The Abdus Salam
International Centre for Theoretical Physics

United Nations
Educational, Scientific and
Cultural Organization

IAEA
International Atomic Energy Agency

2064-2

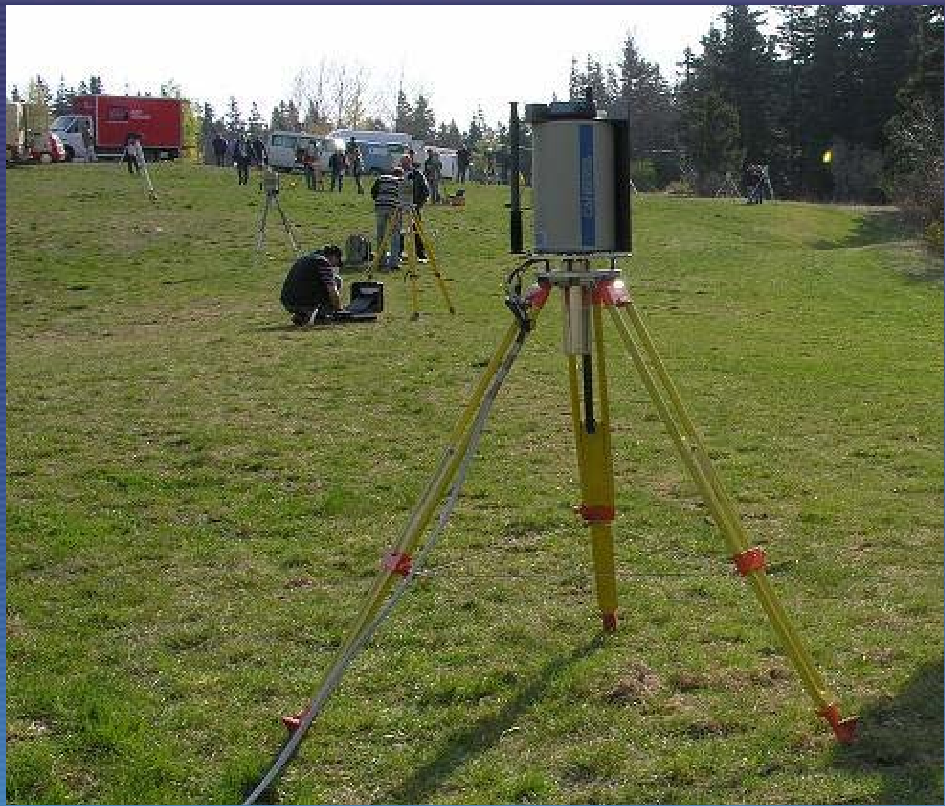
Joint ICTP/IAEA Advanced School on in-situ X-ray Fluorescence and Gamma Ray Spectrometry

26 - 30 October 2009

Different type of detectors for different applications (advantages and disadvantages)

G. Kis-Benekek
IAEA.
Seibersdorf
Austria

IN SITU GAMMA RAY-SPECTROMETRY

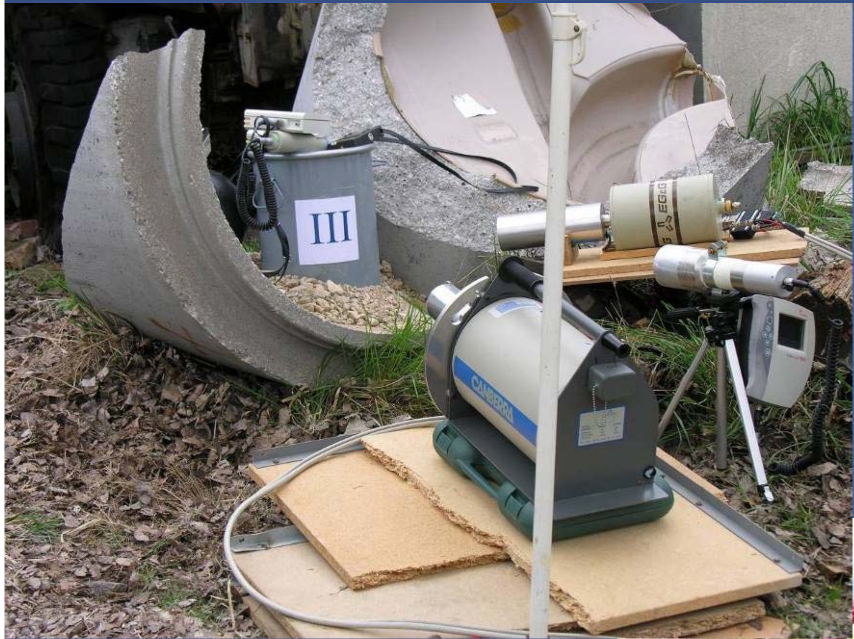


IAEA

Atoms for Peace: The First Half Century

1957–2007

IN SITU GAMMA RAY-SPECTROMETRY



IAEA
Atoms for Peace: The First Half Century
1957–2007

Applications

- Rapid identification and determination of gamma emitting radionuclides in the environment (in the field)
    - activity concentration ($Bq kg^{-1}$) or
    - deposition ($Bq m^{-2}$)
    - activity concentration ($Bq kg^{-1}$)
- Indoor radiation studies

  Analysis of power reactor plumes

NOTE: Spatial distribution of the radionuclide(s) of interest (source geometry) has to be taken into account.

Directly measureable natural radionuclides

Nuclide	E (keV)	Intensity I_γ	Comments
Be-7	477.60	10.44%	Spallation product Continuous deposition at varying rate
K-40	1460.83	10.67%	
Pb-210	46.54	4.24	Low-energy Inhomogeneous distribution Note ^{222}Rn
Ra-226	186.10	3.51	Interference with ^{235}U (185.72 keV)
Pa-231	300.07 302.65	2.47% 2.2%	Interference with ^{212}Pb (300.09 keV)
U-235	143.76 185.72	10.96% 57.2%	Interference with ^{226}Ra (186.10 keV)

Natural radionuclides measurable through daughter products

Nuclide	Gamma-emitting nuclide	E (keV)	Intensity I_γ	Comments
Ra-226	Pb-214 Bi-214	295.22 351.93 609.31 1120.29 1764.49	18.15% 35.1% 44.6% 14.7% 15.1%	Equilibrium assumed between ^{226}Ra, ^{222}Rn, ^{218}Po, ^{214}Pb, and ^{214}Bi Note ^{222}Rn
Ac-227	Th-227 Ra-223	235.97 269.46	12.3% 13.7%	Equilibrium assumed between ^{227}Ac, ^{227}Th, and ^{223}Ra Interference with ^{228}Ac at 270.24 keV
Th-228	Ra-224 Pb-212 Bi-212 Tl-208	240.99 238.63 727.33 583.19 2614.35	4.10% 43.3% 6.58% 30.4% 35.64%	Equilibrium assumed between ^{228}Th, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, and ^{208}Tl Note ^{220}Rn
Th-232	Ac-228	338.32 911.20 968.97	11.27% 25.8% 15.8%	Equilibrium assumed between ^{232}Th, ^{228}Ra, and ^{228}Ac
U-238	Th-234 Pa-234m	63.28 766.37 1001.03	4.1% 0.316% 0.839%	Equilibrium assumed between ^{238}U, ^{234}Th, and ^{234}Pa(m) Note low-energy gamma line of ^{234}Th

- **Analyte:** gamma emitting radionuclides
- **Geometry:** HPGe (or other type) gamma detector usually at 1m above the ground
- **Matrix:** Soil and air
- **Measurement time: usually 20 to 40 min**
- **MDA:** about 100 Bqm^{-2}, depending on detector efficiency, radionuclide of interest and other radionuclides present
- **Accuracy:** 10-50 % depending on calibration accuracy and environmental conditions.
- **Prerequisite:** Calibration of the gamma-ray spectrometric system for in situ measurements

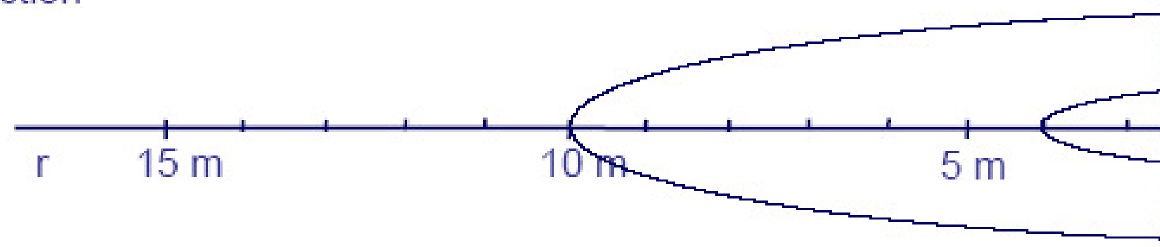
Effective field of view – Sample size

Example

photon energy	662 keV (^{137}Cs)
detector at height	h=1m
soil density	ρ=1.6 g cm^{-3}
relaxation length	L=3.0 cm

r	Fraction of total flux $\Phi(r)/\Phi$	Surface area πr^2
4 m	65%	~50 m^2
10 m	85%	~310 m^2
>10 m	15%#	~710 m^2 (at r=15m)

