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Remediating radium contaminated legacy sites: Advances made through machine learning in routine monitoring of "hot" particles

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HIGHLIGHTS

- Land contaminated with radium is hazardous to human health.
- Routine monitoring permits identification and removal of radioactive *hot particles*.
- Current alarm algorithms do not provide reliable *hot particle* detection.
- Spectral processing using Machine Learning significantly improves detection.

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ABSTRACT

The extensive use of radium during the 20th century for industrial, military and pharmaceutical purposes has led to a large number of contaminated legacy sites across Europe and North America. Sites that pose a high risk to the general public can present expensive and long-term remediation projects. Often the most pragmatic remediation approach is through routine monitoring operating gamma-ray detectors to identify, in real-time, the signal from the most hazardous heterogeneous contamination (*hot particles*); thus facilitating their removal and safe disposal. However, current detection systems do not fully utilise all spectral information resulting in low detection rates and ultimately an increased risk to the human health. The aim of this study was to establish an optimised detector-algorithm combination. To achieve this, field data was collected using two handheld detectors (sodium iodide and lanthanum bromide) and a number of Monte Carlo simulated *hot particles* were randomly injected into the field data. This allowed for the detection rate of conventional deterministic (gross counts) and machine learning (neural networks and support vector machines) algorithms to be assessed. The results demonstrated that a Neural Network operated on a sodium iodide detector provided the best detection capability. Compared to deterministic approaches, this optimised detection system could detect a *hot particle* on average 10 cm deeper into the soil column or with half of the activity at the same depth. It was also found that noise presented by internal contamination restricted lanthanum bromide for this application.

1. Introduction

1.1. Radium contamination

Radium (^{226}Ra) was used extensively during the 20th century predominantly in the form of luminescent paint. Waste generated from military, industrial and pharmaceutical products was regularly buried with little record of its location and inventory (Harvie, 1999). With a half-life of 1600 years, ^{226}Ra contamination is a multigenerational issue. In the UK, a recent government report conservatively estimated there to be 150 to 250 contaminated legacy sites, whilst acknowledging there could be as many as a 1000 (DECC, 2012). Similar extents of ^{226}Ra contamination have been found across Europe and North America (Harvie, 1999). Ultimately, the risk of human exposure at these sites is dependent on a number of potential pathways and the form of contamination and not exclusively on external dose (Dale et al., 2008).

One such pathway, that has the potential to cause significant radiological harm, is ingestion of small highly radioactive items often referred to as *hot particles* (Baker and Toque, 2005). One study explored the committed dose that could be received by a member of the public through simulated stomach acid digestions of a range of radium *hot particles*

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found in Scotland (Tyler et al., 2013). It was concluded that ingestion of a *hot particle* with an activity higher than 20 kBq could result in a committed dose to an infant exceeding the 100 mSv threshold deemed to cause significant radiological harm (ICRP, 2007). At particular sites in Scotland, it has been recognised by the Scottish Environment Protection Agency (SEPA) that there is the possibility of a member of the public coming into contact with such a *hot particle*. To safeguard against this at Dalgety Bay, Fife, Scotland, routine monitoring is undertaken to detect and retrieve any significant radioactive items (Dale et al., 2013). To confirm monitoring is undertaken with sufficient accuracy, SEPA have outlined the following criterion:

- A 20 kBq *hot particle* at a burial depth of 0.1 m must be detected 95% of the time.

This paper aims to develop an optimised detection system that provides better detection capability than systems currently available. This will allow more effective identification of *hot particles* at radium legacy sites, ultimately reducing the risk posed to the general public in both the short and long terms.

1.2. Challenges of "hot" particle detection

The most effective method of detecting *hot particles* in real-time is through a series of mobile measurements using either handheld or vehicle mounted gamma-ray sensors (Tyler, 2008). At many sites where vehicular access is limited handheld detectors can be the only option. Handheld detectors produce gamma-ray spectra, the shape and magnitude of which will provide information of the localised radiation field that the detector has passed through during acquisition. To ensure that an area is adequately covered by a survey in a reasonable time frame, typically a spectrum is acquired every second and a walking speed of 0.5 m s^{-1} is maintained. This maximises the spatial density of measurements and ultimately the probability of detecting any *hot particles*. To initiate the immediate identification of a *hot particle* real-time analysis of the spectral time series is critical (Kock et al., 2012).

