FDA U.S. FOOD & DRUG ADMINISTRATION
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Application of Liquid Scintillation and Gas-flow Proportional Counting Techniques for Simultaneous Detection of Alpha/beta Radioactivity in Food

Stephanie Healey, Zhichao Lin, Clarence Rolle, Jingjing Pan, Kelly Garnick, Anthony Wetherby, Patrick Regan

Winchester Engineering and Analytical Center, Winchester, MA 01890 USA
Office of Regulatory Science, Office of Regulatory Affairs, U.S. Food and Drug Administration

Abstract

Detection of anthropogenic alpha (α) and beta (β) radioactivity in food is difficult due to interferences from sample matrix and natural radionuclides. Recent interlaboratory studies on detection of α & β radioactivity in foods revealed that diverse methods used by the Food Emergency Response Network (FERN) are deficient, and a simple, reliable, and versatile screening method needs to be developed.

Both liquid scintillation counting (LSC) and gas-flow proportional counting (GPC), in concert with proper sample preparation, can detect α & β radioactivity in food. With many FERN radiological laboratories possess only one of the detection techniques, developing a versatile method fitting both LSC and GPC will enable full leverage of existing radioanalytical resources.

A radiochemical procedure that combines rapid food ashing and group extraction of Am, Pu, Cm, and Y was studied for screening of anthropogenic α and β radioactivity in food. To enable LSC counting, the extracted analyte radionuclides were dissolved in 0.5M HCl and then mixed with Ultima Gold AB cocktail. For GPC counting, the extracted analyte radionuclides were transferred onto stainless steel planchet and evaporated to dryness. Alpha and beta standard pairs (i.e., ^{90}Y/^{239}Pu for LSC and ^{90}Y/^{209}Po for GPC), prepared to match sample characteristics, were used to calibrate α and β counting efficiencies as well as percent α/β spillovers for LSC and GPC, respectively.

The method applicability for triage of contaminated foods at levels of regulatory significance was demonstrated by analyzing different types of foods spiked with known α and β radioactivity. The study showed that the method can detect ~0.6 Bq/kg of α radioactivity and ~0.4 Bq/kg of β radioactivity based on analyzing 35-g food and 1-hour sample count time. All analysis results were found to be within ±30 % of the known values.

This poster details the method development and shows the merits of this method on improving the FERN's radioanalytical capability and testing capacity for safeguarding the nation's food supply against radioactive contamination.

Objectives

The overall goal is to establish FERN radioanalytical capability and surge capacity for triaging contaminated foods in the event of a large-scale nuclear or radiological emergency.

❖ Objective 1

Develop a rapid, versatile, and efficient radioanalytical method for simultaneous screening of α and β radioactivity in a variety of foods

❖ Objective 2

Implement the developed method within FERN radiological laboratory network through collaborative matrix extension study

❖ Objective 3

Demonstrate FERN radiological laboratory network preparedness and readiness through radiological proficiency evaluation

Methodological Consideration and Approach

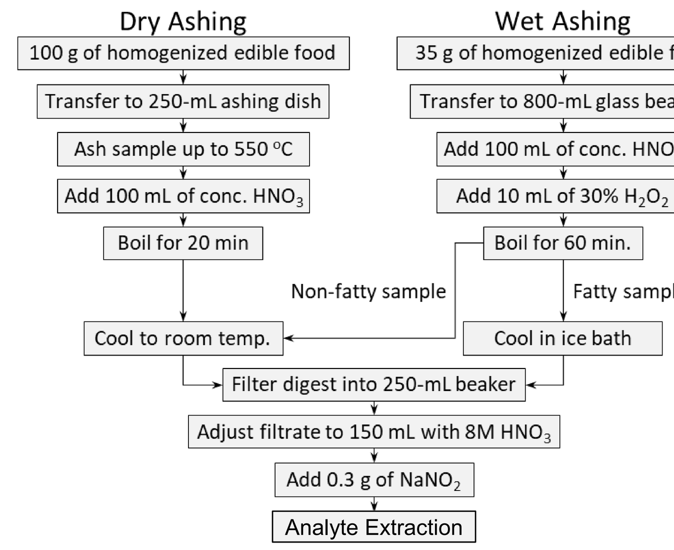
❖ Scope of Method

- **Intended Use:**
  Screening of anthropogenic α & β radioactivity in foods
- **Applicability:**
  Vegetable, dairy, meat, grain, and composite meal
- **Analyte:**
  α-emitter: ^{241}Am, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{243}Cm, and ^{244}Cm
  β-emitter: ^{90}Sr
- **Applicable Instrument:**
  LSC and GPC

