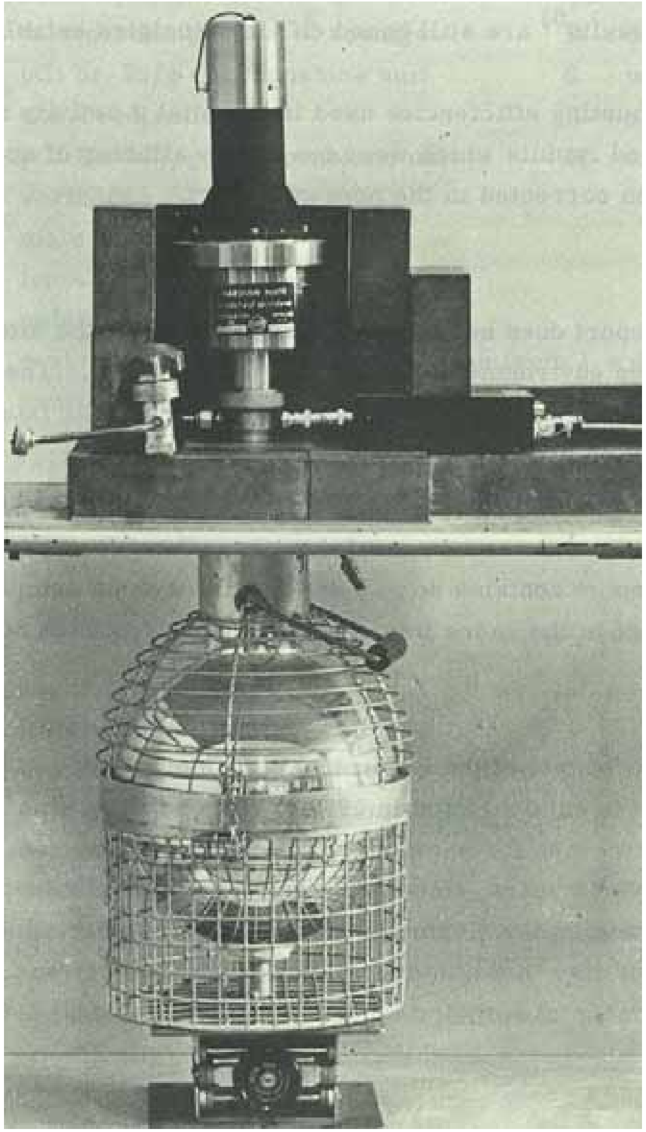
Gamma Spectrometry at Risø for Environmental Radioactivity

Sven Nielsen

Risø DTU
National Laboratory for Sustainable Energy

Gamma Background

- Measurements of environmental radioactivity started at Risø in 1956 using Geiger-Müller equipment (e.g. Anton Electronic Laboratories, New York)
- Gamma spectrometry (air filters) started in 1960 using a 4-inch NaI well detector and a 100-channel pulse height analyzer
- Gamma spectrometry using small home-made Ge(Li) detectors (e.g. 2 cm^3) started in 1965 with 1024 channel analyzer
- Commercial Ge(Li) detectors used from 1974, own production stopped



