Rapid Detection of Processed Uranium in Food

Council on Ionizing Radiation Measurements and Standards
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Gaithersburg, MD

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Goal

Develop a method to test food for uranium contamination in the aftermath of an incident involving processed uranium

- Method must be simple
- Able to be used by non-radiological laboratories
- No need for licensed material (spikes, tracers, etc.)
- Fast/easy sample preparation
  - Applicable for emergency response
  - High throughput

Uranium

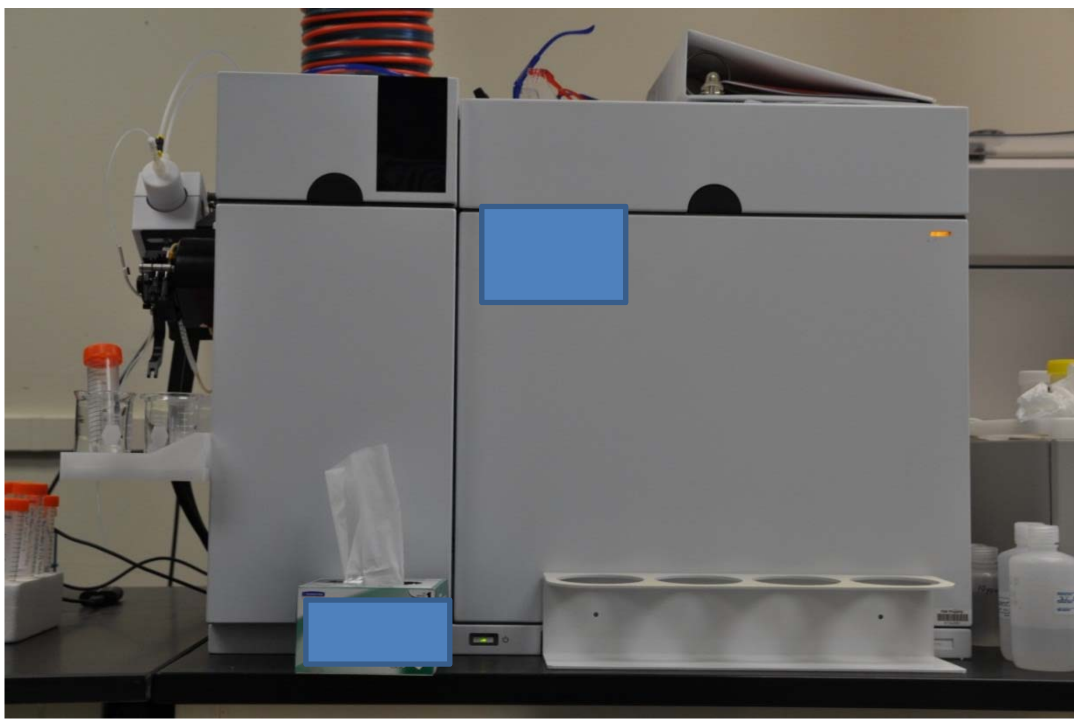
- ^{238}U ~ 99.27 %
- ^{235}U ~ 0.7198 %
- ^{234}U ~ 0.0050 %
- $^{238}U/^{235}U$ natural ratio ~ 138
- ^{235}U enrichment 3-5 % for power and ~ 90 % for military applications

Radiochemical Methods

- Traditional radiochemistry methods are employed to measure alpha, beta, and gamma emitting radionuclides
  - Alpha and beta emitters are more challenging to measure
  - Often must separate elements and isotope

ICP-MS

- Technique is very common in analytical labs
- Relatively simple sample prep



Traditional ICP-MS Methods for Uranium

- Often used for geochemistry
  - Extremely detailed studies
  - Very little error
  - Thermal-ionization mass spectrometry (TIMS) and multiple-collector ICP Mass Spectrometry
- Used by Health Physicists and for bioassay
  - "Dilute and shoot" methods developed by CDC
  - Sector Field-Mass Spectrometry (SF-ICP-MS)
- Quadrupole ICP-MS
  - cheaper instrument, but widely used
  - Organic material must be removed

Parrish, R.R.; Thirlwall, M.F.; Pickford, C.; Horstwood, M.; Gerdes, A.; Anderson, J.; Coggon, D. *Health Phys*. **2006**, *90*, 127-138.