❖ Mineralization of Food Sample

Rapid transformation of food into solution readily for analyte extraction

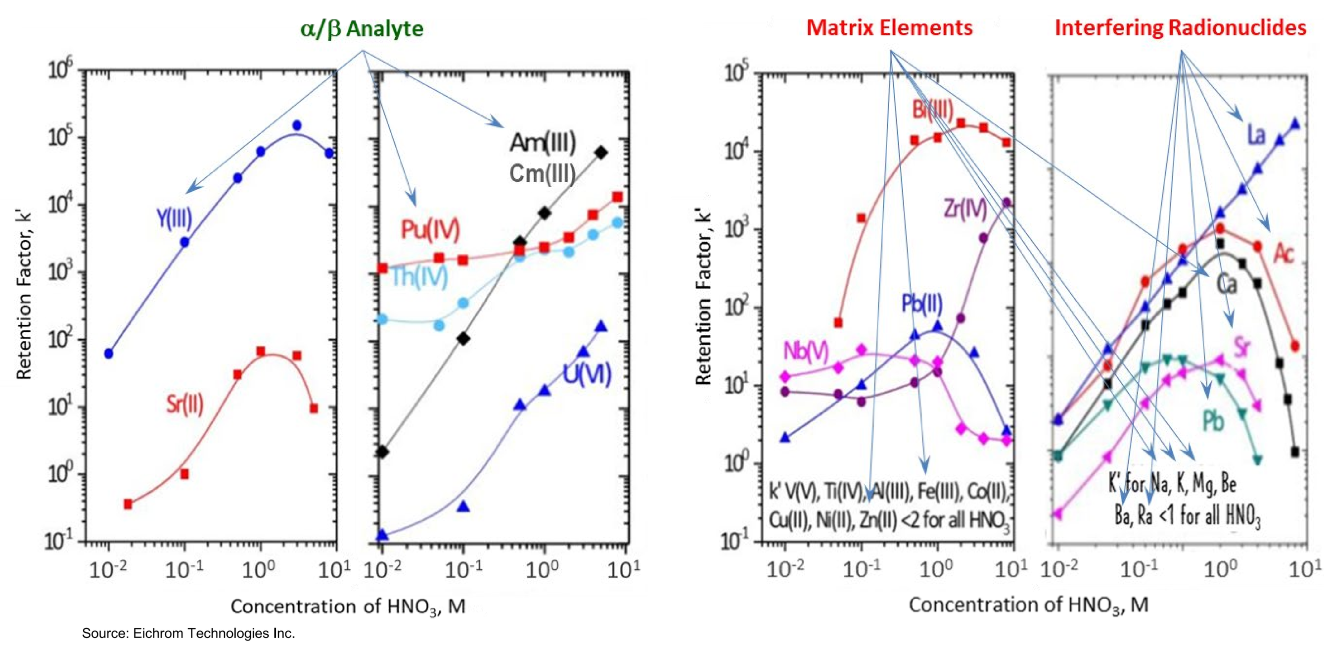
- **Use wet & dry ashing techniques to:**
  - ✓ Expedite sample analysis when analytical results are urgently needed
  - ✓ Enable high-throughout and sustainable sample analysis during emergency operation