remaining fraction



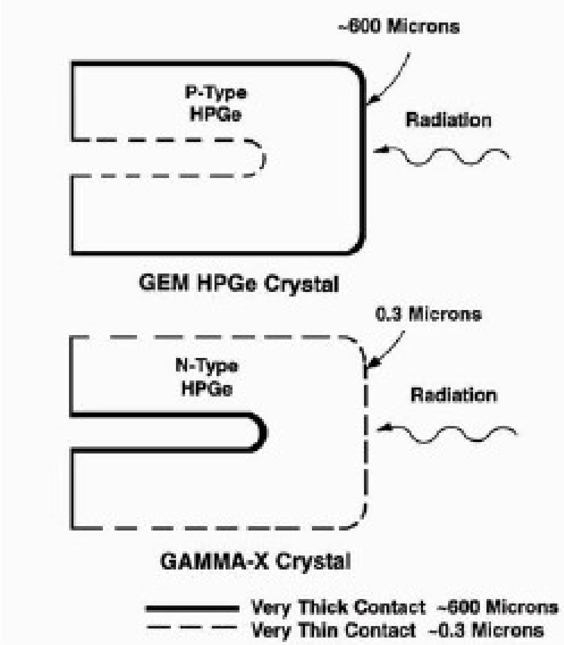
In situ / laboratory mesurements

- Shorter measurement time
- Prompt availability of results (without sampling, sample transport and preparation)
- Averaging radionuclide activity over large area
- Often large errors are observed (intercomparison exercises)
- Using laboratory results (soil profiles – depths distribution) better results can be obtained
- Based on in situ results better sampling plan can be prepared

Basic instrumentation

Detector

- HPGe (hyper-pure germanium detector)
  - p-type for Eγ > 100 keV (3-60 keV up to 3-10 MeV)
  - n-type for Eγ > 45 keV (3 keV up to 300 keV-3 MeV)
- Efficiency of 20%-50% relative to 3”x3” NaI(TI) at 1.3 MeV
- Portable (hand-held) cryostat



Electronics

- portable, battery operated, or
- lab instruments and power generator

Detector holder

- tripod, or
- trolley

IN SITU GAMMA RAY-SPECTROMETRY BASIC EQUIPMENT

QA: In-situ γ-ray spectrometry Checklist

Instrumentation

- Detector
- Electronics (MCA, Notebook PC, set of cables, power supply)
- GPS
- Dose rate meter
- Distance meter
- Camera

Supplies

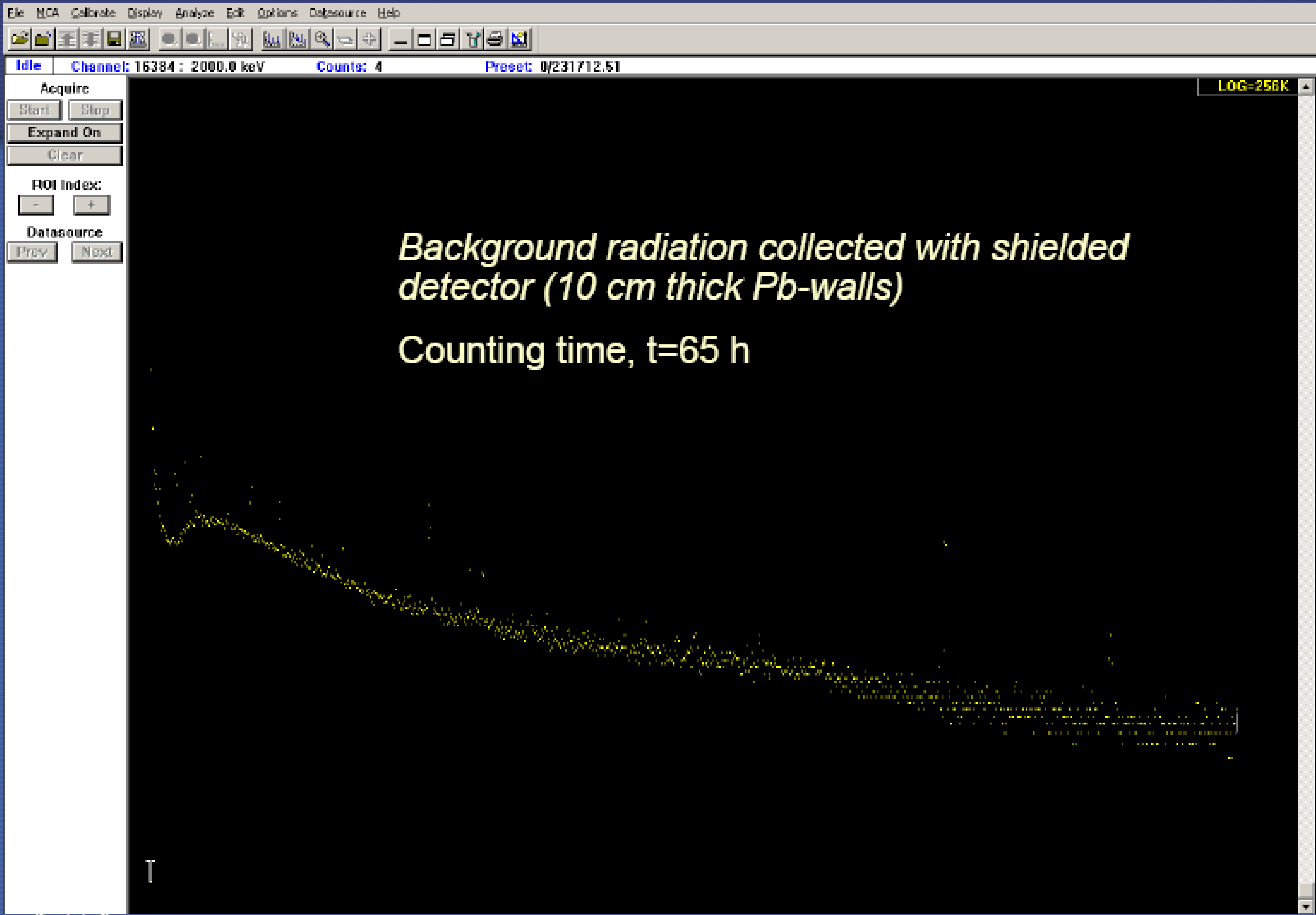
- Detector support (tripod)
- Spare batteries, Spare cables, Check sources
- Liquid nitrogen (fittings for filling)
- Protective clothes, plastic bags,
- Basic tools

