

AN EASY, CHEAP AND SCALABLE METHOD TO CHECK FOR RADIOACTIVITY IN INPUT COMMODITIES AND RAW MATERIAL

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ABSTRACT

Some release of radionuclides into the environment can be expected from the growing number of nuclear plants, either in or out of service, most of them obsolete.

The citizen and the big organization both could be interested in methods for checking the radiological safety of commodities, starting from foods and of their environment. Current technology allows almost everybody to become an active node of a network of environmental sensors, with a huge production of valuable data.

Four methods to detect radioactivity are discussed and compared in this work. They are different in technology, sensitivity, resolution and cost but interesting to build a working detection and measurement chain which can start from a diffused and networked first screening to the final high resolution analysis.

The results of the analyses of food samples are presented with gamma spectra of reference sources. The reported results can be usefully transferred into a radiological emergency scenario, but their more interesting value could be found in the implications on current everyday life.

Keywords : Radionuclides, gamma-rays, gamma-spectrometry, gamma-counter, food safety

INTRODUCTION

The number of nuclear plants in the world, either in or out of service, is growing and many of them, built with obsolete technology are becoming older and older. Therefore some release of radionuclides into the environment can be expected in the future.

For this reason, the common (informed) citizen and the big organization both could be interested in methods for checking the radiological safety of the local environment and of commodities, starting from foods.

BACKGROUND

In recent years, an astounding availability of portable and wearable (Zheng 2014) sensors has flooded the consumer market, as an effect of the explosion of the cell phone market.

Almost everyone can currently monitor georeferenced environmental parameters and can know and share their measurements in real time (Wang 2011).

The parameters that can be more easily measured by a smart-phone and the proper apps installed and running are: local magnetic field (Intensity and direction), visible light (intensity and composition in terms of three-component colorimetric measurement), sound (level and spectrum), atmospheric pressure, atmospheric humidity, latitude, longitude, speed, acceleration, local gravity. In the same way is better known that almost everyone can take an share in real time georeferenced photos, videos and sound registrations.

It is less widely known that with the same hardware (a current generation smart-phone with a camera) and the proper software, everyone can monitor radioactivity and take some measurement of its intensity, although with some sensitivity limitation (Auzinger 2012, Dosiek 2013, Willems 2015, Drukier 2011). Furthermore, with a proper setting and sufficient data, limited spectroscopy information can be determined (Cogliati 2014).

MAIN FOCUS OF THE PAPER

In this work, four methods for radioactivity detection and measurement in commodities and in the environment are discussed and compared. They are different in technology, sensitivity, resolution, and cost but interesting to build a working detection and measurement chain which can start from a diffused and networked first screening to the final high sensitivity and high resolution analysis (Chiozzi 2000).

Two method rely on the current smart-phone technology, with no or minimal additional hardware: for this reason they are really cheap, widely available and could be precious for a wide first screening activity.

Two further methods are well known laboratory methods that depends on specialized instruments. These are much more sensitive, expensive and involve heavy instrument but in a new possible scenario characterized by huge amounts of data produced by the new first-step methods, they can play a new role for measurements validation.

One of them is presented here in a tested, working, networked and remotely controlled configuration.

METHOD, APPROACH AND LIMITATIONS

Reference Sources

To test and calibrate the detectors adopted in this work, a reference standard, characterized by a weak and safe (NRC/IAEA/EU exempt quantity), but easily detectable gamma ray emission, was needed. It was chosen a set of eight reference point source: the RSS8UN by Spectrum Techniques, LLC., including Ba-133, Cd-109, Co-57, Co-60, Mn-54, Na-22 and Zn-65 at 3.7×10^4 Bq each and Cs-137 at 3.7×10^3 Bq (below limits for declaration).

The smartphone as a CMOS gamma-ray detector

The first detector adopted was a Samsung S4 smartphone (<http://www.samsung.com/global/microsite/galaxys4/>), equipped with two specialized apps: “GammaPix” (<http://gammapij.com/sites/>) (Drukier 2011) and RadioactivityCounter (<http://www.hotray-info.de/>) (Auzinger 2012, Van Hoey 2015). Both apps are available for Android and iOS systems.

