



**Quantification of Activity from
Volumetric Samples Using
Mathematical Efficiency Approaches**

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Introduction

- ▶ Background on Mathematical Modeling
- ▶ Validity of Mathematical Modeling as a General Purpose Efficiency Calibration Tool
- ▶ Advantages of Mathematical Modeling
 - ◆ Flexibility of Analysis
 - ◆ Corrections for Cascade Summing Losses
 - ◆ Handling Uncertainty
 - ◆ Measurement Optimization
- ▶ Advanced Features/Future Developments
- ▶ Summary



Setting up a detector for measurement

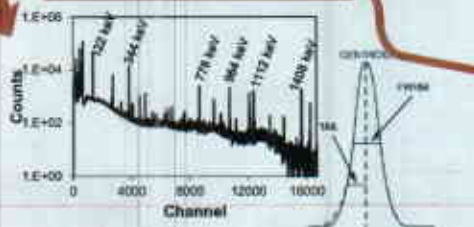
▶ Setup Hardware

- ◆ Peak Shaping Parameters
 - Rise Time
 - Flat Top
- ◆ Pole-zero
- ◆ Number of Channels
- ◆ Signal Gain



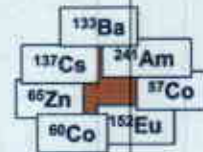
▶ Setup Software

- ◆ Energy Calibration
- ◆ Shape Calibration
- ◆ Efficiency Calibration



◆ Select Sources

- Energy Range
- Half-life
- Cascade effects
- Activity



◆ Select Geometries

- Beakers
- Vials

◆ Pay \$\$\$ for these sources



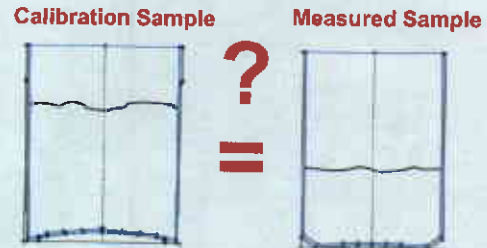
Source-based Efficiency Calibrations



► Cost of Source-based Calibration

- ◆ Purchase of Sources
- ◆ Replacement of Sources
- ◆ Disposal of Sources
- ◆ Licensing for Sources
- ◆ Calibration Program
- ◆ Sample Preparation

Expensive



► What if you can't make the sample match the calibration

- ◆ Not enough material
- ◆ Unusual material composition
- ◆ Locked in sealed container

Corrections required



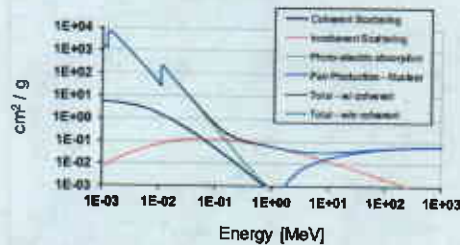
Gamma-ray Interactions in Material



► Gamma-ray Interactions are very well understood.

- ◆ Photoelectric absorption
- ◆ Compton Scattering
- ◆ Pair Production

Energy dependence of gamma ray interaction in Germanium



- We can exploit this knowledge to create efficiency responses based on the physical parameters of the geometry.
- Use mathematical models to accurately compute the transport of gamma-rays through these geometries.

