -3×10^{-5} per annum, if the contamination was continuous and affected area was occupied at all times by the most exposed individual. However, with localised areas of contamination and more restricted occupancy, e.g. garden use, the risk level would reduce to dose to 10^{-6} per annum.

8. REMEDIATION IN PRACTICE.

AEA Technology Nuclear Engineering/RWE Nukem has recently remediated areas within a UK fertiliser plant, which previously produced phosphate - based fertilisers. The phosphate ores used as feedstocks contained elevated levels of naturally occurring radionuclides from the uranium series elements. In the production process radium dissolved preferentially and was then deposited in association with calcium in scale on some process pipework. During maintenance this scale was removed. Some of the scale became deposited on grassed areas and hard standing, resulting in significant levels of radioactive contamination. The soil in this area contained elevated levels of radium and its daughters. The maximum level measured small samples was 1000 Bq(²²⁶Ra) /g.

8.1. Gamma radiation areal survey.

A characterisation was undertaken of the potential area of contaminated ground. This involved first monitoring of the whole area with AEA Technology's Groundhog system (Davies, 1998). This utilises a NaI-based, gamma scintillation detection system, linked to a GPS location system. The output was a sequence of gamma intensity measurements with associated three-dimensional positional co-ordinates. The results were processed and used to produce a map of the radiation levels over the surface. This contamination map was overlain on a map of the site layout. The survey covered the grassed area, the adjacent hard standing and roads. The detection system is sufficiently sensitive to distinguish between the gamma signatures of uncontaminated materials, e.g. concrete, tarmac, granite chippings due to their different contents of natural radionuclides. Hence different areas of the site, such as roads and covered areas were readily detectable on the radiation survey maps.

The survey showed that the bulk of the grassed area had a basic background level of radiation in the range 0.03-0.09 uSv/h. Background levels of up to 0.2 uSv/h are not untypical in low radon areas in England. There was one region of very high gamma intensity with count rates of >25,000 cps (max. 29,400), which may be compared to the lowest background levels on the site in the range of 0-150 cps. This gamma intensity saturated the NaI detector. Hence the actual gamma intensity was likely considerably higher. The Groundhog probe was then calibrated for ^{226}Ra on the basis of various postulated distributions of the contamination. Peak gamma dose rates were > 8.2 uSv/h, compared to the lowest background at 0.03-0.05 uSv/h. Such levels required the contaminated area to be fenced off to prevent general access. The area was subjected to controlled area conditions under IRR 99.

Levels of gamma radiation above 0.3 uSv/h could be detected up to 21 m along one side of the grassed area and 28 m in the direction at right angles away from the highest spot. The results show that contamination levels decreased away more rapidly than to the east. The survey also revealed a few spots of higher activity along the far side of the road, which runs south from the south-eastern corner grassed area. The highest gamma intensities recorded along this roadside were approximately 60 m south of the grassed area. They were in the range 5000-7500 cps, i.e. 1.4-2.1 uSv/h, with the maximum at 5400 cps, i.e. 1.5 uSv/h. Further monitoring and inspection of the roadway revealed that these hot spots appeared to be fixed contamination on the actual road surface.

8.2. Intrusive characterisation survey.

The next phase of the characterisation involved core sampling of the area using a small window sampler on a rectangular grid, followed by gamma spectroscopy of each of the removed cores. This work was undertaken in accordance with a method statement and risk assessment, which was approved by the Environment Agency's inspector for the site. Larger volumes of soil was also removed from the most active area. This soil was placed into two IP-2, black 200 L drums. These drums were assayed on site using a calibrated, portable gamma spectroscopy system, as were the individual core samples.

The results showed that the activity is due to the ^{226}Ra natural decay chain. There was no evidence for ^{228}Ra or its daughters nor for higher members of the uranium series. The daughters of ^{226}Ra were present at 70-85% of their secular equilibrium values. This degree of secular equilibrium is consistent with that frequently found in soil samples. The most active material in the two drums had mean ^{226}Ra specific activities of 190-200 Bq/g. These values are of the order of a factor of 40 greater than the 4.9 Bq/g for radium, which is the current interpretation by the Environment Agency of the limit for radium under the Phosphatic Substances Exemption Order (McHugh, 1999). On this basis it was concluded that the soil would be classified as low-level waste and required disposal at the BNFL Drigg site.

The intrusive characterisation led to estimates of the LLW and exempt waste arising from clearance of the area in the ranges. Following this characterisation work the results were presented to the Environment Agency and an application was made to them for an Authorisation to Accumulate and Dispose of Radioactive Waste at the site. This was granted. At the same time the necessary waste characterisation, quality programme and quality plans were prepared and submitted to BNFL for approval in order to despatch the low-level waste arising from the characterisation and later remedial works. After final approval the remedial works began.

8.3. Remediation works

The remedial work was undertaken using a 6 Te tracked excavator. The latter was fitted with a narrow bucket, which enabled the waste to be loaded directly into 200 L drums, as necessary.

8.3.1. Hot spot removal

The work was undertaken in accordance with the approved method statement and under the conditions of a Permit to Work issued by the plant manager. The work first involved the further removal of the contamination in the vicinity of the main hot spot. The soil was removed and carefully placed into IP-2 drums using the excavator. After each bucketful was removed, the excavation face was re-monitored with a Groundhog probe (3" NaI scintillation detector) and a Harwell Instruments 1667, "pancake-type", by scintillation probe.

The first objective was to remove contamination down to a level of $<0.4 \text{ Bq} (^{226}\text{Ra}) / \text{g}$ and a radiation level of $\leq 0.15 \text{ uSv/h}$. These corresponded to the clean-up levels agreed with the Environment Agency inspector and used earlier for the remediation of Ditton Park, which was similarly contaminated with ^{226}Ra (Graham, 2001). Those levels were deemed adequate for unrestricted public access to that site for recreational purposes. The residual activity limit also corresponds to the elemental radium level (0.37 Bq/g) under Schedule 1 of the Radioactive Substances Act 1993 at which the soil would not be legally judged to be radioactive under British Law.

