A COMPARISON OF COMPUTATIONAL APPROACHES FOR THE DETECTION OF GAMMA-EMITTING RADIONUCLIDES IN FOODS

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Food Emergency Response Network

FDA U.S. FOOD & DRUG ADMINISTRATION

Interest : Quick and sound food safety decision-making in the event of nuclear or radiological emergency

Objectives: Develop rapid and high-throughput screening methods in support of large-scale emergency response and consequence management

Increase radioanalytical capability and sample-surge capacity with greater geographic coverage and operational flexibility

Approach: Communicate and collaborate with state laboratories in method development, validation, and standardization

Provide resources, trainings, and proficiency tests

Deliverable: Full range of radioanalytical capability for detecting all radionuclides concerning food safety

Ability of providing sufficient & reliable analytical results to enable prompt decision-making on food safety

Public Health Impact

Radiation in Foods

- Long-term consumption of food contaminated with high-level radioactivity increases health risk from exposure to radiation.
- Iodine 131, a gamma & beta-emitting radionuclide, can rapidly incorporated into milk and accumulate in thyroid if ingested. It increases risk of thyroid cancer, particularly in young children who consume large amount of milk products.
- Cesium 137, another gamma & beta-emitting radionuclide, behaves like potassium and can appear in meat. Cesium-137 has a half-life of 30 years, which can cause long-term food contamination. Prolonged exposure to Cs-137 can lead to a wide range of cancers.
- Strontium 90, a beta-emitting radionuclide, has chemical similarity to calcium, which can accumulate in bone. A direct linkage between strontium 90 accumulation and bone cancer and leukemia has been found.

Radionuclides of Concern

Radionuclide	Principal Emission	Radionuclide	Principal Emission	Radionuclide	Principal Emission
^{241}Am	α	^{137}Cs	γ	^{89}Sr	β
^{238}Pu	α	^{60}Co	γ	$^{90}Sr/^{90}Y$	β
^{239}Pu	α	^{153}Gd	γ	^{147}Pm	β
^{240}Pu	α	^{192}Ir	γ	^{227}Ac	β
^{210}Po	α	^{75}Se	γ	^{3}H	β
^{242}Cm	α	^{170}Tm	γ	^{32}P	β
^{243}Cm	α	^{169}Yb	γ	^{241}Pu	β
^{244}Cm	α	^{141}Ce	γ	^{228}Ra	β
^{252}Cf	α	^{144}Ce	γ	^{63}Ni	β
^{226}Ra	α	^{57}Co	γ	^{99}Tc	β
^{237}Np	α	^{134}Cs	γ	^{204}Tl	β
^{228}Th	α	^{125}I	γ	^{14}C	β
^{230}Th	α	^{129}I	γ	^{85}Kr	β
^{232}Th	α	^{131}I	γ	^{68}Ge	*EC*
^{234}U	α	^{99}Mo	γ	^{55}Fe	*EC*
^{235}U	α	^{103}Pd	γ		
^{238}U	α	^{103}Ru	γ		
		^{106}Ru	γ		
		^{198}Au	γ		
		^{109}Cd	γ		
		^{99m}Tc	γ		
		^{140}Ba	γ		
		^{140}La	γ		
		^{65}Zn	γ		
		^{85}Sr	γ		
		^{7}Be	γ		

Sources of Radionuclides:
- Natural Origin
- Weapon Tests
- Nuclear Power
- Mining Industries
- Space Explorations
- Nuclear Medicine
- Waste Disposals
- Terrorist Activities

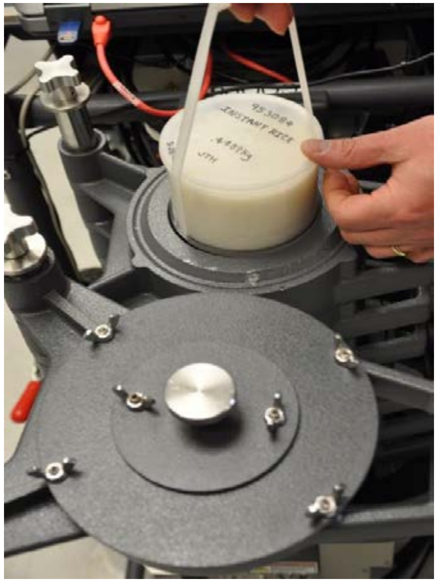
Detector System & Sample Preparation

FDA

Front of Detector

Back of Detector

Sample & 400-mL Container



Loading Test Sample

CURRENT APPROACH

Gamma Spectrometry
Can be used to identify and quantify radionuclides that are gamma emitters

Advantages
Non-destructive technique
Multi-radionuclide analysis

For Identification – Energy calibration
Standard sources containing multiple radionuclides with known photon energies

For Quantification – Efficiency calibration
Requires the equivalent volume geometry between the calibration standard and the test samples

Computational Approach

Software packages have been developed and can be applied to calculate counting efficiency without using calibration standards

