The Abdus Salam
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Workshop on Understanding and Evaluating Radioanalytical Measurement Uncertainty

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In-situ-gamma Spectrometry

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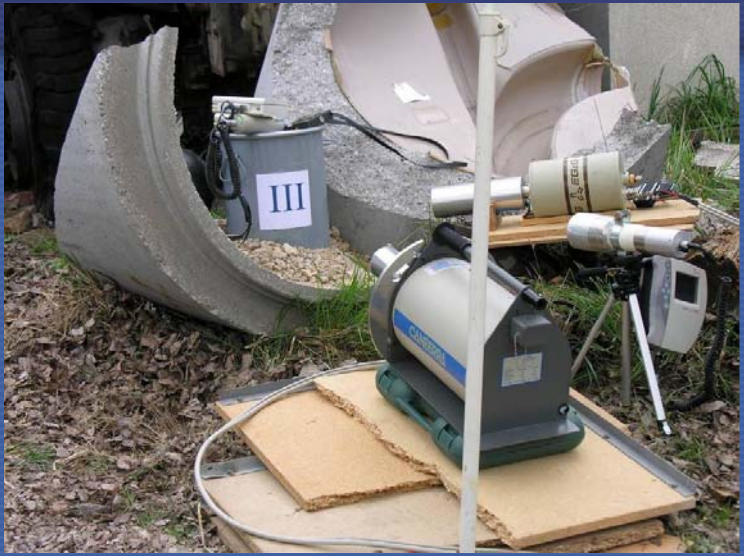
IN SITU GAMMA RAY-SPECTROMETRY

IAEA

Atoms for Peace: The First Half Century

1957–2007

IN SITU GAMMA RAY-SPECTROMETRY



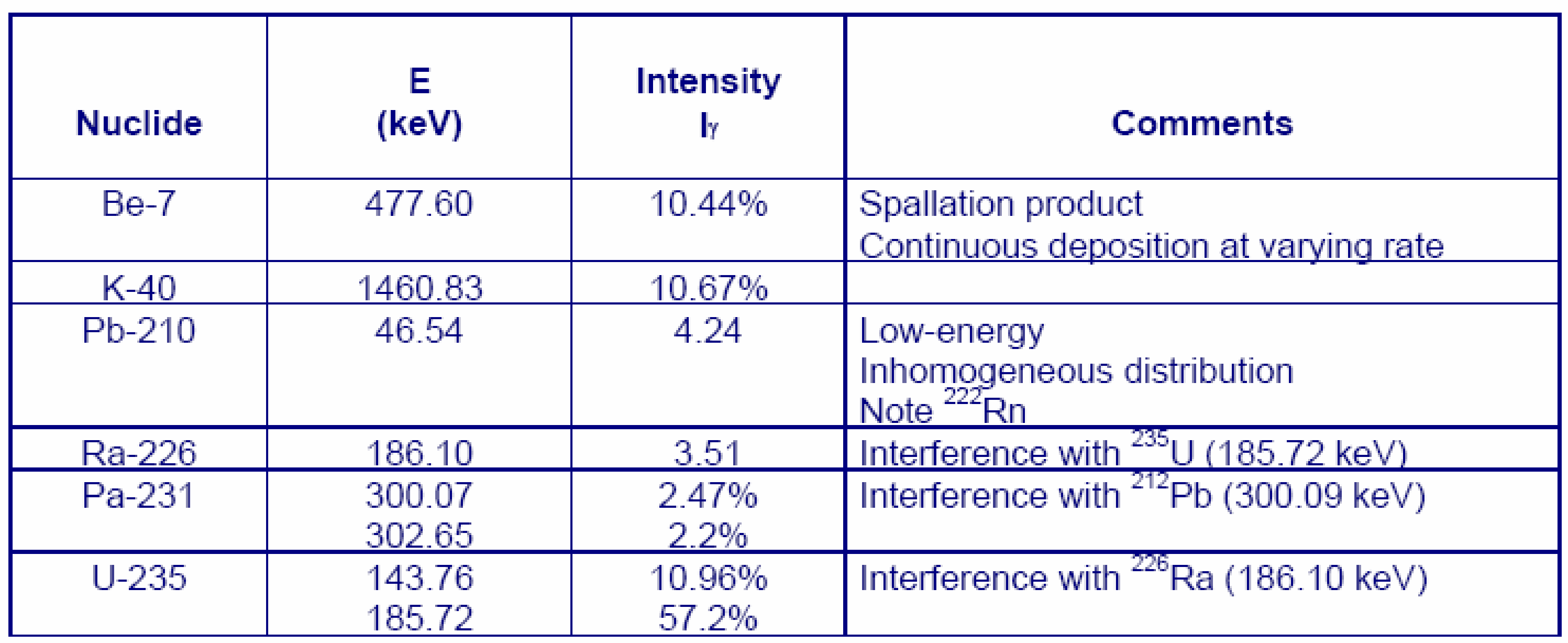
Applications

- Rapid identification and determination of gamma emitting radionuclides in the environment (in the field)
  - activity concentration ($Bqkg^{-1}$) or
  - deposition (Bqm^{-2})
  - activity concentration ($Bqkg^{-1}$)
- 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

