LLW Repository Ltd

LLWR Environmental Safety Case

Examination of International Radioisotope Assay Practice

This report was prepared by the Low Level Waste Repository Site Licence Company.

LLWR/ESC/R(10)10015 Issue 1 Date: 16th March 2010

Title	Name	Signature	Date
Environmental Advisor	Author: Phil Carson	[signature]	16/3/2010
Environmental & Technical Manager	Checked by: Simon Hunter	[signature]	16/3/10
ESC Project Manager	Approved by: Richard Cummings	[signature]	19/3/10

This page is left blank intentionally.

LLW Repository Ltd

Examination of international radioisotope assay practice.

Phil Carson

Executive Summary

The examination of international best practices has shown that LLWR is largely aligned with radioisotope assay techniques at sites with low but yet continuous radioactive discharges. Best alignment was found with the Hanford Site monitoring programme in the United States and alignment with assay techniques at France's Centre de la Manche site is also good.

One area of discrepancy appears to be in the use of alpha scintillation counting (ASC), however, examination of differences between the requirements of monitoring programmes at sites (e.g. gross vs isotope specific assay) reveals that ASC, although not necessarily as widely used internationally, is not inappropriate for use on the LLWR site.

Introduction to Study

Schedule 9 of LLWR's RSA Authorisation from the Environment Agency (EA), places a number of requirements for improvement of the LLWR site. Requirement 3 necessitates:

The Operator shall provide the Agency with a full report of a comprehensive review of the means used to assess the activity of radionuclides in disposals and to determine compliance with this Authorisation including consideration of national and international best practice.

An initial report complied by NNL and approved by LLWR (April 2009) addressed the majority of points under Requirement 3. However, a review of the document by EA in September 2009 revealed this report had not included a sufficient survey of international best practice and EA were therefore not confident that LLWR had taken on board learning from similar facilities internationally; i.e. those with low but yet continuous radioactive discharges. This study aims to address these shortcomings by looking at radioactive waste assay techniques at sites in North America and within the European Union. Sites and international regulatory advice examined include:

- Hanford, WA, USA - The Hanford site is a mostly decommissioned nuclear production complex on the banks of the Columbia River in Washington State. Historical activities including plutonium production are similar to the Sellafield Site. Today, much of the activity on the vast site is related to cleanup of these legacy facilities. In addition to decommissioning, Hanford includes a generating site (Columbia Generating Station), a seat of scientific research (PNNL) and the location of one of the United States' three licensed LLW disposal facilities (operated by American Ecology Inc).

- Centre de la Manche, Normandy, Northern France - Centre de la Manche, located 1km east of the AREVA La Hague reprocessing plant, was the French national LLW and short-lived ILW facility from 1969 until 1994 when it was superseded by the Centre de l'Aube site. Centre de l'Aube consists of two separate disposal facilities, Centre de Soulaines (engineered disposal for LLW) and Centre de Movilliers (trench-based disposal for VLLW) located to the east of Paris in central France. Centre de la Manche is now capped with a bitumen based membrane and is under institutional control by ANDRA, the French national radioactive waste management agency.

- Recommended best practice in the United States for measuring radionuclide emissions (alpha, beta and gamma counting), as issued by the US Environmental Protection Agency (US EPA) Technology Transfer Network.

- Approved tritium assay methods at the Savannah River Site, Georgia (Tritium Extraction Facility - TEF) and other US Department of Energy (DoE) owned sites. TEF extracts tritium for the US nuclear deterrent from materials irradiated in commercial power generation reactors.

- Overview of monitoring at the Swedish Final Repository (SFR) geological disposal facility in Forsmark, Sweden.