Lower Cost than Source-based Calibration

Purchasing Sources – For calibrations

Disposal of Sources – Radioactive Waste

Replacing Sources – Radioactive Decay

Time

Less Labor Intensive - Easier to match the sample for calibration

Flexibility

Limited amount of sample

Unusual material composition

Software Packages For Efficiency Calculations

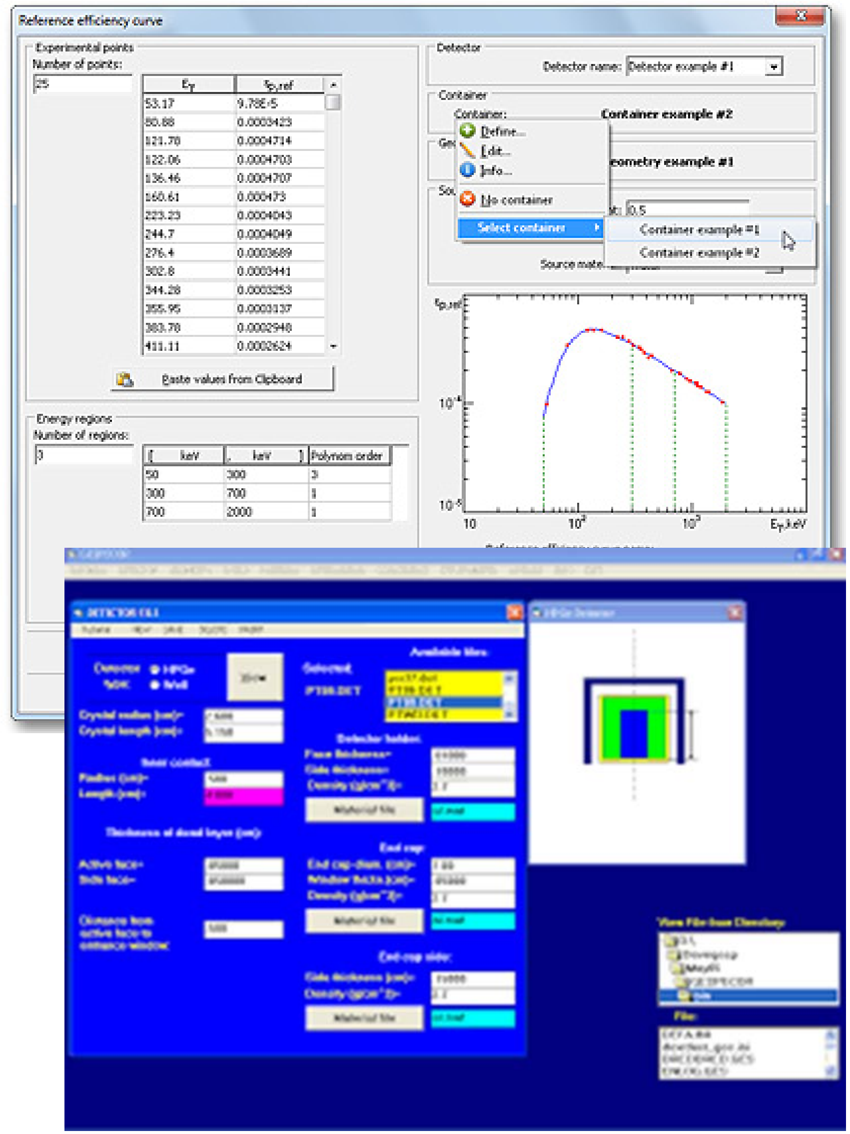
ANGLE combines advantages of both absolute (methods – minimizing potential for systematic errors in the former and reducing practical limitations of the latter.

GESPECOR

The software is very flexible and allows the user to change many variables, including the detector, shield and geometry all from a friendly user interface.

LABSOCS/ISOCS

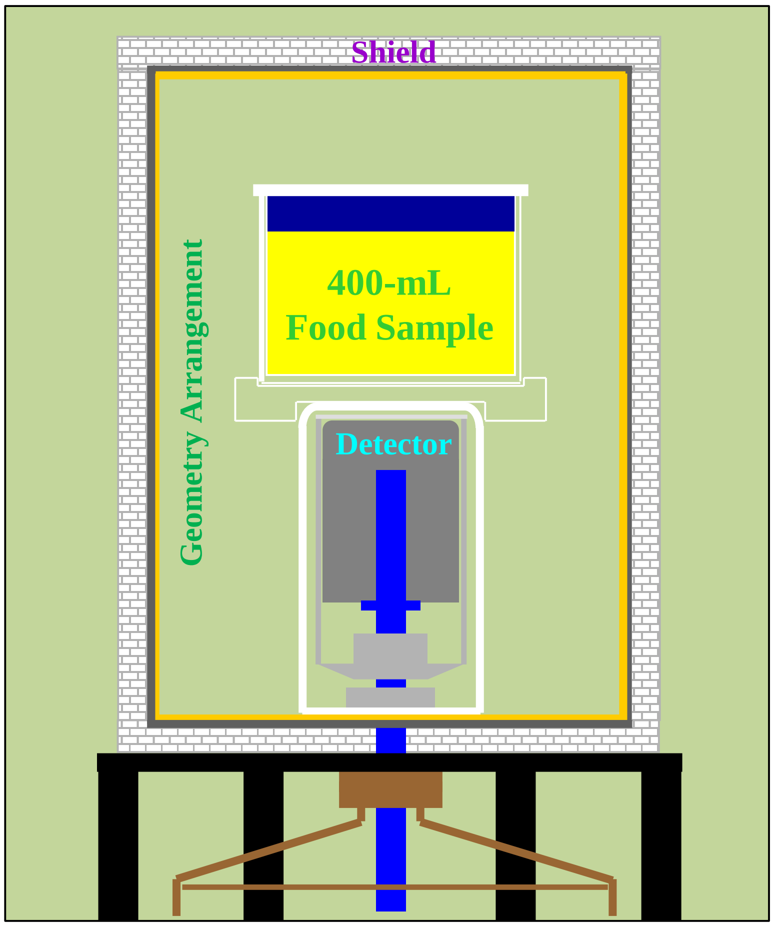
The difference between LabSOCS and other available mathematical calibration packages is that the detector used to perform activity measurements is initially characterized by the manufacturer.