CCD or CMOS chips, used as digital image sensors in surveillance or in smartphone cameras, are sensitive not only to visible light but also to higher energy photons. The software analyses the signals produced by the front or by the rear camera of the smartphone, previously shielded from visible light, subtracts the thermal noise and estimates the gamma-ray exposure of the sensor. Also the background emission (measured before) can be subtracted.

A cheap, lightweight, advanced and networked Geiger counter

The second detector was the Geiger counter PRD 100 (<http://www.prd100.com/>), made by the Italian firm ITS srl. This device is equipped with a Geiger Muller tube of 111 mm length x 11 mm diameter (max. section 1221 mm^2), which is small enough to allow full portability but whose section is much larger (82 times) than the section of the smartphone sensor ($4.4 \times 3.4 = 15 \text{ mm}^2$) and can intercept more radiation.

This detector shows further interesting features: is cheap (around 100 eur), is small and easily portable (190g, $123 \times 91 \times 35 \text{ mm}$), works with rechargeable standard batteries (3 AA), can work in stand-alone mode and/or connected with a smartphone via a Bluetooth radio interface.

A dedicated app (Marie pro PRD-100: <https://play.google.com/store/apps/details?id=it.ipom.marieprd100&hl=en>) running on the smartphone provides some essential real time radiation counting and data displaying, saving, sharing and geo-referencing.

The core of the classical gamma-pectrometry system: the NaI(Tl) scintillator

The third instrument adopted in this work was a NaI(Tl) gamma spectrometer made by Ortec, priced around 18000eur. The system hardware consists of a thallium-doped sodium iodide detector enclosed in a low-background lead shield (30 mm thick), an analog-to-digital converter (ORTEC DigiBase) integrated in an all-in-one spectrometer, and a laptop PC. The digiBASE supplies the multichannel analyzer function, the high voltage for the NaI(Tl) detector, and all the signal processing electronics. The internal stabilization electronics and the internal check source (K-40 4500 Bq/kg) allow the system to be used over a wide range of environmental conditions. Anyway it can be hardly defined portable, if the 80 kg lead shield is considered. The NaI(Tl) crystal is a 3” x 3” standard. The digiBase is connected to the control computer via a USB interface, which powers the whole system.

Several proprietary software components control the instrument, from the first setting to final analyses. Ortec MCB Connections-32 acts as a first-level connection driver for the DigiBase. Maestro-32 MCA Emulation Software provides the second-level control of the DigiBase, the live spectral display and the automatic control of acquisition and analysis. This is achieved via a graphical user-programmable interface or via preprogrammed job streams. The software provides also data and results printing and storage. NuclideNavigator is an interactive gamma-ray reference and library program to view, query, and extract gamma-ray energies and yields, half-lives and parent/daughter relations from databases. It can be used to build application libraries or working libraries. ScintiVision-32 is an integrated multichannel analyzer (MCA) emulator and gamma-spectrum analysis program. It integrates Maestro-32 functions and manages the collection and analysis of gamma-ray spectra. It includes commands that allow you to edit nuclide libraries and automated command sequences or “job streams.”

The set of technologies described above makes gamma-spectroscopy with sodium iodide scintillators an excellent tool for the identification and quantitative determination of gamma ray emitting radioisotopes, either the natural K-40, U-238, Th-232 or the anthropic Cs-137 and I-131 (Caciolli 2012).

The top level technology for gamma spectrometry: the (HPGe) detector

The fourth method introduced here, but not experienced in this work, is based on High Purity Germanium (HPGe) detectors. High Purity Germanium detectors can give excellent gamma signal resolution for energies as low as 3 keV, where NaI(Tl) detectors cannot usefully work.

The major drawback of germanium detectors is that they must be cooled to liquid nitrogen temperatures to produce spectroscopic data. An HPGe system is more complex to manage and on average is priced five to ten times a NaI(Tl) system.