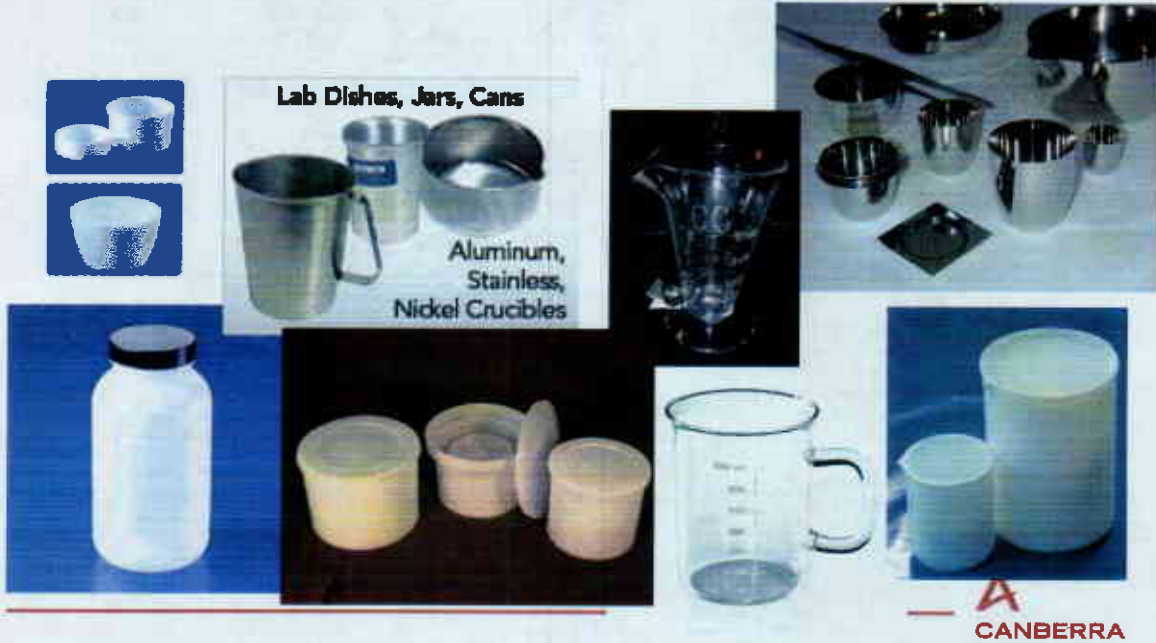




Modeling of Laboratory Samples



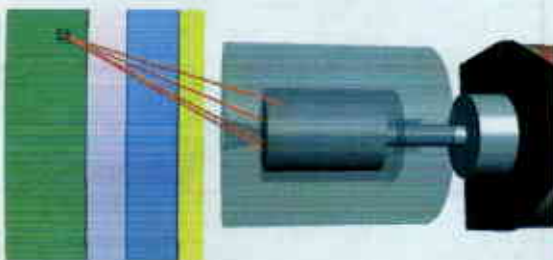
- ▶ How to handle varieties of samples and containers?



ISOCS/LabSOCS: A Generalized Efficiency Computation Method



- ▶ ISOCS/LabSOCS method is designed to quickly and accurately compute efficiencies for a wide range of geometries.
- ▶ Relies on a factory characterization of the intrinsic efficiency response of the detector.
- ▶ Does not require that the customer have a large inventory of calibrated sources (although nominal sources for quality control tracking is highly recommended).



$$\epsilon = \sum_i^{voxels} N_i \epsilon_i^{vac} \sum_j^{paths} W_{ij} e^{-\sum_k^{abs} \mu_{ijk} T_{ijk}}$$

U.S. Patent 6,228,664 B1

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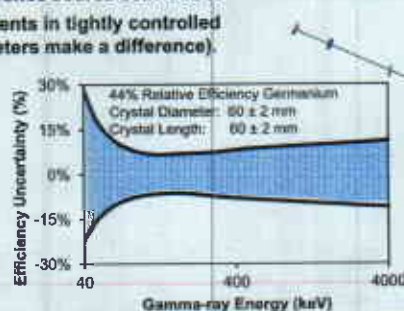
Regulatory Acceptance of Modeling Approaches

- ▶ **"Calibration of Germanium Detectors for In-Situ Gamma-ray Measurements", N42.28-2002 American National Standards Institute, Inc., 1430 Broadway, New York 10018.**
"One such application [of Monte Carlo Methods] is the calculation of the efficiency or response function for an HPGe detector. Using this approach, detectors can be calibrated for a variety of applications using models and simulations."
- ▶ **"Measuring, Evaluating, and Reporting Radioactive Material In Liquid and Gaseous Effluents and Solid Waste", U.S. Nuclear Regulatory Commission Guide 1.21 rev. 2 (June 2009):**
"The use of NIST-traceable sources combined with mathematical efficiency calibrations may be applied to instrumentation used for radiochemical analysis (e.g., gamma spectroscopy systems) if employing a method provided by the instrument manufacturer."
- ▶ **"A Good Practice Guide for the use of Modelling Codes in Non Destructive Assay of Nuclear Materials", ESARDA Bulletin No. 42 (November 2009) 26.**
- ▶ **[Proposed revision of N42.14-1999. In committee]**
"Calibration and Use of Germanium Spectrometers for the Measurement of Gamma-Ray Emission Rates of Radionuclides", N42.14-201x American National Standards Institute, Inc., 1430 Broadway, New York 10018:
"The following approaches may be considered for the calibration of the detector efficiency:
 - a) *Measurement of a standardization coefficient for a specific gamma ray and radionuclide by direct comparison with a standard source of known activity;*
 - b) *Measurement of the full-energy peak efficiency as a function of energy;*
 - c) *Calculation of the peak efficiency as a function of energy with the use of Monte Carlo or other calculation techniques."*



