Is the Holy Grail Plastic? Radiation Identification from Plastic Scintillators

Laura J. C. Butchins, Jonathan M. Gosling, Matthew R. W. Hogbin, Daniel C. Jones, Richard J. Lacey and James G. Stearn

Abstract— Radiation monitoring at national borders is becoming more common place and so efforts to reduce the burden of false positives (or nuisance alarms) at checkpoint operations without reducing the ability to detect potential threats is now a key objective for many nations. Plastic scintillators are relatively cheap to deploy in large volumes, are rugged and sensitive to a range of gamma energies. However, at present, plastic detectors that are available for deployment for security applications provide only minimal or no energy discrimination as the response to gamma radiation is primarily via Compton scattering so photopeaks are usually not visible. Thousands of shipping containers containing Naturally Occurring Radioactive Materials (NORM) made from ceramics, stoneware and other natural products are transported worldwide on a daily basis. Some of these NORM loads are sufficiently radioactive to trigger alarms from plastic scintillator detectors which have limited ability to also identify the radionuclides present thus necessitating secondary inspection which increases the operational overhead. Previous studies have been carried out to ascertain if radionuclide discrimination using plastic scintillators is possible with a variety of approaches including deconvolution and computer learning. In this paper, a two stage algorithm is described. An example implementation of the algorithm is presented, applied to operational data, and has been installed in real time operation on a polyvinyltoluene (PVT) detector.

The approach requires the collection of a large library of spectra using examples of the detectors to be deployed. In this study, data from both actual freight loads passing through a port and predefined freight containing various radionuclides were collected. To ascertain the 'ground truth' content of the freight loads, a sodium iodide detector was deployed along side the PVT detector. A support vector machine (SVM) was used to classify the transformed data into subcategories. This is done by forming a curved surface that separates the principle components when they are plotted in n-dimensions.

The library represents freight loads that may contain industrial, medical, nuclear, and NORM radionuclides. The radionuclides in the predefined freight were placed in various orientations and in various amounts of shielding to mimic many different scenarios. Spectra from mixed sources and sources in the presence of NORMs were also taken in typical quantities seen in freight. Steel and aluminium shielding was used in 1mm incremental steps to achieve scattering and shielding effects that may be seen in typical freight loads.

Manuscript received May 9, 2009. This work was supported by the Home Office, Scientific Development Branch, U.K.

L. J. C. Butchins, and all co-authors are with the Home Office Scientific Development Branch, UK. (phone: +44 (0)1727 816 500; e-mail: laura.butchins@homeoffice.gsi.gov.uk).

Preliminary results on an initial subset of data containing industrial and NORM sources show the number of misclassifications to be less than 1% of the total test data. The training set of data was chosen at random each time the analysis was undertaken, and comprised of approximately 50% of the total data set; the remaining 50% made up the test data. Good initial results were obtained even for low energy radionuclides such as ²⁴¹Am. Where discrimination is not possible, and principle components overlap, this region or "cloud" of the n-dimensional plot can be put aside. Those spectra that fall in the "cloud" can be regarded as suspect and in these cases, some secondary screening will still be necessary. It is predicted that the algorithm will enable recognition of NORM loads by plastic scintillator detectors to be increased by as much as 80% and for high energy industrial radionuclides to be identified with accuracy approaching 100%. A demonstration system has been produced which provides classification in real time.

Index Terms—radiation detection, NORM, principal components analysis, support vector machine.

I. INTRODUCTION

CREENING for radiation at points of entry and exit at international borders has become important in the past ten years. Systems for screening freight, vehicles, rail or pedestrian traffic are all important and depend upon rugged and reliable radiation detection technology. The detectors must also be sensitive to detect any threat material that may be smuggled in the load, as well as cost effective, whilst causing minimal impact on operations. Currently, a two tier system is often used when screening large quantities of goods. Typically, the first stage relies upon large volume plastic scintillation detectors for gamma detection, which provide the sensitivity to allow a high throughput of traffic. For those loads that give a positive indication, the vehicle driver or pedestrian is diverted for secondary screening. The gamma detection method for secondary screening has a spectroscopic response and provides specificity to provide the screeners with identification of the radionuclides. This can then be reconciled with the manifest or any supporting documentation. Particularly in freight traffic, there are a large proportion of loads that require secondary screening because they contain Naturally Occurring Radiological Materials (NORM). NORM is of primordial origin, with the most common radionuclides being 40K, that tends to be found in fertilizer, tobacco, and foodstuffs, ²³⁸U, ²³²Th and their daughters, which tend to be found in ceramics

and stoneware. Large numbers of loads that require secondary screening can put a strain on operations and can cause delays to commerce, and therefore, better methods of screening at the primary stage would be advantageous.

