Measurement of Naturally Occurring Radionuclides with Several Detectors: Advantages and Disadvantages

C. Papastefanou
Aristotle University of Thessaloniki, Atomic and Nuclear Physics Laboratory, Thessaloniki 54124, Greece

Measurement of naturally occurring radionuclides in fields (in-situ) or in Lab (in-vitro) is made by sodium iodide thallium activated, NaI(Tl) detectors and or solid state (semiconductor diode) Ge high purity detectors coaxial, planar or well type by using various spectroscopic systems linked with accumulation data processing units. For spectral analysis of gamma-ray photon peaks, various libraries with softwares have been developed and established and are available to the users. Those gamma-ray spectrometers are classified according to their resolution and the efficiency in detecting and measuring the radionuclide concentrations through the gamma-ray peaks of the gamma-ray photons emitted by the naturally occurring and or man-made produced radionuclides in various kinds of environmental samples.

This article is designed to remove the "black box" approach to gamma-ray spectroscopy results, i.e.,

Put the sample on the detector
Push the button
Read the printed report
Accept the results.

A solid basis is provided in the fundamentals of gamma-ray spectroscopy while focusing on the areas that permit the operator and or user to :

Prepare a representative sample
Optimize system parameters
Understand the effects of cascade summing, interference peaks, geometry, and libraries parameters

Class exercises guide the student through the interpretation of results with consideration of peak fit, source term and process knowledge of the sample. Laboratory quality assurance, QA and good practices are also discussed. The users might be introduced to the concepts and benefits of modeled geometries and field (in-situ) or in Lab (in-vitro) measurements.

The article will review the interaction of radiation with matter to explain spectral features and their interpretation, including peak identification and energy determination, backscattering peaks, single and double escaping peaks and proper use of control charts of radionuclides. It is designed to provide a practical introduction to gamma-ray spectroscopy for those new to the field of gamma-ray spectroscopy, but also provide practical applications to those who are currently performing gamma-ray spectroscopy. It is intended for anybody who will be doing routine and specialized gamma-ray spectroscopy, as well as quality assurance officers and data validators who may have a need to understand gamma-ray spectroscopy measurements.

1. SCINTILLATION DETECTORS

The more usual way of detection and measurement of gamma-rays for a long time period in the decades of '50s to'70s was by using a scintillation detector (Heath, 1962, Heath, 1964, Siegbahn, 1968, Knoll, 1979, Tsulfanidis, 1983). The scintillation detector spectroscopy system is consisted of a scintillation crystal of sodium iodide thallium activated, NaI(Tl) of various sizes, a photomultiplier, a series

Correspondence to: C. Papastefanou, Aristotle University of Thessaloniki, Atomic and Nuclear Physics Laboratory, Thessaloniki 54124, Greece
Phone: ...., E-mail: ....

of nuclear electronic instruments, such as high voltage bias supply, a linear spectroscopic amplifier, a pulse counting unit which could be a single channel analyzer, SCA or multichannel analyzer, MCA . In the decades of '80s and '90s, a PCA card started using for analyzing the gamma-ray spectra in PC computing facilities (Tennelec, 1988) and in nowadays picospec digital multichannel analyzer units, dMCA-pro-card are used (Target, 2000). These units include a spectroscopy amplifier, a digital multichannel analyzer and a high voltage bias supply altogether in one unit.

The main advantages of the scintillation detectors, NaI(Tl) against the other type detectors, e.g. of Ge detectors used for detection and measurements in the gamma-ray spectroscopy are the following:

i) High efficiency
ii) Possibility of counting of a high number of events of gamma-ray decays
iii) Possibility of measurement of gamma-ray photon energy
iv) Low cost of scintillation detectors against the solid state (semiconductor diode) Ge detectors.

This of course, implies bad resolution which compensates for the high efficiency.

When a gamma-ray photon falls onto the scintillation crystal NaI(Tl) and inserts in its volume, it causes the excitation effect. That means, the photon interacts with the matter of crystal by

the photoelectric effect
the Compton effect, and
the pair production effect.

The final result of excitation of the scintillation crystal is the emitting of photons (fluorescent light photons) by the crystal which is lead to the photocathode of a photomultiplier, PM by contributing of the reflector surrounding the scintillation crystal except one side, i.e. the side of connection of crystal with the PM. The fluorescent light photons have a wavelength ranging from 3900 to 5100 Angstroms.

The number of the emitted fluorescent light photons is proportional to the energy of the gamma-ray photon which fell onto the crystal and absorbed in the crystal matter by the electrons of the previously mentioned three effects. These fluorescent light photons after their production fall onto the photocathode of PM, which has a small work function and extract or produce photoelectrons which are then multiplied by the dynodes of PM. Finally, in the anode of PM arrive 10^6 to 10^8 electrons which consist an electronic pulse of about 50 mV. The electronic pulses due to the gamma-ray photons fallen onto the scintillation detector are linearly amplified and by energy discrimination and analysis are then counted.

The energy response of NaI(Tl) crystal is shown in Fig. 1. In fact in this figure, the cross section of absorption of gamma-ray photons, ì as a function of their energy is shown. The gamma-ray photon absorption coefficient, ì is the sum of three coefficients, that is σ_f (photoelectric effect), σ_c (Compton effect) and σ_p (pair production effect).

For gamma-ray photons with energy higher than 300 keV, absorption of gamma-ray photons is mainly due to the Compton effect, while for gamma-ray photons of low energies their absorption is due to the photoelectric effect. For the gamma-ray photons with energies above 1.02 MeV (2x0.511 MeV, the rest mass of the electron and positron), their absorption is due to the pair production effect.

In the case that a gamma-ray photon with energy E_o falls onto the scintillation crystal, NaI(Tl) if photoelectric effect is taking place, then photoelectrons are emitted with energy $E_p = E_o - b$, where b is the binding energy of electrons in the K, L, M., ... shells in the atoms of Na and I. b_K is 1.072 keV for Na, 33.170 keV for I and 85.531 keV for Tl. That energy is attributed in the form of Roentgen radiation photons during the cascades and the filling of shells of atoms. These Roentgen photons might be considered as prompt with the photoelectric effect . These Roentgen photons have two alternative possibilities. The first one and mainly dominant is to be absorbed in the crystal. The second is to escape. In the case of absorption, the fluorescent light photons that are emitted in the crystal matter and excite the PM photocathode have energy proportional to E_o. If they escape, the fluorescent light photons have energy proportional to E_{es} and

$$E_{es}=E_o-\Delta b \tag{1}$$

In the case of NaI (crystal), it is

$$E_{es}=E_o-28\ \text{keV} \tag{2}$$

The peak due to the gamma-ray photons with energy E_o is called photopeak. The peak due to the escape of Roentgen photons with energy E_{es} is called escaped peak of the photoelectric effect. This escape peak of the photoelectric effect is too weak and not easily observed in the ordinary crystal.

