A comparison of emerging gamma detector technologies for airborne radiation monitoring

S J Bell¹, P Aitken-Smith¹, S Beeke¹⁺², S M Collins¹, P H Regan¹⁺² and R Shearman¹⁺²

¹AIR Division, National Physical Laboratory, Hampton Road, Teddington, TW11 0LW, UK
²Department of Physics, University of Surrey, Guildford, GU2 7XH, UK

Email: steven.bell@npl.co.uk

Abstract. This paper presents a comparison of new and emerging gamma detector technologies that have the potential to improve in-situ dose and radioactivity-in-air measurements for national monitoring networks. Five detectors were chosen for investigation; LaBr₃(Ce), CeBr₃, SiPM-CsI(Tl), Cd(Zn)Te and electromechanically-cooled HPGe. These detectors represent the full range of the price-performance matrix. Comparisons have been made of energy resolution, detection efficiency and minimum detectable activity by exposing each detector to a mixed radionuclide source drop-deposited across a filter. Other factors, such as internal radioactivity, linearity, size and cost have also been considered.

1. Introduction
Article 35 of the EURATOM treaty mandates continuous environmental monitoring of radioactivity-in-air [1]. The Chernobyl accident led to increased verification of treaty compliance and wide investment in radiation monitoring networks across Europe. These networks use gamma detectors to measure gamma dose-in-air and gamma emitting airborne particulates collected on filters. In the event of a radiological incident, the measurements made by these networks must be rapidly collected, analysed and assessed. The results are then forwarded to national governments where decisions with far reaching consequences may have to be made. For this reason it is critical that the gamma measurements are as accurate and complete as possible.

The majority of in-field gamma detectors currently used represent tried and tested but dated technology, such as Geiger Müller counters and NaI(Tl) spectrometers. The disaster at Fukushima has intensified research and investment in environmental monitoring and many European authorities are looking to upgrade their gamma radiation monitoring networks. New portable radioactivity-in-air monitoring systems developed by IJS (Slovenia) [2] and CIEMAT (Spain) [3] are examples of this new technology.

Presented is a comparison of new and emerging gamma detector technologies that have the potential to improve in-field dose and radioactivity-in-air measurements for national monitoring networks. The detectors investigated include LaBr₃(Ce), CeBr₃, SiPM-CsI(Tl), Cd(Zn)Te and electromechanically-cooled HPGe. These detectors cover the full price-performance matrix. The detectors have been compared in terms of energy resolution, detection efficiency, linearity, internal radioactivity and minimum detectable activity (MDA). These properties are of vital importance when choosing a suitable detector for in-situ gamma spectrometry of filters.
This work is part of the Euramet EMRP MetroERM project; a project to harmonise the measurement and analysis of airborne radiation following a radiological event [4].

2. Method

The experimental arrangement was chosen to mimic the deployment of a gamma spectrometer in-situ with a high-volume air sampler. It was envisaged that the air sampler would cease pumping after a given time and then the gamma measurement would commence. To achieve this, a spiked filter was prepared and placed 3 cm from the centre of the detector front window.

Measurements of the filter were made within a Cu/Cd lined Pb coffin, and taken over 24 hr. Full energy peak centroids and net areas were extracted from the spectra in order to perform energy and efficiency calibrations. The resolution, efficiency and linearity were assessed. Background measurements were completed over 24 hr to determine MDA values, according to the Currie method [5]. The calculated MDA values represented the “best possible scenario” as Cu/Cd/Pb shielding is unlikely to be available for deployable air monitors.

Details of the five gamma spectrometers investigated are given in section 2.1. Details of the filters prepared for the experiment are given in section 2.2.

2.1. Detectors

The five gamma spectrometers selected for comparison are listed in Table 1. The HPGe and Cd(Zn)Te detectors were self-contained units with integrated high voltage (HV), pre-amplifier, shaping amplifier, analogue-to-digital convertor (ADC) and multi-channel analyser (MCA). The CsI(Tl) detector was coupled to a silicon photomultiplier (SiPM) and was also mounted in a self-contained unit with integrated readout electronics. The amplifier gain and shaping time settings for the HPGe, Cd(Zn)Te and CsI(Tl) detectors were optimised by the manufacturers and not adjusted. The LaBr₃(Ce) [6] and CeBr₃ detectors were coupled to Hamamatsu R9779 PMTs and were read out through an Ortec 672 shaping amplifier and an Ortec 926 MCA. A discreet pre-amplifier was not used. The shaping time and gain were optimised to improve energy resolution. Data acquisition was controlled with MAESTRO or KSPECT depending on the detector being tested. Acquired spectra were exported to Excel and processed with a custom script to extract peak centroids and net counts to ensure continuity of spectral analysis.