Computational Setup

Computational approach requires descriptive inputs with regard to:

- Detector Characteristics
- Sample Properties
- Shielding Material
- Counting Geometry Arrangement



List of Input Variables

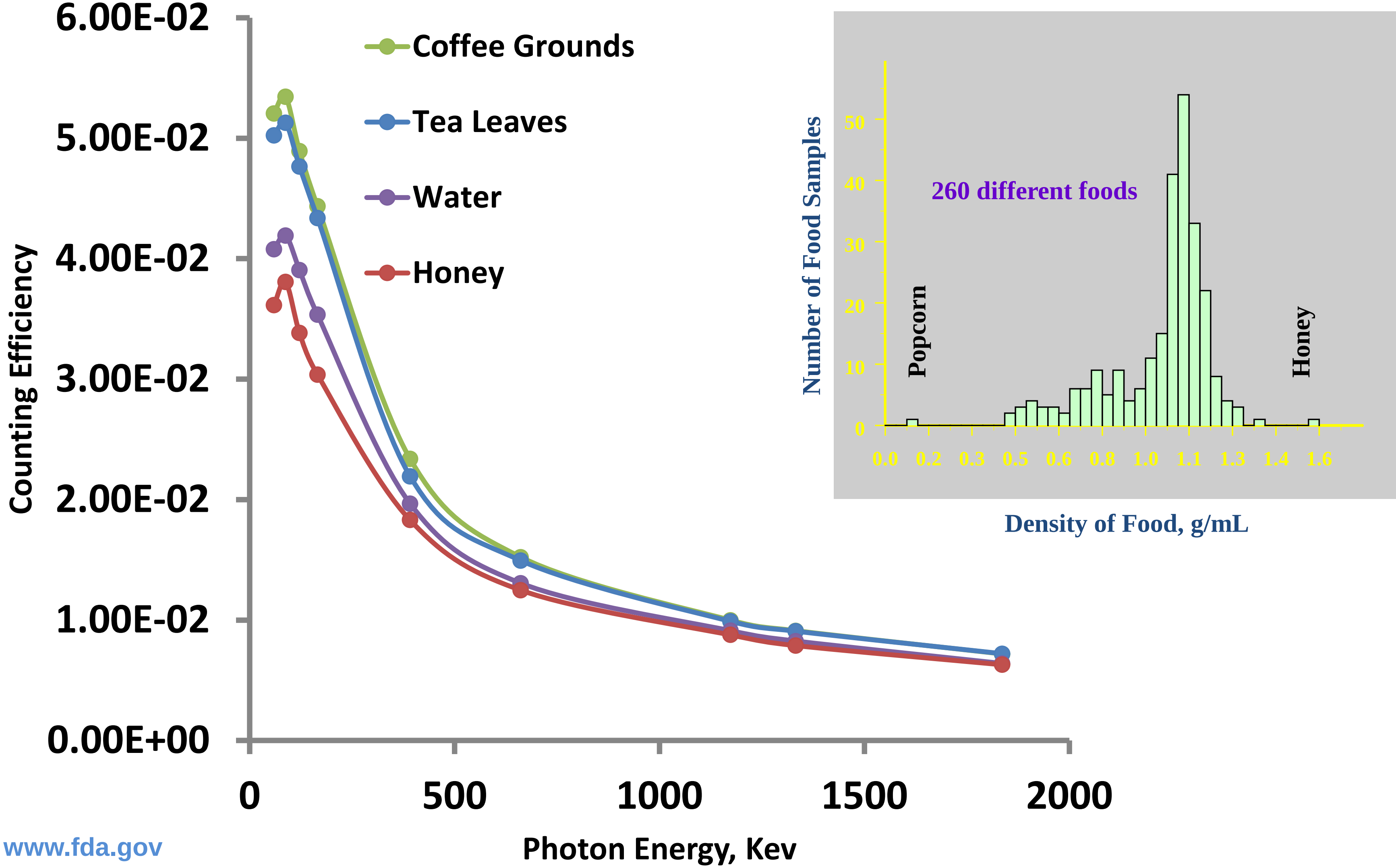
Crystal Radius
Crystal Length
Crystal Inner Contact Radius
Crystal Inner Contact Length
Crystal Holder Face Thickness
Crystal Holder Side Thickness
Crystal Holder Material Density
Face Dead Layer Thickness
Side Dead Layer Thickness
End Cap Diameter
End Cap Window Thickness
End Cap Side Thickness
End Cap Material Density

