Applied Nuclear Science at Pacific Northwest National Laboratory; The Diverse Work of National Security, International Treaties, and Basic Research

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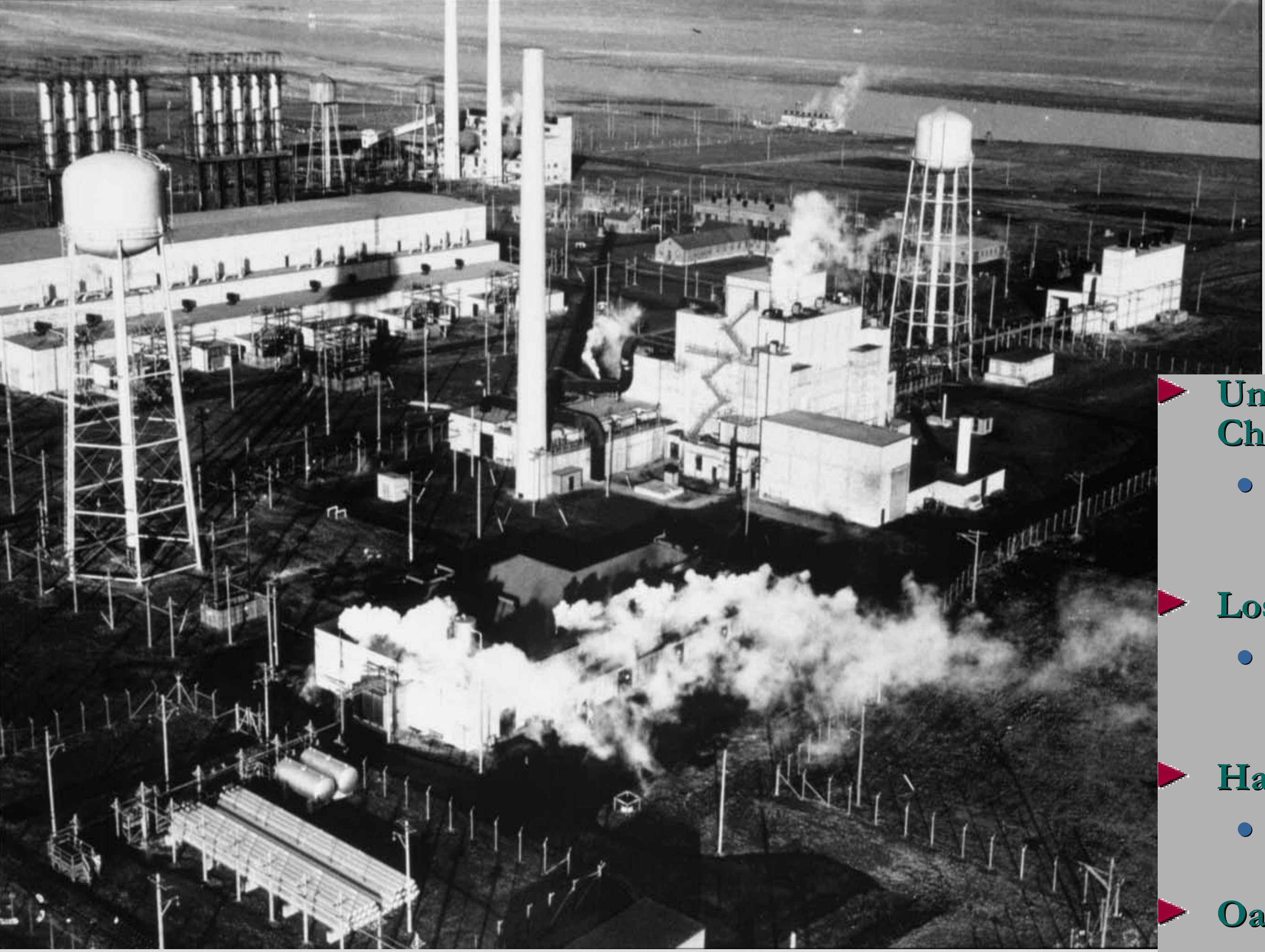
Outline

- Hanford site: a brief history
- Radiological and Chemical Sciences Group
  - Capabilities and Projects
- Comprehensive Nuclear-Test-Ban-Treaty Organization
  - Radioxenon Monitoring



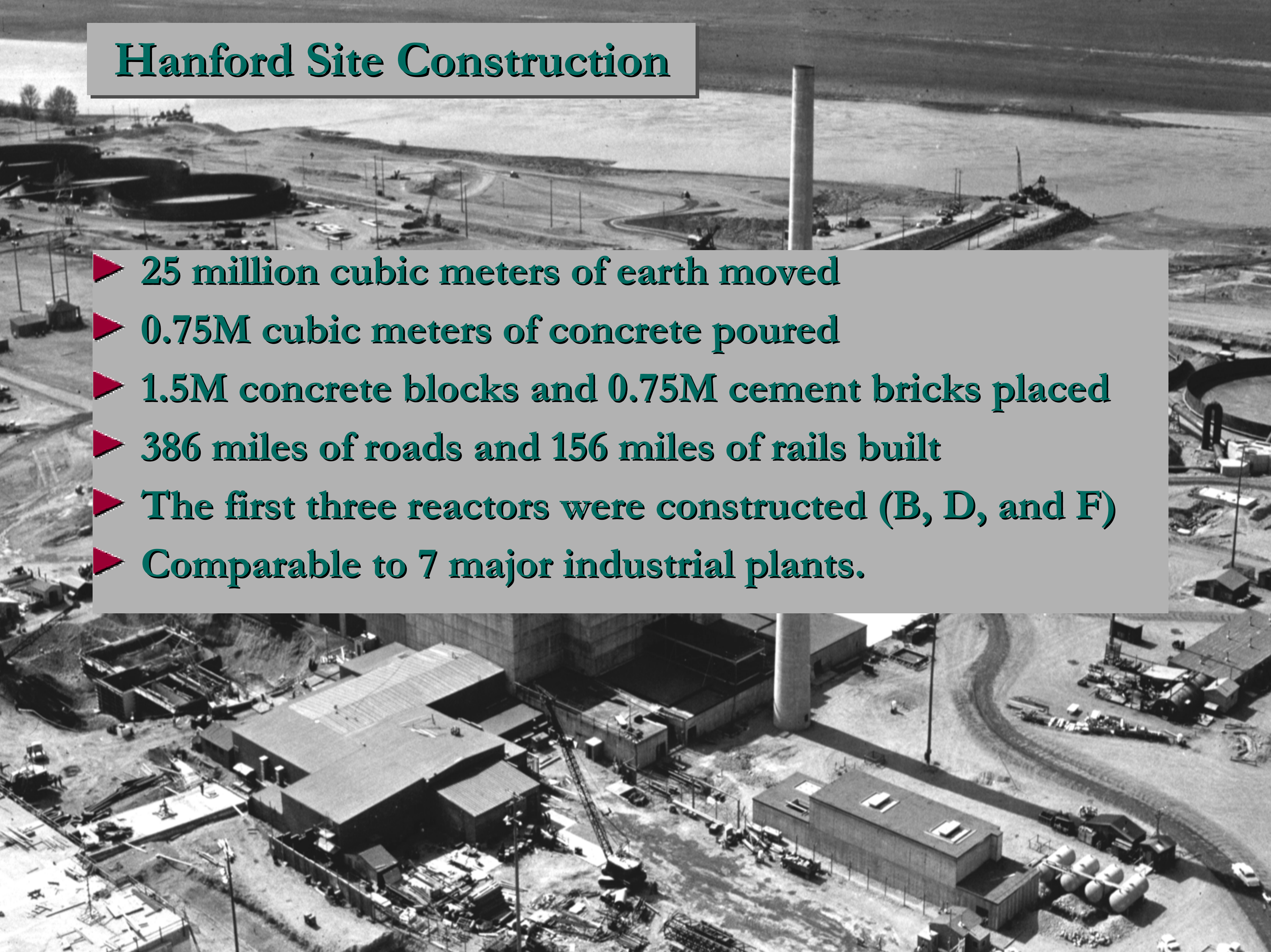
- University of Chicago
  - Theoretical underpinnings a nuclear weapon
- Los Alamos
  - Weapons design and manufacture
- Hanford
  - Reactors and Pu production
- Oak Ridge
  - Isotopic ^{235}U separation

