Detection of Alpha/beta Radioactivity in Food for Safeguarding the Nation's Food Supply

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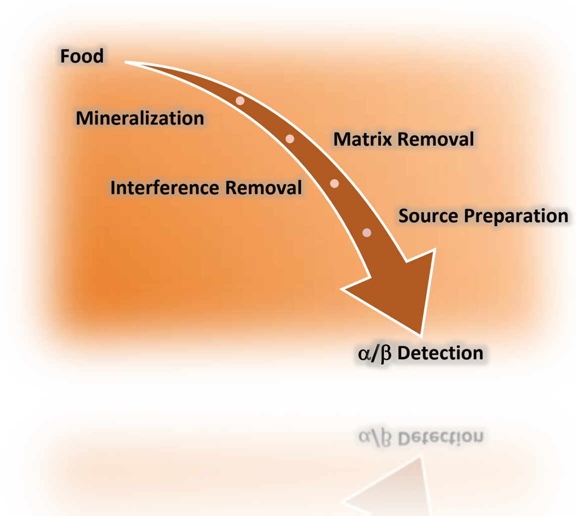
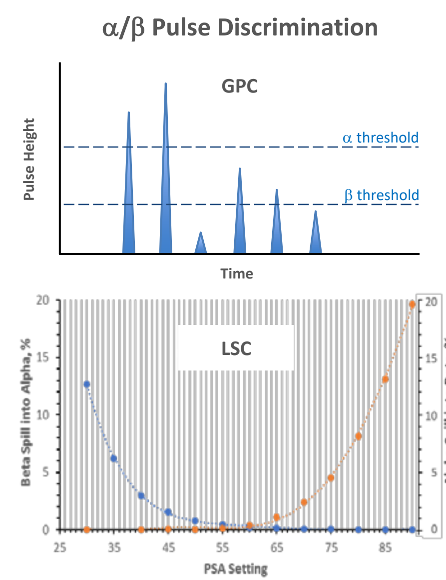
Food and Drug Administration (FDA)





Outline

- **Public's Perception/Expectation on Radiological Food Safety**
  - Perception gap on radiation and risk
  - Insistent public demand on monitoring of radioactivity in food
- **Challenges in Detection of Alpha/Beta Radioactivity in Food**
  - Lack of robust and efficient method
  - Decline in radioanalytical skill and expertise
- **Methodological Research and Development Efforts**
  - Developing rapid/versatile α/β screening method
  - Ensuring broad method applicability
- **Enhancing National Testing Capability and Capacity**
  - Leverage available radioanalytical resources
  - Assessing method acceptability and laboratory proficiency
- **Summary**
  - Current status
  - Future development

▪ Public's Perception/Expectation on Radiological Food Safety

Perception gap on radiation and risk

Misconception-driven psyche:

- ✓ *Radiation exposure causes genetic defect, cancer, and death*

Insistent demand on monitoring radioactivity in food

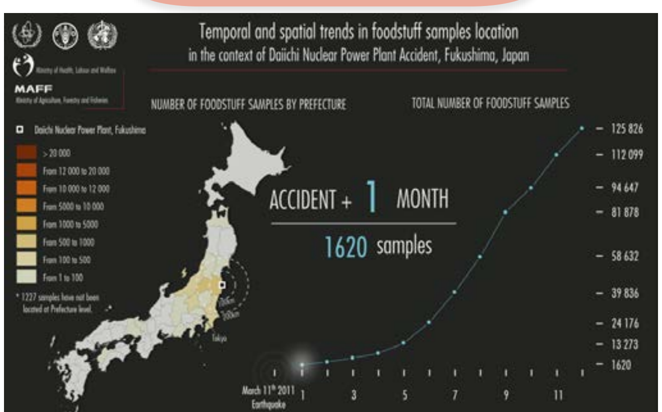
See-to-believe mentality:

- ✓ *Right-to-know sentiment*
- ✓ *Comprehensive food screening*
- ✓ *Data-driven trust and confidence*

Food Monitoring after Fukushima Nuclear Accident

Demand on Rapid Testing:

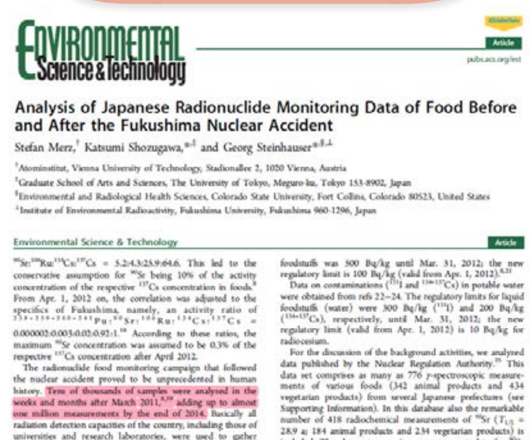
~1620 food tests in 1st month



Courtesy of United Nation Scientific Committee (UNSC)

Demand on Testing Capacity:

~1,000,000 food tests in 4 years



Environmental Science & Technology

Article

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Analysis of Japanese Radionuclide Monitoring Data of Food Before and After the Fukushima Nuclear Accident

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Environmental Science & Technology — Article

$^{90}Sr:^{106}Ru:^{134}Cs:^{137}Cs$ = 5.2:4.3:25.9:64.6. This led to the conservative assumption for ^{90}Sr being 10% of the activity concentration of the respective ^{137}Cs concentration in foods.[8] From Apr. 1, 2012 on, the correlation was adjusted to the specifics of Fukushima, namely, an activity ratio of $^{238+239+240+241}Pu:^{90}Sr:^{106}Ru:^{134}Cs:^{137}Cs$ = 0.000002:0.003:0.02:0.92:1.[18] According to these ratios, the maximum ^{90}Sr concentration was assumed to be 0.3% of the respective ^{137}Cs concentration after April 2012.

The radionuclide food monitoring campaign that followed the nuclear accident proved to be unprecedented in human history. Tens of thousands of samples were analyzed in the weeks and months after March 2011,[8,19] adding up to almost one million measurements by the end of 2014. Basically all radiation detection capacities of the country, including those of universities and research laboratories, were used to gather crucial information on the radionuclide contamination levels in

foodstuffs was 500 Bq/kg until Mar. 31, 2012; the new regulatory limit is 100 Bq/kg (valid from Apr. 1, 2012).[8,21]

Data on contaminations (^{131}I and $^{134+137}Cs$) in potable water were obtained from refs 22−24. The regulatory limits for liquid foodstuffs (water) were 300 Bq/kg (^{131}I) and 200 Bq/kg ($^{134+137}Cs$), respectively, until Mar. 31, 2012; the new regulatory limit (valid from Apr. 1, 2012) is 10 Bq/kg for radiocesium.