| Detector | Size (cm) | Volume (cm³) | Notes |
|----------|-----------|--------------|-------|
| HPGe     | 8.5 x 3.0 | 170          | Electro-mechanically cooled Ortec Detective200 with p-type coaxial crystal |
| CeBr₃    | 3.8 x 5.1 | 58           | Saint Gobain crystals coupled to Hamamatsu R9779 PMTs [6] |
| LaBr₃(Ce)| 3.8 x 5.1 | 58           | USB powered Kromek GR1 with co-planar grid electrodes |
| Cd(Zn)Te | 1.0 x 1.0 x 1.0 | 1 | USB powered Kromek Sigma50 with SiPM |
| CsI(Tl)  | 2.5 x 2.5 x 5.1 | 33 | |

2.2. Filters

The 20.5 × 10.5 cm² filter was prepared by drop-deposition of a mixed radionuclide solution. The solution was deposited evenly across a 1 × 1 cm² grid. The 200 point grid was divided into eight subsections and the dispensing bottle weighed after completion of each subsection to ensure uniformity. The maximum deviation of mass deposited to each subsection was no more than ± 0.7% from the average across the eight subsections. A total of just under 3 g of solution was deposited, providing a total activity of 29.11 ± 0.34 kBq. A photo of the filter is shown in figure 1. The activity concentration of the filter was chosen to mimic the activity collected on a filter being pumped at 1,000
m³/hr for 48 hr in an atmosphere of 0.03 Bq/m³ of ¹³⁷Cs; the minimum reporting level according to the EURATOM treaty [7]. The radionuclide content of the filter is given in table 2.

**Figure 1.** Photograph of 20.5 × 10.5 cm² “half-RASA” filter with 1 x 1 cm² grid. The mixed radionuclide solution was drop-deposited evenly across the 200 grid points.

| Radionuclide | Characteristic Gamma-ray Decay Energy (keV) | Activity (kBq) |
|--------------|-------------------------------------------|----------------|
| ²⁴¹Am        | 59.5                                      | 1.968 ± 0.028  |
| ¹⁰⁹Cd        | 88.0                                      | 8.12 ± 0.31    |
| ⁵⁷Co         | 122.1                                     | 0.4024 ± 0.010 |
| ¹³⁷Ce        | 165.9                                     | 0.3308 ± 0.0067|
| ⁵¹Cr         | 320.1                                     | 6.04 ± 0.11    |
| ¹¹¹Sn        | 391.7                                     | 1.191 ± 0.021  |
| ⁸⁵Sr         | 514.0                                     | 1.113 ± 0.020  |
| ¹³⁷Cs        | 661.7                                     | 1.472 ± 0.026  |
| ⁵⁴Mn         | 834.8                                     | 1.452 ± 0.019  |
| ⁸⁸Y          | 898.0                                     | 2.368 ± 0.031  |
| ⁶⁵Zn         | 1115.5                                    | 2.990 ± 0.053  |
| ⁶⁰Co         | 1332.5                                    | 1.671 ± 0.020  |

**Table 2.** Details of the mixed radionuclide source drop-deposited on the filter. Uncertainties quoted with $k = 2$ coverage.

### 3. Results

**3.1. Energy spectra, resolution and efficiency**

Early warning air monitors should provide rapid and accurate information in the event of an incident. Fine energy resolution is vital for accurate radionuclide identification, which can assist in determining the source of radiation. The efficiency of a detector largely determines the speed with which radionuclide identification can be performed.

Significant differences in terms of energy resolution and detection efficiency were observed across the five detectors. This is illustrated in the comparison of energy spectra in figure 2. As expected, the HPGe detector produced the best FWHM energy resolution; 0.25% at 662 keV, followed by the Cd(Zn)Te detector; 1.50% at 662 keV. The energy resolution of the LaBr₃(Ce) detector was 3.78%; marginally better than that of the CeBr₃, which was 5.0%. The CsI(Tl) detector produced the poorest
resolution, for example, failing to separate the 1115.5 keV $^{65}$Zn peak from the 1173.2 keV $^{60}$Co peak. A full resolution comparison is presented in figure 3.

