Surrogate Post-Detonation Urban Debris (SPUD):

Standard Reference Materials 4600 & 4601

ICRM-LLRMT
September 26-30, 2016

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Outline

- Nuclear Forensics (NF) and Attribution (A)
- Issues with CRMs/SRMs for Post-Det Nuclear Forensics
- SPUD - Nuclear Forensics Post-Detonation SRMs
  - What is it?
  - How would they work?
  - Purpose?
- SRM Certification Status (focus - Homogeneity)
- Next Steps

Nuclear Forensics (NF) & Attribution (A)

- **(NF) Nuclear forensics** is the use of physical and chemical properties to help determine the age, origin, and history of nuclear materials and the design of nuclear devices.

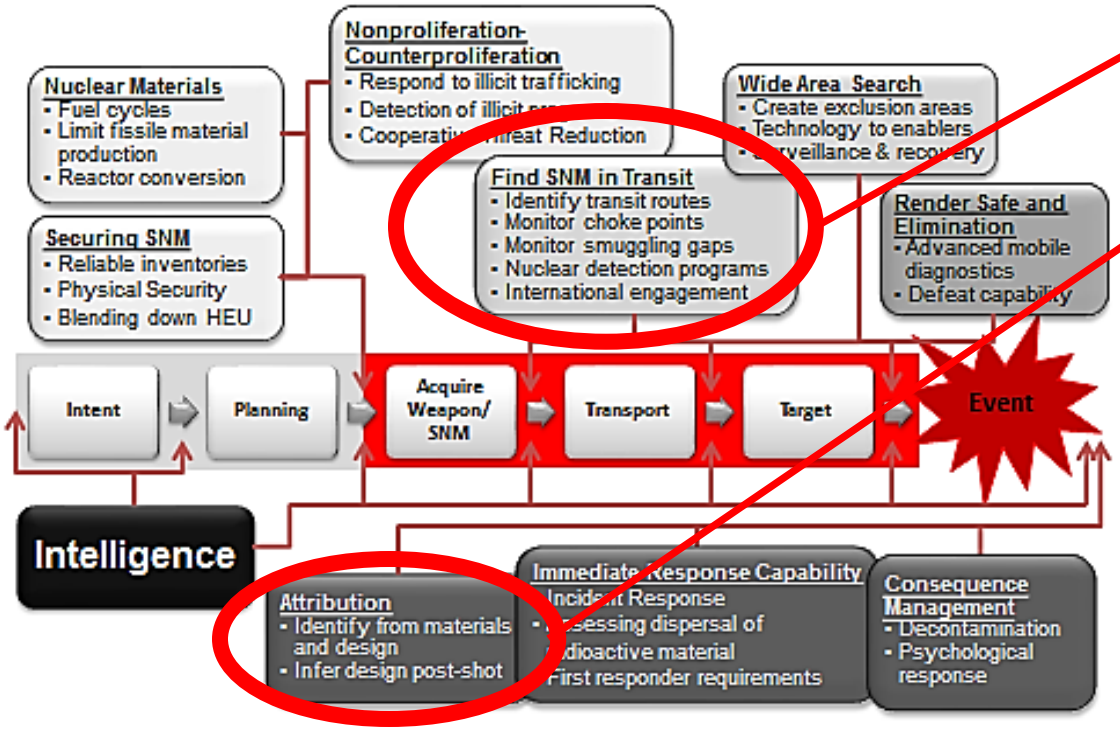
- **Attribution (A)** integrates all information, including nuclear forensic measurement data, law enforcement and intelligence data, to suggest or exclude the origin of nuclear materials and devices, routes of transit, and responsible groups or individuals.

Information that can be obtained from nuclear* material
*uranium (U), plutonium (Pu)

Parameter	Information	Analytical technique
Appearance	Material type (e.g., powder, pellet)	Optical microscopy
Dimensions (pellet)	Reactor type	Database
U, Pu content	Chemical composition	Titration, HKED, IDMS
Isotopic composition	Enrichment → intended use; reactor type	HRGS, TIMS, ICP-MS, SIMS
Impurities	Production process; geolocation	ICP-MS, GDMS
Age	Production date	AS, TIMS, ICP-MS
$^{18}O/^{16}O$ ratio	Geolocation	TIMS, SIMS
Surface roughness	Production plant	Profilometry
Microstructure	Production process	SEM, TEM

HKED (hybrid K-edge densitometry), IDMS (isotope dilution mass spectrometry), HRGS (high-resolution gamma spectrometry), TIMS (thermal ionization mass spectrometry), ICP-MS (inductively coupled plasma mass spectrometry), SIMS (secondary ion mass spectrometry), GDMS (glow discharge mass spectrometry), AS (alpha spectrometry), SEM (scanning electron microscopy), TEM (transmission electron microscopy)

Issues with CRMs/SRMs for Nuclear Forensics



Pre-Det Nuclear Forensics

Post-Det Nuclear Forensics

- **Short Supply**
- **Exceedingly Rare**
- **Long Development Time**
- **Very Expensive**

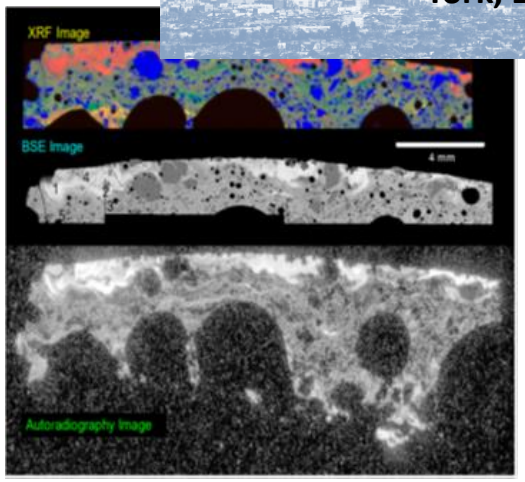
REQUIRED FOR LEGAL DEFENSIBILITY

(method development, validation, accuracy and precision, traceability)

Post-Detonation Nuclear Forensics CRMs/SRMs

"What will happen when the explosions come—when a part of New York or Cairo or Adelaide has been hollowed out by a device in the kiloton range? Since even a so called fizzle yield could kill a number of thousands of people, how many nuclear detonations can the world tolerate?"
—John McPhee, *The Curve of Binding Energy* (Farrar, Straus and Giroux, New York, 1974).

