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A portable real-time in situ gamma-ray analysis system

Cong Wei^{a,*}, Kelly Garnick^a, Thomas Scott^a, Elon Malkin^a, Jennifer Szymanski^a, Steve Laskos^b, Jeffrey Raimondi^b, James Cocks^b, Kara Morris^b, Wilhelm Mueller^b, and James Zickefoose^b

^aThe US Food and Drug Administration, Office of Regulatory Affairs (ORA), Winchester Engineering and Analytical Center (WEAC), 109 Holton Street, Winchester, MA 01890, USA

^bMirion Technologies (Canberra), Inc., 800 Research Parkway, Meriden, CT 06451, USA

Abstract

A portable CeBr₃ based gamma-ray detection system was designed and built for rapid turnaround, high throughput, real-time, and in situ sample analysis. The new technique allows automated data transmission from the field unit to a central laboratory controller to ensure laboratory quality of the data collected by field users without gamma-ray spectroscopy expertise. The method validation data indicates that the system's data quality objectives are adequate for radiological or nuclear emergency response or targeted surveillance programs where gamma-ray analysis is needed.

Keywords

Emergency response; Gamma-ray analysis; High throughput; Laboratory quality; Portable; Rapid turnaround

1. Introduction

A radiological or a nuclear incident, accidental or intentional, could not only cause a catastrophic effect on public safety and health, but it also can have impact on a nation's economy and security. For consequence management decision making, laboratory analysis of samples are needed. In these situations, the expected number of samples required for laboratory analysis will be on the order of 100,000 clinical samples and on the order of another 100,000 environmental samples resulting from a terrorist attack with a Radiological Dispersal Device (RDD) (U.S. House, Subcommittee on Investigations and Oversight, Committee on Science and Technology, 2007). There will be a substantial number of food samples required for laboratory analysis from the same RDD event. In the event of an Improvised Nuclear Device (IND), the damaging effect will be much more overwhelming.

*Corresponding author. Cong.Wei@fda.hhs.gov (C. Wei).

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Japan's 2011 nuclear power plant accident reaffirmed the scale of negative impact of a nuclear accident.

Efforts should be made for preparedness so that the adverse impact can be mitigated. During a radiological or nuclear emergency, a laboratory's capacity for analyzing a large number of samples and providing reliable laboratory analytical results with rapid turnaround time are critical for consequence management decision making. Gamma-ray analysis of samples is an efficient way for determining if the samples are contaminated by gamma-ray emitting radionuclides released due to the incident. A Cerium Bromide (CeBr_3) detector was chosen for the portable gamma-ray analysis system for its stability, high resolution, even though less than that of a high purity germanium (HPGe) detector, and room temperature application (Guss et al., 2009; Quarati et al., 2013).

With respect to sample preparation and data collection, conventional gamma-ray analysis demands experienced laboratory analysts to perform calibrations, process samples, perform data processing and analysis for laboratory conclusions as well as ensure quality control requirements are met. Sample preparation and data collection take substantial time from laboratory analysts and they are limiting factors for achieving rapid turnaround and high throughput analytical results. In addition to expertise shortage (The National Academies Press, 2012; Latshaw et al., 2011; Ware, 2002; Warwick et al.), the technical difficulties and required financial resources for establishing and maintaining a radio-analytical laboratory are also limiting factors for achieving the needed capacity for emergency response to a radiological or nuclear incident. While gamma-ray analysis requires expertise, utilization of a portable analytical system will help to achieve the needed rapid turnaround and high throughput laboratory grade analytical results if the analytical system is built with such features that it allows field personnel to perform sample preparations and data collections. This technique allows portability and operation by laymen without gamma-ray spectroscopy expertise.