❖ Method Selectivity

Detect anthropogenic α & β radioactivity without interferences

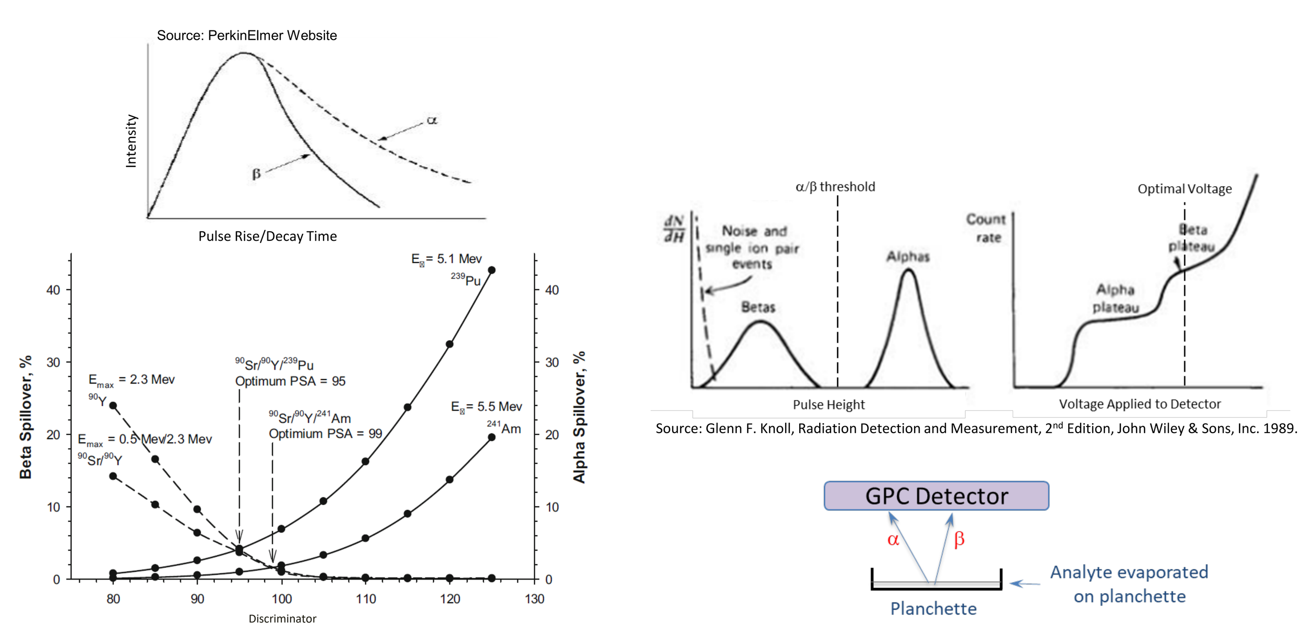
- **Use DGA resin to:**
  - ✓ Extract Am, Pu, Cm, Y
  - ✓ Remove matrix elements
  - ✓ Remove α & β interferences



❖ Method Specificity

Discriminative detection of α & β radioactivity from a single measurement

- **Use LSC or GPC to:**
  - ✓ Discriminate α & β radioactivity based on pulse shape or pulse height
  - ✓ Detect total α radioactivity if ^{241}Am, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{243}Cm, & ^{244}Cm coexist
  - ✓ Detect ^{90}Sr β radioactivity through its progeny ^{90}Y



❖ Method Robustness:

- **Use a wide variety of foods to:**
  - ✓ Achieve broad matrix tolerance
  - ✓ Ensure applicability for priority and staple foods

Candidate Foods

Dairy: 2% Milk, Whole Milk, American Cheese, Cheddar Cheese, Cottage Cheese, Swiss Cheese, Infant Formula, Toddler Formula, Follow-on Formula, Drinkable Yogurt, Light Yogurt, Regular Yogurt, Coffee Cream, Clotted Cream, Whipped Cream

Vegetable: Spinach, Carrot, Celery, Broccoli, Mixed Vegetables, Red Potato, Russet Potato, White Potato, Italian Black Olive, Spanish Green Olive, Olive, Cherry, Blueberry, Strawberry, Peach, Apple Sauce, Fruit Salad, Orange Juice, Sake, Spring Water