The most common figure used to express detector resolution is Full Width at Half Maximum (FWHM). This is the width of a gamma ray peak at half of the highest point on the peak distribution. Resolution can be expressed in absolute (eV) or relative terms. For example, a sodium iodide detector may have a FWHM of 9.15 keV at 122 keV (in absolute terms). To express the resolution in relative terms, the FWHM is divided by the energy of the gamma ray and multiplied by 100. The resolution of that detector is 7.5% at 122 keV. A germanium detector may give resolution of 560 eV at 122

keV, yielding a relative resolution of 0.46%, i.e. 16 times better than NaI(Tl). For this reason the multichannel analyzer connected to a HPGe detector can be usefully set to 16384 channels instead of the 1024 related to the NaI(Tl) technique.

The HPGe systems represent the state of the art in current gamma spectroscopy, and can be set at the top end of a working detection and measurement branched chain, which can start from a diffused and networked first screening to the final high resolution analysis by HPGe.

The radon issue

The Radon is an ubiquitous gas, mainly derived from the natural U-238 decay chain.

Radon isotopes emit alpha particles but some radionuclides from Radon's progeny (Pb-214, Bi-214) emit gamma rays and can add their signals to the spectrum of a sample. To address this problem an independent Radon sensor was adopted to monitor the radon concentration in real time during the analyses by the NaI(Tl) system.

The sensor adopted is the Rstone by the Italian firm Rsens. It can be connected to a computer by a proprietary USB pen. A proprietary program can read and analyze the data stored in the sensor's memory. The sensor itself runs on a battery whose charge can guarantee two weeks of continuous autonomy and monitoring, and has a display to check the current Radon concentration as measured during the last 30'.

Background management and Calibration

With the "RadioactivityCounter" app running on a smart.phone, the sensor's noise and the background emission can be measured and stored in memory, and can be subtracted from every sample's measure, that is expressed and saved as total and by-minute CPM, together with the sensor temperature, that can affect the measure a lot. Similar features are offered also by GammaPix, but by a less clear user interface.

No background compensation is provided by the PRD-100 Geiger counter and by the corresponding Marie PRO PRD-100 app running on a smart phone connected to the instrument. The counting is only instantaneous.

The NaI(Tl) and the HPGe gamma spectroscopy systems allow full control of the background and the former will be described in some detail hereafter.

The lead shield effectively prevents the variable environmental background radiation from hitting the sensor, but the shield itself is a source of radiation mainly from U-238, and Th-232 decay series; anyway this radiation is constant and can be measured and subtracted with high reliability and precision, acquiring "blank" spectra periodically. The K-40 internal standard can also be included in the background. For this reason a spectrum of the background and of the internal standard was acquired for 963933 s, checking for its stability and saving the result every 1800 s. The stability was assessed by keeping the laboratory temperature constantly as near as possible to 295K and monitoring the stabilizer of the instrument, locked to the K-40 peak: if some adjustment occurred, then the corresponding 1800 s set of data were discarded. This way, the centroid of the photopeak of the K-40 at 1461 keV was kept always corresponding to the channel 552.20 +/- 0.20.

This spectrum was used either for the background subtraction or for the first calibration step (Calin 2011,) and all the following spectra (of the reference sources and of the samples under study) were acquired with the constraint of the K-40 centroid corresponding to channel 552.20 as described above.

The energy calibration of the NaI(Tl) system was done in several steps. For a first energy calibration the K-40 peak (4518 Bq) was considered with some clearly recognizable peak of the background: the Tl-208 peak at 2614 keV, near the high energy end of the spectral window, from the Th-232 decay chain, which is widely used as gamma tracer of natural thorium (IAEA 2003, van der Graaf 2011) and the Bi-214 peak at 1765 keV which is widely used as gamma tracer of natural uranium (cit. lit.).

During a second step the energy calibration was refined by considering, from the same background spectrum, the Ac-228 peak at 969 keV, the Tl-208 peak at 511 keV and the Pb-212 at 239 keV from the Th-232 decay chain.