1940's: Building the Atomic Bomb
The Manhattan Project

1943: Selection of the Hanford site

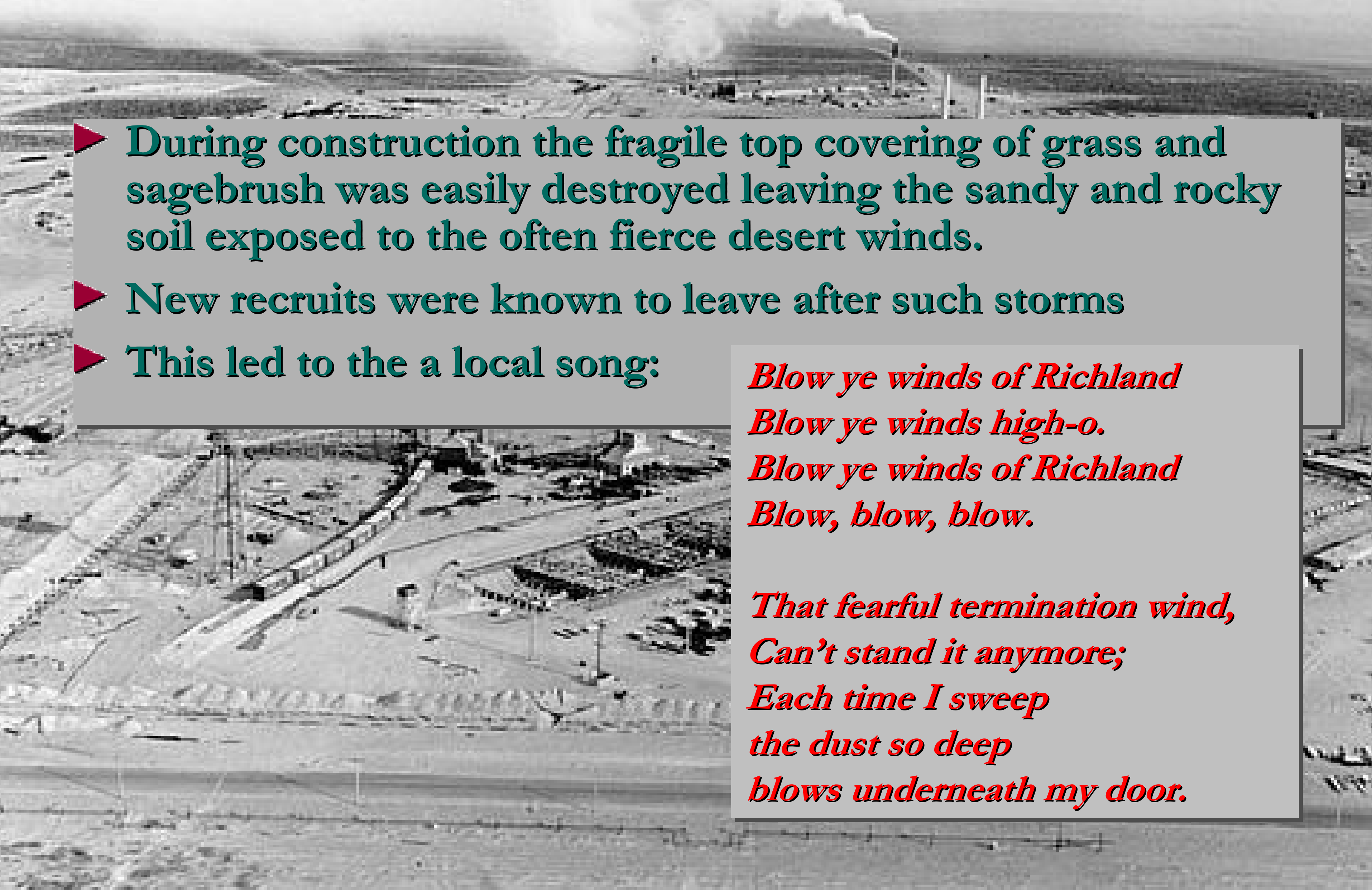
- Extremely low population density minimized the need for displacement
- Fast flowing river to cool the production reactors
- Abundant supplies of cheap electricity from large hydroelectric dams
- Remote… to maintain secrecy
- Existing transportation – railroad, barge, and roads

Hanford Site Construction



- 25 million cubic meters of earth moved
- 0.75M cubic meters of concrete poured
- 1.5M concrete blocks and 0.75M cement bricks placed
- 386 miles of roads and 156 miles of rails built
- The first three reactors were constructed (B, D, and F)
- Comparable to 7 major industrial plants.

Termination Winds



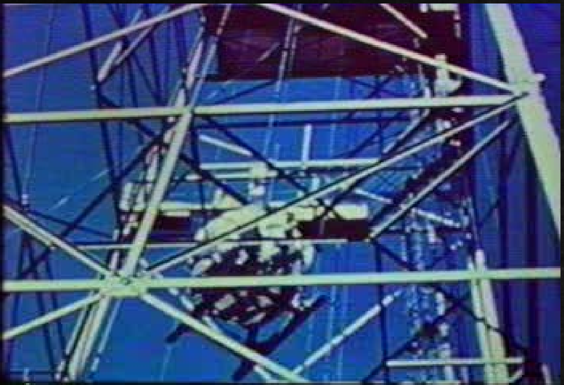
- During construction the fragile top covering of grass and sagebrush was easily destroyed leaving the sandy and rocky soil exposed to the often fierce desert winds.
- New recruits were known to leave after such storms
- This led to the a local song:

Blow ye winds of Richland
Blow ye winds high-o.
Blow ye winds of Richland
Blow, blow, blow.

That fearful termination wind,
Can't stand it anymore;
Each time I sweep
the dust so deep
blows underneath my door.

Hanford's Primary Mission: Produce Plutonium

- 1944: The first Hanford reactor to come on line was B reactor.
  - Photo: workman loading the core of B reactor with uranium-slug-filled rods
  - In the initial hours of operation the reactor began to cool significantly due to ^{135}Xe poisoning (2,500,000 barn thermal neutron cross-section).
  - The reactor operators where able to overcome this problem by loading extra uranium into the reactor core
  - The core capacity had been increased by the reactor engineers unbeknownst to the physicists at the time.



- In its first nine months of operation, B reactor produced the Pu used in the Trinity test and the Fatman bomb

Radiological and Chemical Sciences Group

RC&S Staff Composition

- 80 staff members
- Physics, all kinds but especially nuclear
- Chemistry
  - Radiochemistry / Nuclear Chemistry
  - Analytical Chemistry
  - Physical Chemistry
- Engineering
  - Chemical, Electrical, Mechanical, Nuclear
- Other
  - Materials Science
  - Mathematics
  - Computer Science

Projects & Programs

- Homeland Security-- WMD search, interdiction, & consequences.
- Nuclear Material Detection and Characterization
- Process, waste, environment characterization
- Neutrino Physics -- Majorana
- Proliferation detection, especially nuclear test detection

U.S. Customs Support

- Instrument US Border Crossings with Radiation Detection Capability

High throughpu t systems

Gamma & neutron detectors

- Characterize threat
  - Gamma and Neutron Detectors
- Apply Commercial of the shelf interdiction technology
- Engineer Installation at USCS point of entry
- Train US Customs Staff
- Work with industry and operations experts for successful technology deployment

WMD Screening for Cargo Containers

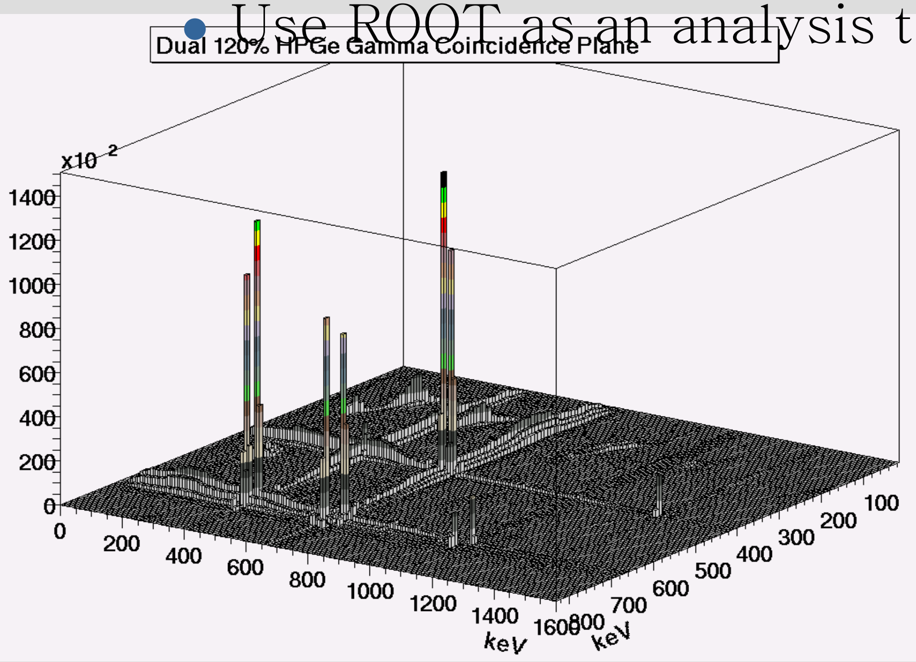
- Need effective yet operationally acceptable sensors to detect threats in inbound cargo containers
  - Assess expected nuclear signatures given realistic cargo, leading to performance specifications
  - Use network of low-cost sensors exploiting long measurement time

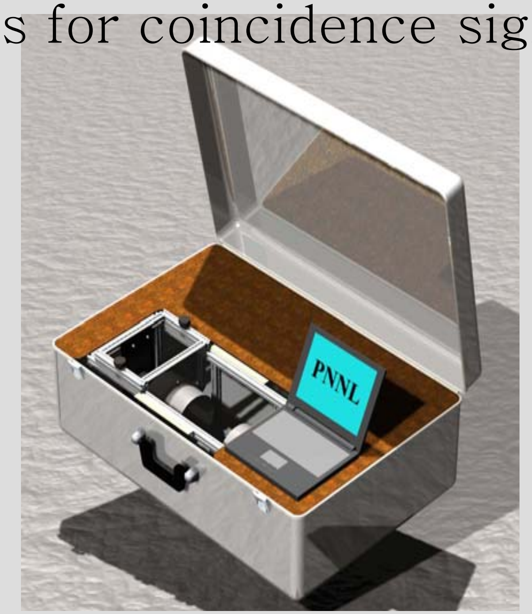
Rapidly Deployable Sensor Networks

- Need for effective yet operationally acceptable sensor to detect threats in inbound cargo containers
  - Explore commercial software and hardware capabilities
  - Develop “ad-hoc” network architecture
  - Develop & demonstrate rapid deployment of sensor network

Portable Radionuclide Analysis System

- Rapid quantification of radionuclides in a variety of sample matrices at or near point of collection.
  - Most radioactive decays have a coincidence signature.
  - Portable β–γ–γ coincidence systems
  - Active shielding methods to reduce ambient background effects.
  - Use ROOT as an analysis tools for coincidence signatures

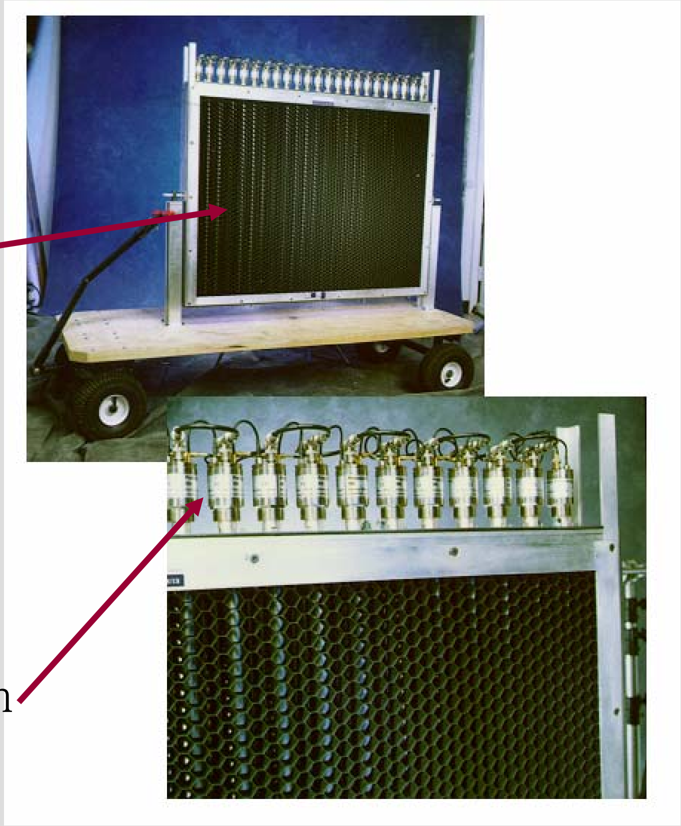




Long Range Neutron Detector

- Most special nuclear materials U, Pu, Am emit high fluxes of neutrons.
- Field teams need
  - A light weight, directional detector for neutron sources at distances of 10 – 100 meters
  - A moderator-free detection method for slow neutrons. Air is the moderator.
  - Theory: build model describing slow neutron transport to long distances
  - Boron shielding and collimation
  - Standard $^3He(n,p)^3H$ neutron detection.