Composite Meal: Macaroni & Cheese, Spaghetti Meatball, Roasted Honey Chicken & Vegetables, Vegetables, Turkey, & Barley, Mixed Vegetables & Beef, Chicken Noodle, Cheese Pizza, Veggie Pizza, Supreme Pizza, Roasted Beef Sandwich, Sausage, Egg, & Cheese Sandwich, Tuna Sandwich, Hamburger, Chicken Sandwich, Bacon Cheeseburger, Baby Food Rice Pudding, Baby Food Peach, Baby Food Turkey

Meat: 85% Lean Beef, Branded Beef, Conventional Beef, Grass-Finished Beef, Beef Salami, Beef Sausage, Lamb, Pork, Chicken, Turkey, Duck, Cod, Salmon, Scallop, Tuna, Shrimp, Oyster, Mussel, Hot Dog

Grain: Baked Bean, Black Bean, Kidney Bean, Oatmeal Plain, Oatmeal Maple, Oatmeal Raisin, Wheat Cereal, Corn Cereal, Rice Cereal, Whole Wheat Bread, Corn Bread, Macaroni, Wheat Spaghetti, Rice Pudding, Rice Krispies, Cooked Rice, Breadcrumbs, Corn Flakes

Results and Discussions

❖ α & β counting efficiencies and α/β spillover

- ✓ ^{239}Pu/^{90}Y standard pair was found preferable for optimizing LSC
- ✓ ^{209}Po/^{90}Y standard pair was found preferable for optimizing GPC

LSC

α Emitter	Energy MeV	α Efficiency %	α into β %
^{241}Am	5.45	99.8±1.0	2
^{239}Pu	5.49	98.5±0.3	4
^{244}Cm	5.80	99.6±0.3	1.7
β Emitter	**Energy MeV**	**β Efficiency %**	**β into α %**
^{90}Sr/^{90}Y	0.55 2.28	96.3±1.3	2
^{90}Y	2.28	98.1±0.6	2

GPC

α Emitter	Energy MeV	α Efficiency %	α spill into β %
^{241}Am	5.45	39.8	-
^{238}Pu	5.49	35.6	-
^{239}Pu	5.14	35.9	-
^{240}Pu	5.15	36.1	-
^{243}Cm	5.76	36.2	-
^{244}Cm	5.80	36.1	-
^{209}Po	4.87	-	1.98
	Mean ± 1s	36.6 ± 1.6	
β Emitter	**Energy MeV**	**β Efficiency %**	**β spill into α %**
^{90}Y	2.28	55.8	0.04
^{90}Y	2.28	56.5	0.04
	Mean ± 1s	56.2 ± 0.5	