For the discussion of the background activities, we analyzed data published by the Nuclear Regulation Authority.[25] This data set comprises as many as 776 γ-spectroscopic measurements of various foods (342 animal products and 434 vegetarian products) from several Japanese prefectures (see Supporting Information). In this database also the remarkable number of 418 radiochemical measurements of ^{90}Sr ($T_{1/2}$ = 28.9 a; 184 animal products and 234 vegetarian products) is included. The data set comprises measurement data for ^{90}Sr

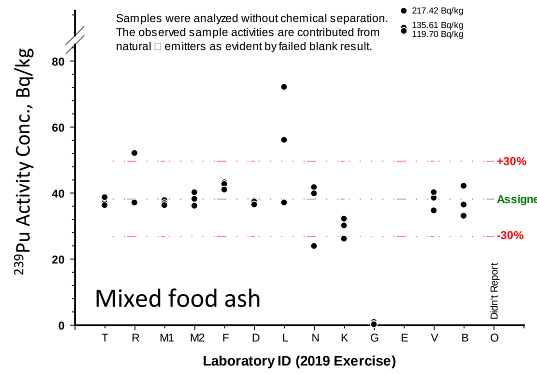
Challenges in Detection of Alpha/Beta Radioactivity in Food

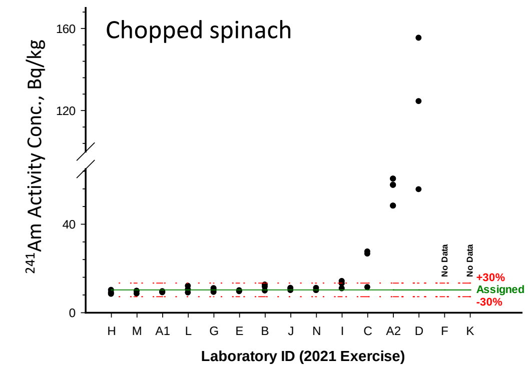
Lack of robust and efficient method

α method

- Screening
  - SM7110-C
  - Lab in-house methods
  - WEAC-NR-Method 7.0
  - Modified EPA 900.0
  - ASTM D7283
- Quantitative
  - EPA 402-R-10-001b
  - Lab in-house methods
  - Modified Eichrom ACW17VBS

α Pass Rate: 69 - 77%

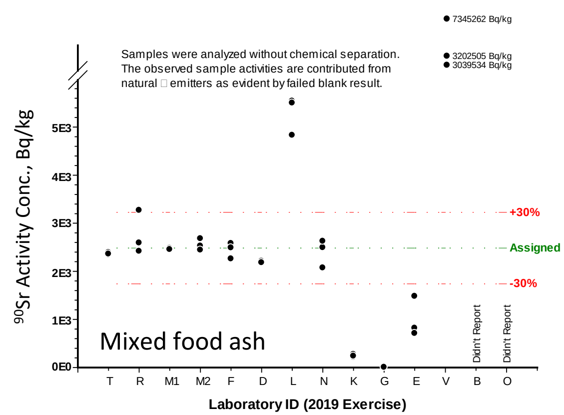


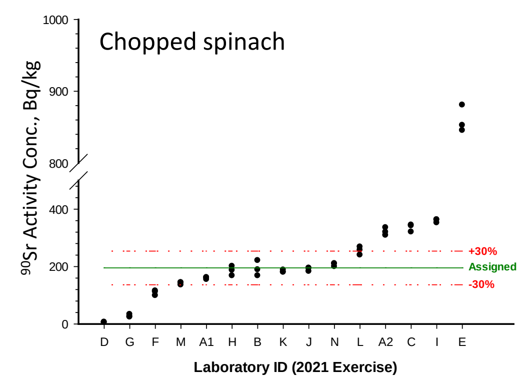


β method

- Screening
  - SM7110-B/EPA 900
  - Lab in-house methods
  - WEAC-NR-Method 7.0
  - Modified EPA 900.0
  - ASTM D7283
- Quantitative
  - EPA 402-R-10-001d
  - EPA 905.0
  - Lab in-house methods
  - Modified Eichrom ACW17VBS

β Pass Rate: 50 - 53%

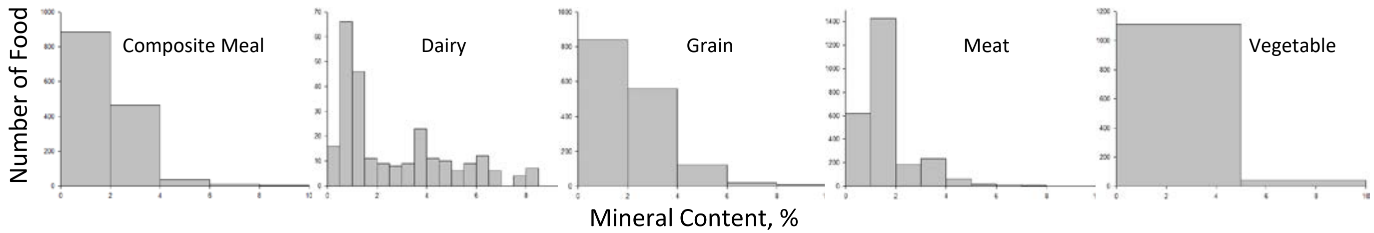




Observations:

✓ Method deficiency

- **Severe matrix attenuation due to lack of matrix removal**



- **Low tolerance to matrix effects**

Fresh food:	Water, carbohydrates, protein, fat, minerals		
Food ash:	Major:	Ca, Cl, Mg, K, Na, P, S	
	Minor:	Cr, Co, Cu, Fe, Mn, Mo, Ni, Se, Si, Sn, V, Zn, Tl, Pd, Pb, Cd, Be, Hg, As, F, I,	
	Compound:	Oxides, sulfates, carbonates, phosphates, chlorides, silicates	

- **Subject to interferences of natural radionuclides**

^{40}K Series
- 1 β-emitter

^{232}Th Series
- 7 α-emitters
- 5 β-emitters

^{238}U Series
- 8 α-emitters
- 6 β-emitters

- **Dissimilarity between calibration standard and sample**

✓ Measurement Inefficiency

- **Sample prepared separately for detection of α and β radioactivity**
- **Tedious and time-consuming radiochemical separations**
- **Only suitable for detecting specific radionuclides**
- **Complex counting source preparation**