Boron coated aluminum collimator



^{3}He tubes, ~5000 barn cross-section

Advanced Land Mine Detection Method

- Need for a low-cost, portable instrument capable of effective and efficient detection of modern buried land mines.

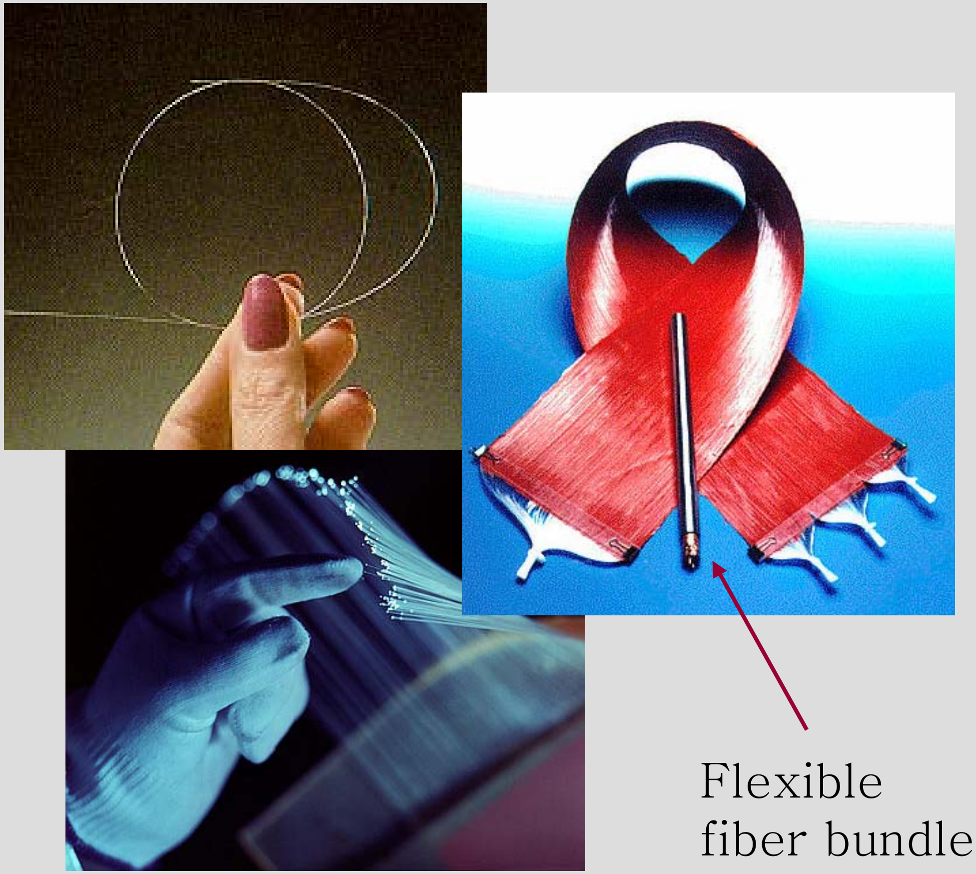
High end weed eater chassis

Tagged *n* source with ^{3}He tube array

- Use timed neutron detection method to rapidly find mines in arid & semi-arid soil (scan rates > 6 m^2/min.)
- Technique shown to work will for real mines (disabled for testing).
- Developed instrument weighs ~5 lbs and costs less than $5K.
- Sensitive to organic and water content of soil, (don't rely on this unit as only technique)

Fiber Optic Development for Sensors

- Specialized optical fibers that are sensitive to neutrons and insensitive to gamma's



Flexible fiber bundle

- Developed scintillating glass formulations that meet neutron detection application requirements
- Use the high neutron cross-section of the ^{6}Li(n, t, α)
- Built a fiber draw tower to fabricated specialized optical fibers from scintillating glass
- Individual fibers have low gamma-ray detection efficiency
- Fibers have been deployed in a number of applications

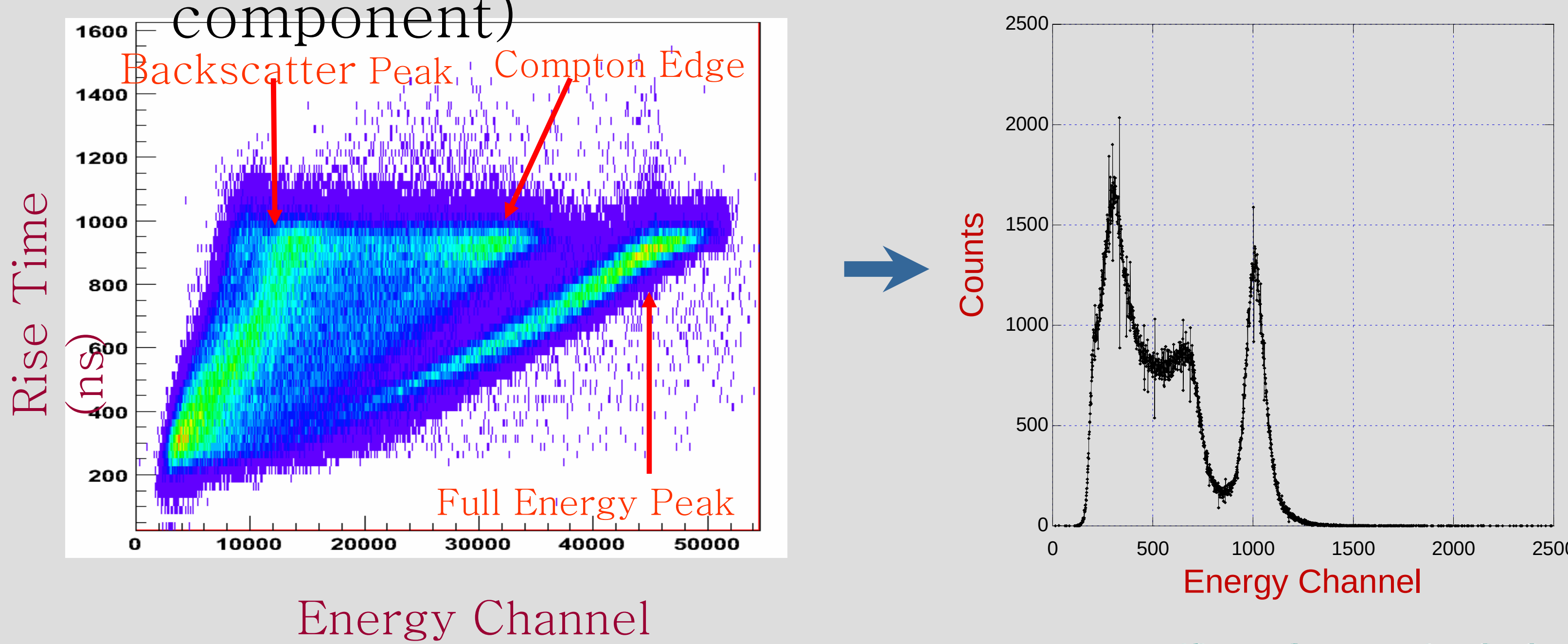
Portable Neutron Spectrometer

- Need field-appropriate instrument that permits measurement / exploitation of neutron energy spectrum.

- Develop, construct, & test field-portable neutron spectrometer.
- Exploit fiber-optic neutron detection medium for novel spectrometer design
- Uses layers of fibers and layers of plastic, and multiplicity counting to discriminate out gamma's
- Can be used for a variety of purposes: Treaty verification, smuggled weapons search, Environmental remediation….

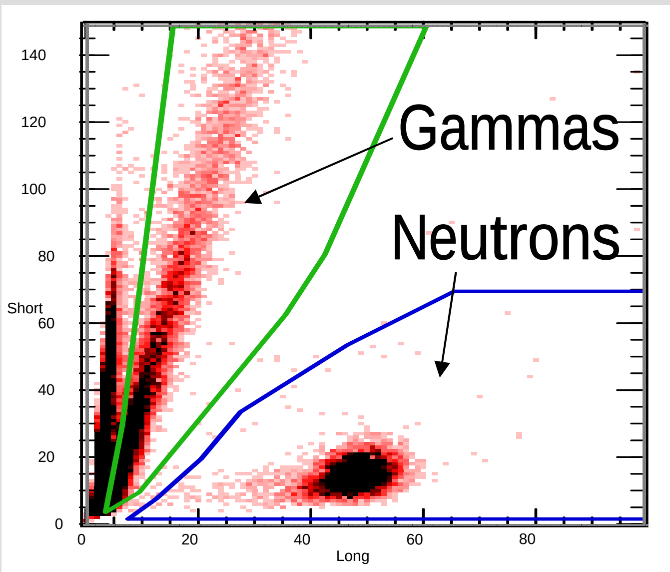
Pulse Shape Processing

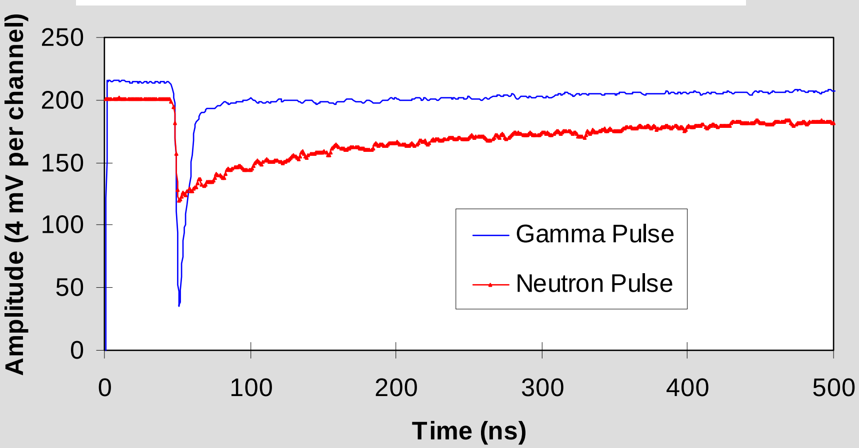
- Use list mode for post data acquisition analysis
  - Energy and time stamps
  - Pulse shape analysis (better resolution)
  - Analysis individual wave forms (dual component)



Dual Optical Component Scintillators

- Need effective γ–ray rejection or neutron energy determination.