Xiao G.; Jones, R.L.; Saunders, D.; Caldwell, K.L. *Radiat. Prot. Dosimetry* **2014**, *162*, 618-624, and references cited

Typical Quadrupole ICP-MS Procedures for Uranium

– Dry food overnight then weigh into container

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– Digest with 8 mL Aqua Regia and 2 mL of hydrofluoric acid (HF)
  - Extremely corrosive mixture
  - Can not be stored (made then destroyed after each use)

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- Digest with 8 mL Aqua Regia and 2 mL of hydrofluoric acid (HF)
  - Extremely corrosive mixture
  - Can not be stored (made then destroyed after each use)
- Microwave heating
- Cool, quench HF with aqueous Boric Acid
- Microwave heating
- Cool, inject into ICP-MS
- Excellent results but suboptimal for emergency response

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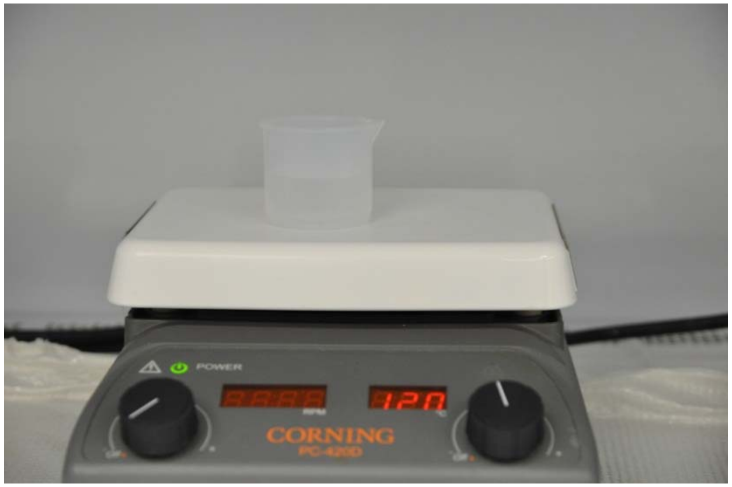
Proof of Principle

- Reference materials measured on multiple days
- Known ratios for $^{238}U/^{235}U$:
  - CRM U500 1.0003
  - CRM U970 0.0054
  - SRM4321C 137.82

	Average	Standard Deviation	Observed bias %
CRM U500 (N = 5)	0.9998	0.0024	-0.05
CRM U970 (N = 5)	0.0053	0.0001	-1.0
SRM4321C (N = 5)	138.87	1.55	0.76

Method

- 5 g food in Teflon Beaker
- Acid leach with 10 mL high purity nitric acid
- Heat



Can We “Dilute and Shoot”

- No!
- Error up to 60 % for pome fruits and stone fruit
  - Residual organic content
- OK for water (~ 0.06 % error)

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- Filter through plug DGA resin plug

