An Intercomparison Study on Radiological Methods Used by FDA Food Emergency Response Radiological Laboratory Network

Stephanie Healey, Kathryn Emanuele, Lindsey Mckee, Zhichao Lin, Patrick Regan

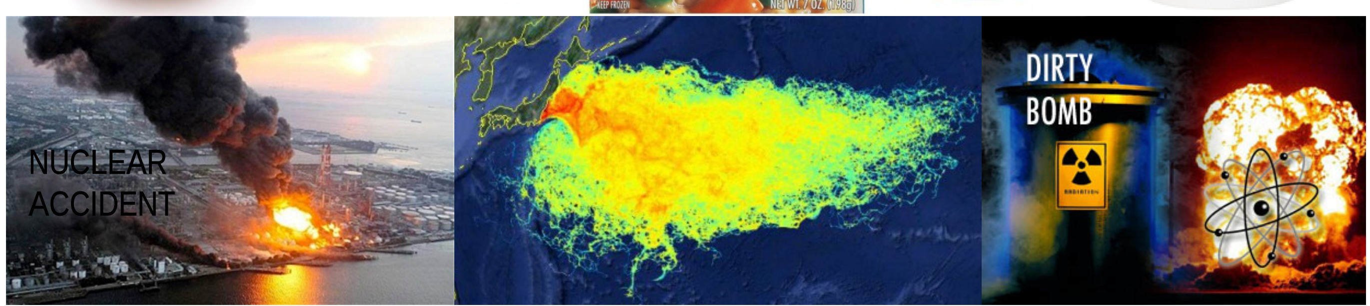
FDA, WEAC, Analytical Branch

Susanne Brooks, Donald H. Burr

FDA, FERN, National Program Office

William Cunningham

Center for Food Safety and Applied Nutrition, FDA



Presentation Outline

- Introduction
- Approach
- Intercomparison studies: Gamma and Tritium
  - Sample Preparation and Verification
  - Results
  - Conclusions
- Future Directions: Materials needed

What is FERN?

- Food Emergency Response Network
  - Chemical, Biological, Radiological
- Network of Laboratories coordinated to respond to threats to the nation’s food supply
  - FDA/ USDA
- Need for Radiological Proficiency Testing
  - Alpha
  - Beta
  - Gamma

Detection of Gamma-emitting Radionuclides in Foods

Background

- Radioactive contamination of food is mostly heterogeneous from either direct (by surface deposition), indirect (via food chain), or induced (expose to neutrons) contaminations.
- Reliable detection of radioactive contamination in food requires test sample to be taken from homogenized food composite
- High-throughput food screening calls for exercising simple, efficient, and practical sample preparation procedure
- Rapid screening capability needs be demonstrated at a measureable activity significantly below the maximum permissible level to minimize the need for confirmatory analysis

Objectives

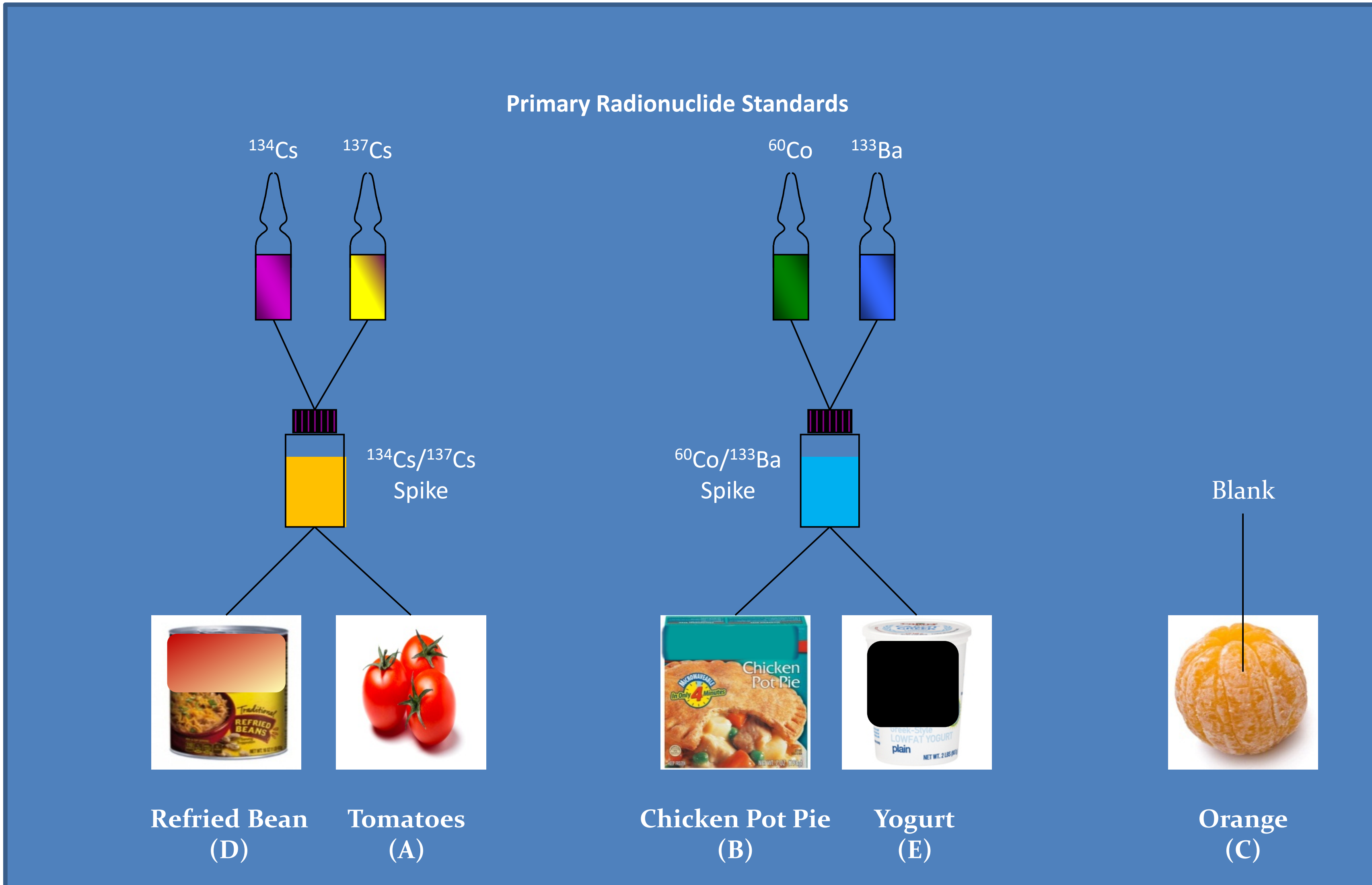
- Practice handling and preparation of different types of radioactive foods in real-world conditions
- Validate FERN radiological labs' ability to correctly identify gamma radionuclides in foods
- Prove capability of g-ray spectrometry methods with heterogeneous samples and quick 10-min counting
- To test speed and efficiency of receiving samples by the FERN labs during emergency response

Approach

- Make use of composite food samples consisting of contaminated and uncontaminated subs
- Conduct proficiency test with regular food products containing blind gamma radionuclides
- Inclusion of blind blank sample to detect potential sample cross contamination
- Test method's detectability at gamma activity levels <1/3 FDA derived intervention levels (DILs)
- Avoid radioactive contamination by limiting sample's activity under the DOT exempt quantity

2015 FERN Gamma Exercise

2015 FERN Gamma Sample Preparation Scheme



Test Sample Basics

Test sample sizes: 0.5 – 1.5 kg

^{134}Cs = 27.47±0.60 Bq/sample
^{137}Cs = 30.09±0.71 Bq/sample — $^{134}Cs/^{137}Cs$ activity ratio = 0.91
^{60}Co = 99.88±1.81 Bq/sample
^{133}Ba = 17.25±0.47 Bq/sample (Outside the scope of study)