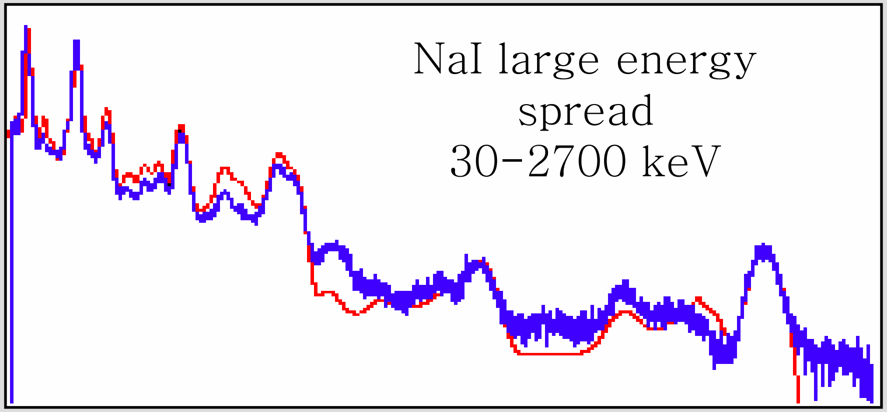




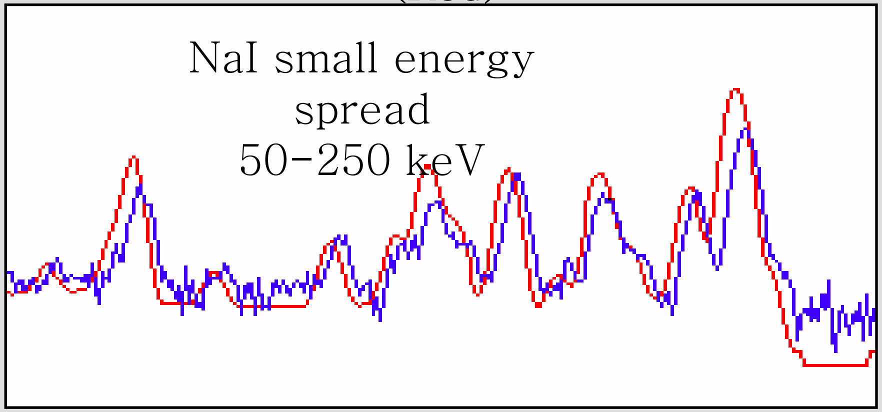
- Explore *n* and γ–detection capability of LiBaF scintillator:
  - a) Determine optical performance,
  - b) Evaluate neutron detection capability,
  - c) Grow larger crystals for practical uses.
- Can uses same analysis techniques for Phoswich detectors to measure coincident β–γ, α–x–ray, etc signatures.

Synthetic Gamma-ray Spectra

- The ability to collect reasonable gamma spectrum for a given source, detector type and geometry aids R&D effort

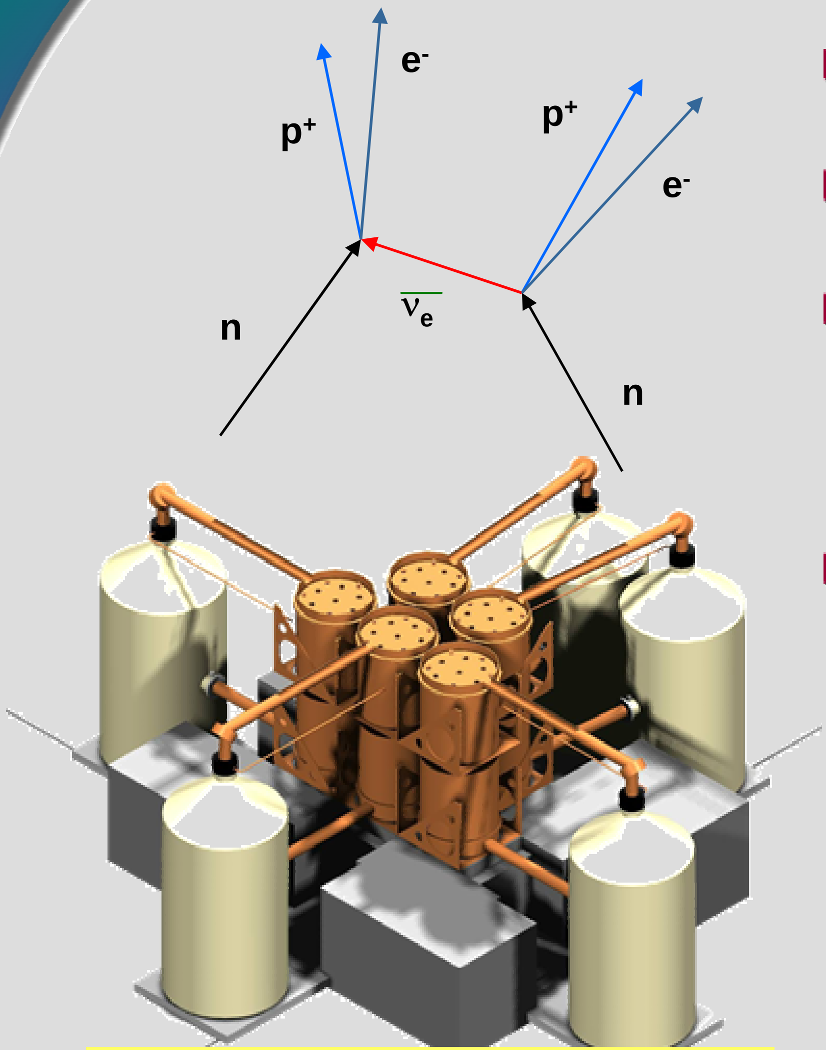


Experimental (Blue) & Predicted Spectra (Red)



- Developed a set of computational algorithms representing the physics of gamma-ray detection
  - NaI, HpGe, CdZnTe, $LaCl_3$
- Develop user-friendly software to provide rapid calculation of synthetic "spectra"
  - Runs on Windows
- Validate predicted spectra
- Commercialized & support software "Synth"

Neutrino Research: Majorana Overview

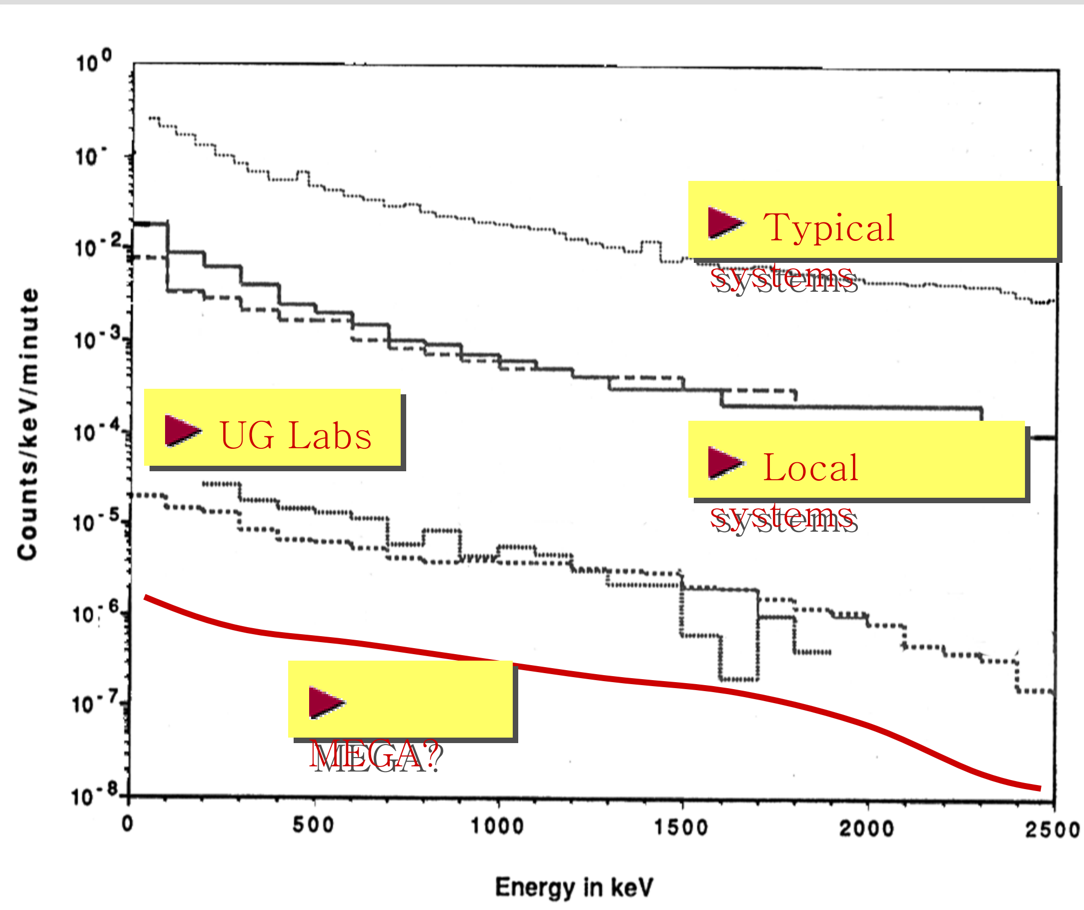


- Reference Configuration

- GOAL: Sensitive to effective Majorana ν mass near 50 meV
- neutrinoless **double** β decay of ^{76}Ge potentially measured at 2039 keV
- Based on well known ^{76}Ge detector technology plus:
  - Pulse-shape analysis
  - Detector segmentation
- Requires:
  - Deep underground location
  - 500 kg enriched 86% ^{76}Ge
  - many crystals, segmentation
  - Pulse shape discrimination
  - Time/Spatial Correlation
  - Special low-background materials

Background comparison of several PNNL systems

- Above ground or shallow underground (100 mwe) sites can be quite useful in extending sensitivity, but limited
- Sensitivities deep underground can be 100x better or more



JRNC, Articles, Vol. 160, No. 2 (1992) 371-385.