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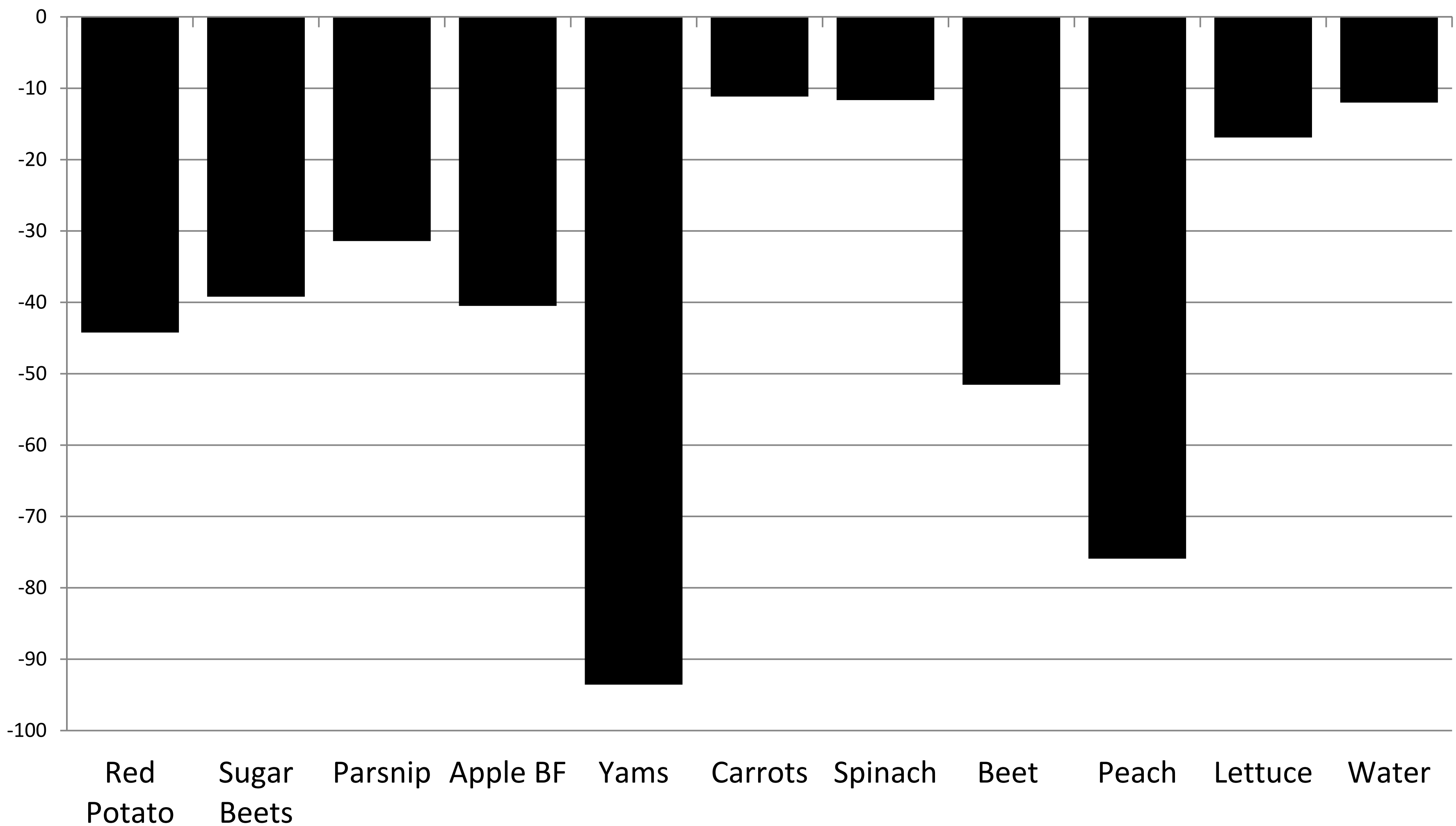
- Filter through plug DGA resin plug
  - Elute with concentrated nitric acid first
  - Strip column with 5 % nitric acid
- Collect and inject into ICP-MS

Intended use

- Fresh fruits, vegetables, dairy
- 25 different types of fruit and vegetables were analyzed in triplicate. Maximum deviation from known ratio: ± 3 %
  - Required no spikes, tracers, or correction
  - Any laboratory with ICP-MS capability could perform this analysis
- How sensitive is this measurement for the detection of ^{235}U?

% Change in $^{238}U/^{235}U$ Ratio with 1.2 picogram spike in 5 g Food



Limitations

- Cereal grains: E.g. Wheat, rice, breakfast cereal, pasta
- Fats from food of animal origin: E.g. Butter, lard, oil
- High Starch and/or protein: E.g. lentils, beans
- High oil content: E.g. Tree nuts, Peanut butter, tahini
- High sugar, low water: E.g. raisins, dried apricots

This method is for use during emergency. Fresh foods are more germane.

Conclusions

- Developed a quadrupole ICP-MS method capable of interrogating uranium fingerprint
- Method not limited to radiochemical laboratories
- Simple, user friendly
- Potential automation with in line HPLC/Ion Chromatography

Acknowledgements

- WEAC laboratory members
- CFSAN collaborator
- Audience for listening