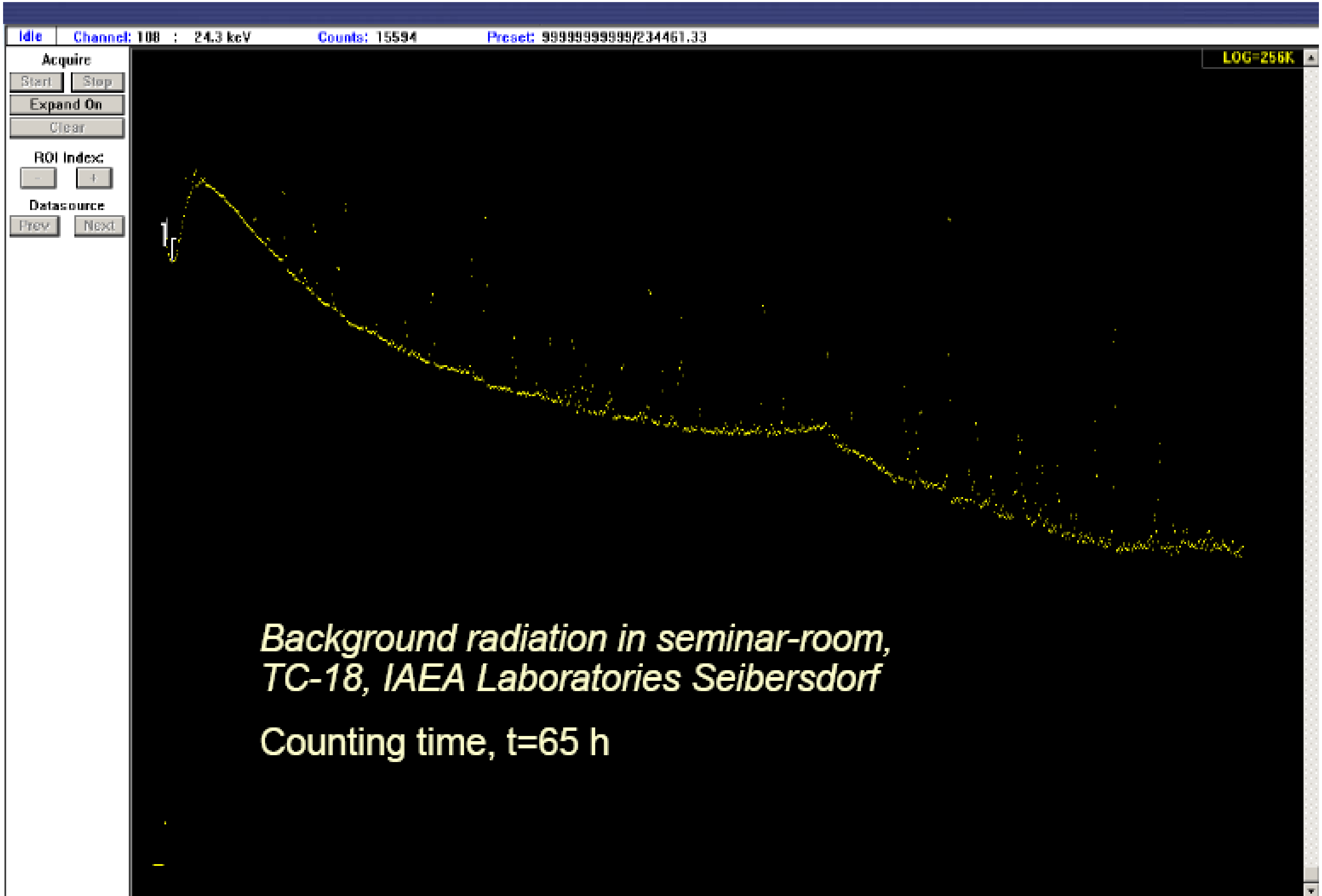
Software

- MCA and Evaluation
- Databases
- Nuclide library, conversion factors

Documentation

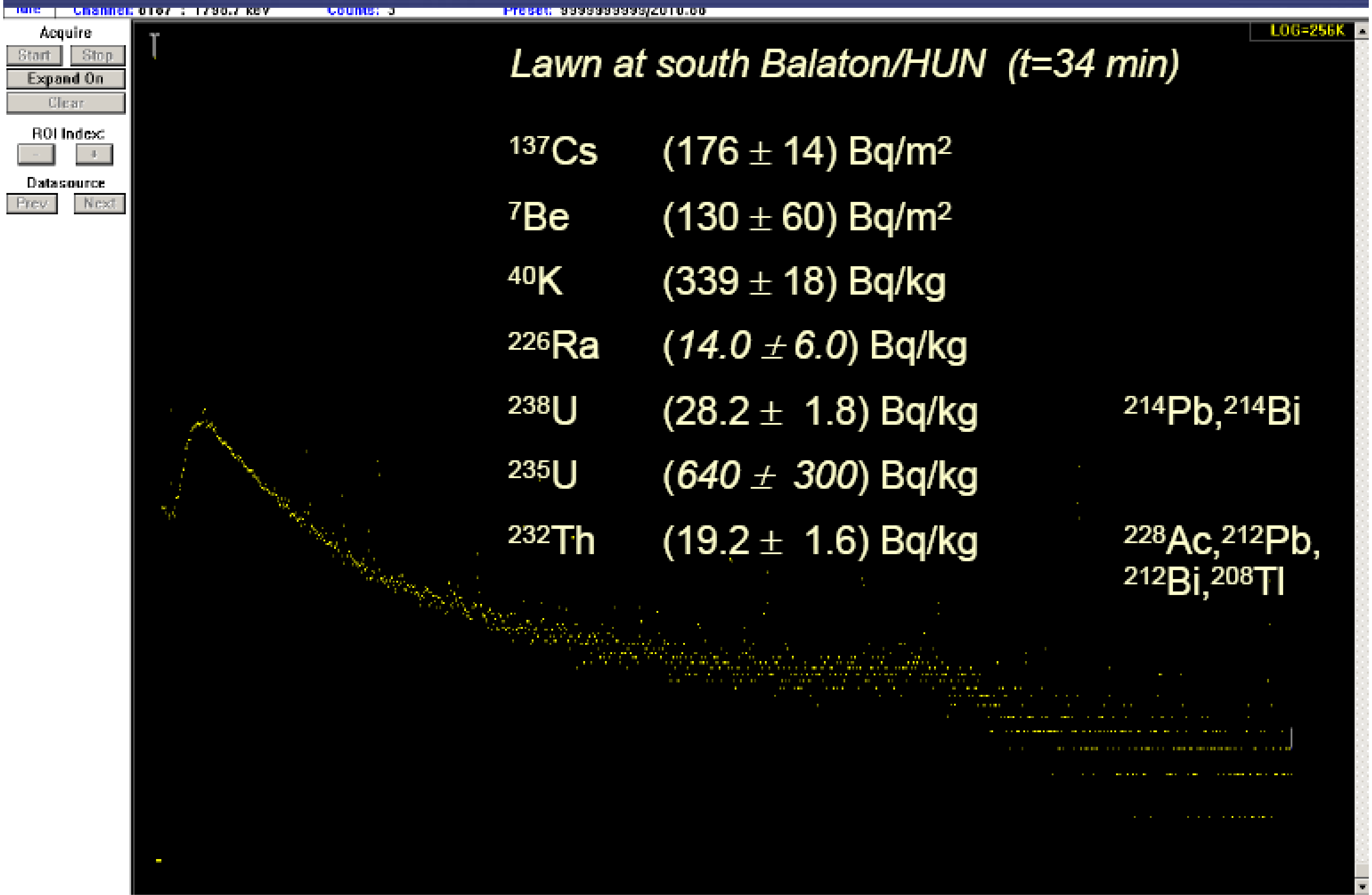
- Manuals (operation, procedures)
- Logbook
- Nuclide library tables, conversion factors
- Map


File NCA Calibrate Display Analyze Edit Options Datasource Help
Idle
Channel: 16384 : 2000.0 keV
Counts: 4
Preset: 0/231712.51
Acquire
Start
Stop
Expand On
Clear
ROI Index:
Datasource
Prev
Next
LOG=256K
Background radiation collected with shielded detector (10 cm thick Pb-walls)
Counting time, t=65 h



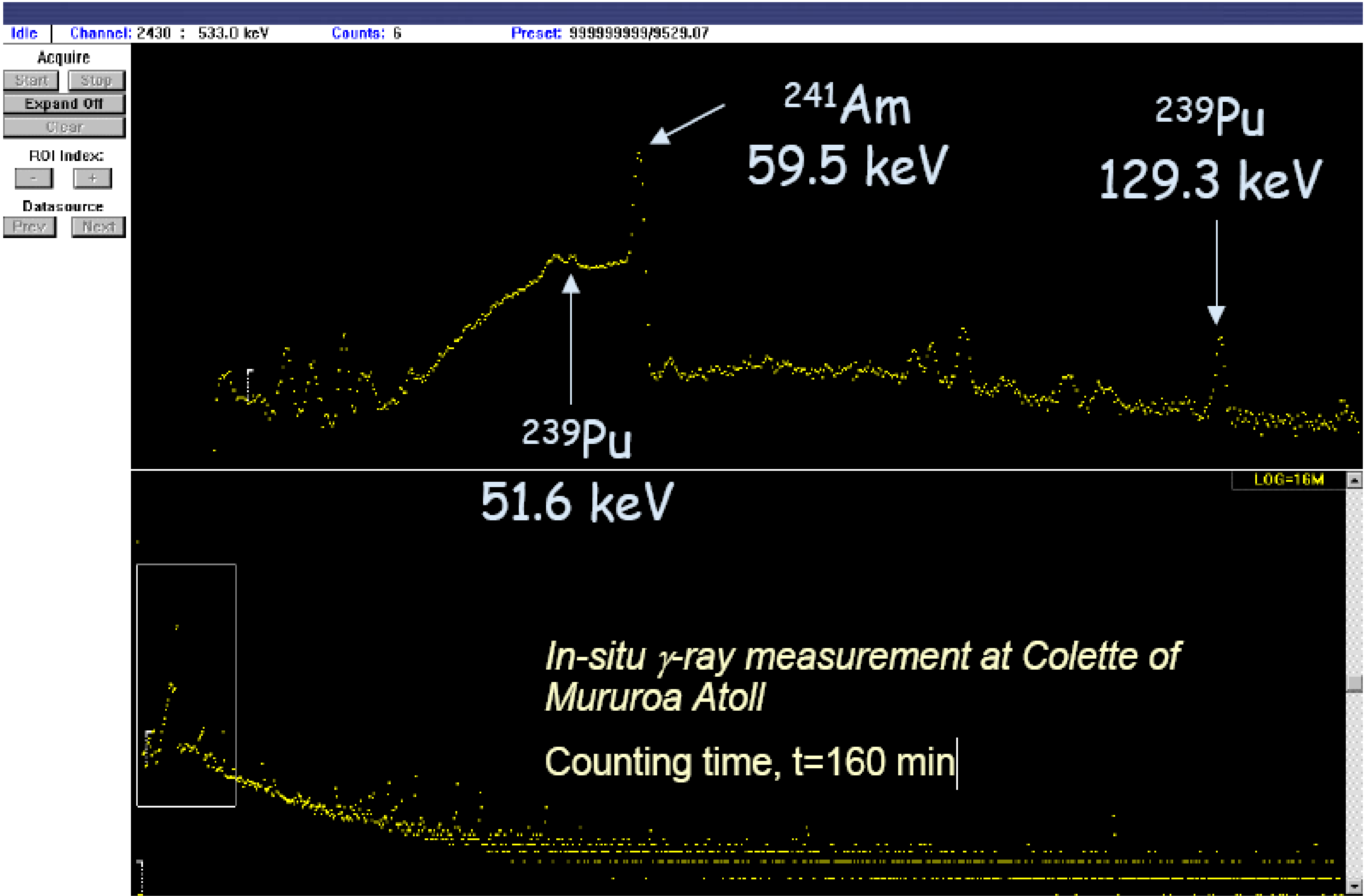
Background radiation in seminar-room, TC-18, IAEA Laboratories Seibersdorf

