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Determination of ^{137}Cs contamination depth distribution in building structures using geostatistical modeling of ISOCS measurements

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HIGHLIGHTS

- ^{137}Cs depth contamination was determined using the multiple photo peak method.
- Geostatistical modeling was used to determine treatment depth areas and perform risk analysis.
- Results were evaluated using laser scanning and long term gamma-ray spectroscopy.
- Waste volume reduction of about 1/3 compared to a more traditional approach.

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ABSTRACT

Decommissioning of nuclear building structures usually leads to large amounts of low level radioactive waste. Using a reliable method to determine the contamination depth is indispensable prior to the start of decontamination works and also for minimizing the radioactive waste volume and the total workload. The method described in this paper is based on geostatistical modeling of in situ gamma-ray spectroscopy measurements using the multiple photo peak method. The method has been tested on the floor of the waste gas surge tank room within the BR3 (Belgian Reactor 3) decommissioning project and has delivered adequate results.

1. Introduction

In June 2012, 435 nuclear power reactors were in operation, 62 under construction and 140 in permanent shutdown (IAEA, 2012). According to the International Atomic Energy Agency (IAEA, 2011) as of December 2010, for only five reactors the license had been terminated, which is the legal act at the end of the decommissioning. The number of nuclear facility decommissioning projects is therefore increasing. Besides the nuclear installations to be dismantled, the building structures containing the installations take part of decommissioning as well. Contamination of the building structure, usually resulting from leakages during operation, releases due to maintenance works or even during dismantling operations, may result in large volumes of predominantly low level radioactive waste. In order to minimize the radioactive waste volume, it is indispensable to determine the contamination depth prior to initiate decontamination works.

Some decommissioning projects use iterative cycles consisting of decontamination treatment steps and radiological control measurements until release levels are reached. The radiological control measurements are usually conducted with straightforward handheld equipment such as gas proportional or scintillation counters for surface contamination measurements. Despite being straightforward, this method might lead to multiple iterative cycles and is therefore not always practical and efficient. Most decommissioning projects also use sampling by core drilling and destructive analysis which is time consuming and costly. Moreover, it only provides information on small discrete spots and spatial extrapolation to the complete area of a structure might cause problems in case of an inappropriate sampling scheme. This could go from non-representativeness of the samples for the complete area or the absence of certain important small or narrow

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discrete features in the sampling scheme, to inaccurate estimation of the spatial variability and hence a misleading idea about the spatial continuity of the contamination. A second iteration of the decontamination might be required when important high contaminated spots or features are missed, while an erroneous estimate of the spatial continuity might heavily affect decontamination volumes. Therefore, the use of larger spatial supports (area/volume corresponding to a measurement location) is investigated in this paper as a possible methodology for optimizing a decommissioning project.

Determining the radionuclide depth in soil and concrete using Non Destructive Assay based on In-Situ Gamma Spectroscopy is a way to obtain prompt results and larger measurement supports. This has already been studied based on various principles (Whetstone et al., 2011):

- Using different special designed multiple collimators and/or shields (Whetstone et al., 2011; Benke and Kearfott, 2002; Benke and Kearfott, 2001; Van Riper et al., 2002),
- Using the peak to valley ratio method (Zombori et al., 1992; Tyler, 1999),
- Using the multiple photo peak method (Sowa et al., 1989; Beck et al., 1972; Karlberg, 1990) or the primary photo peak and the X-ray lines (Rybacek et al., 1992).

Each of these methods has its particular advantages and disadvantages. According to the most recent publication listed above (Whetstone et al., 2011), the first method seems to deliver the most accurate results. A drawback of this method is that one usually considers that the activity distribution in the horizontal direction is relatively homogeneous which is not always justified for example when hot spots are present. Moreover, multiple measurements on one spot are required to determine the radionuclide contamination depth. Therefore, based on experience from the remediation of the BR3 (Belgian Reactor 3) building structure at the Belgian Nuclear Research Centre (SCK•CEN, Mol, Belgium), we evaluated the third methodology using the In Situ Object Counting System (ISOCS) of Canberra (Canberra, 2012). In the case of a Pressurized Water Reactor such as the BR3, the key radionuclide to determine the contamination depth in building structures is ^{137}Cs (Boden and Cantrel, 2007).

