Radiometric Measurements of Environmental Radioactivity
Beta Counting, Alpha and Gamma Spectrometry

Nielsen, Sven Poul

Publication date: 2013

Citation (APA):
Radiometric Measurements of Environmental Radioactivity

Beta Counting, Alpha and Gamma Spectrometry

Sven Nielsen

DTU Nutech
Center for Nuclear Technologies
Early start at Risø

- Measurements of environmental radioactivity started at Risø in 1956 using Geiger-Müller equipment (e.g. Anton Electronic Laboratories, New York)
- Alpha/beta measurements using proportional counters from 1960
- 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 Ge(Li) detectors (e.g. 2 cm³) made at Risø started in 1965 with 1024 channel analyzers
- Alpha spectrometry using Si detectors and 256-ch MCAs from 1968
- Commercial Ge(Li) detectors used from 1974, own production stopped
Risø Low-Level Beta GM Multicounter
Description

• Low-level gas-flow beta multicounter for simultaneous measurements of 5 samples.
• Incorporates 5 individual counter elements and a common guard counter
• Guard counter uses anti-coincidence technique to reduce background from cosmic radiation by more than a factor 700
• Aluminized Mylar window of thickness less than 1 mg/cm²
• Counter gas 99% Ar / 1% isobutane or propane
• Counter placed in 10-cm lead shield
Specifications and Applications

- Multicounters used for total beta counting of $^{90}$Sr and $^{99}$Tc
- Guard count rates typically about 2 cps
- Detector background count rates typically about 0.2 cpm
- Detector efficiencies checked monthly using reference sources, $^{36}$Cl and $^{99}$Tc
- Counting efficiencies about 58% for $^{90}$Y sources mounted on steel cores and about 40% for $^{99}$Tc sources on steel disks
- Detection limits for routine applications about 2 mBq for $^{90}$Sr and 4 mBq for $^{99}$Tc
- Results from spreadsheet calculations

---

**Multicounter 10 Background**

**Strontium-90 in Danes**
Alpha Spectrometry
Risø Vacuum Chamber
Description

- 32 Si detectors
- Vacuum chambers made at Risø
- 20 mm diameter sources electrodeposited on stainless steel disks
- Counting times typically 3-7 days
- Detection limits 0.1-0.2 mBq
- Results from spreadsheet calculations
- Analyses of Po, U, Pu, Am, (Np)
Gamma Laboratory

DTU Nutech, Technical University of Denmark
Lead Shields
## Ge Detector Specifications

<table>
<thead>
<tr>
<th>Risø id.</th>
<th>Producer</th>
<th>Year</th>
<th>Efficiency</th>
<th>Fwhm (keV)</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ortec</td>
<td>1986</td>
<td>38%</td>
<td>1.8</td>
<td>1.3 mm Al window</td>
</tr>
<tr>
<td>2</td>
<td>Ortec</td>
<td>1986</td>
<td>35%</td>
<td>1.8</td>
<td>1.3 mm Al window</td>
</tr>
<tr>
<td>3</td>
<td>Ortec</td>
<td>1986</td>
<td>33%</td>
<td>1.9</td>
<td>Low energy, 0.5 mm Be window</td>
</tr>
<tr>
<td>4</td>
<td>Ortec</td>
<td>1986</td>
<td>33%</td>
<td>1.9</td>
<td>Low energy, 0.5 mm Be window</td>
</tr>
<tr>
<td>5</td>
<td>Canberra</td>
<td>1987</td>
<td>35%/180 cm³</td>
<td>2.0</td>
<td>Low energy, Mg well, low background</td>
</tr>
<tr>
<td>6</td>
<td>Canberra</td>
<td>1998</td>
<td>118 cm³</td>
<td>1.8</td>
<td>Low energy, 0.5 mm carbon epoxy, low background</td>
</tr>
<tr>
<td>7</td>
<td>Canberra</td>
<td>2001</td>
<td>260 cm³</td>
<td>2.3</td>
<td>Low energy, Al well, low background</td>
</tr>
<tr>
<td>414</td>
<td>PGT</td>
<td>1979</td>
<td>25%</td>
<td>1.8</td>
<td>Ge(Li)</td>
</tr>
<tr>
<td>423</td>
<td>PGT</td>
<td>1978</td>
<td>27%</td>
<td>2.0</td>
<td>Ge(Li)</td>
</tr>
<tr>
<td>952</td>
<td>Ortec</td>
<td>1995</td>
<td>37%</td>
<td>1.8</td>
<td>Low energy, 0.5 mm Be window, low background</td>
</tr>
</tbody>
</table>
Electronics
Sample Geometries

- 1-L Marinelli beaker (1 L)
- 210 mL cylindrical beaker (range 20-200 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

- Efficiency and energy resolution of detectors checked monthly with reference sources $^{241}\text{Am}$, $^{137}\text{Cs}$, $^{60}\text{Co}$ and $^{40}\text{K}$
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)