Test sample gamma activities were 5 – 36 folds below regulatory limits

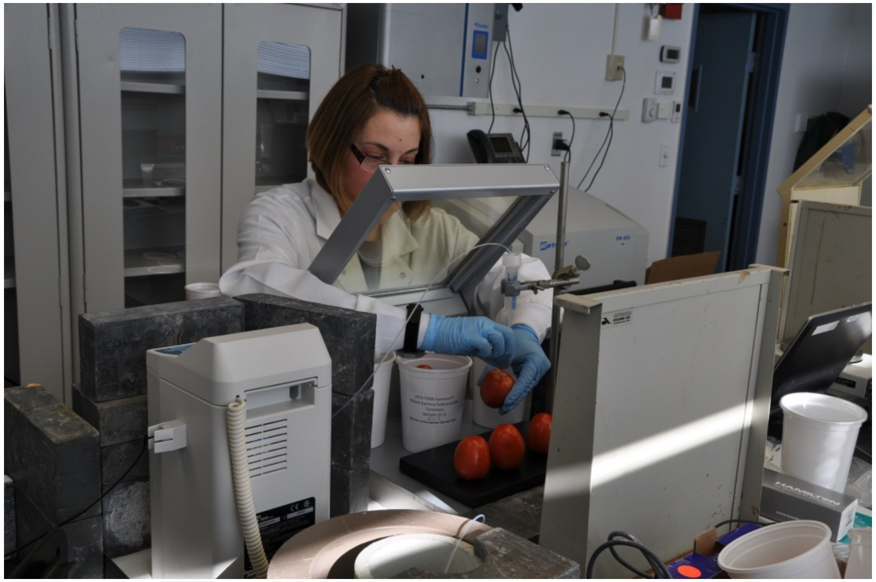
Codex Guideline Level:

^{134}Cs = 1000 Bq/kg
^{137}Cs = 1000 Bq/kg
^{60}Co = 1000 Bq/kg

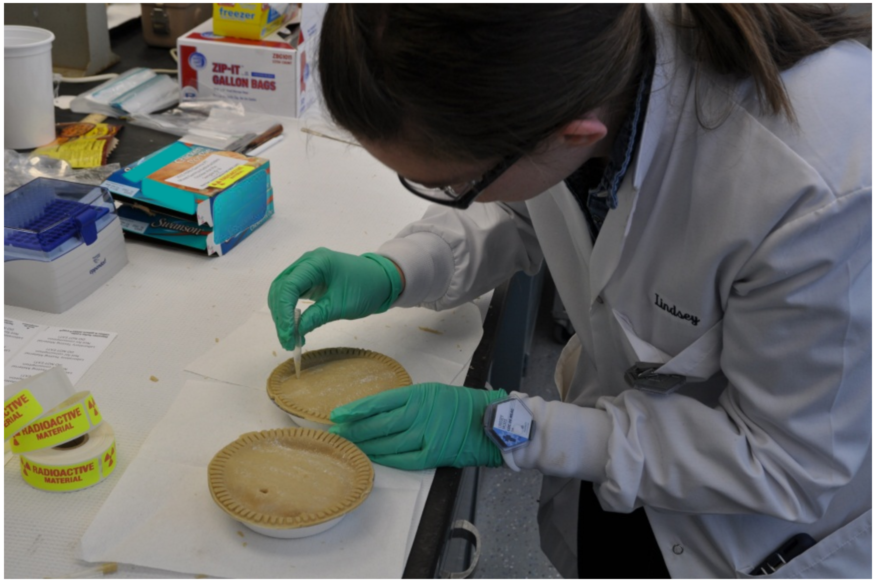
FDA DIL Level:

$^{134}Cs+^{137}Cs$ = 1200 Bq/kg

Spiking Tomatoes

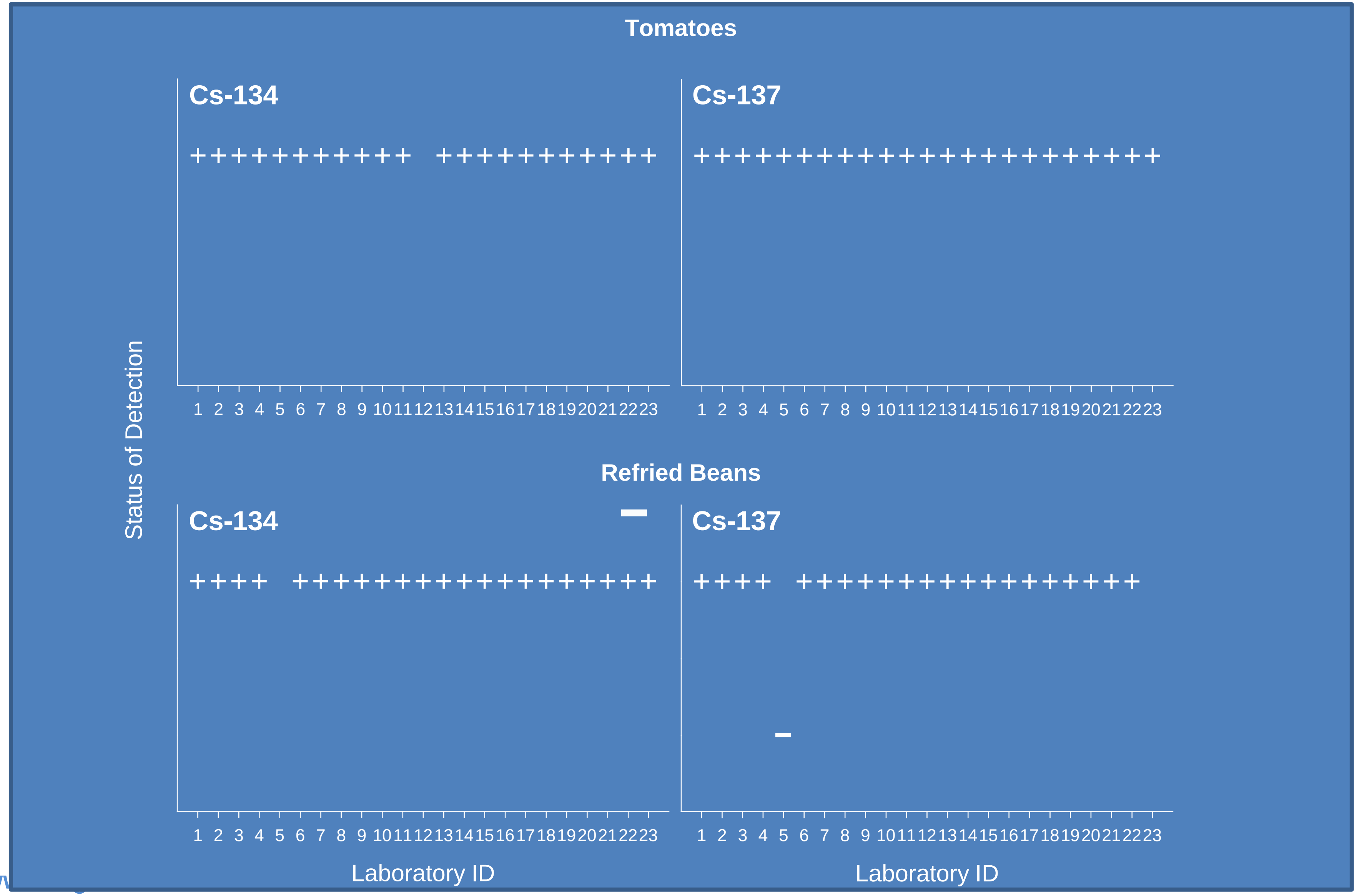


Spiking Chicken Pot Pie

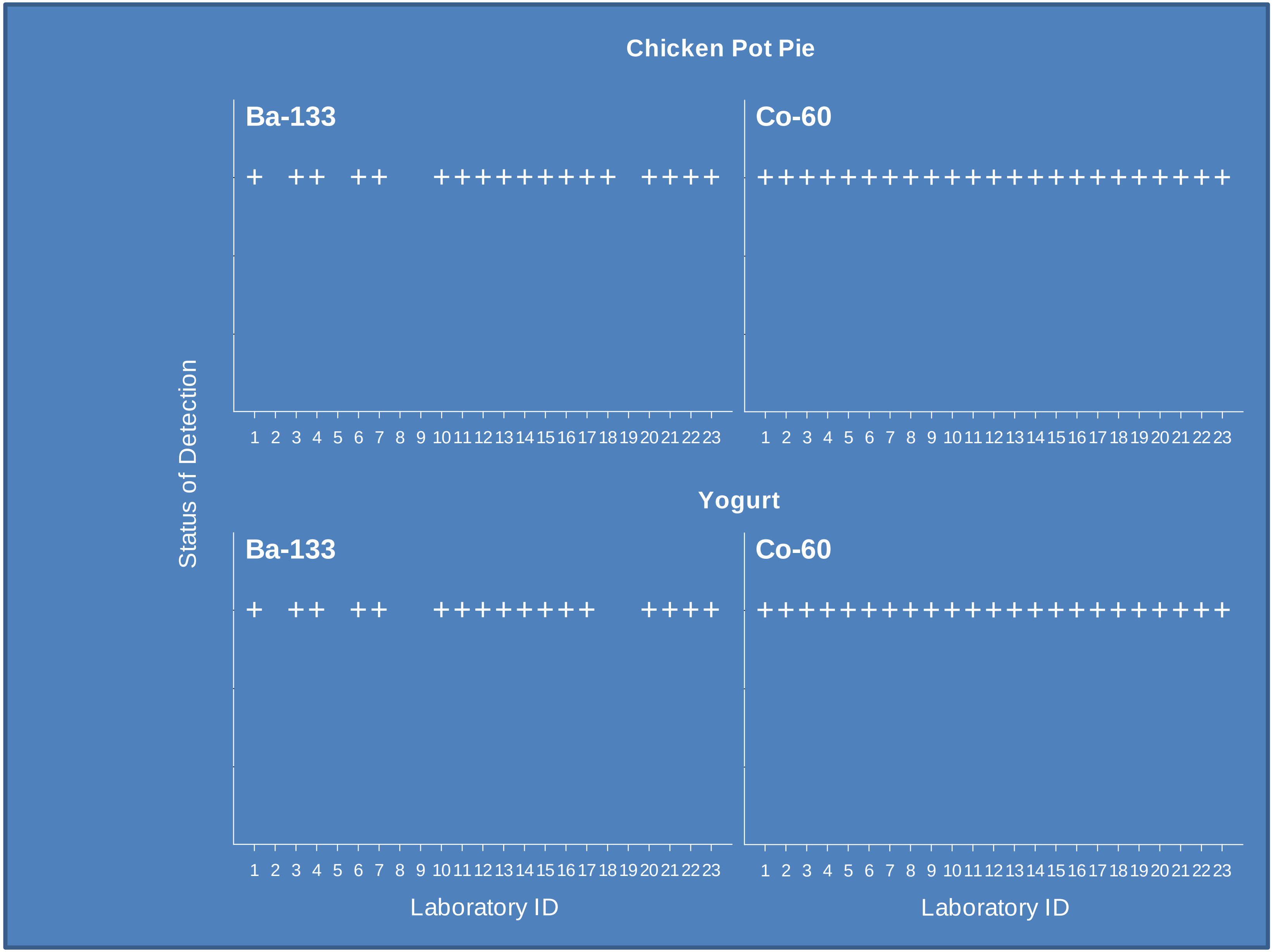


No Spike addition for Oranges

Method Detectability



Method Detectability

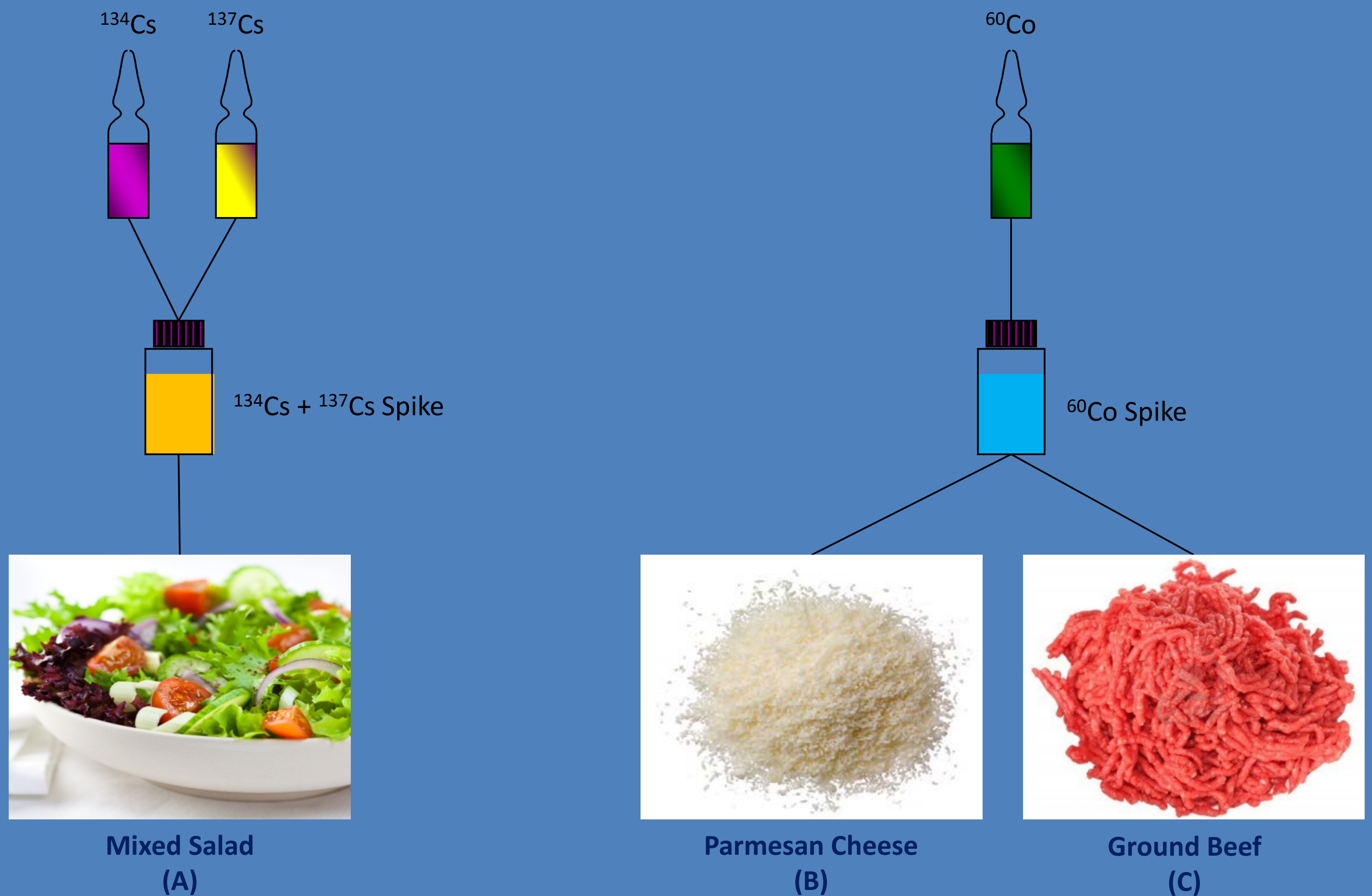


Radionuclide Identification

^{134}Cs	^{137}Cs	^{60}Co	^{133}Ba
100% Correct Identification Rate	**100%** Correct Identification Rate	**100%** Correct Identification Rate	**100%** Correct Identification Rate

2016 FERN Exercise on Detection and Identification of Gamma-Emitting Radionuclides in Food

Primary Radionuclide Standards



Test Sample Basics

Types of food samples: (A) Mixed salad
(B) Parmesan cheese
(C) Ground beef

Range of sample weights: 0.3 – 2.4 kg

Sample Activity:
^{134}Cs = ~229 Bq/kg
^{137}Cs = ~271 Bq/kg
^{60}Co = 176 – 212 Ba/kg

Sample activities were 4.4 – 5.7 times below regulatory limits

Codex Guideline Levels:

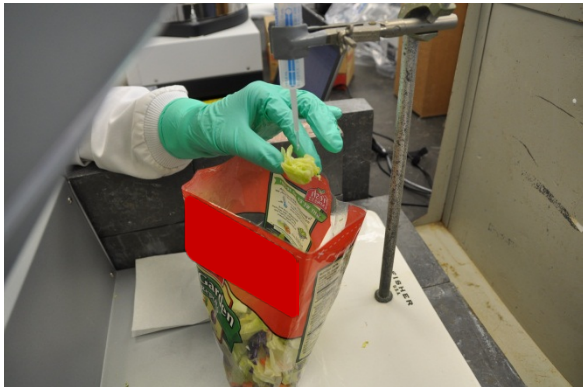
^{134}Cs = 1000 Bq/kg
^{137}Cs = 1000 Bq/kg
^{60}Co = 1000 Bq/kg

FDA DIL Level:

$^{134}Cs+^{137}Cs$ = 1200 Bq/kg

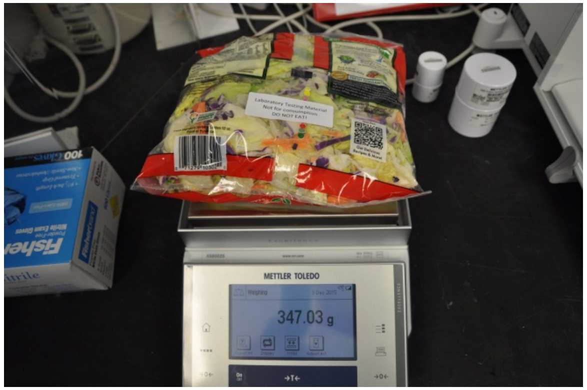
Preparation of Spiked Mixed Salad Samples

Spiking
One piece of lettuce per bag



Sealing
Use of original commercial packing

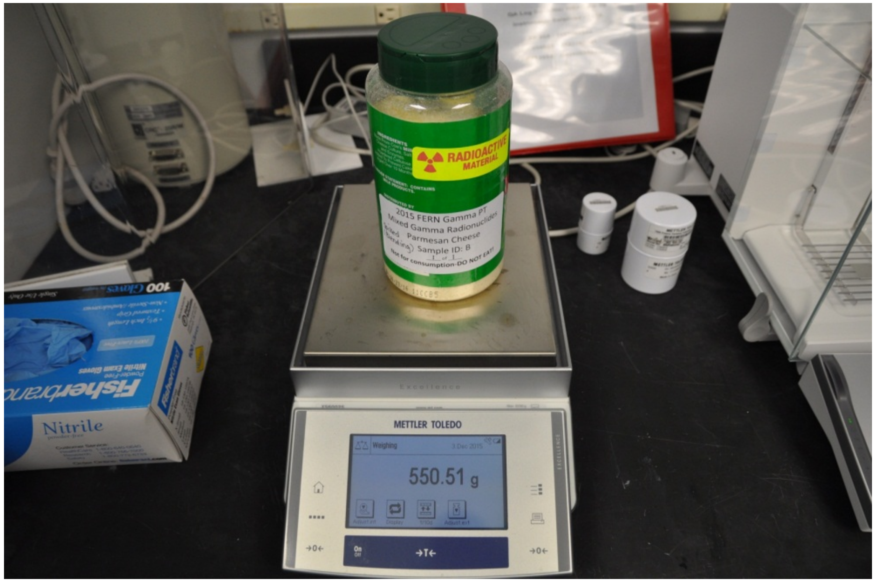
Weighing
Sample activity concentration per fresh weight



Preparation of Spiked Parmesan Cheese Samples

Spiking
One spot at the center of product

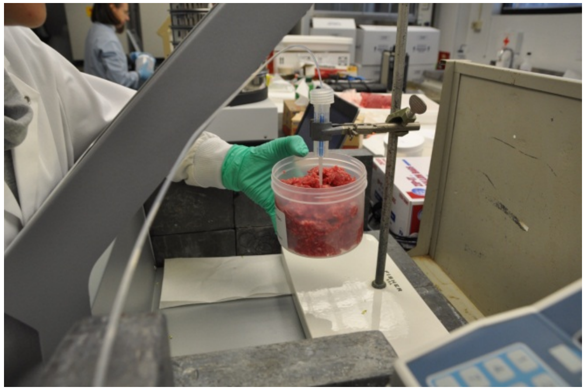
Weighing
Sample activity concentration per sample net weight



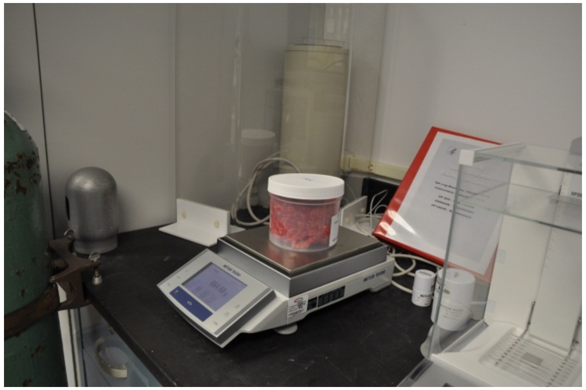
Preparation of Spiked Ground Beef Samples

Preparation
Make one hole at the center of the product

Spiking
Add spike to the hole



Weighing
Activity concentration per net sample weight



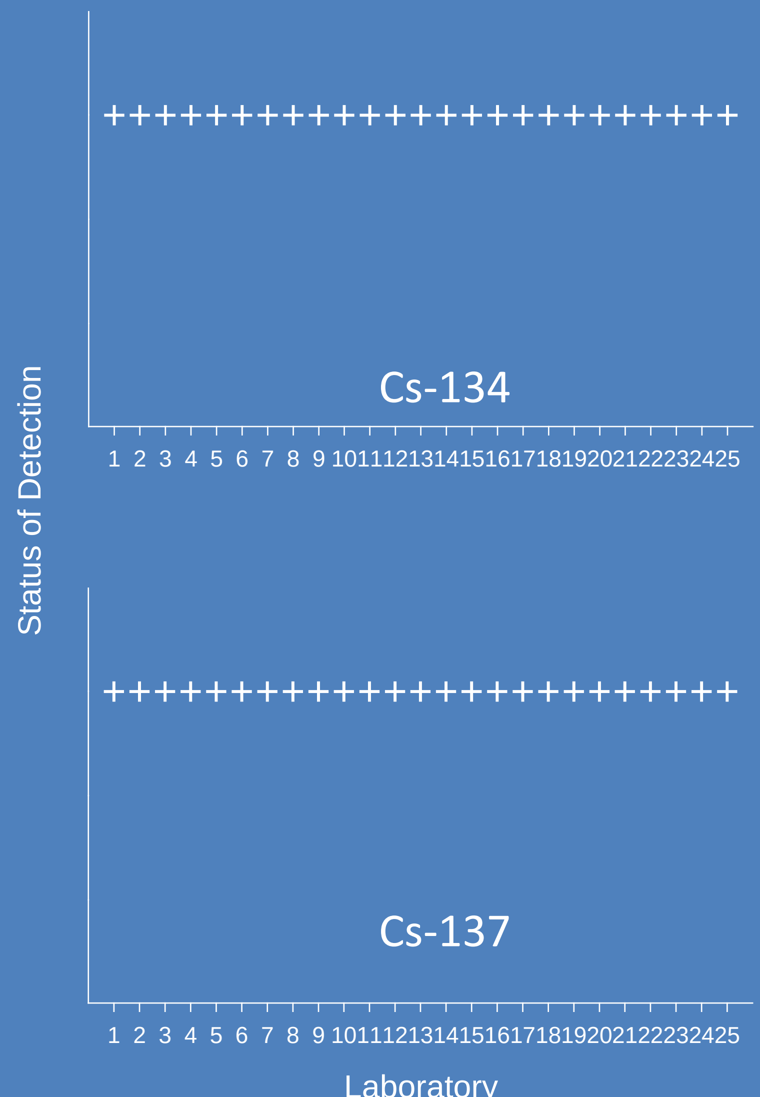
Radionuclide Identification

^{134}Cs	^{137}Cs	^{60}Co
100% Correct	**100%** Correct	**100%** Correct

All participants correctly identified the radionuclides presented in the test samples