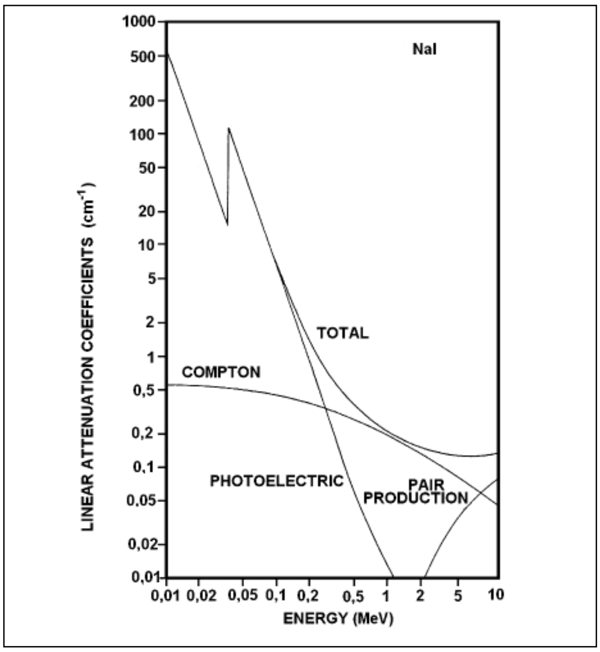

Fig. 1. Mass absorption coefficient of gamma-ray photons for a scintillation detector, NaI(Tl) as a function of their energy.

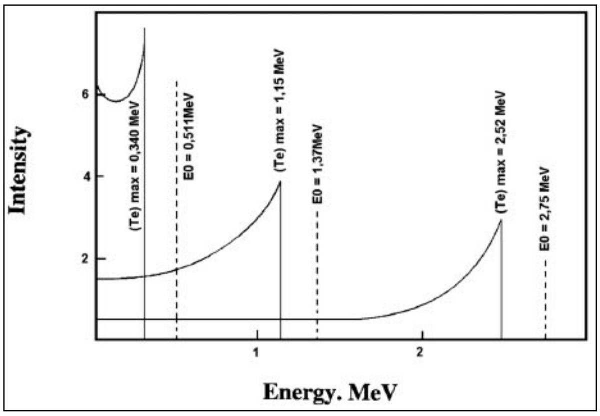

Fig. 2. Distribution of Compton electrons for the marked energies of the gamma-ray photons.

The electrons produced by the Compton effect have continuous distribution for their energy in the gamma-ray spectrum. The energy of these electrons is given by

$$T_e = E_o\,[1–1/(1+(1–\cos\theta)\,E_o/0.511)] \qquad (3)$$

Where θ is the angle of scattered photons. So, the energy of electrons, T_e is varying between 0 and $(T_e)_{max}$ which is

$$(T_e)_{max} = E_o/(1+0.511/2E_o) \qquad (4)$$

Where $\theta = 0^0$ in Eq. (3)

The theoretical distribution of electron energy due to Compton effect is shown in Fig. 2. The distribution of electron energy is appeared in the gamma-ray spectrum as “Compton edge”.

As the Compton effect is also taking place in the materials surrounding the scintillation crystal NaI(Tl), while the scattered photons in these interactions have “supplementary” energy distributions of those of Compton electrons , it is concluded that in the gamma-ray spectrum the peak of the backscattered photons due to Compton effect by the surrounding materials of the crystal will be appeared with energy $(E_{bs})_{min}$ as

$$(E_{bs})_{min} = E_o/(1+2E_o/0.511) \qquad (5)$$

Where $\theta = 180^o$ in Eq (3)

The gamma-ray spectrum of ^{137}Cs is shown in Fig. 3. The photopeak due to $E_o = 0.662$ MeV, the Compton edge $(T_e)_{max} = 0.477$ MeV and the backscattering Compton peak $(E_{bs})_{min} = 0.184$ MeV. In the case that the gamma-ray photon energy E_o is greater than 1.02 MeV, it is possible for pair production to be taken place. The two charged particles, electron and positron of pair production have total kinetic energy E_o - 1.02 Me V. Positron of pair production is thermalized in the NaI(Tl) crystal in about 10^{-11} s. Finally, it annihilates by an electron by producing two photons of 0.511 MeV in energy. These two photons could be absorbed in the crystal material and the total energy deposited in the crystal is equal to E_o. It is also possible for one of the photons to be absorbed and the other to be escaped, or both the photons to be escaped.

As the pair production effect is possible to take place in the materials surrounding the scintillation crystal NaI(Tl), two photons of 0.511 MeV to be escaped. So, in the case that $E_o > 1.02$ MeV, the escaping peak of 0.511 MeV and the double escaping peak of 1.02 MeV will be appeared in the gamma-ray spectrum. In the case that $E_o >> 1.02$ MeV, except the Compton edge and the backscattering Compton peak, three other peaks will be appeared in the gamma-ray spectrum:

i) Eo
ii) Eo – 0.511 MeV single escaping
iii) Eo – 1.02 MeV double escaping

The fact that the photons will escape or not, and consequently the above peaks will be appeared strong or not, depends on the crystal size.

Figure 4 shows the gamma-ray spectrum of ^{24}Na which

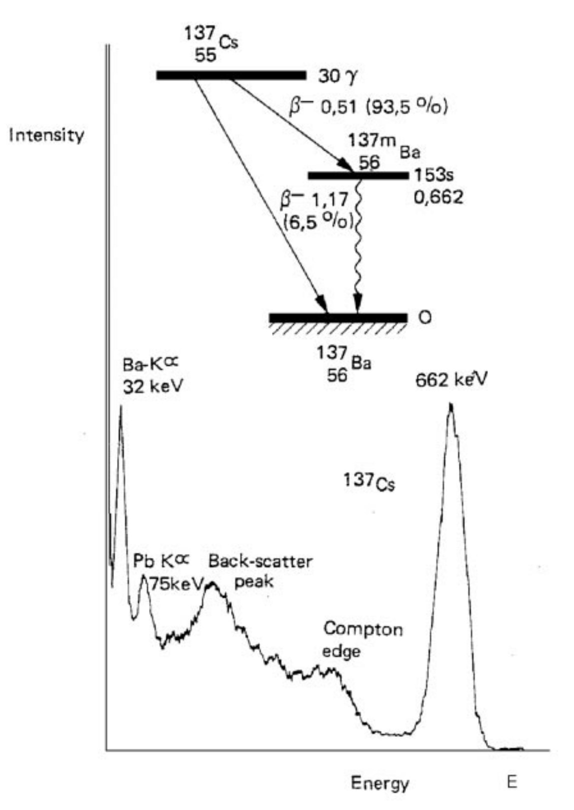

Fig. 3. Gamma-ray spectrum of ^{137}Cs obtained by a scintillation detector, NaI(Tl) and showing the photopeak of 662 keV, the Compton edge of 477 keV and the backscattering Compton peak of 184 keV.