In calibration trials AEA determined that a 0.2 m thick layer of soil contaminated with ^{226}Ra and its daughters (77% secular equilibrium) at a level of $0.4 \text{ Bq} (^{226}\text{Ra}) / \text{g}$ produced a response on the Groundhog detector of -1100 cps. This was used as the

target level for the remedial work. It was noted that if the same layer were covered by 0.2 and 0.4 m of dean soil, the detector responses would decrease to 140 and 80 cps, respectively.

After the removal of the bulk of the contamination at the main hot spot, the work focused on a second hot spot area to the northern edge of the grassed area. The ground in this area was found to be quite springy, when monitored. This was unlike the other soil on the grassed area and was not consistent with compacted soil. Upon excavation it was found to contain some waste debris, including gloves, clothe from clothing and plastic sheeting. The same approach of monitoring, excavation and remonitoring was used to remove this area of contamination. With the primary and secondary hot spots removed, the work concentrated on the areas of lower contamination. This involved some careful hand excavation around the electrical cables to the street lighting, etc. In total 31 drums were filled with waste in addition to the three drums from the original characterisation.

8.3.2. General contamination removal

The contamination levels were reduced very considerably from the original levels. However, it was apparent that significant contamination still remained. Given that the levels remaining were consistent with exempt waste, i.e. $\leq 4.9 \text{ Bq } (^{226}\text{Ra}) / \text{g}$, rather than low-level waste and the volumes appeared to be substantial, it was decided that this waste would be removed more appropriately in covered skips. The waste was loaded directly into the skips after being monitored. Upon filling the contents of each skip were monitored to determine the gamma dose rates. Their sides, etc, were also monitored to ensure that there was no radioactive contamination. They were then covered to prevent any possible loss of contaminated soil.

The data from the monitoring of the contaminated area was then reassessed to determine the need and extent of any further remedial work. The residual gamma radiation and contamination levels in the area were measured following conclusion of the remedial work. The results showed that the maximum gamma intensity and ^{226}Ra specific activity were in accordance with the remediation target and in most cases very substantially lower.

8.3.3. Waste assaying and monitoring

The next task was to undertake confirmatory analyses of the wastes in all of the waste drums and the skips. Each of the waste drums was assayed using a Scintrex GRS-2000 portable gamma ray spectrometer in a predefined assay geometry. The drums were rotated successively through 90° and the measurements repeated. The average of these multiple measurements on each drum was then used in determining its radionuclide inventory. Two of the drums had been assayed previously during the characterisation study, using an EG&G Ortec high resolution gamma spectrometer. Samples of the waste in these drums were also separately assayed by Harwell Scientifics using the same high resolution, gamma spectrometer. These earlier analyses, which were undertaken using a NAMAS accredited method, were repeated and used a check standard for the assay of the remaining drums, etc.

For the skips the waste was assayed at two equispaced points along the centreline on the top and side surfaces, treating the viewed volume as effectively an infinite source. Given the skip dimensions and the volume of waste in each skip, this was a valid approximation. In order to determine the specific activity of the waste in each drum or skip, it was also necessary to determine the mass of waste in each drum, etc. For the skips, this was done on the basis of filled volume and a mean density determined from the drum measurements. Each drum was weighed using a calibrated set of mechanical scales. The average specific activity estimates for the wastes in each container were then used to segregate those wastes into the LLW and Exempt categories. Measurements were also made of the maximum dose rates on the surface of each drum. The top and cylindrical surfaces of each drum and the sides and tops of each skip were monitored with an Eberline RO2 by monitor. The highest dose rates were recorded.

8.3.4. Waste disposal

The results of all of the analyses and waste volumes were sent to the Environment Agency prior to any disposal. Details of the low-level waste volumes and activity levels were also forwarded to BNFL Low Level Waste Management Services at Sellafield. With the agreement of the EA the skips and drums of exempt waste were sent to a landfill in the region, which was authorised to accept and dispose of such wastes. Upon final authorisation from BNFL fifteen IP -2 drums, which contained low-level waste, were despatched to BNFL Drigg for final disposal.

S.3.5. Clean-up verification survey.

After removal of all of the contamination a further Groundhog gamma radiation survey was carried out to verify the extent of the remediation. This survey was considerably more extensive than the original survey. In addition, to the immediate area of contamination, it included the entirety of the hard-standing area and the site of the former phosphoric acid plant. It also encompassed all of the grassed area and the roadway between that area and the former acid plant compound. Two very limited areas could not be surveyed. These were associated with small outbuildings and storage containers.

The radiation levels on the hard-standing corresponded to the lowest background levels measured on the site, i.e. 0.03-0.055 uSv/h. On the area occupied by the former phosphoric acid plant there were elevated gamma dose rates in the eastern third, which might have been indicative of some contamination below the surface covering, as no loose contamination could be detected with contamination probes. Within this part there are three regions of higher activity. The maximum dose rates measured in these regions were 0.9, 0.5 and 0.6 uSv/h, respectively. It is not possible to determine from the initial gamma measurements whether these areas had low-level surface contamination or higher levels of contamination beneath the granite chippings. The sources of the elevated dose rates were checked by using an excavator to remove the surface covering and remonitoring. It was confirmed that only low-level contamination existed. The associated risks were assessed to be very low and the area was judged to be acceptable for its intended use after covering with

a fixed surface for car parking.

8.4. Conclusions

The contamination within the grassed area was removed. The remediation targets in terms of both residual specific activity and dose rates were met throughout the contaminated area. The bulk of contaminated soil was disposed of as Exempt waste to an approved landfill. The remaining low-level radioactive waste was transported to Sellafield for packaging by BNFL for disposal at its Drigg site.