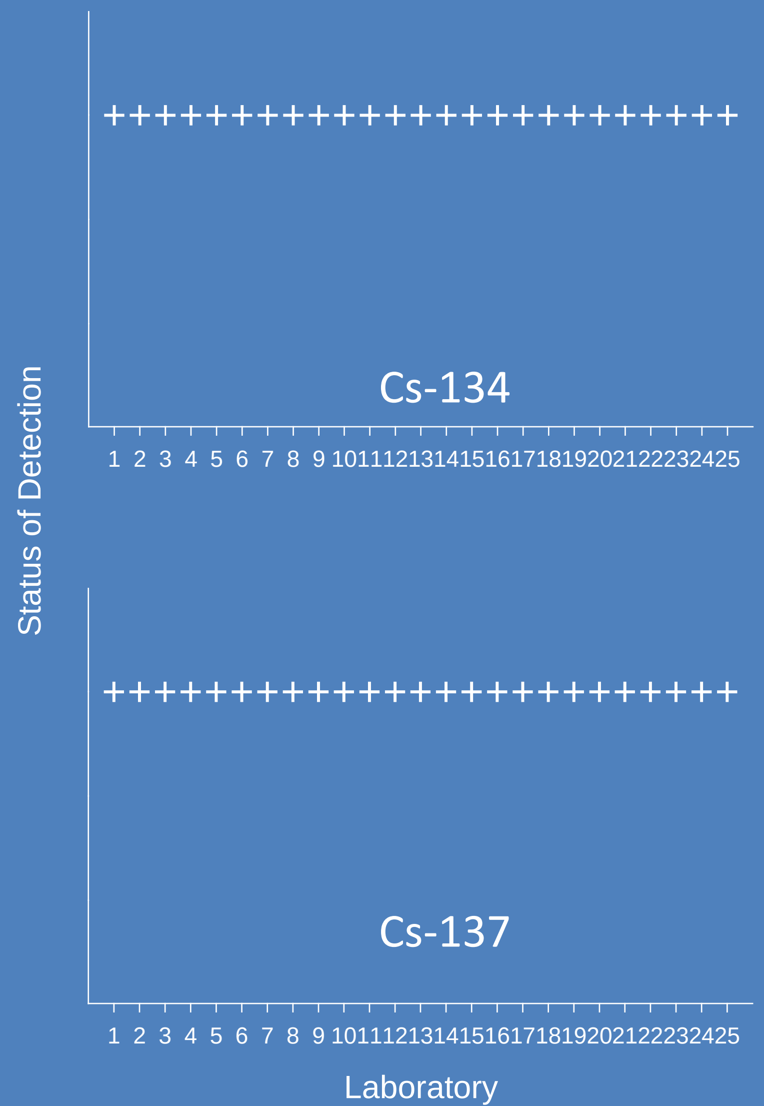
Method Detectability

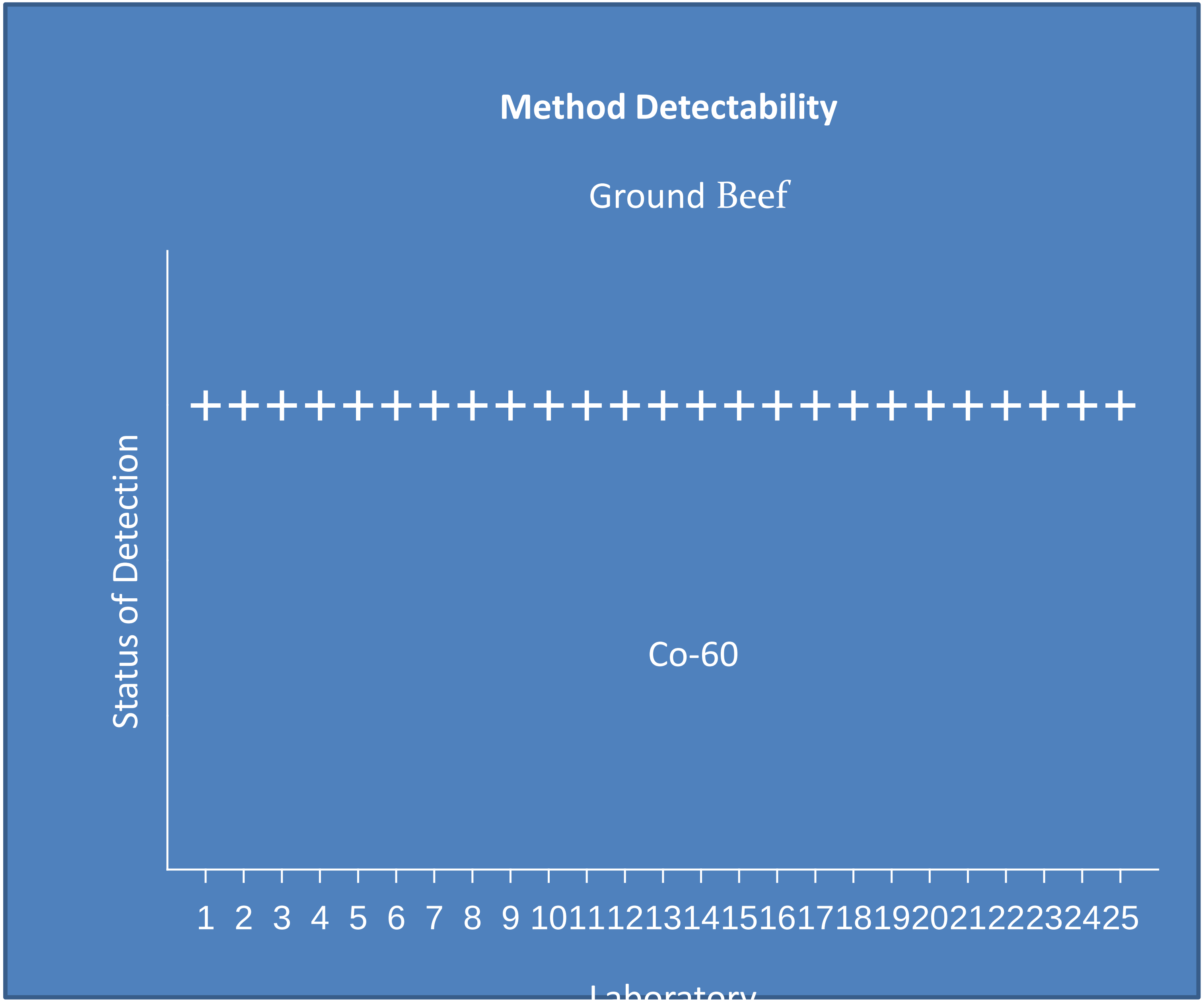
Mixed Salad



Method Detectability

Mixed Salad

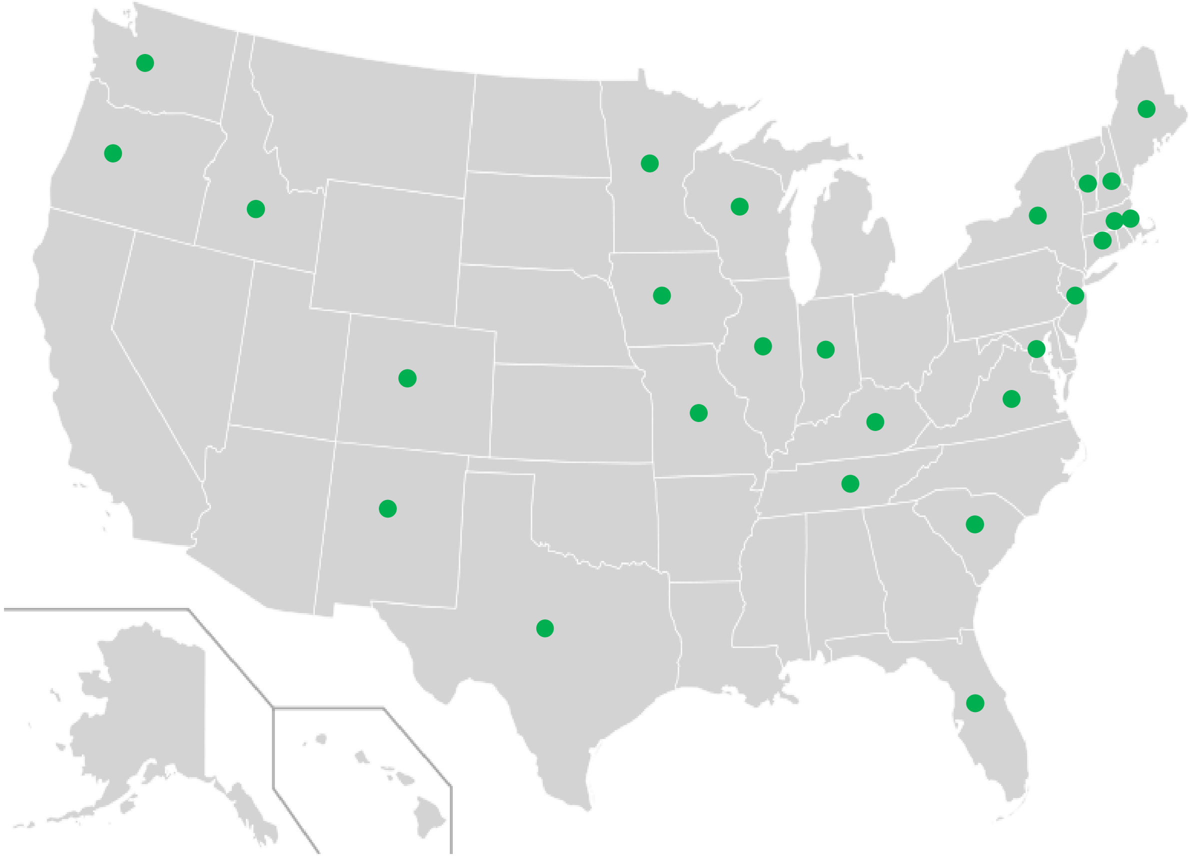



Method Detectability
Ground Beef
Status of Detection
+++++++++++++++++++++++++
Co-60
1 2 3 4 5 6 7 8 9 10111213141516171819202122232425
Laboratory