Benefitting from a large volume and surface area, the HPGe detector produced the best detection efficiency. However the higher effective atomic numbers of the other detector materials went some way to compensate. This is demonstrated in table 3 where the efficiency and volume of each detector relative to that of HPGe is compared. For in-situ filter measurements, a compact detector is a necessity to avoid disruption of the air flow.

![Figure 2. Comparison of energy spectra measured by detectors exposed to 29 kBq mixed radionuclide filter source for 24 hr.](image1)

![Figure 3. Comparison of detector energy resolution from 60 keV to 1.8 MeV. Each dataset has been fitted with a power function.](image2)

**Table 3.** Efficiency and relative volume of each detector. Uncertainties quoted with $k = 2$ coverage.

| Detector     | Absolute efficiency at 662 keV | Efficiency relative to HPGe | Volume relative to HPGe |
|--------------|--------------------------------|-----------------------------|-------------------------|
| HPGe         | 1.161(50)%                     | [n/a]                       | [n/a]                   |
| CeBr$_3$     | 0.649(11)%                     | 56%                         | 34%                     |
| LaBr$_3$(Ce) | 0.658(11)%                     | 56%                         | 34%                     |
| Cd(Zn)Te     | 0.01167(28)%                   | 1.0%                        | 0.59%                   |
| CsI(Tl)      | 0.3116(61)%                    | 27%                         | 19%                     |

3.2. Linearity

Knowledge of the linearity of a detector allows the user to determine the most appropriate energy calibration to perform. The linearity of each detector was assessed by comparing how the proportionality (channels per keV) varied across the energy range. The plot in figure 4 demonstrates the non-linearity of the LaBr$_3$(Ce), CeBr$_3$ and CsI(Tl) detectors, compared to the highly linear HPGe detector. A high order polynomial function was required to produce a satisfactory calibration of these detectors.

It was observed that the primary source of non-linearity for the LaBr$_3$(Ce)/CeBr$_3$ detectors was the lack of magnetic shield on the PMT. A comparison with a shielded LaBr$_3$(Ce) is shown in figure 4. To mitigate non-linearity, an energy calibration must be performed with gamma lines covering the full energy range of interest. Extrapolation outside of the energy range or interpolation between distant energy points is likely to result in significant errors.
3.3. Internal radioactivity

The background radiation detected by a gamma spectrometer produces interference that can mask the presence of a radionuclide of interest. Typically the background is produced by cosmic-rays and naturally occurring radioactive materials (NORM), such as $^{40}$K with an emission at 1461 keV. If the detector material itself contains radioactive material then a significant background will result from the high detection efficiency for internally emitted radiation.

A comparison of the background energy spectra measured by each detector is shown in figure 5. The background of Cd(Zn)Te is the lowest, due to its low absolute efficiency and low internal radioactivity. Several NORM gamma lines are evident in the HPGe spectrum, also suggesting low internal radioactivity. Evidence for alpha-emitting contaminants was found in the LaBr$_3$(Ce) and CeBr$_3$ spectra (between 2,000-2,500 keV for LaBr$_3$(Ce) and between 1,400-2,000 keV for CeBr$_3$). This was due to the presence of $^{227}$Ac, which is part of the natural $^{235}$U decay series. The contamination of CeBr$_3$ varies from batch-to-batch with certain detectors showing almost no evidence for internal alpha emission [8]. Due to the chemical similarity of La and Ac, $^{227}$Ac contamination is harder to remove from LaBr$_3$(Ce). LaBr$_3$(Ce) also contains the naturally abundant radioactive isotope $^{138}$La. Evidence was found for this in the LaBr$_3$(Ce) spectrum, in the form of a gamma peak at 1436 keV and gamma/beta sum peak at 789 keV [6]. The benefit of internal radioactivity is the presence of a consistent reference peak for gain stabilisation (which is required for detectors such as LaBr$_3$(Ce)/CeBr$_3$ that are known to drift with temperature).

