



National Standards for Radioactivity Measurements

Historical Overview & Current Status

R. Collé

*Ionizing Radiation Division
Physics Laboratory*

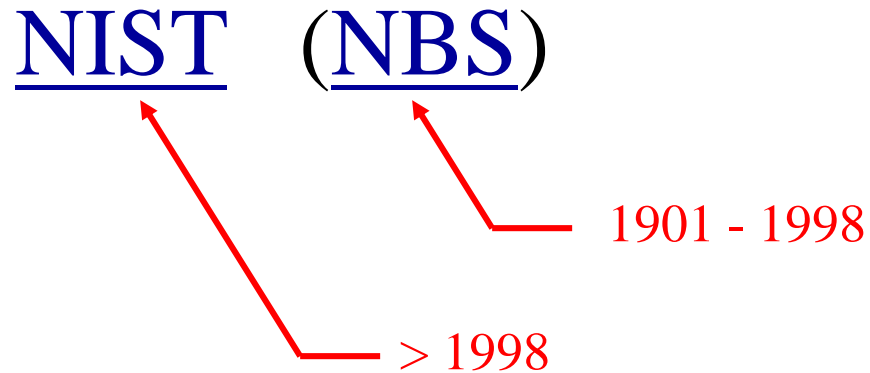
*National Institute of Standards and Technology
Gaithersburg, MD 20899 USA*



ANS / ENS International Meeting
American Nuclear Society
European Nuclear society

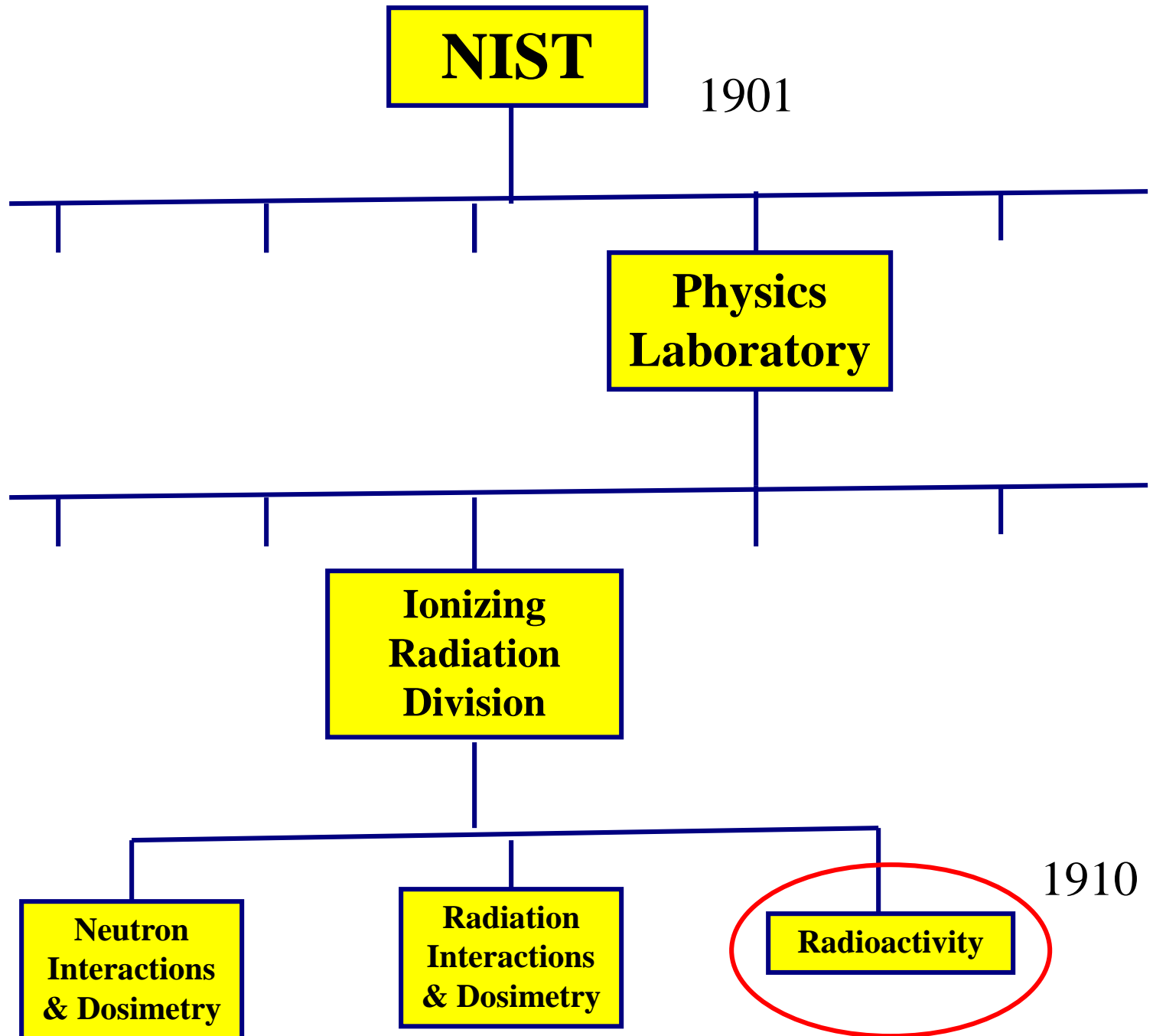
15 November 2007
Washington, DC





Highest authority in USA

- for setting physical measurement standards
- and ensuring accurate measurements



OUR ROLES

primary objectives

- ➡ develop and maintain national standards through primary calibrations
- ➡ develop secondary standards and transfer standards;
- ➡ disseminate standards;

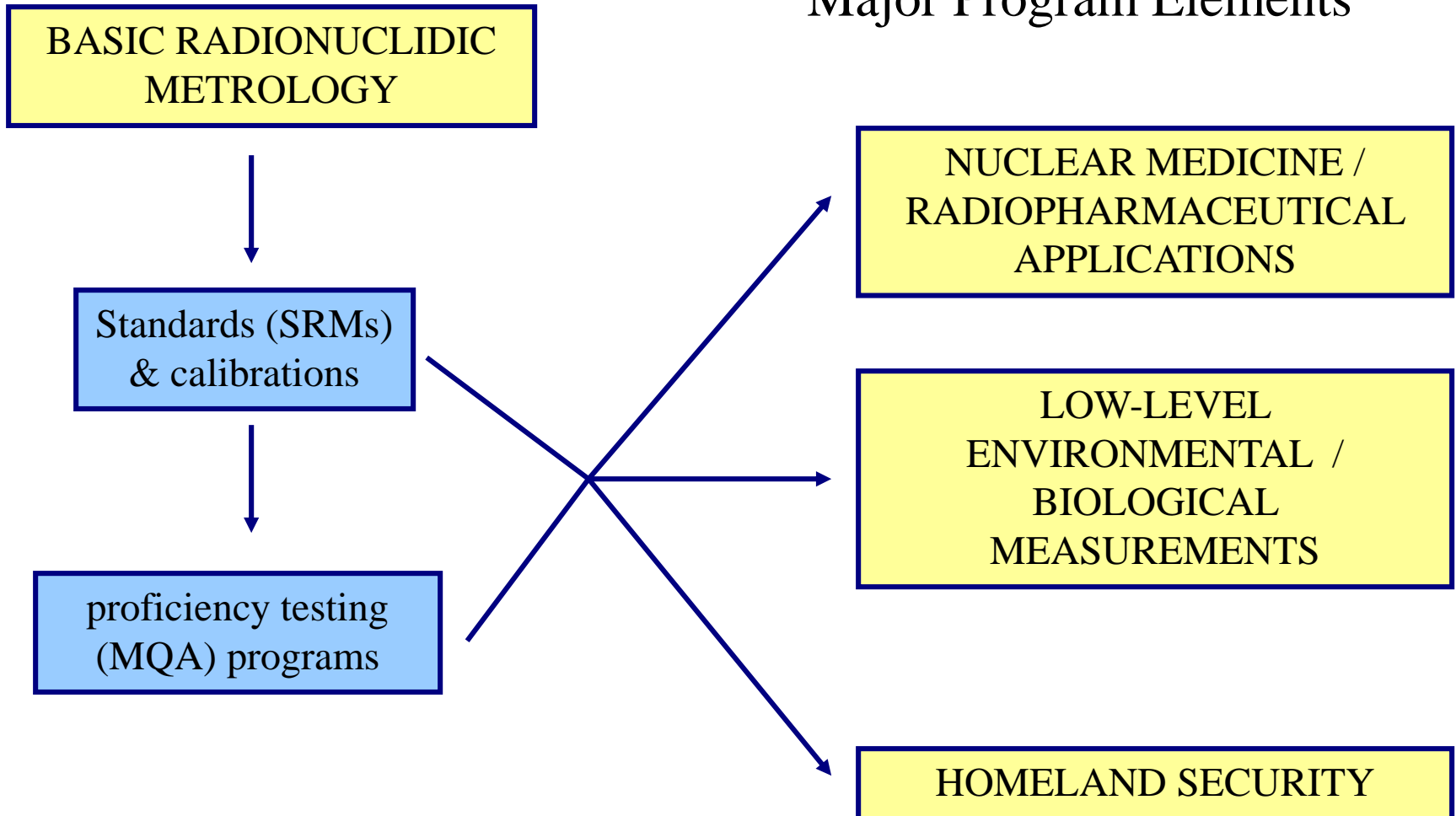
and *provide mechanisms* for insuring the quality of measurements

principal elements of what we do

- ➡ basic & applied research
 - includes technology & method development
- ➡ standards (SRMs) and calibrations
- ➡ proficiency testing (MQA)
- ➡ international intercomparisons & collaborations