Summary of Hanford Site Environmental Monitoring Assay Protocols, including the Richland LLW facility (American Ecology) and Environmental Restoration Disposal Facility (ERDF)*

* Note: ERDF is a disposal facility (opened 1995) for LLW and limited volumes of mixed waste arising from Hanford site decommissioning and remediation

ANALYTICAL TECHNIQUES

Sample Type	Analysis	Method	LLWR Techniques
Air-collected on glass fibre (GF) filters. LLWR collect aerial stacks discharges on GF filters, bulk air samples on borosilicate microfibres.	Gross alpha	Gas-flow proportional counting	Alpha scintillation counting.
	Gross beta	Gas-flow proportional counting	Gas-flow proportional counting
	Gamma	HPGe gamma spectroscopy	HPGe gamma spectroscopy. (bulk air only)
	U, Pu isotopes	Alpha energy analysis	Alpha spectrometry.
Surface water	Gross alpha	Gas-flow proportional counting	Alpha scintillation counting.
	Gross beta	Gas-flow proportional counting	Geiger-Müller. (proportional counting used for Drigg Stream water)
	Gamma	HPGe gamma spectroscopy	HPGe gamma spectroscopy
	U, Pu isotopes	Alpha energy analysis	Alpha spectrometry.
	Tritium	Liquid scintillation counting	Liquid scintillation counting
Ground water	Gross alpha	Gas-flow proportional counting	Alpha scintillation counting
	Gross beta	Gas-flow proportional counting	Cerenkov counting, liquid Geiger-Müller, proportional counting
	Tritium	Liquid	Liquid

		scintillation counting	scintillation counting
	Hanford also implements an extensive trace organics, trace inorganics and trace metals monitoring programme in ground water.		
Food products	Gamma	HPGe gamma spectroscopy	HPGe gamma spectroscopy
	Tritium	Liquid scintillation counting	Liquid scintillation counting
	Note: LLWR also include alpha and beta counting in milk analysis.		
Vegetation	Gamma	HPGe gamma spectroscopy	HPGe gamma spectroscopy
	U, Pu isotopes	Alpha energy analysis	Alpha spectrometry.
	Note: LLWR also include alpha and beta counting and tritium in herbage analysis.		
Sediments	Alpha	Gas-flow proportional counting	Alpha scintillation counting
	Beta	Gas-flow proportional counting	Geiger-Müller counting.

Summary of Radioisotope Assay Protocols in France, ANDRA: Centre de la Manche.

Surface Water Monitoring (Cap Runoff)

Analyte	Centre de la Manche	LLWR
Alpha Analytes: Ra-226 (trapped on MnO_2), U-234, U-235, U-238, Pu-238, Pu-239, Pu-240 Am-241, Cm-244	Alpha spectrometry	Total alpha relative to Pu-239 and Pu-240 using alpha scintillation counting
Beta Analytes: Tritium (H-3), C-14, Cl-36, Ni-63, Sr-90, Tc-99, Sm-151, Pu-241	Liquid scintillation counting	3-H by liquid scintillation counting
		Total beta relative to Sr-90/Y-90 or K-40 using Cerenkov counting, liquid Geiger-Müller counting and gas-flow proportional counting.
Gamma Analytes: K-40, Co-60, Nb-94, Ag-108,Ag-110,Cs-134,Cs-137, Pb-210, Pb-212, Pb-214, Ra-226, Th-234, U-235, Am-241	HPGe gamma spectrometry	LLWR do not assay for gamma nuclides in surface waters, gamma scan of Drigg Stream waters is by HPGe gamma Spectrometry.

Sediment Monitoring

Analyte	Centre de la Manche	LLWR
Alpha Analytes: U-234, U-235, U-238, Pu-238, Pu-239, Pu-240, Am-241, Cm-244	Alpha spectrometry	Total alpha relative to Pu-239 and Pu-240 using alpha scintillation counting
Beta Analytes: C-14, Cl-36, Ni-63, Sr-90, Tc-99, Sm-151, Pu-241	Liquid scintillation counting	Total beta relative to K-40 using Geiger-Müller counting.
Gamma Analytes: K-40, Co-60, Nb-94, Ag-108, Ag-110, Cs-134, Cs-137, Pb-210, Pb-212, Pb-214, Ra-226, Th-234	HPGe gamma spectrometry	HPGe gamma spectrometry.