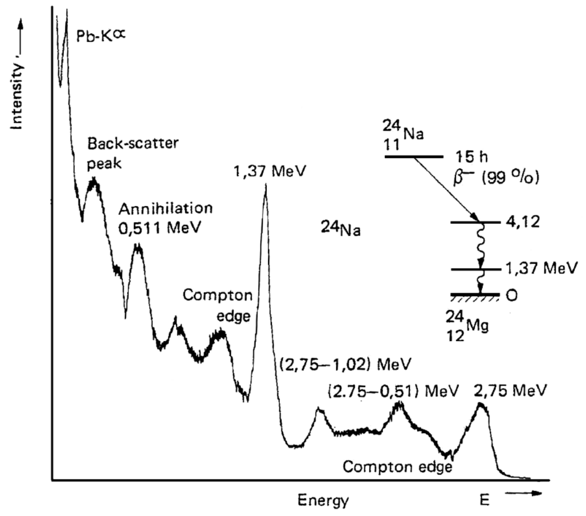

Fig. 4. Gamma-ray spectrum of ^{24}Na obtained by a scintillation detector, NaI(Tl) and showing the 511 keV annihilation peak , the 1.37 MeV and 2.75 MeV gamma-ray photon peaks and the single and double escaping peaks.

emits in a decay two gamma-ray photons with energies 2.75 MeV and 1.37 MeV, respectively. So, except the photopeaks and other peaks, the single escaping of 0.511 MeV and the double escaping of 1.02 MeV as well as the peak of gamma-ray photons of 0.511 MeV are shown in the gamma-ray spectrum of Fig. 4. Additionally, in the gamma-ray spectra there are the following peaks:

i) Peaks due to Roentgen radiation of the material surrounding the crystal, e.g. Pb (Figures 3 and 4),
ii) Peaks due to Roentgen radiation of the decay product of a nucleus if this is an alpha- or beta-emitter, e.g Ba for the case of ^{137}Cs (Fig. 3),
iii) Sum peaks. These peaks are due to the possibility of two accidental photons or two prompt photons to be absorbed simultaneously in the crystal or not. The latter case is rather due to the appropriate geometry of crystal – source of radiation. For example, if a source of ^{60}Co is set in a well type NaI(Tl) crystal, then the two different gamma-ray photons with energies of 1.17 MeV and 1.33 MeV, respectively, is possible to be absorbed simultaneously in the crystal and a sum peak of 2.5 MeV will be appeared in the gamma-ray spectrum as shown in Fig. 5, together with the gamma-ray photon peaks of 1.17 MeV and 1.33 MeV.

The scintillation detector is subject in various statistical fluctuations resulting that the total efficiency and its background to be depended on the measuring conditions. The high voltage bias supply and the pulse height of the low discriminator to be selected appropriately, so that, the measuring system to have high efficiency, E and low background, B, that means, the figure-of-merit (FOM), E^2/B to be maximum, where E is the efficiency counting

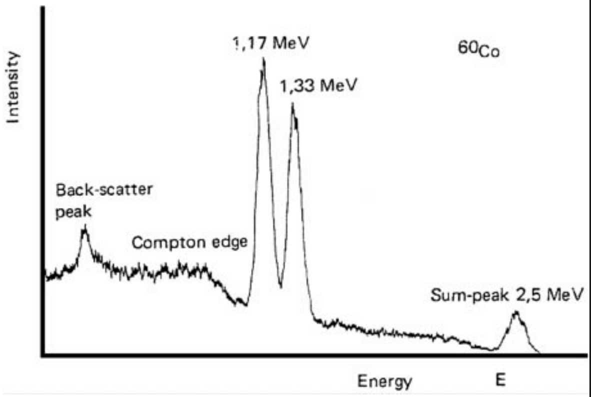

Fig. 5. Gamma-ray spectrum of ^{60}Co obtained with a well type scintillation detector, NaI(Tl) and showing the 1.17 MeV and 1.33 MeV gamma-ray photon peaks and the sum peak of 2.5 MeV.

rate, i.e. the ratio of net count rate to the real activity of source of radiation.

The output electronic pulses of the spectroscopy linear amplifier due to the detection of gamma-ray photons are having different heights from 0 to 10 V and producing corresponding peaks in the gamma-ray spectrum. The width of a gamma-ray photon peak is due to:

i) The energy dispersion in the detector crystal,
ii) The natural width of the gamma-ray peak which is of the order of a keV (0.001 MeV). This is due to the uncertainty of the excited state of the nucleus when decays according to the Heisenberg principle, $\Delta E \, \Delta t >= h$,
iii) The noise introducing by the electronic devices of the gamma-ray spectroscopy system.

The resolution of the scintillation detector, δ is determined by the equation

$$\delta = \Delta E/E \; 100\,\% \qquad (6)$$

Where ΔE is the full width at the half maximum of the gamma-ray peak (photopeak), FWHM . It is evident that the best resolution means low value of δ.

The factors that affect the resolution of the scintillation detector NaI(Tl) are due to:

i) light dispersion in the NaI(Tl) crystal,
ii) light dispersion in the photocathode of photomultiplier, PM,
iii) electron production in the photocathode of PM,
iv) electron collection by the first dynode,
v) electron multiplication successively by the dynodes.

Therefore, the resolution of the scintillation detector, δ depends on:

i) The quality of the scintillation crystal and the photomultiplier, PM,
ii) The energy of the gamma-ray photons. The dependency of the resolution, δ on the energy of the gamma-ray photon there is an empirical equation

$$\delta^2 = a+b/E \qquad (7)$$

Where a, b are constants, i.e. $a = 1.11 \times 10^{-3}$ and $b = 4.06 \times 10^{-3}$.

iii) the high voltage supply in the PM through the spectroscopy preamplifier.

A best resolution for a scintillation detector NaI(Tl) is that of 6 %, while the resolution of 8 % is considered as reference for the gamma-ray photons of $E_o = 0.662$ MeV of ^{137}Cs.

In the gamma-ray spectroscopy system for analyzing the gamma-ray photon peaks in the spectra, a single channel analyzer, SCA or a multichannel analyzer, MCA with a PCA card or a picospec digital multichannel analyzer, DMCA-pro-card with 1024 or 4096 channels (Target, 2000) is used.

2. SOLID STATE (SEMICONDUCTOR DIODE) DETECTORS

(i) Ge detectors

Germanium, Ge detectors are semiconductor diodes having a p-i-n structure in which the intrinsic (i) region is sensitive to ionizing radiation particularly X-ray and gamma-rays. Under reverse bias, an electric field extends across the intrinsic or depleted region. When, for example, gamma-ray photons interact with the material within the depleted volume of the detector, charge carriers (holes (+) and electrons (-) are produced and are swept by the electric field to the p and n electrodes. This charge, which is in proportion to the energy deposited in the detector by the incoming gamma-ray photon, is converted into a voltage pulse by an integral charge sensitive preamplifier.

Because Germanium has relatively low band gap, these detectors must be cooled in order to reduce the thermal generation of charge carriers (thus reverse leakage current) to an acceptable level. Otherwise, leakage current of about 0.01 nA induced noise destroys the energy resolution of the detector. Liquid nitrogen, which has a temperature of 77° K is the common cooling medium for such detectors. The detector is mounted in a vacuum chamber which is attached to or inserted into an LN_2 Dewar. The sensitive detector surfaces are thus protected from moisture and condensable contaminants. By use of both p-type and n-type Germanium, there are used diffused, implanted, and barrier contacts.