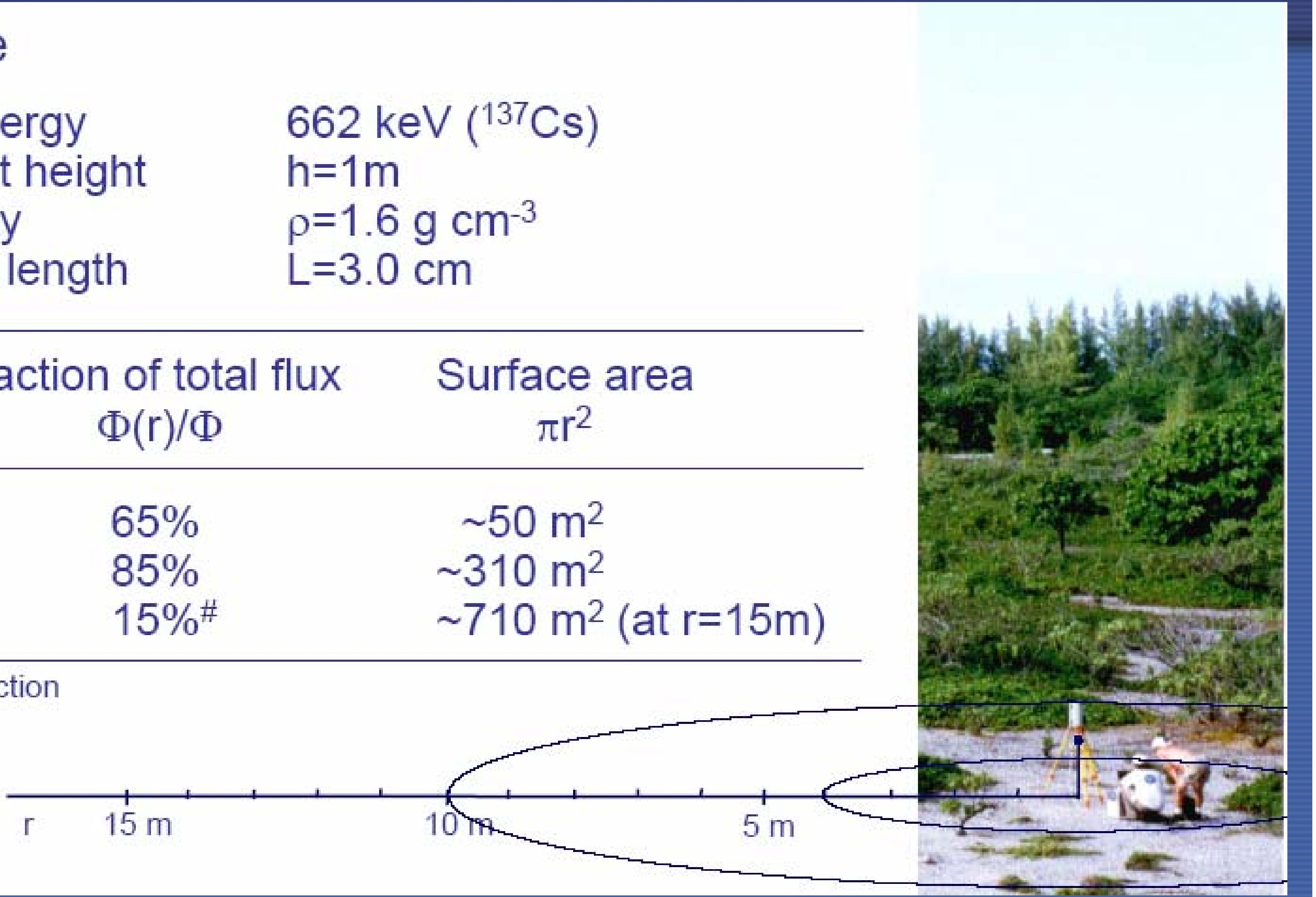
Effective field of view – Sample size

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

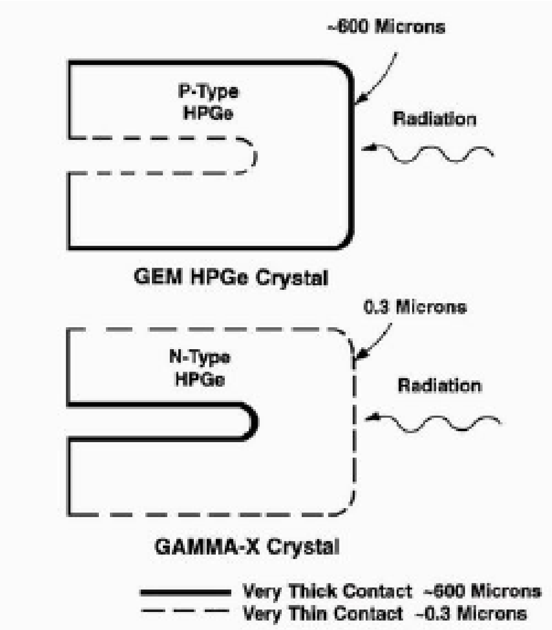
- HPGe (hyper-pure germanium detector)
  - p-type for Eγ > 100 keV (3-60 keV up to 3-10 MeV)
  - n-type for Eγ > 45 keV (3 keV up to 300 keV-3 MeV)
- Efficiency of 20%-50% relative to 3”x3” NaI(Tl) at 1.3 MeV
- Portable (hand-held) cryostat