ROOT CAUSE ANALYSIS

▪ Challenges in Detection of Alpha/Beta Radioactivity in Food

Decline in radioanalytical skill and expertise

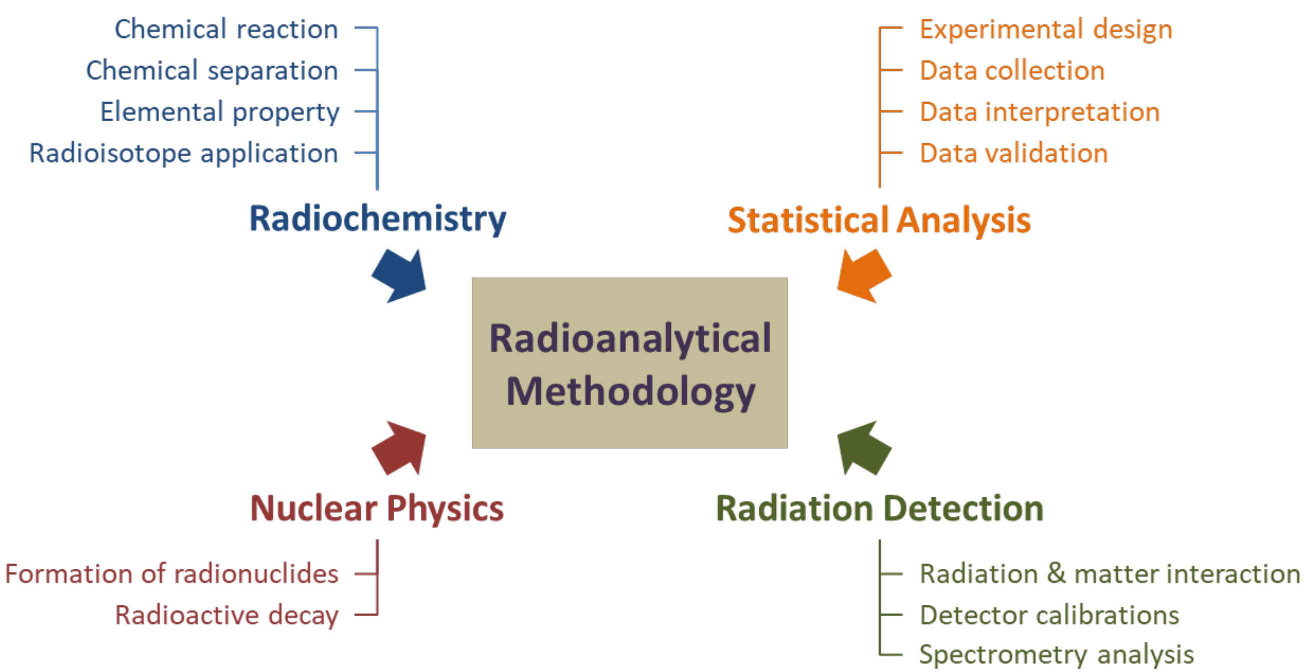
✓ **Loss of radioanalytical expertise due to retirement and turnover**

Senior scientists retired or experienced radiochemists left for better job opportunities

✓ **Empty pipeline**

Little or no radiochemistry course offered at universities due to lack of interest and funding

Radioanalytical Methodology - Interdisciplinary Science



▪ Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

✓ **General process for analyzing α/β radioactivity in food**

Edible Food ⇨ Sample Mineralization ⇨ Analyte Extraction ⇨ Radioactivity Detection ⇨ Result Calculation

Preferable Method Characteristics

Sample Mineralization:

- Rapid conversion of food into ionic solution
- Suitable for analyte extraction

Analyte Extraction:

- Simple matrix/ interference removal
- Robust for a wide variety of foods
- Adaptable procedure for different types of instruments

Radioactivity Detection:

- Simultaneous detection of α/β radioactivity
- High detection efficiency

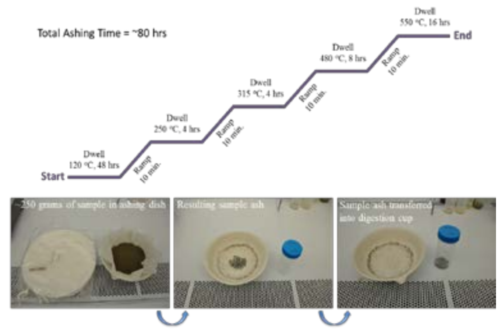
▪ Methodological Research and Development Efforts

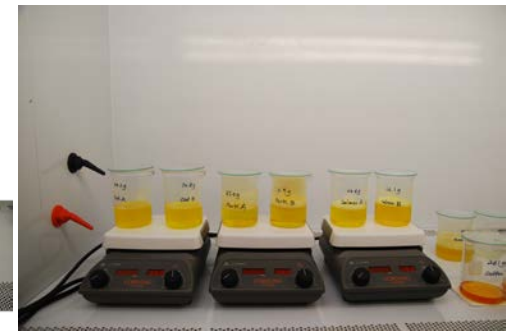
Developing rapid/versatile α/β screening method

✓ **Mineralization of Food**

Wet Ashing

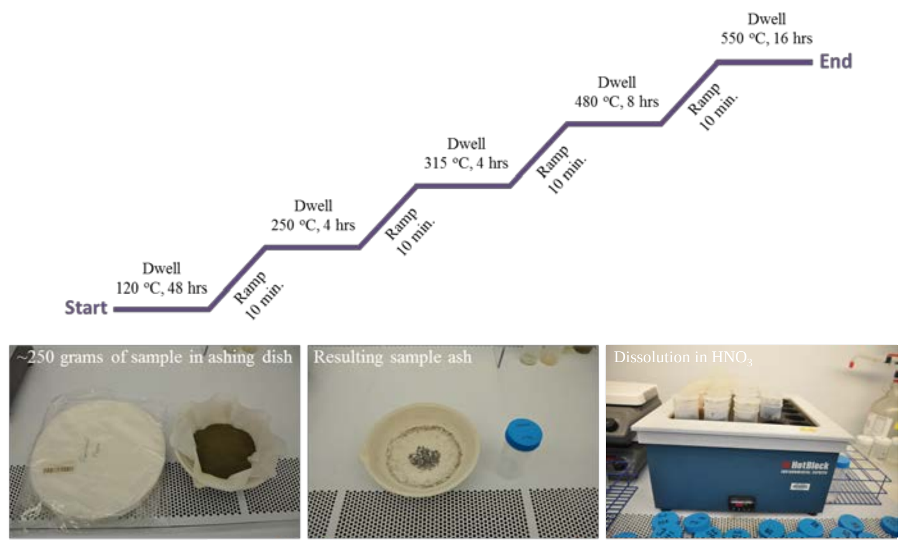
- Batch of 8 samples
- ~35 g per sample
- 2.5-hr process
- 8M HNO_3 solution





Dry Ashing

- Large number of samples
- ~250 g per sample
- Room temp to 550 ^{o}C
- 80-hr process
- 8M HNO_3 solution



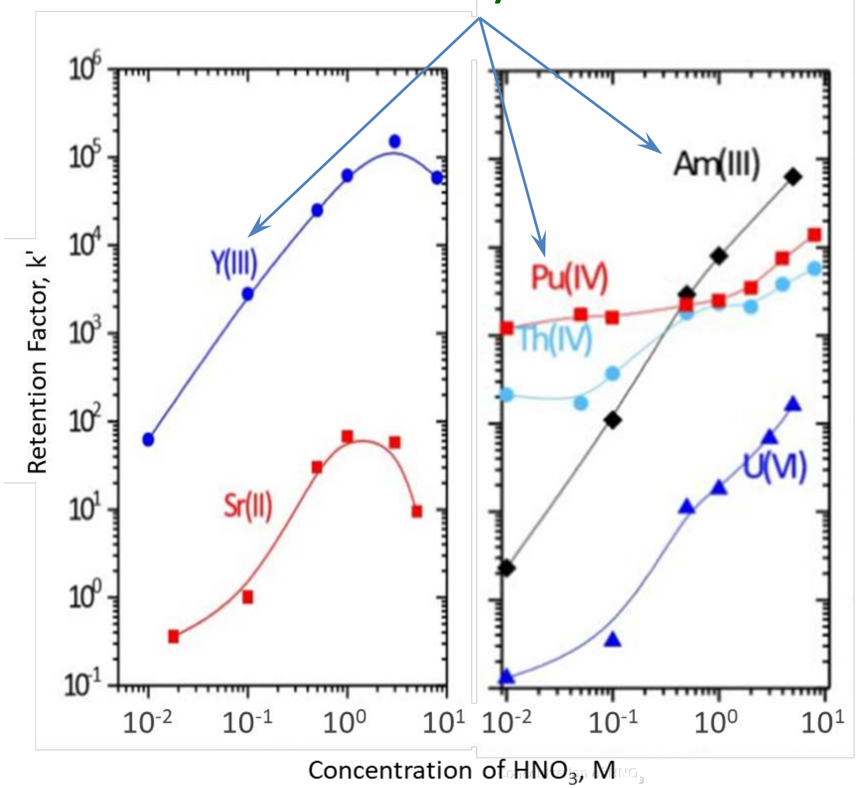
▪ Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