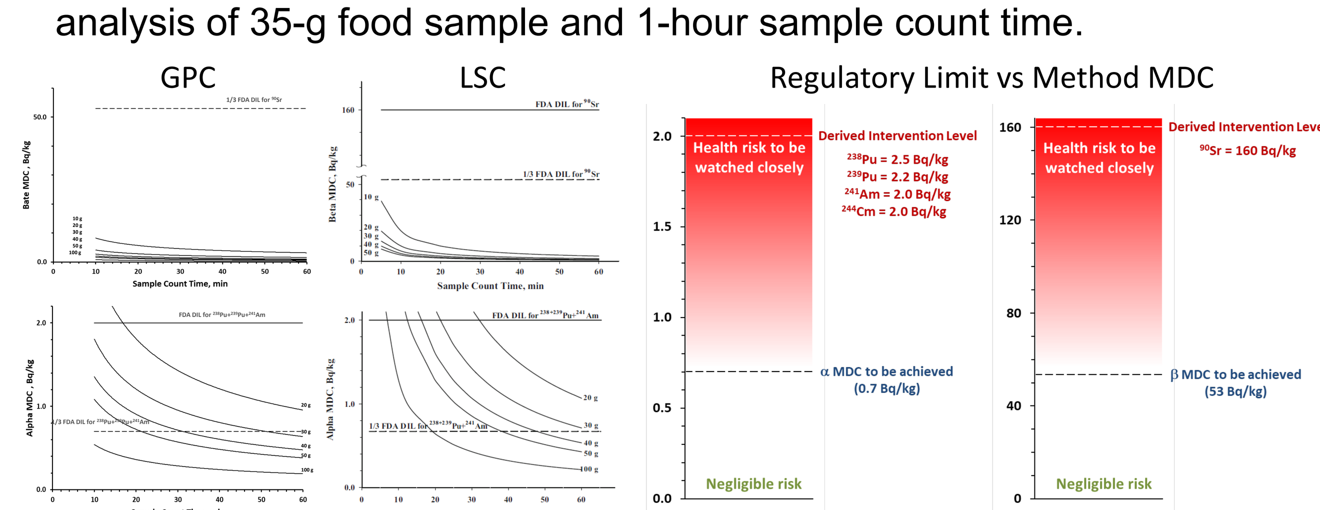
❖ Procedure Yields

- ✓ An average procedure yield determined with different α and β radionuclides could be used to correct procedure loss
- ✓ It is essential to avoid disproportionation and polymerization of Pu in order to minimize Pu loss
- ✓ Manipulating sample in quantitative manner throughout analysis is necessary to produce a result within ±30% of known value

Food Group	Food Description	Weight, g	Procedure Yield, % ^{241}Am	^{239}Pu	^{244}Cm	^{90}Y
Composite meal	Chicken noodle	100.2	-	88.2	-	85.8
	Macaroni & cheese	100.5	-	98.8	-	93.9
	Vegetables, turkey, & barley	100.0	-	89.5	-	96.2
	Mixed vegetables & beef	55.5	96.5	-	-	93.6
	Vegetable beef pie 1	36.0	-	-	94.4	-
	Vegetable beef pie 1	36.0	-	-	101.7	-
	Vegetable beef pie 1	36.0	-	-	98.3	-
Dairy	2% milk	118.0	-	92.5	-	91.9
	Whole milk	111.3	95.8	-	-	95.4
	Baby formula	60.0	104.3	88.7	-	97.3
	Formula 1	36.0	-	-	98.9	-
	Formula 2	36.0	-	-	99.6	-
	Formula 3	36.0	-	-	99.9	-
Grain	Rice cereal	30.0	-	-	-	92.1
	Corn flakes	24.9	91.1	-	-	92.6
	Cooked rice	63.8	95.3	74.2	-	97.8
	Breadcrumbs	30.1	97.4	76.9	-	94.8
	Baked bean 1	36.0	-	-	99.1	-
	Baked bean 2	36.0	-	-	99.0	-
	Baked bean 3	36.0	-	-	101.6	-
Meat	85% lean beef	108.0	94.5	85.0	-	94.5
	White tuna	51.1	100.4	89.3	-	94.2
	Cod	63.2	98.1	76.8	-	96.3
	Shrimp	65.1	102.0	-	-	90.9
	Pork	67.0	97.2	77.5	-	-
	Salmon	63.6	98.2	80.3	-	-
	Scallop	71.3	99.1	-	-	78.7
	Chicken	66.1	96.5	-	-	-
	Mussel	63.4	105.9	91.7	-	92.3
	Ground beef 1	36.0	-	-	93.8	-
	Ground beef 2	36.0	-	-	97.2	-
	Ground beef 3	36.0	-	-	99.0	-
Vegetable & fruit	Spinach	50.0	-	99.3	-	99.2
	Diced peaches 1	100.9	-	91.4	-	90.0
	Mixed vegetables	100.1	95.5	97.2	-	96.2
	Diced peaches 2	100.2	-	-	-	95.6
	Carrot	53.9	98.1	-	-	95.0
	Apple mango kiwi	50.9	99.3	-	-	96.8
	Apple sauce	87.0	107.3	79.7	-	97.4
	Mixed salad 1	36.0	-	-	94.1	-
	Mixed salad 2	36.0	-	-	100.9	-
	Mixed salad 3	36.0	-	-	91.1	-
		Average Procedure Yield:	**98.6**	**86.9**	**97.9**	**93.7**
		1 sigma:	**4.0**	**8.1**	**3.2**	**4.3**

❖ Method Detectability:

As plots shown below, the method could detect anthropogenic α and β radioactivity below 1/3 of FDA's derived intervention levels based on analysis of 35-g food sample and 1-hour sample count time.