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

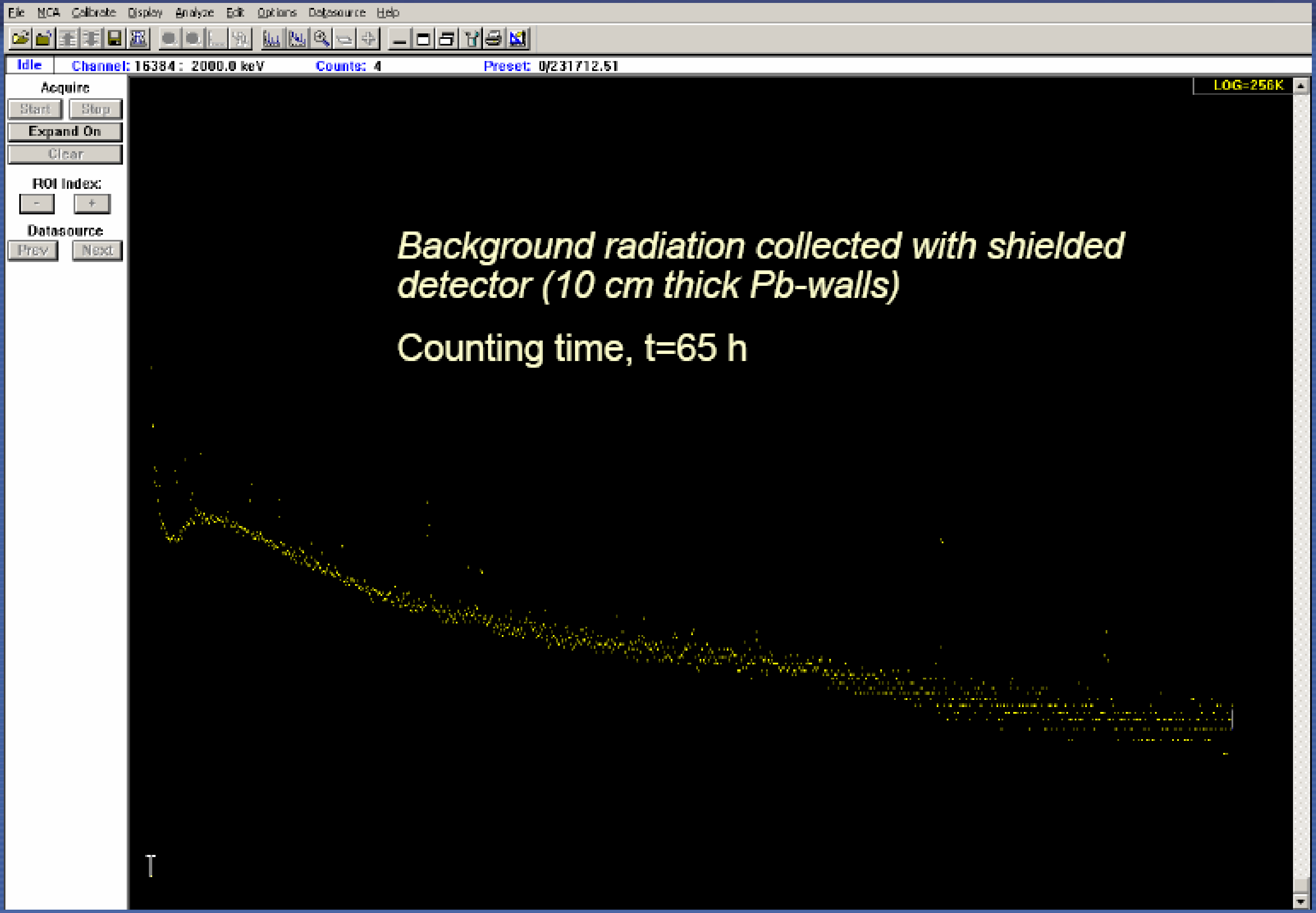
Software

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

Documentation

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

Shielded background

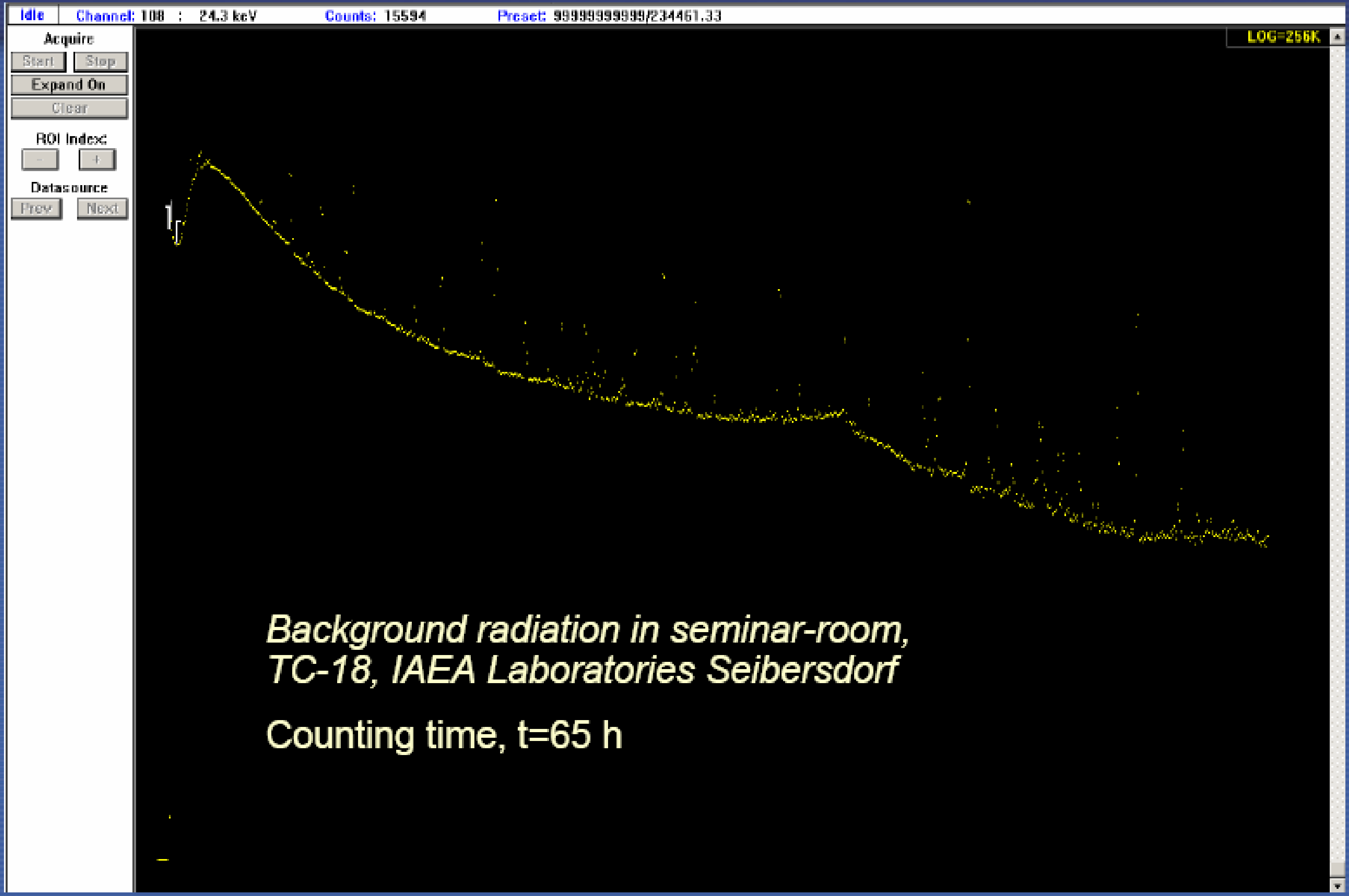


Idle Channel: 16384 : 2000.0 keV Counts: 4 Preset: 0/231712.51

Background radiation collected with shielded detector (10 cm thick Pb-walls)

Counting time, t=65 h

Seminar room



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Idle   Channel: 108  :  24.3 keV     Counts: 15594      Preset: 999999999999/234461.33