✓ Analyte Extraction

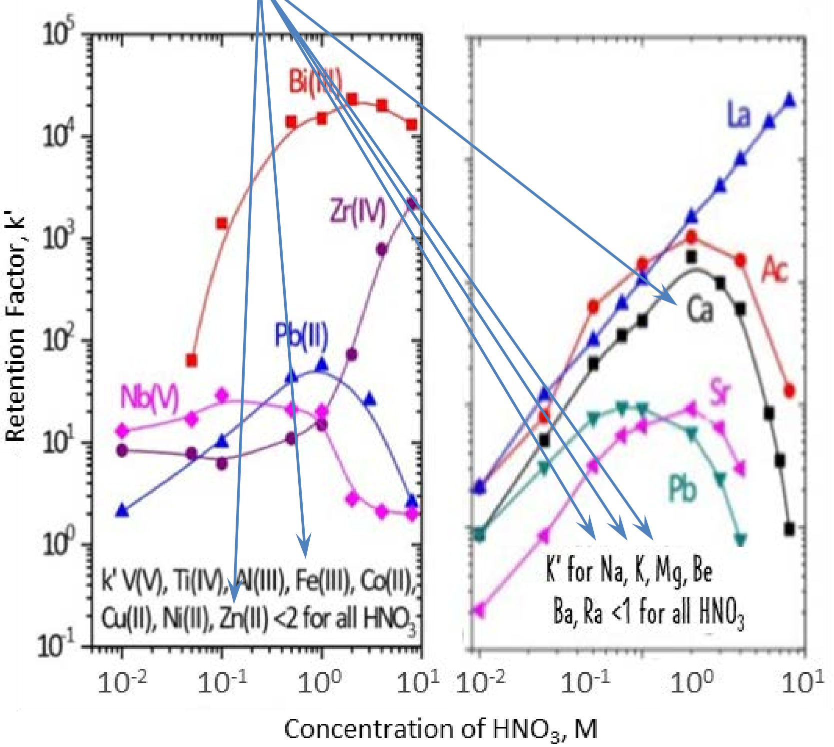
One-Step Cleanup using DGA Resin

α/β Analyte



Matrix Elements

Interfering Radionuclides



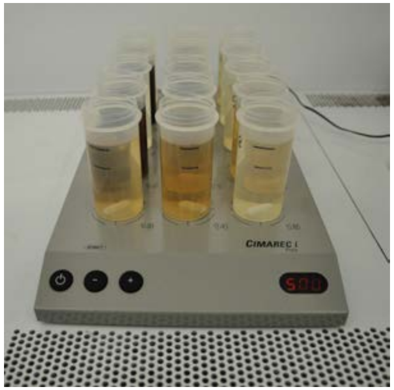
▪ Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

✓ **Analyte Extraction**

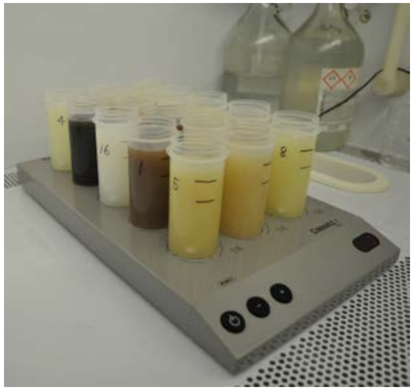
Simple batch extraction

Sample filtrates on stir plate

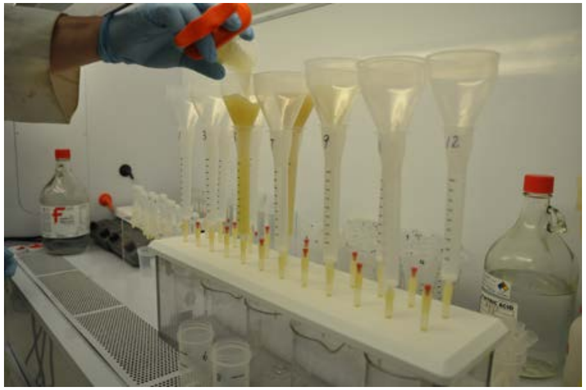


DGA resin added to sample filtrate

Stir sample filtrate for 15 min



Collect analyte loaded DGA resin filter column

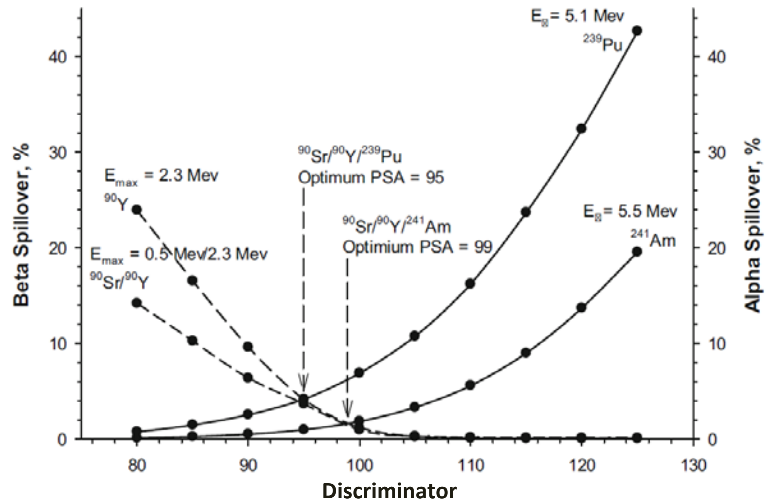


▪ Methodological Research and Development Efforts

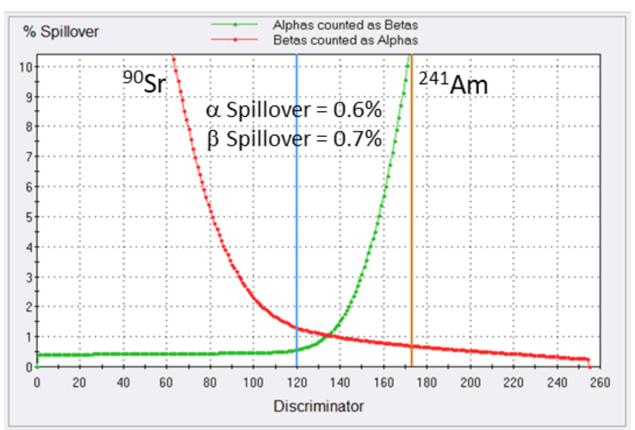
Developing rapid/versatile α/β screening method