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APPENDIX 1. INTERNATIONAL GUIDANCE ON SUITABLE CLEAN-UP LEVELS FOR SITES CONTAMINATED WITH ELEVATED LEVELS OF NORM

A1.1. Introduction.

Advice currently exists on the disposal of radioactive wastes (NRPB, 1992) and on recovery countermeasures after land has become contaminated after a serious accident involving the release of radioactivity (NRPB, 1990; ICRP, 1992). However, there is an absence of internationally agreed advice concerning the rehabilitation and management of land, which has been contaminated with radioactive materials from past practices. Such guidance is being developed by international organisations, such as the International Commission on Radiological Protection (ICRP) and International Atomic Energy Agency (IAEA). It will cover a wide range of remediation situations. However, due to the need for extensive consultation and consensus, definitive advice from these sources is unlikely to be available in the near future. In the ICRP case it is known that their views are still very much in the formative stage and it will be at least two years before such advice will be available. The IAEA is more advanced and has issued an interim report for comment. However, it is also likely to be a minimum of two further years before that guidance is definitive.

A12. Application of the ICRP framework for radiological protection.

The framework for radiological protection recommended by the ICRP (ICRP, 1991) and endorsed by national radiological protection authorities does exist and is equally applicable to contaminated land exposures as to any other radiological exposure situation. It recognises two types of situation involving radiological exposures. The first situation is a practice, where radiation exposures are being **increased** beyond the currently prevailing level for a particular group of people, be they workers or members of the general public. The second is an intervention, where action is being taken to *reduce* radiation exposures below the prevailing level for all or a selected group of people.

The system of radiological protection recommended by the ICRP for proposed and continuing practices is based on the following general principles:

- a) No practice involving exposures to radiation should be adopted unless it produces sufficient benefit to the exposed individuals or to society to offset the radiation detriment it causes. This is the justification of the practice;
- b) In relation to any particular source within a practice the magnitude of individual doses, the number of people exposed, and the likelihood of incurring exposures where these are not certain to be received should all be kept as low as reasonably achievable, economic and social factors being taken into account. This procedure should be constrained by restrictions on the doses to individuals (dose constraints), or the risks to individuals in the case of potential exposures (risk constraints), so as to limit the inequity likely to result from the inherent economic and social judgements. This is the optimisation of protection; and
- c) The exposure of individuals resulting from the combination of all the relevant practices should be subject to dose limits, or to some control of risk in the case of potential exposures. These are aimed at ensuring that no individual is exposed to

radiation risks that are judged to be unacceptable from these practices in any normal circumstances. Not all sources are susceptible to control by action at the source and it is necessary to specify the sources to be included as relevant before selecting a dose limit. (Individual dose and risk limits).

For intervention, the ICRP recommends that two general principles be followed:

- a) The proposed intervention should do more good than harm, i.e. the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and the costs, including social costs, of the intervention; and
- b) The form, scale and duration of the intervention should be chosen so that the net benefit of the reduction in dose, i.e. the benefit in the reduction in radiation detriment, less the detriment associated with the intervention, should be maximised.

The ICRP qualified this advice with the statement that :

"The dose limits recommended by the Commission are intended for use in the control of practices. The use of these dose limits or of any other pre-determined dose limits, as the basis for deciding on intervention might involve measures that would be out of all proportion with the benefit obtained. This would then conflict with the principle of justification. The Commission therefore recommends against the application of dose limits for deciding on the need for, or scope of, intervention. Nevertheless, at some level of dose, approaching that which would cause serious deterministic effects, some kind of intervention will become almost mandatory."

Although generally helpful the ICRP distinction between practices and intervention is not always clear in respect of land contaminated with radioactive materials. Where contaminated land is currently accessible to the public, remedial action taken to reduce exposures would constitute intervention. However, if the land is not accessible at the time remediation is undertaken, but is then immediately redeveloped in such a way as to allow public access, then this would introduce new exposures from the activity remaining after remedial action. Such exposures might reasonably be expected to be subject to similar criteria to those applied to practices. Hence, for fertiliser plants and storage areas it is reasonable to assume that the remediation of the contaminated zones within the operating areas before their final release to the public should be treated as per practices. Thus, as a minimum the additional exposures to any member of the public beyond background levels after final remediation and release of the plant areas, etc, should not exceed 1 mSv/yr.

The extent of any remediation of contamination should be such that the future exposures to occupants of the site, as well as other exposures to workers performing remedial work, are as low as reasonably achievable, economic and social factors being taken into account. Optimisation will, in general, require consideration of the full range of options from no remedial action to complete decontamination. Other options may include actions to immobilise or isolate the radioactive material without removing it from the site. In evaluating these options, the expected future use of the site will be important. For this will determine the likely future exposures as well as the costs and doses associated with any remedial work.

A1.3. Guidance from the UK national radiological protection board.

In the United Kingdom outside of the nuclear industry the most common cause of radioactive contamination on land involves radium salts, primarily from luminising operations. The second major class of contamination has originated from the industrial scale development and processing of minerals, chemicals and metals. In these natural radionuclides, again primarily from the uranium and thorium series of elements, have become concentrated.

To provide guidance on the remediation of these sites UK National Radiological Protection Board (NRPB) has recently provided advice in a consultative document (Barradough, 1996) on radioactively contaminated land. It defines the latter as soil or other materials in the ground at or near the surface. "Contamination" is defined to include artificial radionuclides and natural radionuclides where their concentrations have been enhanced by human actions.

In order to ensure that risks to the public will not be unacceptable, the NRPB has recommended that the excess risk to the critical group, attributable to the residual contamination, resulting from the future use of the site should not exceed a maximum risk constraint of 10^{-5} /yr. This corresponds to the same level of risk constraint as they have recommended for future exposures arising from the disposal of solid radioactive wastes on land (NRPB, 1990). For year on year exposures over a lifetime, this also corresponds to the risk associated with an annual effective dose of 0.3 mSv/yr, the maximum dose constraint which the NRPB recommends for the operation of new controlled practices.