Sample output

--------- 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, detector 4, kalibrering 4
Kan: KeV: w:(w0) b: Bagg: τ: Eta: cps*1000: Eta: Bq(100%): Vfk.: Bq(vf):
438761 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

-----------------------
Tests of accuracy versus that of data set no. 4

<table>
<thead>
<tr>
<th>Data no.</th>
<th>Software</th>
<th>Type</th>
<th>DF</th>
<th>T</th>
<th>χ²-Reduced</th>
<th>Sign.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CompAct</td>
<td>Simple</td>
<td>21</td>
<td>15.9</td>
<td>0.76</td>
<td>ns</td>
</tr>
<tr>
<td>2</td>
<td>GammaVision</td>
<td>Simple</td>
<td>21</td>
<td>202</td>
<td>9.62</td>
<td>***</td>
</tr>
<tr>
<td>3</td>
<td>GammaVision</td>
<td>Simple</td>
<td>21</td>
<td>195</td>
<td>9.29</td>
<td>***</td>
</tr>
<tr>
<td>4</td>
<td>GammaVision</td>
<td>Simple</td>
<td>21</td>
<td>21.9</td>
<td>1.04</td>
<td>ns</td>
</tr>
<tr>
<td>5</td>
<td>Genie-PC</td>
<td>Simple</td>
<td>21</td>
<td>40.0</td>
<td>1.90</td>
<td>*</td>
</tr>
<tr>
<td>6</td>
<td>Genie-PC</td>
<td>Simple</td>
<td>21</td>
<td>38.5</td>
<td>1.83</td>
<td>*</td>
</tr>
<tr>
<td>7</td>
<td>Genie-PC</td>
<td>Simple</td>
<td>21</td>
<td>38.2</td>
<td>1.82</td>
<td>*</td>
</tr>
<tr>
<td>8</td>
<td>Genie-PC</td>
<td>Fitting</td>
<td>21</td>
<td>18.4</td>
<td>0.88</td>
<td>ns</td>
</tr>
<tr>
<td>9</td>
<td>GAMANAL</td>
<td>Fitting</td>
<td>20</td>
<td>32.1</td>
<td>1.61</td>
<td>ns</td>
</tr>
<tr>
<td>10</td>
<td>GRILS</td>
<td>Fitting</td>
<td>20</td>
<td>269</td>
<td>13.5</td>
<td>***</td>
</tr>
<tr>
<td>11</td>
<td>EMCAPLUS</td>
<td>Fitting</td>
<td>21</td>
<td>11.0</td>
<td>0.52</td>
<td>ns</td>
</tr>
<tr>
<td>12</td>
<td>ANSP</td>
<td>Fitting</td>
<td>21</td>
<td>9.8</td>
<td>0.47</td>
<td>*</td>
</tr>
<tr>
<td>13</td>
<td>GammaTrac</td>
<td>Fitting</td>
<td>21</td>
<td>21.0</td>
<td>1.00</td>
<td>ns</td>
</tr>
<tr>
<td>14</td>
<td>GammaTrac</td>
<td>Fitting</td>
<td>21</td>
<td>53.9</td>
<td>2.57</td>
<td>***</td>
</tr>
<tr>
<td>15</td>
<td>GAMMA-96</td>
<td>Other</td>
<td>21</td>
<td>19.1</td>
<td>0.91</td>
<td>ns</td>
</tr>
</tbody>
</table>

![Fig. 3. Plot of relative average accuracies of peak-area ratios for the data sets.](image)

DTU Nutech, Technical University of Denmark
Efficiency Calibration

- Calibrations based on measurements in standardized geometries of known activities of mixed radionuclide gamma-ray reference solutions and K$_2$CO$_3$ 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
True Coincidence Summing Correction

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

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

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

\[ CF = e^{\rho - 1} \cdot x_{abs} \cdot e^{m_0 - m_1 \ln E_{\gamma}} \]

- Where \( \rho \) is sample density, \( x_{abs} \) characteristic length for sample geometry, \( m_0 \) and \( m_1 \) constants, and \( E_{\gamma} \) gamma energy.
- Example correction factors

<table>
<thead>
<tr>
<th>Gamma energy (keV)</th>
<th>210 mL cylinder 178 mL</th>
<th>210 mL cylinder 59 mL</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>1.33</td>
<td>1.13</td>
</tr>
<tr>
<td>500</td>
<td>1.14</td>
<td>1.06</td>
</tr>
<tr>
<td>1000</td>
<td>1.10</td>
<td>1.04</td>
</tr>
<tr>
<td>1500</td>
<td>1.08</td>
<td>1.03</td>
</tr>
</tbody>
</table>

- 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
Commercial software for gamma analysis

- Using Genie2000 for single application under formal QA/QC
  - Test of radiometric purity of eluate from $^{99}$Mo/$^{99m}$Tc generator
- Advantages of commercial software
  - More user friendly and with better future prospects than home made software
  - Features and documentation prepared for formal QA/QC
  - Documentation available
Future

- Full use of Genie2000 software