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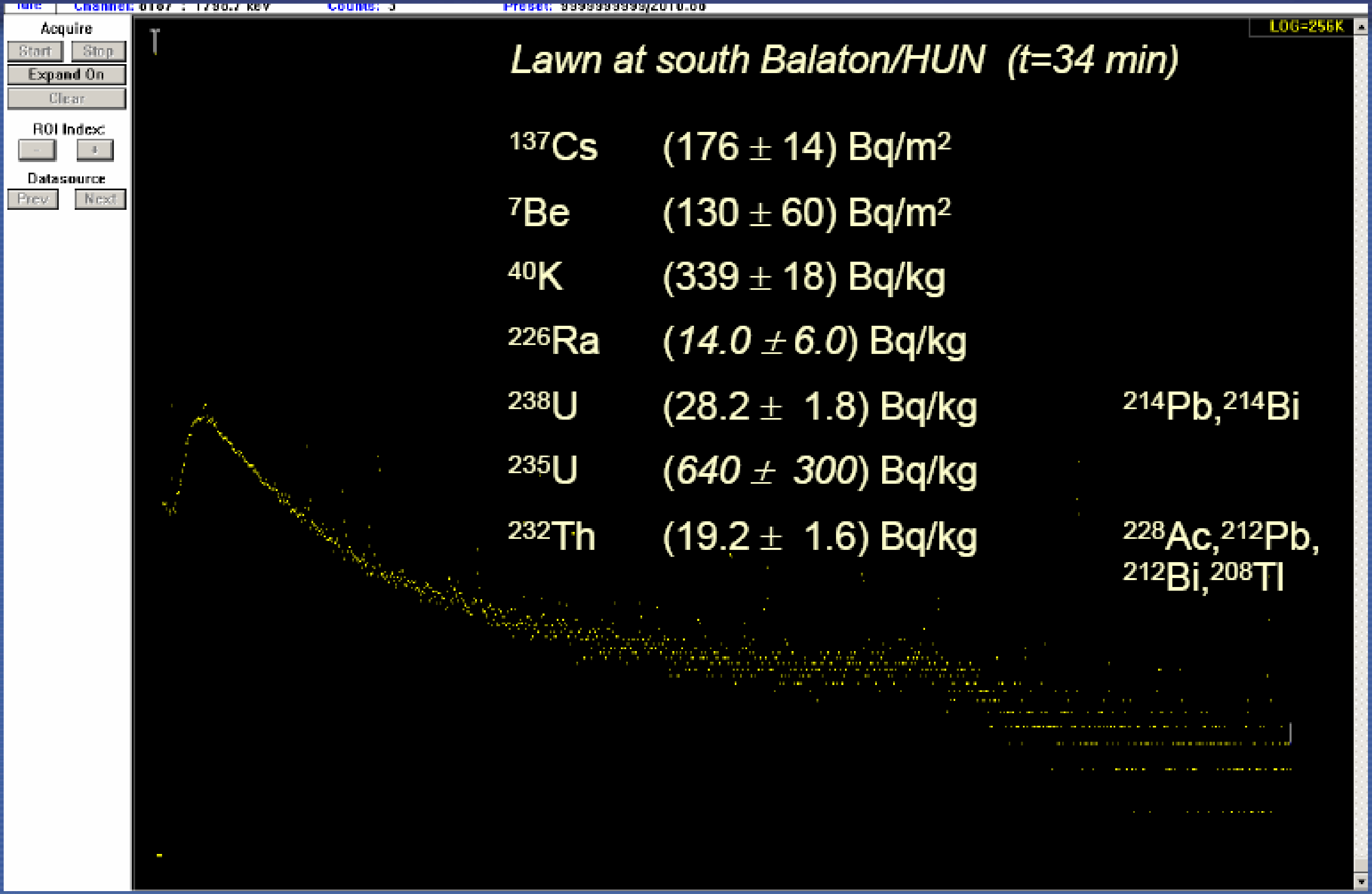
Background radiation in seminar-room, TC-18, IAEA Laboratories Seibersdorf

Counting time, t=65 h

Typical environment



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

^{137}Cs	(176 ± 14) Bq/m^2	
^{7}Be	(130 ± 60) Bq/m^2	
^{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

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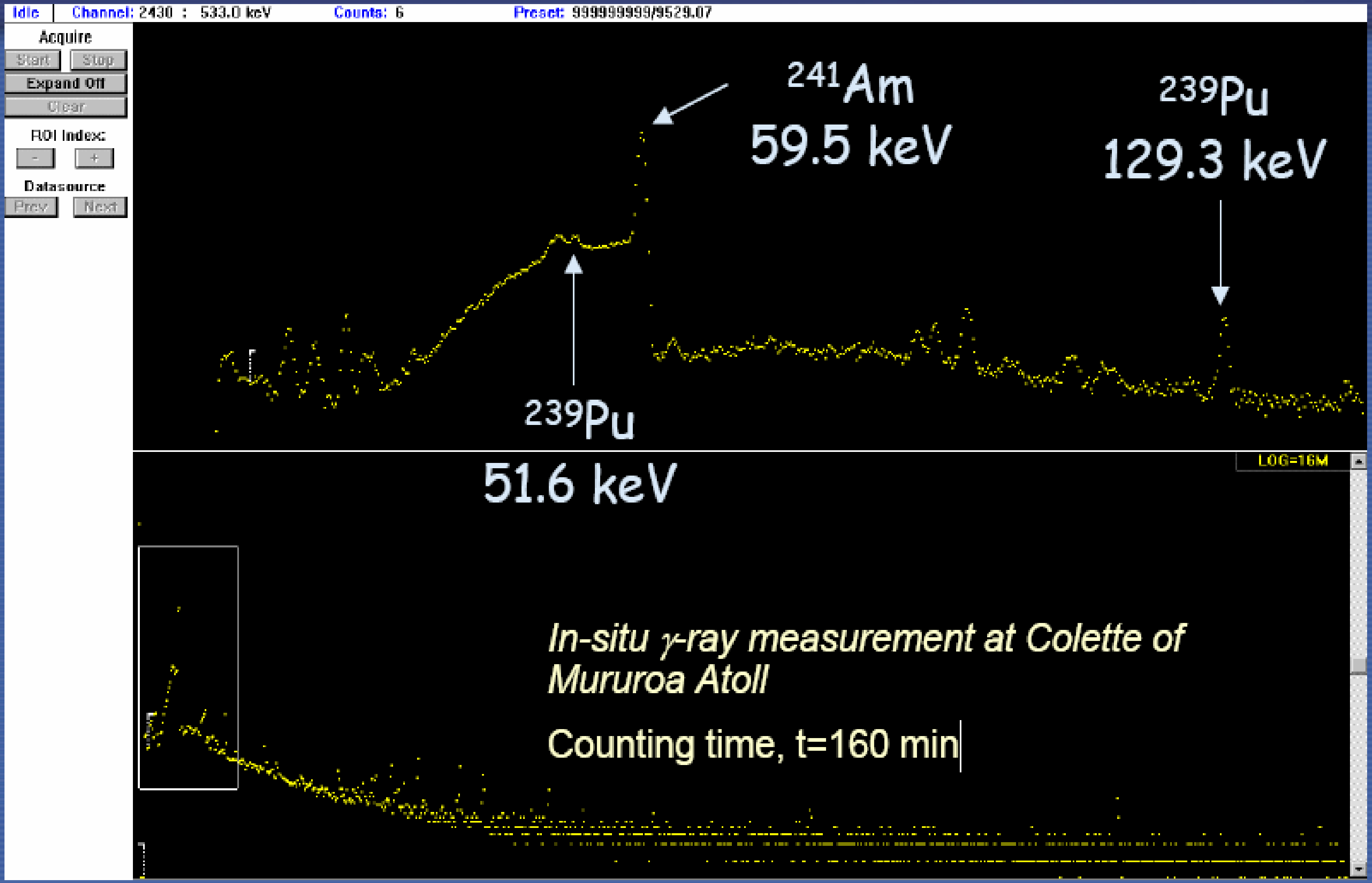
Muroroa