Container Radius
Container Height
Container Wall Thickness
Container Material Density
Sample Composition
Sample Density

Shield Inner Radius
Shield Inner Height
Shield Outer Radius
Shield Outer Height
Shielding Material Density

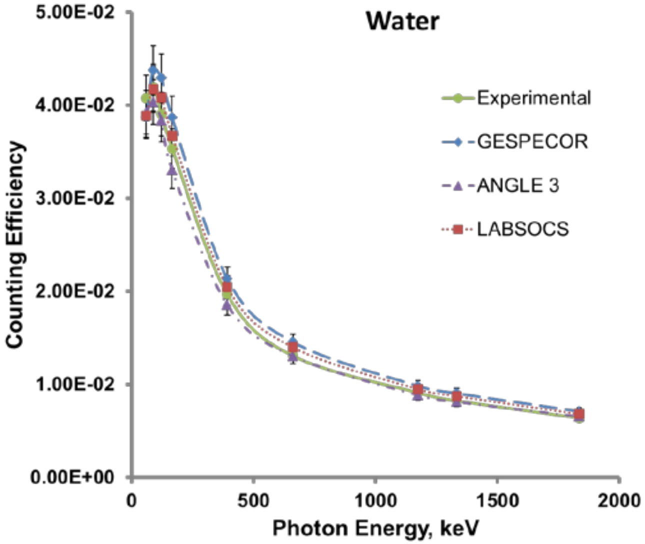
End Cap-to-Inner Shield Top Distance
Active Face-to-Entrance Window Distance

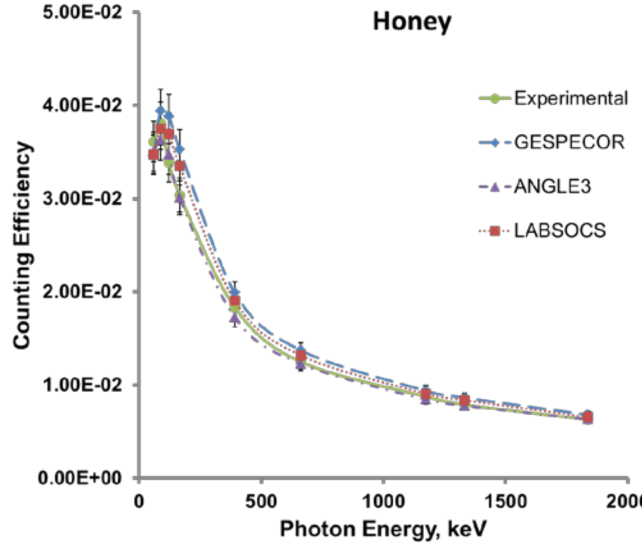
The Effect of Food Density on Efficiency