<u>ISSUE:</u> No fresh fission product post-detonation urban RM that exists to test and validate nuclear forensics measurements.



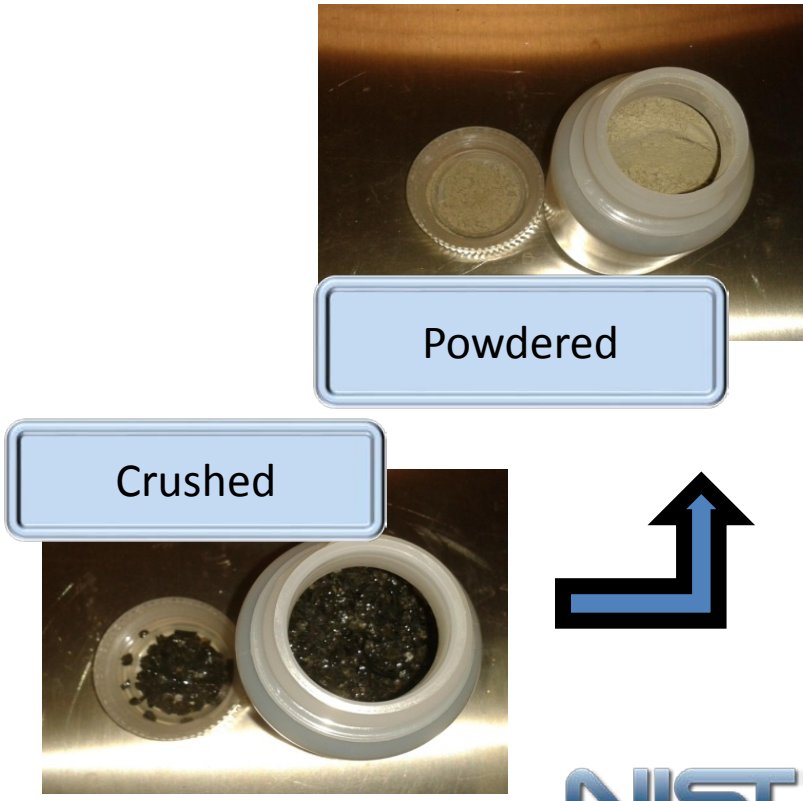
What is it?

Surrogate urban debris material that mimics “rubble” of a blown up city after a IND event

- vitrified mixture (cement, concrete, steel) material (CaAlFeSi)
  1) Batch 1 - doped with natural U (blank) – **SRM 4600**
  2) Batch 2 - doped with HEU (U235-22%) – **SRM 4601**
- 100 units of 25 g each

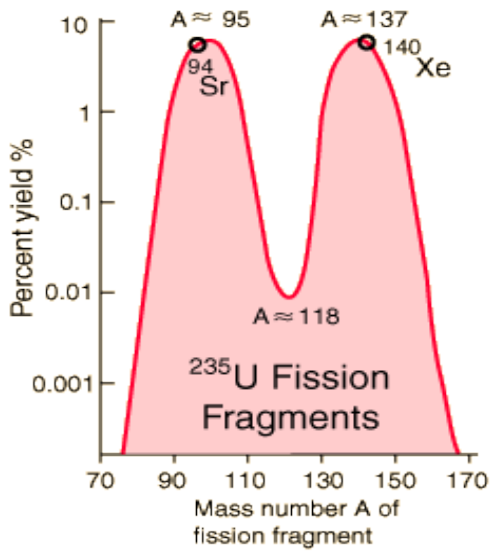
- glass was prepared in several batches with all the reagents mixed prior to producing the batches so that all batches were produced from the same mix.

- reagents were mixed using a ‘cone and quartering’ approach followed by grinding (gyro mill) and re-mixing (rotary mixer), to produce a homogeneous product.



How Would It Work?

The SPUD SRM contains enriched U in a complex matrix of TE, that upon irradiation would produce both fission and activation products.