1966 Compton rejection setup

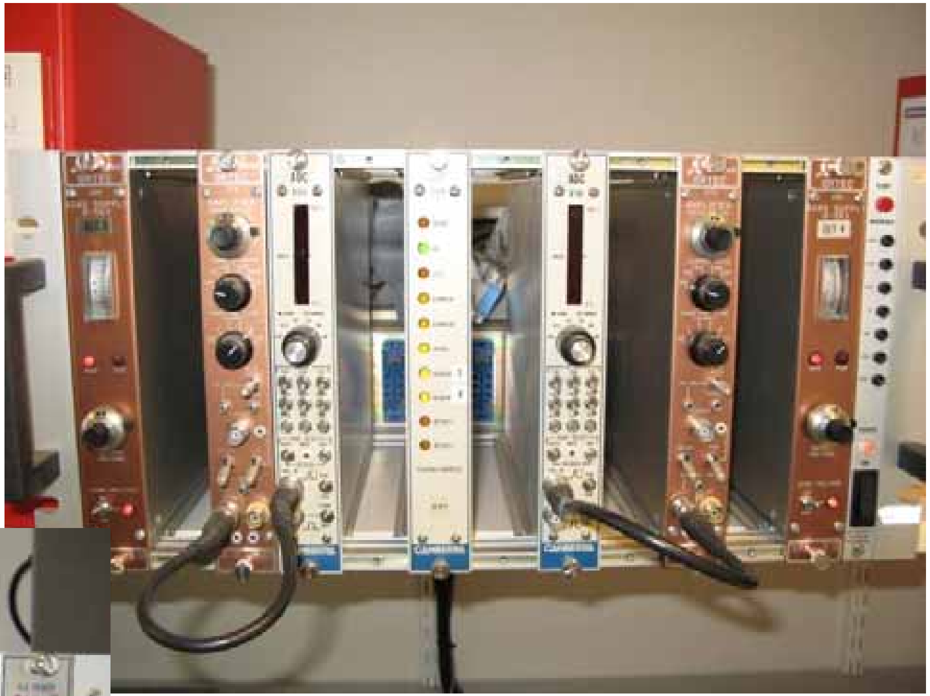
Gamma Laboratory

Lead Shields

Ge Detector Specifications

Risø id.	Producer	Year	Efficiency	Fwhm (keV)	Other
1	Ortec	1986	38%	1.8	1.3 mm Al window
2	Ortec	1986	35%	1.8	1.3 mm Al window
3	Ortec	1986	33%	1.9	Low energy, 0.5 mm Be window
4	Ortec	1986	33%	1.9	Low energy, 0.5 mm Be window
5	Canberra	1987	35%/180 cm^3	2.0	Low energy, Mg well, low background
6	Canberra	1998	118 cm^3	1.8	Low energy, 0.5 mm carbon epoxy, low background
7	Canberra	2001	260 cm^3	2.3	Low energy, Al well, low background
414	PGT	1979	25%	1.8	
423	PGT	1978	27%	2.0	
952	Ortec	1995	37%	1.8	Low energy, 0.5 mm Be window, low background