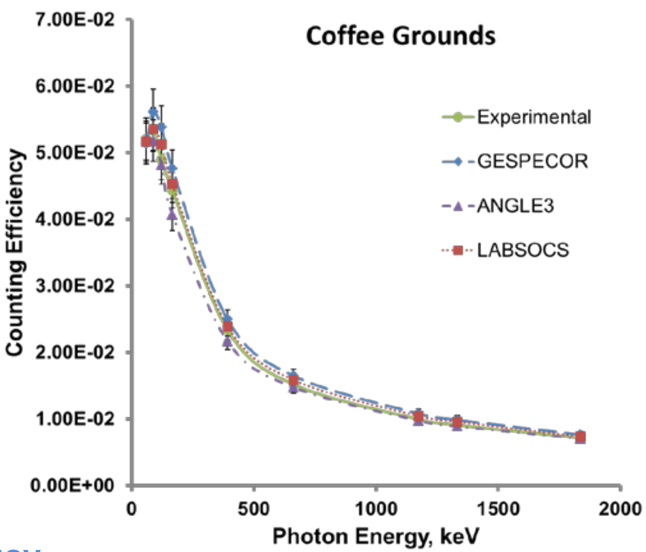


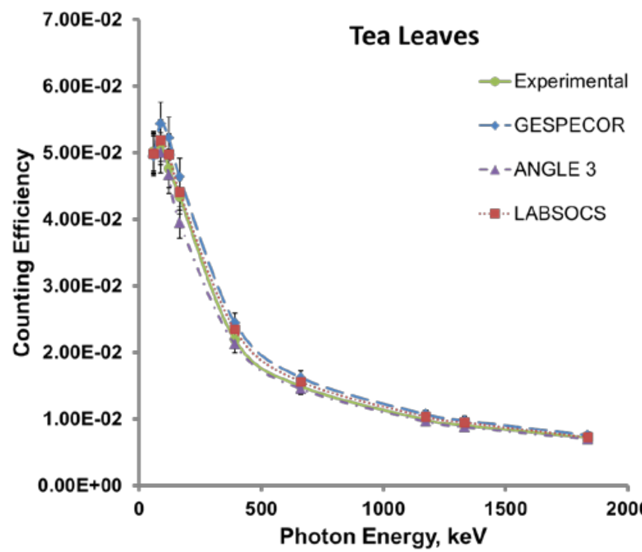
Results- Sample Density

Effective at reproducing experimental results and limit cost





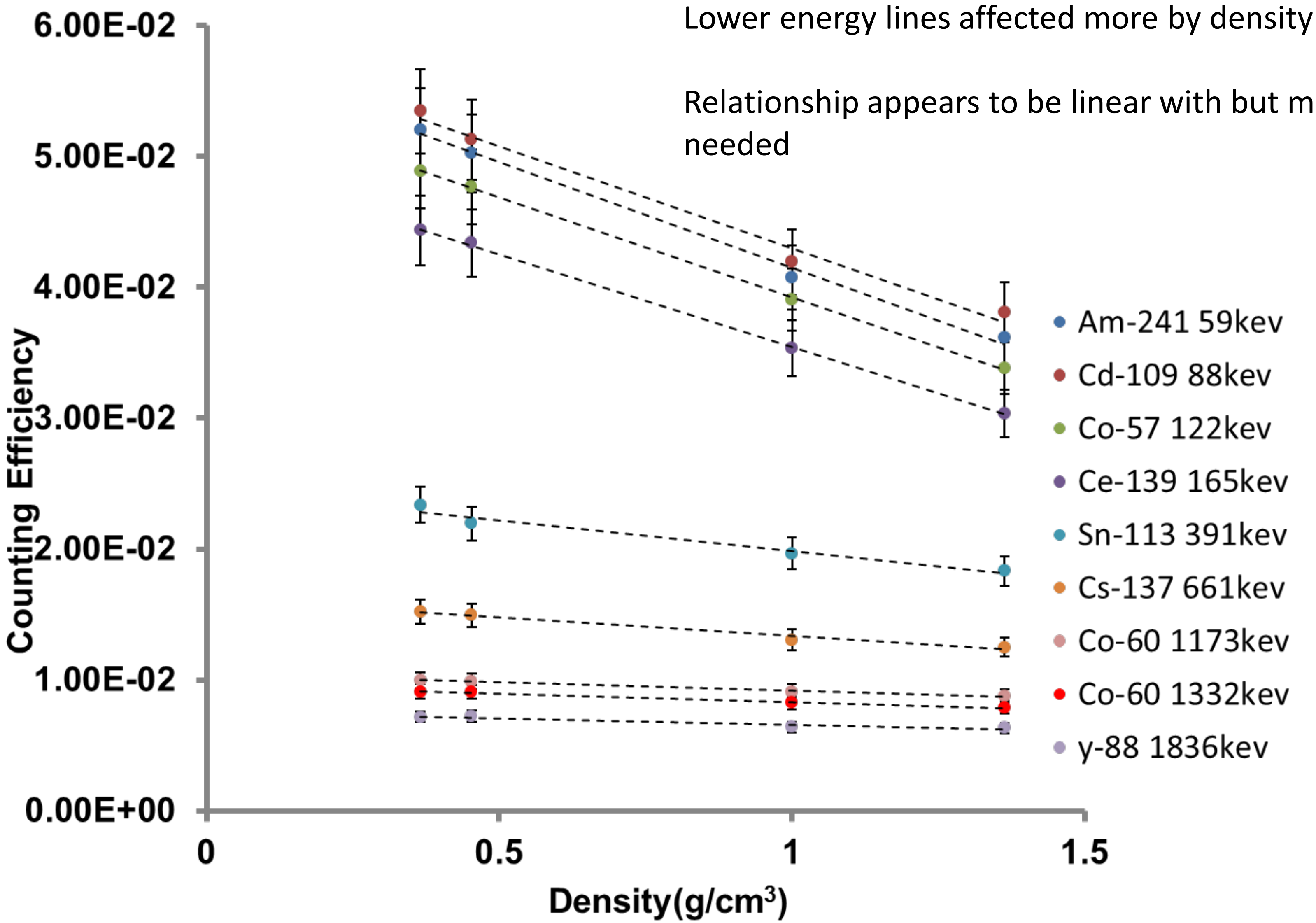




Comparison of Measured Efficiencies with Change in Density

Lower energy lines affected more by density

Relationship appears to be linear with but more data needed



Comparison of Measured Efficiencies with Change in Density

Food Product	Density	Elemental Analysis
Coffee	0.365	**C%** 57.00 **H%** 7.00 **O%** 35.00 **N%** 1.00
Tea	0.453	**C%** 55.59 **H%** 6.10 **O%** 35.16 N**%** 3.02 **S%** 0.07 **P%** 0.07
Water	1	**H%** 11.19 **O%** 88.81
Honey	1.363	**C%** 40.00 **H%** 7.00 **O%** 53.00