The main advantage of Ge detectors against the scintillation detectors, NaI(Tl) used for detection and measurement in the gamma ray spectroscopy is the resolution (Fig. 6). The resolution of Ge detectors is one order of magnitude higher than that of the scintillation detectors, NaI(Tl). This is due to the fact that the energy needed to produce a pair of a hole (+) and an electron (-) is just about 2 eV. For example, the full width at half maximum, FWHM of the 0.662 MeV photopeak of ^{137}Cs is

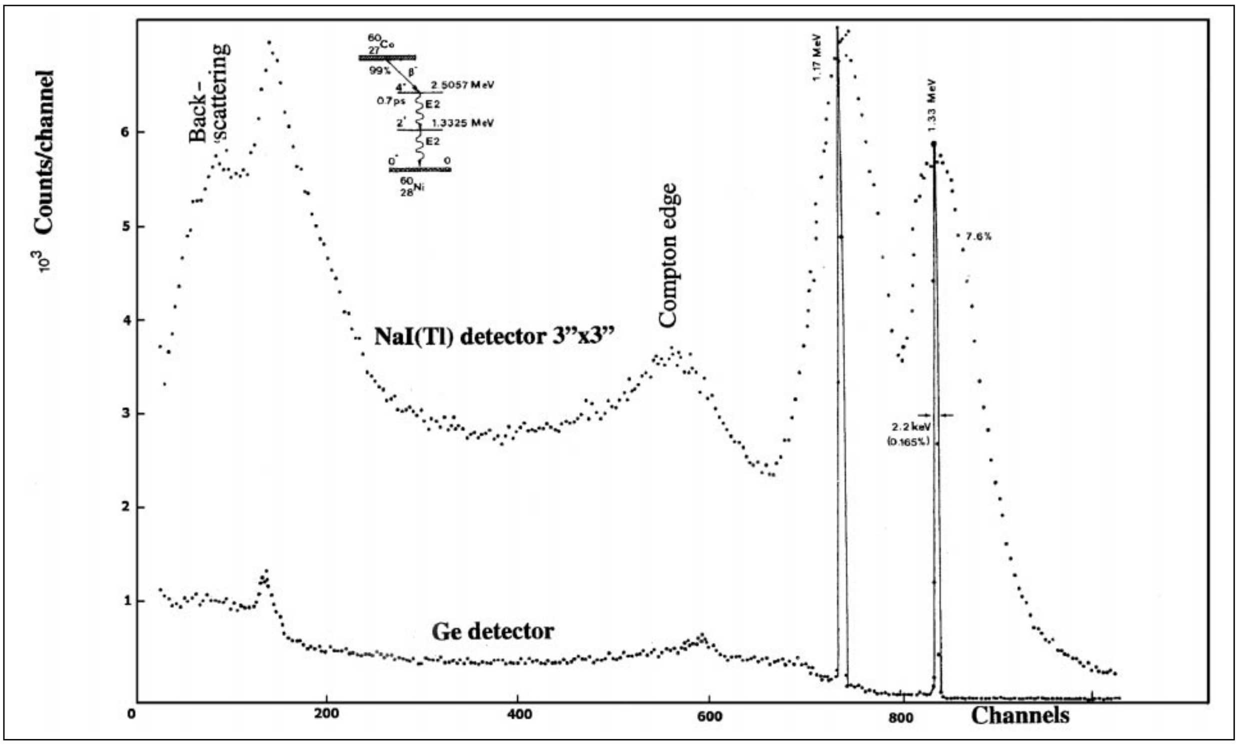

Fig. 6. Gamma-ray spectra of ^{60}Co obtained by a Ge detector and a scintillation detector, NaI(Tl), respectively, for comparison and distinguished the resolution of the detectors.

about 0.002 MeV (2 keV) or less for Ge detectors. This equivalent with a resolution value of 0.002/0.662 = 0.3 %.

The specific characteristics of Ge detectors are the following:

(i) the resolution which is as low as 1.8 keV at 1.33 MeV gamma-ray photons of ^{60}Co
(ii) the peak-to-Compton ratio, P/C which is as high as 70 : 1. The peak-to-Compton ratio, P/C of Ge detector is the mean height of Compton edge in the gamma-ray spectrum of ^{60}Co (1.040 to 1.096 MeV) to the height of photopeak of 1.33 MeV of ^{60}Co.
(iii) the efficiency which is as high as 45 %. The efficiency of Ge detector is the relative efficiency of a photopeak, i.e. the area under the photopeak (counting rate) of Ge detector compared with the respective area of the photopeak of a scintillation detector, NaI(Tl) 3"x3" in size of crystal for the gamma-ray photons of 1.33 MeV of ^{60}Co with the source of radiation set in a 25 cm distance from the end cap of the detector. Figure 7 shows a comparison in the efficiency between two typical Ge detectors of p-type and n-type.

To increase the efficiency of gamma spectroscopy system linked by a Ge detector, the system is surrounded by an Anti-Compton, suppression system which operates as a mantle of shielding and consisted of a series of scintillation detectors, NaI(Tl) with large volumes appropriately mounted to reject the background counts.

(ii) Silicon detectors

Si(Li) detectors cover the energy range mainly of X-ray photons and of course the low energy gamma-ray photons from a few hundred eV to 50 keV or so. These are surface barrier lithium drifted detectors.

The specific characteristics of the silicon detectors are:

(i) the sensitive area ranging from 12.5 up to 200 mm^2
(ii) the depletion depth ranging from 2 to 5 mm
(iii) the Be window about 25μm thick
(iv) the resolution (full width at half maximum, FWHM). This is considered for α-rays of ^{241}Am with energy 5.486 MeV (85 %), ranging from 12 to 42 keV, averaging 20 keV, for β-rays of ^{137}Cs with energy maximum 0.514 MeV (94 %) averaging 18 keV, for γ-rays of 0.122 MeV (85.6 %) of ^{57}Co ranging from 150 to 200 eV and for X-rays of 5.9 keV of ^{55}Fe averaging to 250 eV.

3. IN SITU-GAMMA-RAY SPECTROMETRY

The term *in situ* is taken from Latin and translates to "in the original place". Thus, given a site where radioactivity and radiation levels are under investigation, the term "in situ gamma-ray spectrometry" implies that a spectrum of the ambient gamma-ray flux would be collected at the site and analyzed, principally to identify and quantify the radionuclides present. Sometimes, a less academic term is used, i.e. "field gamma-ray spectrometry", which implying that one is in the outdoor environment.

The technique of *in situ* gamma-ray spectrometry had its origin during the time of atmospheric nuclear weapons testing where it was found to provide quick, reliable information on the components of the outdoor radiation environment. It provided a means to separate natural background radiation from man-made sources of radiation and give quantitative results. Over the years, it has been employed by various groups for assessing sources of radiation in the environment not only through group based detectors, but with aircraft systems as well. It proved particularly useful following the Chernobyl accident and was employed by a number of Laboratories in Europe. It should prove adaptable to site assessments in the current era of environmental restoration.