Counting time, t=65 h



Lawn at south Balaton/HUN (t=34 min)

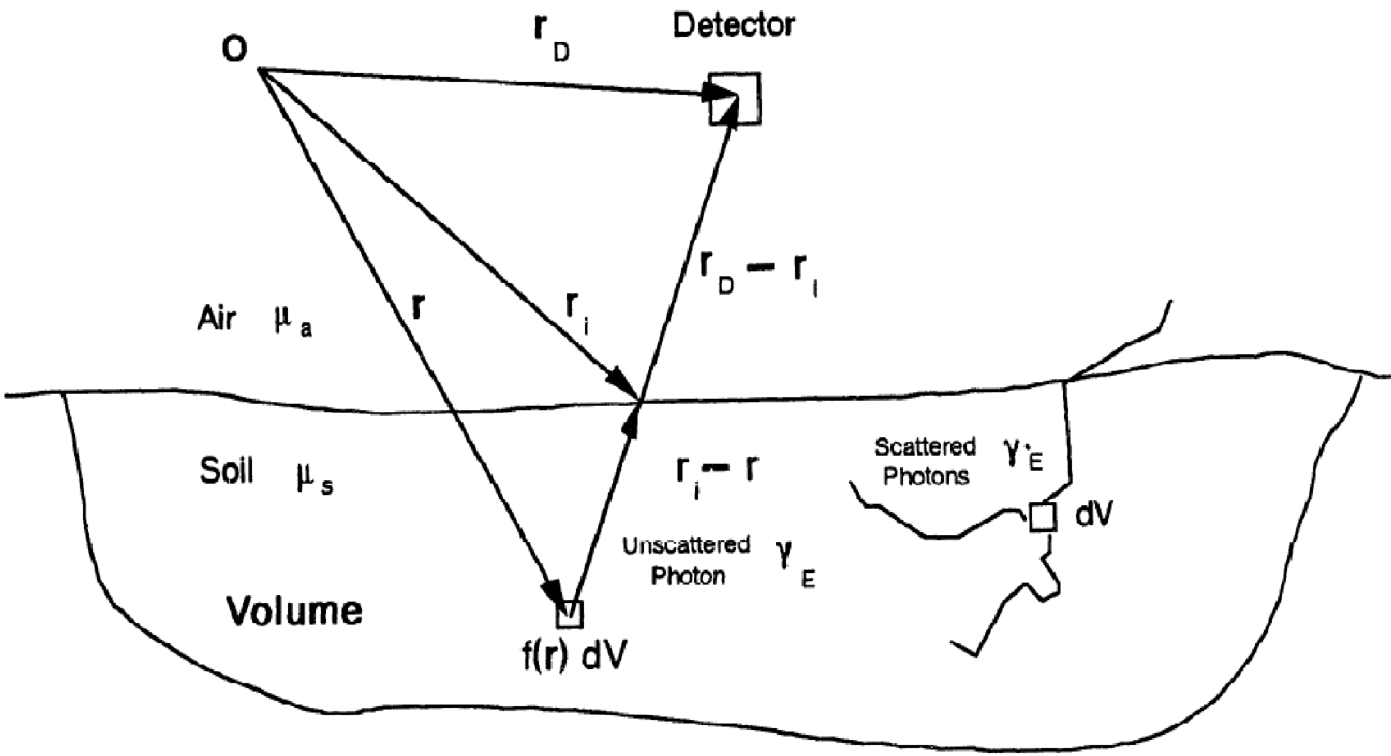
^{137}Cs	(176 ± 14) Bq/m²	
^{7}Be	(130 ± 60) Bq/m²	
^{40}K	(339 ± 18) Bq/kg	
^{226}Ra	(*14.0 ± 6.0*) Bq/kg	
^{238}U	(28.2 ± 1.8) Bq/kg	^{214}Pb, ^{214}Bi
^{235}U	(*640 ± 300*) Bq/kg	
^{232}Th	(19.2 ± 1.6) Bq/kg	^{228}Ac, ^{212}Pb, ^{212}Bi, ^{208}Tl


Idle
Channel: 2430 : 533.0 keV
Counts: 6
Preset: 999999999/9529.07
Acquire
Start
Stop
Expand Off
Clear
ROI Index:
Datasource
Prev
Next
241Am
59.5 keV
239Pu
129.3 keV
239Pu
51.6 keV
LOG=16M
In-situ γ-ray measurement at Colette of Mururoa Atoll
Counting time, t=160 min

Theoretical model for photon flux calculation in an in situ measurement

$$\Phi = \int_V \frac{f(r)}{4\pi(r_D - r)^2} \exp\left[-\frac{\mu_s}{\rho}\rho(r_i - r) - \frac{\mu_a}{\rho_a}\rho_a(r_D - r_i)\right] dV \qquad (2.2)$$

where f(r) is the source strength at r, μ_s/ρ is the mass attenuation coefficient for soil ($cm^2\ g^{-1}$) and μ_a/ρ is the mass attenuation coefficient for air.



Commonly used depth distributions and units

Exponential	$f(z) = \frac{S_0}{L} e^{-z/L}$	S_0 in (Bq m^{-2})
Uniform L=∞	$f(z) = S_V$	S_V in e.g. (Bq m^{-3}), (Bq cm^{-3}), or (Bq kg^{-1})
Plane L=0	$f(z) = S_A$	S_A in (Bq m^{-2})

Plane at depth z_0

$$f(z) = S_A\, \delta(z - z_0)$$

S_A in (Bq m^{-2})

In general

$$f(z) = \sum_i S_i\, \delta(z - z_i)$$

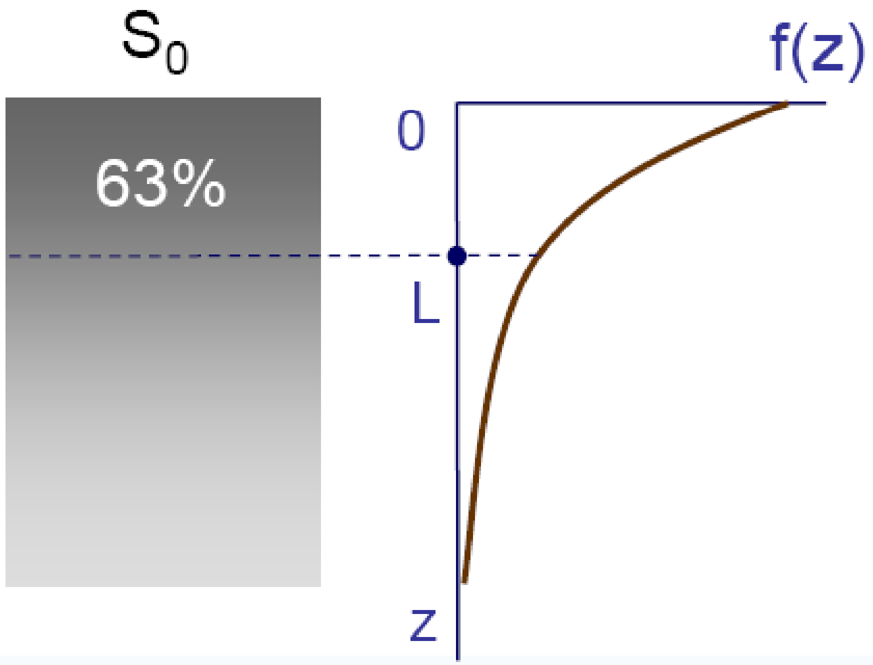
S_i in (Bq m^{-2})

Exponential distribution of a radionuclide

$$f(z) = \frac{S_0}{L} e^{-z/L}$$

S_0 – activity in a soil column, total inventory (Bq m^{-2})

L – relaxation length, e.g. (cm)



Interpretation of S_0:

$$\int_0^\infty f(z)dz = S_0$$

Interpretation of L:

$$\int_0^L f(z)dz \approx 0.63\, S_0$$

Example values for relaxation length (Soil density: 1.6 gcm^{-3})

L (cm)	Applications
0.10	used for **fresh fallout** where all the radioactivity is deposited on the soil surface
1.25	used for **global fallout on wooded or desert sites** (1.25 cm – 3.1 cm)
12.5	used for **global fallout on open undisturbed fields** (7 cm – 12.5 cm)
infinity	used for uniformly distributed radionuclides, e.g. **natural radioactivity**

Calculation of unscattered photon flux for different radionuclide depth distributions

Exponential

$$\Phi = \frac{1}{2} S_0 \left\{ E_1(\mu_a h) - e^{\frac{\mu_a}{\mu}\frac{h}{L}} E_1\left[\left(1 + \frac{1}{\mu L}\right)\mu_a h\right]\right\}$$

Uniform

$$\Phi = \frac{1}{2} S_V \frac{\mu_a}{\mu}\left[\frac{1}{\mu_a h} e^{-\mu_a h} - E_1(\mu_a h)\right]$$