Basis Measurements for Modeling

- ▶ Even mathematical modeling approaches requires validation to a basis set of a source-based efficiency measurements.
- ▶ The most difficult part of the model validation is the full-energy response of the gamma-ray detector.
- ▶ The efficiency response depends on the scattering properties of the gamma-ray within the active part of the crystal. Uncertainties even on a millimeter scale can cause errors as much as 15% or more.
- ▶ **Keys to validating mathematical models:**
 - Benchmark all reference source activities to **Standard Reference Materials**.
 - Perform measurements in tightly controlled geometries (millimeters make a difference).
 - Include enough variety of measurements to accurately capture all the important detector features.
 - Simultaneously validate all measurements to a common model.



A comment on the ISOCS characterization uncertainty

Estimated uncertainties (%) for the 30 cm point source validation measurement

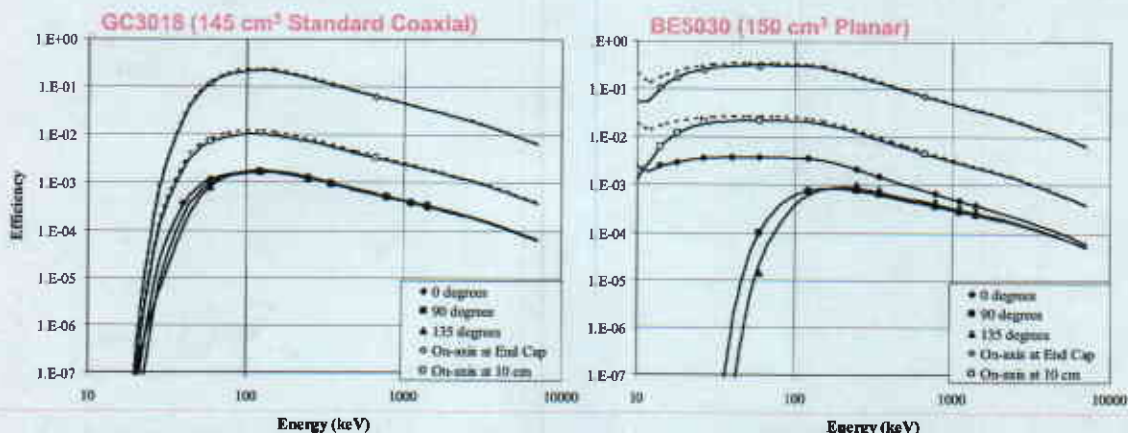
	13.9	59.5	1408	Comment
Decay Yield	3.4	1.5	1.1	Source activity and per decay Yield
Peak Analysis	7.0	0.5	0.5	Peak analysis techniques and statistical precision
Geometrical	0.25	0.25	0.25	Source positioning
Electronic	1.0	1.0	1.0	Pileup treatment, thresholds, dead time, etc.
Simulation Precision	1.0	1.0	1.0	Monte Carlo Precision
Model Approx.	4.0	2.0	2.0	Accuracy of the MCNP model
Total Uncertainty	8.9	2.9	2.7	

- ▶ With every ISOCS characterization report, we include a summary of the process uncertainty budget.
- ▶ This is not the overall ISOCS uncertainty. The recommended uncertainties are still represented by the ISOCS/LabSOCS V&V report.
- ▶ We take our measurements seriously and work hard to control all our sources of error.