✓ **Radioactivity Detection**

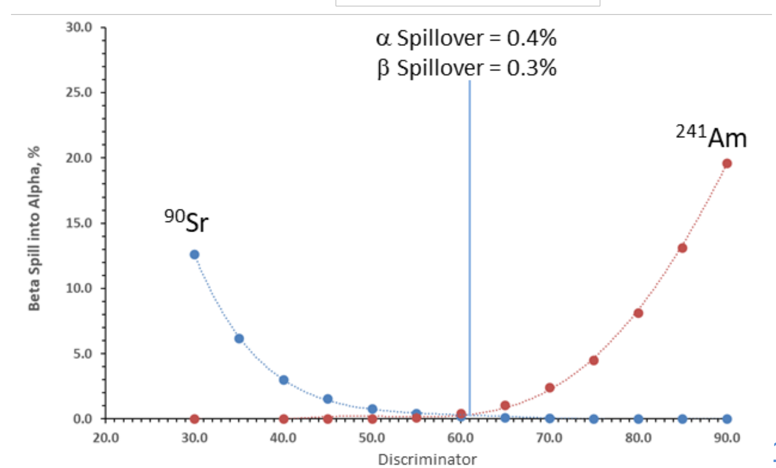
Calibration of liquid scintillation counting (LSC) for simultaneous detection of α/β radioactivity



Quantulus GCT 6220



Quantulus 1220

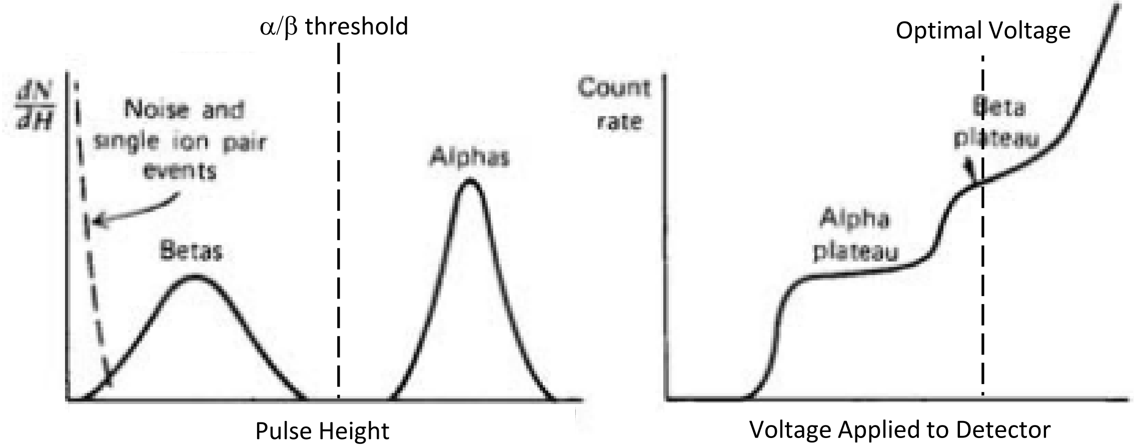


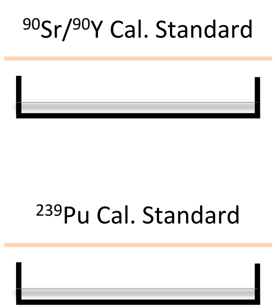
▪ Methodological Research and Development Efforts

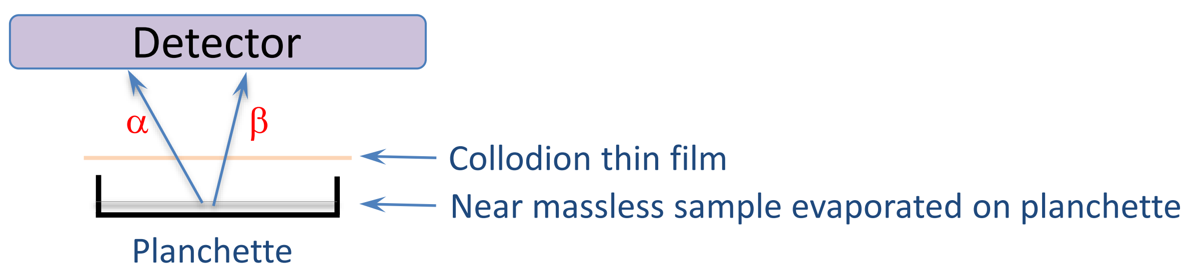
Developing rapid/versatile α/β screening method

✓ Radioactivity Detection

Calibration of gas-flow proportional counter (GPC) for simultaneous detection of α/β radioactivity