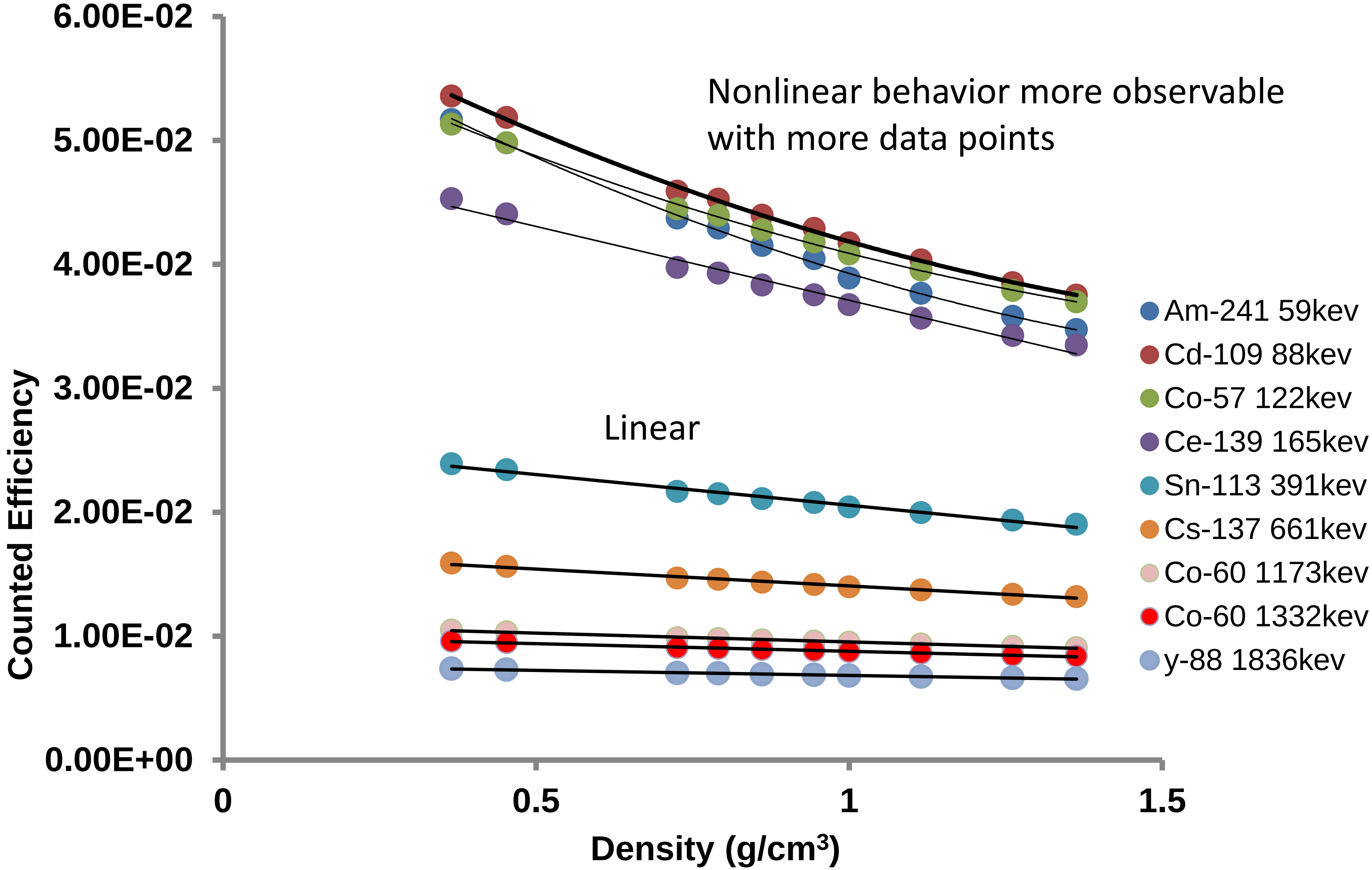
Organic Solvent	Density	Elemental Analysis
Triethylamine	0.7255	**C%** 71.22 **H%** 14.94 **N%** 13.84
Acetone	0.791	**C%** 62.04 **H%** 10.41 **O%** 27.55
DMF	0.944	**C%** 49.30 **H%** 9.65 **O%** 21.89 **N%**19.16
Glyme	0.9637	**C%** 53.31 **H%** 11.18 **O%** 35.50
Ethylene Glycol	1.115	**C%** 38.70 **H%** 9.74 **O%** 51.55
Glycerol	1.261	**C%** 39.13 **H%** 8.76 **O%** 52.12

Principle components in food are C, H, O, N

Using organic solutions to mimic food components, while improving range of densities that can be studied

Simulate radioactive solutions to minimize generating waste.

Comparison of Computed Efficiencies with Change in Density



Results of Food Spiked With ^{137}Cs and ^{60}Co

Spiked Activity: The amount of radioactive material added to the food sample.