The ALARA Principle is also to be applied, so those actual doses resulting after clean-up may be considerably lower. NRPB has provided additional guidance on a level of future risk below which the optimisation requirement can be relaxed. This is the level below which it is not necessary to consider the application of significant resources, i.e. any money, to achieve further dose reductions. This level is taken to be an excess risk of 10^{-6} /yr, i.e. 20-30 uSv/yr for an average member of the critical group of future site occupants. NRPB recognised that there are likely to be situations where the measures necessary to reduce risks to 10^{-6} /yr are not warranted. For these situations the optimum level of residual risk will lie between 10^{-5} and 10^{-6} /yr.

A1.3.1. Guidance from UK Nuclear Installations Inspectorate (NII)..

Consideration is currently being given in the United Kingdom to the decommissioning of former nuclear industry facilities. Part of that process involves the potential release of the land on which such facilities were built for public use. The regulatory body with responsibility for regulating the operation of major nuclear facilities in the United Kingdom is the Nuclear Installations Inspectorate (NII), which is part of the Health and Safety Executive. It has a responsibility to ensure that no such land is released for unrestricted public use until it can be demonstrated that the land presents no significant risk to the health of any member of the public. The NII has judged that a residual risk of serious harm of $\leq 10^{-6}$ per annum meets that criterion. From its

assessments it has recommended that a residual activity level above the local background of ≤ 0.4 Bq/g is adequate to meet that risk limit for all radionuclides (Delaney, 1999).

A1.4. Regulatory guidance in Germany.

In Germany guidance on the remediation of sites contaminated by naturally occurring radionuclides, such as radium, thorium and uranium, has also been influenced by the extensive areas in the former East Germany, which are affected by the residues from uranium mining operations (Ettenhuber, 1993). Thus the criteria and reference levels recommended by the German Commission on Radiological Protection are:

a) Dose criterion. Reference level 1 mSv/yr in addition to natural regional level; and
b) Radioactivity criteria. Measurable quantities expressed as soil specific activity, c_a and dose rate with the natural level *included*.

$C_a \leq 0.2$ Bq $^{226}\text{Ra}/\text{g}$ for unrestricted use;

$0.2 < C_a \leq 1$ Bq $^{226}\text{Ra}/\text{g}$ for restricted use, e.g. for industrial sites with dose rate ≤ 0.3 $\mu\text{Sv}/\text{h}$

$C_a > 1$ Bq $^{226}\text{Ra}/\text{g}$. This requires site-specific investigation. The limits are calculated for volumes and areas of dumped material, etc., of $\leq 10^5 \text{m}^3$ and 10^4m^2 , respectively.

A1.5. Regulatory Guidance in the United States

In the United States the situation is less clear with the various federal and state environmental protection agencies having slightly different clean-up levels. Thus the U.S. Environmental Protection Agency (EPA) applies several criteria to cleanup actions, such as maximum contaminant levels (MCLs), maximum permissible concentration (MPC), national ambient air quality standards (NAAQS) and ambient water quality criteria (WQS). It has defined levels for various chemical contaminants and radionuclides. Directly applicable numerical limits have been used in certain cases. Some clean-ups under the jurisdiction of the U.S. Nuclear Regulatory Agency (USNRC) have been based on limits specified in NRC Regulatory Guide 1.86 (Devgun, 1993). This defined acceptable surface contamination levels for ^{226}Ra and ^{228}Ra of $0.15 \text{Bq}/\text{cm}^2$ average with a maximum of $0.45 \text{Bq}/\text{cm}^2$ and $0.03 \text{Bq}/\text{cm}^2$ for removable material. The U.S. Department of Energy (USDOE) uses cleanup limits for ^{226}Ra , ^{228}Ra , ^{230}Th and ^{232}Th of $0.185 \text{Bq}/\text{g}$ ($5 \text{pCi}/\text{g}$) for surface layer contamination, which is taken as averaged over the first 0.15m of soil, with $0.56 \text{Bq}/\text{g}$ for deeper material again averaged over 0.15m thick layers. An ALARA ("As low as reasonably achievable") evaluation is also performed to determine if it is possible to cleanup to levels below these standards.

Site-specific cleanup levels are also derived through risk assessments using pathway analysis methods and radiation dose limits. The USDOE uses the RESRAD computer code and an individual dose limit of $1 \text{mSv}/\text{yr}$ for future land use scenarios to derive acceptable residual radioactivity levels. However, actual cleanup levels are frequently lower than these levels in practice as a result of the application of the ALARA principle. For its pathway analyses the USEPA employs the analogous PRESTO and PATHRAE codes.

The situation with respect to individual annual dose limits following remedial works is equally complex. The ICRP recommends an effective dose limit for an individual member of the public of 1 mSv/yr. The USDOE through its Order 5400.5, 1990 implements the same effective dose limit with ALARA. It also uses a guideline value of 0.2 uGy/h above background for γ exposure rates with values in dpm per 100 cm for surface contamination. In applying the provisions of 10 CFR 20 for the possession or use of radioactive materials, the USNRC specifies an annual effective dose limit of 4 mSv for unrestricted areas. However, for uranium fuel cycle operations the provisions of 40 CFR 190 apply, which provides for an annual effective dose limit of 0.25 mSv. Under 10 CFR 20 the reference annual effective dose level for a member of the general public is 1 mSv. However, prior authorisation may be sought under certain conditions for an operation resulting in individual public exposures as high as 5 mSv. The USEPA uses a limiting criterion of 0.1 mSv/yr for emission of radionuclides to ambient air under 40 CFR 61. The USNRC licensing requirements for land disposal of radioactive wastes, (40 CFR 61), specify an effective whole body dose limit of 0.25 mSv/yr. The USEPA's standards for radioactive waste disposal include a groundwater protection requirement, (40 CFR 191), which specifies an effective dose limit of 0.04 mSv/yr.