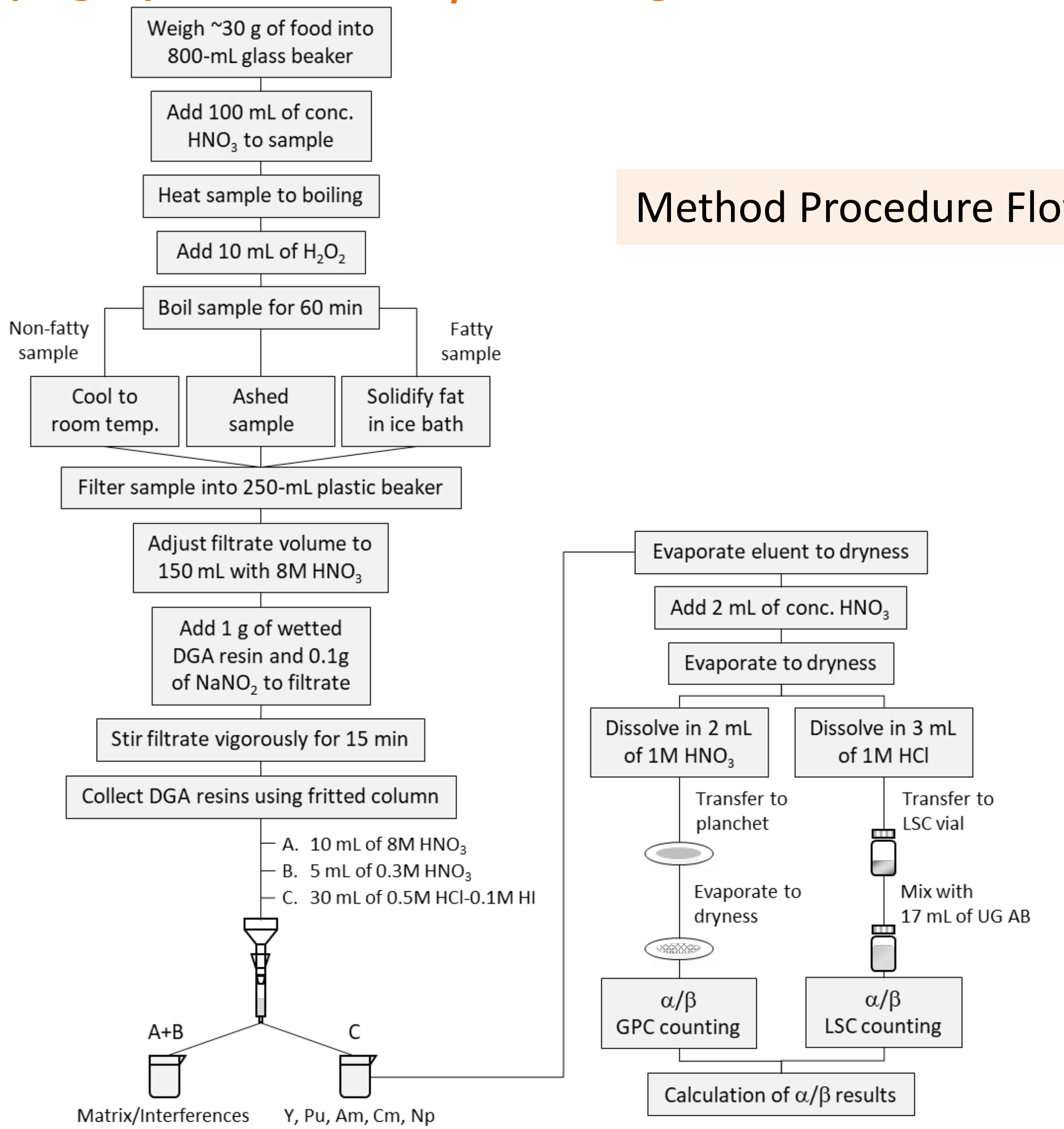




▪ Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

Method Procedure Flowchart

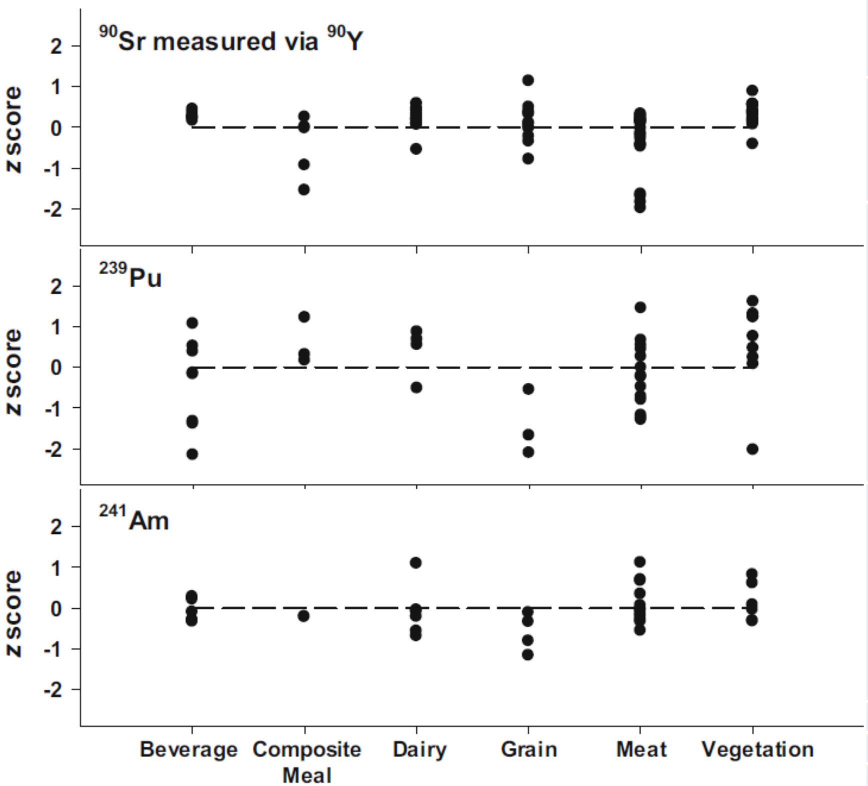


Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

Results of single laboratory study:

Analysis of Spiked Foods



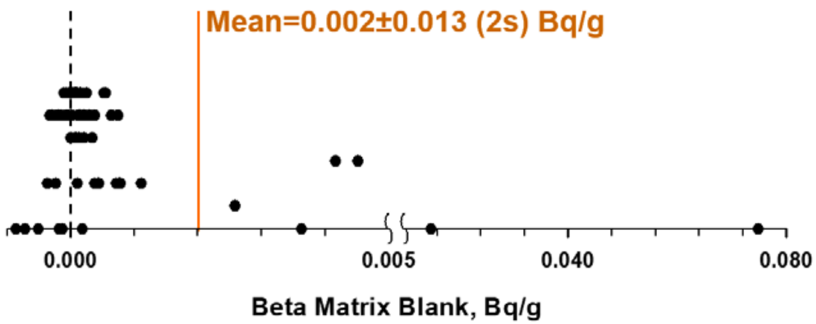
Food Group	Sample Description	Sample Size, g	Extraction Yield, % ^{241}Am	^{239}Pu	^{90}Y
Beverage	Spring water	527.0	-	90.9	95.8
	Tang juice, orange	100.3	97.7	-	97.2
	Sake	100.0	-	80.1	-
Composite Meal	Chicken noodle	100.2	-	88.2	85.8
	Macaroni & cheese	100.5	-	98.8	93.9
	Vegetables, turkey, & barley	100.0	-	89.5	96.2
	Mixed vegetables & beef	55.5	96.5	-	93.6
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
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
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
Vegetation & 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
		Average	98.6	86.7	93.9
		1s	3.9	8.0	4.3
		%, 1s	4.0	9.2	4.6

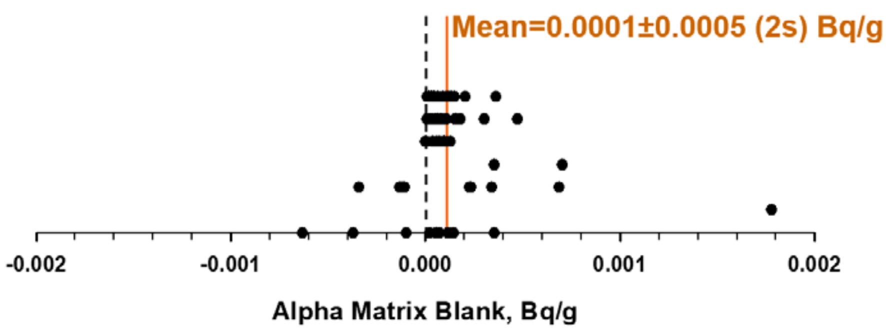
▪ Methodological Research and Development Efforts

Developing rapid/versatile α/β screening method

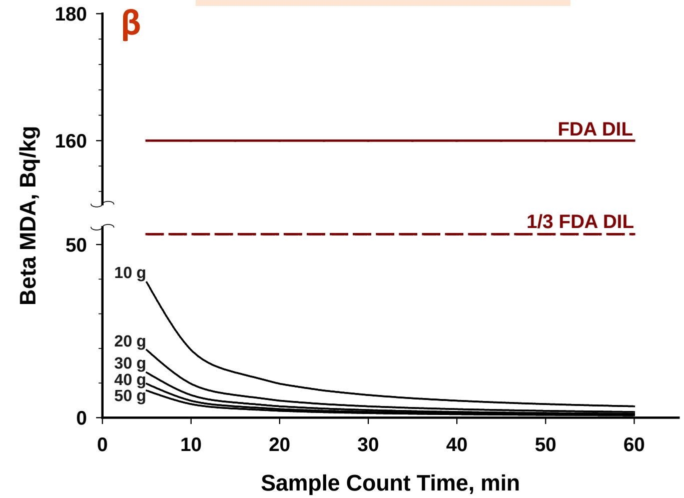
Results of single laboratory study:

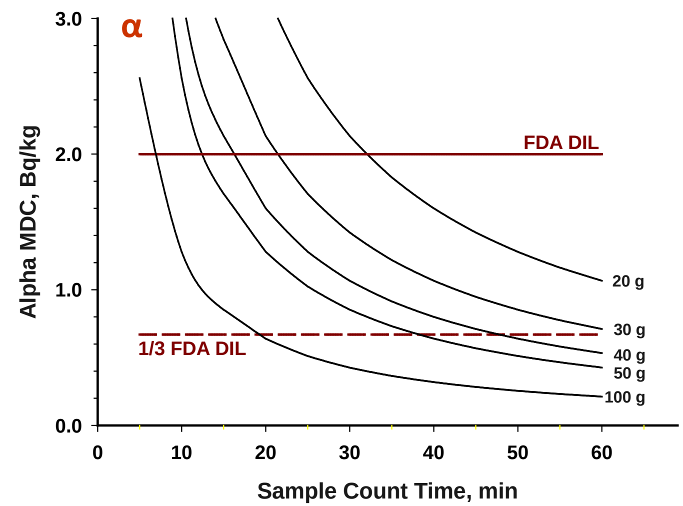
Analysis of Food Matrix Blank





Method Detectability

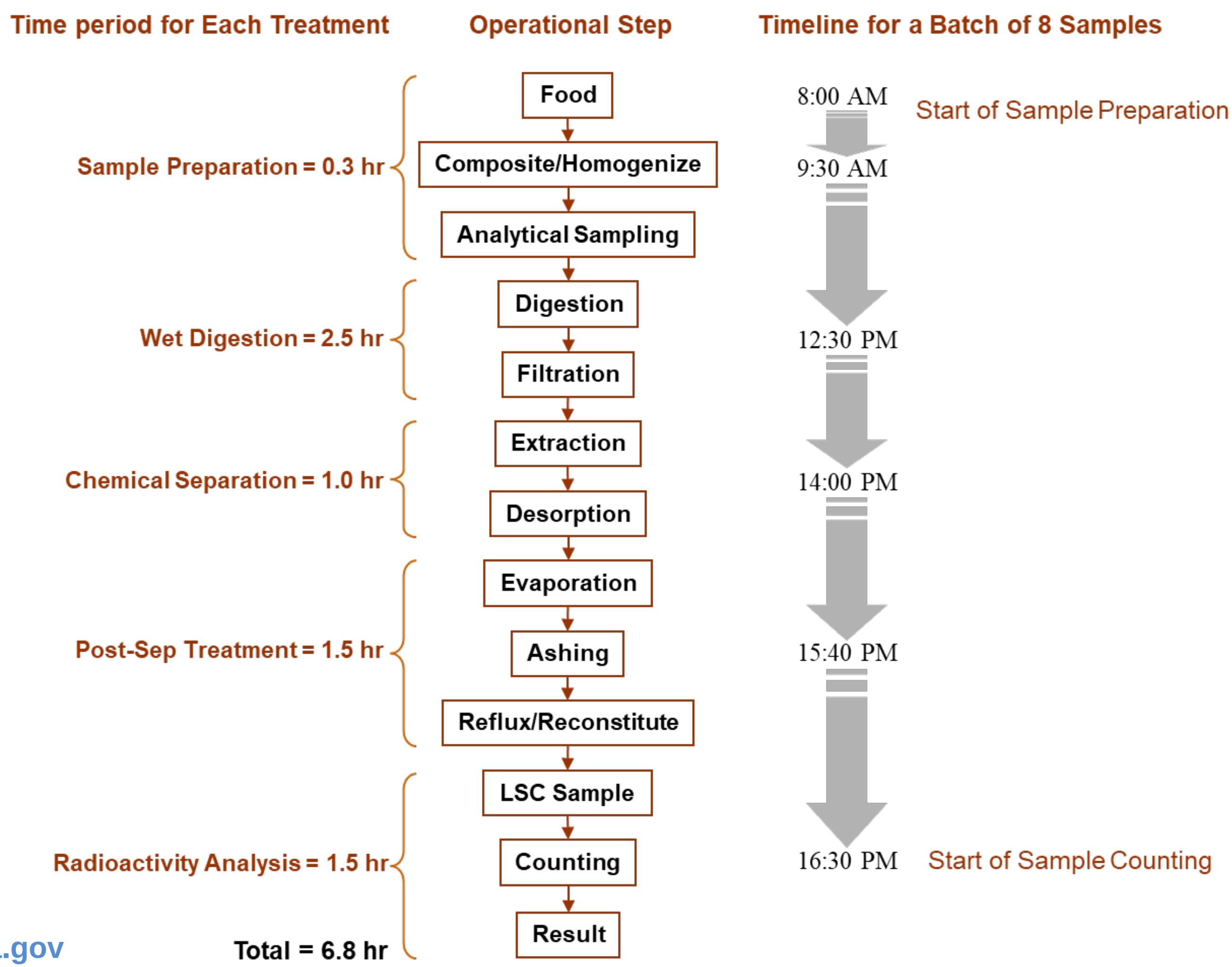




Enhancing National Testing Capability and Capacity

Developing rapid/versatile α/β screening method

Timeline for analysis of a batch of 8 samples



▪ Methodological Research and Development Efforts

Ensuring broad method applicability

Foods selected for multi-lab matrix extension study

Food

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
California olive
Cherry
Blueberry
Strawberry
Peach
Apple sauce
Apple mango kiwi
Tang 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
Big mac
Beef burger
Bacon beef cheeseburger
Baby food rice pudding
Baby food peach
Baby food turkey

Meat

85% lean beef
Branded beef
Conventional beef
Grass-fed 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
Wheat bread
Corn bread
Macaroni
Wheat spaghetti
Rice pudding
Rice cereal
Cooked rice
Breadcrumbs
Corn flakes

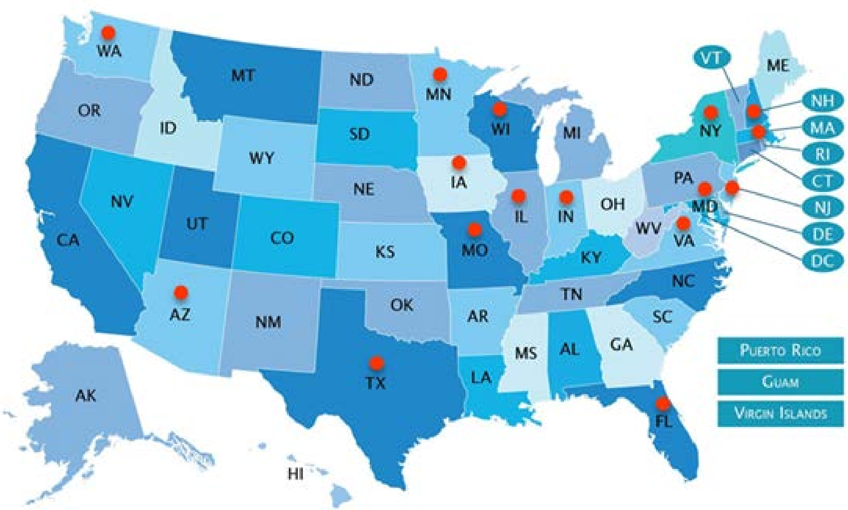
▪ Enhancing National Testing Capability and Capacity