Aqueous Discharge (Leachate) Monitoring

Analyte	Centre de la Manche	LLWR
Alpha Analytes: Ra-226 (trapped on MnO_2), U-234, U-235, U-238, Pu-238, Pu-239, Pu-240, Am-241, Cm-244	Alpha spectrometry	Total alpha relative to Pu-239 and Pu-240 using alpha scintillation counting
Beta Analytes: Tritium (H-3), C-14, Cl-36, Ni-63, Sr-90, Tc-99, Sm-151, Pu-241	Liquid scintillation counting	3-H by liquid scintillation counting Total Beta relative to Sr-90/Y-90 or K-40 using Cerenkov counting, liquid Geiger-Müller counting and gas-flow proportional counting
Gamma K-40, Co-60, Nb-94, Ag-108, Ag-110, Cs-134, Cs-137, Sb-210, Pb-212, Pb-214, Ra-226, Th-234, U-235, Am-241	HPGe gamma spectrometry	VTNSEL and EA carry out a gamma scan (by HPGe gamma spectrometry) as part of quarterly check monitoring on leachate from the marine holding tanks.

Aerial Discharges

Unlike the LLWR, the Centre de la Manche Site does not have any controlled stacks (e.g. LLWR's PCM magazine stacks and DGF). Air samples, which are collected on GF filters are restricted to gross alpha, gross beta and tritium by liquid scintillation counting. At LLWR air is counted for gross alpha (alpha scintillation), gross beta (proportional counting), gamma (gamma scan) plus total Pu-alpha, Am-241, U-alpha (alpha spectrometry) and Pu-241 (liquid scintillation counting). In addition, radon monitoring is carried out at monthly intervals at sample points at the northerly and southerly extremes of the site. This is in line with the intermittent radon monitoring of trenches, vaults and buildings within 250m of both at LLWR.

Vegetation Monitoring

Historical monitoring of the site reveals that K-40 and Be-7 are the predominant radioisotopes present in vegetation on the Centre de la Manche; K-40 is assayed for by gamma spectrometry whilst Be-7 decays by electron capture and must be assayed for by different methods. Other gamma emitting analytes identified by gamma spectrometry are Cs-134, Cs-

137, Co-60. LLWR vegetation monitoring includes gamma spectrometry, but also extends to total alpha (by alpha scintillation counting), total beta (by Geiger-Müller) and tritium (liquid scintillation counting).

Agricultural Produce Monitoring

Milk, potatoes, vegetables, meat and fish produce are all monitored as part of the dose assessment for the site, however methodologies and assay technique information was not available. It has therefore not been possible to make direct comparisons between LLWR and Centre de la Manche.

United States EPA Approved Assay Methods in use at LLWR for Environmental Monitoring.

Alpha

Name	LLWR Use
Direct alpha spectrometry Method A-3	Bulk air sampling (collected on Borosilicate microfibres) uses AS to determine Pu-Alpha. (VT Method QAAM 835)
Direct alpha counting (gross alpha determination). Method A-4	Used to count alpha emissions from GF filters on stacks e.g. PCM Magazines and DGF. Monitoring carried out onsite immediately and by VT following a decay period. (VT Method QAAM 523)
Chemical determination of uranium Method A-5	LLWR do not assay for uranium isotopes. Historically, uranium breakdowns from total alpha in bulk air samples have been provided but this is not routine. Uranium is a component of the acceptance limits for waste consigned to LLWR.
Radon-222: Continuous gas monitor Method A-6	LLWR carry out handheld intermittent radon monitoring on the trench cap, trench cap probe holes, vault perimeter and buildings within 250 metres of the vaults.
Radon-222: Alpha track detectors Method A-7	As above