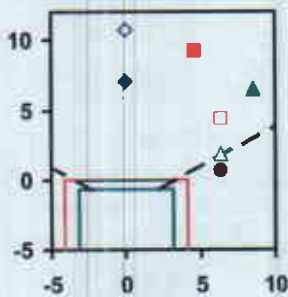
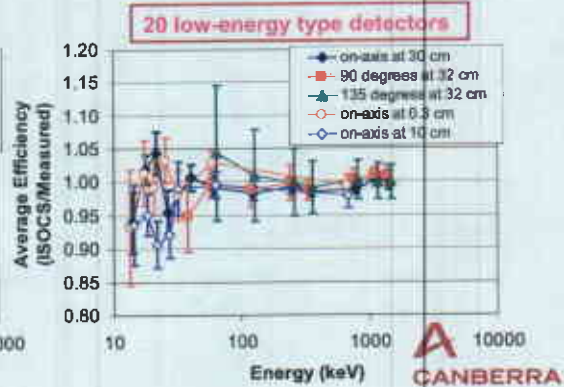
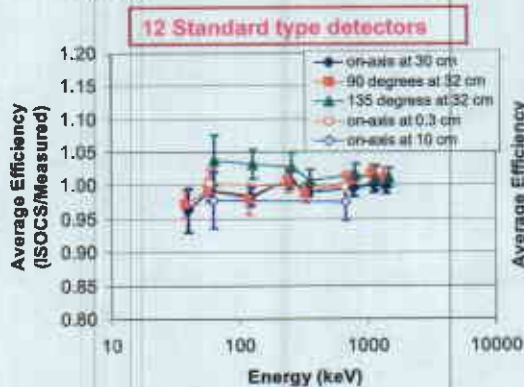
Comparison Measured and Modeled Absolute Efficiency

- ▶ Possible to perform characterizations for all detectors down to 10 keV.
- ▶ For standard coaxial detectors, it is only possible to validate the characterizations down to 40 keV from the front and side and 60 keV from the back.
- ▶ For low-energy sensitive detectors, validations are possible down to 14 keV from the front, 40 keV from the side, and 60 keV from the back.



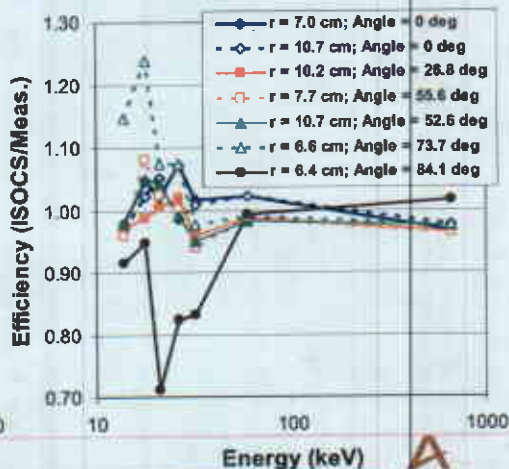
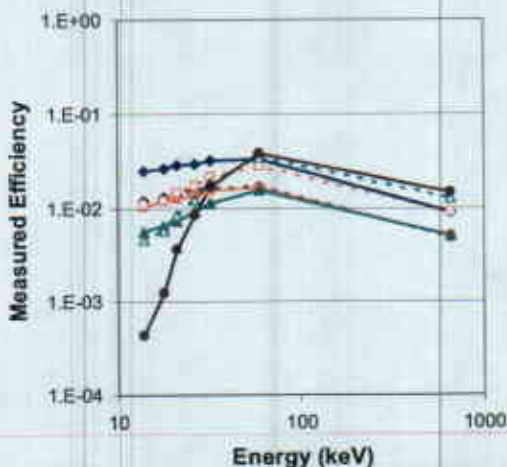
Comparison of Analysis Process Over a Large Variety of Detectors

- ▶ Comparison of modeled efficiencies (ISOCS) to measured efficiencies for 32 detectors of various types
- ▶ 20 of these detectors are low-energy type (REGE, XTRA, or BEGE)
- ▶ At high energies (≥ 50 keV) agreement between modeled and measured efficiency is better than 2.5% for forward and side geometries for all detector types
- ▶ Backward geometries are more difficult for low energy detectors because of the highly attenuating holder structures
- ▶ Measurements below 40 keV are not possible for standard SEGe detectors because of the germanium dead layer.
- ▶ For energies less than 60 keV, the agreement is within about 7% two of the geometries
- ▶ For the "on-axis 10 cm" measurement, there is an observed bias of about 7.5% between 14 and 20 keV. At this position the source passes through about 9 mm of plastic, and the bias is due to difficulties modeling the mounting structure.