Dose-based cleanup limits used by the USDOE and USNRC inherently involve pathway analysis methodologies (Devgun, 1993; Wolbarst, 1996) and the use of dose conversion factors to derive cleanup criteria. These limits can allow greater flexibility in site clean-up, while achieving protection goals, because they are based on site-specific analyses and considerations. Such limits also lead to application of different cleanup criteria to different sites, which may have regulatory, social and political implications with respect to their public acceptability.

A1.6. Regulatory guidance in Canada.

In Canada there has been a long programme of remediating sites contaminated with naturally occurring radioactive materials, resulting from the early years of radium and uranium production. This work has been undertaken by the Low-Level Radioactive Waste Management Office (LLRWMO) of the Federal Government. The criteria derived by the Federal/Provincial Task Force for remediating these sites (Pollock, 1993) were:

- | | |
|-----------------------------------|---------------------|
| a) Radon progeny levels (indoors) | < 0.02 WL. |
| b) Gamma radiation (outdoors) | < 1 uGy/h at 1 m. |
| c) Gamma radiation (inside) | < 0.5 uGy/h at 1 m. |

These criteria include background and were intended to be applied independently. There were no explicit criteria for non-radioactive contaminants.

In later work a primary cleanup criteria of 1 mSv/yr dose above background, but excluding the dose from radon progeny, and an average annual indoor radon progeny concentration of 0.02 WL have been applied. A risk assessment using a pathway analysis has then been used to derive soil criteria (Pollock, 1993). The derived criterion was developed assuming a sufficiently large areal and volumetric extent of contamination for both a house and garden to be constructed on the

contaminated soil. Typical site specific, derived soil limits for²²⁶Ra were 0.2-0.3 Bq/g.

A1.7. Proposed IAEA guidance.

The draft IAEA guidance on the application of radiation protection principles to the clean-up of contaminated areas (IAEA, 1997) is summarised in the following table. Sites are banded according to the annual doses received by average members of the critical group **before** clean-up. The suggested clean-up levels are then, in principle, to be determined by optimisation. However, with the possible exception of situations initially in the upper end of Band 4, where a justified and optimised clean-up might conceivably leave a situation towards the lower end of Band 4, any clean-up would normally need to produce an end-point, which was at least one band lower and no higher than Band 4.

Table1. Proposed IAEA clean-up levels

| Band No. | Range of Annual Doses (to average member of critical group) | Is Clean-up needed | |
|-----------------|--|---------------------------|---------------------------|
| | | With constraint | Without constraint |
| Band 6 | > 100 mSv/yr | Always | Always |
| Band 5 | 10-100 mSv/yr | Always | Almost always |
| Band 4 | 1-10 mSv/yr | Almost always | Usually |
| Band 3 | 0.1-1 mSv/yr | Usually | Sometimes |
| Band 2 | 10-100 µSv/yr | Sometimes | Rarely |
| Band 1 | < 10 µSv/yr | Almost never | Almost never |

A1.8. Exemption limits

The principle of exemption was developed jointly by the IAEA and NEA (IAEA, 1988) and is also discussed in ICRP 60 (ICRP, 1991). It was developed through international recognition that regulatory systems need to include provisions for granting exemptions in cases where it is clear that a practice involving radioactive materials is justified, but regulatory provisions are unnecessary or unwarranted. This is the case where the risks are assessed as low.

The exemption system has been used to allow organisations to use and dispose of low levels of radioactive materials without the need to notify national regulatory bodies. In the United Kingdom an Exemption Order currently applies to phosphatic substances. These are materials with a low specific activity, containing enhanced levels of natural radionuclides from the uranium and thorium decay chains. The quantities of material involved here are usually of the order of a few tonnes. However, the underlying assumption behind the exemption system is that the quantities involved were small. It is questionable whether under the strictest interpretation of the associated legislation exemption levels should have been applied to large scale

operations. In practice, individual regulators allowed the Phosphatic Substances Exemption Order to be applied to much greater quantities of contaminated material, particularly during the clean-up of radioactively contaminated land. In these cases the ultimate level of risk has still be judged to be low, as the waste was only allowed to go to selected landfill disposal sites. These disposal sites were selected for the higher quality of their containment.

The basis for exemption limits and their values have recently been revised (Harvey, 1993; Robb, 1994; IAEA, 1996), following the ICRP guidance in its publication ICRP60 (ICRP, 1991). For countries within the European Community the new limits are incorporated into the European Community Directive, which lays down the "Basic Safety Standards (BSS) for the Protection of the Health of Workers and the General Public against the Dangers of Ionising Radiation (CEC, 1993)". Similar limits are being proposed by the IAEA in its interim report for comment (IAEA, 1996).

These new limits have been derived using pathway quantitative risk assessments. The scenarios considered in the analyses included land filling, incineration, recycling and reuse of the materials. The exempt levels have been derived on the basis of an individual dose criterion of 10 $\mu\text{Sv}/\text{yr}$, as this is deemed to correspond to a trivial dose. Under accident or misuse scenarios the public dose limits of 1 mSv/yr effective dose and 50 mSv/yr skin dose are used with a probability of occurrence of 0.01. The definition of "moderate amounts" of material is again not stated explicitly. However, a value of 1 tonne is often referred to as the interpretation and this is used in the supporting pathway analyses. Quantities of this order may be of value in the management of scales from decontamination operations, but are much smaller than are likely to arise from any site remediation works. Hence, exemption levels may not be of explicit value in the sentencing of contaminated material in the remediation of the contaminated land areas. However, they do justify the methodology used in determining the clean-up levels.