Beta

Name	LLWR Use
Direct beta counting (gross beta determination) Method B-4	Gaseous discharges from the magazine stacks and DGF are collected on a GF filter which is dissolved and subject to beta counting. Bulk air sample (borosilicate filter) subject to same method. (VT Method QAAM: 523)
Liquid scintillation spectrometry Method B-5	Use throughout the environmental monitoring programme (tritium) and analysis of liquid discharges from MHT. Cerenkov counting (direct counting of β emission without fluors) also used. (VT Method QAAM: 131). Scintillation counting also for Alpha counting (VT Methods QAAM: 834, 482,131)

Gamma

Name	LLWR Use
High resolution gamma spectrometry. Method G-1	Part of check monitoring sample (quarterly) for discharges from MHT and environmental monitoring samples (sediment, milk, herbage, ambient air, surface and river waters). High resolution P-type coaxial HPGe detectors are used. VT are UKAS accredited for γ photons of energy 100keV to 1450keV. Combined with LEPS (Low Energy Photon Spectrometry) for I-129 and Am-241. (VT Methods QAAM: 620, 626, 627)

Tritium Assay methods the Savannah River Site (Tritium Extraction Facility) and other United Stated Deparment of Energy (DoE) sites.

The US DoE approves a variety of methods including mass spectrometry and calorimetry for tritium assay on its sites. Such methods are normally utilized when tritium is present in higher concentrations.

On DoE sites, low concentrations of tritium in liquid form, such as in aqueous discharges are generally measured by using a scintillation counter. A sample of the liquid is mixed in a scintillation cocktail (normally primary and secondary fluorescent species in an aromatic organic solvent such as naphthalene), and the quantity of tritium in the sample is measured by counting the resulting photons with a calibrated photomultiplier tube and counter adjusted to identify pulses of light energy related to the tritium low energy β-emission.

Relevance to LLWR site

Tritium on the LLWR is generally found in low concentrations in liquid discharges, thus methods such as calorimetry are inappropriate as total heat emission from a soft β emission characteristic of tritium would be small in comparison to experimental error of the method. Tritium concentrations in discharges through the marine pipeline (leachate system) are therefore monitored using the liquid scintillation counting method (VT UKAS accredited method QAAM 130) in line with US DoE practices. Under the monitoring programme for herbage, milk, river (Drigg Stream), surface and ground waters, low tritium concentrations are assayed by liquid scintillation counting (under the same UKAS accredited method).

SFR Forsmark (Swedish Final Repository; geological disposal for LLW and ILW from Swedish nuclear operations). Operated by SKB.

Sweden has a unique policy in the European Union of geological disposal for its LLW (and ILW) at the Forsmark site on Sweden's east coast some 50 metres below the Baltic Sea. As a result of this very different disposal method, radionuclide discharges and monitoring are very different to sites discussed previously. A detailed comparison between LLWR and SFR practices is therefore of limited value. However, a brief comparison of groundwater and aerial discharge measurement techniques is made here.

Water monitoring is restricted to the groundwaters around the SFR. Groundwaters are subdivided into areas where contamination is possible (few m^3 per annum) and areas where contamination is not possible. All groundwater from contaminated areas is assayed at the Forsmark Nuclear Power Plant and treated if necessary. Groundwater from uncontaminated areas is assayed on a quarterly basis and is pumped out of the SFR and used as cooling water at Forsmark. Note that groundwater in the area is extremely immobile due to the pressure exerted by the Baltic Sea, groundwater surrounding the SFR is around 7000 years old.

Aerial discharges from the site are obviously limited, however, discharges from the repository's exhaust air system are collected on a filter and analysed every 2-4 weeks.

Conclusions of Study

In general LLWR radioisotope assay practices appear largely in line with relevant international best practice in the United States and France. It does appear that the use of scintillation counting for alpha emitters is not used or recommended by the sites/regulators featured. This is possibly because scintillation as an (alpha) spectroscopic method has historically suffered from high background count rates in its energy range caused by cosmic radiation and environmental radiation events in the scintillation cocktail.