Idle | Channel: 2430 : 533.0 keV Counts: 6 Preset: 9999999999/9529.07

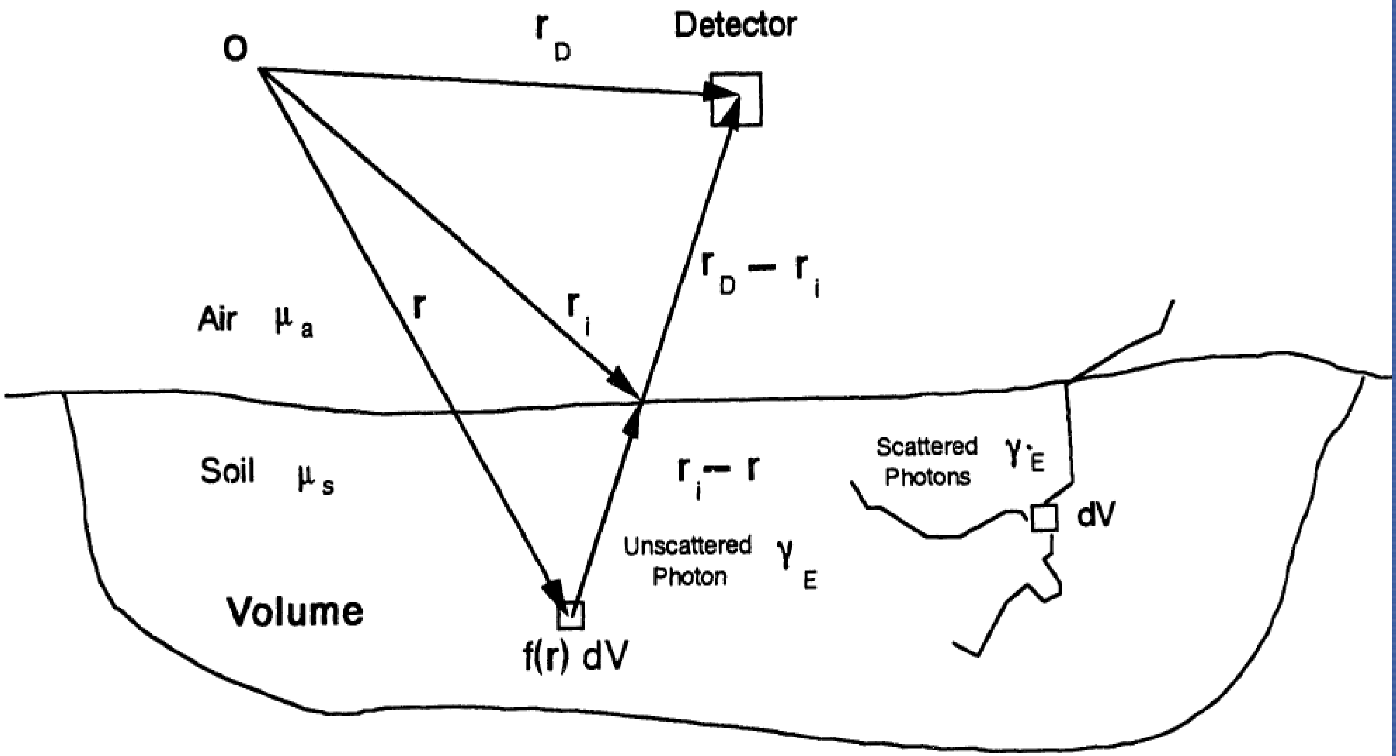
In-situ γ-ray measurement at Colette of Mururoa Atoll

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

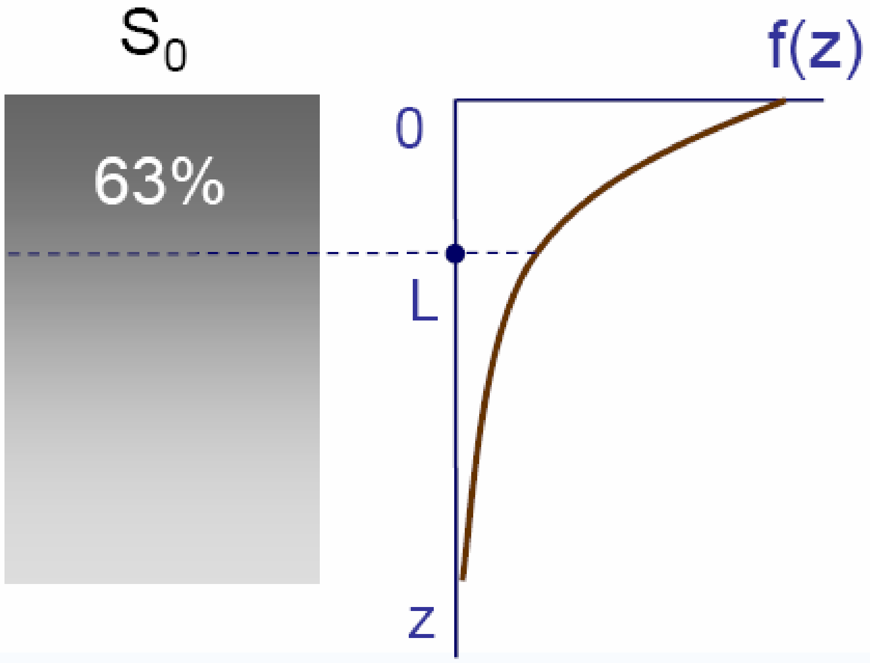
Exponential	$f(z)=\frac{S_0}{L}e^{-z/L}$	S_0 in (Bq m^{-2})
Uniform L=∞	$f(z)=S_V$	S_V in e.g. (Bq m^{-3}), (Bq cm^{-3}), or (Bq kg^{-1})
Plane L=0	$f(z)=S_A$	S_A in (Bq m^{-2})
Plane at depth z_0	$f(z)=S_A\,\delta(z-z_0)$	S_A in (Bq m^{-2})
In general	$f(z)=\sum_i S_i\,\delta(z-z_i)$	S_i in (Bq m^{-2})