❖ Results of Interlaboratory Analysis

- ✓ Screening of ^{90}Sr, ^{214}Am, ^{239}Pu, and ^{244}Cm in foods using LSC
- ✓ Acceptable results were produced by different analysts and laboratories

^{244}Cm & ^{90}Sr in food – Lab A

Sample Description	Sample Size, g	Measured Value, Bq/kg ^{244}Cm	Measured Value, Bq/kg ^{90}Sr	Known Value, Bq/kg ^{244}Cm	Known Value, Bq/kg ^{90}Sr
Apple juice ^{244}Cm	35.49	65.94±1.07	-0.80±0.41		
		66.52±1.08	-0.23±1.13		
	Mean±1s	66.23±0.41	ND	65.69±0.60	-
Apple juice ^{90}Sr	35.71	0.41±0.09	37.00±0.75		
		0.23±0.08	37.12±0.79		
		ND	37.06±0.08	-	38.25±0.09
Apple juice ^{244}Cm, ^{90}Sr	35.66	66.86±1.09	11.15±0.40		
		67.66±1.10	11.00±0.44		
	Mean±1s	67.26±0.57	11.08±0.11	68.91±0.62	11.16±0.03
Apple juice ^{244}Cm, ^{90}Sr	35.34	954.02±12.96	11.19±0.57		
		953.92±12.96	11.78±0.66		
	Mean±1s	953.97±0.07	11.49±0.42	945.14±0.85	9.83±0.02
Apple juice ^{244}Cm, ^{90}Sr	35.46	60.59±1.00	5.59±0.33		
		60.36±1.00	5.71±0.38		
	Mean±1s	60.48±0.16	5.65±0.08	59.29±0.53	5.23±0.01

^{241}Am & ^{90}Sr in food – Lab B

Sample ID	Matrix	Analyte	Value ± 2s, Bq/kg Measured	Known	Difference %	Acceptability
Matrix Blank A10	Chopped Spinach	NA	0.50±0.42	0	NA	Acceptable
Matrix Spike B10		^{90}Sr	194.64±6.46	195.2±3.0	-0.3	Acceptable
Matrix Spike C10		^{90}Sr	194.18±6.45		-0.5	Acceptable
Matrix Spike D10		^{90}Sr	183.15±6.12		-6.2	Acceptable
Matrix Blank A10	Chopped Spinach	NA	0.15±0.14	0	NA	Acceptable
Matrix Spike B10		^{241}Am	10.75±0.57	10.3±0.2	4.4	Acceptable
Matrix Spike C10		^{241}Am	11.03±0.58		7.1	Acceptable
Matrix Spike D10		^{241}Am	9.98±0.55		-3.1	Acceptable