Plane

$$\Phi = \frac{1}{2} S_0 \, E_1(\mu_a h)$$

The function $E_1(x)$ is the 1st order exponential integral

$$E_1(x) = \int_x^{\infty} \frac{e^{-t}}{t} dt$$

Series expansion

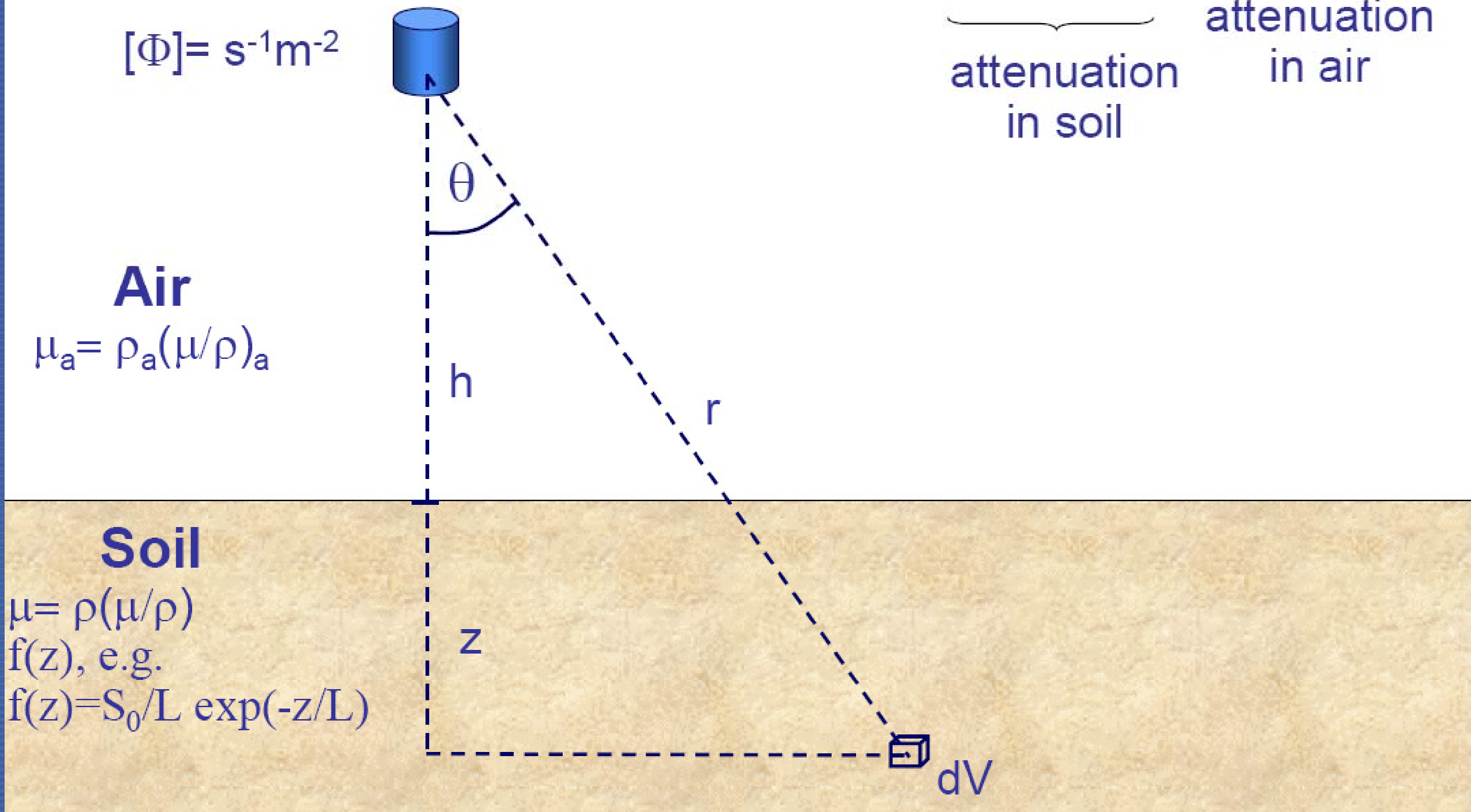
$$E_1(x) = -\gamma - \ln x - \sum_{n=1}^{\infty} \frac{(-1)^n x^n}{n \, n!}$$

$$\gamma = 0.57721\,56649...$$

Theoretical model for photon flux calculation in an in situ measurement

$$\Phi = \int_0^{\pi/2} d\theta \int_{h/\cos\theta}^{\infty} \frac{S_0}{4\pi r^2} e^{-z/L} \cdot 2\pi r^2 \sin\theta \cdot \underbrace{e^{-\mu(r-h/\cos\theta)}}_{\text{attenuation in soil}} \cdot \underbrace{e^{-\mu_a h/\cos\theta}}_{\text{attenuation in air}} dr$$

$[\Phi] = s^{-1}m^{-2}$



Calculation of radionuclide deposition

$$C = 10 \frac{N}{t} \frac{1}{C_f I_\gamma}$$

- C – deposition – surface concentration of a radionuclide, (kBq m^{-2})
- 10 – unit conversion factor
- N – net counts in the full-energy peak at energy E
- t – counting time, live time, (s)
- C_f – detector calibration factor at energy E, (cm^2)
- I_γ – intensity of a gamma line at energy E (emission probability)

Detector calibration factor

$$C_f = \frac{R_f}{A_s} = \left(\frac{R_f}{R_0}\right)\left(\frac{R_0}{\Phi}\right)\left(\frac{\Phi}{A_s}\right)$$

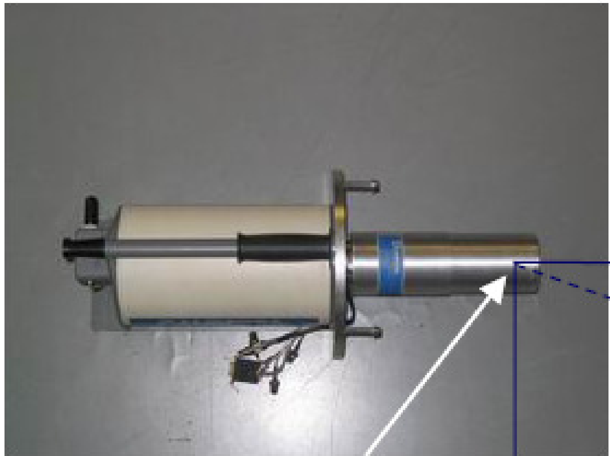
geometrical factor – total photon flux density at the detector per unit concentration or deposition inventory of the radionuclide

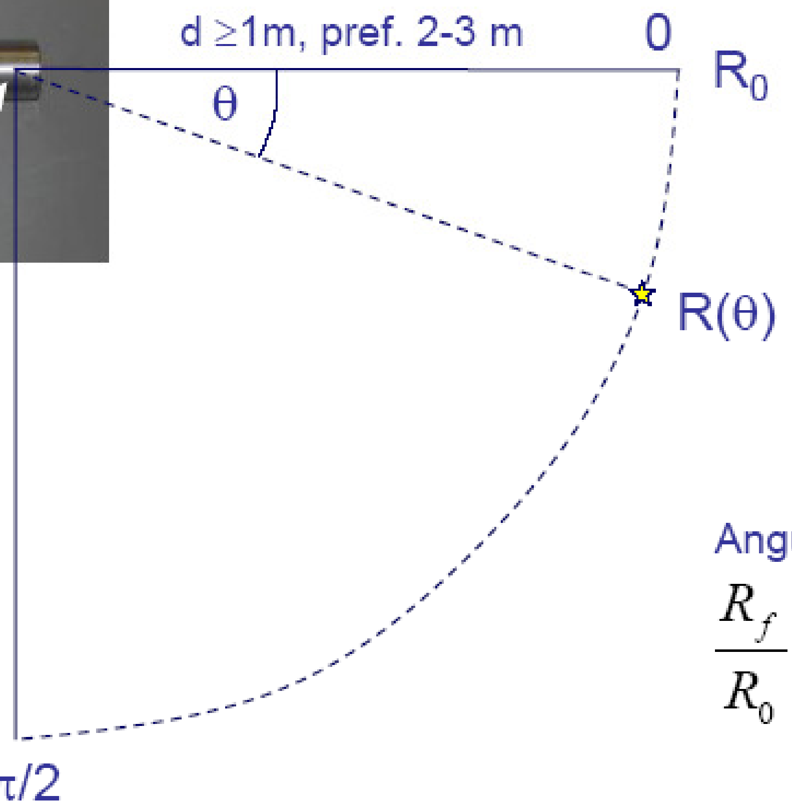
angular correction factor – correction factor required to account for the detector angular response

response factor - neat peak count rate due to a unit primary photon flux density of energy E incident on the detector (normal to the detector face)

Calibration – detector characteristics

R(θ) – net count rate in a full-energy peak, at angle θ





Flux at the detector's effective center:

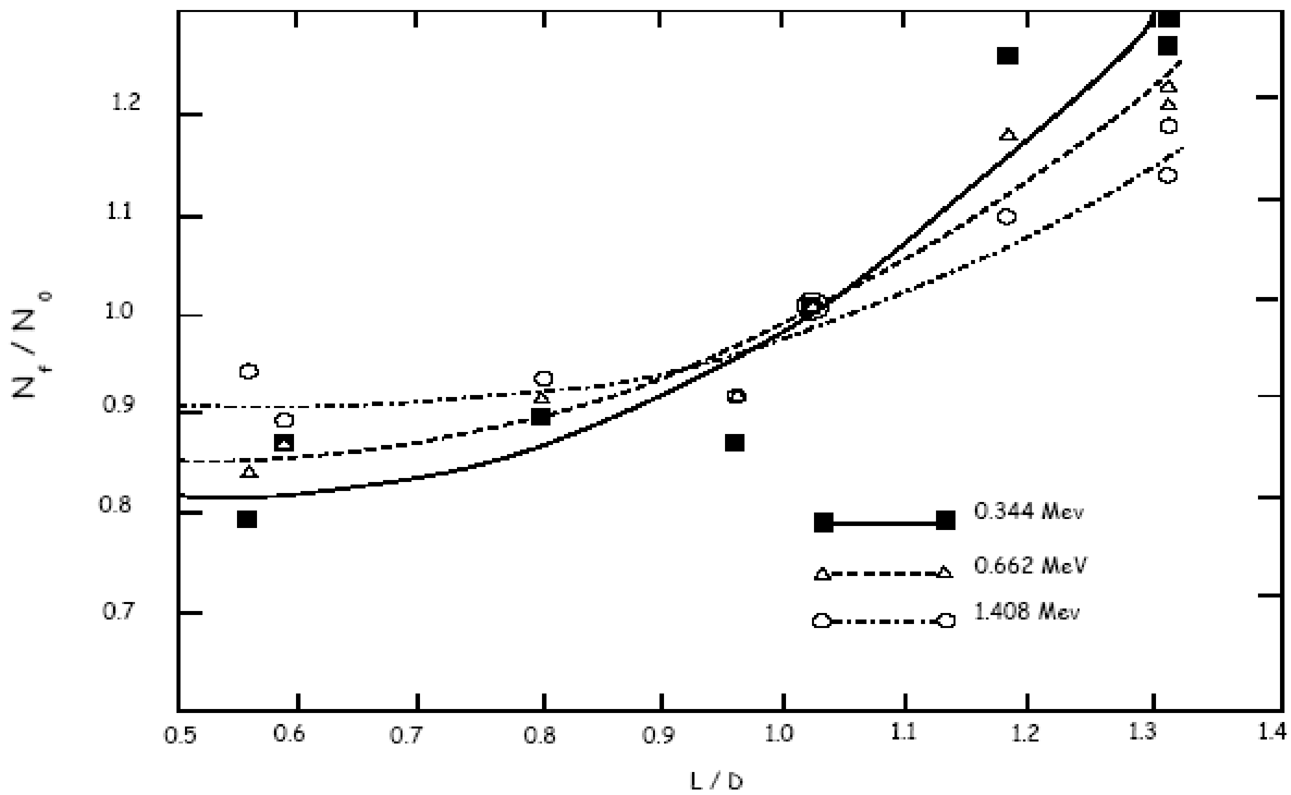
$$\Phi_0 = \frac{AI_\gamma}{4\pi r^2} e^{-\mu_a (r-x)}$$

Angular correction factor:

$$\frac{R_f}{R_0} = \int_0^{\pi/2} \frac{\Phi(\theta)}{\Phi} \frac{R(\theta)}{R_0} d\theta$$

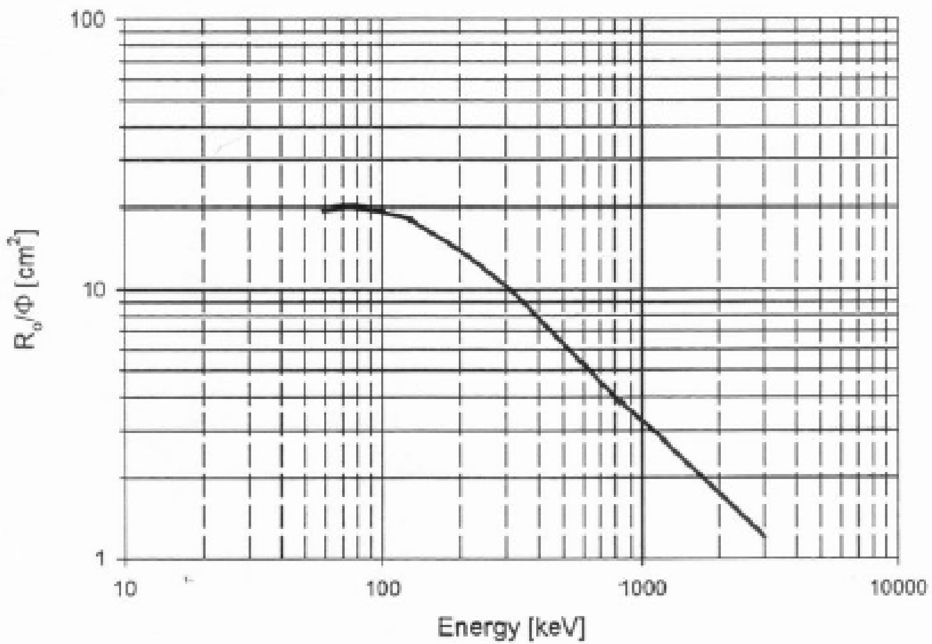
Angular correction factor

Angular correction factor R_f/R_0 as a function of Ge crystal length/diameter L/D ratio at three different energies for a downward facing detector for a uniform with depth source profile in the soil.

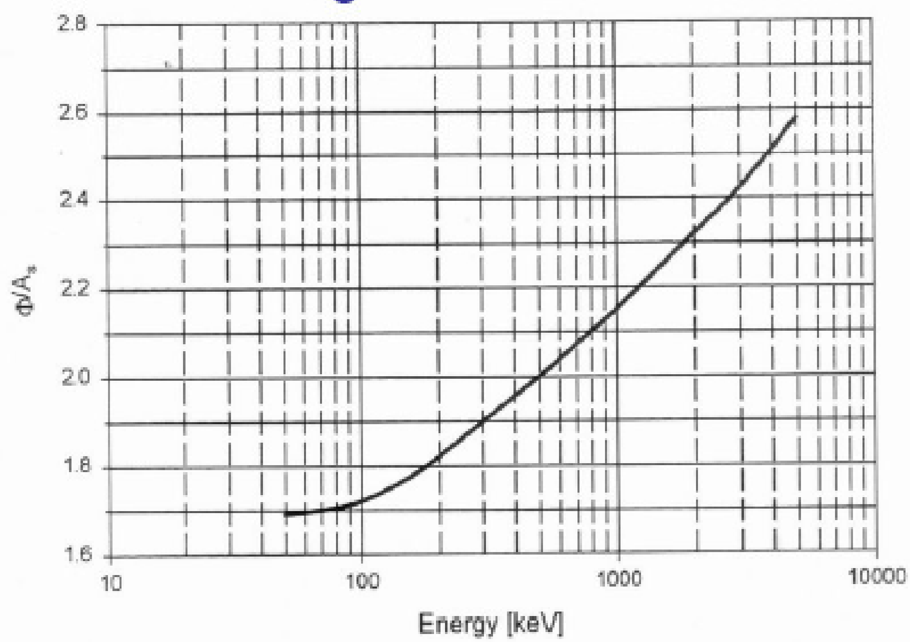


Examples of a detector characteristics

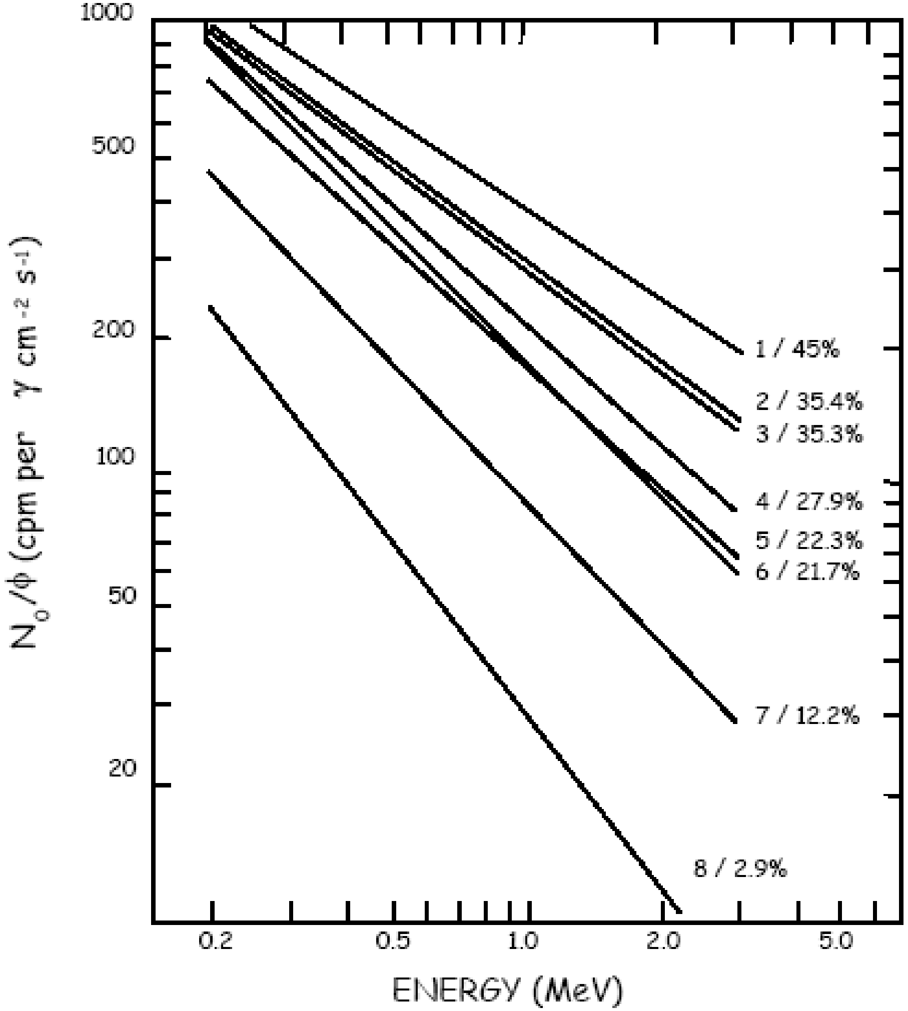
Typical **response factor**, R_0/Φ, for a Ge detector of 22% relative efficiency