A1.9. Recommended dose constraints and activity levels for clean-up work

The international criteria currently being used for the remediation of radioactively contaminated land are summarised in Table 2 below.

Table 2 : *Summary of international remediation criteria.*

| Country | Criterion | Dose rate. (Additional to background) | Specific Activity. ^{226}Ra Bq/g | Comments |
|----------|-------------------|---|---|----------------------------------|
| ICRP | ALARP | < 1 mSv/yr | | |
| IAEA | Critical Group | < 100 $\mu\text{Sv}/\text{yr}$ $\leq 10 \mu\text{Sv}/\text{yr}$. No further optimisation needed | | |
| UK(NRPB) | Critical Group | $\leq 30 \mu\text{Sv}/\text{yr}$ | | No further optimisation needed |
| USA | Public | 0.25-1 mSv/yr and $\leq 0.2 \mu\text{Gy}/\text{h}$ | ≤ 0.185 in top 0.15 m of soil ≤ 0.56 below top layer | |
| Canada | ^{226}Ra | 1 mSv/yr 1 $\mu\text{Gy}/\text{h}$ (outdoors) 0.5 $\mu\text{Gy}/\text{h}$ (indoors) | 0.2-0.3 | <0.02 WL (Radon progeny indoors) |
| Germany | ^{226}Ra | 1 mSv/yr | ≤ 0.2 (unrestricted) 0.2 < ≤ 1.0 (restricted use) | $\leq 0.3 \mu\text{Sv}/\text{h}$ |

The implication of the IAEA guidance is that where possible residual doses should be reduced to below approximately 100 uSv/yr for final clearance of the sites. No further optimisation is likely to be justified, if the doses can be reduced below 10 uSv/yr. In practice, these limits are similar to those proposed by the NRPB in the United Kingdom and are appropriate for use in determining clean-up criteria for fertiliser plant sites.

For clean-up of any areas already open to the public, a higher dose limit approaching 1 mSv would be justifiable under the intervention criteria. However, in practice little additional effort and expense is likely to be involved in reducing the doses in these areas to considerably lower levels. Hence the ALARA optimisation principle would support clean-up levels of closer to 100 uSv/yr as the dose target.

International practice is that no further optimisation can be justified, if remediation results in a residual risk limit of $\leq 10^{-6}$ per annum of serious harm to any member of the public. Risk assessments by the UK Nuclear Installations Inspectorate show that a residual contamination level above the local background of ≤ 0.4 Bq/g will satisfy this condition.

APPENDIX 2.: REMEDIATION APPROACHES FOR RADIO- ACTIVELY CONTAMNATED SITES.

A2.1. Introduction.

A range of remedial actions, which have the potential to improve the land contamination situation, are available (USEPA, 198S, 1990; Vandenhove, 1999). There are two stages to remediation, each of which has options associated with them. These are:

- a) The removal, redistribution or segregation of radioactive material on the site to reduce exposure to operators and the public; and
- b) The storage or disposal of any radioactive material removed from the site.

Options for the first stage include:

- Do nothing.
- Control access to the site and handle during facility decommissioning
- Dilute and disperse by mixing with uncontaminated soil, such as by deep ploughing, or land spreading
- Cap by covering with clean material
- Stabilise in-situ by selectively encapsulating and even vitrifying highly contaminated areas.
- Excavate selectively and treat/store/dispose on- or off-site.

Variations or combinations of these options are possible and may help to alleviate shortcomings in any individual option.

Options for the storage and disposal of material removed during site remediation include:

- Long term storage in an engineered facility, which could include storage pits.
- Burial in an shallow land disposal facility.

Any proposed treatment would be directed at reducing the volumes of contaminated material to be handled, concentrating the radioactivity further by physical and/or chemical means.

A22. Do nothing option.

The "Do nothing" option implies controlling each site during its operational life to prevent any operator from receiving doses in excess of 1 mSv per annum and then abandoning the site after that period. Control would be necessary during the operational phase, as the external radiation rates are such that operators could receive doses in excess of 1 mSv per annum. On closure and decommissioning of the production facilities, the utilisation of the site should decrease to a very low level. This is because there will be nothing of value in material, agricultural, mineralogical or tourism terms to warrant visits. This option must always be considered. However, it is unlikely to be justifiable for most sites in the United Kingdom and Europe, as its

underlying premise for justification is very low occupancy for thousands of years. It could possibly have application elsewhere in remote phosphate mining areas, etc.

Such a management approach is likely to lead to the activity on the site being slowly redistributed with time both within and beyond the site as a result primarily of wind and rain erosion. This will reduce peak and average activity concentrations, but will extend the areas for potential exposures. Behind this option is the requirement that likely occupancy rates for the sites do not increase faster than the rates of radionuclide decay, which provides a factor of 2 over the first 100 years and a further factor of 3 over the succeeding 1000 years. This in turn assumes that the attractiveness of the site, as a result of technological changes, improvements in water availability, etc., do not change significantly from present day conditions. This option offers no additional mitigation against the impacts of any of the other pathways.

Such an approach has the advantage of being very cheap to implement. However, it is heavily reliant on factors beyond management influence for its long-term viability. It offers no protection against the possibility of individuals staying on the sites for longer periods than those required to exceed the annual public dose limits. Hence, it is unlikely to be acceptable to regulatory authorities and would always leave the operator open to claims from affected parties for compensation for personal injury. It is, therefore, a potentially a high risk option and is very unlikely to be attractive.

A2.3. Dilute and disperse option.