The power in the technique of *in situ* gamma-ray spectrometry lies in the fact that a detector placed over a ground surface measures gamma radiation from radioactive sources over an area of several hundred square meters. As an example of the effective ground area being measured by a detector at 1 m above the ground, Figure 8 shows the relative contribution to the fluence from different rings of ground area about the detector for a typical source of fallout ^{137}Cs (gamma rays energy of 0.662 MeV in the environment. The "field of view" for the detector would be larger for higher energy sources closer to the soil surface. In contrast, a soil sample would represent an area of but a few tens or hundreds of square centimeters. In practice, an effective characterization of a site would involve *in situ* gamma-ray spectrometry in conjunction with soil sampling. As part of an overall program, *in situ* gamma-ray spectrometry provides a means to assess the degree of contamination in areas during the course of operations in the field, thus guiding the investigator on where to collect samples. It can also substantially reduce the number of samples that need to be collected and subsequently analyzed.

Some of the limitations of *in situ* gamma-ray spectrometry need to be pointed out from the start. Due to the nature of radiation transport through matter (the soil and air), it is for the most part limited to the measurement of gamma emitters and, to some extent, X-ray emitters. Even so, the attenuation properties of soil are such that buried sources are not likely to be detected with measurements performed above ground. At 0.662 MeV, 30 cm of soil will cut out about the 97 % of the primary flux from a buried point source. Of course, a detector can be lowered into a bore hole for measuring a buried source and techniques applied for interpreting a collected gamma-ray spectrum. While this constitutes an *in situ* measurement in the broad sense of the term, it will not be addressed in this article. We will instead limit the subject area to measurements performed near ground level, where the distribution of the source of radiation is expected to be spread out over a fairly large area and where the source of radiation is at or near the surface of the soil.

Although measurements can be conducted with scintillation detectors NaI(Tl), as they were in the 60s, the energy resolution of solid state Ge detectors and the fact that they are available with efficiencies as great as that of a 3"x3" NaI(Tl) detector make them the detectors of choice. As with any counting system, the size of the detector that is needed is related to the source strength, the counting time, and the desired statistical counting error. For typical environmental radiation fields, a detector with a quoted 25 % efficiency would be large enough to give a 5 % (1σ) counting error for natural gamma emitters given a 1 hour counting time. A quick 10 minute counting time would be sufficient to provide lower limits of detection on the order of 100 Bq m^{-2} for many common fission product radionuclides residing at the surface of the soil. Higher sensitivity and/or reduced counting times can be achieved with larger detectors. Depending upon the application, a smaller detector might actually be a better choice in order to reduce counting time when making measurements in highly contaminated areas.

Another consideration is the choice between a p-type and n-type Ge detector regarding the crystal. For applications that involve the measurement of low energy gamma rays, such as from ^{241}Am (59.5 keV), the n-type detector has better sensitivity (Fig. 7). Older lithium drifted Ge detectors can function perfectly well, however, the fact that intrinsic or high-purity Ge detectors can warm up without damage makes them the most suitable for field measurements. Quality Ge detectors can be expected to have resolutions of 2 keV or better at 1.332 MeV. Better energy resolution allows a greater separation of two peaks that are close in energy. Also, each individual peak is narrower and therefore lower statistical counting errors are achieved since there is less continuum counts under the peak. Modern Ge detectors are equipped with built-in

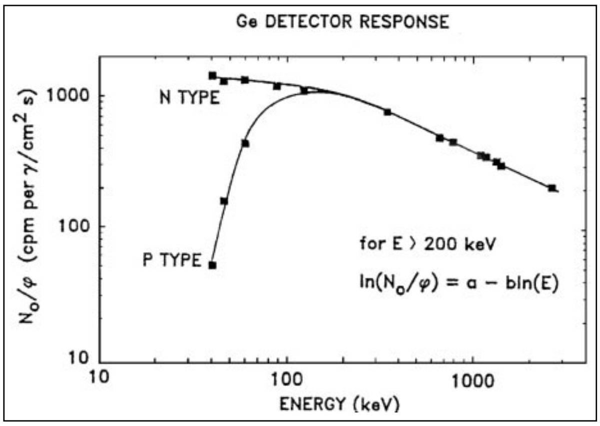

Fig. 7. Comparison in the efficiency between the p-type and n-type Ge detectors.

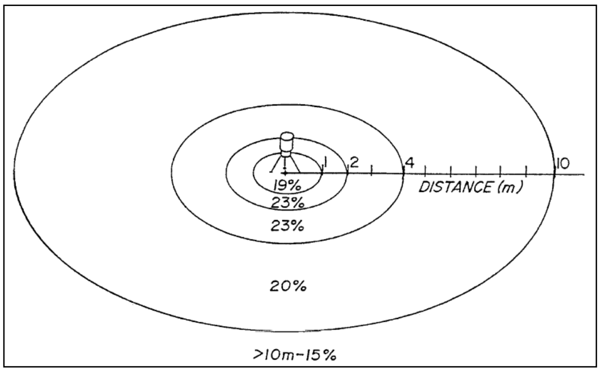

Fig. 8. Contribution to total 0.662 MeV primary gamma-ray photon flux at 1 m above ground for a typical ^{137}Cs source distribution.

preamplifiers. For field measurements where battery power is used, it is important to specify a low-power preamplifier when ordering a detector. This will extend the operational time in the filed since the preamplifier is a principal draw on power.

Although measurements in the field can be performed with a Ge detector in almost any type cryostat-dewar configuration, performance and ease of handling is best achieved with a small dewar (1 to 2 liters) that can be tripod mounted with the detector facing down, For convenience, a 24 hour liquid nitrogen holding time is desirable as this requires filling only once a day, although it may be safer to maintain a twice a day schedule. Ge detectors can also be cooled with electrically powered apparatus, however, this may not be as convenient for field measurements with battery-powered equipment. It is possible to mate small dewars to automatic filling apparatus in the laboratory or to larger gravity-feed storage dewars. As for orientation, a detector facing sideways (the axis of symmetry parallel to the ground) should be avoided because it introduces complicated angular corrections.

A Ge detector can be connected to a full laboratory instrumentation package that is carried in a van and powered with a motor generator of battery bank. This was the norm in the early days of field gamma-ray spectrometry. Today, it is far more convenient to make use of portable battery-powered analyzers that are specifically designed for field measurements. These units not only serve as multichannel pulse height analyzers, but they also provide preamplifier power and high voltage to the detector. This type of analyzer with the Ge detector and a set of connecting cables is all that is needed for a complete gamma-ray spectrometry system. Also available now are portable laptop computer-based systems which have the capability to run more sophisticated analysis programs. An overnight recharge is generally sufficient to provide 8 hours of operational time in the field for either the full-function analyzer ot the computer-based system. In the latter case, the computer can be shut off to conserve its own limited battery supply, while a gamma-ray spectrum continues to collect in the memory of the analyzer base unit.

One additional component needed for practical application of *in situ* gamma-ray spectrometry is a method of spectrum storage since it is likely that many spectra will be collected during the course of a site investigation. Some portable analyzers have built-in compact disk (CD) or mini cassette data storage capability, while others rely on an external portable audio digital video (DVD) or audio cassette recorder. The PC-based systems have the advantage of being able to store numerous gamma-ray spectra directly on the internal disk drive.