Electronics

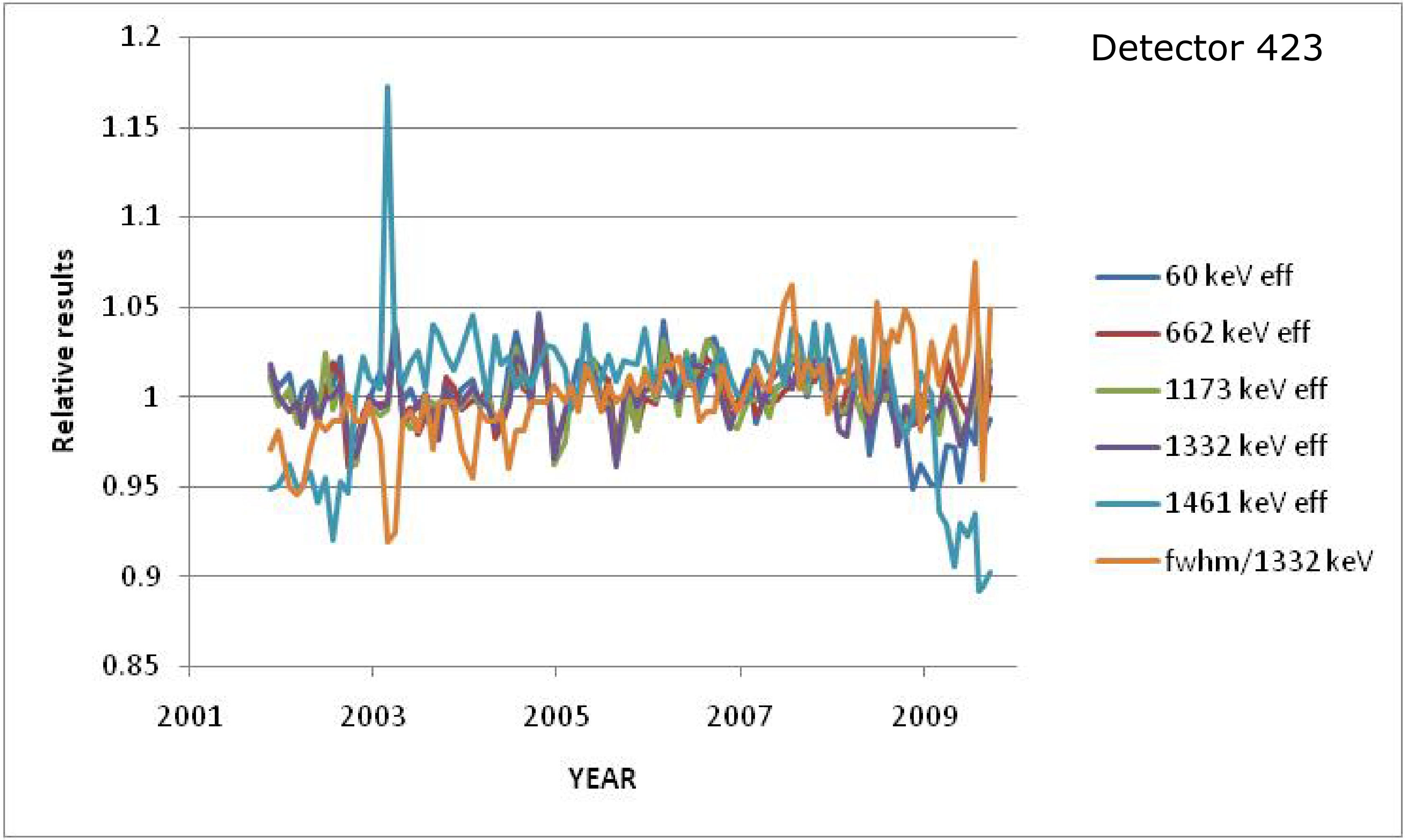


Sample Geometries

- 1-L Marinelli beaker (1 L)
- 210 mL cylindrical beaker (range 20-210 mL)
- 25 mL Petri dish (5, 10 and 15 mL)
- 10 mL vial (range 1-8 mL)
- 2 mL vial (range 0.2-2 mL)

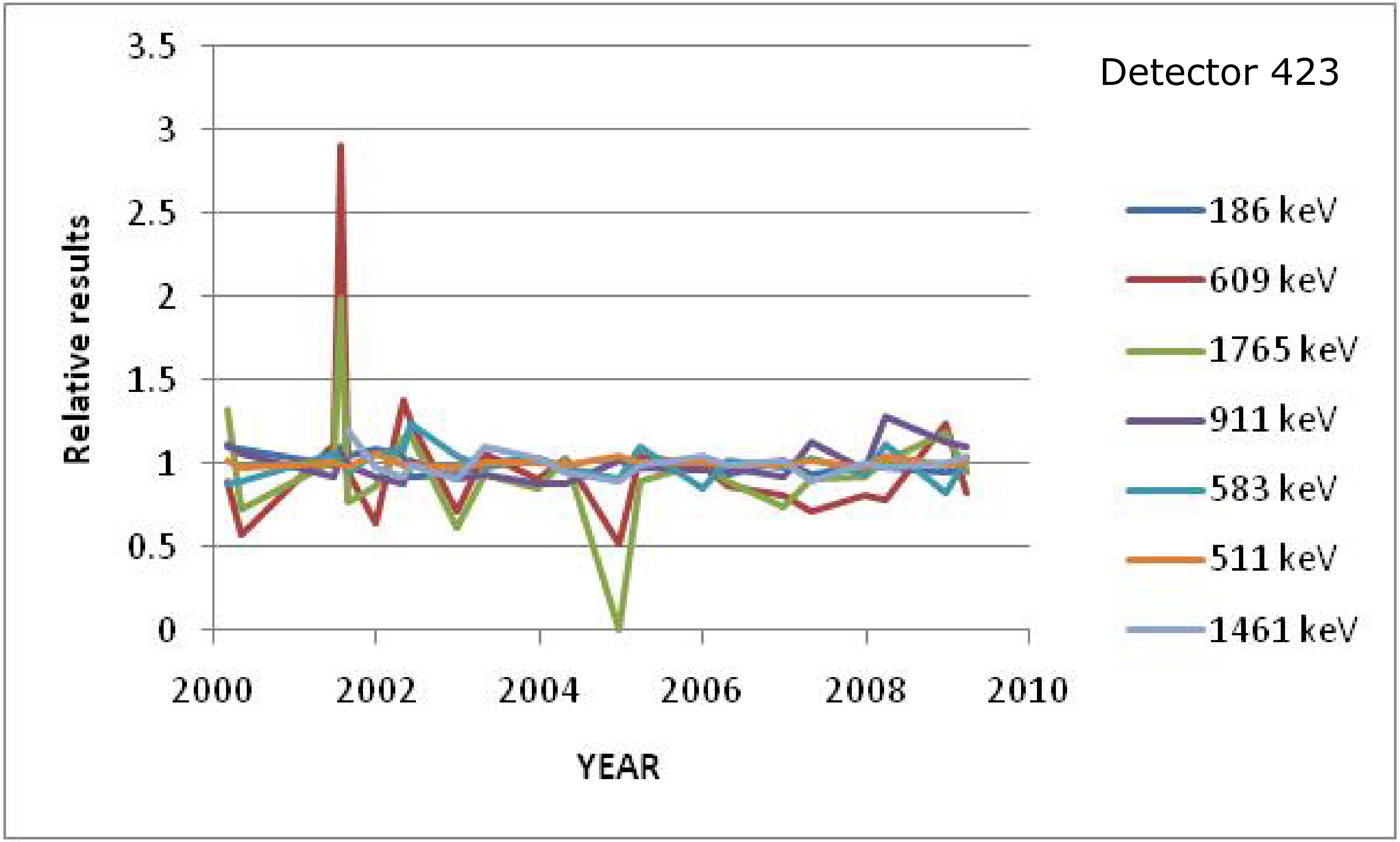
Detector Efficiency and Energy Resolution