Geometrical factor, Φ/A_s, as a function of photon energy in case of surface source distribution for 1m above ground

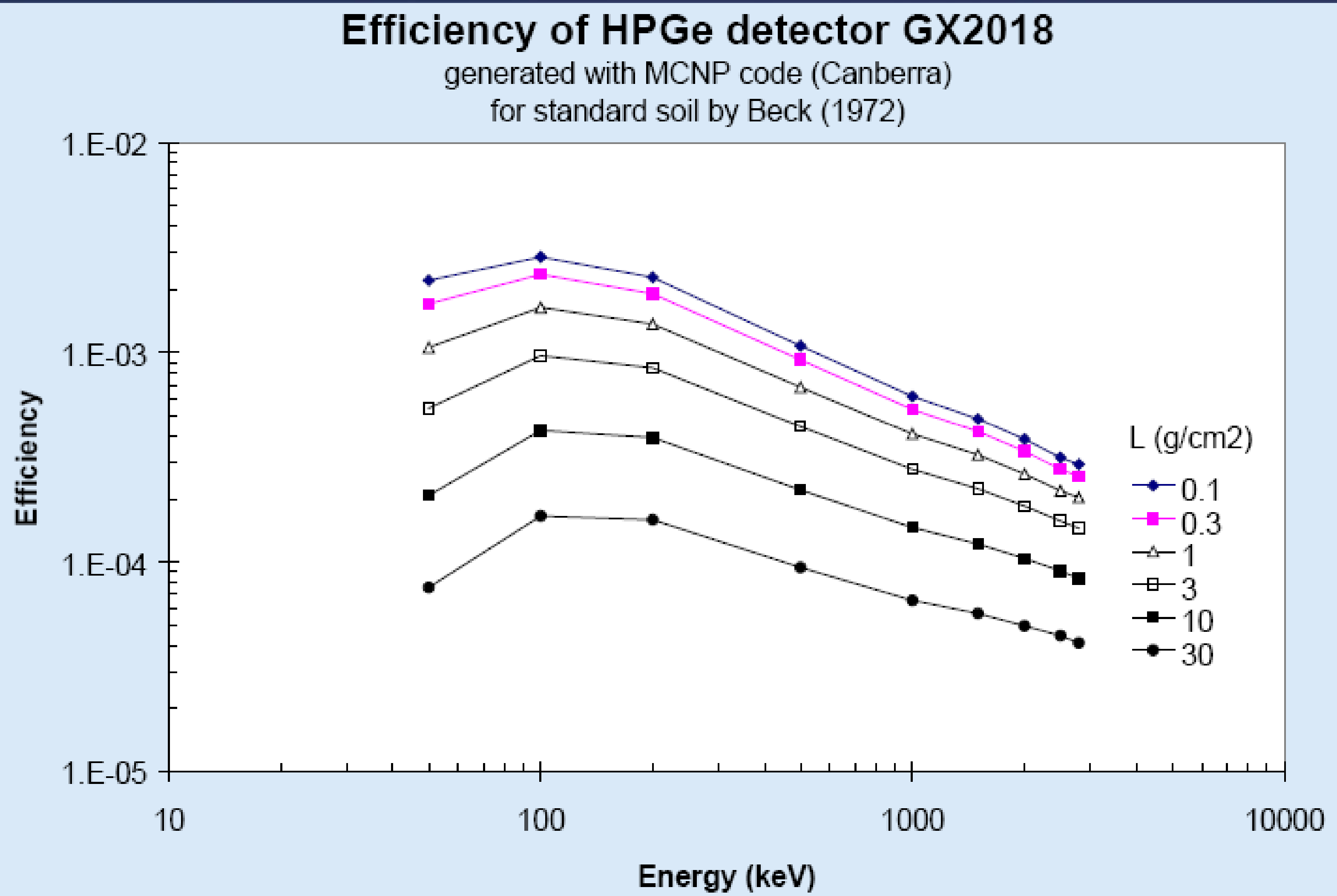


Response factor – a quick reference



Count rate per unit incident flux at normal incidence, R_0/Φ, as a function of energy for different detectors.

Different approach – mathematical detector efficiency calculation

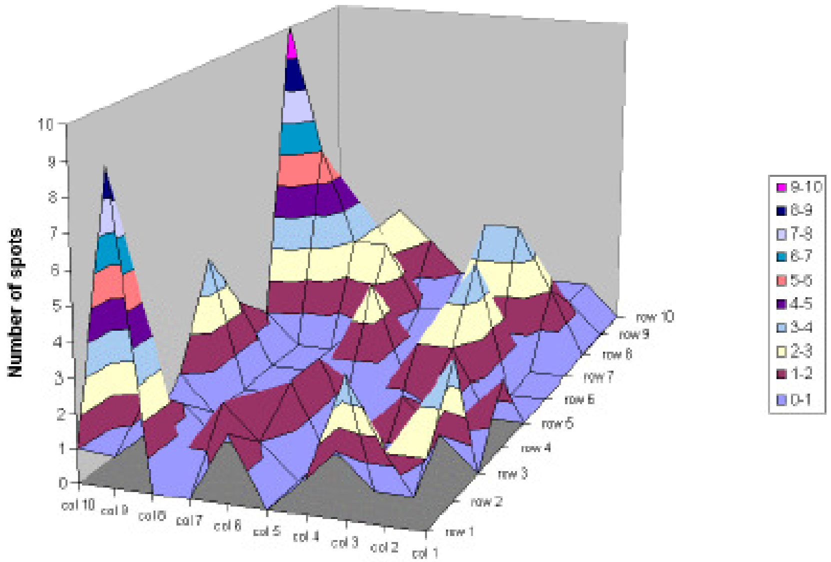


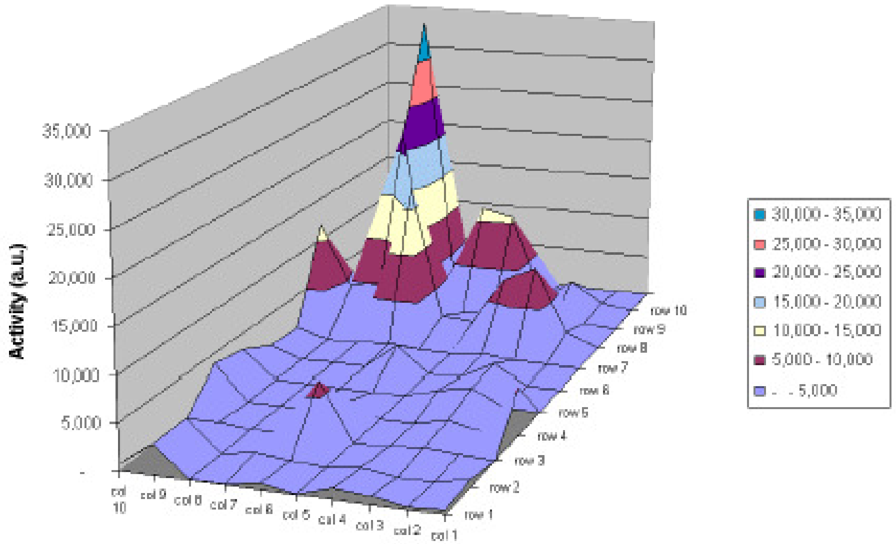
Sources of errors

	Error
• Radiation situation is not adequately well described by a mathematical model used/available (lack of proper model) - departure of real depth distribution from the model used, - obstacles, e.g. building, wall, etc. (modified "field-of-view" –departure from a conical shape) - nonuniform surface, e.g. grassland and a road in the field-of-view - roughness of the soil surface (if not considered in the model)	10%-50% (larger are not uncommon)
• Poor experimental arrangement in the field (geometry)	10%-20%
• Incorrect or poor calibration for in-situ measurements	15%-20% (?)
• Human errors due to e.g. typing for reporting	a factor >10
• Counting statistics of net count rate	5%-10%
• Nuclear constants used	1%-5%