Location	Background	Detection Limit
Shallow Underground	0.01	0.1
Well Shielded Surface	0.01	0.1
Deep Underground	0.0001	0.01

- 100 times better Detection Limit!

Low-Background Electroformed Copper

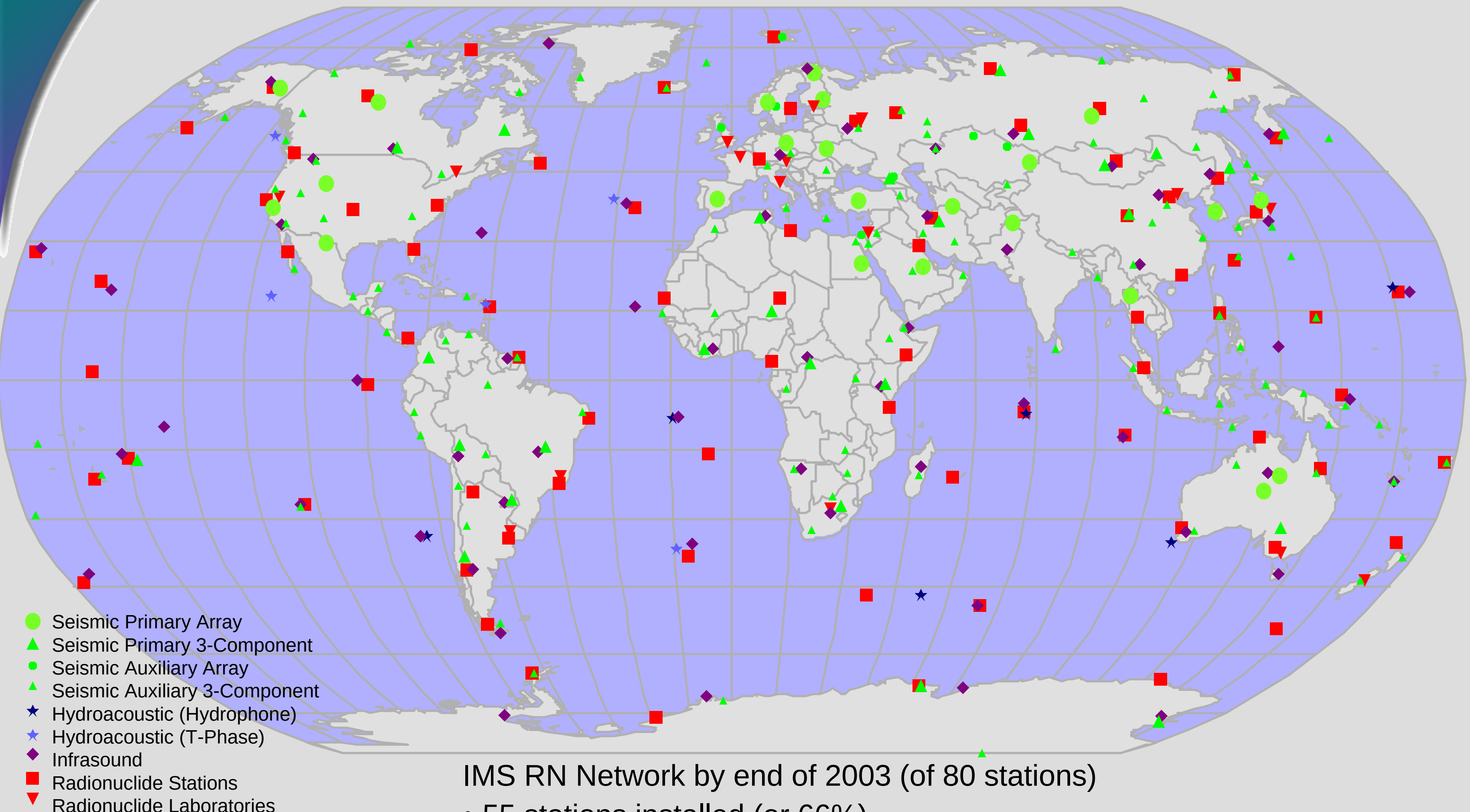
Low-background detector and electroformed cryostat during assembly

- Semiconductor-grade acids
- Glassware-free handling
- Copper sulfate purified by recrystallization
- Baths circulated with continuous microfiltration to remove oxides and precipitates
- Continuous barium scavenge removes radium
- Cover gas in plating tanks reduces oxide formation
- Periodic surface machining during production minimizes dendritic growth

CTBT-IMS: what is it?

- Comprehensive Nuclear-Test-Ban-Treaty (CTBT): eliminate nuclear weapons testing
- International Monitoring System (IMS) will be implemented to produce verification data for the CTBT
- Four types of networks
  - Infrasound, Hydro-acoustic, Seismic
  - Radiation: Particulate, Radioxenon
  - The **A**utomated **R**adioxenon **S**ampler-**A**nalyzer (ARSA) is a radionuclide sampling station developed for application to the IMS

The International Monitoring Sensor Network



IMS RN Network by end of 2003 (of 80 stations)

- 55 stations installed (or 66%)
- 22 stations certified (or 28%)

Production Mechanism

Radionuclides are produced directly from the fission device (fission products) or secondarily (activation products)

- Below ground testing traps most of the fission products.
- Weather carries the escaped radionuclides to the samplers.
- Samplers collection a large amount of air and determine the radioactivity per volume yielding an activity concentration .
- No direct measure of device yield nor device type.

Advantages of Monitoring Radioxenon

- Radioxenon is in mass range with large fission yield
- Several RXe isotopes are produced with convenient half-lives (^{131m}Xe, ^{133m}Xe, ^{133g}Xe, and ^{135}Xe)

fission yields for A=130-135

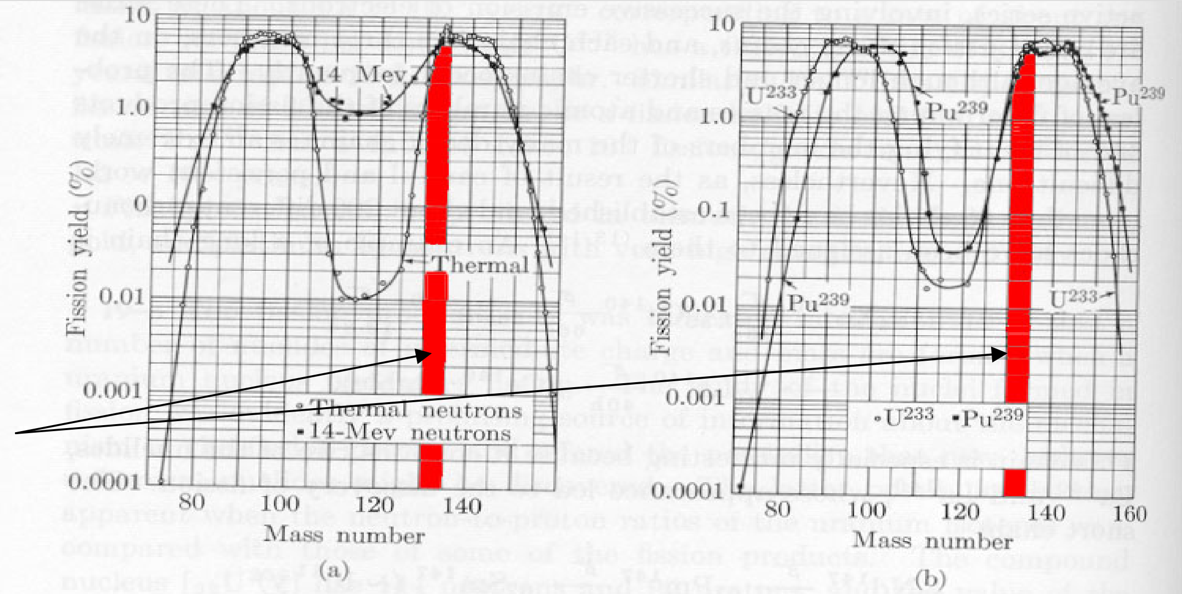


FIG. 19–3. Fission yields from U^{235}, U^{233}, and Pu^{239}: (a) thermal and fast fission of U^{235}; (b) thermal fission of U^{233} and Pu^{239}.[15]

Plot from Kaplan, "Nuclear Physics" (1963)

Advantages of Radioxenon Monitoring II

- Xenon is an inert noble gas, likely to escape even from an underground explosion
- Doesn't combine with other gases in atmosphere (no chemical effects)
- Easy to extract from air using traps at moderate temperatures (~163K)
- Dominant background: radon, easily removed with proper processing

Automated Radioxenon Sampler/Analyzer

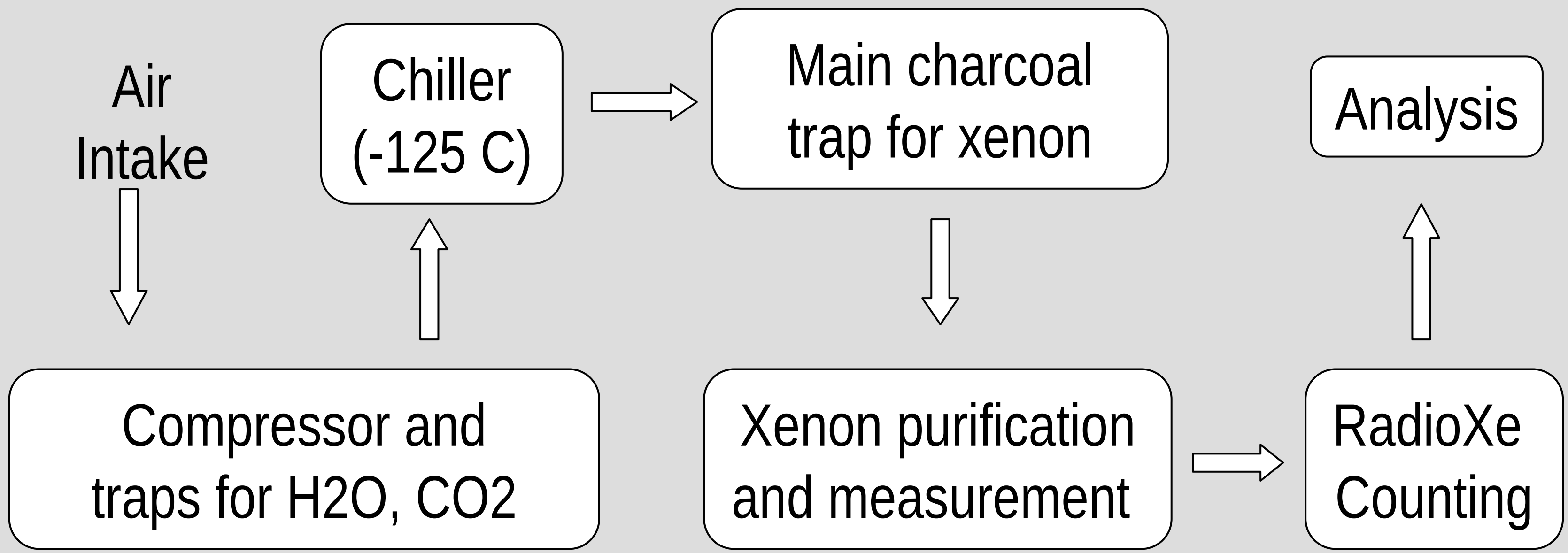
- **Separates and concentrates Xe from 87 ppb to 65 % in a 6.6 cc volume**
- **Highest system sensitivity ever produced ~100 μBq/m^3**
- **Only system capable of measuring ^{135}Xe in the environment**
- **Fully automated, automatic data transfer and high throughput 48 m^3/ 8 hours - 144 m^3/ day**