Radioactivity Group

Major Program Elements



Some of our ongoing activities

Radionuclide Metrology

- Microcalorimetry
- Coincidence & anticoincidence counting methods
- Liquid-scintillation methods: CIEMAT/NIST & TDCR
- LS cocktail composition effects
- Mass spectrometric methods: RIMS & TIMS

Nuclear Medicine/Radiopharmaceutical Standards

- Diagnostic radionuclides (^{18}F , ^{76}Br , $^{68}\text{Ge}/^{68}\text{Ga}$)
- Therapeutic radionuclides (^{211}At , ^{223}Ra , ^{177}Lu , ^{90}Y)
- Brachytherapy sources (calibrations)
- **SRM** production & distribution
- MQA programs (NEI)

Environmental Radioactivity

- Chemical speciation
- Traceability programs DOE, EPA, NRIP
- **Natural matrix SRM** production & certification
- Mass spectrometric methods

Homeland Security

- Cargo container inspection station
- Portal monitors
- Protocols for hand-held detector calibrations
- Radionuclidic forensic methods

Metrology

- ⇒ 130+ nuclides
- ⇒ Many geometries
- ⇒ > 30 systems

Standards

- ⇒ 60+ nuclides
- ⇒ 9 natural matrix (multi-nuclide)
- ⇒ 500 – 1000 units per year
- ⇒ by over 200 users
- ⇒ \$ 300K - \$ 600K USD in sales per year (depends on availability)
- ⇒ 20 users buy 10 or more SRMs per year

Calibrations

- ⇒ > 100 routine per year
- ⇒ 20 – 30 nuclides; many geometries
- ⇒ \$ 100K – 200K USD per year
- ⇒ few special \$ 30 - \$ 100K USD **EACH**

MQA programs

- ⇒ 17 unique
- ⇒ from monthly to 2x per year

Our measurement methods and instruments include:

- Basic (primary) standardizations
- Secondary standardization methods
- Routine measurements
(e.g., for monitoring, QC, etc.)
- Ancillary measurements
(e.g., for impurity analyses, research, etc.)

high-pressure re-entrant ion chamber

our SRM work horse -- calibration factors for 40+ nuclides for 30+ years

LS counters (Beckam, Wallac, Packard) – our other SRM work horses!

3-phototube TDCR system (under development)

isothermal microcalorimeter (a low-temp 8K calorimeter is inactive)

high-resolution γ spectrometry with HPGe & Si (4 systems)

defined-geometry $0.8\pi\alpha$ counter

Nal(Tl) x-ray counting system

$4\pi\beta$ (LS)- γ (Nal) anticoincidence counting system

$4\pi\beta$ -coincidence counting system (inactive)

sum-peak -- 2 Nal(Tl) -- counting system

Nal(Tl) pin-well detector

coupled large (8") Nal(Tl) detectors in 4π geometry for total efficiency counting and γ - γ coincidence counting

length-compensated proportional counters for gas counting

RIMS + TIMS

primary radon measurement system (pulse ionization chambers + gas handling)

secondary Nal(Tl)-based radon counting system

α -table with PIPS detector

large-area proportional counter & the α “pancake” counter

Various $2\pi\alpha$ surface-barrier detectors, gas-flow β proportional counters; and γ Nal(Tl) well counters

Fuji phosphor-imaging system

commercial “dose calibrator” ion chambers (6+)

windowless Si(Li) x-ray detector

2025
January 13, 2025

Who are our users ?

And how do we know their needs?

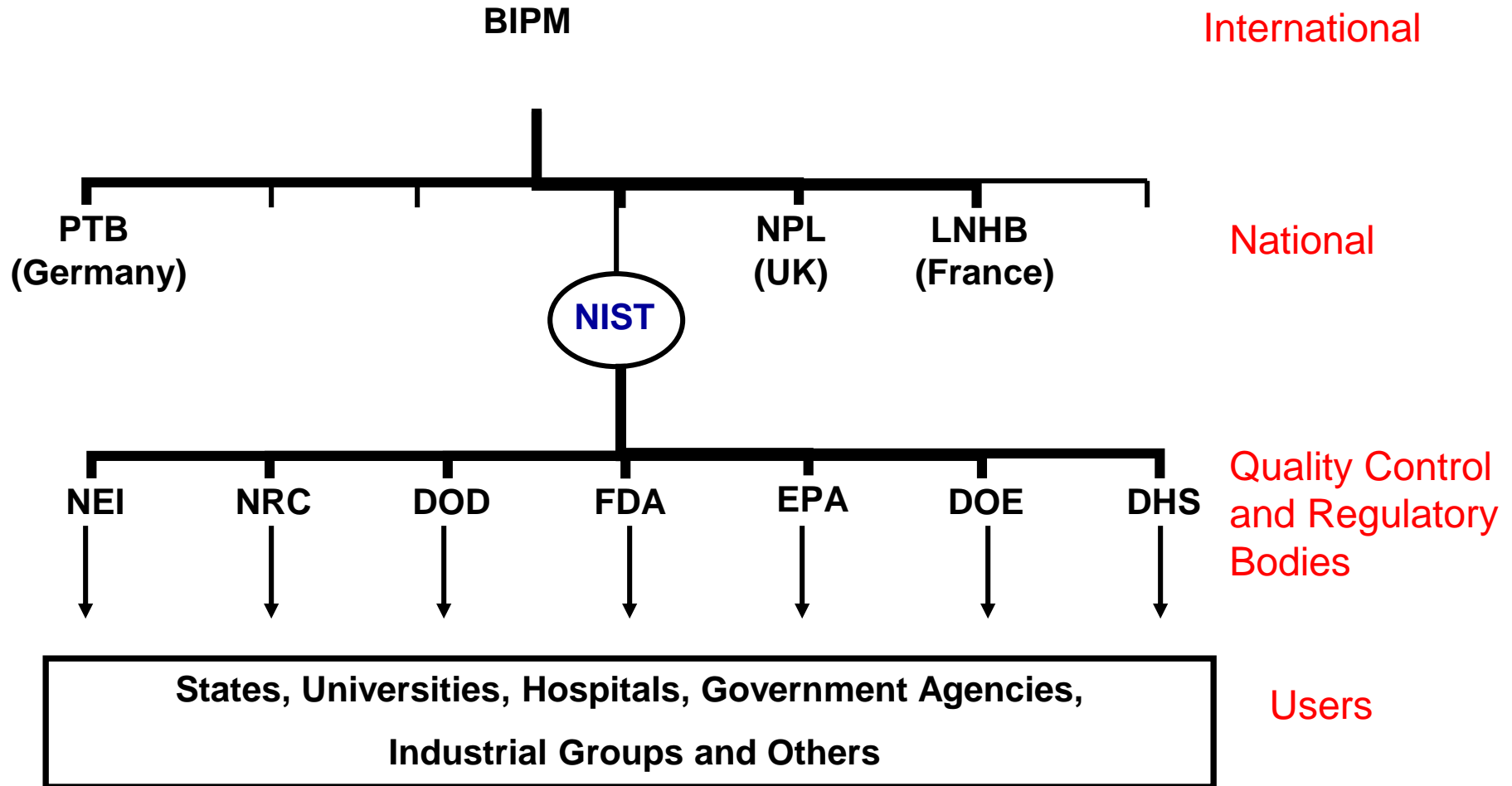
Council on Ionizing Radiation Measurements & Standards