The spectral response or signal quality of a detector will largely be governed by the composition of the detector's active volume. Lightweight scintillators tend to be used for handheld detectors, of these sodium iodide (NaI:Tl) is the standard as it is relatively cheap and robust (Knoll, 2010). Nevertheless alternatives are available. One such detector, lanthanum bromide (LaBr:Ce), has recently received much attention (Guss et al., 2010). It has better energy resolution (~2.5% at 662 keV) than the NaI:Tl (~7% at 662 keV) (Fig. 1), greater photon efficiencies and better temperature stability. However, a relatively large intrinsic background signal attributed to internal isotopes (^{138}La and ^{227}Ac) can be found distributed throughout the spectrum (Iltis et al., 2006). Notice considerable contamination contributions at 1468 keV (gamma and x-ray summation peak), 786–1100 keV (beta continuum) and over 1700 keV (alpha) (Menge et al., 2007). This raises concerns about its potential in low source signal situations as these are frequently encountered in the environment.

The second element of a detection system is the algorithm used to process the detector's signal in real-time to determine whether there are signal contributions from a *hot particle* (Fig. 1). If the algorithm is tripped, an alarm is sounded, allowing the operator to locate and retrieve any potential radioactive items (Jarman et al., 2008). Yet processing environmental gamma-ray spectra is not straightforward.

First of all the vast majority of spectral changes are benign and can occur over the scale of a few metres (Fagan et al., 2012). This spatial variation is brought about by changes in the natural radioelements (^{40} K, and the ^{238}U and ^{232}Th series) contained within the local geology. Changes can also occur temporally due to variations in the density and chemical composition of the geological matrix and radon exhalation (IAEA, 2003). Temporal fluctuations can be very challenging to account for particularly as they can occur on varying time scales, for example deviations in density over a tidal cycle and radon exhalation caused by atmospheric pressure changes (Ball et al., 1991; De Groot et al., 2009). Furthermore, weak source signals can appear very similar to background signals given that ^{226}Ra forms part of the ^{238}U series. Consequently, in an attempt to isolate source signal from benign signal, it is often appropriate to use the most recent observations in the time series as estimates of background (Ely et al., 2004).

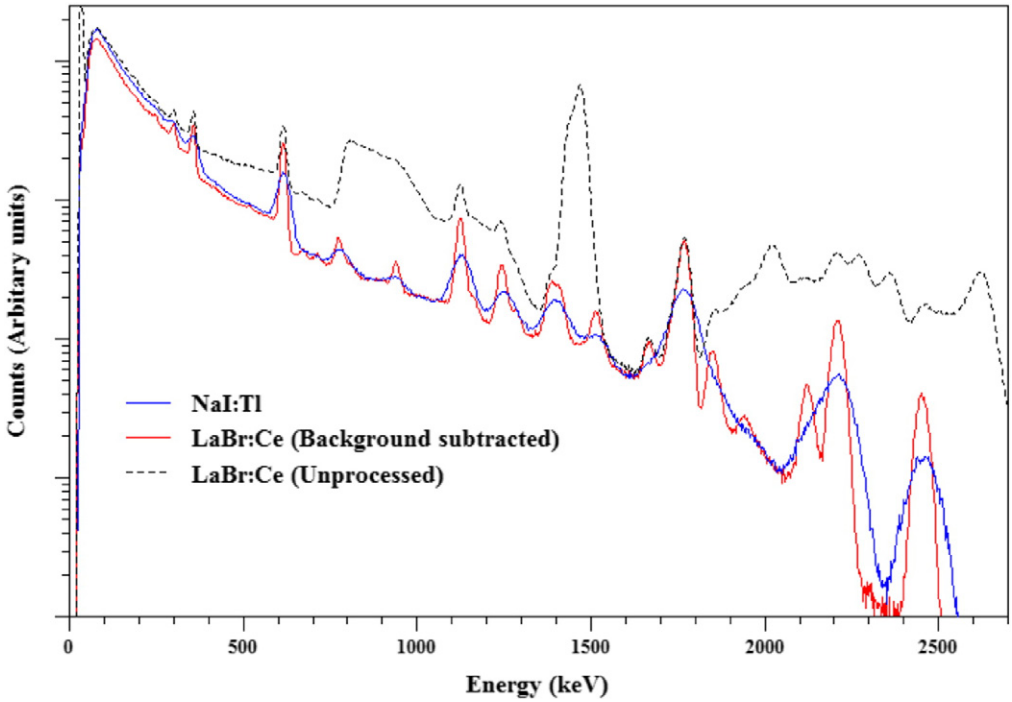

Fig. 1. Detector responses to a ^{226}Ra *hot particle*: NaI:Tl (blue), LaBr:Ce background subtracted (red) and LaBr:Ce unprocessed (broken black).