



Mapping the spatial distribution and activity of ^{226}Ra at legacy sites through Machine Learning interpretation of gamma-ray spectrometry data

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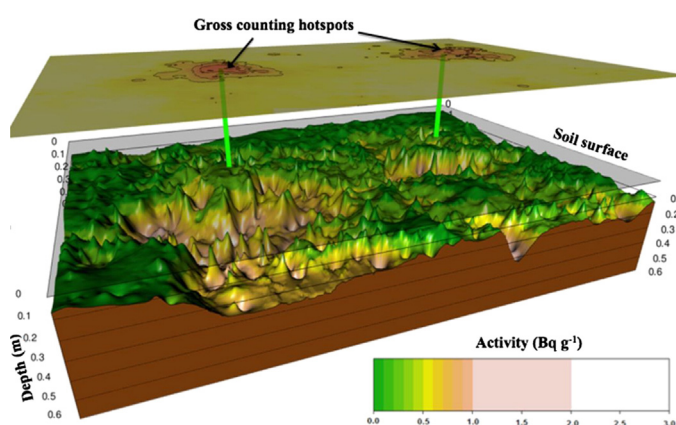
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HIGHLIGHTS

- Land contaminated with radium is hazardous to human health.
- Contamination characterised with gamma-ray spectrometry.
- Machine Learning to derive activity and depth estimated from spectral shape.
- Lanthanum bromide and Neural Network provided optimum performance.
- The developed approach demonstrates a powerful assaying tool.

GRAPHICAL ABSTRACT



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ABSTRACT

Radium (^{226}Ra) contamination derived from military, industrial, and pharmaceutical products can be found at a number of historical sites across the world posing a risk to human health. The analysis of spectral data derived using gamma-ray spectrometry can offer a powerful tool to rapidly estimate and map the activity, depth, and lateral distribution of ^{226}Ra contamination covering an extensive area. Subsequently, reliable risk assessments can be developed for individual sites in a fraction of the timeframe compared to traditional labour-intensive sampling techniques: for example soil coring. However, local heterogeneity of the natural background, statistical counting uncertainty, and non-linear source response are confounding problems associated with gamma-ray spectral analysis. This is particularly challenging, when attempting to deal with enhanced concentrations of a naturally occurring radionuclide such as ^{226}Ra . As a result, conventional surveys tend to attribute the highest activities to the largest total signal received by a detector (Gross counts): an assumption that tends to neglect higher activities at depth. To overcome these limitations, a methodology was developed making use of Monte Carlo simulations, Principal Component Analysis and Machine Learning based algorithms to derive depth and activity estimates for ^{226}Ra contamination. The approach was applied on spectra taken using two gamma-ray detectors (Lanthanum

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Bromide and Sodium Iodide), with the aim of identifying an optimised combination of detector and spectral processing routine. It was confirmed that, through a combination of Neural Networks and Lanthanum Bromide, the most accurate depth and activity estimates could be found. The advantage of the method was demonstrated by mapping depth and activity estimates at a case study site in Scotland. There the method identified significantly higher activity ($<3 \text{ Bq g}^{-1}$) occurring at depth ($>0.4 \text{ m}$), that conventional gross counting algorithms failed to identify. It was concluded that the method could easily be employed to identify areas of high activity potentially occurring at depth, prior to intrusive investigation using conventional sampling techniques.

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1. Introduction

1.1. Regulation of radium contaminated land

Radium (^{226}Ra) was used extensively during the 20th century in military, industrial, and pharmaceutical products (Tyler et al., 2013). ^{226}Ra has a half-life of over 1600 years, and is the parent of an additional 8 radioactive elements that together produce a complex array of alpha, beta and gamma emissions (Pratt, 1993). Follow-up studies into the health implications of radium dial workers in the US typify the risks associated with long-term radium exposure (Stebbins, 2001). Within the UK, radium was used extensively as a composite in luminescent paint principally by the military during wartime periods. Prior to the Radioactive Substances Act 1960 (HMSO, 1996) vast inventories of waste were routinely burned and buried with little record presenting complex remediation challenges (Wilson et al., 2013).