This problem could be overcome by replacing PVT primary detectors with detectors exhibiting better spectroscopic resolution, therefore providing identification, but significantly adding to the cost of the detector hardware to retain the required sensitivity. Another approach is to refine the plastic scintillator material; by loading the material, for example with nanoparticles [1], or to change the geometry of the plastic, or by enhancing the spectral output by deconvolution methods [2]. The approach presented here relies upon statistical methods to provide better discrimination at primary screening by making use of all the information in the spectra produced by the large plastic scintillation detectors. Multivariate analysis techniques have been employed for this purpose. These techniques have previously been used in a variety of applications over many years since they were first derived [3] but were only applied to spectroscopy and spectrometry in more recent years (often referred to as chemometrics) to make use of subtle variations and similarities in spectra that may not be obvious to the human eye or simple peak picking techniques. A recent study carried out by Runkle et al. [4] applied principal components analysis (PCA) with the use of the Mahalonobis distance metric as the classifier for spectra from both sodium iodide and PVT scintillator detectors showing that NaI performs better than PVT with this combination of statistical techniques. It was also shown that PCA could usefully be employed with PVT for anomaly detection when compared to using gross counts.

The approach taken here is to use PCA with a support vector machine (SVM) [5], a powerful classifier that requires a large data set that accurately represents the entire population. SVM classifies the principle components by building hypersurfaces between each set to establish "population clouds".

II. DATA COLLECTION

A. Operational Data Collection

Data were collected for a period of several weeks from loads that passed through a secondary inspection system providing 645 "real" NORM spectra.

B. Industrial Radionuclide Data Collection

A set of 537 spectra were collected from industrial and other radioactive sources. A number of experimental variables were altered including; the activity, the shielding thickness and the shielding material, which was oriented at 360° around the source. (The shielding not only changes the activity but also increases the Compton scattering contribution to the spectra and skews the spectra at low energies). Another variable was the addition of NORM material alongside the radionuclides. The variables were changed incrementally, for example, each time the shielding was changed, 2mm were added before the

next spectrum was collected. With this systematic approach, a reasonably comprehensive spectrum set was collected.

C. Data Processing

The full set of data was normalized, split into two roughly equal parts, a training and a testing set, and reduced using PCA. Tests showed that using different split ratios had little effect on the results. Figure 1 is an example plot of the first two principal components (PCs) for the full data set. A support vector machine (SVM) classifier was found to produce good results for this data set. Classification to separate all radionuclides was assessed, as well as classification of NORM versus all other radionuclides. In all cases, the first four PCs were used. Different kernel types were tested and the best performance was found using the radial basis function. The cost parameter was optimized and set to 30, and the gamma parameter similarly optimized and set to 0.25. In so doing, the decision boundaries around the training data points were optimized for the sample set. Figure 2 demonstrates the radial basis function in two dimensions overlaid by a plot of principal for a sample set.

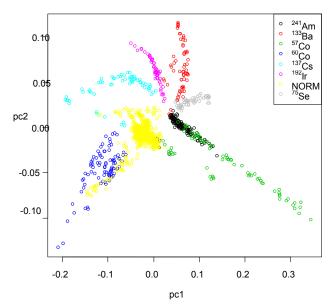


Fig. 1. Plot of the first two principle components (PCs) for 7 radionuclides and NORMs.

D. Classification of NORM from all Other Radionuclides

In an operational situation, it may be beneficial to separate the NORMs from all other radionuclides, without the requirement to identify those other radionuclides. Figure 3 provides an illustration of the separation of NORM from other radionuclides by plotting the first two PCs. On applying the SVM to the data, the correct and incorrect identifications can be determined, which correspond to o and □ accordingly. On

classification of NORM from all other radionuclides, 99% were classified correctly (with a standard error <1%, n=12).

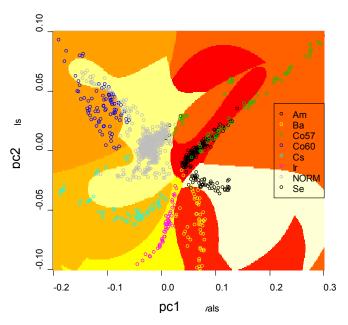


Fig. 2. Image demonstrating the hyperplanes formed by the radial basis function. The principal components for a sample data set are overlaid to demonstrate how the boundaries are formed.