Difference Between Measured and Known Values, %

	Mixed Salad		Parmesan Cheese	Ground Beef
Laboratory	**Cs-134**	**Cs-137**	**Co-60**	**Co-60**
1	-7	1	-5	8
2	63	83	27	-82
3	-	-	-	-
4	-18	2	2	4
5	-15	-7	8	3
6	-17	-8	-16	2
7	-8	-3	23	39
8	-24	-15	-5	-6
9	33	36	47	-12
10	13	29	19	32
11	-23	-15	-2	-12
12	-9	-9	-1	-19
13	42	-16	18	-16
14	-7	0	-5	-9
15	-11	-8	-8	-14
16	-9	1	14	-1
17	-31	-33	1	-30
18	-	-	-	-
19	-14	-14	-18	-16
20	-10	-7	8	-2
21	-	-	-	-
22	-2	7	5	1
23	68	81	43	8
24	-7	8	6	-3
25	-	-	-	-

Participation and Sample Shipping Around Nation



Summary of Study

- Composite and homogenization of heterogeneously contaminated food products for gamma spectrometric analysis were realistically practiced
- Sample homogenization methods used by some participating laboratories need improvement
- All laboratory's methods were able to correctly identify gamma radionuclides presented in the test food samples
- Radionuclides of interest were detectable down to 1/4 of regulatory limits with 10-min count time
- Previous problems on sample shipping were resolved

2016 FERN Exercise on Detection of Tritium in Food

Mission Relevance

- In the aftermath of a nuclear or radiological emergency, FDA's decision-making on ensuring food safety and protecting public health will be based on large pools of data from its FERN laboratory network that uses diversified analytical methods. This project enables agency:
- To evaluate method performance and acceptability per FDA data quality objectives
- To improve network laboratory proficiency
- To ensure data comparability based on measurement traceability
- To promote method improvement and harmonization

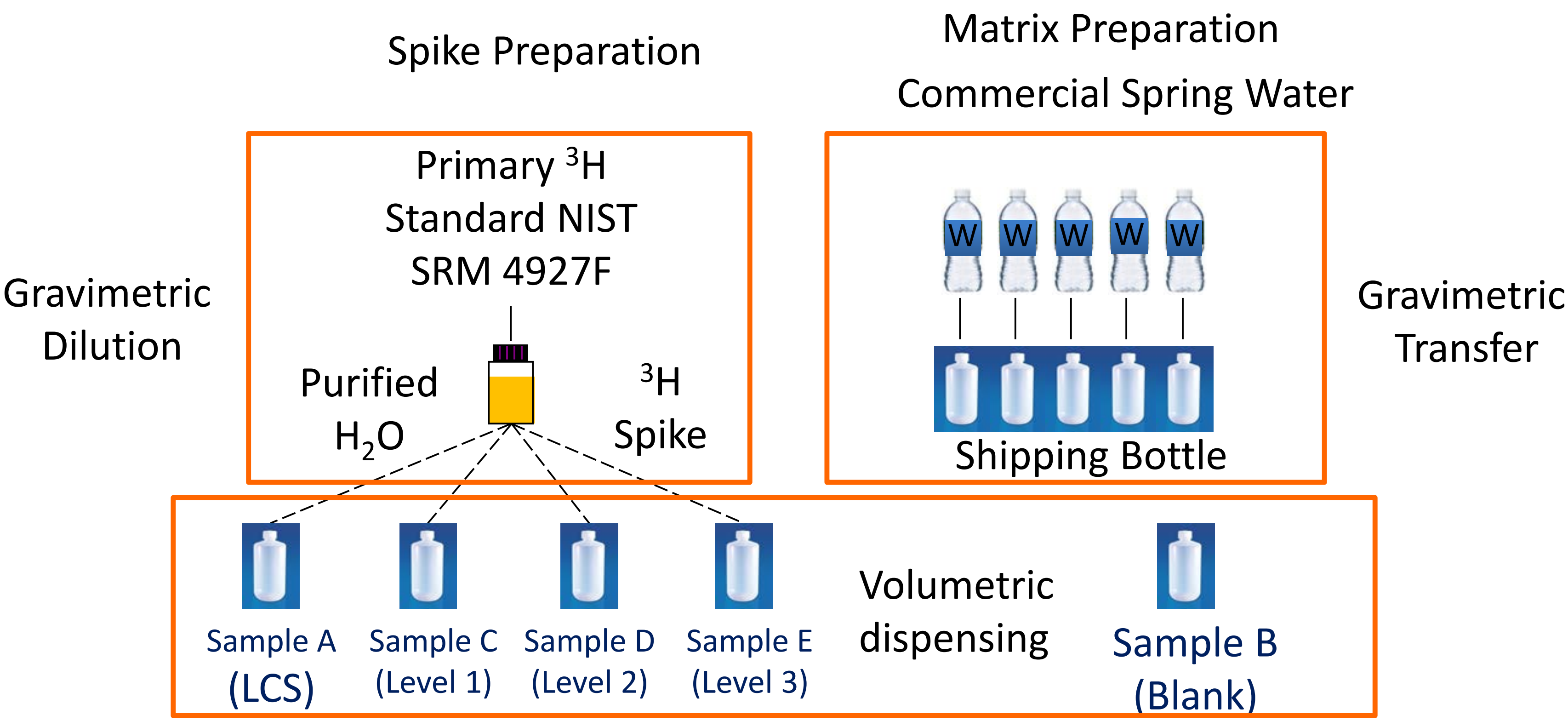
Introduction

- Proven analytical methods and a competent laboratory network are essential for FDA to implement food defense and safety measures under the Food Safety Modernization Act.
- Ambiguous findings from cooperative laboratories would inhibit FDA's ability to take prompt action on protecting food safety and public health
- Method acceptability, laboratory proficiency, and data comparability are essential for effective and efficient risk assessment and management
- Among all radionuclides of concern, those emitting α- and β-radiation pose the greatest health risk via internal radiation exposure from ingestion.
- Tritium (^{3}H) is a β-emitter made both naturally by interactions between nitrogen and cosmic rays and artificially by nuclear explosion/power production
- Tritiated water is the most common form of ^{3}H in the environment and ecosystem
- ^{3}H has ability to incorporate with the DNA and raise cancer risk thus its presence in aquafer, groundwater, and food must be monitored per regulatory guidelines

Approach

- Prepare test samples using commercial bottled water that presents typical matrix and radiometric interferences
- Assess method performance and acceptance at multiple concentration levels
- Include blank and LCS samples for quality control
- Open choice on selection of method and instrument

Sample Preparation and Verification



^3H Blank in Commercial Bottled Water

Bottle ID	Sample Vol. mL	Count Time min	UGLLT mL	LSC Vial Type	CPM	1s%	Quench Index
1	10	60	10	Plastic	2.65	8.01	694.62
2	10	60	10	Plastic	2.82	7.76	697.12
3	10	60	10	Plastic	2.29	8.61	695.84
4	10	60	10	Plastic	2.82	7.76	696.57
5	10	60	10	Plastic	2.48	8.28	694.92
6	10	60	10	Plastic	2.64	8.03	697.03
7	10	60	10	Plastic	2.72	7.91	699.52
8	10	60	10	Plastic	2.43	8.36	699.04
9	10	60	10	Plastic	2.65	8.01	701.25
10	10	60	10	Plastic	2.64	8.03	696.42
11	10	60	10	Plastic	2.64	8.03	699.40
12	10	60	10	Plastic	2.89	7.67	697.72
13	10	60	10	Plastic	2.38	8.45	696.20
14	10	60	10	Plastic	2.72	7.91	697.48
MilliQ H_2O	10	60	10	Plastic	3.28	7.20	697.50
				Mean	2.67		697.38
				1s	0.24		1.81
				1s%	8.93		0.26

- The degrees of sample quench as shown by the Quench Index were found to be consistent among 14 commercial bottled water, which indicated that the commercial bottled water has homogenous composition
- ^3H blank count rates observed in 14 commercial bottled water were found to be very low and quite consistent.
- It appeared that the ^3H blank count rate for commercial bottled water was even lower than the ^3H blank count rate for MilliQ H_2O.