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

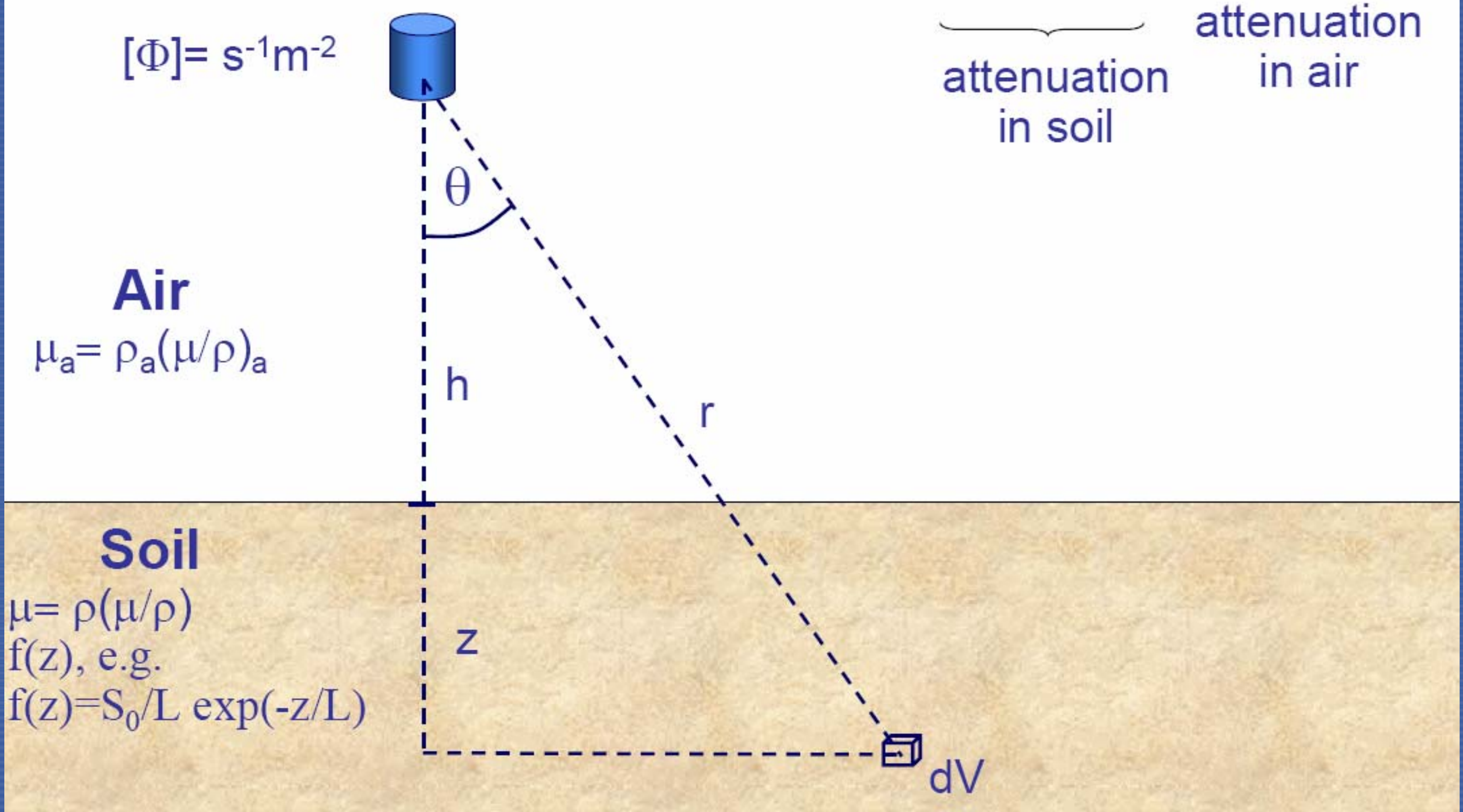
$$E_1(x) = -\gamma - \ln x - \sum_{n=1}^{\infty} \frac{(-1)^n x^n}{n\, n!}$$

$$\gamma = 0.57721\,56649\ldots$$

Theoretical model for photon flux calculation in an in situ measurement

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

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



Air

$\mu_a = \rho_a(\mu/\rho)_a$

Soil

$\mu = \rho(\mu/\rho)$

$f(z)$, e.g.

$f(z) = S_0/L \exp(-z/L)$

Calculation of radionuclide deposition

unit conversion factor

net counts in the full-energy peak at energy E

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

deposition – surface concentration of a radionuclide, (kBq m^{-2})

counting time, live time, (s)

detector calibration factor at energy E, (cm^2)

intensity of a gamma line at energy E (emission probability)

Detector calibration factor

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

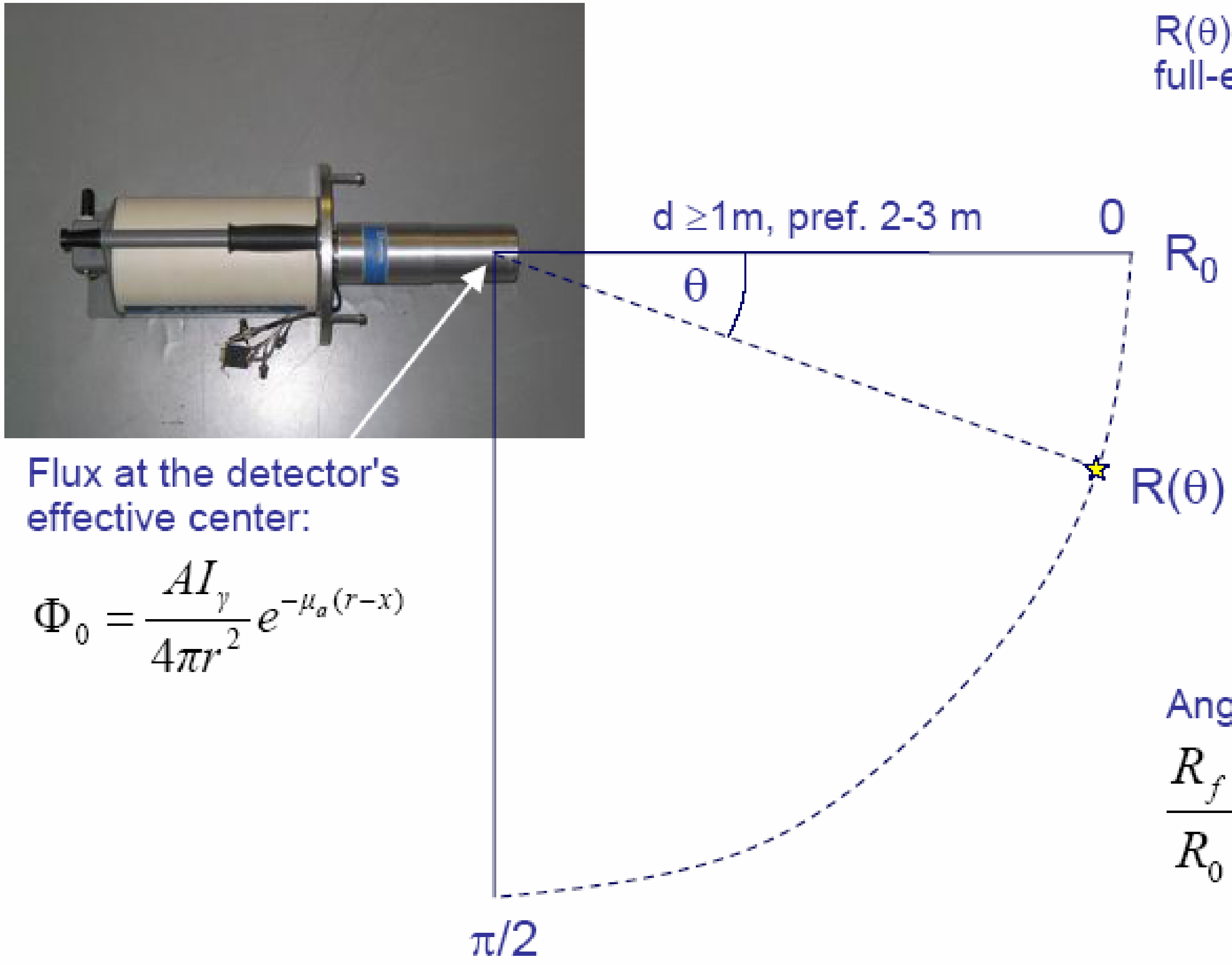
angular correction factor – correction factor required to account for the detector angular response

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

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:

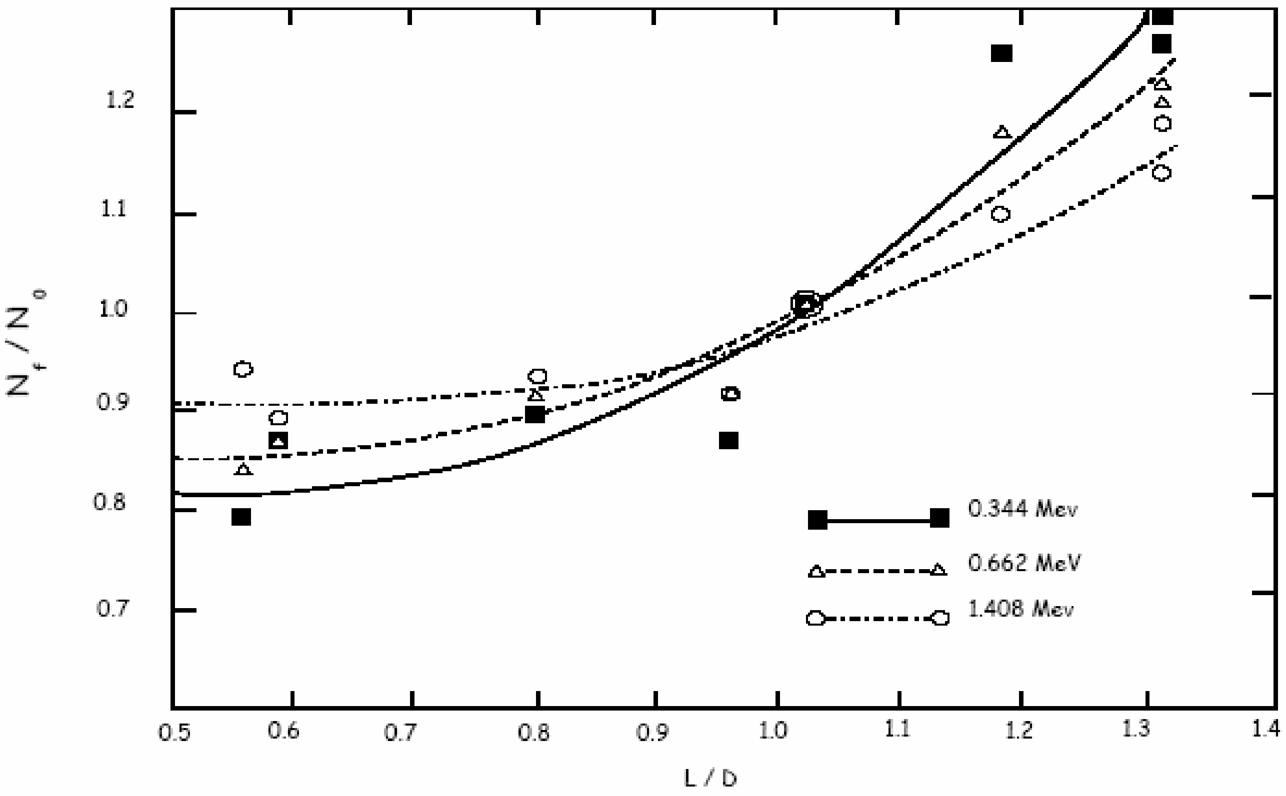
$$\Phi_0 = \frac{A I_\gamma}{4\pi r^2} e^{-\mu_a (r-x)}$$

Angular correction factor:

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

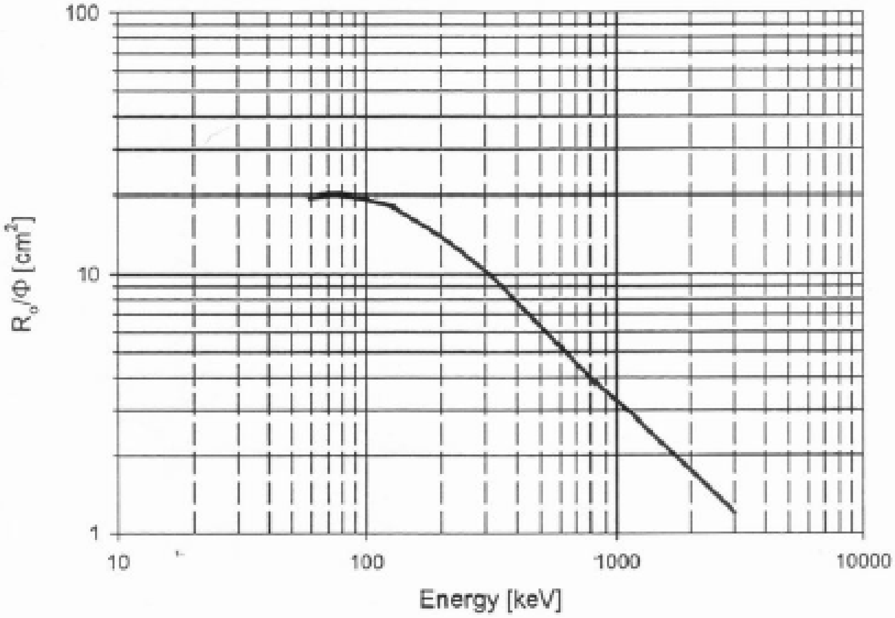
Angular correction factor

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.

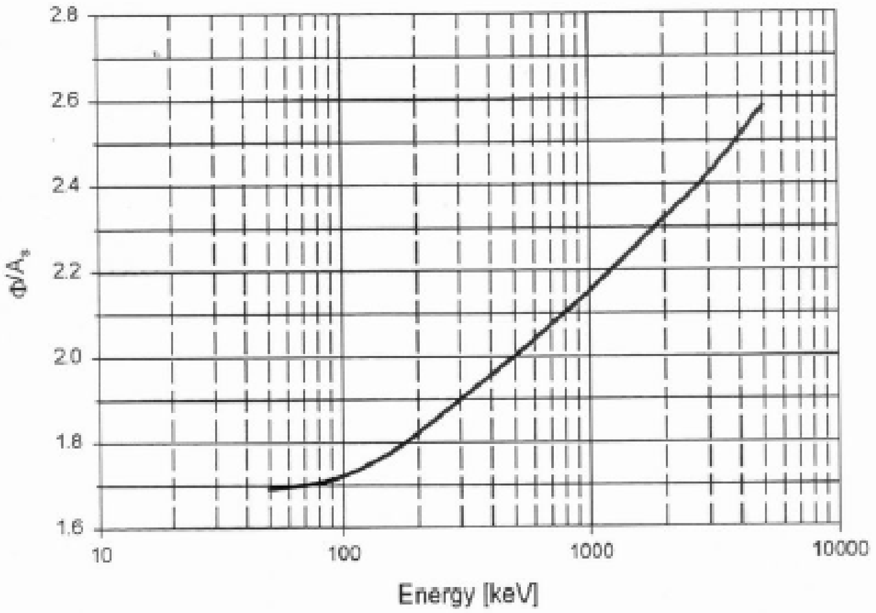


Examples of a detector characteristics

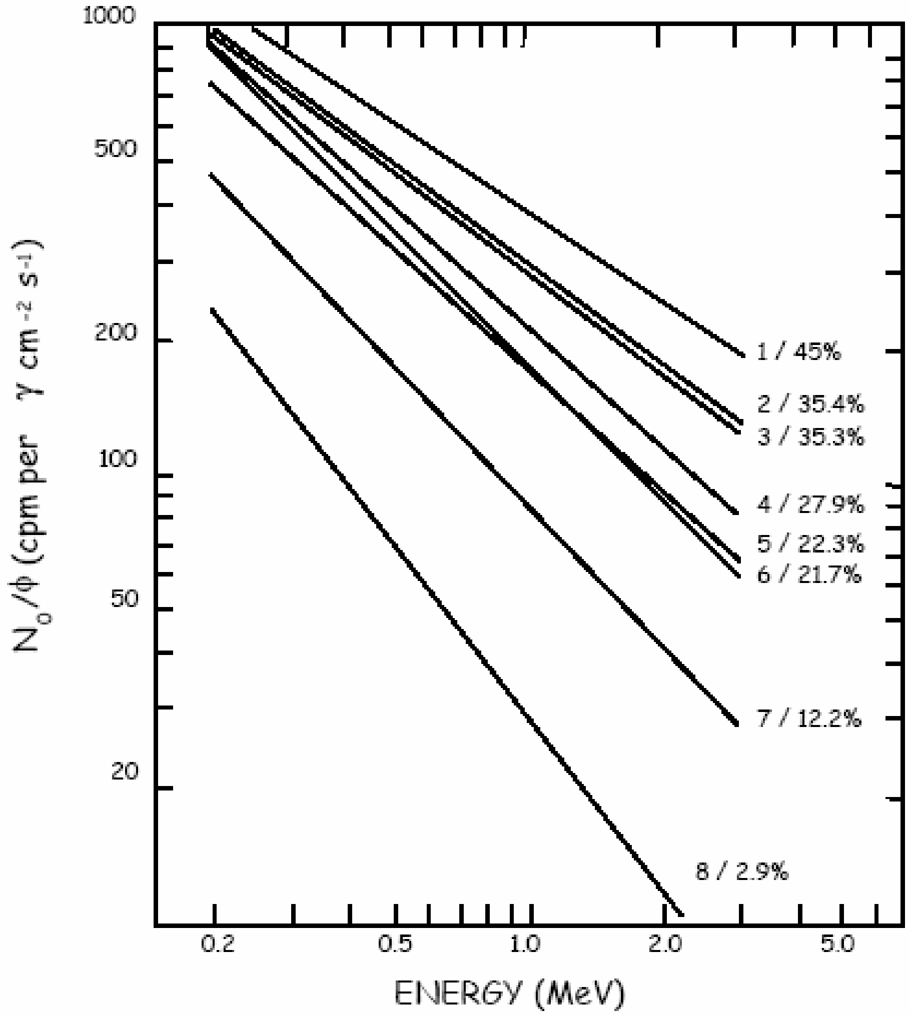
Typical **response factor**, R_0/Φ, for a Ge detector of 22% relative efficiency