General Specifications of the ARSA

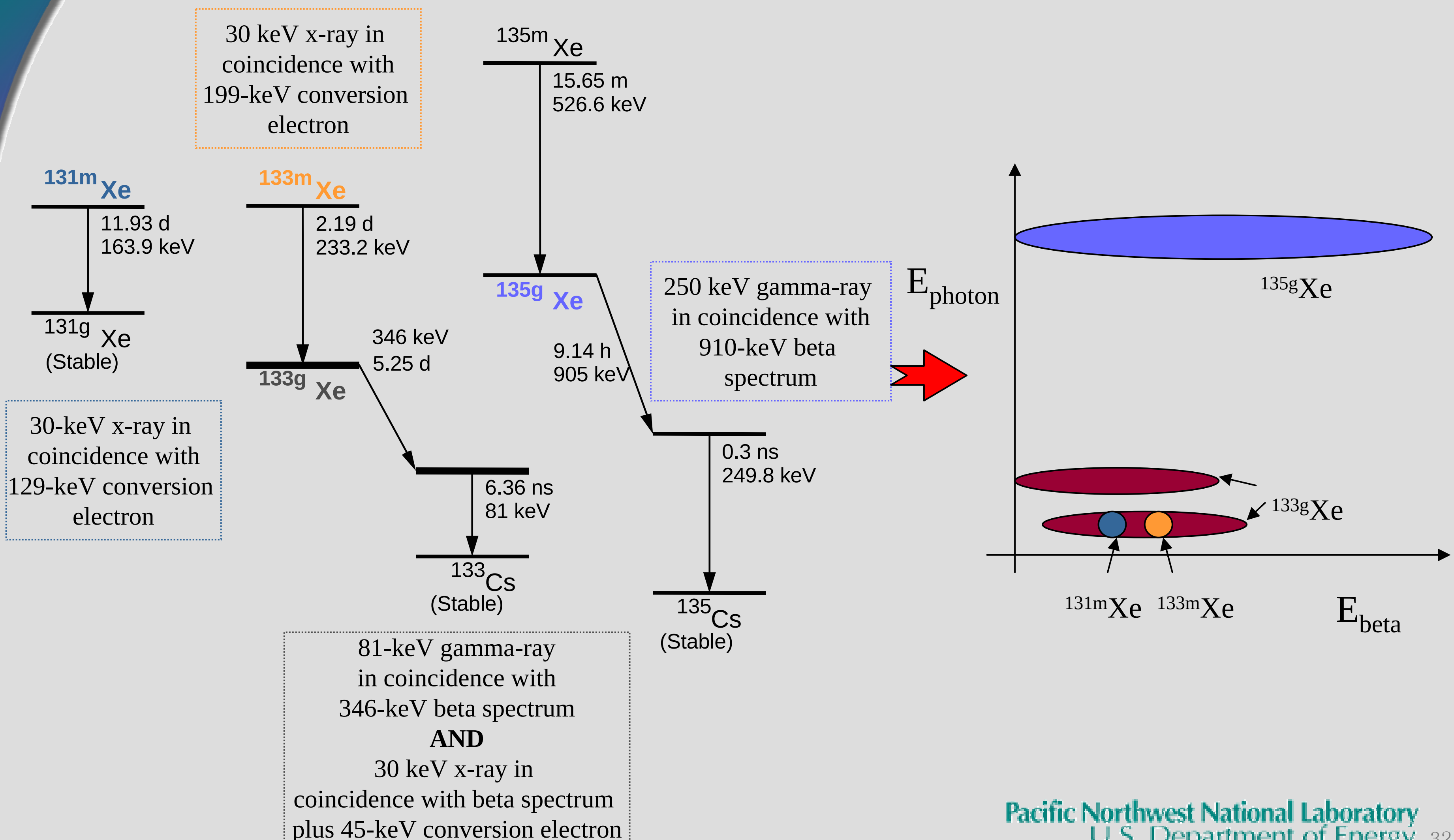
- Automatic collection, purification, and analysis of radioactive xenon from the atmosphere
  - Remotely controllable over the internet or modem (or GCI)
  - Self-monitoring software
  - Analysis software included
- Radioxenon concentration measured every 8 hours
- Detection sensitivity for ^{133}Xe better (<0.1 mBq/m^3) than CTBT IMS specifications
  - Sensitive detection of ^{131m}Xe, ^{133m}Xe and ^{135}Xe

How does the ARSA Work?

- Collect air, separate, purify and measure xenon, count RXe decays, analyze isotope concentration

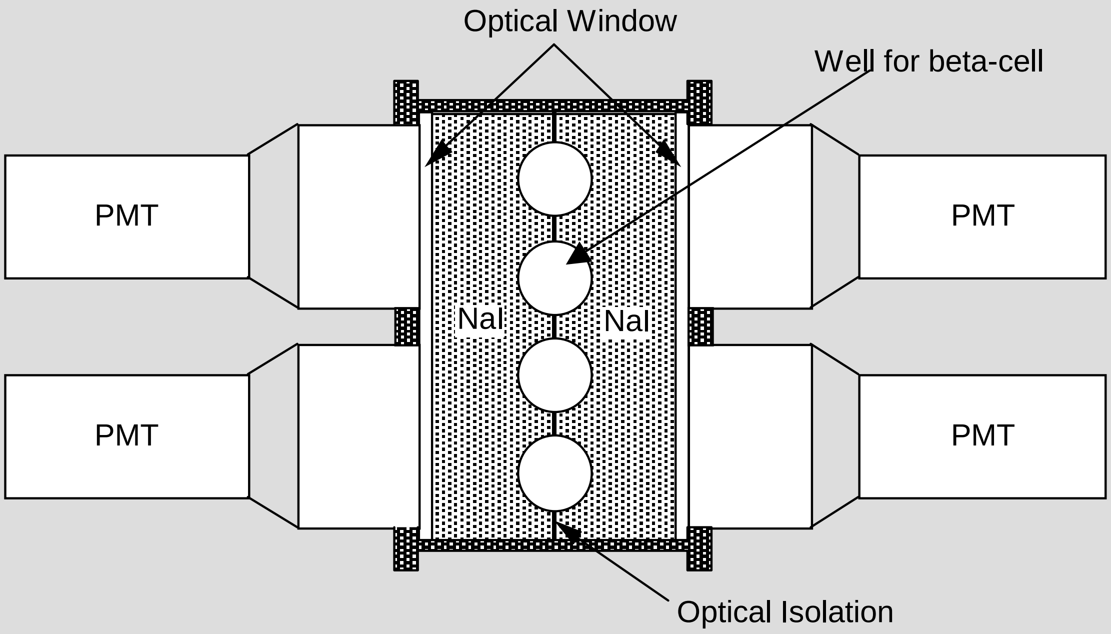


Radio Xenon Gamma/Beta Signatures

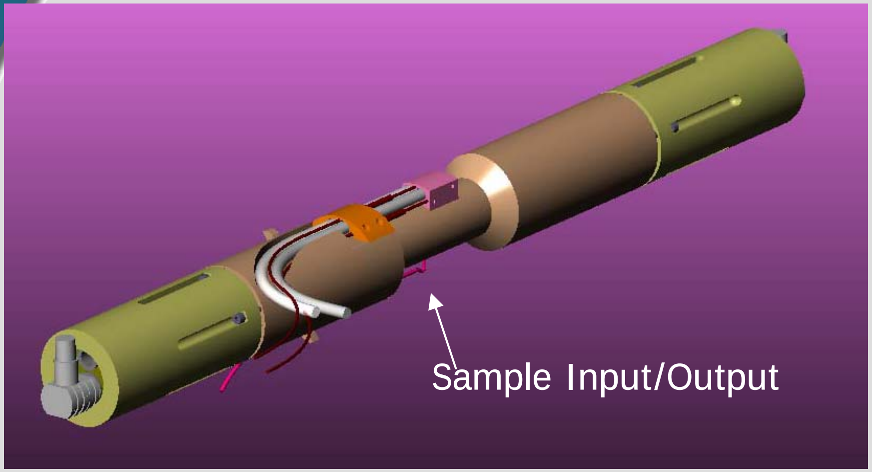


β–γ Spectrometer: NaI(Tl)

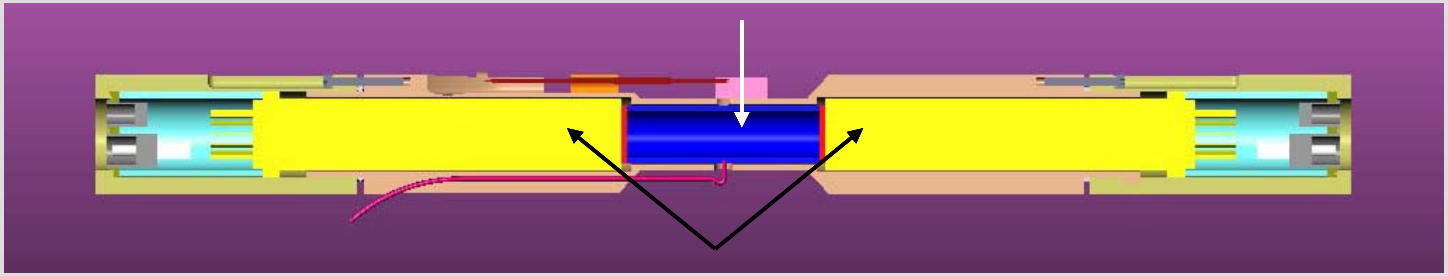
- NaI(Tl) detector with 4 wells
  - NaI(Tl) crystal is nominally 1” thick
  - Two crystals optically separated
  - Each crystal viewed with two 3” PMT’s
  - Size:
    - 18 cm tall
    - 35 cm wide
    - 10 cm thick