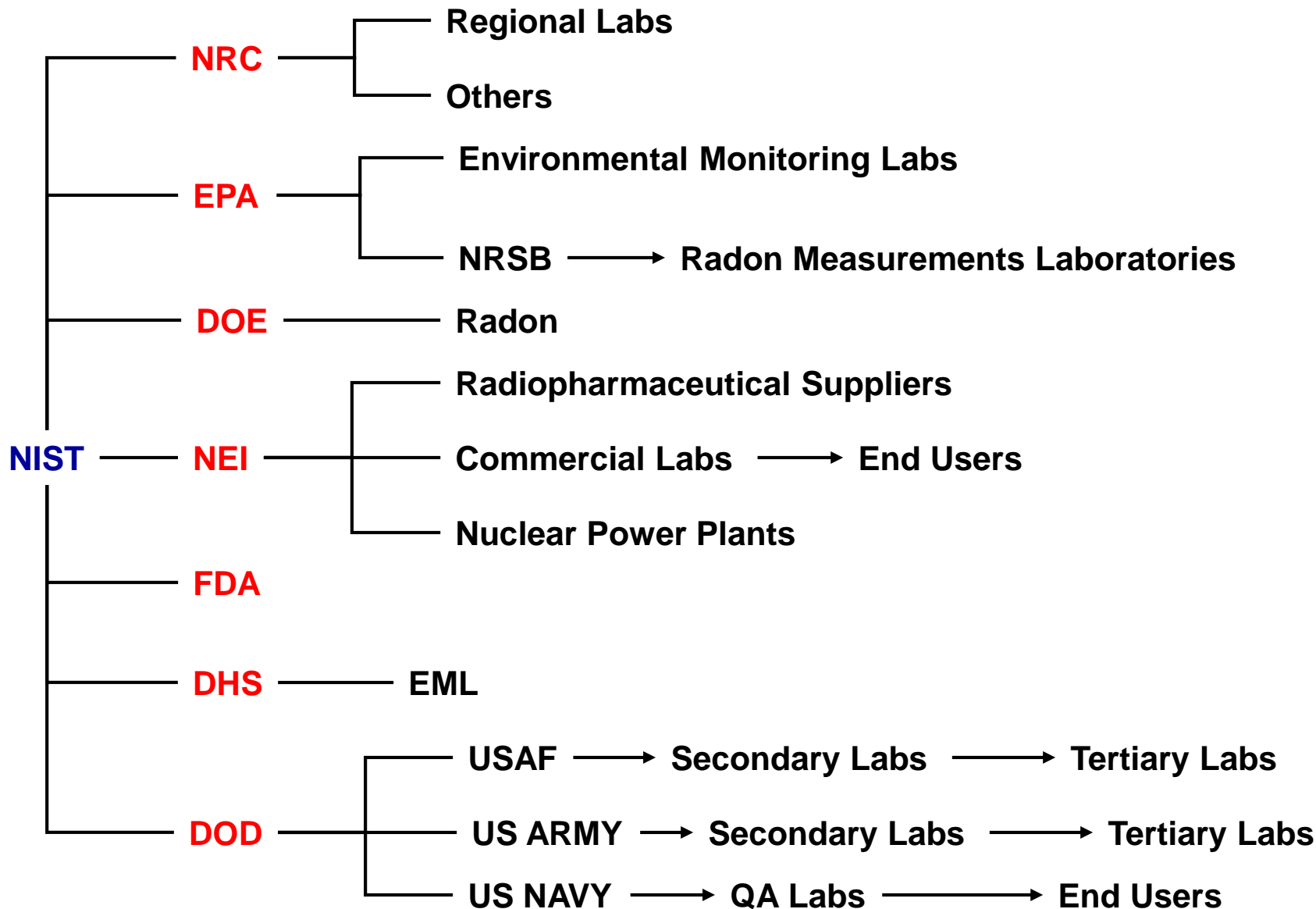
Community includes:

- National laboratories
- Monitoring laboratories
- Secondary calibration laboratories
- Radioactive source suppliers
- Radiopharmaceutical manufacturers
- Instrument manufacturers
- University research
- Nuclear power industry
- Environmental monitoring
- Governmental regulators
- Health Physics
- Military
- Homeland Security

International and National Radioactivity Measurements “Traceability Tree”



Traceability of Radioactivity Measurements in the U.S



1896

← radioactivity

1898

← polonium / radium

1901

← NBS (NIST)

1914

← 1st calibrations of radium

radium / radon

Natural series

Ores/ minerals

c. 1950

← Anthropogenic nuclides

nuclear power

international comparisons

golden age radionuclidic metrology

1970's -

← Specialization (“needs” studies)

radiopharmaceuticals

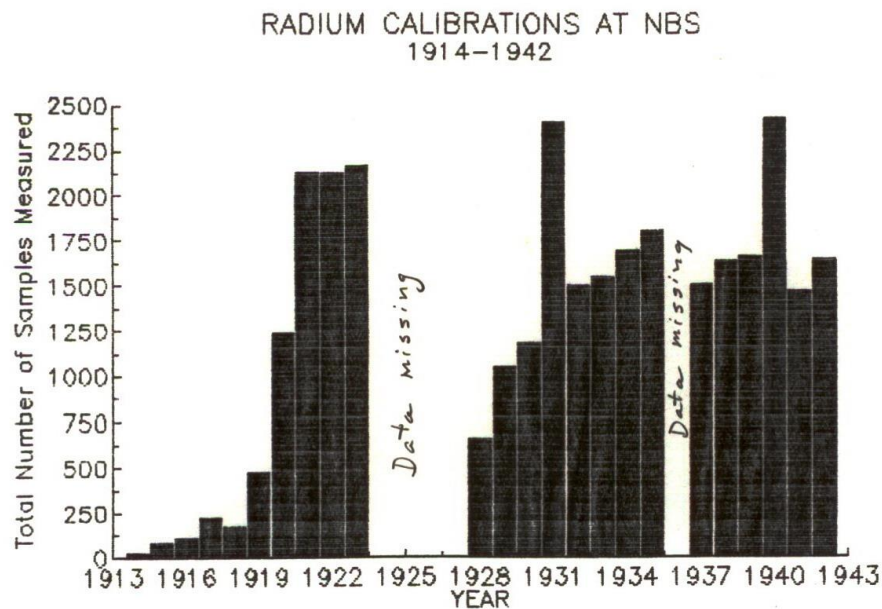
environment --- Natural Matrix Stds --

power plant

nuclear medicine – Therapy

forensics -- homeland security

Radium and Radon (1914 – WW2)

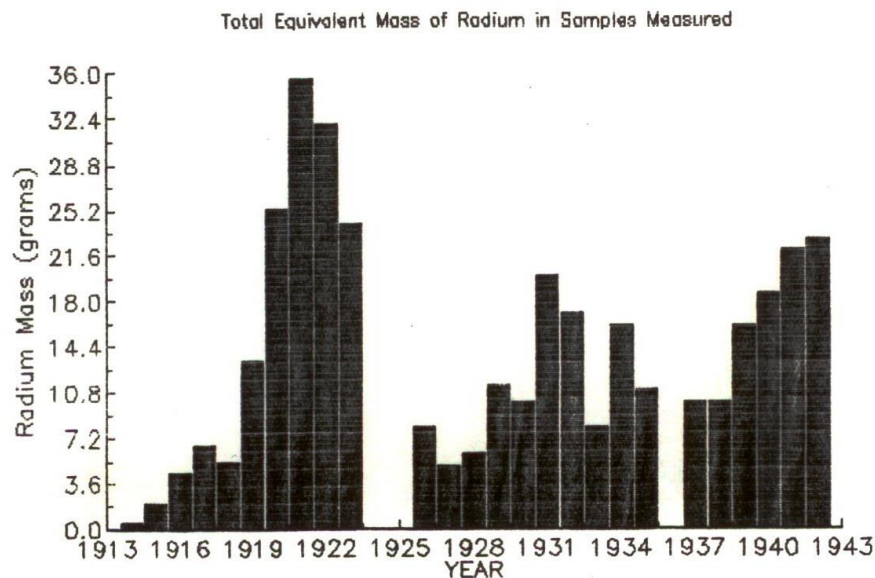


Typically 100 samples
per month

+ Breath samples
(miners & dial painters)

1942- 1952

1000 to 2000 per year



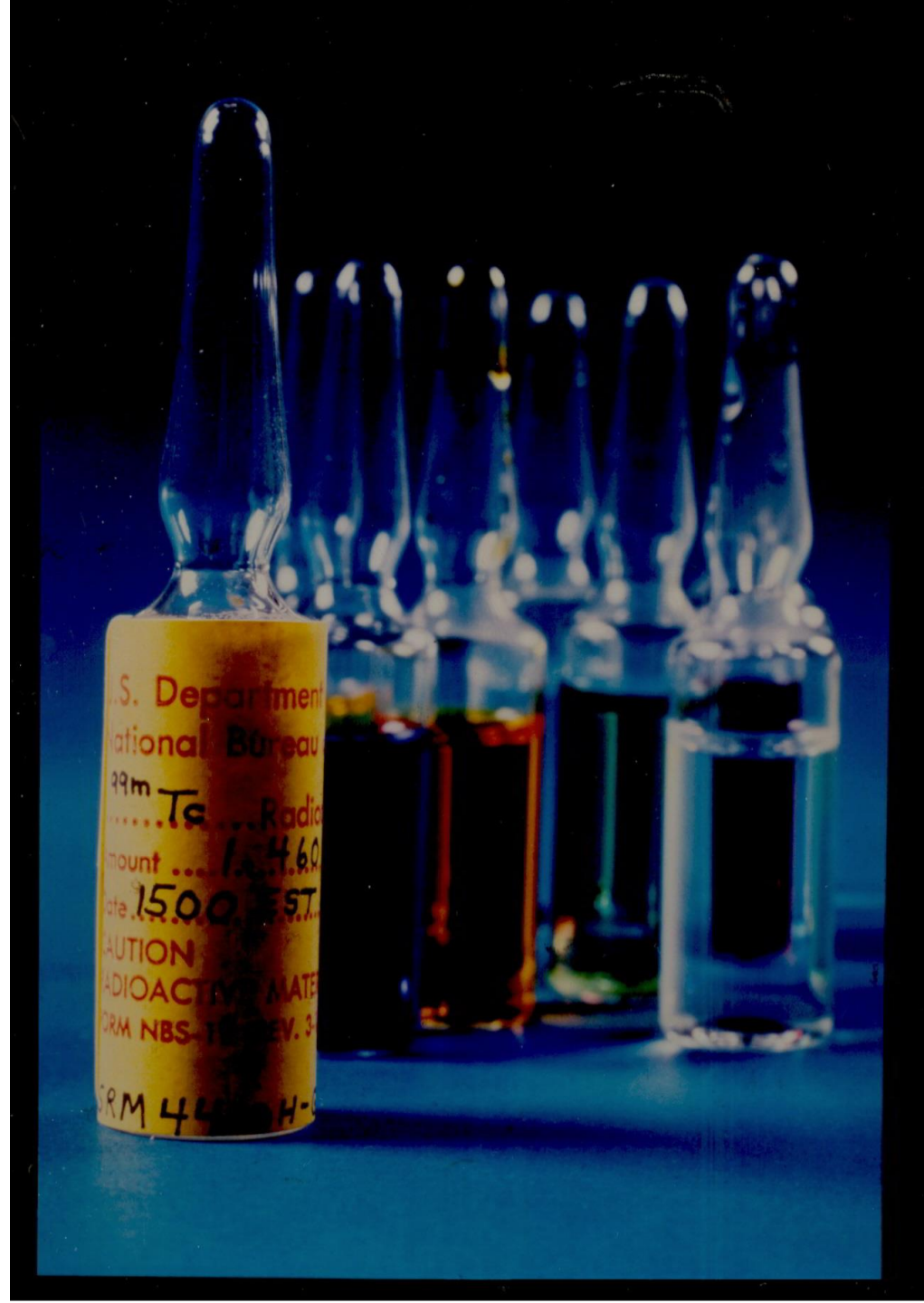
Typically > 1 gram
per month

1 gram = 1 Ci = 37 GBq

(825mR/hr at 1 m; 98 mSv/hr at 1 m)