- Effeciency and energy resolution of detectors checked monthly with reference sources ^{241}Am, ^{137}Cs, ^{60}Co and ^{40}K



Detector Gamma Background

- Ge detector background counts performed during prolonged holidays, i.e. Easter and Christmas



Software for Gamma Spectrum Analysis

- Home made software, developed since 1970’s, implemented first in Algol programming language on main-frame computer, later in C on personal computer
- Peak search based on values of second derivative of smoothed spectrum
- Peak area calculation based on simple summation of smoothed spectrum counts over peak channels minus background, fitting of doublets
- Accuracy of peak-area calculation method compared with other procedures (1998)

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--------- Måling nr. 405486 ---------
1: Sample type: Milk
2: Date       : 2009-Aug
3: Location   : W-Jutland 3
6: Sample ID  : 20090327
Res.el.vægt:             2.0000  kg dry
1: Detektor   :       4,       4
2: Måleperiode: 20090812.1138,20090817.0847
3: Fyldning   :            -0.4000
4: Vægtfylde  :             0.6100
5: Energikal. :   2.0606,  0.6687
6: Måletid    :            421797
Spektrum: 4000 kanaler
TOPAREAL fil A -> B, t = 1.5, max.eta = 40 %
 41 - 5979 kan., delta = 2.5 keV
  br. fra kalib.  w1: 5, w2: 13, udglat = 3
 Isotoptabel indeholder 140 isotoper
Milk from Videbæk august 2009
Spektrum nr. 405486, detektor 4, kalibrering 4
Kan: KeV:   w:(w0) b: Bagg: Eta: cps*1000: Eta: Bq(100%)  Vfk.: Bq(vf):
438 761  510.6  7295.0  6( 6) 1.2 1.81  9.9     -0.22 100.0
439 ( 7) 2.525.41  1.0   5.25  11.2     0.303 0.932     0.283 ( 3)
 907  608.9  7( 7) 1.9 2.96  5.5     -0.09 100.0
 986  661.2  7( 7) 1.6              10.14   4.4     0.739 0.940     0.695 ( 4)
2180 1459.8  9( 9) 2.2 2.81  3.7    655.66   0.2    92.579 0.958    88.733 ( 6)
2634 1763.0 10(10) 2.9 0.87  9.6     -0.13 100.0
3905 2613.4 13(13) 3.1 1.69  4.5      0.00 100.0
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Sample output

Tests of accuracy versus that of data set no. 4

Data no.	Software	Type	DF	T	χ^2-Reduced	Sign.
1	CompAct	Simple	21	15.9	0.76	ns
2	GammaVision	Simple	21	202	9.62	***
3	GammaVision	Simple	21	195	9.29	***
4	GammaVision	Simple	21	21.9	1.04	ns
5	Genie-PC	Simple	21	40.0	1.90	*
6	Genie-PC	Simple	21	38.5	1.83	*
7	C-Base	Simple	21	38.2	1.82	*
8	Genie-PC	Fitting	21	18.4	0.88	ns
9	GAMANAL	Fitting	20	32.1	1.61	ns
10	GRILS	Fitting	20	269	13.5	***
11	EMCAPLUS	Fitting	21	11.0	0.52	ns
12	ANSP	Fitting	21	9.8	0.47	*
13	GammaTrac	Fitting	21	21.0	1.00	ns
14	GammaTrac	Fitting	21	53.9	2.57	***
15	GAMMA-96	Other	21	19.1	0.91	ns