For the assessment and risk evaluation of surface contamination of an entire floor based on a limited number of measurements, the use of geostatistics could be envisaged. Geostatistics, based on the theory of regionalized variables (Goovaerts, 1997) provides algorithms to estimate expected values, uncertainties, and risk of exceeding a given threshold, based on a limited number of discrete sampling points. The basic notion is the covariance structure of the spatial random variable with the assumption that the correlation between two random variables depends only on their lag (separation) distance. This statistical method, originally applied in the mining industry (Krige, 1951), is applied in many areas where prediction of or uncertainty on regionalized variables is of importance, in particular, in environmental pollution studies (e.g. Goovaerts et al., 2008; Goovaerts, 2010; Zhang et al., 2009; Wu et al., 2009) and hydrology (e.g. De Marsily et al., 2005; El Idrysy and Smedt, 2007; Rogiers et al., 2012, 2010).

Because radiological contamination is also expected to be correlated in space, several projects applied geostatistical methods for the evaluation of radiological contamination such as studies for sampling scheme optimization and for the spatial structure of extreme values (Jeannee et al., 2008; Desnoyers et al., 2011). These two studies focus on the surface activity, and use many small-scale measurements with uniform spatial support (0.03 m²). However, by varying the vertical placement of the ISOCS device, it is possible to perform measurements with different spatial supports at the same location which allows characterizing larger areas with a single measurement as well as smaller-scale details. Moreover, the focus in the present study is on the contamination depth which has a direct consequence on the radioactive waste volume, rather than the surface activity.

The objective of this paper is to apply a new methodology for the decontamination of ^{137}Cs contaminated building structures with a view of minimizing the radioactive waste volume based on a geostatistical interpretation of a profound pre-treatment characterization program using mainly Non Destructive Assay. The scope of the proposed methodology is oriented toward relatively high ^{137}Cs contamination levels, signifying building structures categorized as 2 and 3 (Cantrel and Boden, 2008). Building structures categorized as 2 are located at known contaminated areas where the contamination hazard mainly concerned aerosols and/or dust. In those areas only limited migration of ^{137}Cs is expected, with contamination depths limited to 5 mm. Contaminated areas where the contamination hazard concerns liquids as well are categorized as 3. In those areas contamination depths up to several hundreds of millimeters are expected. This paper describes a test case of the floor of the waste gas surge tank room within the BR3 decommissioning project (SCK•CEN), where ISOCS measurements combined with geostatistics have been applied in order to define the decontamination depth of ^{137}Cs and divide the floor into several decontamination areas. Results obtained are compared with the actual end result of the treatment and final release measurements.

2. Decontamination methodology and test case

The floor of the waste gas surge tank room has been used to evaluate the decontamination methodology proposed.

2.1. Decontamination methodology

The proposed process for the decontamination of ^{137}Cs contaminated building structures is presented in Fig. 1 and consists of following steps:

(i) *Pre-treatment characterization*: For clearly in-depth ^{137}Cs contaminated building structures, the pre-treatment characterization consists of the determination of the ^{137}Cs contamination depth by Non Destructive Assay using ISOCS.
(ii) *Geostatistical analysis*: A proper analysis of the spatial distribution of the ISOCS measurement results is carried out, accounting for the different spatial supports (area represented by the measurement) that are used.
(iii) *Probability maps and expected mean depth*: The risks are mapped using the spatial correlation characteristics from step (ii) and geostatistical simulations. Final risk maps are presented at the decontamination scale (scale on which the removal of material can be performed).
(iv) *Check with cores*: Sampling by core drilling at step (i) is used to validate the results obtained in (iii) and to check the the building structure (composition and thicknesses of different layers).
(v) *Decontamination plan*: Based on the results from (iii), previous experiences in the decontamination of rooms and input from the decontamination expert, the decommissioning plan is agreed upon.
(vi) *Post-treatment characterization*: After the execution of the decontamination plan radiological control measurements are applied to demonstrate that the contamination has been completely removed.

In order to evaluate the process proposed, potential remaining traces of ^{137}Cs contamination are determined using long term