Our principal product...

solution standards of
radionuclides



Radionuclides standardized at NIST

H-3	Fe-55	Sr-90	Sb-124	Ce-141	Hg-197	Th-229
Be-10	Co-56	Nb-93	I-124	Ce-144	Au-198	Th-230
C-14	Mn-56	Nb-94	I-125	Pm-147	Tl-201	Th-232
F-18	Co-57	Nb-95	Sb-125	Gd-148	Hg-203	U-232
Na-22	Co-58	Zr-95	Te-125m	Eu-152	Pb-203	U-234
Na-24	Fe-59	Mo-99	I-126	Gd-153	Tl-204	U-235
Al-26	Co-60	Tc-99	Xe-127	Sm-153	Bi-207	Np-237
P-32	Cu-62	Tc-99m	I-129	Eu-154	Po-208	U-238
P-33	Ni-63	Pd-103	I-130	Eu-155	Po-209	Pu-238
S-35	Zn-65	Ru-106	I-131	Eu-156	Bi-210	Pu-239
Cl-36	Ga-67	Ag-108m	Ba-131	Ho-166	Po-210	Pu-240
Ar-37	Se-75	Cd-109	Xe-131m	Ho-166m	Pb-210	Pu-241
Ar-39	Kr-79	Ag-110m	Ba-133	Yb-169	At-211	Am-241
K-40	Sr-82	In-111	Xe-133	Lu-177	Bi-214	Pu-242
K-42	Kr-85	In-113m	Xe-133m	Re-184	Pb-214	Am-243
Ca-45	Sr-85	Sn-113	Cs-134	Re-186	Rn-222	Cm-243
Sc-46	Rb-86	Sn-117m	Cs-137	Re-188	Ra-223	Cm-244
V-49	Y-88	Sn-121m	Ce-139	W-188	Ra-226	
Cr-51	Sr-89	I-123	Ba-140	Ir-192	Ra-228	
Mn-54	Y-90	Te-123m	La-140	Au-195	Th-228	

2007 Schedule for NEI Program

January	I-131	4401, lot 32	0.8 GBq	30 MBq
February	Mo-99	4412, lot 31	3.0 GBq	75 MBq
March	<i>open</i>			
April	Ga-67	4416, lot 27	0.4 GBq	20 MBq
May	Tc-99m	4410, lot 32	7.4 GBq	
June	Tl-201	4404, lot 29	0.4 GBq	33 MBq
July	<i>open</i>			
August	In-111	4417, lot 26	0.4 GBq	19 MBq
September	Xe-133	4415, lot 30	7.4 GBq	740 MBq
October	Y-90	4427, lot 10	0.2 GBq	19 MBq
November	<i>open</i>			
December	I-125	4407, lot 31	0.8 GBq	6 MBq

NIST Standard Reference Materials (SRMs) for Radionuclides Used in Nuclear Medicine and Biology

Radionuclide	SRM ID	Last Issued	Radionuclide	SRM ID	Last Issued
Chromium-51	4400N	July 1992	Xenon-133	4415, lot 29	September 2005
Iodine-131	4401, lot 32	January 2006	Gallium-67	4416, lot 27	April 2006
Tin-113-indium-113m	4402C	October 1980	Indium-111	4417, lot 25	August 2005
Strontium-85	4403B	April 1977	Mercury-203	4418A	November 1976
Thallium-201	4404, lot 28	June 2005	Ytterbium-169	4419C	October 1986
Gold-198	4405B	August 1978	Lead-203	4420B	November 1984
Phosphorus-32	4406O	October 1997	Gold-195	4421A	December 1979
Iodine-125	4407, lot 31	December 2005	Chlorine-36	4422A	April 1980
Cobalt-57	4408F	July 1995	Strontium-90	4423A	November 1985
Selenium-75	4409D	August 1981	Sulfur-35	4424A	October 1988
Technetium-99m	4410, lot 32	May 2006	Samarium-153	4425D	July 2002
Iron-59	4411B	January 1979	Strontium-89	4426A	April 1995
Molybdenum-99	4412, lot 31	February 2006	Yttrium-90	4427, lot 9	October 2005
Mercury-197	4413A	May 1976	Gadolinium-153	4428A	October 1998
Iodine-123	4414C	June 1980			