Beta-Cell

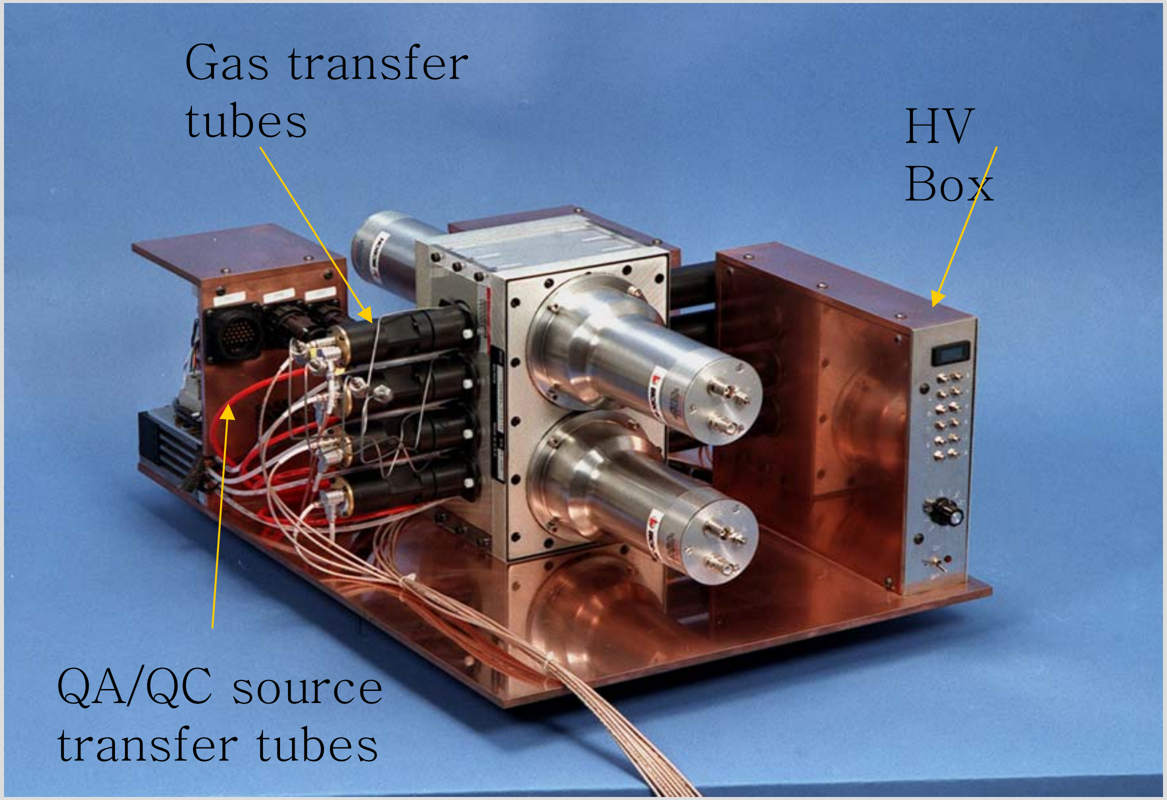


- Beta-cells made of plastic scintillator, 4π geometry for β's
  - External sources provide QA/QC checks
  - Size:
    - 1 cm right cylinder
    - 5 cm long
    - Wall 1.2 mm thick



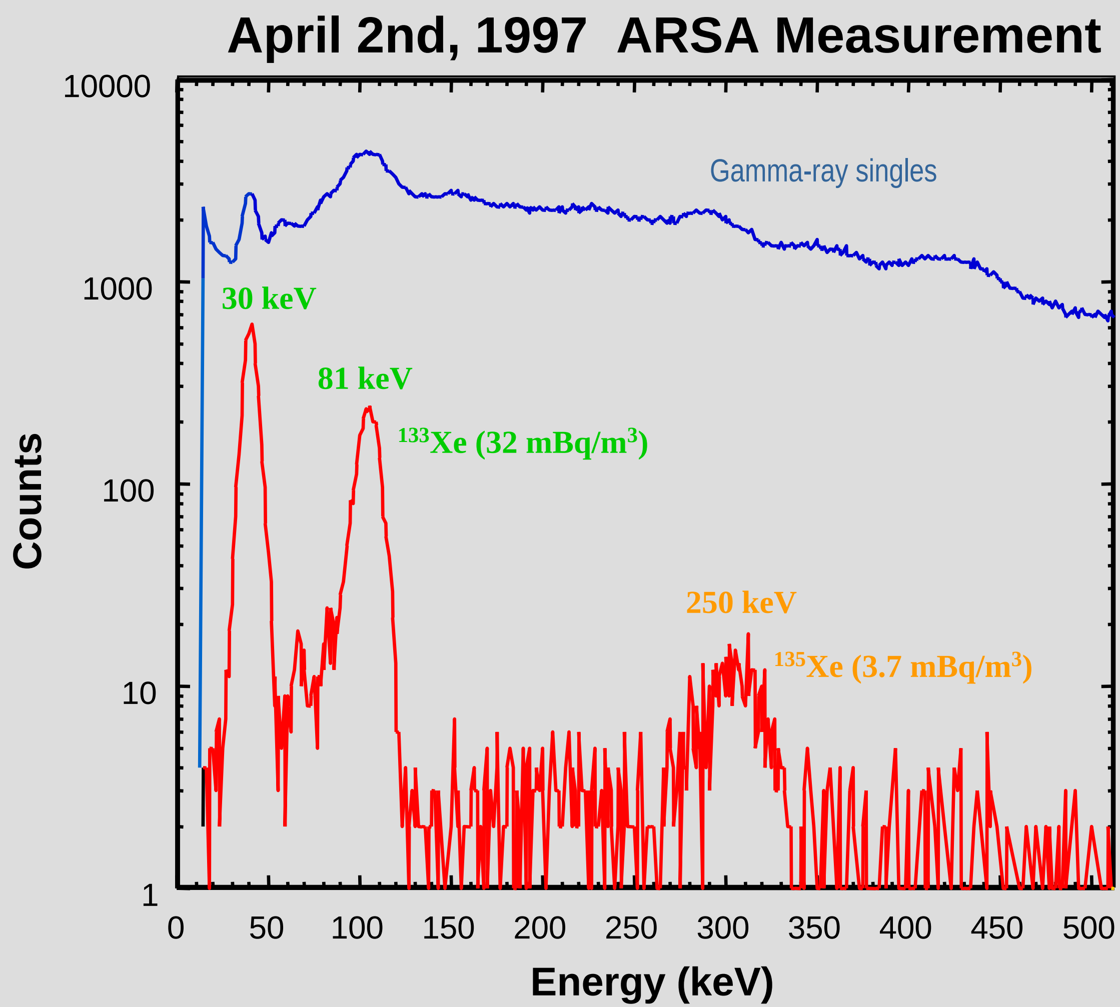
ARSA : Detector Basics

- Entire assembly enclosed in copper cave (5 mm thick) surrounded by 5 cm of lead.



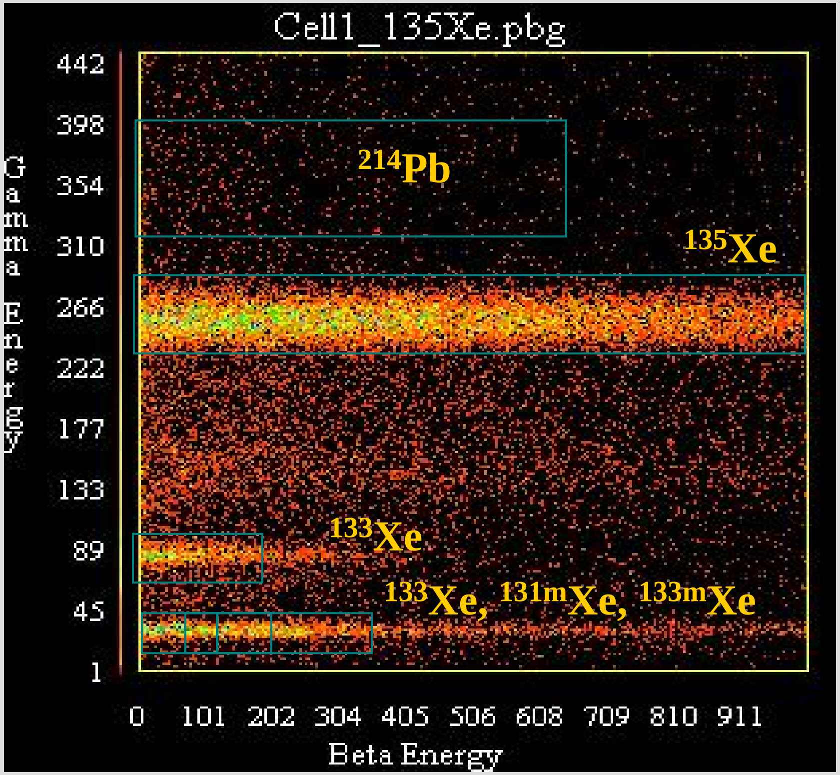
Background Suppression

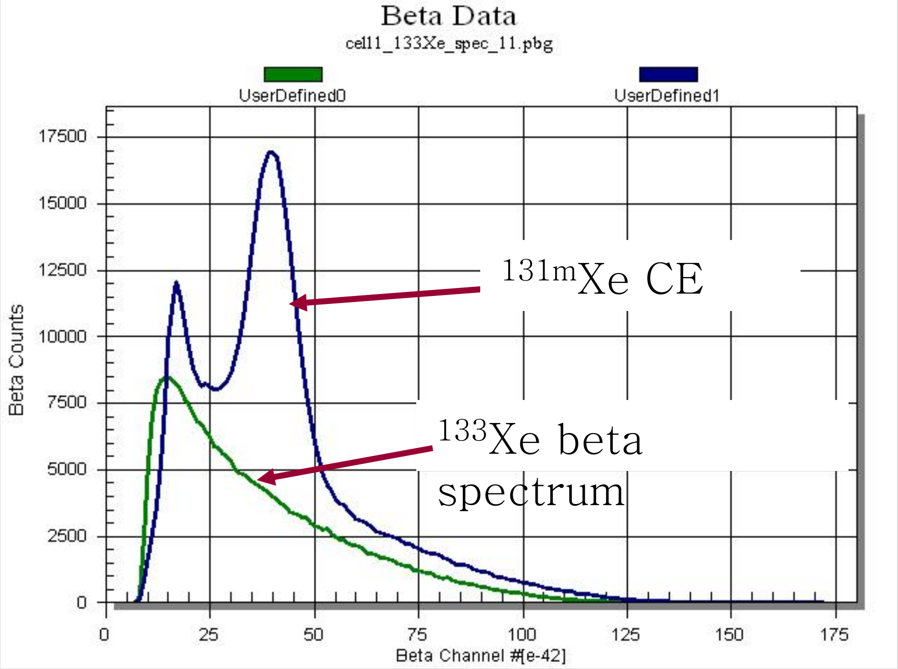
- ARSA: data from EML field test in NYC
- Background is reduced with coincident detection of both the photon and electron