2. System description and discussions

The portable gamma-ray analysis system consists of a central laboratory server controller and a field gamma-ray analysis system or field unit with a CeBr_3 detector. The CeBr_3 based gamma-ray detection system covers gamma-ray energy range from 30 keV to 3 MeV with a typical relative resolution 4.3% at 662 keV. Canberra Genie 2000, Apex-GammaTM and customized software are used in the central laboratory controller. Customized software with an application focused graphic user interface (GUI) based on Canberra's FoodProTM is used in the field unit. This software is preconfigured with specific counting times, analyses, and geometry calibrations using Canberra's LabSOCSTM (Bronson and Atrashkevich, 2001) efficiency calibration methodology. The field unit has the following features:

2.1. Light weight, modular, scalable, mobile

The field gamma-ray analysis system shown in Fig. 1 weighs about 50 kg with each modular component weighing less than 10 kg shown in Fig. 2. The size and material of each component can be changed (scaled) per application needs. For meeting data quality and design objectives such as robustness, and portability, a series of measurements and

numerical simulations using Monte Carlo N-Particle code (MCNPTM) (Briesmeister, 2000) were performed to determine the size and material for constructing the modular shield. Both simulation and experimental data were obtained from measurements of 450 mL samples in a deli container (approximately 475 mL) using an approximate 5 cm diameter CeBr₃ detector. Comparisons of experimental and simulation data for detection limit ($y^{\#}$) are shown in Fig. 3. Based on the data, about 1.3 cm thickness stainless steel was selected as the shield material for meeting the design requirements and data quality objectives. Two 20 cm (diameter) soft rubber back wheels and two 7.6 cm (diameter) soft rubber front caster wheels provide easy mobility.

2.2. Robustness adequate for field and in situ operations

The field unit, composed of the stainless steel shield and galvanized aluminum frame, may be assembled from component pieces in less than 10 min without hand tools. The temperature-stabilized CeBr₃ detector eliminates the need for cooling and the entire field system can be powered by a laptop computer via USB port for approximately 8 h before charging. The detector chamber has a removable/disposable spill guard that reduces the possibility of cross contamination.

2.3. Real-time in situ high throughput sample data collections

In a scenario of surge capacity needed for gamma-ray analysis of food samples, counting time can be decreased for rapid turnaround and increasing sample analysis throughput. However, a sample analysis process also involves laboratory analysts physically receiving, preparing samples to be analyzed, while also documenting the chain of custody and sample preparation. It is the authors' experience that the quickest turnaround time for completing gamma-ray analysis of a sample is about 30 min with 10 min spent on counting the sample. Assuming there are sufficient gamma-ray spectrometry systems available in a laboratory, the limiting factor for sample throughput is the number of analysts qualified for performing gamma-ray analysis. Using the portable gamma-ray analysis system, the central laboratory instrument controller allows qualified laboratory analysts to efficiently pull data for processing and analysis, typically completed within minutes. Furthermore, physical samples will not need to be shipped to laboratories if the portable gamma-ray analysis system is used in the field. Therefore, using the portable gamma-ray analysis system has the potential to provide much quicker turnaround analytical results for consequence management decision making, which is significant given the global shortage in radio-analytical expertise and costs associated with establishing and maintaining a radio-analytical laboratory infrastructure. While the field does increase the potential for variability in sample preparation and background, if these variations are controlled and contained within the system's data quality objectives, the field application of the system shall realize high throughput of sample analysis.

2.4. Free of knowledge and experience requirement of a field user

The design rule for the field gamma-ray spectrometer system is accomplishing data collection of a sample with a minimum of GUI clicks and entries (see Fig. 4). A field user primarily needs to composite and fill a sample into a gamma-ray counting container per a sample preparation procedure. Once the sample counting container is placed in the detection

unit, data collection of the sample is completed with a few operating steps. As shown in Fig. 4, a field user enters a sample number and sample description, performs background and calibration checks, and presses the “COUNT SAMPLE” button to accomplish data collection for a sample. Once the data collection is completed, a screen will appear to prompt the field user to take a photograph of the sample counting container as a part of quality control measures to allow a central laboratory analyst to ensure that the sample was composited and filled correctly. Note that use of the correct sample geometry is one of the important factors for meeting accuracy and precision components of the data quality objectives in gamma-ray analysis. The gamma-ray counting data, quality control data, sample information, and the photo of the sample container will be automatically packaged and transmitted to the central laboratory instrument controller if both the field unit and central laboratory instrument controller are connected to the internet or local network. If transmission of sample files to the central laboratory fails due to unavailability of the internet or interruption of data transmission, a screen will appear to prompt the field user to send the files to the central laboratory. The central laboratory analysts will perform data processing, analysis, and draw laboratory conclusions. Central laboratory analysts will ensure that a correct efficiency (corresponding to low, medium and high density) correction is applied based on the sample compositing and matrix information. The laboratory conclusions will be based on the measured values determined by central laboratory analysts and will be used for consequence management decision making. Therefore the gamma-ray spectroscopy knowledge and experience are not required of a field user. Normally if the measured activity concentration of a radionuclide of concern in a sample is above its corresponding regulatory guideline value, a check analysis of the sample will be performed either with the same analytical system or a central laboratory’s HPGe gamma-ray analysis system. If the check analysis confirms the result of the original analysis, the decision making personnel will decide whether the product shall be withheld from being released into the commerce based on the analytical result and other relevant factors. However, the decision making protocols may be revised during an emergency response to a nuclear incident.