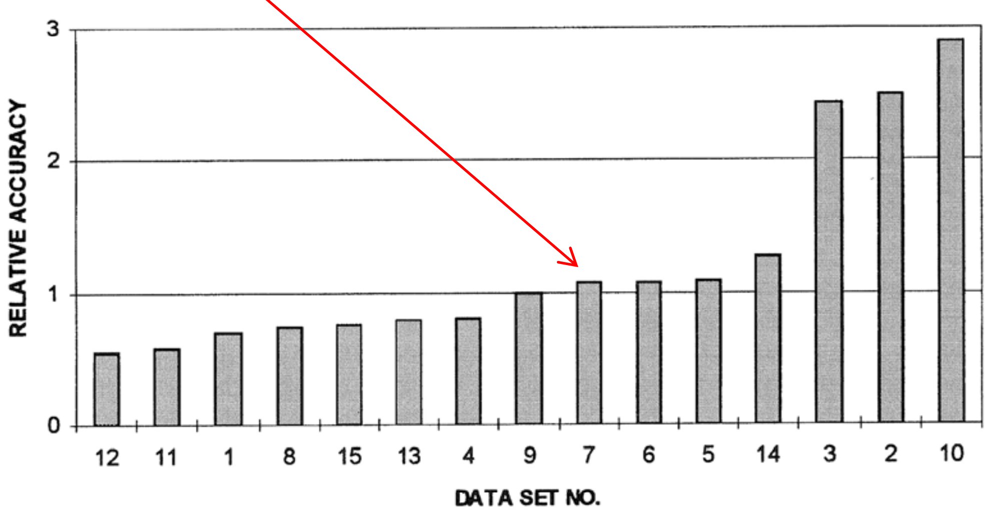
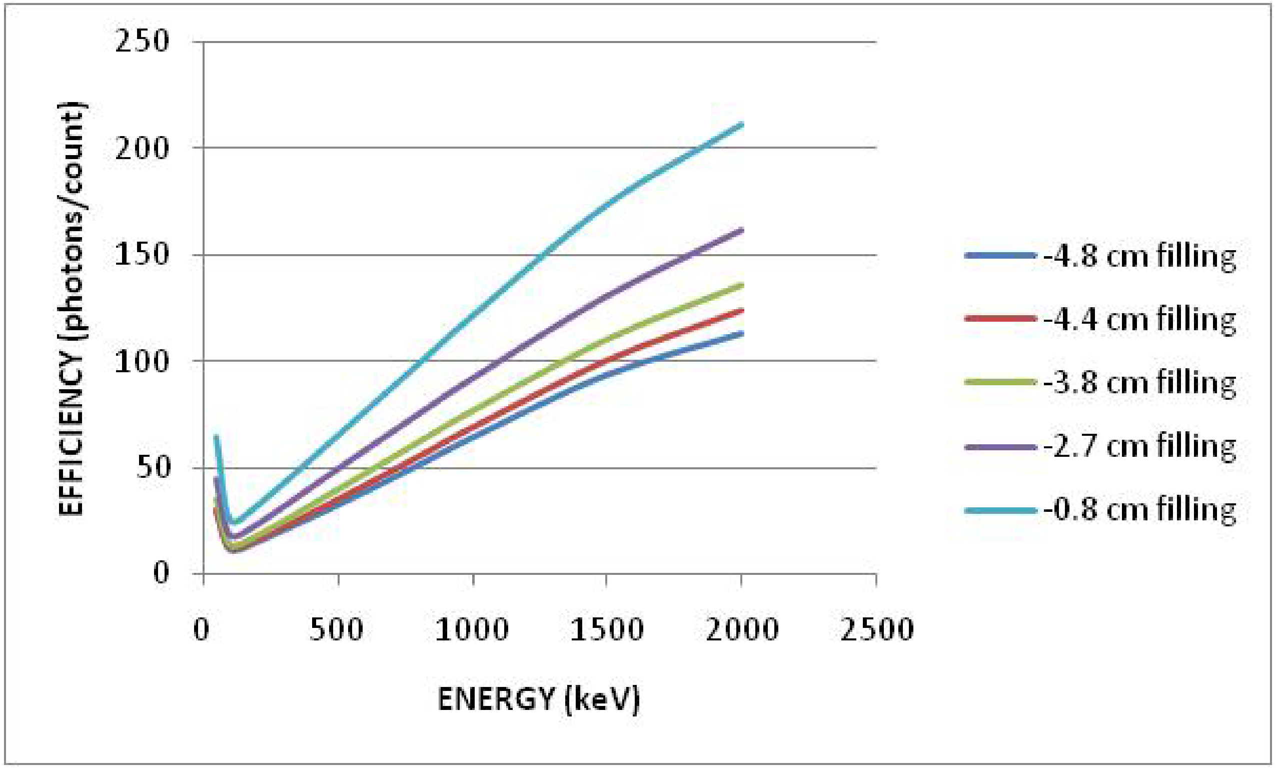