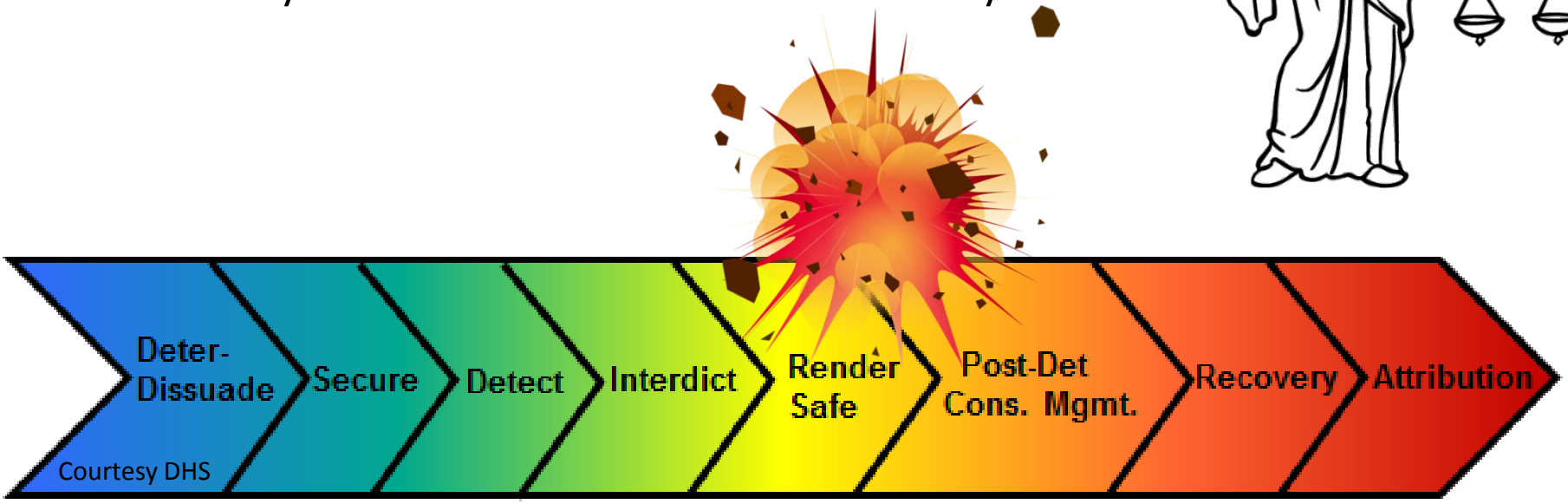
	SRM 4600	SRM 4601
element	µg/g	µg/g
Aluminium	51800	55690
Arsenic	20.3	18.7
Calcium	142000	144900
Chromium	575	500
Cobalt	45.7	46.24
Copper	240.8	262.7
Iron	195500	188400
Magnesium	3350	3160
Manganese	1812	1874
Molybdenum	263	261
Nickel	360	611
Niobium	77	78.5
Phosphorus	331	349
Potassium	2610	2718
Silicon	168800	172800
Sodium	749	734
Strontium	295	308.3
Tantalum	40.3	43.3
Thorium	16.38	16.16
Tin	29.4	28.13
Titanium	1205	1248
Tungsten	34	19.6
Uranium	62.61	81.072
Vanadium	130.8	141.3
Zinc	14	12.2
Zirconium	53.5	56.8

- start with a known starting material (known chemical concentrations and isotope composition)
- Expose the SRM material to known neutron energy & fluence
- “Known” activation and fission product yields can be predicted as they are directly related to the energy and flux

Purpose?

To test nuclear forensic measurement capability by providing a surrogate post-detonation material to enable:

- method development and validation,
- testing and readiness,
- and enable laboratories to legally and scientifically demonstrate the efficacy, accuracy, and precision of analysis methods and establish traceability.

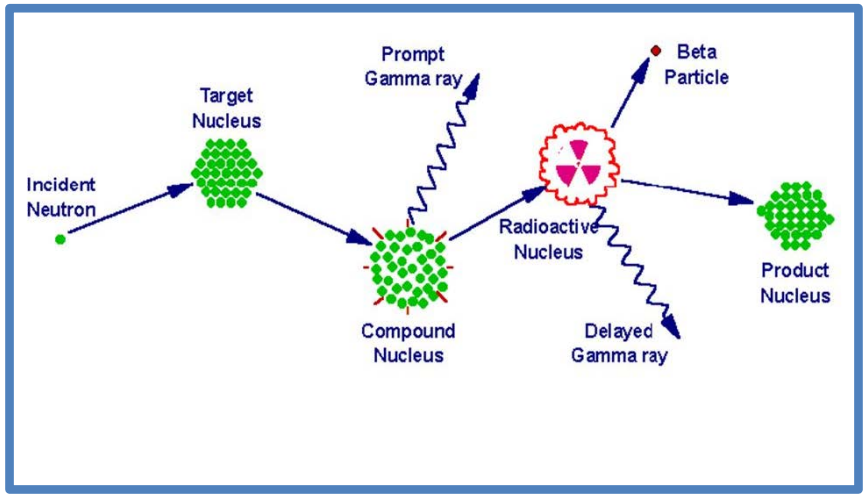


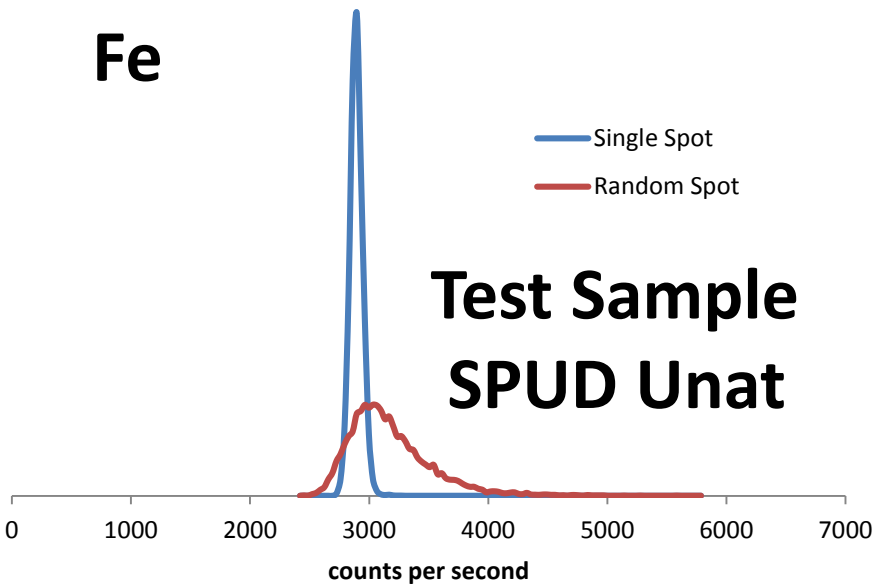
Nuclear Defense Spectrum

Homogeneity Assessment

Methods:

INAA/PGAA Analysis (NIST)
µXRF Analysis (FBI & AFIT)