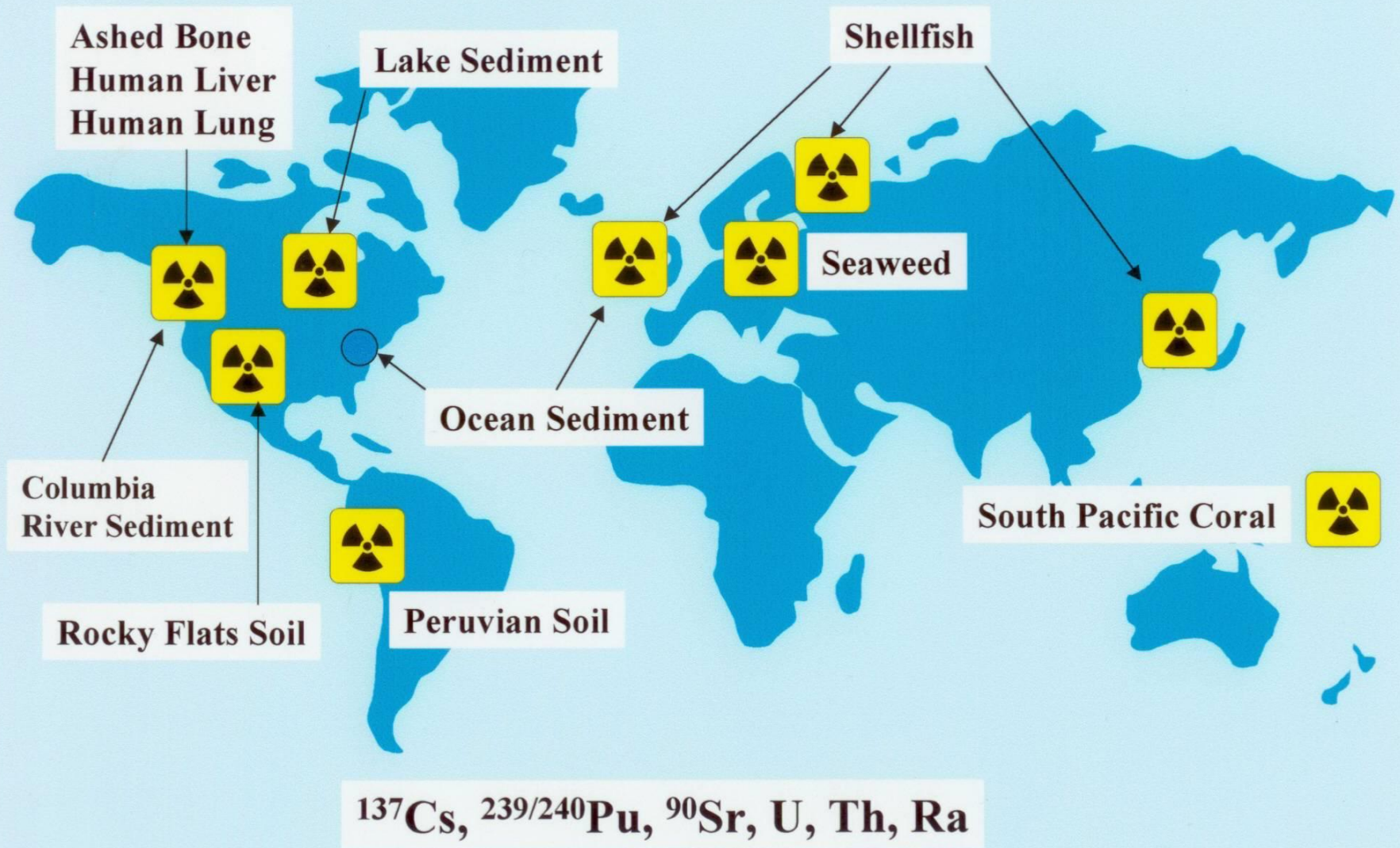
Natural Matrix SRMs

for Environmental Radioactivity Measurement

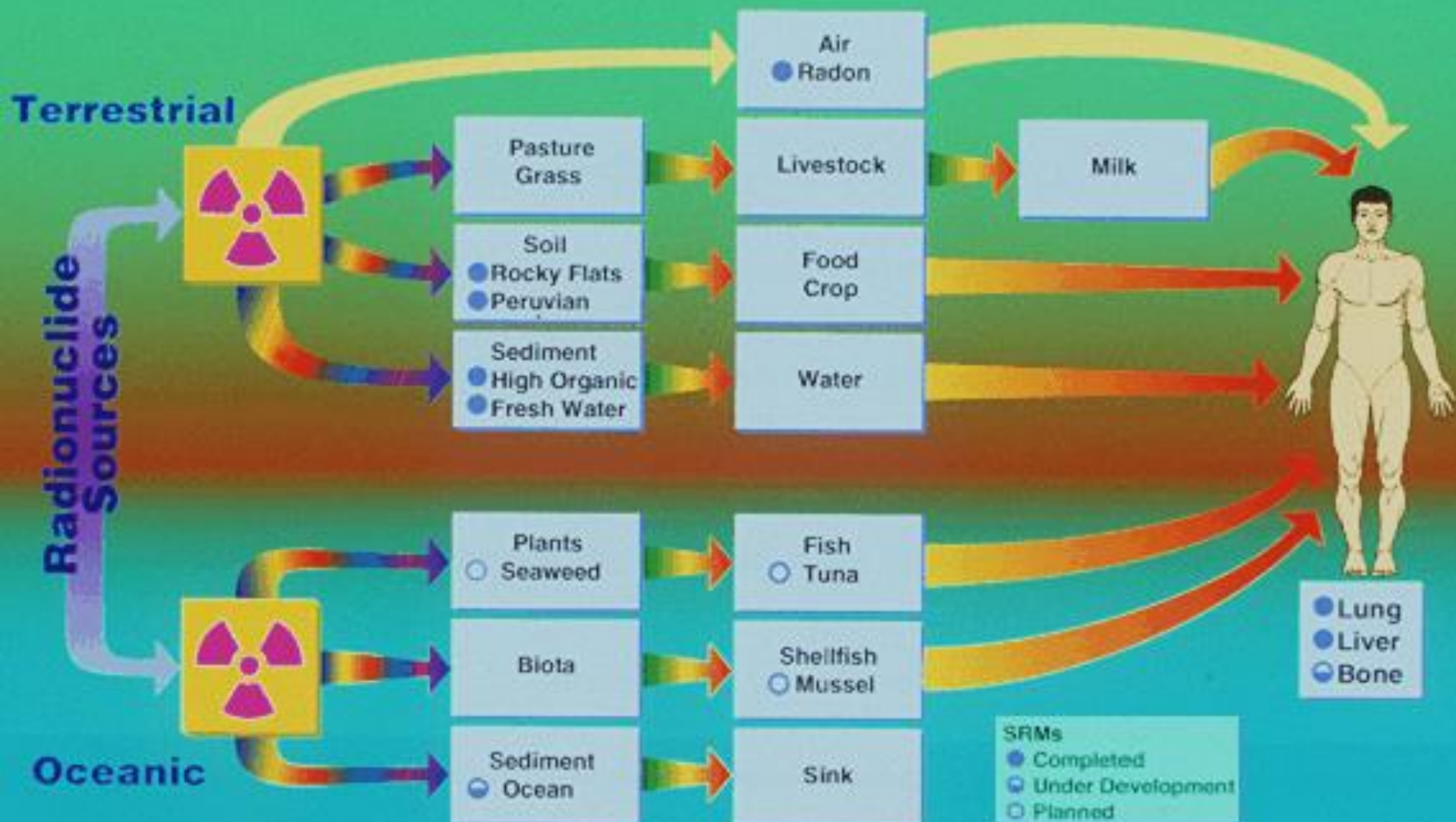


- Rocky Flat Soil I
- River Sediment
- Peruvian Soil
- Human Lung
- Human Liver
- Lake Sediment
- Ocean Sediment
- Bone Ash
- Shell Fish

Environmental Radionuclide SRM's



Environmental Radioactivity



Natural Matrix Standard Needs

Matrix

Radionuclides

10-Yr Requirements

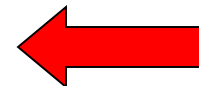
Needs --- last 30 years

Soil:

**High Ca
Low Ca**

**^{90}Sr , ^{137}Cs , ^{210}Pb ,
Alpha Emitters**

**5,000 Aliquants
(1 kg Samples)**



Sediments:

**High Ca
Low Ca
Mill Tailings**

**Alpha- & Beta-Particle, and
Photon Emitters**

**1,600 Aliquants
(100 g Samples)**

Water:

**^3H , ^{60}Co , ^{90}Sr , ^{106}Ru , ^{134}Cs ,
 ^{137}Cs , Natural Radionuclides,
Alpha-Particle Emitters**

**Thousands of Aliquants
(50-100 mL Samples)**

Biological: Aliquants

**Lung
Liver
Bone
Milk
Sea Clam
Sea Hare
Seaweed**

^3H , ^{14}C , Fission & Activation

Several Hundred

**Products, Alpha-Particle
Emitters**

Forensics (chronology)

Isotopic standards natural materials

Wastes (water, sludges, concretes, etc.

Foodstuffs

Particles (sizes, nuclides, ionic state)

Radiobioassays (people +)

more

Air: Filters

Natural Radionuclides

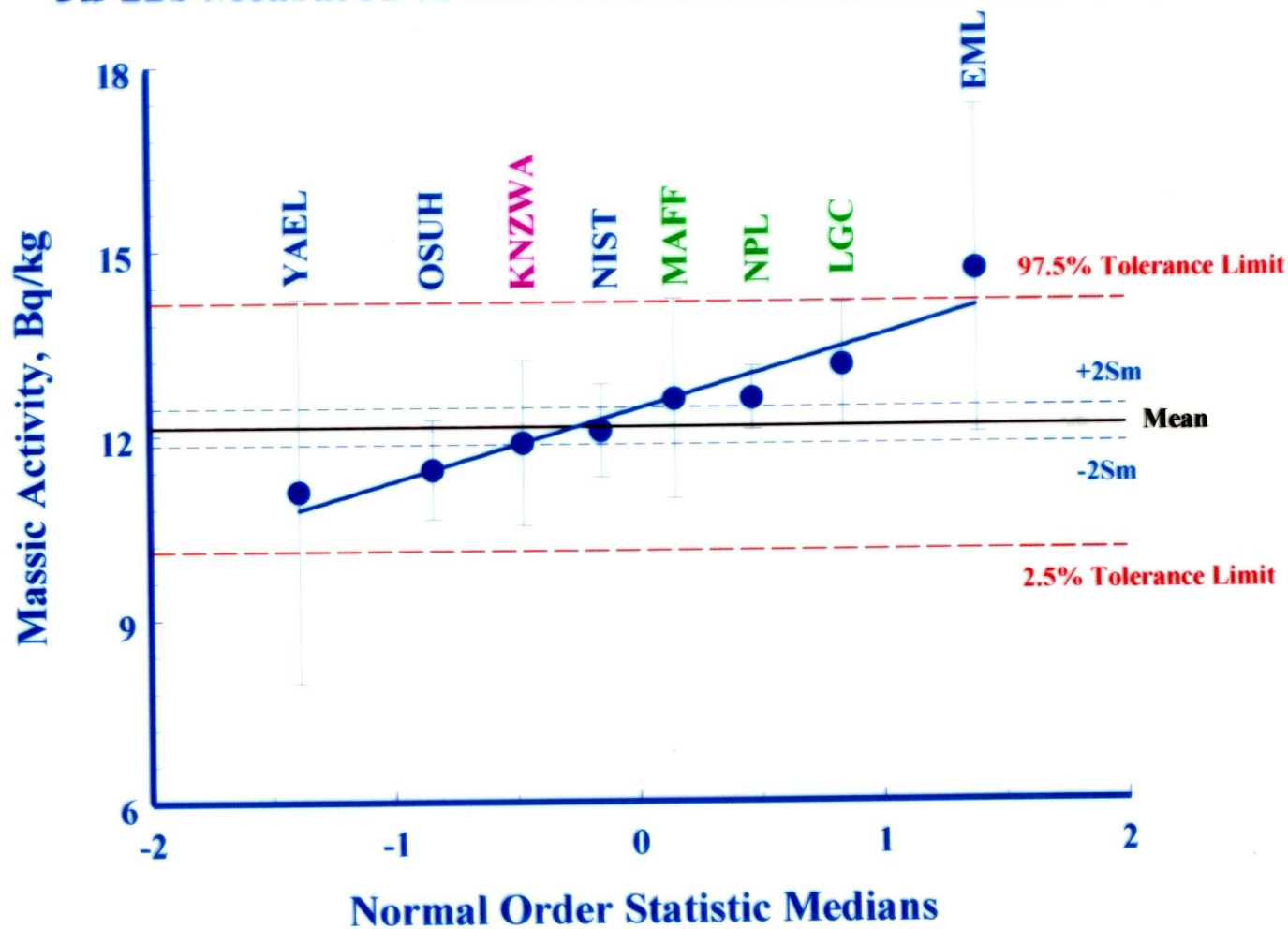
New areas (next 20 years)



Process of Developing a NIST Natural-Matrix Radionuclide SRM

- Matrix Selection
- Matrix Collection
- Matrix Preparation
 - Size
 - Drying
 - Milling/Pulverization
 - Blending
 - Final Drying
 - Sterilization
- Interlaboratory Comparison
 - Traceability
- Data and Statistical Analysis
- Discrepancy Resolution
- Write Certificate

Th-228 Measured in the NIST Ocean Sediment SRM-4357



The digestion and detection methods used for the radionuclides of interest in the NIST bone ash SRM