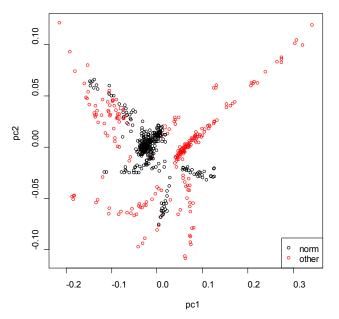


Fig. 3. Plot of the first two principle components (PCs) for the test set of data from spectra of 7 radionuclides (red) and NORM (black); o = correct classification, $\Box = incorrect$ classification.

In general, for all security systems, the balance between threat detection (TD) and false alarm rate (FAR) is important and must be finely tuned in each case. A Receiver Operating Characteristic (ROC) curve, which is a plot of the probability of detection against the probability of a false alarm is able to demonstrate the trade off in detection for an increase in the false alarm rate and is plotted for this data set in Figure 4. To isolate the area of interest in the ROC curve, a Detection Error

Trade-off (DET) curve is plotted. This is a plot of the probability of missed detection versus the probability of a false alarm (log₁₀ scale on both axes). The critical area of the ROC curve for a high performing classifier is the top left hand corner so to zoom in, the data from this area are plotted on a DET curve in Figure 5. Although the resulting line is not smooth due to the limited number of samples, a trend can be seen showing the trade-off between detection and false alarm rate. For example, these data shows that, for a probability of missed detection of 1 in 10 000, the probability of a false alarm is 0.27 for this pre-selected, higher than background activity population.

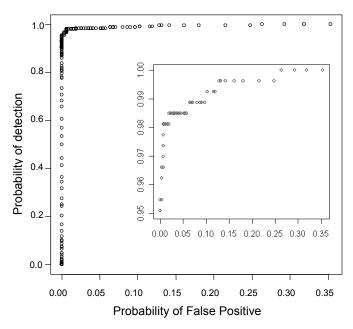


Fig. 4. Receiver Operating Characteristic (ROC) curve and zoom-in (inset) for detection of non-NORMs.

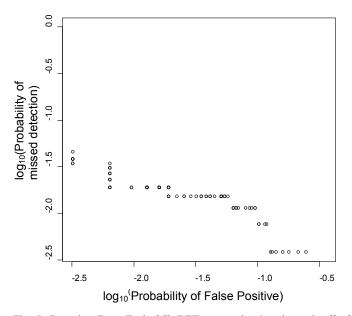


Fig. 5. Detection Error Trade-Off (DET) curve showing the trade-off of probability of detection with probability of false alarms for non-NORMs.

Four PCs were chosen in this study which attributed to only 14% of the total variation in spectra, however, subsequent PCs added fractions of a percent of variability so were not included. With a more rigorous data set, it may be necessary to use more PCs.

A drawback of this technique is that the data set is specific to the detector and set up, and to some extent the radiation background in which the data were taken. To be extended to another system, a mapping process would need to be carried out in order to apply the current trained model. Ideally, and more simply, a new training data set could be collected. Likewise, if the system was modified, for example, shielding added to the exterior of the detector, the training data set would also need to be retaken.

One way to overcome the variation in natural background from site to site, and from detector to detector, is to set up a period of data collection when the detector is first installed in each location (whilst doing routine screening with gross counts) so that each portal had its own unique training set that it could match future spectra to.

III. FINAL REMARK

This study demonstrates that if multivariate techniques are to be used for discrimination of radionuclides with plastic scintillation, it is of vital importance to ensure a complete data set is obtained and used for "live" data matching. This data set must be made up from all envisaged scenarios that could be encountered as far as is reasonable. Though a time consuming task, the method may be worth consideration where significant savings in operational costs can be made.

REFERENCES

- [1] E. A. Mckigney, R. E. Del Sesto, L. G. Jacobson, P. A. Santi, R. E. Muenchausen, K. C. Ott, T. M. McCleskey, B. L. Bennett, J. F. Smith and D. W Cooke, "Nanocomposite scintillators for radiation detection and nuclear spectroscopy" *Nucl. Inst. Meth A.579* vol. 579 pp.15-18, Apr. 2007.
- [2] D. Ramsden, M. Dallimore, "Gamma ray detectors", U.S. Patent 11 597 535, May 13, 2005.
- [3] K. Pearson, "On lines and planes of closest fit to systems of points in space" *Phil. Mag* vol. 2 pp. 559-575 1901.
- [4] R. C. Runkle, M. F. Tardiff, K. K. Anderson, D. K. Carlson and L. E. Smith, *IEEE Trans*. Nuclear Science, vol. 53, pp. 1418 – 1423, 2006.
- [5] V. Vapnik, "The Nature of Statistical Learning Theory". Springer N. Y. ISBN 0-387-94559-8, 1995.