The ideal site for collecting a gamma-ray spectrum would be a large (20 m diameter or more) flat, open area with little or no natural or man-made obstructions. The area to be measured can be scanned first with a suitably sensitive survey meter to insure that there is rough uniformity in background dose rate. It is also possible to move the Ge detector about and obtain quick gamma-ray spectra (1 to 5 minutes), observing that a full absorption peak counting rate does not change substantially for a radionuclide under study. For measuring fallout radionuclides that were deposited. In the past, the land should not have been disturbed by plowing or by wind or water erosion. For standard measurements, the Ge detector should be at height of 1 m above the ground, although a variation of as much as 50 cm in either direction will not introduce a large error. While collecting a gamma-ray spectrum, personnel should stand away from the detector. Since the operator may wish to examine the gamma-ray spectrum during collection, it is best to position the analyzer away from the detector using cable lengths of a few meters.

As with any gamma-ray spectrometry system, the amplifier gain and analyzer conversion rate must be adjusted to provide a gamma-ray spectrum in the energy region of interest (ROI). For environmental gamma radiation, this would be from 50 keV out to 2.615 MeV, normally the highest energy gamma line seen. For a 4096 channel analyzer, a conversion rate of 1 keV per channel will suffice in most cases, although 0.5 keV per channel may be desirable in certain situations to take advantage of the higher energy resolution of the detector at low gamma-ray photon energies.

4. INFERRED QUANTITIES

(i) Concentration in soil

The fundamental quantities used for *in situ* gamma-ray spectrometry include full absorption peak counting rate (N), fluence rate (Φ), and radiation source activity (A). In practice, one would like a single factor to convert from the measured peak counting rate, N in a gamma-ray spectrum to the radiation source activity level in the soil or the dose rate in air. This factor can be calculated from three separately determined terms as follows:

$$N_f / A = (N_f / N_o)\ (N_o / \Phi)\ (\Phi / A) \qquad (8)$$

Where N_f /A is the full absorption peak counting rate at some energy, E, from a gamma-ray transition for a particular radioisotope per unit activity of that radioisotope in the soil;

N_o/Φ is the full absorption peak counting rate per unit fluence rate for a plane parallel beam of gamma-ray photons at energy, E, that is normal to the detector face;

N_f/N_o is the correction factor for the detector response at energy, E, to account for the fact that the fluence from an extended radiation source in the environment will not be normal to the detector face but rather distributed across some range in angles; and Φ/A is the fluence rate at energy, E, from gamma-ray photons arriving at the detector unscattered due to a gamma-ray transition for a parallel radioisotope per unit activity of that radioisotope in the soil.

The fluence rate, Φ(E) at the detector is given by the expression

$$\Phi(E) = R(E) / 4\pi x^2 \qquad (9)$$

where R(E) is the gamma-ray emission rate at that energy and x is the source to detector distance. The attenuation effect of the source encapsulation should be taken into account along with that of the air between the source and detector, particularly for low energy gamma rays and large values of x.

The term N_o/Φ is purely detector dependent, while the term N_f/N_o is dependent on both the detector characteristics and the radiation source geometry. The term Φ/A is not dependent on the detector characteristics but rather on the radiation source distribution in the soil.

Having determined the three separate quantities, N_o / Φ, N_f / N_o, and Φ / A, their product yields the desired conversion factor, N_f / A. for radionuclides uniformly distributed with depth in the soil ($\alpha / p = 0$, where α is the inverse of the relaxation length, (cm), p is the soil density (g cm^{-3}), the term A is in units of activity per unit mass. As such, there is no need to determine the soil density.

Although the assumption of a uniform profile in the soil for natural gamma emitters is generally safe, unusual situations where there is markedly different soil strata of varying radionuclide concentration may produce anomalous results. This situation been used. Also, evaluations of the ^{238}U series must be done with the awareness that ^{222}Rn escapes from the soil and that the important gamma-emitting decay products, ^{214}Pb and ^{214}Bi, may not be in equilibrium with ^{226}Ra in the soil. In fact, there may be a measurable contribution to the fluence rate at 1 m above the soil from the decay products in the air, particularly under atmospheric inversion conditions. Disequilibrium is also possible for the ^{232}Th series due to the exhalation of ^{220}Rn (thoron), although this is less likely to be as severe due to its relatively short half-life.

Another effect that may interfere with the interpretation of a gamma-ray spectrum is that of ^{222}Rn decay products scavenging during precipitation. In this situation the ^{214}Pb and ^{214}Bi assume a surface radiation source distribution that can considerably alter the flux and dose rate. For this reason (and to keep people and equipment dry!), it is best to avoid measurements during and for about 2 to 3 hours following rain.

It is possible to consider a fallout radionuclide product as having a uniform profile if it is deeply distributed or has been mixed through soil cultivation. Depending upon the radiation source gamma-ray photon energy, plowing to depth of 15 to 30 cm essentially accomplishes this. Although the distribution does not extend to infinity in a situation such as this, in terms of the total gamma-ray photon flux seen above ground, it is effectively infinite in depth. For in situ applications such as this, the concentration that is measured can be considered as representative of the surface soil.

(ii) Deposition/inventory

For radionuclides that are exponentially distributed with depth ($\alpha/p>0$), the “term A is in units of activity per unit area. Although the results of analyses of environmental samples are frequently reported in terms of concentration, the fundamental quantity that is of most use for assessing fallout radionuclide products is the deposition (sometimes referred to as deposition density or inventory). Whereas the deposition remains a constant, the concentration of a fallout radionuclide product will vary depending upon the depth distribution. To illustrate this point, consider a radionuclide such as ^{137}Cs that was deposited in an area 45 years ago from atmospheric nuclear weapons testing. Where the surface soil has retained it, a sample down to 5 cm will yield some concentration, x . On an adjacent strip of land that was plowed deeply, the same sampling protocol will yield a concentration of perhaps only 0.2x. Obviously, this would be a flawed scheme for investigating a potential local radiation source of contamination. Instead, consider a soil core that was taken down to 30 cm. The measured concentration of an aliquot of this sample should be multiplied by the entire sample mass to give the total activity in the core and then divided by the sample area to give activity per unit area. This would yield the same result for both sites. The only precaution is to sample to a great enough depth to collect essentially all of the deposited activity.

In order to make an accurate assessment of deposited activity with *in situ* gamma-ray spectrometry, an estimate or actual measurement of α/ρ must be made. As such, the time of deposition must be taken into account and assurances that no erosional or human activities such as plowing have disturbed the site. For fresh fallout that is dry deposited, the assumption of a surface source of radiation ($\alpha/\rho = \infty$) is generally not justified due to the effects of soil surface roughness which effectively buries the source and lowers the fluence at the detector. Wet deposition processes will also tend to distribute the fallout within the surface soil layer such that the assumption of a surface source of radiation would not be correct. Experience has shown that a more realistic assumption of α/ρ would be on the order of 1 to 10 cm^2 g^{-1}. Depending upon the degree of uncertainty that is acceptable, experimental determination of the profile may be required through soil sampling. For deposition that occurred in the past, soil sampling is generally required to obtain an accurate value of α/ρ.