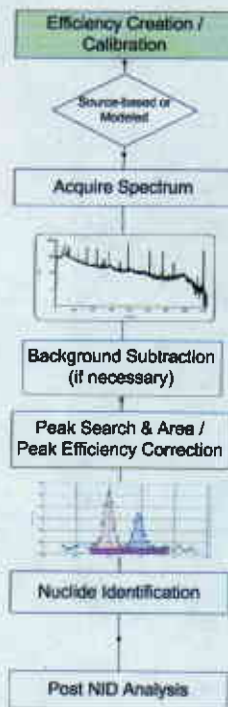
Efficiency response at different positions

- ▶ Measurements performed with a GR5020
- ▶ *Am-241/Cs-137* point source
- ▶ ISOCS eff. in good agreement with measured eff. in forward geometries.

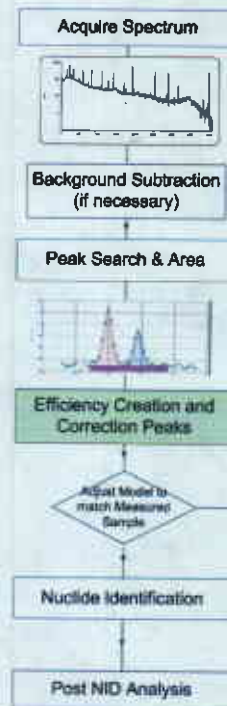


With Modeling, Efficiency in an Analysis Step

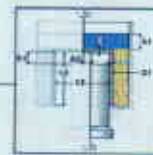
▶ Traditional Analysis Flow



▶ Alternative Analysis Flow



- ▶ The user can create the efficiency model at the efficiency correction step.
- ▶ Match directly the source being measured.



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Flexibility of Modeling

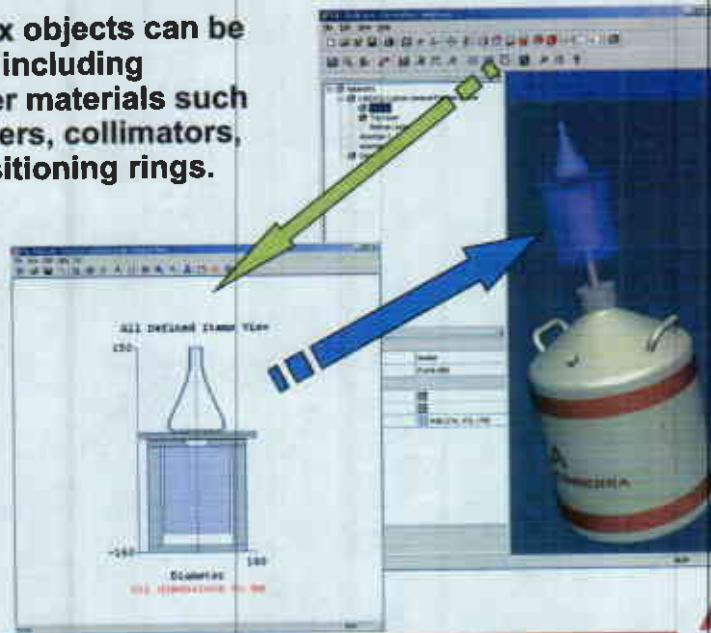
- ▶ With modeling, it is possible to rapidly produce geometries that represent many usual shapes for which source standards may not be readily available.



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Include Features Beyond just the Sample

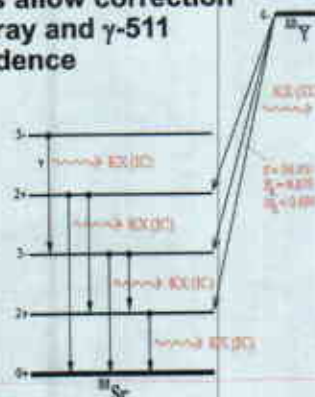
- ▶ Complex objects can be created including absorber materials such as spacers, collimators, and positioning rings.