2.5. Laboratory analytical data quality

One of the analytical laboratory’s major products is the analytical data. Naturally there is always a pressure and demand for providing rapid turnaround of analytical results for consequence management decision making in an emergency response situation. However, data quality must not be compromised because inaccurate data will result in making inappropriate consequent management decisions. For the portable gamma-ray analysis system, analytical data quality is ensured by implementing the same quality control procedures per ISO/IEC 17025 requirements as applied in an ISO/IEC 17025 accredited laboratory. The unit is efficiency calibrated using a deli container filled to the top (equivalent to the 450 mL standards). The efficiencies are created assuming that the density of the sample being analyzed is the density midpoint of the low, medium and high density ranges. A factor, k , the midpoint sample weight divided by the actual sample weight, is used for correcting sample density effect on efficiency if the sample weight information is available. For analysis in the field where a balance is not available, the value of k is assumed to be 1. The uncertainty in the assigned sample weight is included in the k factor. Because of the opposing effects of sample weight and density on efficiency, the uncertainty in activity

concentration within a density range due to the assignment of $k = 1$ for field applications where a balance is not available should be smaller than the deviation in the k factor. Spiked samples used for method validation with known activity values were used to assess the uncertainty in activity concentration caused by the k factor in the density range from 0.4 g/mL to 1.4 g/mL. Based on the obtained uncertainty data, the low, medium, and high density ranges were assigned to limit the uncertainty caused by the k factor not to exceed 10% within each density range. We have also established a database that contains the information on density and food correlations. Based on the sample description provided by a field operator and the knowledge of density and food correlations, a central laboratory analyst will be able to use an appropriate density range for data processing. Before performing analysis of samples, the field unit GUI enforces an automated background check for verifying the absence of the analytes of interest in the background and calibration check with an exempt ^{152}Eu standard (Eckert and Ziegler Isotope Products, Certificate Number: 1868-63-1, activity: 1.02 ± 0.03 kBq). An epoxy matrix standard (Eckert and Ziegler Isotope Products, Certificate Number: 1868-63-2) with ^{133}Ba (1091 Bq/kg at both 303 keV and 356 keV), ^{137}Cs (1138 Bq/kg at 662 keV), and ^{60}Co (1149 Bq/kg at both 1173 keV and 1333 keV) is used as a laboratory control sample and analyzed with a batch of samples. The relative uncertainties of the activities in the standard were certified as 2.3% with 95% confidence interval. All the quality control data files with the sample counting geometry information are reviewed by central laboratory analysts to ensure the quality of the analytical system, sample preparation, and data collections. Fig. 5 shows a spectrum obtained with the portable gamma-ray analysis system using a sample spiked with ^{134}Cs at 590 Bq/kg and ^{137}Cs at 620 Bq/kg. As shown in Fig. 5, ^{134}Cs and ^{137}Cs were clearly resolved for obtaining quantitative results. Spike levels were confirmed with HPGe assay while homogeneity was confirmed by splitting the sample and confirming agreement in activity. For ^{134}Cs , all activity determinations are calculated using the 795 keV line only due to the potential interference of the 609 keV (^{214}Bi) background and relative closeness of less abundant ^{134}Cs peaks, e.g., the 604 keV of ^{134}Cs peak. Fig. 6 shows a spectrum obtained from the epoxy matrix standard (Eckert and Ziegler Isotope Products, Certificate Number: 1868-63-2) with ^{133}Ba , ^{137}Cs , and ^{60}Co covering the energy range of interest using the portable gamma-ray analysis system. The spectrum also indicated that the radioisotopes were clearly resolved for obtaining quantitative results.

3. Data quality objectives and method validation data

The detection limits, trueness, precision, and uncertainty were evaluated using food matrices with densities ranging from 0.4 kg/L to 1.4 kg/L.