Spike and Test Sample Verification

Sample ID	Gravimetric, Bq/g	Measured, Bq/g	Diff, %
^{3}H Spike:	417.34 ± 3.00	419.07 ± 2.73	0.42

Test Samples:	Known, Bq/kg	Measured, Bq/kg	Diff, %
A23	724.61 ± 11.81	722.15 ± 13.72	-0.34
C23	729.01 ± 11.88	718.82 ± 17.54	-1.40
D23	364.37 ± 4.74	366.69 ± 12.39	0.64
E23	239.12 ± 3.35	241.45 ± 9.95	0.97

- For ^{3}H spike, the agreement between its gravimetric and measured values was found to be 0.42%, which demonstrated a traceability between the ^{3}H spike and its primary standard
- For a set of randomly selected test samples, the agreements between the known and measured values were found to be better than -1.4% and 0.97%, which verified that the sample spiking was performed correctly

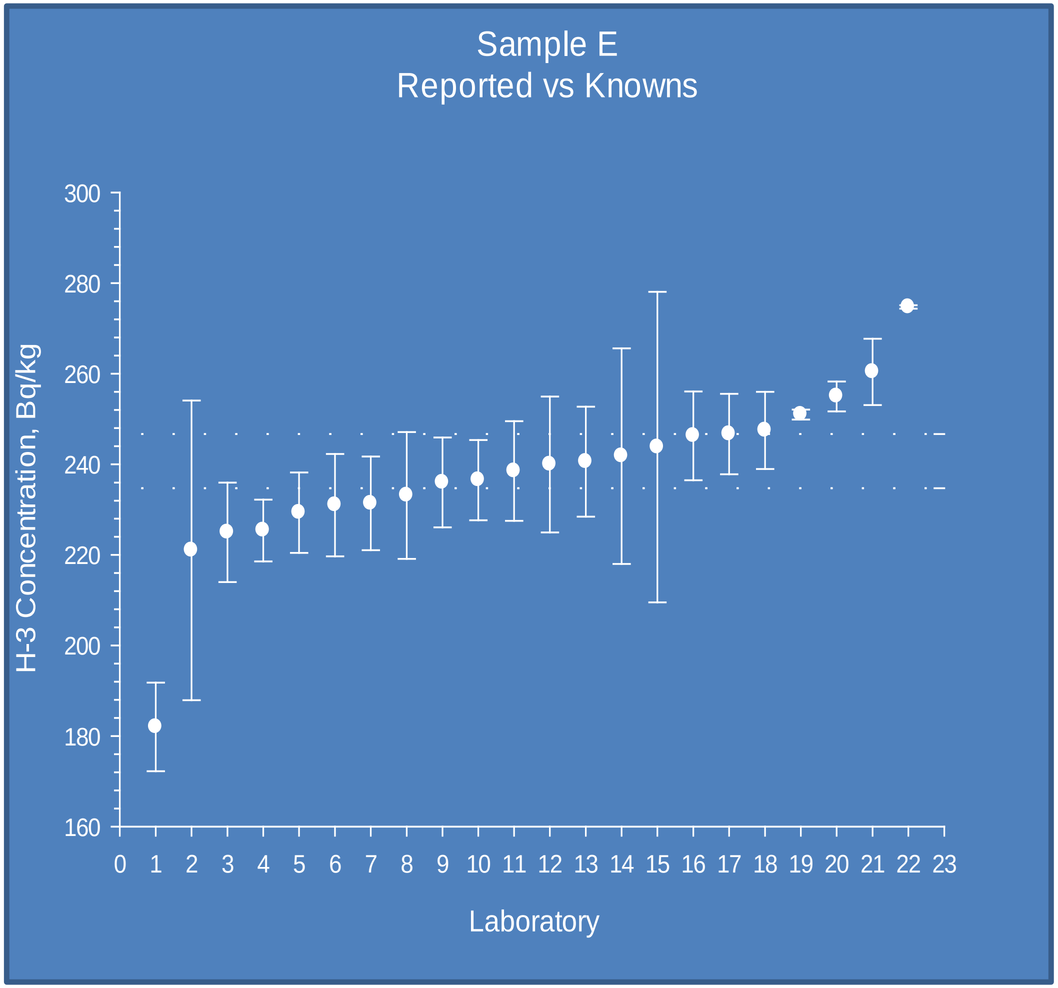
Spike and Test Sample Verification

Sample ID	Gravimetric, Bq/g	Measured, Bq/g	Diff, %
^{3}H Spike:	417.34 ± 3.00	419.07 ± 2.73	0.42

Test Samples:	Known, Bq/kg	Measured, Bq/kg	Diff, %
A23	724.61 ± 11.81	722.15 ± 13.72	-0.34
C23	729.01 ± 11.88	718.82 ± 17.54	-1.40
D23	364.37 ± 4.74	366.69 ± 12.39	0.64
E23	239.12 ± 3.35	241.45 ± 9.95	0.97

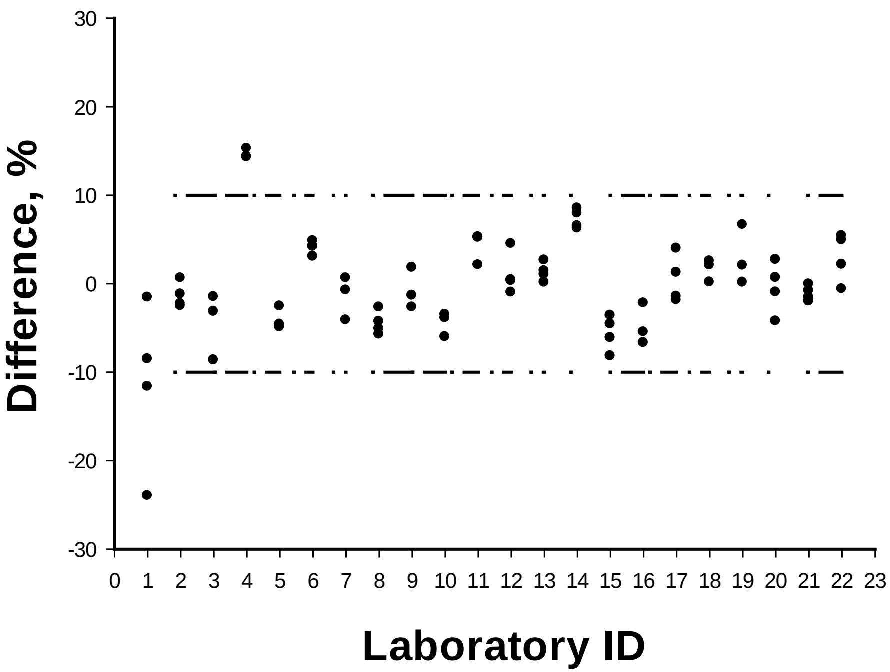
- For ^{3}H spike, the agreement between its gravimetric and measured values was found to be 0.42%, which demonstrated a traceability between the ^{3}H spike and its primary standard
- For a set of randomly selected test samples, the agreements between the known and measured values were found to be better than -1.4% and 0.97%, which verified that the sample spiking was performed correctly