Radio-nuclides	Digestion/Detection Method	Participant
⁴⁰ K	1A	JCAC, PTB
⁹⁰ Sr	(3 + 5)B + 3D	IRMM, JCAC, MAFF, NRPB
²¹⁰ Pb	1A + 3B + 3C	IRMM, JCAC, JSI, MAFF, PTB
²¹⁰ Po	4C	NRPB
²²⁶ Ra	1A + 5D + 3E	IRMM, JCAC, JSI, MAFF, PTB
²²⁸ Ra	1A	JSI
²²⁸ Ac	1A	JCAC, PTB
²²⁸ Th	1A + (2 + 3 + 6)C	IRMM, JCAC, JSI, MAFF, NIST, PTB
²³⁰ Th	(2 + 3 + 6)C	IRMM, JCAC, MAFF, NIST
²³² Th	(2 + 3 + 6)C	IRMM, JCAC, MAFF, NIST
²³⁴ U	(2 + 3 + 6)C	IRMM, JCAC, LDMC, LDML, MAFF, NIST, PTB
²³⁵ U	(2 + 3 + 6)C	JCAC, LDMC, MAFF, NIST, PTB
²³⁸ U	(2 + 3 + 6)C	IRMM, JCAC, LDMC, LDML, MAFF, NIST, PTB
²³⁸ Pu	(2 + 3 + 6 + 7)C	IRMM, JCAC, LDMC, LDML, MAFF, NIST, NRPB, PTB
(²³⁹ + ²⁴⁰)Pu	(2 + 3 + 6 + 7)C	IRMM, JCAC, LDMC, LDML, MAFF, NIST, NRPB, PTB
²⁴¹ Am	1A + (2 + 3 + 6 + 7)C	IRMM, JCAC, JSI, LDMC, LDML, MAFF, NIST, NRPB, PTB
(²⁴³ + ²⁴⁴)Cm	(3 + 6)C	JCAC, LDMC, LDML, MAFF

Sample Digestion Methods:

1. Non-destructive
2. HNO₃ + H₂O₂ + HF + HClO₄
3. HNO₃
4. HNO₃ + HCl
5. HNO₃ + HF
6. HNO₃ + H₂O₂
7. Calcination at 500°C followed by addition of 8M HNO₃

Detection Methods:

- A. (Ge) gamma-ray spectrometer
- B. Beta-particle Geiger counter
- C. Alpha-particle spectrometer
- D. Liquid scintillation counting
- E. Lucas Emanation Cell

Many other geometries for standards & calibrations

- Spiked sources (soils, synthetic urine, water)
- Filter sources
- Point Sources
- Marinelli beakers
- Radioactive gas ampoules
- Polyethylene encapsulated radon emanation sources
- Large area sources
- Dose vials
- Brachytherapy sources

Alpha-Particle Emitters Including Mixed Alpha
Beta-Particle Emitters
X- and γ -ray Emitters

Customers include: Department of Defense, Department of Energy, Environmental Protection Agency, Nuclear Regulatory Agency, National Aeronautics and Space Agency, Department of Homeland Security, Nuclear power industry, Source & instrument manufacturers, Radiopharmacies, etc..

Radioactivity Measurement Assurance Programs (MQAs)

**Nuclear Energy Institute – Radiopharmaceuticals
(31 years)**

**Nuclear Energy Institute – Power Station Radiochemistry
(17 years)**

**NIST Radiochemistry Intercomparison Program (NRIP)
(8 years)**

Recent Standards (2005-2007)

^{63}Ni	$4\pi\beta$ LS – ^3H Std CNET & TDCR
^{55}Fe	microcalorimetry + LS
^{241}Am & ^{230}Th	$4\pi\alpha$ LS
^{60}Co & ^{137}Cs	$4\pi\gamma$ ion chamber ($4\pi\beta(\text{LS})$)- $\gamma(\text{NaI})$ anticoincidence counting
^{90}Sr	$4\pi\beta$ LS -- CNET & TDCR
$^{166\text{m}}\text{Ho}$	HPGe (photon emission only)
^{210}Pb	$4\pi\beta$ LS ^3H Std CNET $4\pi\beta(\text{LS})$)- $\gamma(\text{NaI})$ anticoincidence counting $2\pi\alpha$ spectrometry of ^{210}Po ingrowth HPGe γ spectrometry
^{241}Pu	$4\pi\beta$ LS -- CNET & TDCR $4\pi\alpha$ LS -- ingrowth ^{241}Am (30 years)
Seaweed -- Natural Matrix	(14 nuclides + 15 uncertified; 24 labs)
Rockyflats soil # 2	(10 nuclides + 22 uncertified; 12 labs)

Underway: ^{99}Tc ; ^{209}Po ; ^{226}Ra ; ^{228}Ra ; ^{229}Th ; ^{238}Pu ; ^{239}Pu ; Shellfish; Peruvian soil