3.1. Detection limit

The targeted detection limit for ^{134}Cs , ^{137}Cs , and ^{131}I are below 100 Bq/kg with 20 min gamma-ray counting time. The detection limits were obtained using Currie's method for paired observations (Currie, 1968) and international standard ISO11929 respectively. Per Currie's method for paired observations, the detection limit in activity concentration (Bq/kg) was expressed as follows:

$$y^{\#} = \frac{2.71 + 4.65 \times \sigma_B}{\varepsilon \times ab \times T \times DF \times k} \quad (1)$$

Where, ε is massimetric efficiency (cps/(dps/kg)), ab is the gamma-ray abundance and branching ratio factor, T is count duration in seconds, DF is the decay factor, k is the ratio of the assigned range sample weight to the actual sample weight, which is assumed to be 1 for field analysis when a balance is not available, σ_B (for the sample blank) is the standard deviation of the counts observed within the peak channel range that are not determined to be part of the net peak value.

The term σ_B will be determined differently depending upon whether a peak is identified within the tolerance window of the identifying library energy. When there is not a detected peak, σ_B is the total counts within the peak window defined by the radionuclide library energy with a Full Width at Half Maximum (FWHM) calculated using the shape calibration, whereas when a peak is detected, σ_B is the propagated uncertainty in the continuum counts below the integrated peak and the peak itself.

Following the international standard for determination of the characteristic limits for measurements of ionizing radiation (ISO, 11929; 2010(E)), assuming there are not any shielding factor and additional background corrections, the net signal y in activity concentration (Bq/kg) can be expressed as:

$$y = G(x_1, \dots, x_m) = (x_1 - x_2) \bullet w \quad (2)$$

Where $w = \frac{1}{\varepsilon \times ab \times T \times DF \times k}$, at a certain time interval, x_1 is gross count, x_2 is background count, and w accounts for non-count related input quantities that affect the net signal in activity concentration (Bq/kg). Also assuming both gross and blank counts follow Poisson distribution, and $k_{1-\alpha} = k_{1-\beta} = 1.645$, the $y^{\#}$ can be derived as:

$$y^{\#} = \frac{2.71 + 4.65 \times \sigma_B}{\varepsilon \times ab \times T \times DF \times k \times \{1 - 2.71 \times u_{rel}^2(w)\}} \quad (3)$$

Where

$$u_{rel}^2(w) = \sigma_{\varepsilon}^2 + \sigma_{ab}^2 + \sigma_T^2 + \sigma_{DF}^2 + \sigma_k^2 + \sigma_{CS}^2,$$

σ_{ε} is the uncertainty in the efficiency for the peak used to calculate the radio-activity using Canberra software; σ_{ab} is the uncertainty in the abundance for the peak used to calculate the radio-activity as defined by Brookhaven National Laboratory; σ_T is the uncertainty corresponding to the counting time; σ_{DF} is the uncertainty associated with the decay factor; σ_k is the uncertainty due to the difference in sample matrix both due to weight and

composition used for establishing the efficiency and the sample; and σ_{CS} is the uncertainty due to the coincidence summing correction. Both σ_{DF} and σ_T are very low and negligible in comparison to other uncertainty sources.

Twenty five water blanks, fifty food matrix blanks with 19 different matrices, e.g., vegetables, fruits, grains, cheese, honey etc. and fifty six spiked food samples with 14 different matrices, e.g., grains, meat, jam, honey etc. were used for evaluating the $y^\#$ s and efficiency respectively. Samples were bracketed into 3 density groups for correction of density effect on radio-activities: (1) low density: 0.4 – 0.7 (kg/L); (2) middle density: 0.7 – 1.1 (kg/L); (3) high density: 1.1 – 1.4 (kg/L).

Table 1 shows a summary of average $y^\#$ values for ^{134}Cs at 795 keV, ^{137}Cs at 662 keV, and ^{131}I at 364 keV obtained from the measurement of food matrix blanks. Table 2 gives a summary of average $y^\#$ values for ^{134}Cs , ^{137}Cs and ^{131}I at the same corresponding energies obtained from samples spiked from 90 Bq/kg to 465 Bq/kg for ^{134}Cs , 140–610 Bq/kg for ^{137}Cs . The data indicated a higher $y^\#$ with lower density food matrices and that all $y^\#$ were well below the corresponding FDA recommended derived intervention levels (DILs) (U.S. Food and Drug Administration, 2004).