Despite this, the ease of absorption of alpha particles means that they tend to transfer all their energy into the cocktail (unlike β and γ emissions which transfer only small proportions of their energy). Therefore, if the counter used can discriminate against low energy photons (from β/γ decay) then the remaining higher energy species can be used to make gross alpha determinations. At LLWR, such gross measurements make up much of the monitoring of alpha emissions, it is therefore only necessary to resort to spectroscopic techniques where detailed radioisotope information is required, such as alpha emissions arising from uranium and plutonium in the bulk air sample. It is likely that this is at least partly why US EPA have not approved alpha scintillation counting for radioisotope assay in the environment. However, in circumstances where no other alpha emitting radionuclide is present, alpha scintillation counting can be an appropriate specific assay method; for instance it is approved for use by individual US States' Departments of Health for domestic and workplace radon (Rn-222) assay, as Rn-222 is likely to be the major alpha emitting element identified in such settings.

There is therefore no need to amend LLWR assay practices given that the site is already aligned with international best practice.

Issue 1 LLWR/ESC/R(10)10015

References:

LLWR Authorisation Schedule 9, Requirement 3 Study. NNL (09) 10297 Issue 1.

Environmental Monitoring Plan, DOE/RL-91-50. US DoE Richland Operations Office.

ANDRA, Bilan annuel de la surveillance du centre et de son environnement pour l'année. Paris, 2008

United States Department of Energy, Comparison of Low-Level Waste Disposal programs of US DoE and Selected International Countries. National Low-Level Waste Management Program. June 1996. Washington DC.

US EPA, Method 114- Test Methods for Measuring Radionuclide Emission from Stationary Sources. Technology Transfer Network, Emission Measurement Centre.

United States Department of Energy Handbook: *Tritium Handling and safe storage*. DOE-HDBK-1129-99, Washington DC. 1999.

Email Communication from Ann McCall (Vice President) SKB International Consultants AB 8/12/2009.

SFR Brochure, Svensk Kärnbränslehantering AB, Stockholm Sweden. Retrieved from http://www.skb.se/upload/publications/pdf/Engelsk_low_res.pdf

Ross H, Noakes J, Spalding D. Liquid Scintillation counting and Organic Scintillators. *Chapter 18 Factors affecting Alpha Particle Detection in Liquid Scintillation Spectrometry*. Oak Ridge National Laboratory and University of Georgia. 1991.

Appendix

Sampling Regime at Hanford site (Richland LLW and ERDF)

Air

Bulk air samples are collected on glass fibre filters, counted for gross alpha, gross beta, Sr-90, Pu isotopes, U isotopes and gamma. Analysis for gross alpha and gross beta occur immediately after sample collection to identify any possible uncontrolled release. Sr-90, Pu and U isotopes from biweekly samples are composited for quarterly analysis to track trends not ordinarily detectable with the gross activity measurements. Samples for tritium analysis are collected on silica gels and collected every four weeks.

Surface Water

Surface waters are collected by:

1) A remote system which collects a fixed volume of water at set intervals at each location during a given sampling period.
2) A system which continually collects waterborne radionuclides on a series of filters and a mixed-bed ion exchange column.

Due to the size of the Columbia River flowing through the site it is not practical to collect flow proportional samples.

Surface water analytes include gross alpha, gross beta, gamma scan (for Co-60, Ru-106, Sb-125, Cs-137, Eu-152, Eu-154), tritium, isotopic U, Sr-90, Tc-99, I-129, Pu-238, Pu-239, Pu-240.

Ground Water

Many ground water wells on the Hanford site are sampled for multiple objectives and requirements e.g. US Atomic Energy Act, Comprehensive Environmental Response, Compensation and Liability Act, Resource Conservation and Recovery Act etc. The site utilises the US EPA's DQO process to manage overlap between these requirements, ensuring that the needs of each sampling objective is met at an optimal schedule.