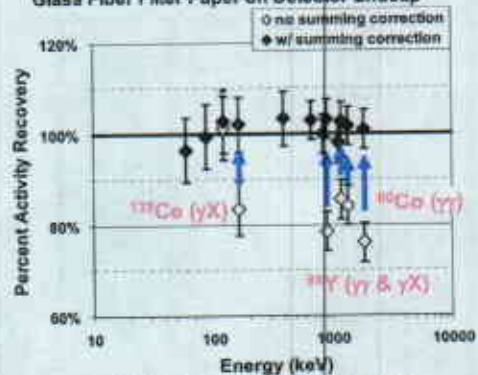
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Complex Corrections: Cascade Summing Effects

- ▶ With mathematical approaches, it is possible to correct for cascade summing losses.
- ▶ No source-based peak-to-total calibration is necessary
- ▶ Algorithms allow correction for γ - γ , γ -X-ray and γ -511 true coincidence summing



Glass Fiber Filter Paper on Detector Endcap



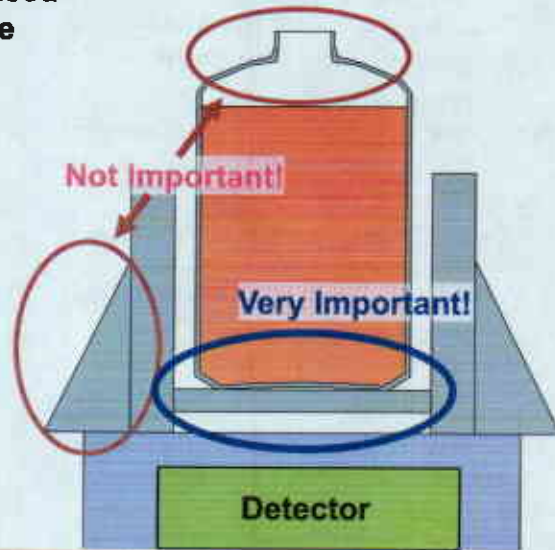
- ▶ Expanded cascade summing nuclide library extracted from ENSDF

- ◆ ENSDF – Internationally recognized Nuclear Structure Database
- ◆ All important nuclides included

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A Word of Caution

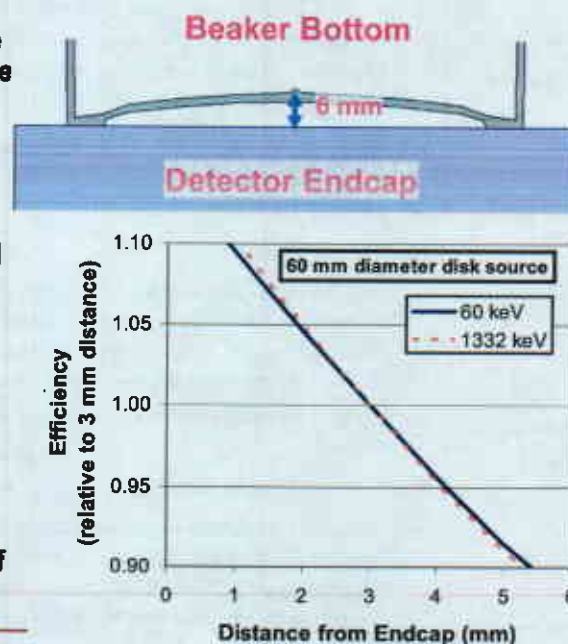
- ▶ While mathematical modeling is a significant time and cost saver compared to source-based calibrations, care must still be taken to model the geometry to an appropriately accurate level.
- ▶ Not all regions are equally important.
- ▶ Some critical parameters are:
 - ◆ Distance from Sample to Detector
 - ◆ Attenuation of gamma-rays by intervening materials