This option involves mixing the contaminated surface layer with less contaminated or clean deeper material. It could also be applied with clean material being added over the contaminated layer. This would be necessary if it were to achieve the required dose reduction factors. It results in a homogenisation of the contamination over the area of the site, etc, so eliminating the impact of hot spots. Depending on the thickness of the contaminated layer and the depth of mixing, it may reduce average contamination levels by typically factors of 2-4, but potentially as high as 10, if the original layer of contamination is very thin. If deep ploughing techniques are used, the mixing depths are usually limited to -1 m. However, greater mixing depths can be achieved using other techniques. Dust inhalation impact will also be reduced in proportion to the decrease in the average contamination concentration on the surface. The ingestion pathways will only be reduced, if the mixing depth includes clean material from below the depth of potential crop routes. Otherwise, since the radionuclide inventory is unchanged, the impact on the other ingestion routes through migration of contaminated groundwater, etc., will be small.

As an approach this is quite cheap to implement. The operators will be working on tractors or bulldozers, which will both raise them above the contaminated ground and likely further shield them from external radiation through the metal body of their plant. Thus with appropriate respiratory precautions to prevent any inhalation doses to the operators, this approach can be implemented safely. It does reduce the key external dose rates, so proportionately increasing potential residence times on-site for the public before the annual dose limits are exceeded.

It has the disadvantages that the radionuclide inventory on-site remains unchanged and little has been done to reduce its long-term impact on the environment. In addition, its acceptance by regulators is very uncertain. It would not generally be accepted in the United Kingdom or Europe. Even if accepted in the near term, the possibility exists that changes in regulatory standards may require further remedial action. Should the latter occur, this strategy will have increased the volume of material requiring treatment by the ratio of the thickness of the contaminated layer to the mixed depth. This will increase the cost and potentially the difficulty of any further remedial action, particularly if excavation is involved. Such an approach has been used in the past in a number of cases and several have required further action at later dates.

The dilute and disperse principle is widely accepted and applied for low-level, liquid and gaseous radioactive wastes. However, as an approach it is not accepted international best practice to deliberately dilute contaminated solid material with a substantially greater volume of clean material. Hence, there may be regulatory difficulties in adopting such an approach, particularly if the contaminated soil after mixing is still above internationally defined exemption limits for radioactive materials.

Land spreading is a disposal option, which is not often considered for radioactive waste. The advantages of land spreading are that the technology is relatively simple and is cheap to implement. It would also result in a permanent solution for the disposed of wastes as recovery would no longer be practical. The disadvantages are that it is relatively untried, although land spreading of radium sludge from drinking water treatment systems has been an allowed policy in Illinois in the USA since 1984 (USEPA, 1988). In addition, there could be considerable political and regulatory difficulties in implementing such a strategy, particularly since it is not accepted best international practice.

A2.4. Capping.

The option exists of covering the contaminated soil at each site with a cap. This approach is frequently used on sites in the United Kingdom with non-radioactively contaminated soil, particularly where the contaminants are effectively insoluble and hence immobile. Such areas are frequently made into decorative features of sites with low human occupancy or risks of intrusion. If the cap were of adequate thickness, then given the domination of doses received by external exposure, this approach could provide the required dose reduction factors. In its simplest form this could be a layer of clean soil, etc. (Cancio, 1993). Alternatively, it could be an engineered multi-layer cap, which is designed to minimise infiltration and radon emanations and to resist erosion and intrusion. Capping could be employed on a temporary basis during the continued operational lives of the production facilities, whereupon final decisions could be taken on the final solution for the waste arisings. Alternatively, it could form the final solution, as has frequently been applied to the radioactive mining and milling wastes arising from the extraction of uranium, radium and rare earths.

The benefits of capping are:

a) It can reduce external radiation exposures to any chosen extent, including negligible proportions, depending on the cap thickness used.

b) It eliminates the dust inhalation pathway as no contaminated material is exposed to the atmosphere.

c) It reduces radon exposures by restricting the rates of radon release.

d) It can reduce radionuclide migration through the groundwater pathway and hence into the human food chain. This is achieved by restricting rain infiltration rates and hence the downward migration of radionuclides, particularly if impermeable materials are incorporated into the capping layer. This in turn reduces the potential ingestion risks.

e) It can reduce the risks of exposures through accidental intrusion by reducing the accessibility of the waste. The degree of protection provided can be increased substantially, depending on the cover design employed.

f) There is very extensive international experience and acceptance of using capping for containing the rates of migration of contaminants from sites to very low and acceptable levels. It is, for example, recommended by the IAEA for remediating uranium mining and milling waste sites.

g) It has high reliability, particularly if maintained properly.

h) It is relatively inexpensive to apply.

The disadvantages of this approach are:

a) The radioactive contamination remains on site until it decays away to negligible proportions, which is ~20,000 years with ²³⁸Pu. In the United Kingdom this could also lead to the requirement for an authorisation to accumulate and store radioactive materials with continued regulatory supervision.

b) The contamination remaining under the cap is potentially retrievable and hence open to future changes in standards or legislation to the requirement for further remedial works.

c) The presence of contaminants permanently disposed on site can restrict potential changes of use of the site, potentially ruling out those which could realise the greatest value for the land.

Given the nature and locations of the sites, a simple soil cap would be unlikely to avoid erosion by the wind and winter rain. Hence, as a minimum any cap would need to incorporate a layer to stabilise the surface. In addition, it would likely need to incorporate some form of impermeable membrane made from natural and/or man-made materials.

A2.5. In-situ stabilisation of selected areas.

Stabilisation of selected highly contaminated areas, such as hot spots, can be achieved by the use of cement encapsulation or in-situ vitrification. The latter has been used in the USA at Hanford and Oak Ridge to treat soil contaminated with various radionuclides and was used to vitrify several disposal pits containing radioactively-contaminated soils and wastes at Maralinga in Australia. It has the advantage that the contaminated soil and scale are melted to form a homogeneous glass block, which contains all of the radionuclides in a very impermeable, monolithic

mass. The leaching rates of the radionuclides from vitrified blocks are extremely low, so effectively preventing any significant migration from them. The monolithic glass structure also effectively eliminates any inhalation pathways by eliminating dust formation and severely retarding radon releases. The vitrification process results in a volume reduction, typically by 30-50 % . Hence, the resulting vitrification pit is usually is backfilled with clean soil, so providing extra attenuation to external radiation. The disadvantages of the technique are that it is both slow to apply and very expensive. The production rate is controlled by the rate of heating the contaminated soil to temperatures -1500°C. Likewise, the bulk of the costs are associated with the electrical energy input required. Typical production rates per unit are -30 m³per day with application costs of -\$4-6,000/m³ treated. Thus, it would be prohibitively expensive to apply on a large scale at the sites.