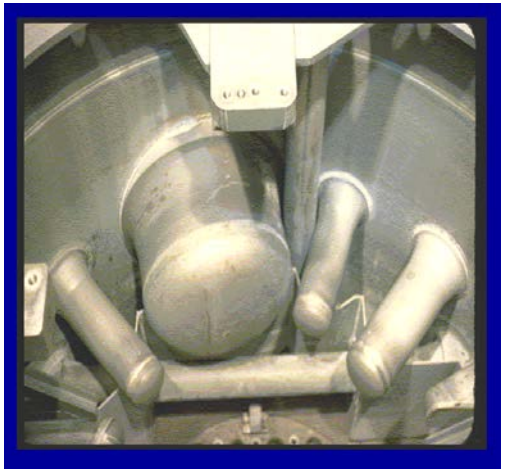
10K points

Data from AFIT

INAA/PGAA Analysis

Irradiation Conditions

- Sample size ~ **250 mg**
- Irradiated for 1 h in NIST 20 MW Neutron Beam Split-Core Reactor (NBSR)
- Neutron fluence rate of approximately (3×10^{13} $n^{-2} \cdot s^{-1}$), thermal energies
- Irradiated at two day intervals



Interior of NBSR reactor, courtesy of Robert Williams, NIST

Quantitative Evaluation of Homogeneity

	SRM 4600 (Unat)			SRM 4601 (U235)		
Element	τ (between bottle STD)	σ (within Bottle STD)	P-value	τ (between bottle STD)	σ (within Bottle STD)	P-value
As	0.075	0.177	0.42	**0.346**	**0.224**	**0.0167**
Co	0.303	0.478	0.41	**0.313**	**0.315**	**0.078**
Cr	**12.65**	**10.91**	**0.062**	6.549	5.959	0.122
Fe	0.158	0.189	0.10	**0.148**	**0.070**	**0.003**
Mo	1.89	2.76	0.42	--	--	--
Ta	0.158	0.322	0.41	0.127	0.176	0.109
Th	0.224	0.360	0.42	0.083	0.319	0.415
W	0.089	0.476	0.42	0.109	0.410	0.419
U	0.314	0.741	0.42	**1.379**	**1.222**	**0.075**
U238	--	--	--	**1.094**	**1.017**	**0.095**
U235	--	--	--	**0.300**	**0.227**	**0.030**