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Sensitivity of Distance

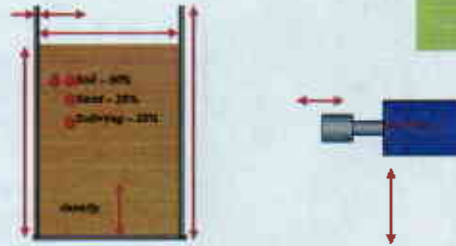
- ▶ The efficiency for sources close to the detector are very sensitive to the position of the source.
- ▶ As a general rule of thumb: 1 mm \approx 5% change in efficiency for close geometries.
- ▶ Many beakers have complicated bases and an "effective distance" may not be clear.
- ▶ Also note: Reference source standards in epoxy matrices may deform beaker (from Rxn heat) thus changing the geometry.
- ▶ Direct modeling of beaker base provides accurate description of the container.



In-Situ Uncertainty Analysis Example

200 liter drums containing soil

- ▶ Drum diameter +/- 1cm maximum
- ▶ Drum height +/- 1cm maximum
- ▶ Drum wall thickness +/- 20% maximum
- ▶ Soil composition
 - ◆ Normal soil 50% of drums
 - ◆ Sand 25% of drums
 - ◆ Soil + Humus 25% of drums
- ▶ Fill height 70-90% full
- ▶ Density via weight 450-750 lbs 95% of time
- ▶ Detector distance 90 – 110 cm
- ▶ Detector height from ground 16 – 36 cm



Variable	% at 60 keV	% at 1001
Drum diameter	+ 2 - 2	+ 2 - 2
Drum height	0	0
Drum wall thick	+ 29 - 29	+ 2 - 2
Sample height	+ 3 - 3	+ 1 - 1
Sample density	+ 39 - 28	+ 31 - 20
Sample comp'n	+ 9 - 6	+ 1 - 1
Detector distance	+ 18 - 14	+ 18 - 15
Detector height	+ 0 - 4	+ 0 - 2

Variable	95% CL at 60 keV	95% CL at 1001 keV
All items variable as per example	36%	20%
Density well known, all other items variable	30%	14%
Density and container wall well known, all other items variable	16%	14%



Sample container of soil

- ▶ Injection molded sample container
- ▶ Container diameter and height and wall thickness well known
- ▶ Concave bottom with +/- 2 mm variation
- ▶ Soil composition
 - ◆ Normal soil 50% of drums
 - ◆ Sand 25% of drums
 - ◆ Soil + Humus 25% of drums
 - ◆ Single calibration used for all
- ▶ Density varies between 1.1 and 1.4 g/cc
- ▶ 100 g always used, therefore fill height varies +/- 14%

Laboratory Uncertainty Analysis Example



Variable	% var at 60 keV	% var at 1001 keV
Container bottom curvature	+ 7 - 7	+ 9 - 8
Sample fill height	+ 13 - 7	+ 10 - 6
Sample density	+ 9 - 5	+ 9 - 8
Sample composition	+ 7 - 0	+ 0 - 0

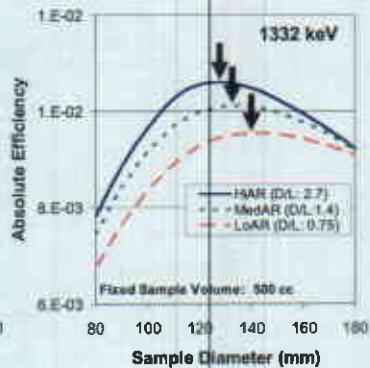
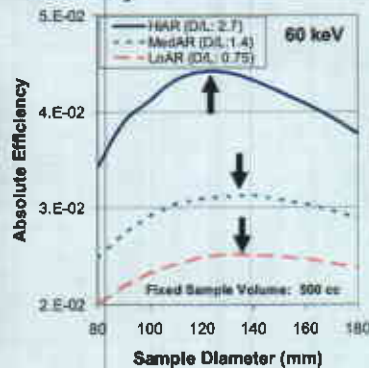
Variable	95% CL at 60 keV	95% CL at 1001 keV
All items variable	13%	9%
Density and sample height fixed, all other items variable	6%	3%