Two Dimensional Histogram

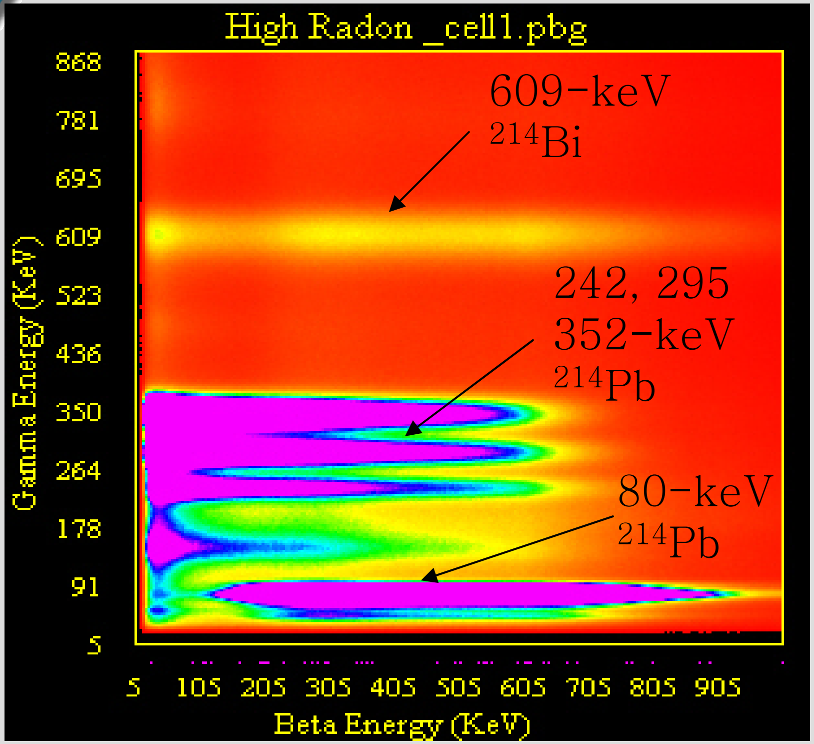
- 2-D Plot: γ vs. β pulse height
- Radioxenon isotopes inhabit well-defined regions (plot)

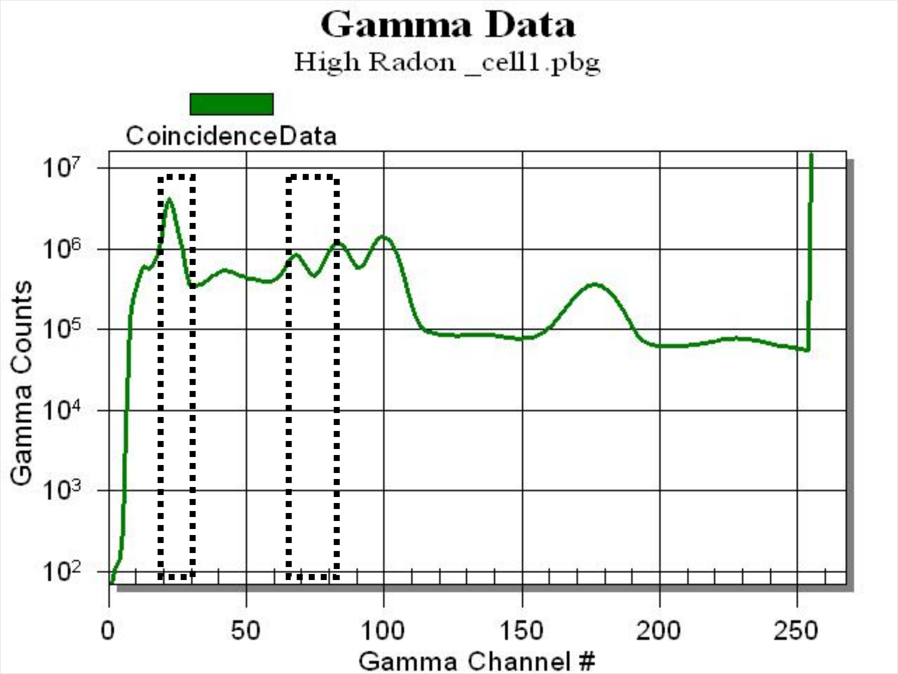




- Beta Resolution clearly shows ^{131m}Xe (gate on the 30-keV and 80-keV region

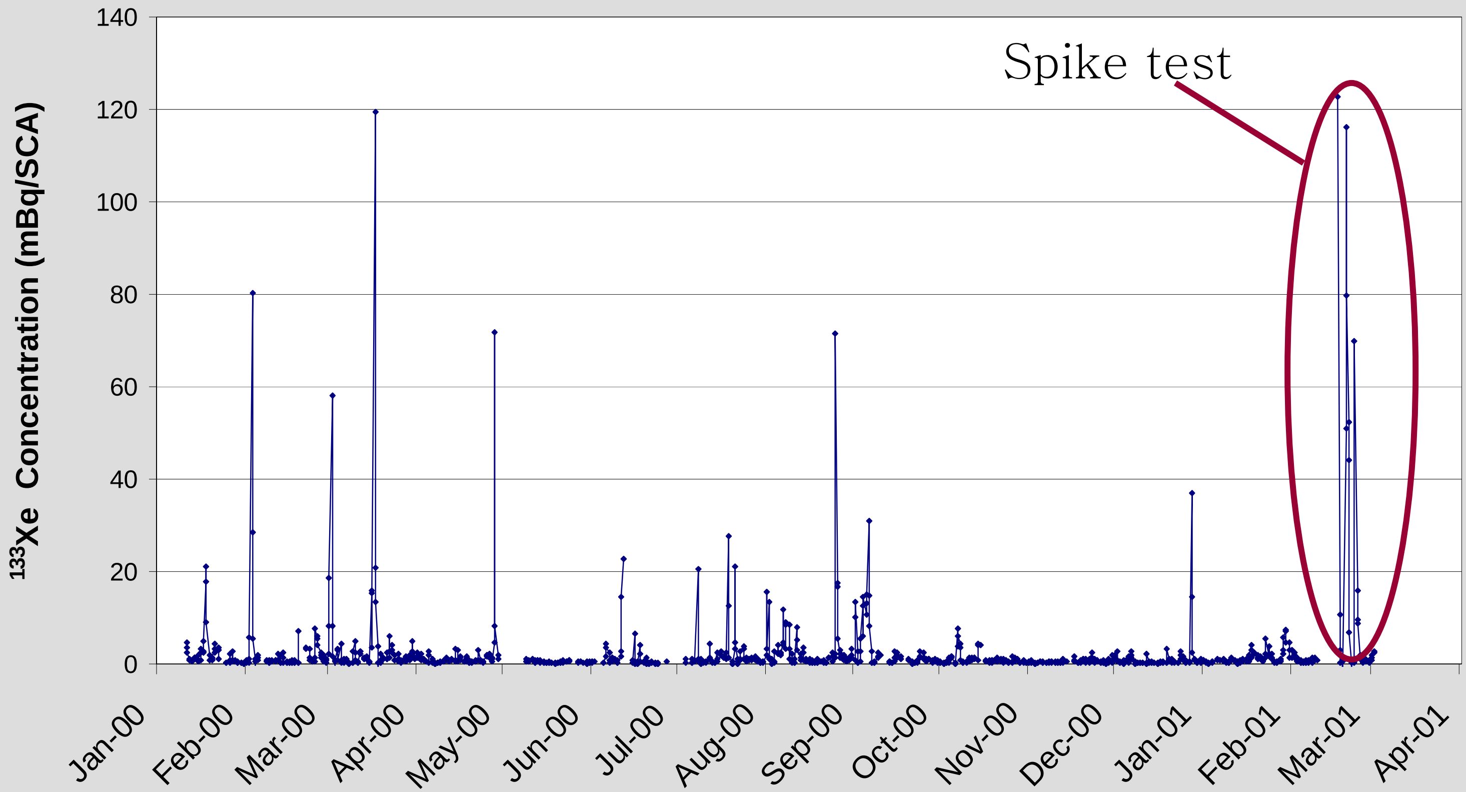
Radon Interference





- $\beta-\gamma$ Energy spectrum from a radon spike
  - 3 α's, several $\beta-\gamma$ coincidences.
- $^{222}Rn \rightarrow {}^{218}Po \rightarrow {}^{214}Pb \rightarrow {}^{214}Bi \rightarrow {}^{214}Po \rightarrow {}^{210}Pb \rightarrow {}^{210}Bi \rightarrow \cdots$.

^{133}Xe Concentrations in Freiburg Germany



- Measured over 1100 samples,
- High concentrations from nuclear reactors or Hospitals

The Commercial ARSA System

- Commercial ARSA will be available soon
  - Produced by DME in Florida, selling price expected near $650K
  - Taking data in Guang Zhou China since 2001
  - First production units available sometime in 2004

Summary

- Lots more work not shown.
  - Modeling
  - Environmental monitoring
  - Hanford cleanup
  - International policy
  - IAEA work
  - Etc. …..
- Applied Nuclear Research alive and well in the Pacific Northwest.