For the final energy calibration steps, three reference sources were adopted and their corresponding spectra were acquired: Ba-133 (53 and 81 keV peaks), Cd-109 (88 keV peak) and Cs-137 (662 keV peak). The spectrum of each source was acquired separately, for 66691 s (Ba-133 and K-40), for 14189 s (Cd-109 and K-40) and for 81453 s (Cs-137 and K-40). The three point sources were acquired without any correction for the geometry, at 75mm from the detector surface, to lower the intensity of their signals and make it possible for the stabilizer to work properly.

The final calibration was as follow:

$$\text{Energy} = -7.7883 + 2.568091 * \text{Channel} + 0.000165948 * \text{Channel}^2$$

The full width at half maximum (FWHM) was calibrated as a linear function of energy:

$$\text{FWHM} = 4.8213 + 0.038887 * \text{Channel} + 0 * \text{Channel}^2$$

Efficiency was factory-calibrated (polynomial with uncertainty = 1.5025 % and coefficients: -0.100632, -4.638859, 1.053616, -0.094965, 0.002519, 0.000000. The efficiency at energy E is given by:

$$\text{Efficiency} = 10^{(a_1 + a_2(\log_{10}(E)) + a_3(\log_{10}(E))^2 + a_4(\log_{10}(E))^3 + \dots)}$$

where every a_n is a fitting coefficient.

The systems to start testing the analysis chain

Four infant food samples were tested and their activity compared with the activity of the Cs-137 and K-40 reference sources. They were analyzed as they were found on the market, without any purification, extraction or concentration or treatment except Sample 5, which was lyophilized.

The infant food samples were organic, from the same brand, but different in composition:

Sample 1) 250 g of homogenized food composed by apple, banana and cookies

Sample 2) 160 g of homogenized food composed by "mixed vegetables" (carrot, potato, cauliflower, pea)

Sample 3) 250 g of homogenized food composed by banana fruit

Sample 4) 160 g of homogenized food composed by cod, carrot, potato

Sample 5) 1375 g of lyophilized Sample 1

RESULTS AND IMPLICATIONS

In the laboratory where the NaI(Tl) spectrometer works and where also the two radiation counter were tested, the Radon concentration varied between 20 and 60 Bq/m³ and at this level, did not required any special attention and the main environmental problem was to keep the temperature stable to have the NaI(Tl) spectrometer stable as well.

All the food systems, tested by the two radiation counters under study, were not different from the background. Only a radioactive reference source could induce a response clearly different from the background in both instruments, as reported in Table 1, which shows the measures obtained at different distances from a point reference source of Zn-65 of 37000 Bq. It can be observed that the smart-phone's sensor performed better than expected from the difference of the two sensitive sections involved.

For the sake of comparison, a reference source of Cs-137 of 3700 Bq (tenfold less than Zn-65), put at 75 mm from the section of the NaI(Tl) sensor (18200 mm²) is "viewed" by the spectrometer as a 1L sample containing 7262 Bq/kg of Cs-137 (Pic. 1 and Tab. 2). This kind of instrument easily recognizes contents as low as the Tl-208 of the background, "viewed" as a concentration of 19.4 Bq/kg (Pic. 2 and Tab. 2).

The five samples, from the radiological point of view are very similar, with some difference in the Potassium contents, that can explain the different levels of K-40 (Tab. 2). The sample 5, 1375 g instead of 250 or 160 g and the longest acquisition time shows its K-40 content very clearly (Pic. 3) and with high reliability.

The Pb-214 value of sample 1 is not confirmed by the more reliable analysis of its lyophilized counterpart and depends only on the 74.81 keV peak: a too low energy to be considered alone with the NaI(Tl) technology.

CONCLUSIONS

The current technology opens the doors to a massive radiation monitoring, where the number of data can compensate for the possible lack of precision and it puts in the hands of the common citizen a direct knowledge that yesterday only specialized entities could access.