A report published in 2012 by the UK government, conservatively estimated there to be 150–250 Radium Contaminated Legacy Sites (RCLS) linked to Ministry of Defence activities within the UK (DECC, 2012). Moreover, it was recognised that there could be as many as 1000 contaminated sites in the UK alone. Similar RCLS can be found across other parts of Europe and North America (IAEA, 1998).

a) UK legislation has now provided the Scottish Environmental Protection Agency (SEPA) with a framework to classify radioactively contaminated land and set guidelines to describe the amount of contamination that may give rise to significant harm to humans (The Radioactive Substances Act 1993 Amendment (Scotland) Regulations, 2011). Homogeneous contamination at RCLS in Scotland is controlled by the following criterion: An effective dose must not exceed 3 mSv per annum (Statutory Guidance to support the Radioactive Contaminated Land (Scotland) Regulations, 2008).

Homogeneous contamination can be defined as contamination that is dissociated or not in particulate form, which can vary significantly in activity over small spatial scales.

Surface flux measurements using dosimetry can be used to rapidly assess the effective dose at a site (IAEA, 1998). Yet, this measurement is somewhat limited in thoroughly assessing the activity and burial depth of homogeneous contamination, which tend to change significantly across a RCLS (Varley et al., 2015b). These factors are critical pieces of information for long-term remediation purposes, particularly at sites that are exposed to the public, where changes in site use or erosion events may occur increasing the risk of contact (Dale et al., 2013). Currently, not one Scottish legislation alone can be used to specifically outline an activity limit that must not be exceeded for suspected homogeneous contamination at a RCLS. Therefore, in this study we accept that ^{226}Ra should be treated under the exemption for Naturally Occurring Radioactive Materials declaring:

b) An activity must not exceed 10 Bq g^{-1} (The Radioactive Substances Act 1993 Amendment (Scotland) Regulations, 2011).

The discrete nature of the items that were initially disposed of can also lead to the formation of *hot particles*. At one RCLS at Dalgety Bay,

Fife, Scotland, a diverse range of *hot particles* and historic artefacts ($<70 \text{ MBq}$) has been found (Dale et al., 2013). If such items were to be picked up by a member of the public this may result in a significant committed dose (Tyler et al., 2013). A method for the real-time identification of ^{226}Ra containing *hot particles* at RCLS has been outlined in our previous work (Varley et al., 2015a).

In light of the uncertainties behind site formation and the lack of disposal records, once a RCLS has been identified the contamination should be systematically characterised to ascertain the risk it poses to long-term human health; thus going beyond limited surface dosimetry estimates (IAEA, 1998). In this paper we propose a method that can be rapidly and inexpensively deployed at a RCLS to provide accurate estimates of near-surface *homogenous* ^{226}Ra contamination depth, activity, and spatial distribution.

1.2. Environmental gamma-ray spectrometry

Handheld gamma-ray spectrometry (HGS) or mobile gamma-ray spectrometry, generally performed using inorganic scintillators, is often the cheapest and most robust technique of characterising RCLS (IAEA, 1998, 2003; Knoll, 2010; Dale et al., 2013; Read et al., 2013; Haddad et al., 2014). Using this method the spatial extent and activity of gamma-emitting radionuclides can be estimated using remote surface measurements without the need for time-consuming invasive methods (Tyler, 2008). Individual energy spectra produced during a survey are representative of the localised radiation field a detector has passed through (Beck et al., 1972).

For mapping purposes each spectrum can be post-processed using an algorithm to unfold spectral information (Kock et al., 2012). The first objective of this unfolding process is to identify whether there are characteristic signals from radium contamination (source), which typically differ in shape from background spectra. For example, notice characteristic peaks at 351, 609, 1120, 1764 and 2244 are generated by a radium source (Fig. 1A). However, source-background separation is often complicated by spatial fluctuations in background (^{40}K , and the ^{238}U and ^{232}Th series) and comparatively benign (^{137}C in the case of RCLS) radioelements, alongside variations in soil density and composition, which together introduce nuisance spectral changes (Runkle, 2006). Fagan et al. (2012) presents an informative review of the challenges associated with, and the techniques employed in, spectral classification.

Once a contaminated spectrum has been identified, the second aim is to identify spectral elements that are symptomatic of source burial depth and activity. Notice non-linear changes take place across the spectrum as the burial depth of a source varies: for example lower energy peaks are attenuated more relative to higher energy peaks with increasing burial depth (Fig. 1A).

The multiple photopeak method aims to capture this occurrence by calculating the area under two background-subtracted full energy peaks with the purpose of comparing the observed ratio to that of a calibrated one to estimate source burial depth and activity (Miller et al., 1994; Thummerer and Jacob, 1998; Haddad et al., 2014). Another method that utilises more spectral information is termed Full Spectral Analysis (FSA). FSA compares the spectral similarity of an obtained spectrum to a calibration library by a weighted least-squares fitting procedure

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