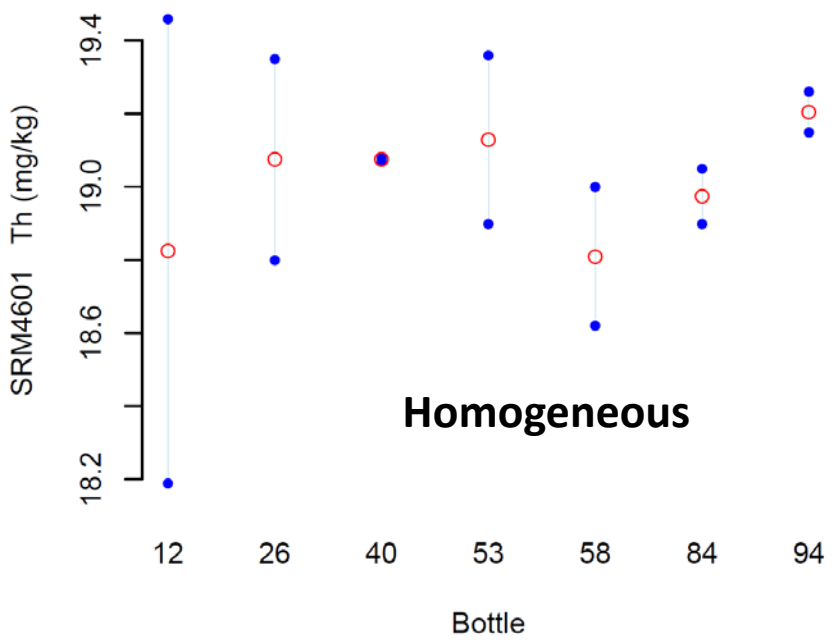
Likely heterogeneous

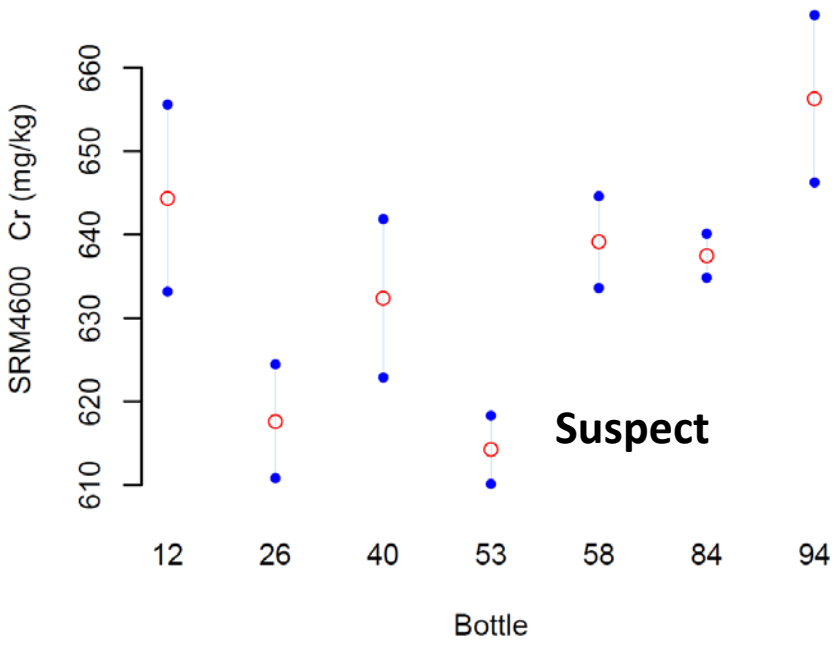
Maybe heterogeneous

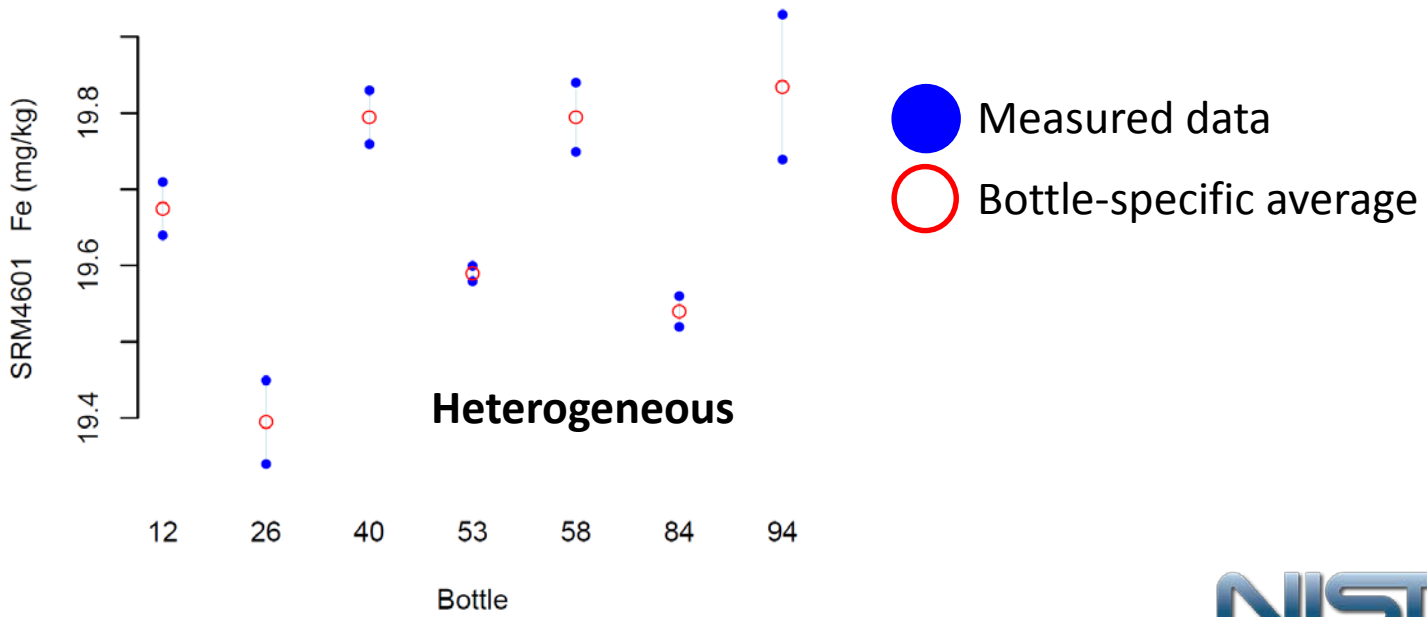
- Closer tau is to zero the more likely homogeneous
- Small p-values suggest significant heterogeneity

*** p-value, in this case, statistical test to decide whether tau is significantly greater than zero or significantly larger than sigma

*** tau is the "extra" component of uncertainty above and beyond what the variability of the replicates (within each bottle) suggests


SRM4601 Th (mg/kg)
18.2
18.6
19.0
19.4
Homogeneous
12
26
40
53
58
84
94
Bottle


SRM4600 Cr (mg/kg)
610
620
630
640
650
660
Suspect
12
26
40
53
58
84
94
Bottle


SRM4601 Fe (mg/kg)
19.4
19.6
19.8
Heterogeneous
12
26
40
53
58
84
94
Bottle
Measured data
Bottle-specific average

Take Aways

- Overall it appears that of the elements measured it seems that SRM 4600 tends to be less heterogeneous then SRM 4601 (for 250 mg sample sizes).
- Only Cr seems to be suspect for SRM 4600.
- As, Fe, and U are definitely suspect for SRM 4601 and Co is also suspicious.

**** Being there are only 2 replicates per bottle the results must be taken with a sense of skepticism, although the statistical test does take into account (to some extent), these limited number of samples.

µXRF Analysis

Goals (µXRF):

- **To determine the minimum mass of sample that can be used with confidence that nugget effects are not contributing to the measurement uncertainty.**