Fig. 3. Plot of relative average accuracies of peak-area ratios for the data sets.

Efficiency Calibration

- Calibrations based on measurements in standardized geometries of known activities of mixed radionuclide gamma-ray reference solutions and K_2CO_3 standard, e.g. ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co
- Calibration curves fitted to measured efficiencies (photons/count) using polynomial expressions



Detector 423 efficiency calibration

Coincidence Summing

- Coincidence summing correction factors determined experimentally as deviations between observed efficiencies and calibration curves

Nuclide	Energy (keV)	-4.8 cm filling	-4.4 cm filling	-3.8 cm filling	-2.7 cm filling	-0.8 cm filling
57Co	122	0.98	0.99	0.99	0.99	1.00
57Co	136	0.93	0.89	0.93	0.96	0.95
60Co	1173	1.00	1.00	1.00	1.00	1.00
60Co	1332	0.99	0.99	1.00	1.00	0.99
134Cs	605	1.18	1.17	1.15	1.14	1.15
134Cs	796	1.13	1.13	1.11	1.10	1.11
134Cs	802	1.22	1.23	1.21	1.18	1.18
226Ra	186	0.47	0.47	0.49	0.48	0.46
226Ra	352	1.05	1.04	1.07	1.04	1.04
226Ra	609	1.17	1.16	1.16	1.15	1.14
226Ra	1765	0.95	0.95	1.00	0.99	0.99

Excerpt of coincidence summing correction factor table for five different fillings of the 210 mL geometry for detector 423

Density Correction

- Density correction based on a mathematical model of Ge detector, sample geometry and density (Lippert 1983)
- Correction factor CF calculated as