Measured Activity: The activity determined by measurement of the sample using a water based efficiency

Corrected Activity: The activity determined in sample using computed efficiencies with density corrections

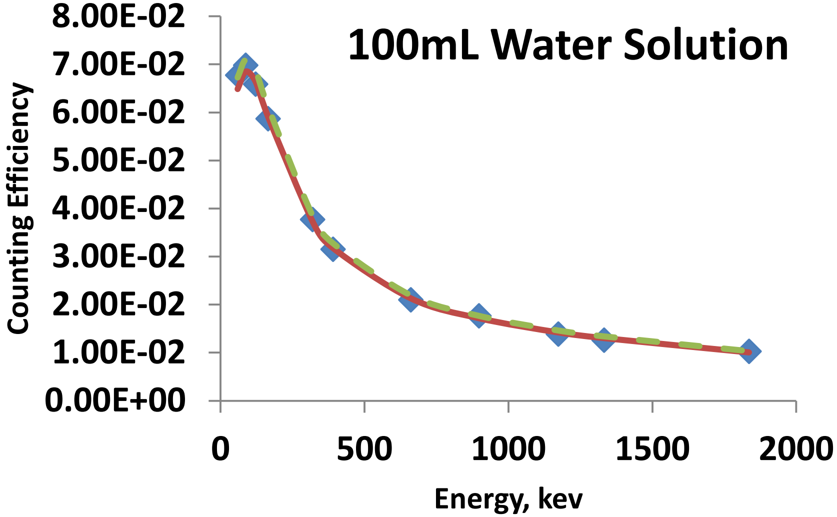
Food Product	Density	Isotope	Spiked Activity (Bq/g)	Measured Activity (Bq/g)	%error	Corrected Activity (Bq/g)	%error
Water	1	^{137}Cs	0.64	0.66	3.29	0.62	2.93
		^{60}Co	1.03	0.96	6.98	0.98	5.41
			1.04	0.95	8.41	0.98	6.00
Coffee	0.365	^{137}Cs	4.09	4.76	16.45	3.94	3.55
		^{60}Co	6.61	6.87	3.99	6.32	4.33
			6.61	6.70	1.34	6.72	1.60
Tea	0.453	^{137}Cs	3.33	3.83	15.10	3.21	3.63
		^{60}Co	5.38	5.51	2.46	5.16	4.08
			5.38	5.41	0.65	5.08	5.47
Unknown Syrup	1.15	^{137}Cs	0.18	0.16	10.68	0.19	1.68
		^{60}Co	0.37	0.35	5.26	0.35	7.01
Honey	1.363	^{137}Cs	0.48	0.47	1.67	0.47	1.91
		^{60}Co	0.77	0.70	10.08	0.74	4.20
			0.78	0.70	9.42	0.74	3.94

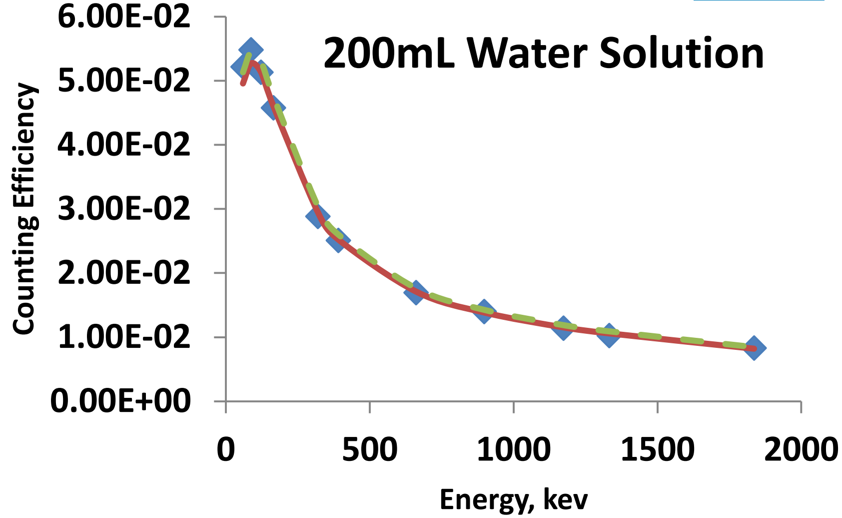
Summing Corrections for ^{133}Ba and ^{134}Cs in Water

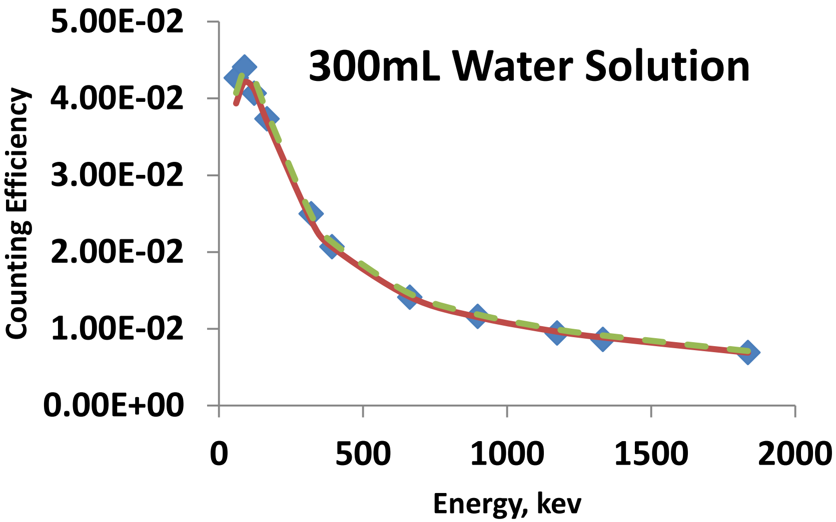
Nuclide	Known Activity	Uncorrected Activity (Bq/g)	% Diff.	LABSOCS Activity (Bq/g)	%Diff.	Gespecor Activity (Bq/g)	%Diff.	Angle Activity (Bq/g)	%Diff.
^{133}Ba	321.3	270	16.0%	330	2.7%	303	5.7%	300	6.6%
^{134}Cs	232.2	204	12.2%	225	3.1%	220	5.3%	223	4.0%