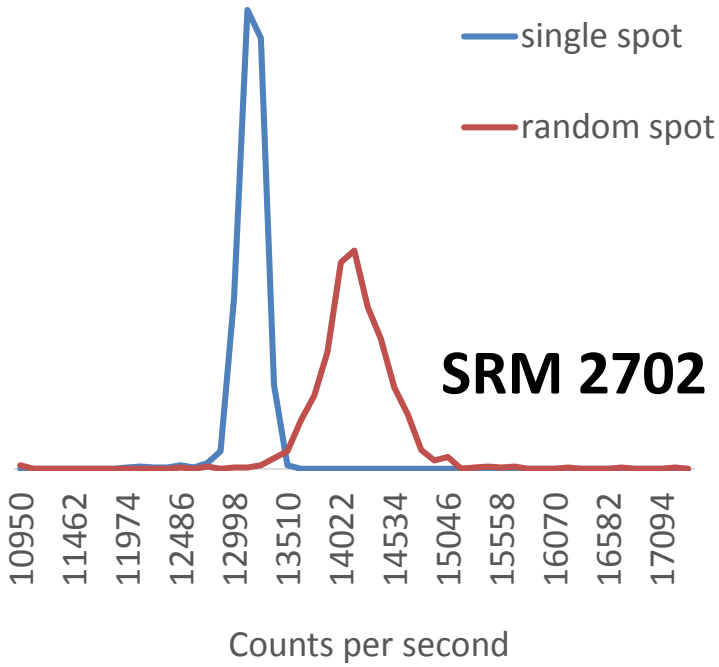
Analysis Methodology

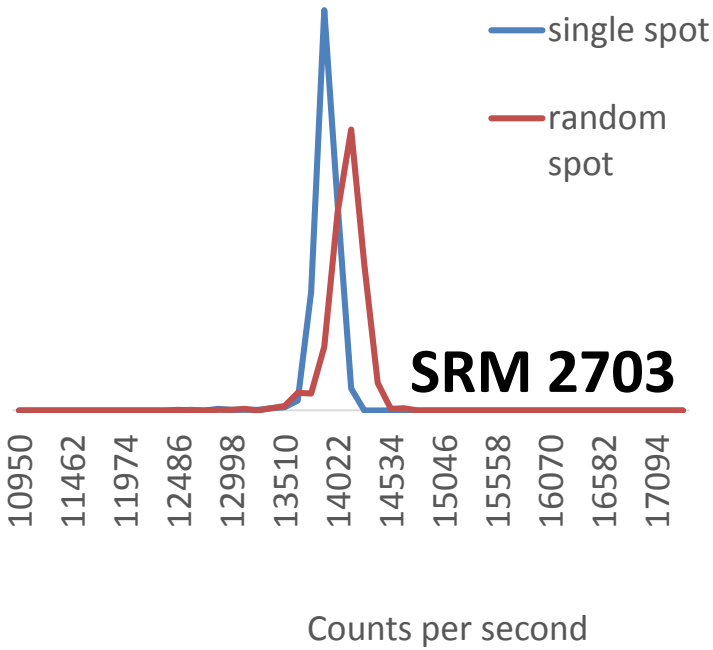
Single location/spot many times

- 5 s analysis, 10000 times, 1 location
- Variation due to instrument and measurement sources

Many locations/spots a single time

- 5 s analysis, 10000 times, 10000 locations
- Variation due to sample, instrument and measurement sources

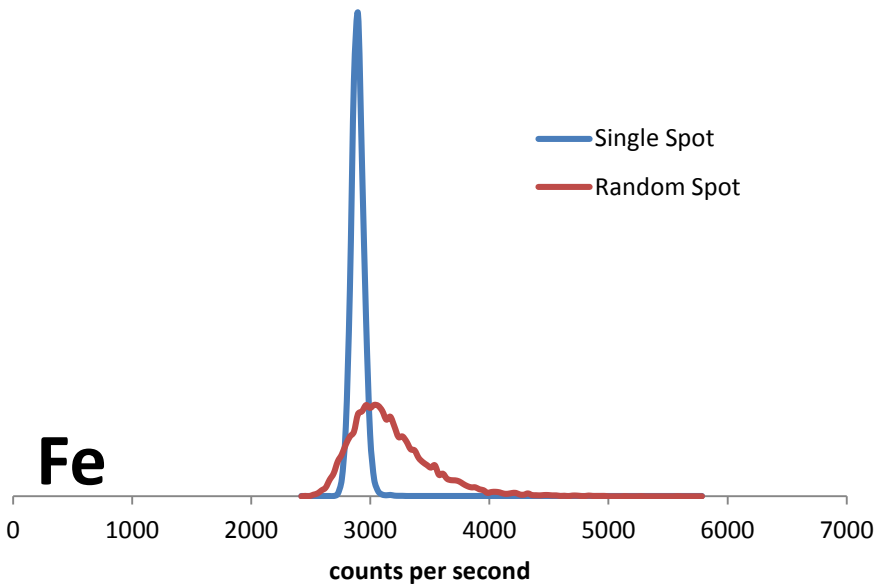




Courtesy of FBI

SRM 4600 (Unat)