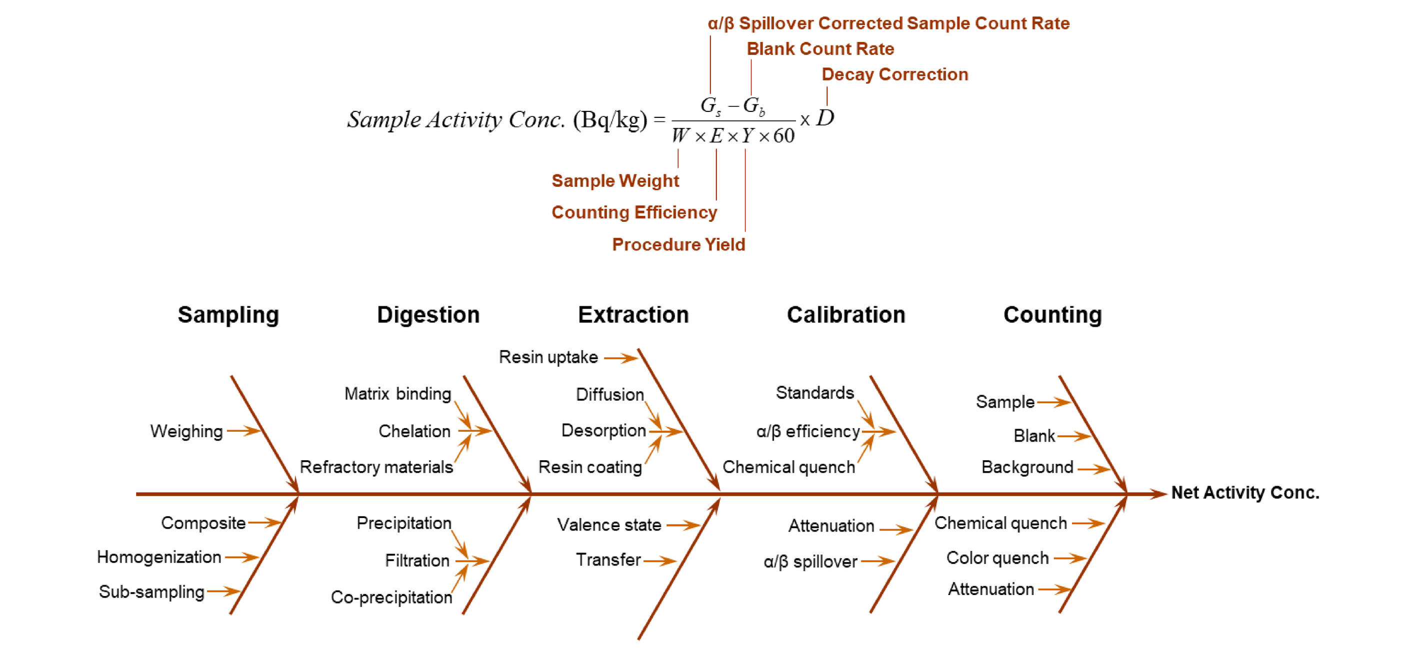
^{239}Pu & ^{90}Sr in food – Lab B

Sample ID	Matrix	Analyte	Value ± 2s, Bq/kg Measured	Known	Difference %	Acceptability
Matrix Blank A9	Food Ash Composite	NA	4.6±2.1	0	NA	Acceptable
Matrix Spike B9		^{90}Sr	2526±42	2483±13	1.7	Acceptable
Matrix Spike C9		^{90}Sr	2441±40		-1.7	Acceptable
Matrix Spike D9		^{90}Sr	2678±44		7.9	Acceptable
Matrix Blank A9	Food Ash Composite	NA	3.26±0.75	0	NA	Acceptable
Matrix Spike B9		^{239}Pu	40.1±2.4	38.2±0.2	5.0	Acceptable
Matrix Spike C9		^{239}Pu	36.0±2.3		-5.8	Acceptable
Matrix Spike D9		^{239}Pu	38.1±2.4		-0.3	Acceptable

❖ Uncertainty Estimate

- ✓ key variables affecting measurement result were identified and estimated

$$\text{Sample Activity Conc. (Bq/kg)} = \frac{G_c - G_b}{W \times E \times Y \times 60} \times D$$