$$CF = e^{\rho-1} \cdot xabs \cdot e^{m_0 - m_1 lnE_\gamma}$$

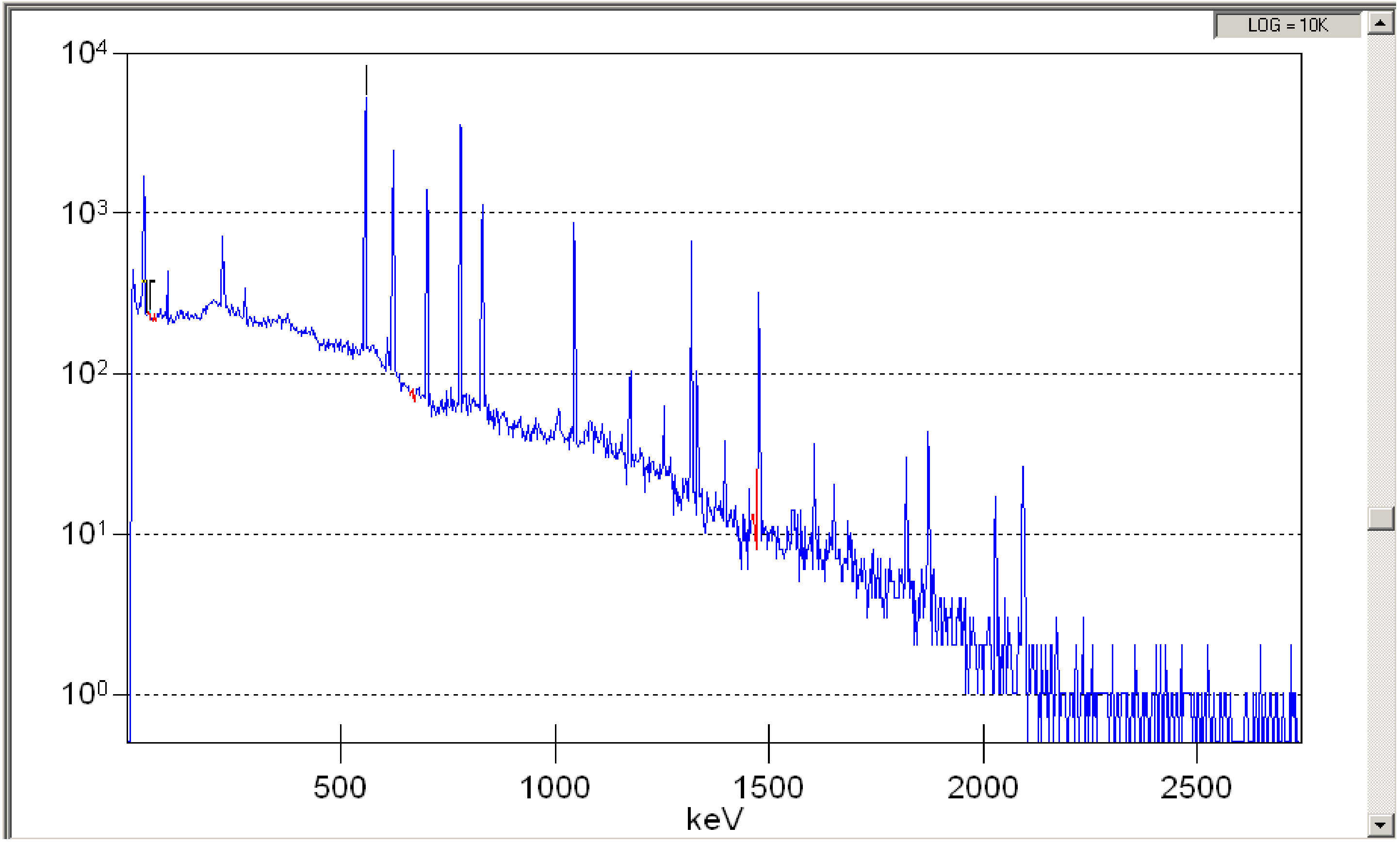
- Where ρ is sample density, xabs characteristic length for sample geometry, m_0 and m_1 constants, and E_γ gamma energy.
- Example correction factors

Gamma energy (keV)	210 mL cylinder 178 mL	210 mL cylinder 59 mL
100	1.33	1.13
500	1.14	1.06
1000	1.10	1.04
1500	1.08	1.03

- Furthermore, for measurements of ^{210}Pb at 47 keV, correction for self absorption is applied by experimental determination of attenuation using a ^{210}Pb point source

Detection Limit (DL)