Test Sample

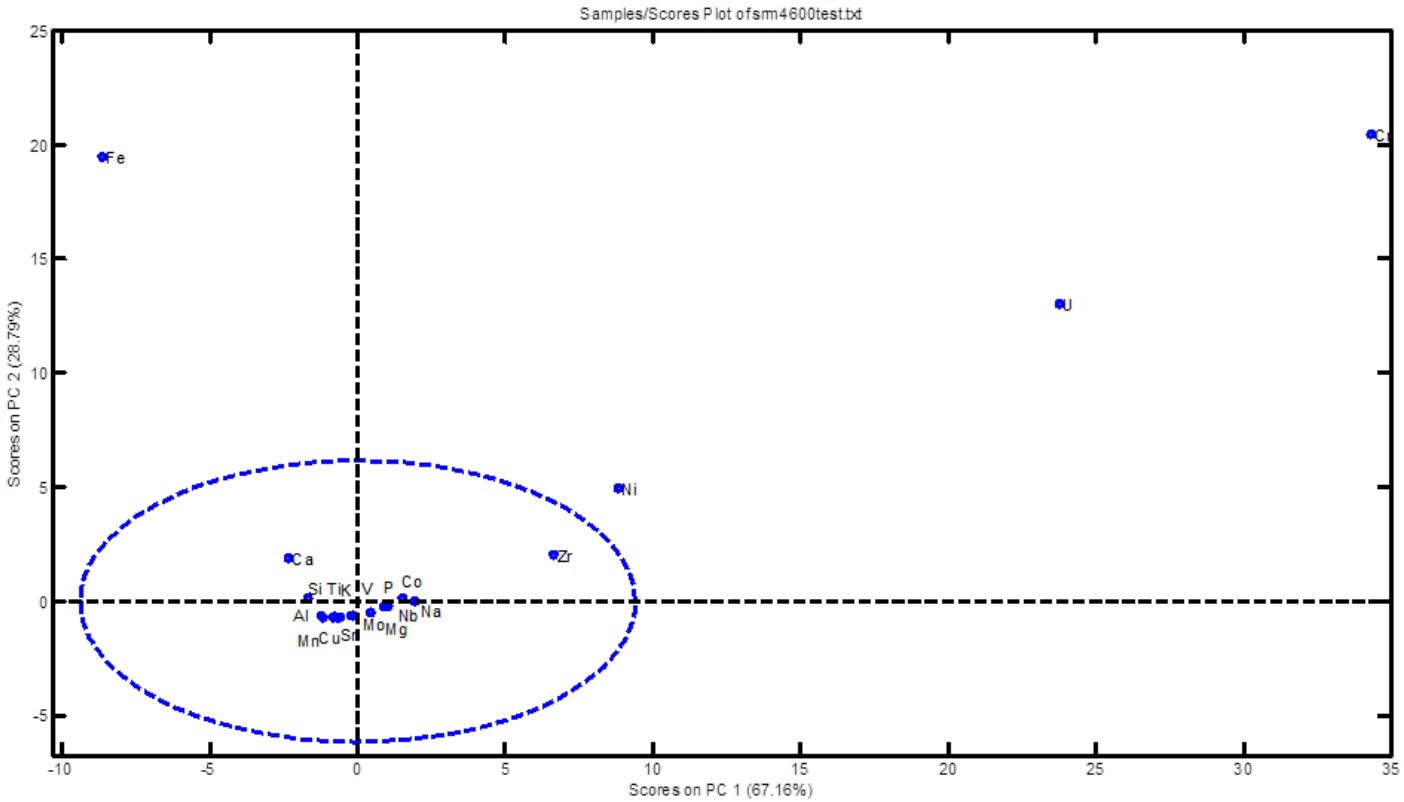


10K points

Data from AFIT

SRM 4600 (Unat)

Test Sample – PCA ANALYSIS

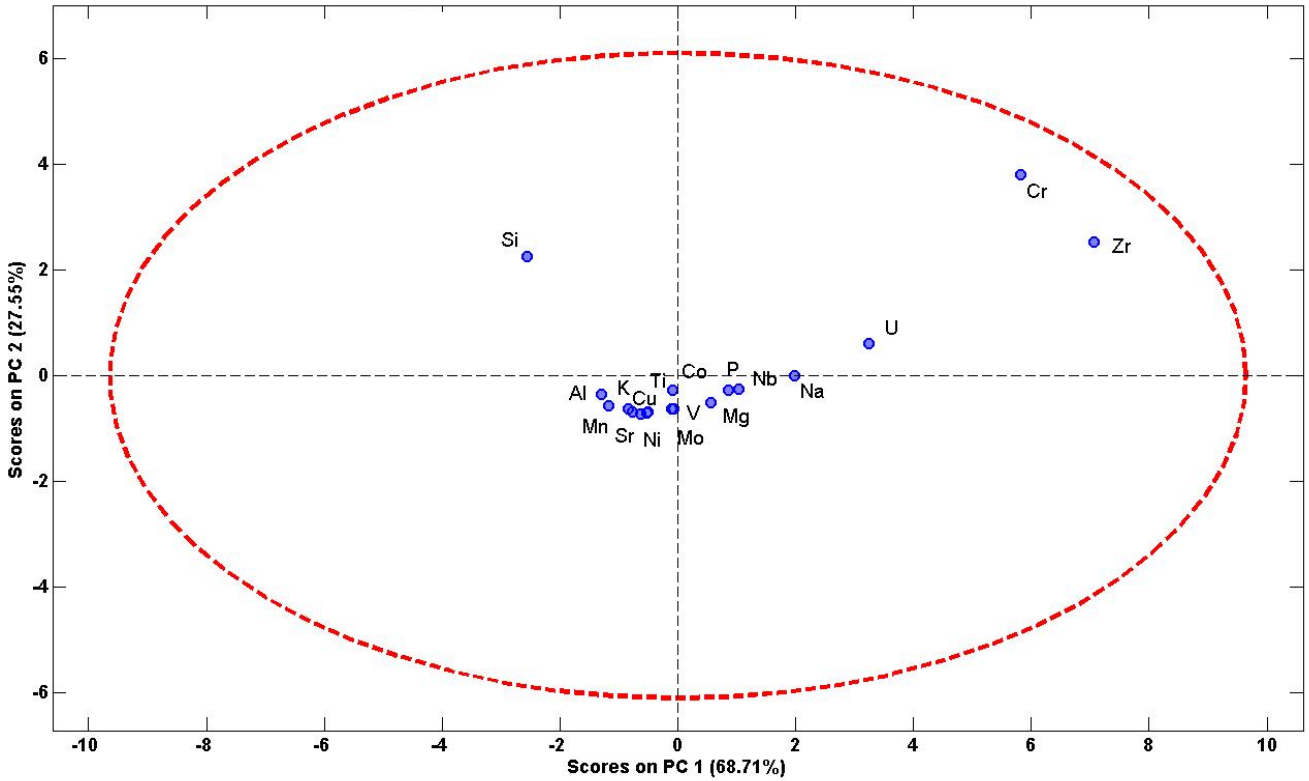


- Sample size interrogated is roughly 0.5 mg (much less than INAA analysis)
- Initial PCA finds that many elements are homogeneous (or too low to detect (e.g. Na).
- Elements outside circle are heterogeneous and have detectable nugget effects.
- Blue circle denotes 3 standard deviations from the center of the PCA model built from data originating from the single spot (know homogeneous).