3.4. Minimum detectable activity

The MDA of a detector is often quoted when a positive identification of a radionuclide is not made. It provides insurance against claiming a radionuclide is not present, simply because it was not detected. An air monitor with large MDA values for key radionuclides is not useful as the operator will be forced to report high concentrations of those radionuclides on a routine basis. Actual releases may well be missed.
MDA values for three key radionuclides were calculated: $^{241}\text{Am}$, $^{137}\text{Cs}$ and $^{60}\text{Co}$, and are presented in figure 6. The HPGe and CeBr$_3$ detectors performed best, with MDA values for 24 hr measurements of 0.1 and 0.2 Bq, respectively. It would take just 100 and 200 s, respectively, for these levels of contamination to be collected on a filter in an environment of 0.03 Bq/m$^3$, even with a modest pump rate of 10 m$^3$/hr (typical of a handheld air sampler). With this level of performance it would be prudent to better balance the sampling-to-measurement times (if not concurrent). The MDA values of Cd(Zn)Te were found to increase with energy as detection efficiency decreased. The MDA values of LaBr$_3$(Ce) were found to decrease with energy. This can be explained with reference to the internal background (see figure 5), which decreases from 60 to 1332 keV. The MDA values produced by the CsI(Tl) detector were reasonable despite the poor energy resolution.

Figure 6. Comparison of MDA values for $^{241}\text{Am}$, $^{137}\text{Cs}$ and $^{60}\text{Co}$ when measured in Cu/Cd lined Pb coffin over 24 hr.

4. Summary and conclusions

A comparison of new and emerging gamma detector technologies for airborne radioactivity measurements has been completed. The key performance metrics of energy resolution, detection efficiency and MDA have been assessed.

The HPGe detector produced superior resolution, efficiency and MDA values compared with the other detectors. However the HPGe detector tested was large, making it impractical to mount within an air sampler unless some form of filter progression was employed. A remote electromechanical cooler would improve practicality. The cost also limits the number of monitors that could be deployed across a network. The Cd(Zn)Te detector demonstrated good potential for radionuclide identification and was the most compact. The poor detection efficiency would however extend measurement times and delay response. The internal radioactivity of LaBr$_3$(Ce) resulted in large MDA values. The significantly lower internal background of CeBr$_3$ compensated for its modestly worse energy resolution. The CeBr$_3$ MDA values were comparable with those of the HPGe detector. The cost of LaBr$_3$(Ce)/CeBr$_3$ (inc. PMT and associated electronics) is typically lower than HPGe so more monitors could be deployed. Despite the poor energy resolution and relatively small volume, the CsI(Tl) detector produced reasonable MDA values. The low cost (over an order of magnitude lower than HPGe) and compact form are both highly desirable when considering in-situ gamma spectrometers for wide-scale deployment.

The detectors investigated covered the full range of the price-performance matrix and the results of the comparison will assist instrument manufacturers and networks operators in determining which technology satisfies their individual requirements.

Acknowledgments

The authors would like to acknowledge funding by the Department of Business Innovation and Skills through the National Measurement System, Euramet through the EMRP programme, the STFC though grant #ST/L005743/1 and the NDA through a PhD bursary. The authors would like to acknowledge Kromek Group PLC for the loan of detectors and support of the project. PHR acknowledges support from the STFC (UK) via grant number ST/L005743/1.
References

[1] Treaty establishing the European Atomic Energy Community
   http://ec.europa.eu/energy/en/topics/nuclear-energy/ last accessed 05/08/16.

[2] Cindro D et al. 2016. http://www.nss.si/nene2016/contents.html#5300 last accessed 05/06/16

[3] Navarro Ortega N et al. 2015
   https://www.ctbto.org/fileadmin/user_upload/SnT2015/SnT2015_Posters/T3.2-P4.pdf last accessed 05/08/16.

[4] MetroERM Consortium http://earlywarning-emrp.eu/ last accessed 05/06/16.

[5] Currie L A 1968 Anal. Chem. 40 (3) pp 586–93 DOI: 10.1021/ac60259a007.

[6] Lorusso G, Shearman R, Regan P H, Judge SM, Bell S, Collins S M, Larijani C, Ivanov P, Jerome SM, Keightley JD, Lalkovski S, Pearce AK, Podolyak Zs., 2016 Applied Radiation and Isotopes 109 pp507-511 DOI: http://dx.doi.org/10.1016/j.apradiso.2015.12.050

[7] Commission recommendation 2000/473/Euratom http://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:32000H0473&from=NL last accessed 05/06/16.

[8] Quarati F G A 2013 NIMA 729 pp 596-604 DOI: 10.1016/j.nima.2013.08.005.