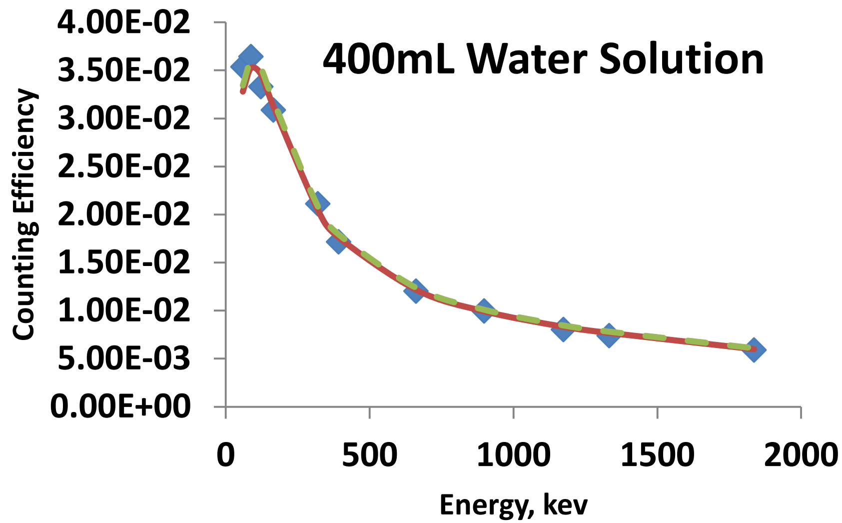
Our regulatory programs require a measurement accuracy to be within ±10%. Efficiency computations with a 2% uncertainty assigned to each of the detector and sample parameters showed that a combined effect on counting efficiencies from all uncertainty components is on the order of 6%.

Results – Sample Height









Fission Products of ^{235}U in 400mL of Water

Radionuclide	Known, Bq/kg	±2s	Experimental Bq/kg	Diff, %	Labsocs Activity Bq/kg	Diff, %	Angle 3 Bq/kg	Diff, %	Gespecor	Diff, %
Ba-140	2.43E+04	7.54E+02	2.47E+04	1.55%	2.44E+04	0.31%	2.36E+04	2.98%	2.18E+04	10.38%
Ce-141	1.91E+04	5.33E+02	1.94E+04	1.83%	1.88E+04	1.32%	1.81E+04	4.99%	1.59E+04	16.54%
Ce-144	2.88E+03	1.27E+02	3.13E+03	8.56%	3.03E+03	5.10%	2.98E+03	3.36%	2.56E+03	11.21%
Cs-137	9.14E+01	3.75E+00	9.47E+01	3.59%	9.42E+01	3.04%	9.16E+01	0.20%	8.45E+01	7.57%
I-132	1.82E+03	8.18E+01	1.81E+03	0.39%	1.84E+03	1.26%	1.72E+03	5.35%	1.69E+03	7.00%
La-140	2.77E+04	7.48E+02	2.79E+04	0.68%	2.76E+04	0.40%	2.70E+04	2.57%	2.47E+04	10.87%
Mo-99	1.33E+03	4.14E+01	1.32E+03	1.11%	1.31E+03	1.86%	1.27E+03	4.86%	1.22E+03	8.60%
Nb-95	5.17E+03	1.55E+02	5.16E+03	0.29%	5.11E+03	1.25%	4.99E+03	3.57%	4.59E+03	11.30%
Nd-147	8.34E+03	4.33E+02	7.96E+03	4.51%	7.97E+03	4.40%	7.53E+03	9.67%	6.69E+03	19.75%
Ru-103	8.50E+03	2.72E+02	8.29E+03	2.49%	8.23E+03	3.20%	7.99E+03	6.02%	7.38E+03	13.20%
Tc-99m	1.40E+03	4.91E+01	1.49E+03	6.22%	1.45E+03	3.37%	1.39E+03	0.91%	1.22E+03	13.03%
Te-132	1.85E+03	6.47E+01	1.73E+03	6.42%	1.79E+03	3.18%	1.76E+03	4.80%	1.54E+03	16.70%
Zr-95	1.29E+04	4.53E+02	1.31E+04	1.26%	1.30E+04	0.49%	1.27E+04	1.83%	1.17E+04	9.56%

Conclusions

- Computational Methods effective at predicting efficiencies over a range of densities
- Software packages are capable of delivering high throughput, accurate results in an emergency scenario
- Simulating radioactive solutions can help minimize generating waste saving cost of preparing samples and improve response time in the case of an emergency.

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