Furthermore, the fact that everyone can check for the safety of the nearby environment allows a new kind of bottom-up control of the information released by the public authorities and a more transparent decision making.

The reported results can be usefully transferred into a radiological emergency scenario, but their more interesting value can be found in the implications on current everyday life.

Current technology allows almost everybody to become an active node in an environmental sensory network, where the value of the output information can only be imagined.

Of course the huge amount of environmental data that could be produced and shared needs some public revision and lab validation, and here a sensitive, traditional and not expensive technology like the NaI(Tl) spectrometry can play a new role, before the definite Hi-Res confirmation that the HPGe technology can give.

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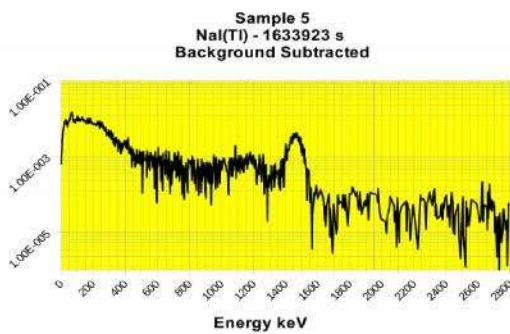
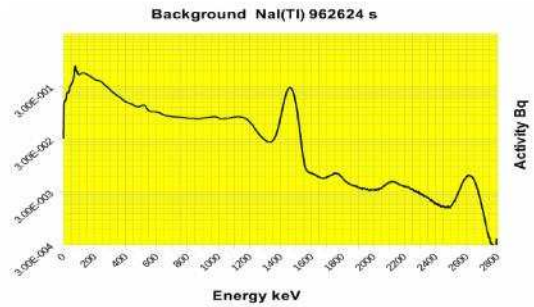
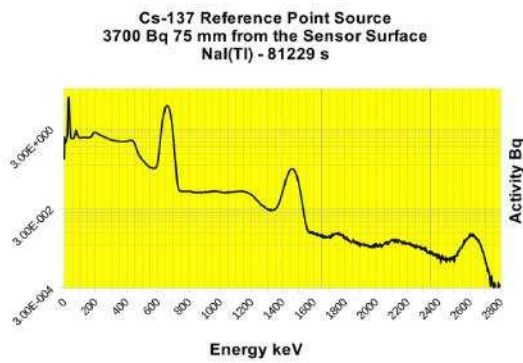
TABLES AND PICTURES WITH CAPTIONS

Source	Bq	37000	Time for each measure: 1200 s				
Distance	mm	(Background)	75	30	15	5	0
Device/Software							
PRD 100/MariePro	Bq	0	0.5	1.5	3	5	7
S4/GammaCounter	Bq	0.03	0.08	0.12	0.2	0.43	0.46

Table 1: Results of the two tested radiation counters put at different distances from a reference source (Zn-65, 37000 Bq).

Sample	Acquisition Time	Total Activity	K-40 Activity	Pb-214 Activity	Pb-212 Activity	Tl-208 Activity	Cs-137 Activity
	s	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg
1	434448	56.69	7.406	44.13	0.1396	5.0132	
2	409147	36.76	29.9	2.708	4.156		
3	599858	5.06				5.06	
4	962624	27.4	27.4				
5	1634627	60.85	60.85				
Background	963033	4571	4538	9.851	4.209	19.4	
Cs-137 3700 Bq	81453	7262					7138

Table 2: Results of the NaI(Tl) analyses of the five samples under study, of the background including the K-40 internal standard and of a Cs 137 reference source of 3700 Bq.



Picture 1 (top left):

The reference source Cs-137 (662 keV) with the internal standard (K-40 at 1461 keV) and the TI-208 background peak at 2614 keV.

Picture 2 (top right):

The background, including the K-40 (1461keV 4538 Bq); also the peak of TI-208 (2614 keV, 19.4 Bq) is very clear.

Picture 3 (bottom left):

The Sample 5, whose content of K-40 (1461 keV) is made clear by the background subtraction.