The option of stabilisation by cementation or other fixing agents does exist (USEPA, 1988). They offer many of the advantages of in-situ vitrification, but without providing such a high quality of containment. They will result in an increase in the volume of waste and hence will intrinsically provide additional shielding to reduce external radiation exposures. In addition, the application technology is much less complex and hence is more suitable for field application without the need for a highly trained, imported workforce.

A2.6. Excavate selectively and treat, store or dispose on- or off-site.

The option exists of selectively excavating areas above acceptable contamination levels and treating, storing or disposing of the resulting wastes. The approach would be to define the areas requiring treatment on the basis of the risk assessment, using surface γ -radiation measurements, calibrated against previous individual radionuclide and depth profiles to define the locations in the field. The contaminated material can then be removed using standard earthmoving equipment, such as excavators, backhoes, front-end loaders, graders and scrapers with trucks or scrapers, if used, for the movements. The operations can be carried out either wet or dry. The wet route has the advantage that wetting minimises dust generation. However, it has the disadvantage that contamination sticks more readily to plant, so increasing final decontamination demands. If the dry route is used, the earthmoving equipment may require special modification to seal and pressurise the cabins and to HEPA filter all air intakes. The advantage of this route is that contamination levels on plant are invariably significantly lower, so reducing final decontamination demands. In damp climates, such as the United Kingdom, damping down is the usual approach. The dry approach is more suitable for semi-arid regions.

After excavation the contaminated soil, scale, etc., could be treated in order to concentrate the activity into a small volume requiring storage and ultimately disposal. The remainder, which would constitute the bulk of the material, could be acceptable for discharge as clean material or would be suitable for disposal on a local landfill. The available treatment technologies include chemical separation using inorganic salts, mineral acids or complexing agents and physical separation using screening, classification, gravity concentration or flotation. Of the chemical extraction processes, leaching with mineral acids and/or complexing agents is most effective (USEPA,

1990). However, the product of this treatment is a liquid stream, which requires further treatment to remove and concentrate the radioactivity into a small volume. This can be achieved by ion exchange, carbon or chemical precipitation, followed by filtration or ultra filtration.

The physical separation processes tend to rely on the contamination being present or enhanced on a discrete phase or component of the particulate mass. This could be a finer particle fraction or a denser or magnetically susceptible phase. However, it is unlikely to be the case with contaminated soils and scale. Hence, it is unlikely that physical separation techniques will provide any significant reduction in volumes of waste for disposal.

A variant of physical separation is where the contaminated soil, etc, is fed as a thin layer on a conveyor belt through an array of gamma radiation monitors. The latter are used to detect soil, etc, with contamination above selected levels. This contamination can then be removed either remotely using variants of gate systems or manually.

The application of chemical and/or physical separation techniques will require laboratory and likely pilot plant trials to determine the optimum processing conditions and to determine the degree of decontamination achievable. If applicable, it will then require the construction of a plant on-site and its decontamination and decommissioning upon conclusion of the work. The degree of decontamination achievable varies with the nature of the material treated. Much of the contamination has effectively been deposited by chemical precipitation, which is itself a key chemical concentration process. Hence, it is unlikely that the degree of decontamination achieved will be high. It is, therefore, unlikely that the additional costs involved with chemical and/or physical treatment can be justified in terms of the reductions achievable in the volumes of waste for storage and disposal.

The costs for disposing of low-level wastes in the UK, France and USA are in the range \$3,000-10,000/m³. This can make the disposal of thousands cubic metres of contaminated soil very expensive.

The options for interim storage include the construction of an engineered store for packaged waste and the construction of one or more covered mounds on-site or at a chosen central location.

An engineered store would likely be a variant on or an extension of the currently proposed interim store to hold the NORM scale wastes from the decontamination operations. Such a store is likely to hold wastes in drums for ease of handling. This would require any wastes to be similarly packaged or the store to be modified to handle half-height ISO containers. The latter development would require the installation of special handling equipment, so increasing the overall cost of the operation. A typical store cost would be \$35- 100/m³ with additional costs for any special handling equipment. In addition, there would still be annual operating costs for the life of the store, followed by the final disposal costs.

The disposal options include an engineered mound, either entirely above ground or initially constructed as a trench and then covered with clean spoil and capped. Such a facility would be very similar to those described previously under the capping option. Recommended designs for such disposal facilities are available through the IAEA and the US EPA and DOE, as they are extensively used for active wastes from uranium, thorium and radium mining. In addition, such facilities have received regulatory approval in many countries, including USA, Canada, France, Germany, Italy, Spain, Sweden and Australia. The mound concept is not expensive to develop and cost scales effectively linearly with capacity.

A2.7. Ranking of options

The choice of preferred management option will depend a number of factors including:

- a) The volumes of contaminated material requiring disposal.
- b) The availability and capacity of existing storage and disposal facilities.
- c) The costs/m of providing new or extra storage or disposal capacity.
- d) The difficulties in obtaining regulatory approval for new disposal facilities.
- e) The ease of retrievability and hence the finality of any disposal option.
- f) The logistics of waste movements between sites.
- g) The level of reduction in risk achieved.

After consideration of these factors the most common approach adopted in the United Kingdom is excavation with prompt disposal off-site in authorised landfills for the exempt waste and at the BNFL Drigg repository for low-level waste.