NIST Figure

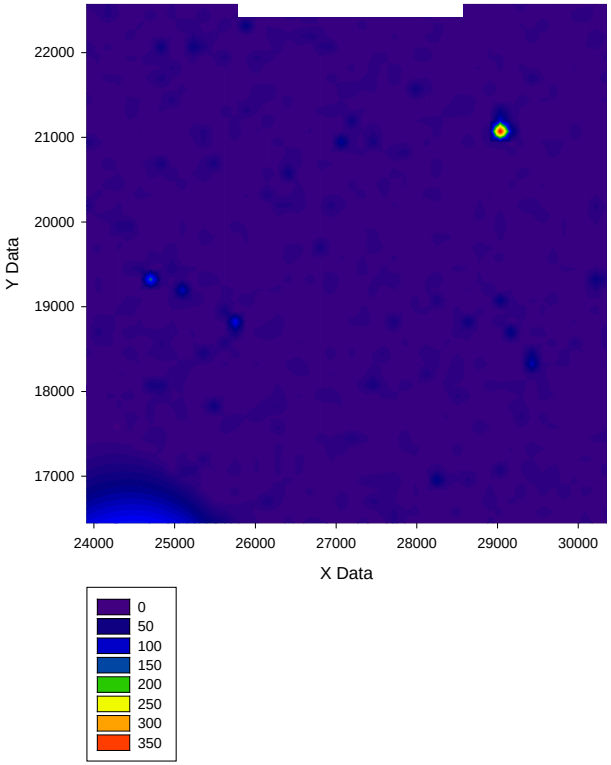
SRM 4600 (Unat)

Test Sample – PCA ANALYSIS

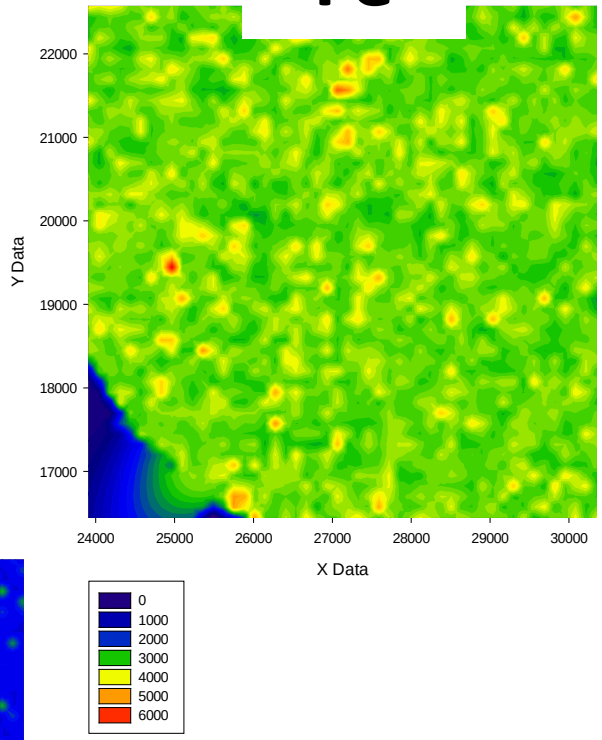


- Simulation of a larger spot size (averaging data from adjacent points in both the 'stability run' and the 'random run' and running this through the PCA model until no elements fall outside the 3s confidence ellipse) allows for the determination of a minimal sample mass needed for all the elements to be homogeneous.
- 5000 spectra were averaged together for all the elements to fall within the 3s confidence ellipse
- Indicating sample mass of roughly 3 mg required for all the elements to be homogeneous

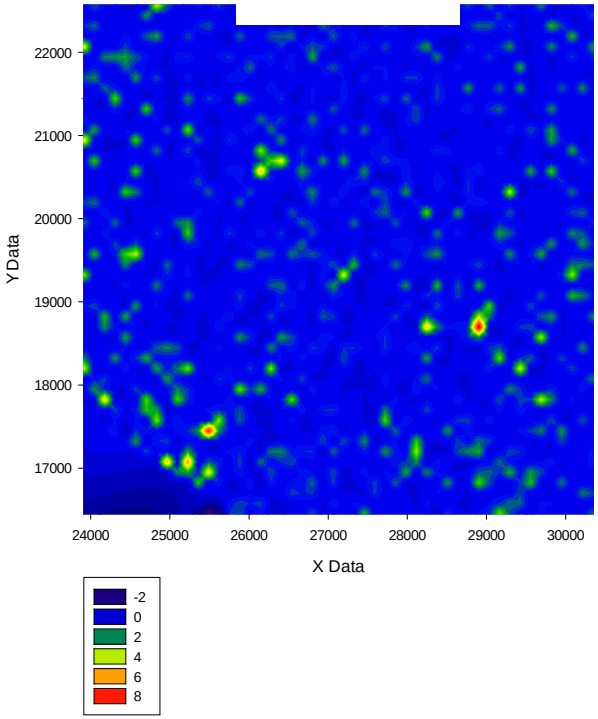
Cr


22000
21000
20000
19000
18000
17000
Y Data
24000
25000
26000
27000
28000
29000
30000
X Data
0
50
100
150
200
250
300
350

Fe


22000
21000
20000
19000
18000
17000
Y Data
24000
25000
26000
27000
28000
29000
30000
X Data
0
1000
2000
3000
4000
5000
6000

U


22000
21000
20000
19000
18000
17000
Y Data
24000
25000
26000
27000
28000
29000
30000
X Data
-2
0
2
4
6
8

Take Aways

- Preliminary PCA analysis of SRM 4600 suggests that Cr, Fe, Ni, and U maybe heterogeneous (for sample size of roughly 0.5 mg).
- Neutron Activation analysis found only Cr to be suspect, however the sample size in this case was 250 mg versus 50 mg sample size of the microXRF.
- The data seem to suggest that Cr is heterogeneous for SRM 4600 at almost any sample size and that for smaller sample sizes (< 250 mg) Fe, Ni and U may also be heterogeneous.
- HOWEVER, thus far only one sample has been run through PCA analysis so the suggestion that only 3 mg is required for all the elements to be homogeneous should be taken with a grain of salt.

Future Work

- Complete remainder of INAA/PGAA analysis (specifically short –lived)
- Complete remainder of XRF analyses – AFIT and FBI
- Complete PCA analysis - NIST
- Commence elemental and isotopic analysis and certification