Modeling for Sample Optimization



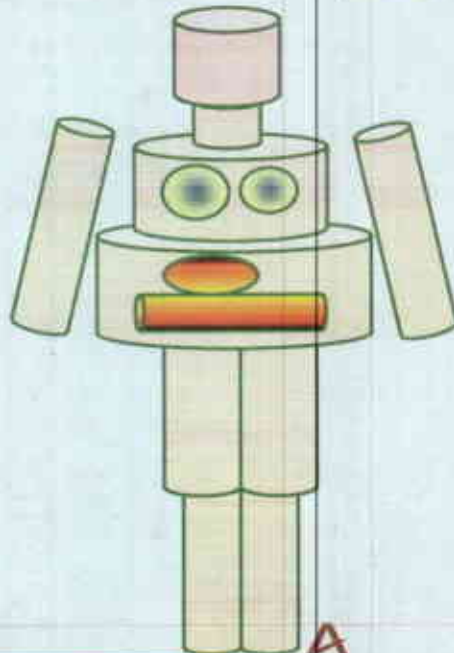
- ▶ Cylindrical sample with 500 cc of 1.6 g/cc Soil.
- ▶ Ran multiple efficiency computations with varying container diameter and height but with fixed sample volume.
- ▶ Determine maximum efficiency.
- ▶ All detectors have maximum efficiency when sample diameter is about 12 – 14 cm, regardless of energy.
- ▶ In all cases a high aspect ratio (e.g. BEGe's) detector has greater efficiency than a low aspect ratio detector of similar relative efficiency.



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What About Complex Objects?

- ▶ Conflicts between multiple objects occupying the same space resolved by object priority level
- ▶ Detector can be anywhere and point in any direction
- ▶ Multiple detector locations can be used to simulate moving detectors or moving sources

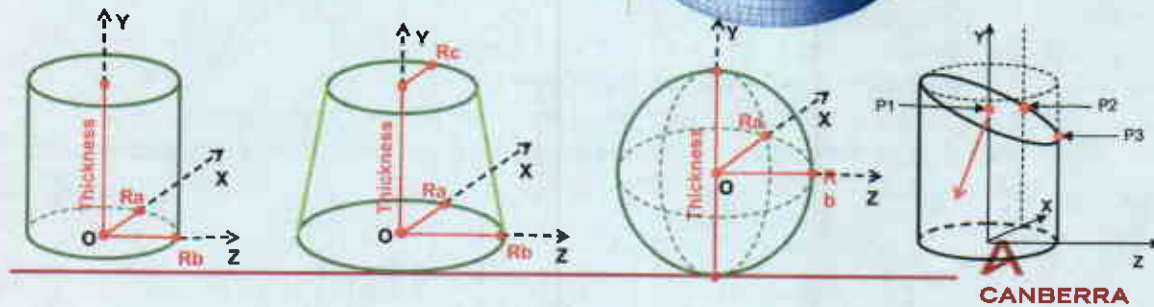


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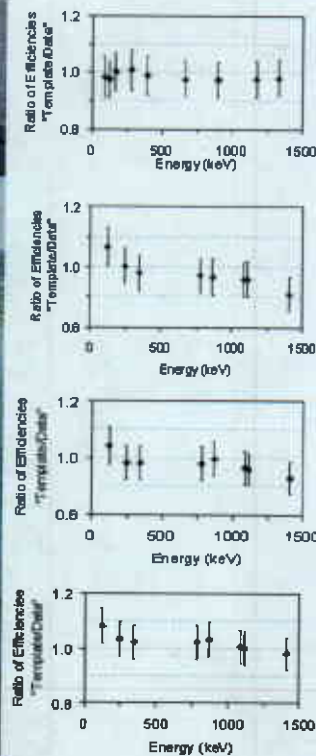
NEW ISOCS TEMPLATE (under development)

► Possible geometries allowed are:

- ◆ Elliptical Cylinder
- ◆ Cone – Circular, Inverted, Elliptical & Elliptical inverted
- ◆ Pyramid – inverted & assymetrical
- ◆ Spheroid & Ellipsoid
- ◆ Torroid – round or oval, full or partial
- ◆ Cut Plane to remove part of object



BOMAB Model



- Advanced modeling techniques can handle exotic geometries.
- Shown here are a variety of cylindrical source geometries.
- The figures represent the ratio of the modeled efficiency compared to the measured (certificate) efficiency
- Agreement is within about 5% for all cases.