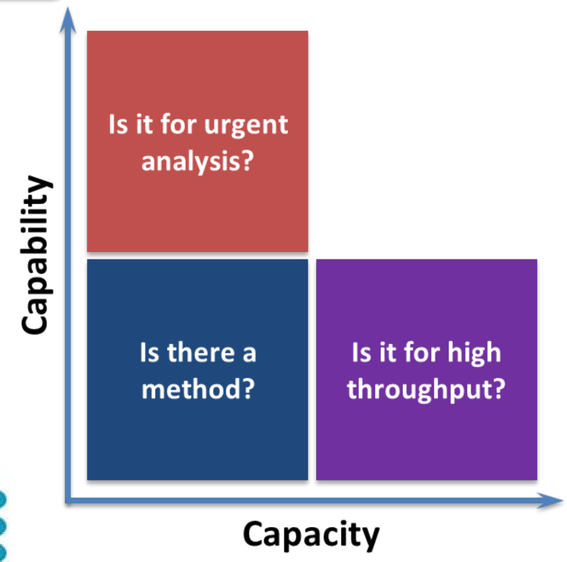
Leverage available radioanalytical resources

Ability to detect α/β radionuclides of interest

- ✓ ^{214}Am
- ✓ ^{238}Pu
- ✓ ^{239}Pu
- ✓ ^{240}Pu
- ✓ ^{242}Cm
- ✓ ^{244}Cm
- ✓ ^{243}Cm
- ✓ ^{89}Sr
- ✓ ^{90}Sr

FERN α/β Capabilities
(16 Labs)



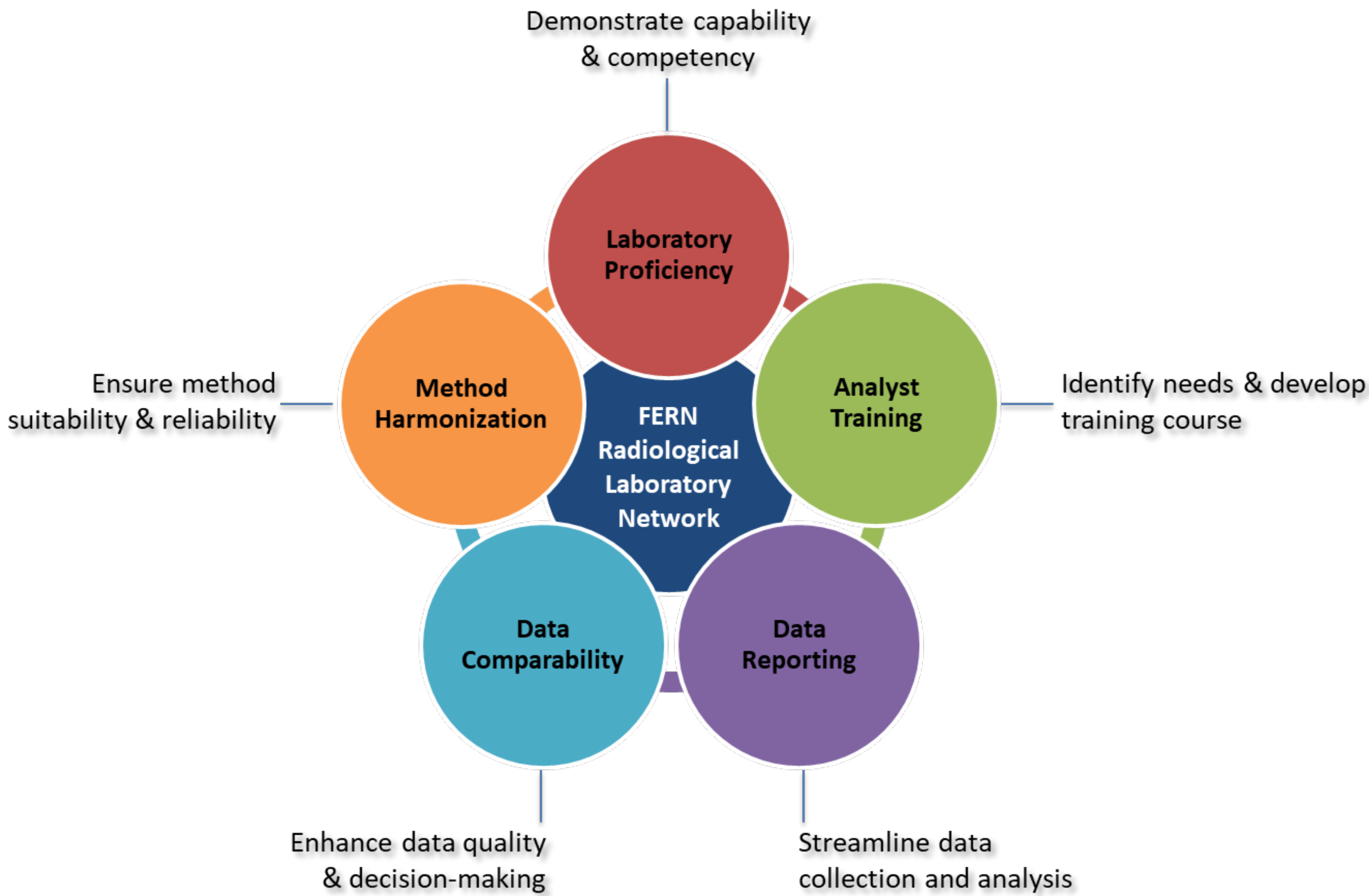


Sufficient testing capacity for perceived emergencies

- ✓ Environmental incident
- ✓ Nuclear power plant accident
- ✓ Nuclear terrorism
- ✓ Nuclear waste leak
- ✓ Nuclear weapon detonation

▪ Enhancing National Testing Capability and Capacity

Assessing method acceptability and laboratory proficiency



Summary

Current status

- ✓ There is certain level of capability and testing capacity for detecting α/β radioactivity in food, but further enhancement is needed
- ✓ The method procedure been studied was found to be robust and suitable for rapid screening of α/β radioactivity in different types of foods
- ✓ The procedure simplicity and versatility enable sustainable operation and maximize use of instruments for high sample throughput
- ✓ Methods established for water analysis were attempted by FERN rad laboratories for detecting α/β radioactivity in food with disappointing results
- ✓ Some methods developed by FERN rad laboratories for food analysis showed limited applicability and need matrix extension study
- ✓ Method development studies taken by FERN rad laboratories are constrained by depleted radiochemistry expertise and absence of training and mentorship
- ✓ Lack of access to suitable food-based reference materials also hampers method development and validation

FERN collaborative study following a fully developed method SOP

▪ Summary

Future Development

FDA/WEAC Single-Lab Validation

- Complete method SOP
- Compile single-lab validation report detailing methodological study
- Transfer the developed method to FERN ad laboratory network
- Provide Q/A on the method SOP and single-lab validation study

FERN Multi-Lab Validation

- Validate individual lab instrument calibrations
- Analyze food samples containing α/β radioactivity by FERN labs
- Assess method for matrix extension based on FERN lab results
- Address the needs for method improvement revealed by FERN inter-lab study
- Compile multi-lab validation report on method performance in real-world environment

Method-Based FERN Radiological Lab Proficiency Evaluation

- Analyze proficiency test samples in 48-hour turnaround time
- Evaluate lab data acceptability
- Identify training needs based on observations and feedback

Enhanced National Preparedness & Readiness for Rapid Screening of α/β Radioactivity

- Proven FERN labs' capability for rapid screening of α/β radioactivity in food
- Sustainable nationwide testing capacity
- Demonstrated lab competency via FERN radiological proficiency testing program
- Improved analyst knowledge, experience, and skill

Thank You!

Questions and Comments

FDA U.S. FOOD & DRUG ADMINISTRATION