3.2. Trueness, precision, and uncertainty

Trueness and precision were obtained from 15 sample matrices with density ranged from 0.4 kg/L to 1.4 kg/L and shown in Table 3. The relative total combined standard uncertainty was estimated using Eq. (4). The combined standard uncertainties were 19% for ^{134}Cs , 16% for ^{137}Cs , and 21% for ^{131}I , with the major contributions coming from counting (uncertainty estimated with 10% counting contribution), the efficiency and the k factor.

$$\sigma_A = \sqrt{\sigma_P^2 + \sigma_{DF}^2 + \sigma_T^2 + \sigma_{ab}^2 + \sigma_\varepsilon^2 + \sigma_k^2 + \sigma_{CS}^2} \quad (4)$$

Where σ_P is the uncertainty in the peak area.

4. Conclusion

The communicated analysis system, to our knowledge, is the first kind of gamma-ray analysis system that enabled an analytical method per ISO/IEC 17025 to ensure laboratory grade data quality without gamma-ray spectroscopy knowledge and experience requirements of the field user. The concept and design features that enabled the removal of gamma-ray spectroscopy knowledge and experience requirements of the field user can also be applied to any other analytical subject areas where laboratory resources are limited and expertise is in shortage. The application of the system will address the global shortage in radio-analytical expertise, dramatically increase efficiency and effectiveness of operations, and reduce the costs for establishing and maintaining gamma-ray analytical capability.

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HIGH LIGHTS

- A portable system for quantitative gamma-ray analysis was developed.
- The system has a modular shield and a user-friendly graphic user interface.
- Gamma-ray spectroscopy expertise is not required for operating the field unit.
- Data quality is ensured by implementing procedures per ISO/IEC 17025 standard.
- The system can dramatically increase the nuclear emergency response capacity.

Fig. 1.
Portable gamma-ray analysis system.

Fig. 2.
Individual modules of the portable gamma-ray analysis system.

Fig. 3. Computed and measured detection limits for ^{134}Cs , ^{137}Cs , and ^{131}I .

Fig. 4.
Field unit graphic user interface.

Fig. 5. Spectrum obtained from a mashed potato sample spiked with ^{134}Cs and ^{137}Cs , count time: 20 min.

Fig. 6.
Spectrum obtained from an epoxy matrix standard, count time: 20 min.

Summary of average detection limit, $y^\#$, values (Bq/kg) obtained from method blanks, measurement time: 20 min.

Table 1

	^{134}Cs 795 keV		^{137}Cs 662 keV		^{131}I 364 keV	
	ISO 11929 $y^\#$	Currie $y^\#$	ISO 11929 $y^\#$	Currie $y^\#$	ISO 11929 $y^\#$	Currie $y^\#$
Water	38	36	36	34	37	35
Low Density Foods	52	49	47	44	48	45
Medium Density Foods	35	33	31	29	32	30
High Density Foods	27	26	25	23	26	24

Summary of average detection limit, $y^\#$, values (Bq/kg) obtained from spiked samples, measurement time: 20 min.

Table 2

	^{134}Cs 795 keV		^{137}Cs 662 keV		^{131}I 364 keV	
	ISO 11929 $y^\#$	Currie $y^\#$	ISO 11929 $y^\#$	Currie $y^\#$	ISO 11929 $y^\#$	Currie $y^\#$
Low Density Foods	62	59	61	58	54	50
Medium Density Foods	51	48	51	48	46	42
High Density Foods	34	32	35	33	32	30

Table 3

Summary of trueness and precision, measurement time: 20 min.

	Radionuclide	Energy (keV)	%Precision	%Bias
Low density	^{134}Cs	795	9	-9
0.4 – 0.7 (g/mL)	^{137}Cs	662	6	-15
Medium density	^{134}Cs	795	15	-3
0.7 – 1.1 (g/mL)	^{137}Cs	662	19	-12
Epoxy matrix standard (1.05 g/mL) (^{131}I Surrogate)	^{133}Ba	356	3	-8
High density	^{134}Cs	795	12	-8
1.1 – 1.4 (g/mL)	^{137}Cs	662	13	-16