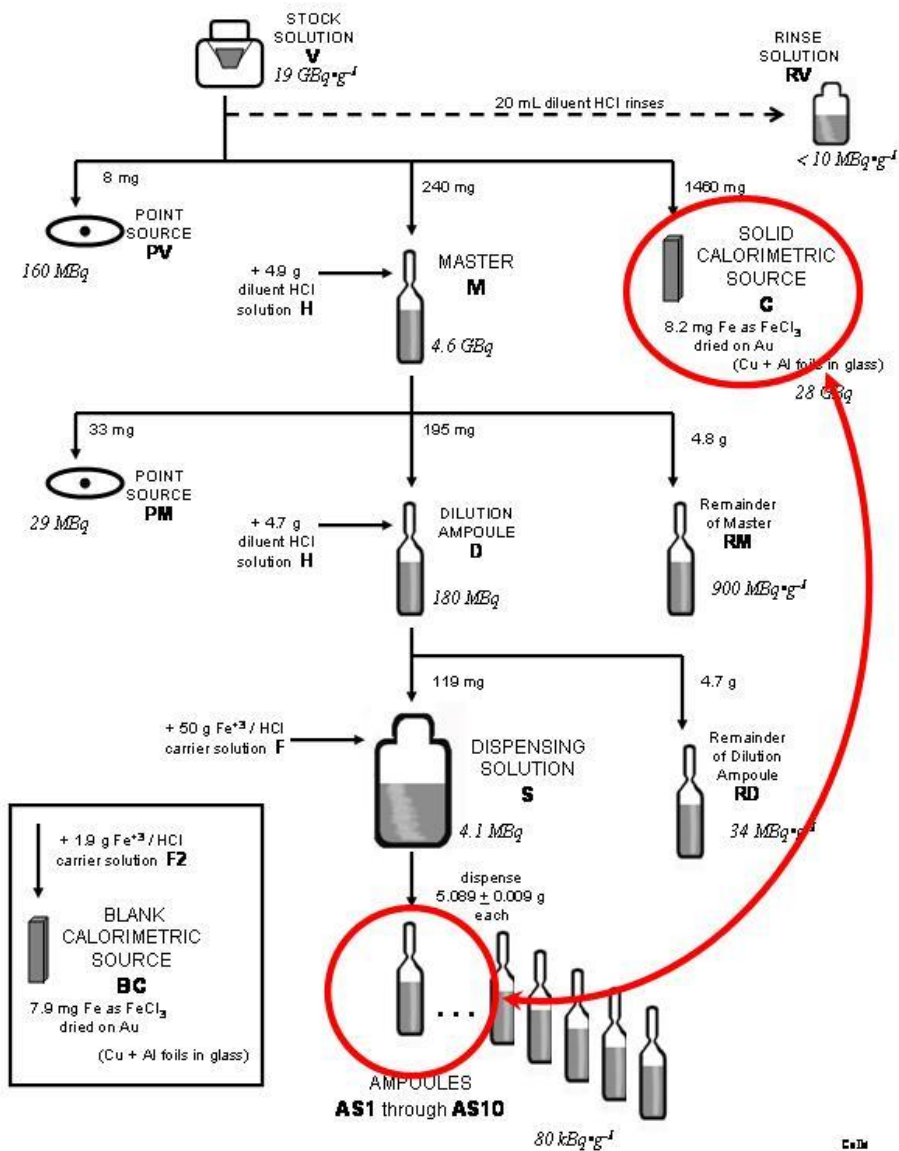
Just one example

^{55}Fe

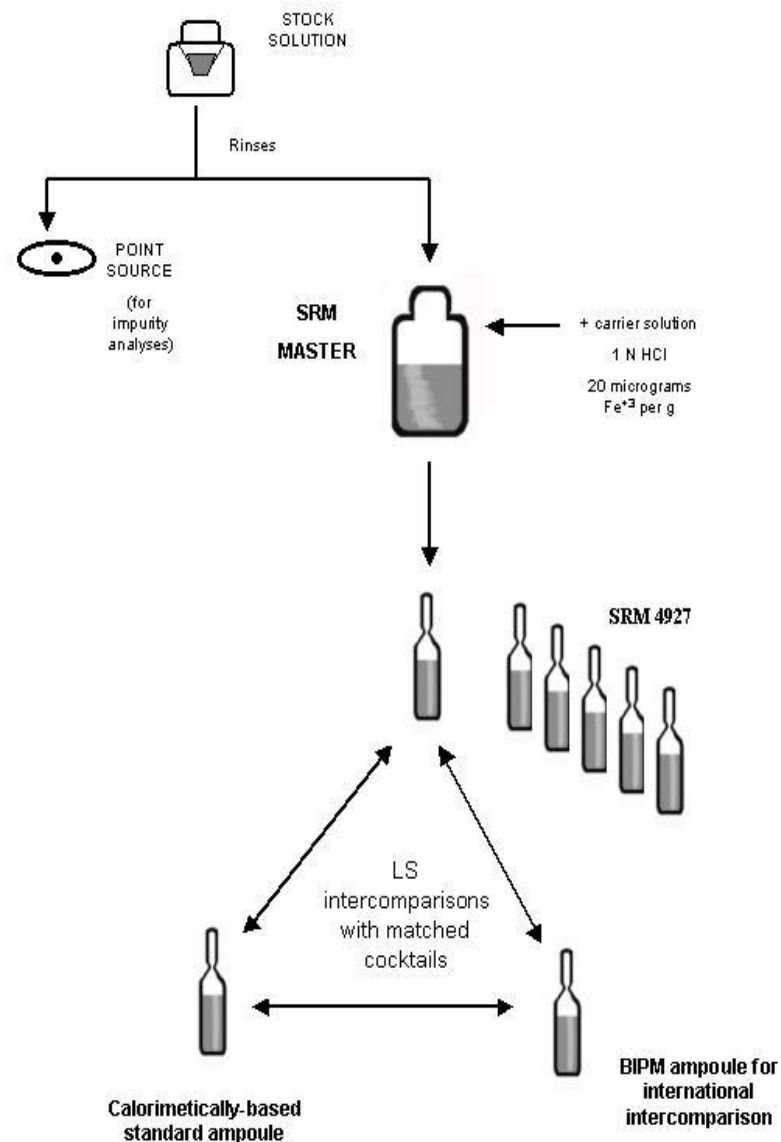
primary standardization by microcalorimetry

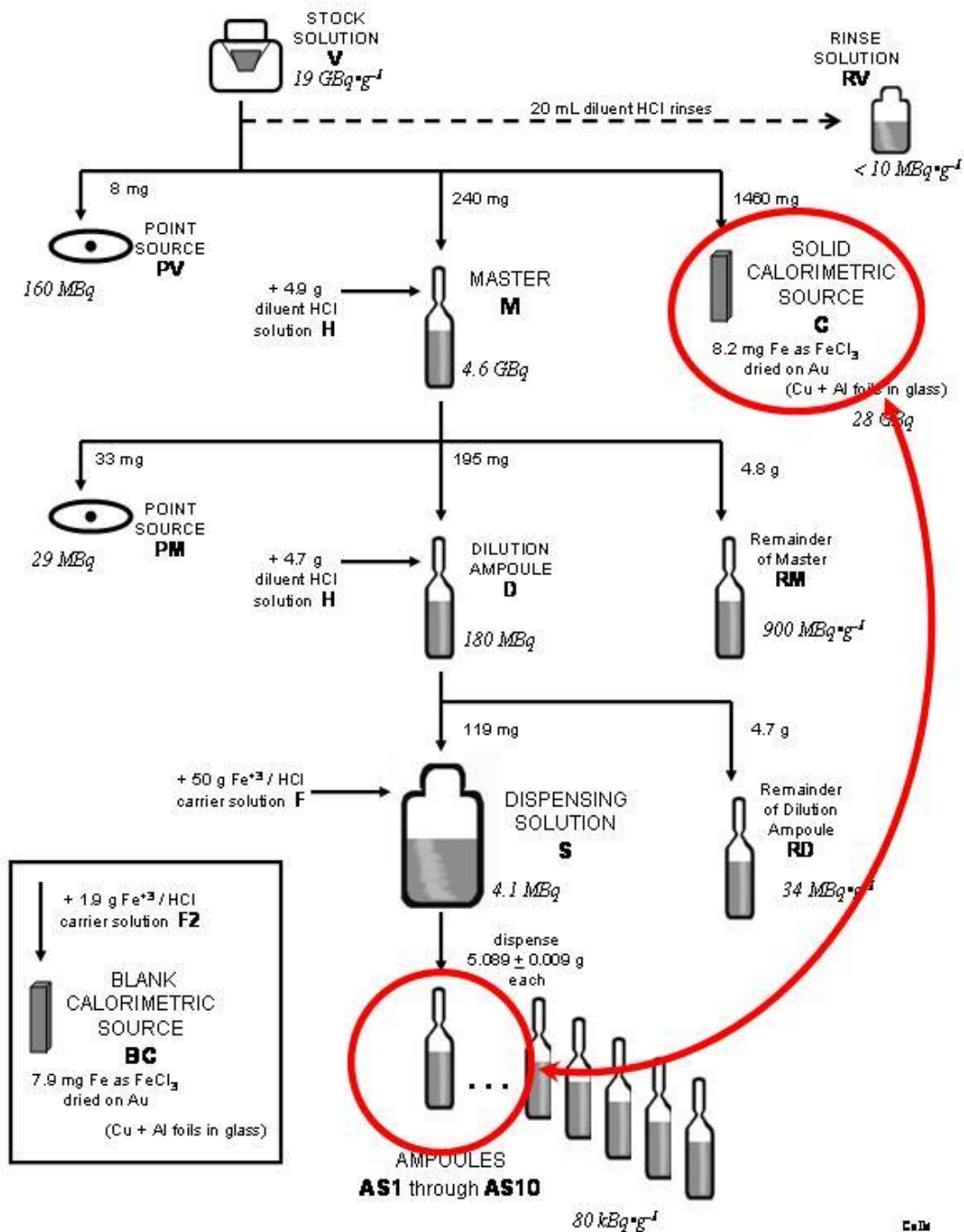
linked to SRM

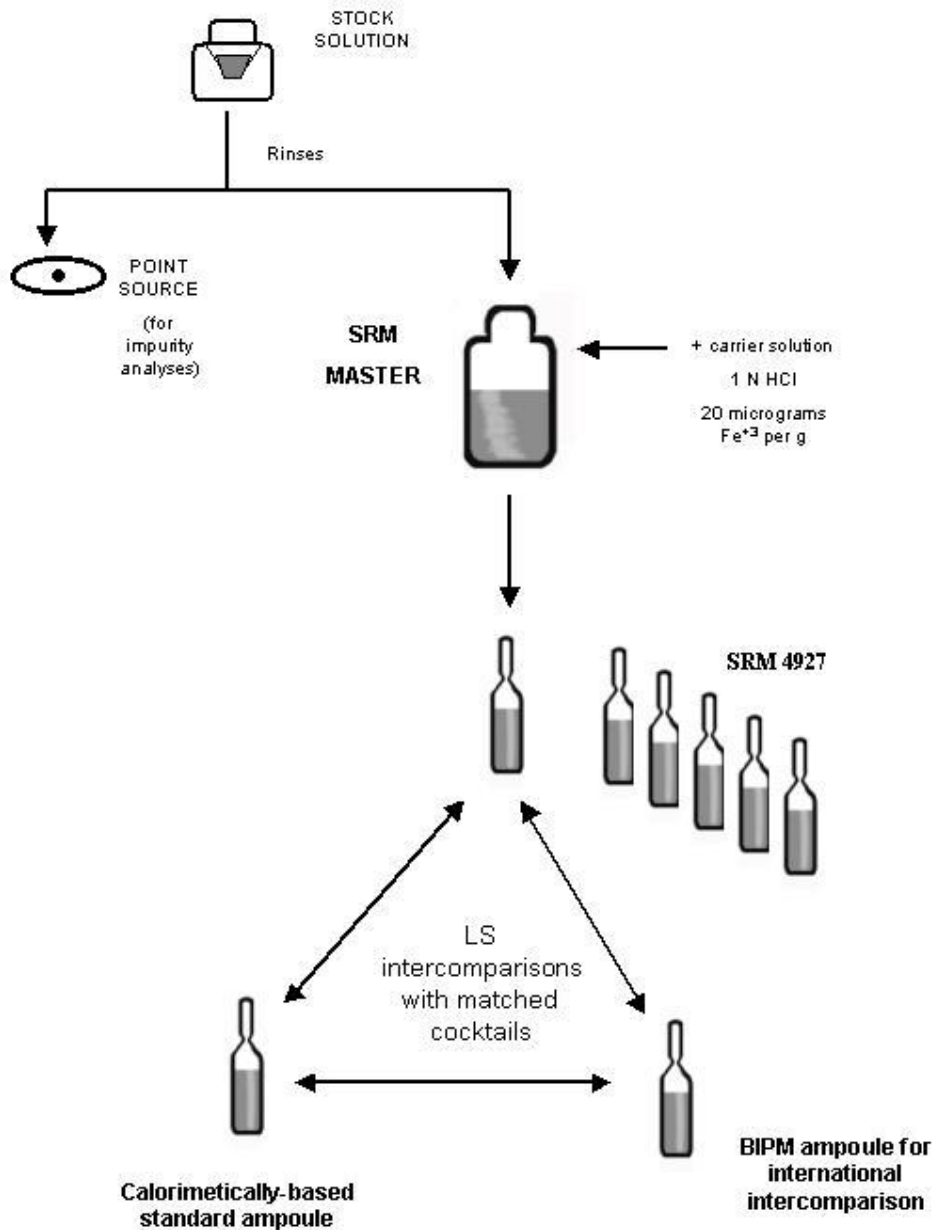
international intercomparison



CaM
2004







Calorimetry

13 independent determinations

LS intercomparisons

776 activity ratios; variables include:

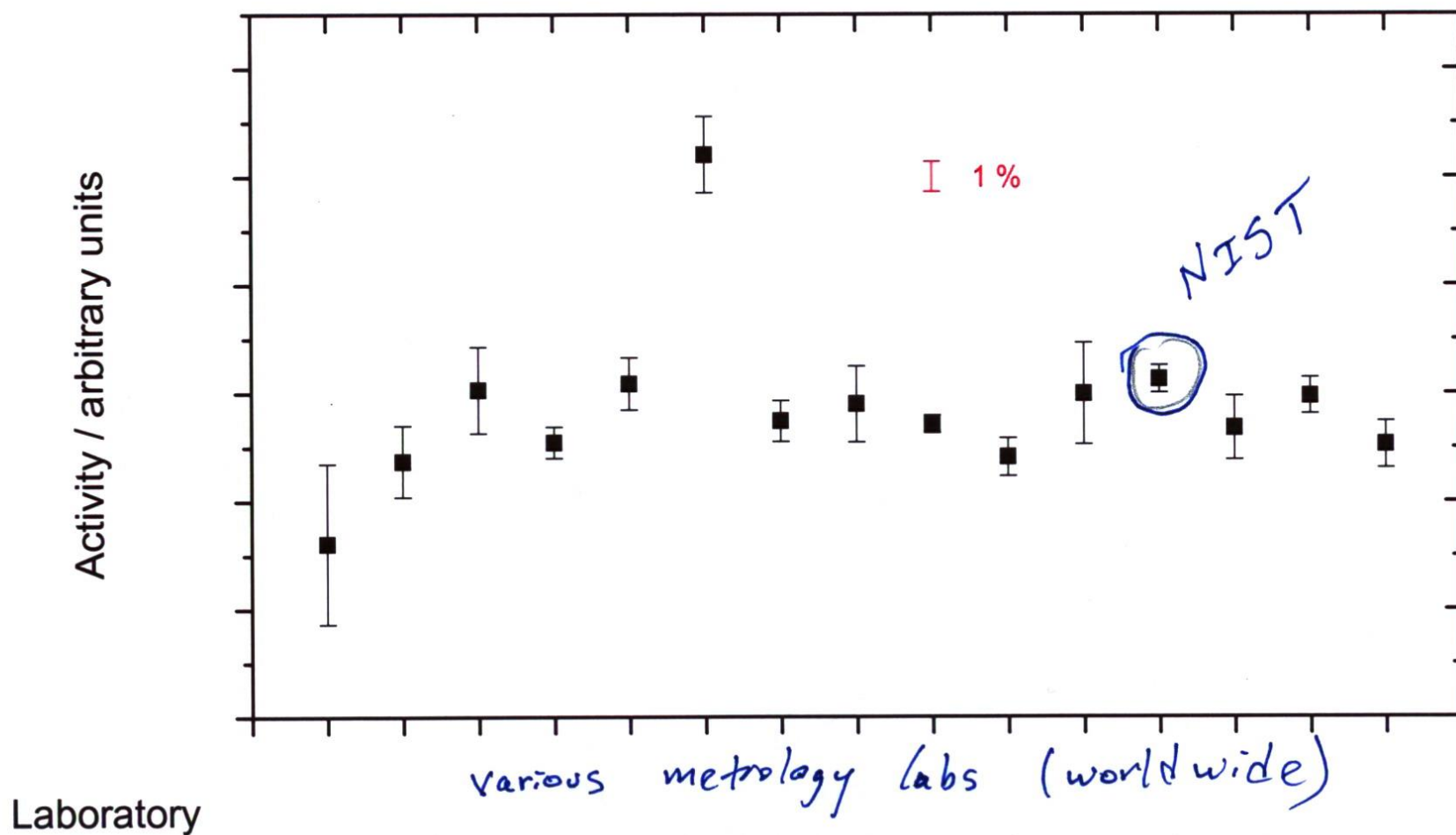
- 3 counters
- 3 scintillators
- 44 matched cocktails
- 4 distinct aq. fraction (+Fe) compositions
- 2 NIST solution dilutions
- 97 days of aging

NIST Uncertainty Analysis for ^{55}Fe Microcalorimetric Standardization of NIST Solution Standards

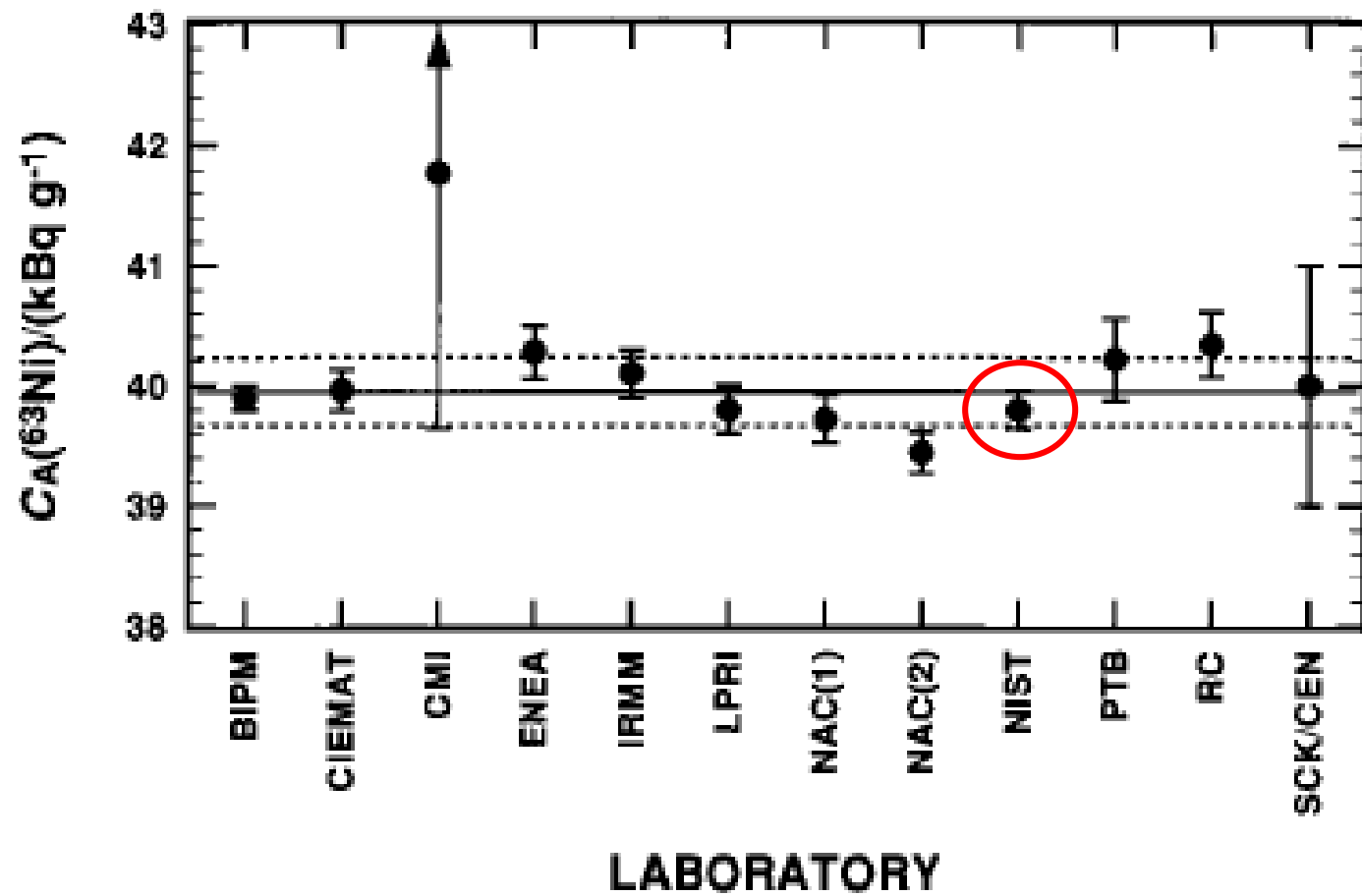
Item	Uncertainty component	Assessment Type	Relative standard uncertainty contribution on massic activity of ^{55}Fe (%)
1	Measurement precision for 13 independent calorimetric determinations of the power of solid source C; includes precision in the calibrations & baseline measurements for each determination; std. dev mean for $\nu=12$ degrees freedom (passes Normal test)	A	0.25
2	Gravimetric (mass) linkage of source C to NIST standard solutions	B	0.07
3	Activity loss in source C preparation	B	0.15
4	Power calibration of calorimeter, includes any systemic heat losses	B	0.05
5	Possible heat defect / excess effects	B	0.1
6	^{55}Fe decay corrections during calorimetric measurements	B	0.02
7	^{55}Fe decay corrections from calorimetric reference time to BIPM reference time.	B	0.08
8	Average energy per decay for ^{55}Fe (to convert calorimetric power to activity)	B	0.17
COMBINED STANDARD UNCERTAINTY			0.39

Results (without the outlier value)

International comparison of activity measurements of a solution of ^{55}Fe
Preliminary results; January 2007



^{63}Ni international intercomparison



Features of our standardization work

Available “transfer standards” (SRMs, etc.) are based on identified “needs”

Standardized by primary method

Usually at least one confirmatory determination

Establish links to previous calibrations, if possible

Develop & maintain secondary calibrations

Uncertainties ($k = 2$) typically $< 1 \%$

u ($k = 1$) few tenths of %

Comparisons with other metrology labs

N.B. → “***National Standards***” are not artifacts but are our (NIST’s) ability to perform primary standardizations with our instruments, procedures, people, etc...