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

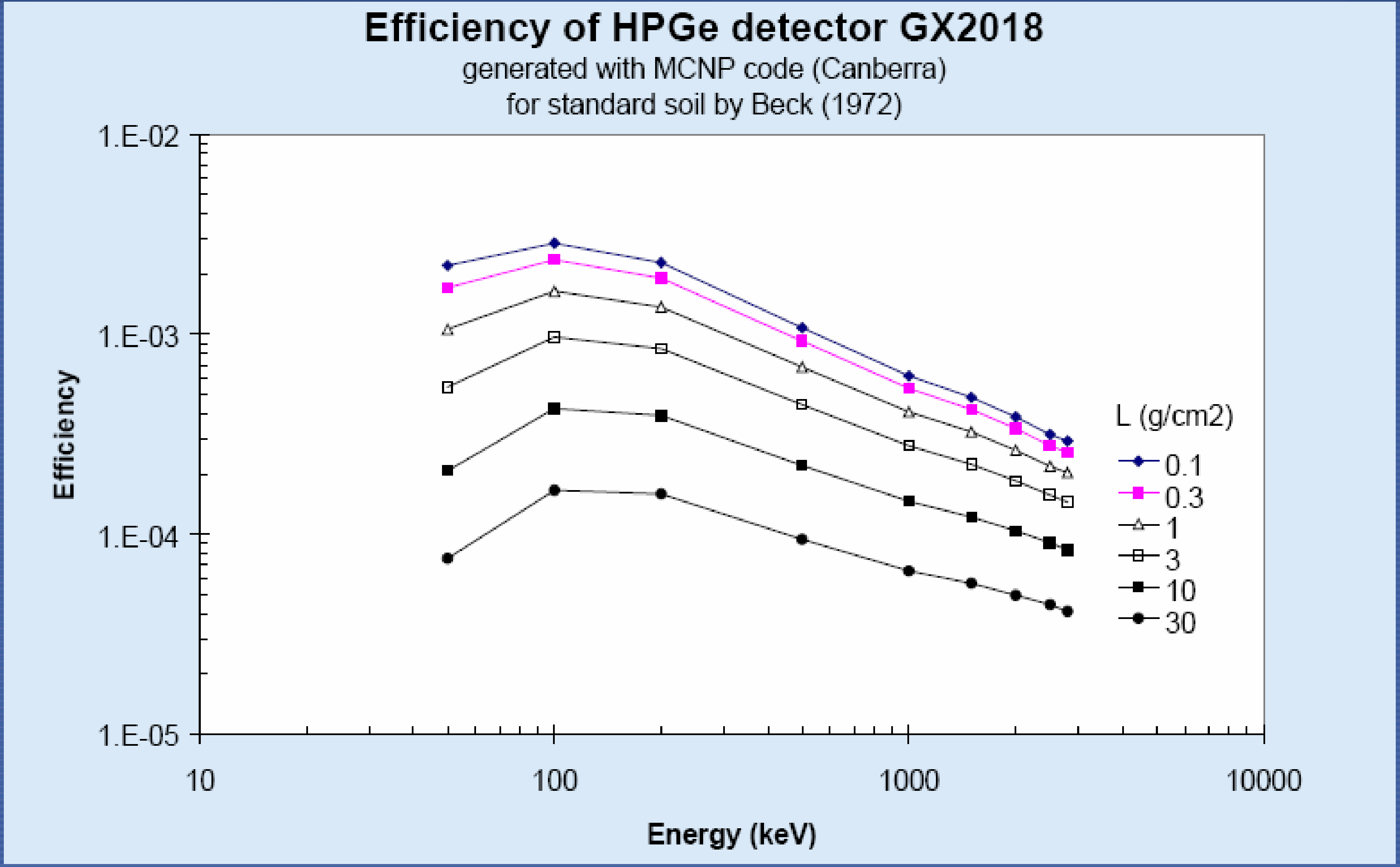


Response factor – a quick reference



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

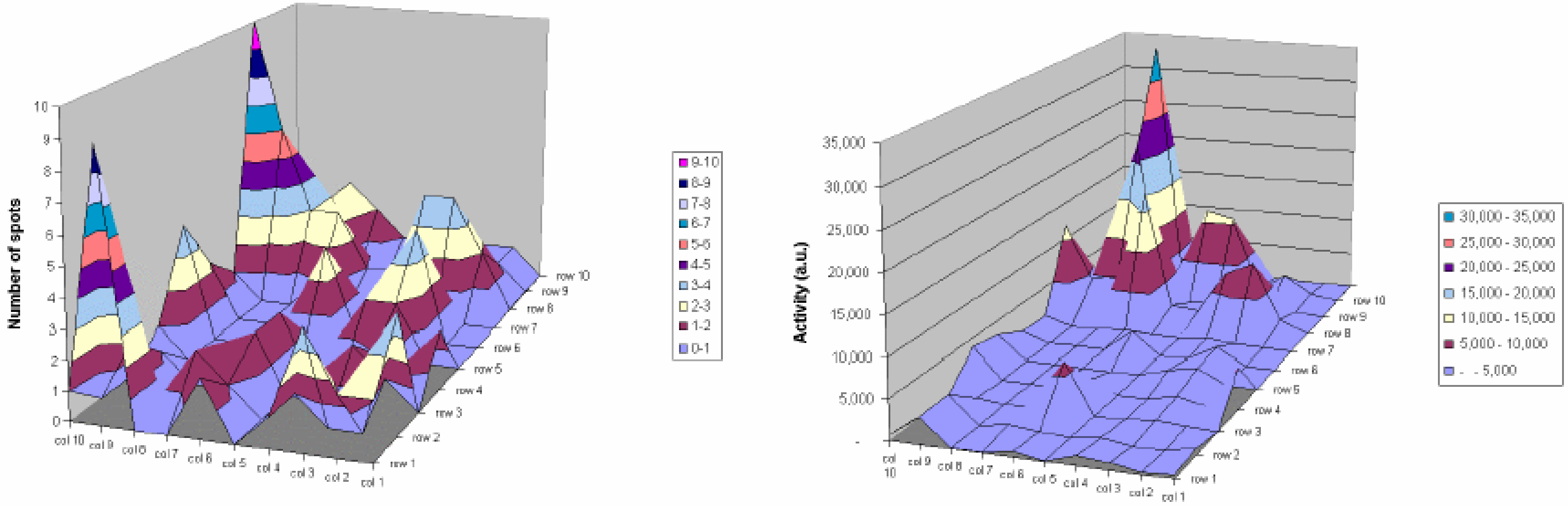


Sources of errors

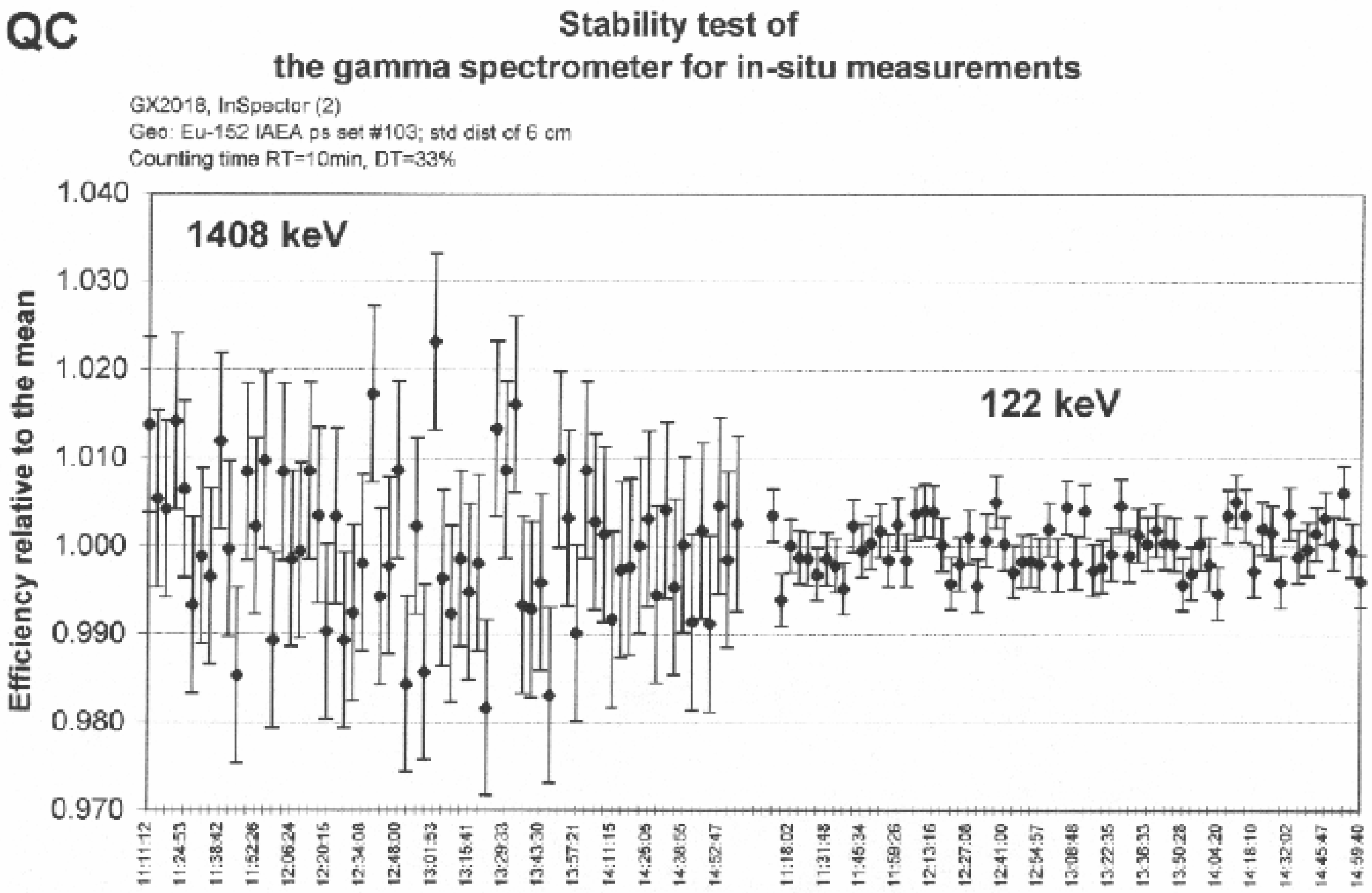
	Error
Radiation situation is not adequately well described by a mathematical model used/available (lack of proper model) - departure of real depth distribution from the model used, - obstacles, e.g. building, wall, etc. (modified "field-of-view" –departure from a conical shape) - nonuniform surface, 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

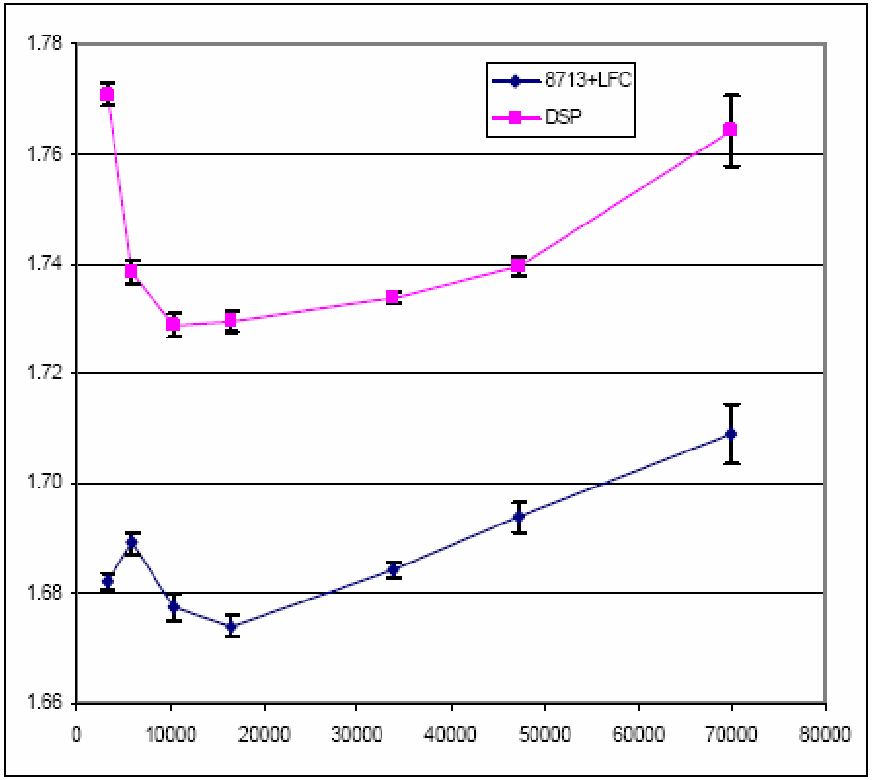


Monitoring the system performance



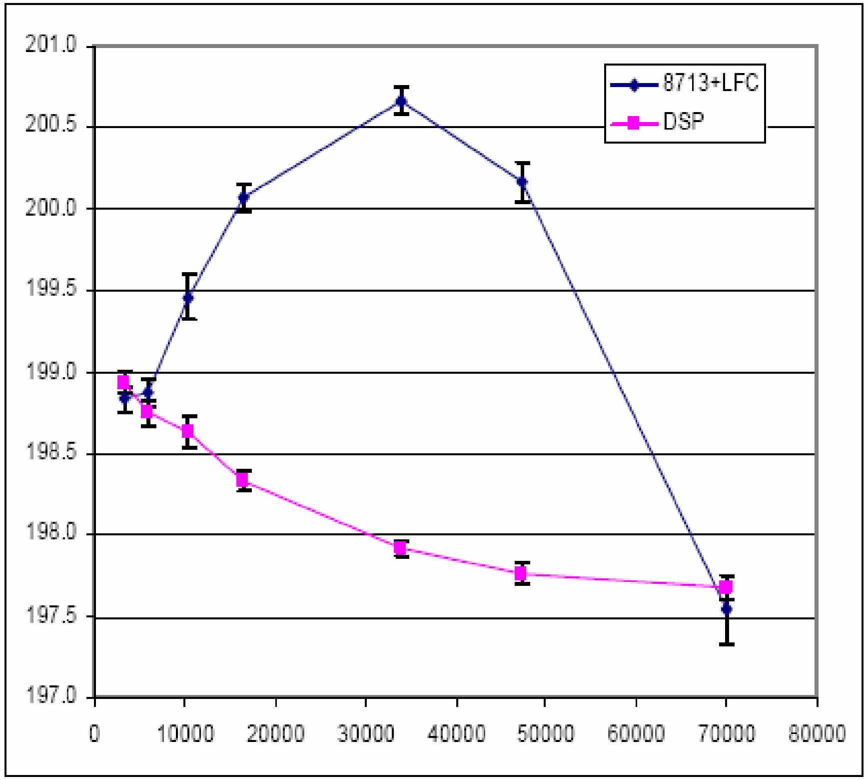
Effect of count rate in well adjusted sytem

Resolution, FWHM (keV)



Input count rate (cps)

Net peak area (cps)



Input count rate (cps)

References

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Marek Makarewicz IAEA

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