In making measurements of deposition of radionuclides in the environment, one must be aware of the sensitivity of the inferred inventory to the value of α/ρ. Figure 9 shows an example of the results of a calibration for a 22 % efficient Ge detector. The conversion factor, N_f / A, is ploted as a function of the source depth constant, α/ρ, for the commonly encountered fission product radionuclide ^{137}Cs. The conversion factor is seen to change relatively little for values of $\alpha/\rho > 1$cm^2 g^{-1} (shallow source depth distribution) as compared to values of $\alpha/\rho < 1$ cm^2 g^{-1} (deep source depth distribution). In effect, the error made in inferring the source activity will not be large for a fresh deposition event even if the profile is not precisely known. Conversely, if a measurement of aged fallout is made, accurate results will only be obtained if the profile is determined by some independent means, i.e. soil sampling.

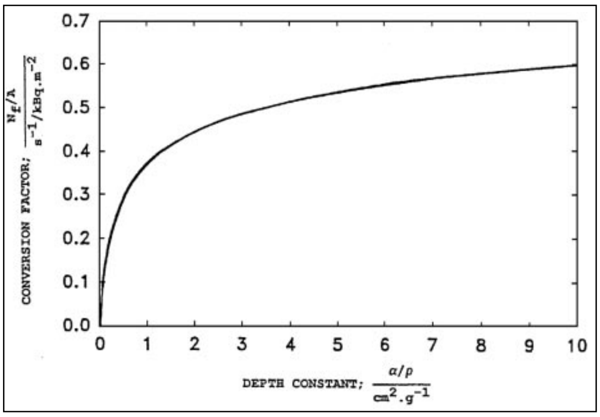

Fig. 9. Conversion factor N_f/A as a function of the source depth parameter α/ρ.

(iii) Dose rate in air

One of the most useful quantities that can be determined with *in situ* gamma-ray spectrometry is the dose rate in air (or the exposure rate) for the individual radionuclides present at a site. To do this, the results of transport calculations are used for the infinite half space geometry and the exponential source distribution. The conversion factors, I/A, exposure rate per unit activity in the soil, can be found in Beck (1972), Beck et al. (1972) and Beck (1980). One can incorporate these factors directly into the detector calibration using the relationship

$$N_f / I = (N_f / A) \,/\, (I / A) \qquad (10)$$

where N_f / I is the full absorption gamma-ray photon peak count rate per unit exposure rate for that radionuclide. The factor I / A takes into account all of the gamma rays

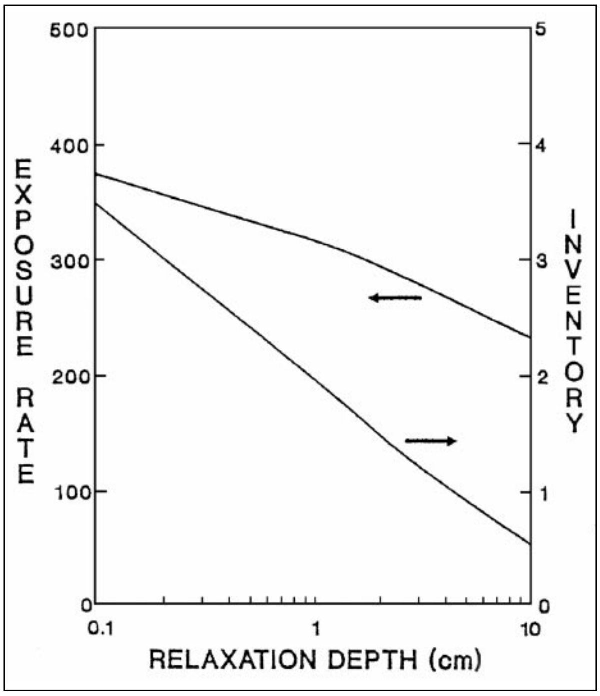

Fig. 10. Exposure rate and inventory conversion factors as a function of the relaxation length for ^{137}Cs.

emitted in the decay of that radionuclide. Therefore, one does not have to analyze every gamma-ray photon peak for that radionuclide. In practice, however, it is best to analyze more than just one peak, especially if they are well separated in energy to check agreement.

What is not obvious in this analysis is the fact that the derived quantity, N_f / I, is less sensitive to α/ρ than is N_f / A. This results from the fact that as the source distribution in the soil gets deeper, the primary flux decreases relatively rapidly compared to the scattered component. This scattered component still contributes to the dose rate. To illustrate this, Figure 10 compares these to the two calibration factors N_f / I and N_f / A as a function of the relaxation depth, α^{-1}, where the soil density $= 1.6$ g cm^{-3}. This range in depth profiles extends from a fresh deposit to one that is perhaps 30 years old. It can be seen that the exposure rate factor varies by only 50 % or so whereas the inventory factor varies by about a factor of 7. Thus, only a rough estimate of the depth profile is needed to predict the dose rate. At the same time, substantial errors can be made in the inventory estimate if the wrong depth profile is used.

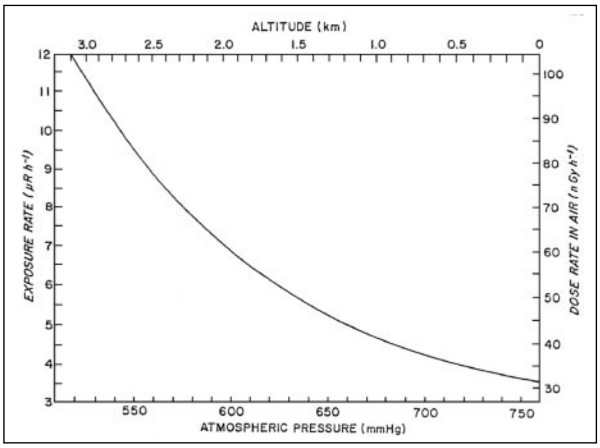

Fig. 11. Mid-latitude conversion factor for cosmic-ray dose and exposure rate as a function of altitude and atmospheric pressure.

5. RADIATION SOURCES IN THE ENVIRONMENT

(i) Natural emitters

Virtually any gamma-ray spectrum collected over soil will reveal the presence of the three primordial natural radionuclides, ^{238}U, ^{232}Th and ^{40}K. In the case of ^{238}U, detection and measurement is made through the analysis of its decay products, principally ^{214}Pb and ^{214}Bi. For ^{232}Th, the decay products ^{228}Ac, ^{228}Th and ^{208}Tl are commonly used. As mentioned previously, these radionuclides are generally distributed uniformly with depth in the soil. As such, the appropriate quantity to report is the concentration, i.e. the specific activity (Bq kg^{-1}). Since these natural radionuclides are likely to contribute substantially to the total gamma-ray flux, the exposure rate or dose rate in air is a useful quantity to report as well. The summation of all contributions to the dose rate should be made and compared to a reading from an instrument such. Table 1 lists some of the more prominent gamma-ray peaks that are seen in the gamma-ray spectrum and which are the best to analyze. As a standard practice, the conversion factors N_f/A and / or N_f/I should be computed for these gamma-ray peaks, as they almost always be used.