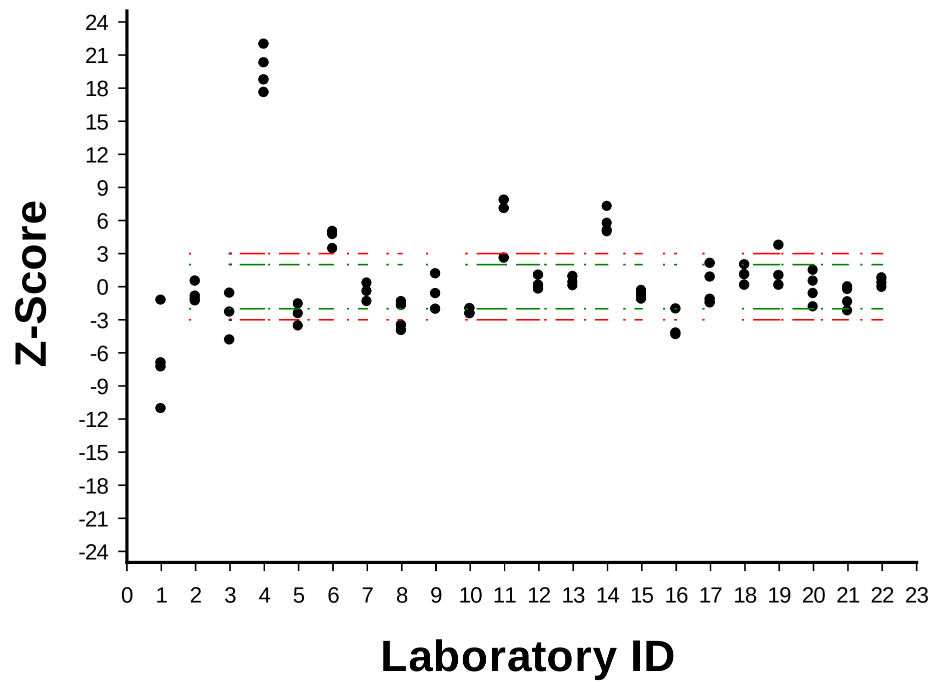
Comparison of ^{3}H Radioactivity Quantified at 1/3 of EPA/FDA Regulatory Limit



- As shown by the error bars, some participants over- or underestimated their measurement uncertainties indicating training needs on measurement uncertainty analysis
- Low or high bias was observed for some methods indicating problematic efficiency calibration and sample preparation
- Up to 50% difference was observed between the lowest and highest values reported by the participants indicating the needs on method improvement and harmonization

Method Performance





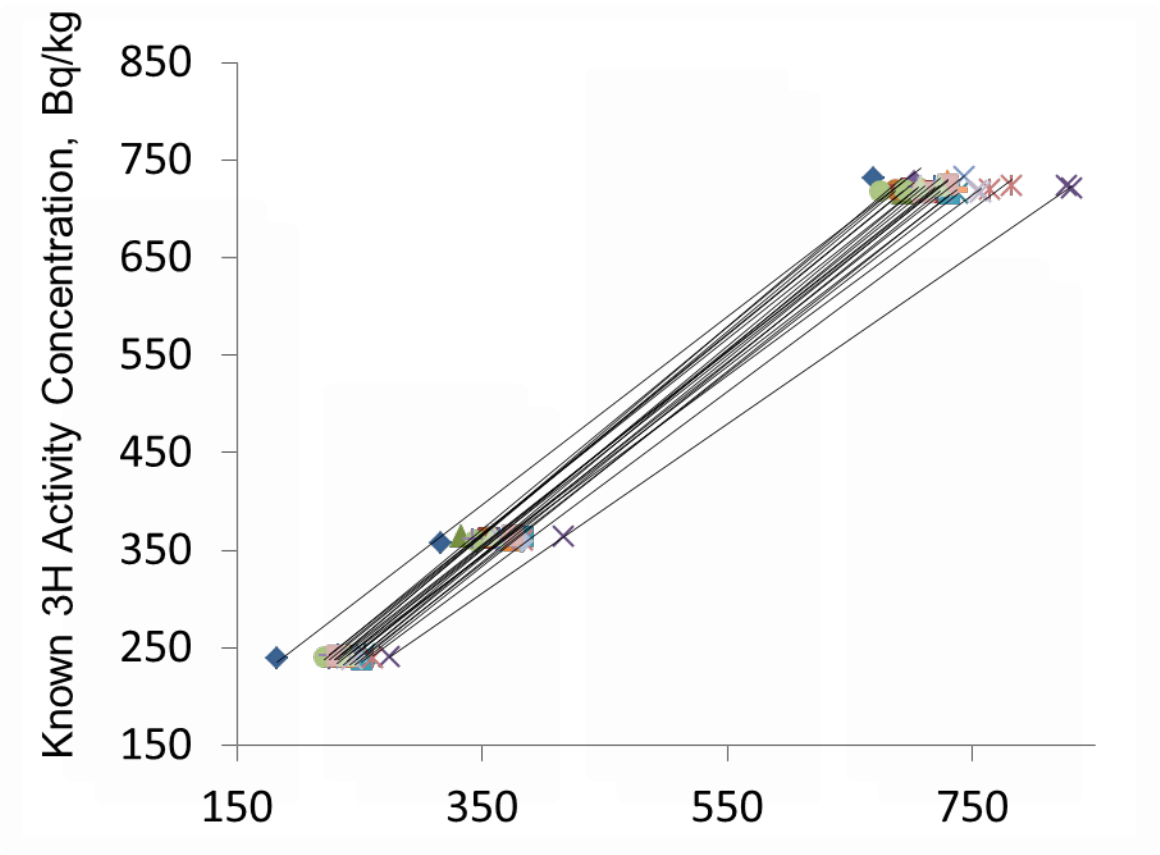
- Majority of the methods showed acceptable performance at all levels of ^3H activities tested
- Most of the results agreed with the knowns within ±10%, indicating their acceptability per the FDA data quality objective
- Comparing to ±10% acceptance criteria, more results were rejected by Z-score test due to inadequate estimates on measurement uncertainty

Method Detectability

Lab ID	MDA, Bq/kg	LOQ, Bq/kg	FDA DIL, Bq/kg
1	365.3	-	740
2	9.6	28.7	740
3	13.1	30.4	740
4	9.3	9.3	740
5	0.5	1.9	740
6	10.2	78.1	740
7	10.6	35.1	740
8	10.4	35.5	740
9	-	-	740
10	16.0	9.7	740
11	-	-	740
12	3.7	-	740
13	5.6	11.1	740
14	8.1	-	740
15	2.6	-	740
16	15.0	51.0	740
17	12.6	38.3	740
18	14.5	19.3	740
19	6.7	33.7	740
20	6.1	18.3	740
21	2.6	5.4	740
22	3.3	14.5	740

- All methods were able to detect and quantify ^{3}H significantly below FDA's DIL, indicating their abilities in meeting the required detection limit with smaller sample size and shorter count time
- All methods, except one, showed a potential for high throughput analysis of ^{3}H in bottled water

Method Linearity



Reported 3H Activity Concentration, Bq/kg

- All methods showed excellent linearity as indicated by their high R^2 values (0.993 – 1.00)
- Consistent results were observed between the methods that use and don't use quench correction

Conclusions

- All methods produce adequate detection limits for analyzing ^{3}H in bottled water
- Trainings on estimating measurement uncertainty per ISO guideline is needed for uniform data reporting and data acceptance evaluation
- All LSC counters were suitable for ^{3}H analysis
- No quench correction was needed with careful control of sample quench levels

List of Participants

Colorado Department of Public Health and Environment
Dr. Katherine A. Kelley Connecticut State Public Health Laboratory
Florida Department of Health Bureau of Radiation Control
Idaho State University Environmental Monitoring Laboratory
Illinois Emergency Management Agency
Indiana State Department of Health Laboratories
State Hygienic Laboratory at the University of Iowa
Kentucky State Public Health Laboratory
Maryland Department of Health and Mental Hygiene
Minnesota Department of Health
New Hampshire Department of Public Health-Radiochemistry Laboratory
New Jersey Department of Health, Public Health and Environmental Laboratories
NY Wadsworth Center- Biggs Laboratory
Commonwealth of Massachusetts Radiation Control Program, MA Environmental Radiation Laboratory
South Carolina Department of Health and Environmental Control Bureau of Environmental Services
Sandia National Laboratoires-Radiation Protection Semple Diagnostics
Tennessee Department of Health Nashville Division of Laboratory Services
Texas Department of State Health Services Laboratory
Vermont Department of Health Laboratory
Virginia Division of Consolidated Laboratory Services
State of Washington, Department of Health Public Health Laboratories
Wisconsin State Laboratory of Hygiene, University of WI-Madison
FDA Winchester Engineering & Analytical Center (WEAC)
Missouri State Public Health Laboratory
State of Maine Health and Environmental Testing Laboratory

Future Directions

- Homogenized Food Ash for alpha beta method development and intercomparison studies

FDA U.S. FOOD & DRUG ADMINISTRATION