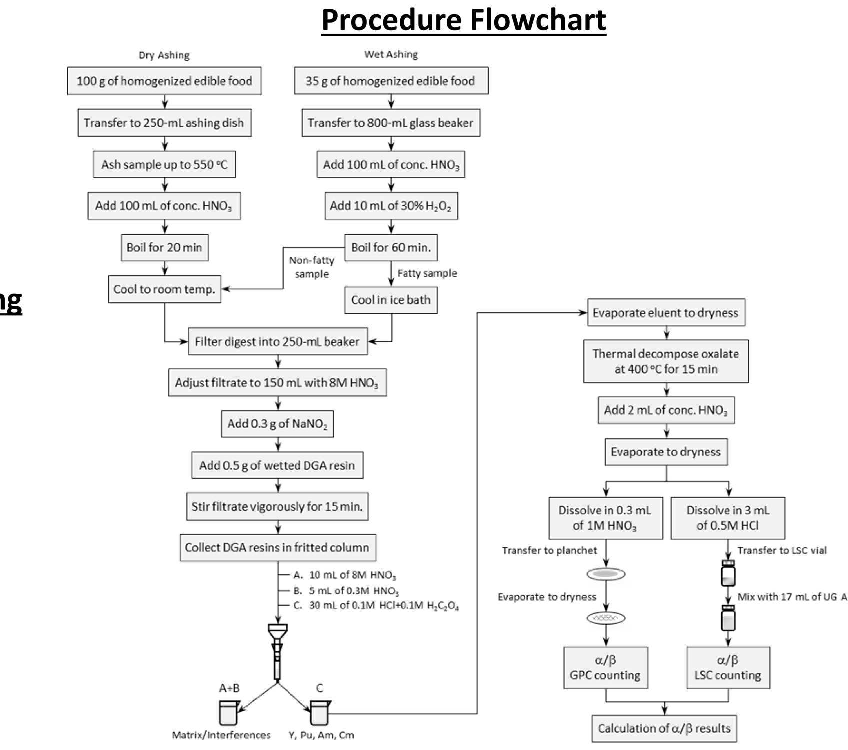
Current Progress and Future Plan

❖ Finalize Method Procedure

- ✓ Procedure for LSC has been developed & validated
- ✓ GPC application has been developed & ready for validation

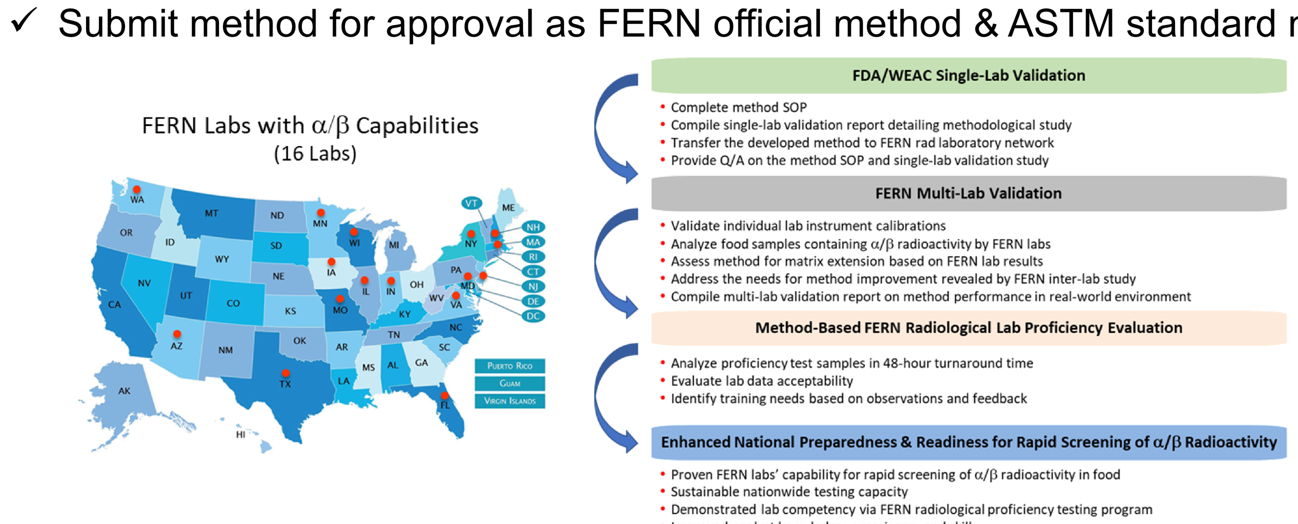
Processed Food Samples for GPC Counting

Procedure Flowchart



❖ Multi-lab Collaborative Study

- ✓ Submit method for approval as FERN official method & ASTM standard method



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