One characteristic of an *in situ* gamma-ray spectrum is that the continuum rises substantially at low gamma-ray energies from the absorption of scattered radiation in the air by the crystal of Ge detector. This makes it difficult to detect and analyze gamma-ray peaks below about 200 keV. For instance, the rather weak 186 keV gamma-ray peak of ^{226}Ra superimposed on this large continuum does not usually give highly precise results due to the counting error.

One cosmogenically produced radioisotope that can sometimes be seen in the gamma-ray spectra is ^{7}Be (477 keV, 53 day half-life). Since it is produced in the atmosphere and deposited on the earth's surface, it can be expected to

Table 1. Principal gamma-ray photon energies of natural radionuclides

Energy (keV)	Nuclide	Parent Series	Comments
186	226Ra	238U	low intensity, high continuum, cannot be resolved from ^{235}U peak at 185 keV
239	212Pb	232Th	strong peak, contribution from ^{224}Ra peak at 241 keV, interference from ^{214}Pb peak at 242 keV
295	214Pb	238U	generally clean peak, fairly strong
352	214Pb	238U	generally clean, strong peak
583	208Tl	232Th	generally clean, strong peak
609	214Bi	238U	strong peak, interference from 605 keV peak if ^{134}Cs is present
911	228Ac	232Th	generally clean, strong peak
965+969	228Ac	232Th	doublet, not as strong as 911 peak
1120	214Bi	238U	reasonably strong, continuum relatively low
1461	40K	-	clean, strong, only peak for this nuclide
1765	214Bi	238U	reasonably strong, continuum low
2615	208Tl	232Th	clean, strong, continuum very low

have an exponential profile like that of a typical fission product radionuclide in fallout. Due to its short half-life, it can be expected to lie close to the soil surface and thus have a high value of α/ρ.

(ii) Fallout emitters

Due to nuclear weapons testing in the atmosphere, measurable amounts of the fission product radionuclide ^{137}Cs can be seen in surface soils around the world. Also, many areas especially in Europe, showed the activation product, ^{134}Cs, along with additional amounts of ^{137}Cs from Chernobyl fallout. Other, less intense, and shorter-lived radioisotopes from Chernobyl, such as ^{125}Sb and ^{106}Ru, can be sometimes seen as well. For common fallout product radionuclides, such as ^{125}Sb and ^{106}Ru and for other radioisotopes that one would expect to encounter, it is useful to determine the conversion factor N_f /A and plot it for several different values of α/ρ. A smooth curve can be drawn through the points or a fit can be applied, as shown in Fig. 9.

For in situ applications where there is potential for inhomogeneity in the horizontal distribution of deposited activity due to sparse ground cover, accurate measurements can still be performed providing that the scale of these inhomogeneities is small in comparison to the field of view of the detector. As an example, fallout in semi-arid regions may tend to clump under scattered plants from the effects of wind blown soil. If the depth distribution of the radionuclides is approximately the same for bare ground, as well as under the plants, no correction is needed as the application of the appropriate conversion factor for that depth distribution will yield the average inventory for that site. However, it is possible that there may be two or more distinct depth profiles associated with the various ground covers in which case separate determinations must be made. The infinite half space in this circumstance can be considered a collection of subspaces, each with its own characteristic radionuclide inventory and depth profile. The conversion factor for field spectrometry is then computed as an average, weighted by the fraction of the total deposited activity associated with each ground cover.

An estimate of this can be made through selected soil sampling to determine the inventory and by measuring the fraction of the half space for each ground cover. In a strict sense, the *in situ* gamma-ray spectrum in this situation does not provide an independent measure of the deposited activity in that there is a reliance on the data provided by the soil samples. However, the average conversion factor is bounded by the range in respective values for each type of ground cover. This range may be small compared to the variation in deposition density so that the in situ gamma-ray spectrum provides a reasonably accurate average without resorting to far more extensive soil sampling. For more details on this subject, the reader is referred to Miller (1984), Miller and Helfer (1985), Helfer and Miller (1988) and Miller and Shebell (1993).

(iii) Cosmic radiation

A portion of the continuum seen in a gamma-ray spectrum obtained by a Ge detector is due to the interaction of cosmic-ray secondary radiation in the crystal of the detector. The degree of this contribution can be estimated from the count rate above the 2.615 –MeV line from ^{208}TI. Generally, it is a small fraction of the count rate due to terrestrial gamma radiation. The overall effect is to increase somewhat the error associated with the analysis of a peak in the gamma-ray spectrum. In that the continuum under that peak is slightly higher.

It is important to realize, however, that a measurement of the external dose rate will included a contribution from the cosmic-ray component. Many survey instruments have some response to cosmic radiation. If a comparison is made between a survey instrument reading and the sum of the dose rates inferred from peak analysis with a Ge detector, it must be remembered that the latter provides only the terrestrial gamma-ray component.

In general, the dose rate from cosmic radiation increases towards the earth's poles and decreases toward the equator (latitudinal effect, Papastefanou, 2007). For mid-latitudes, Fig. 11 provides a useful conversion from altitude/pressure to cosmic-ray dose rate. A reading with a pressure meter would be the preferred method with which to infer the cosmic-ray component. In place of this, a geological survey map can be used to find one's altitude. In using this chart, a limitation on its accuracy must be recognized. There are variations of a few percent with the 11-year solar cycle and somewhat smaller variations with season. During periods of maximum solar activity (as measured by sunspots for instance by Ioannidou and Papastefanou (1994) and Papastefanou and Ioannidou (2004)), the cosmic component tends to be lower, while during period of a "quit" sun it is higher. The overall uncertainty given both these spatial and temporal variations is estimated to be on the order of 10 %.

6. Epilogue

Most of us who deal with aspects of ionizing radiation in the environment are familiar with basic dose rate and or radioactive sample measurements using survey instruments portable or not. Perhaps we can recall those times where we have walked about a site with a meter in our hand and measured external / or internal radiation levels. This constitutes an *in situ* measurement in its most basic form, one which deals with a single parameter such as the exposure rate. For more information, one can take a sample from this same site, perhaps soil, and return it top the laboratory for analysis. Gamma-ray spectrometry by a Ge detector might then be employed to determine the specific radionuclides present in the samples. This could be done for strictly qualitative purposes or perhaps to convert measured concentrations of radionuclides in the samples to the exposure rate at the original site using conversion factors. The technique of *in situ* gamma-ray spectrometry combines radioelements of both of these methods for characterizing the external radiation field. By using high resolution Ge spectrometers placed over the ground, a spectrum of gamma radiation collected in the field can be used to identify radionuclides present in a qualitative manner by simply looking for the presence of peaks at characteristic energies of gamma-ray photons. At a higher level of sophistication, one can convert the measured peak count rate into some more meaningful quantity such as the concentration of these radionuclides in the soil or, in the case of deposited fallout, the activity per unit area. It is also possible to infer the contribution of each individual radionuclide to the dose rate in air.

This article will introduce one to these techniques the individual will be able to adapt the techniques to unique situations. To this end, a basic grounding in the theory is given, however short-cut methods are also presented for those who may use the techniques for approximate measurements. For more details on certain aspects, appropriate references are given.

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