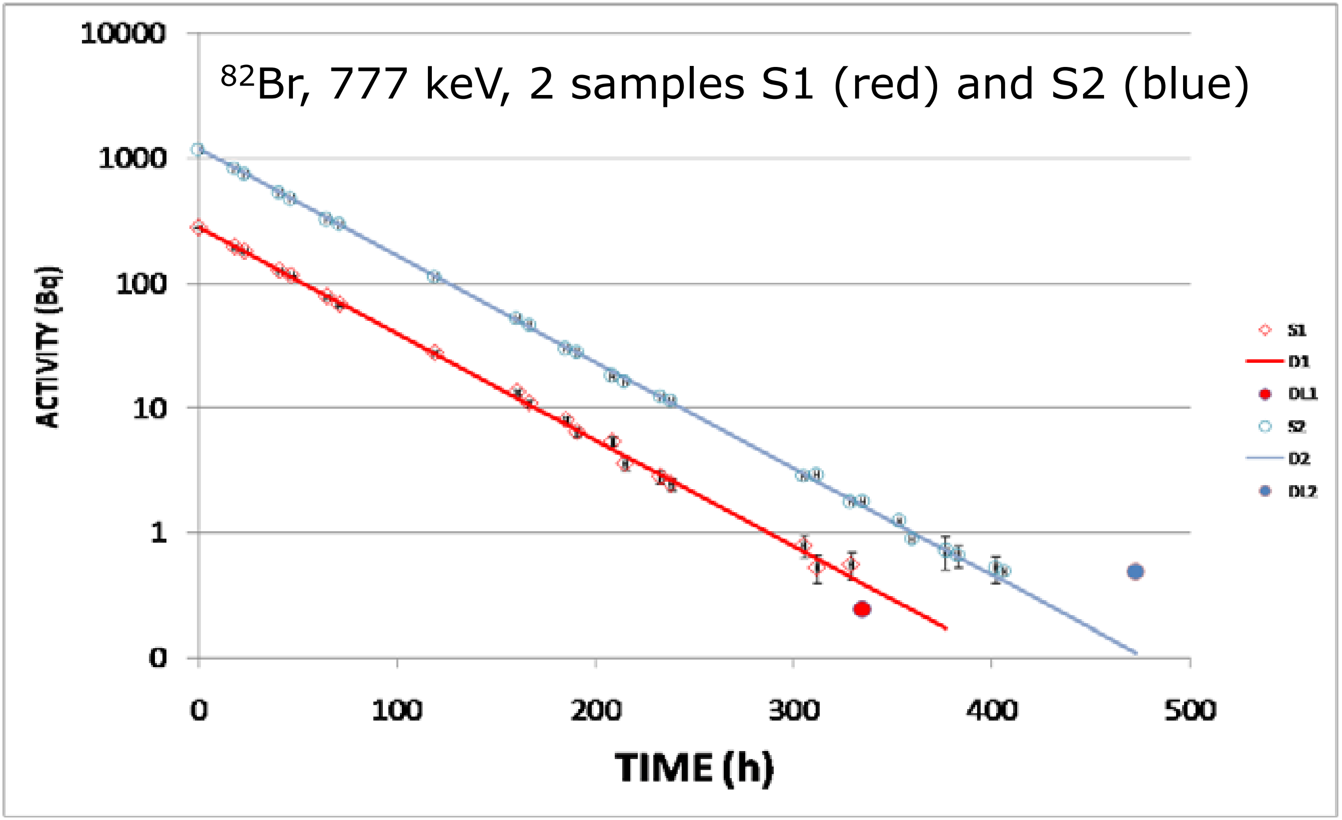
- Calculation based on Currie (1968) : $DL = 2.71 + 4.65 \cdot SD_{bgr}$ (95% probability)
- Applied by summing spectrum counts over 2 fwhm at energy in question
- Test by frequent gamma measurements of ^{82}Br samples ($T_{1/2}$=1.5d) for several weeks until all peaks below detection limit



^{82}Br gamma spectrum

DL Test

- Two ^{82}Br samples measured daily for 30 min and evaluated for prominent gamma energies
- DL calculated when peak disappeared from spectrum
- Value of DL compared with activity value
- DL values were higher than activity values in 22 out of 25 gamma peaks



Future Plans

- Will change to commercial gamma analysis system, probably Genie2000

NKS-B Workshop on

Radioanalytical Chemistry for Radioecology and Waste Management

Risø-DTU, Roskilde, Denmark,

16th-20th November, 2009

Registration deadline: 1st of October

Contact person: Xiaolin Hou

E-mail: xiho@risoe.dtu.dk

Topics of the Lectures (16-18th Nov. 2009):

- General aspects in Radiochemical Analysis for Radionuclides;
- Separation techniques for determination of radionculides
- Radiochemical separation methods of various radionuclides including isotopes of Pu, U, Th, and Ra, ^{237}Np, ^{241}Am, ^{210}Po, 210Pb, ^{99}Tc, ^{90}Sr, ^{137}Cs, ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, and ^{129}I.
- Sampling and pre-concentration techniques for environmental radioactivity analysis
- Radiometric analytical techniques including γ- and α-spectrometry, beta counting with Ultra-low level background G-M counter, and Liquid Scintillation counting.
- Mass spectrometric for long-lived radionuclides including ICP-MS, AMS.
- Automated and rapid analytical technique for radionuclides.
- Techniques for speciation analysis of radionuclides

Laboratory practices (19th-20th Nov. 2009)

Three groups:

- Radiochemcial separation of ^{90}Sr, ^{210}Po, ^{226}Ra and γ- and α-spectrometry measurement
- Radiochemcial separation of ^{99}Tc, Pu isotopes and ICP-MS measurement
- Radiochemcial separation of ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni, and LSC measurement.