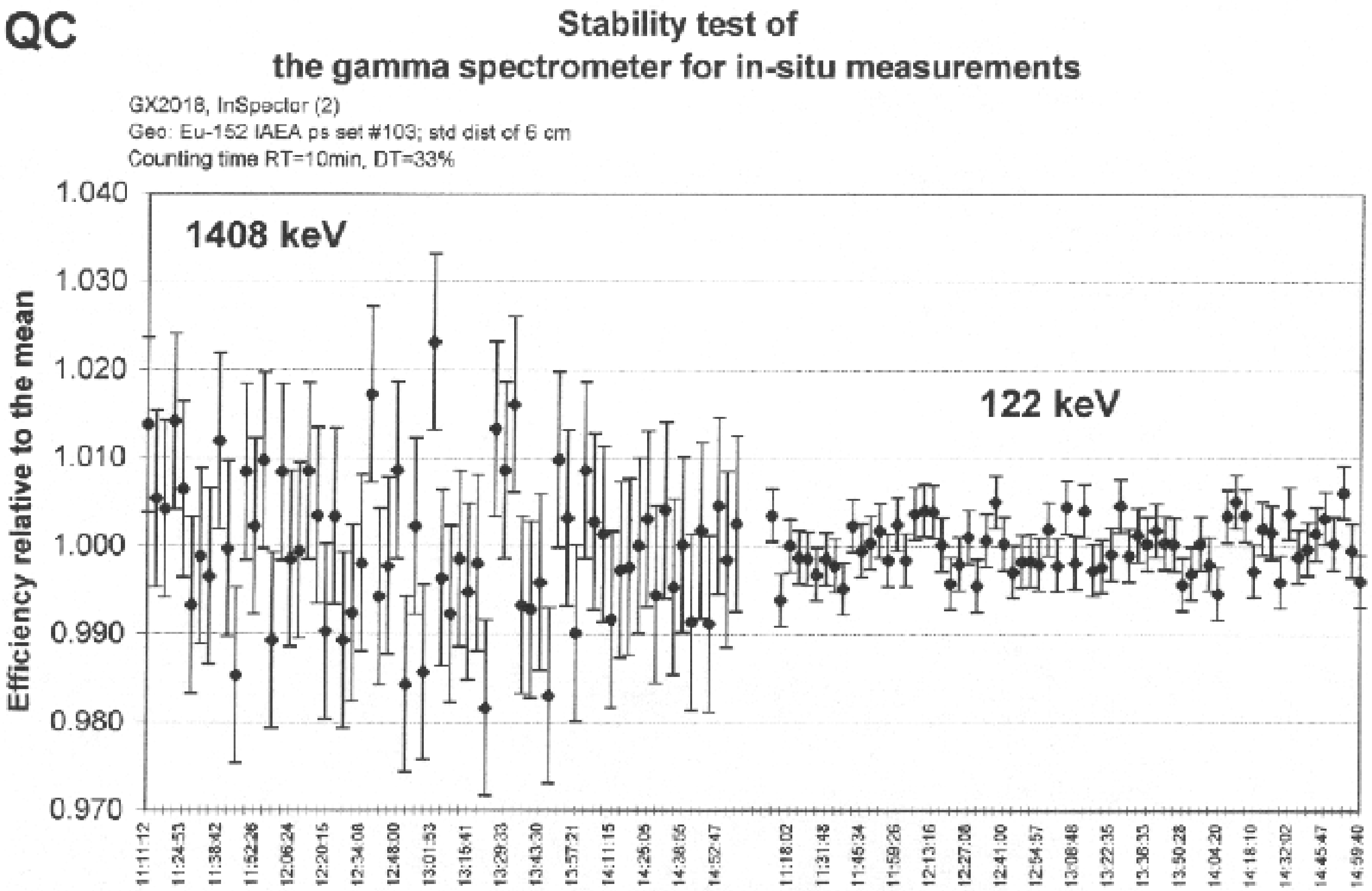
Inhomogenius radionuclide distribution

Surface-distribution of Am-241at Colette of Mururora Atoll
10m x 10m square



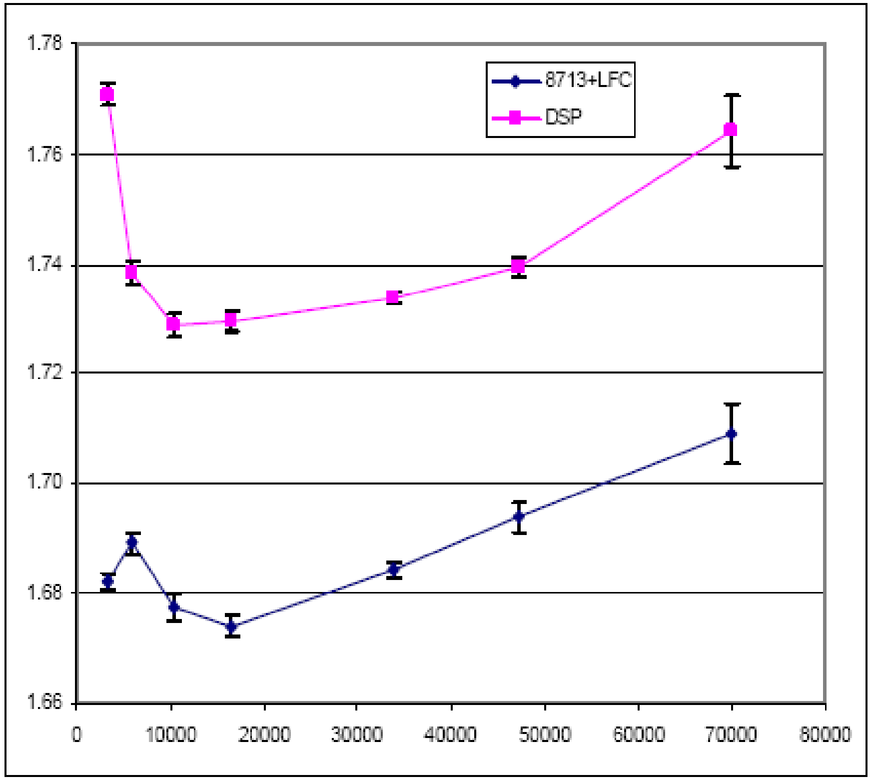


Monitoring the system performance



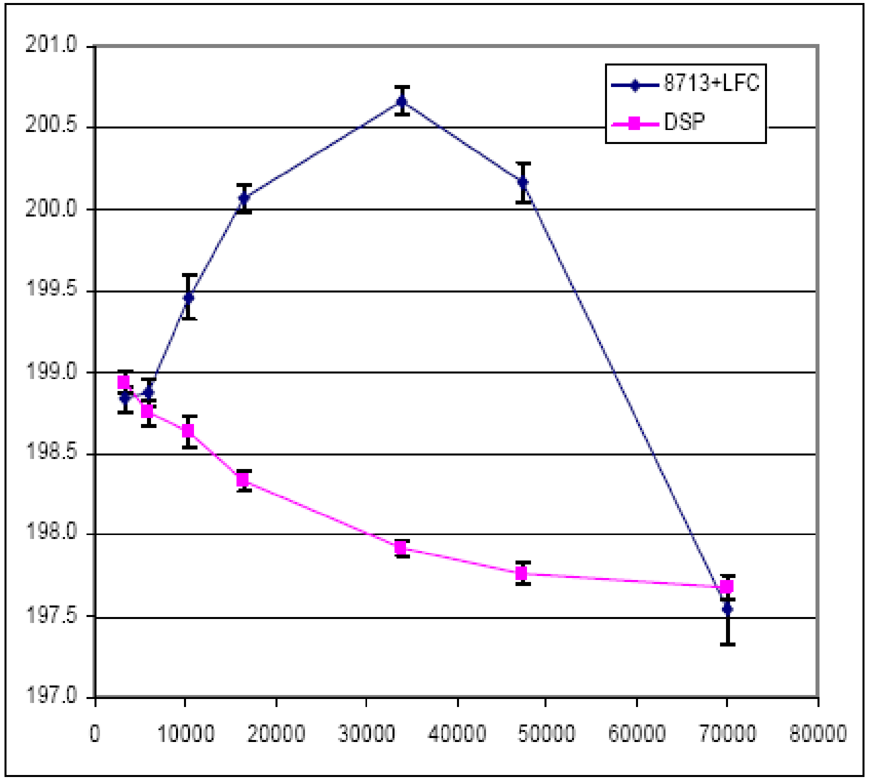
Effect of count rate in well adjusted sytem

Resolution, FWHM (keV)



Input count rate (cps)

Net peak area (cps)



Input count rate (cps)

IAEA

Atoms for Peace: The First Half Century

1957–2007

References

Field measurements of radioactivity: introduction to in-situ γ-ray spectrometry,

Marek Makarewicz IAEA

References

- ICRU REPORT 53 Gamma –Ray Spectrometry in the Environment, Beteheshada(1994)
- EML-557 In situ Gamma –Ray Spectrometry A tutorial for environmental radiation scientist, K.M Miller nad P. Shebell, US DOE 1993
- Generic procedures for monitoring in a nuclear or radiological emergency IAEA-TECDOC-1092, June 1999
- A theoretical comparison of methods of quantification of radioactive contamination in soil using in situ gamma spectrometry; J. MacDonald et. al; J. of Radiol. Prot. 1997 Vol. 17 No.1 3-15
- Uncertainties of in situ gamma spectrometry for environmental monitoring, W. Sowa et. al, Radiation Protection Dosimetry Vol. 27 No. 2 pp. 93-101 (1989)