Samples generally are collected after three casing volumes of water have been purged from the well and after field parameters (pH, temperature, specific conductance, and turbidity) have stabilized (i.e., after two consecutive measurements are within 0.2 units pH, 0.2°C for temperature, 10% for specific conductance, and turbidity; <5 Nephelometric Turbidity Units (NTU). For routine groundwater samples, preservatives are added to the collection bottles before their use in the field according to subcontractor procedures. Samples to be analyzed for metals are usually filtered in the field so that results represent dissolved metals.

Sediment

Sediment samples are collected from the Columbia River bed by boat-deployed dredger, samples from shallower reaches are collected by hand operation of the dredger or hand scoops very shallow areas and ponds. Analytes include, Sr-90, U and Pu isotopes and a gamma scan

Agri-food

Milk is collected from dairies downwind of the site, on the perimeter and at a control location upwind from the site on an annual basis. Analytes include Sr-90, gamma scan, tritium, U and Pu isotopes, Am-241.

LLW Nuclear facilities in France, Centre de la Manche

The original disposal mode at Centre de la Manche was tumble-tip of LLW into unlined trenches, in a much analogous method to historical disposal methods on the LLWR site. Tumble-tip operations were halted in 1976 when routine environmental monitoring of the site revealed leaching of tritium into a nearby stream. This resulted in a change of disposal method to more engineered structures, LLW was subsequently stored on top of a monolith composed of ILW (short lived, high activity radionuclides) and backfilled with gravel. Initial capping materials used include soil and clay. As part of site closure in the early 1990s, the entire site was capped off with a bituminous membrane, soil and seeded with grass as shown in Figure 1:

Environmental Monitoring is now the main activity on the Centre de la Manche site since the last waste was accepted in 1994.

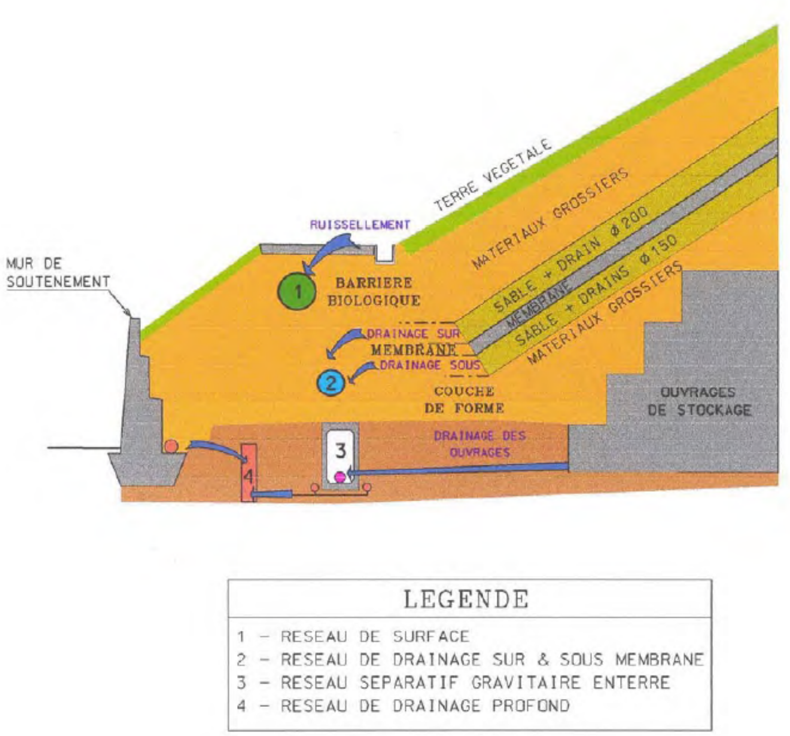

Figure 1: Schematic of capping at Centre de la Manche, leachate and surface water management.

The monitoring programme on the site involves analysis of surface waters, sediments, leachates, radioactivity in the air and vegetation on the cap. ANDRA present analytical techniques on an isotope by isotope basis.