



# Radiochemistry Webinars

## Uranium Resources



*In Cooperation with our University Partners*



UNIVERSITY of CALIFORNIA • IRVINE



## Meet the Presenter...

*Dr. Lindsay Shuller-Nickles*

Dr. Lindsay Shuller-Nickles is an Assistant Professor in Environmental Engineering and Earth Science at Clemson University. She teaches undergraduate courses on subjects of mineralogy, petrology, the nuclear fuel cycle, and nuclear waste management and graduate courses on nuclear environmental engineering, technical nuclear forensics, and applications of quantum-mechanical modeling in environmental science. She received her Ph.D. in Materials Science and Engineering from the University of Michigan working with Rod Ewing and Udo Becker. Dr. Shuller-Nickles' research integrates computational and experimental tools to gain a fundamental understanding of the behavior of radionuclide-containing materials in the environment. She currently supports three undergraduate students, five graduate students, and one post-doc working on two funded projects. The first, funded by the Department of Homeland Security, supports her research in nuclear forensics of the characterization of pre- and post-detonation solid materials. The second is an EPSCoR Implementation grant, which funds Dr. Shuller-Nickles' group as part of a much larger project (~\$5M for three years). Her work on the EPSCoR grant is focused on quantum-mechanical calculations to understand cation ordering, waste loading, and phase stability for advanced ceramic waste forms.



**Contact Information: [lshulle@clemson.edu](mailto:lshulle@clemson.edu)**



# Uranium Resources

Dr. Lindsay Shuller-Nickles  
Clemson University

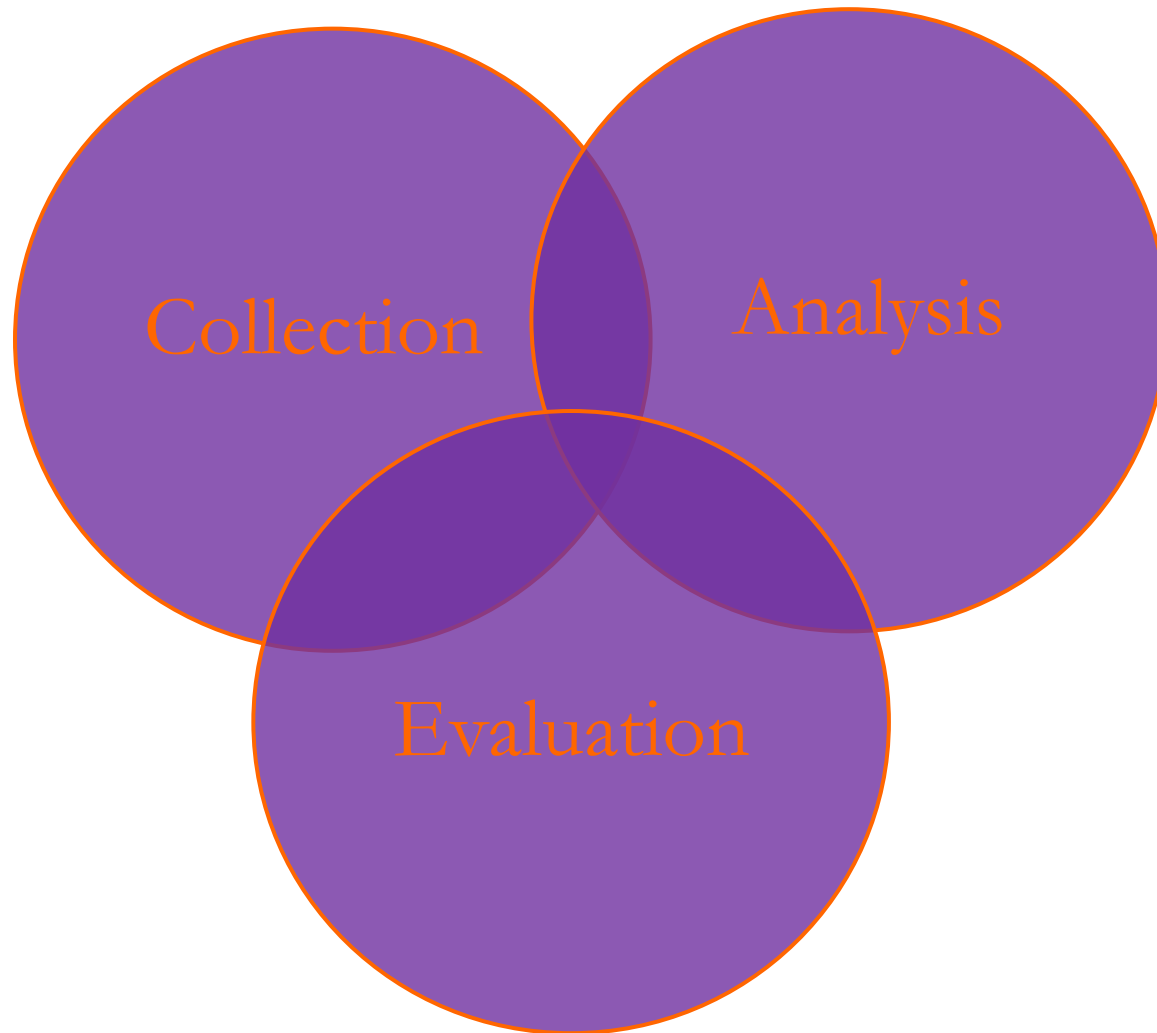


**National Analytical Management Program (NAMP)**  
**U.S. Department of Energy Carlsbad Field Office**

TRAINING AND EDUCATION SUBCOMMITTEE



# What is Nuclear Forensics?



# Nuclear Forensics is ...

- The collection, analysis, and evaluation of pre-detonation (intact) and post-detonation (exploded) radiological or nuclear material, devices, and debris, as well as the immediate effects created by a nuclear detonation.

- National Technical Nuclear  
Forensics Center (NTNFC)

# President Barack Obama, National War College, December 3, 2012

*“There’s still much **too much material** — **nuclear, chemical, biological** — being stored without enough protection. There are still terrorists and criminal gangs doing everything they can to get their hands on it. And make no mistake, if they get it, they will use it; potentially killing hundreds of thousands of innocent people, perhaps triggering a global crisis.”*

# What nuclear material is included?

- Focus today: Special Nuclear Material
- Others:
  - Radiological material (medical isotopes, short/med-lived isotopes, sealed sources)
  - Component materials (casings, containers)
  - Associated materials (boosters, heavy water)

## Special Nuclear Material is ...

(1) **plutonium, uranium** enriched in the isotope **233** or in the isotope **235**, and any other material which the Commission, pursuant to the provisions of section 51, determines to be special nuclear material, but does not include source material; or (2) any material artificially enriched by any of the foregoing, but does *not include source material*.



*Source material* is...(1) Uranium or thorium, or any combination thereof, in any physical or chemical form or (2) ores which contain by weight one-twentieth of one percent (0.05%) or more of: (i) Uranium, (ii) thorium or (iii) any combination thereof. Source material does not include special nuclear material.

Title 1; Atomic Energy Act of 1954

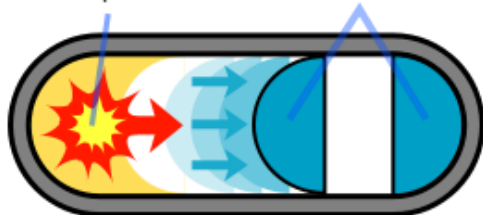


# Why do we care about SNM?

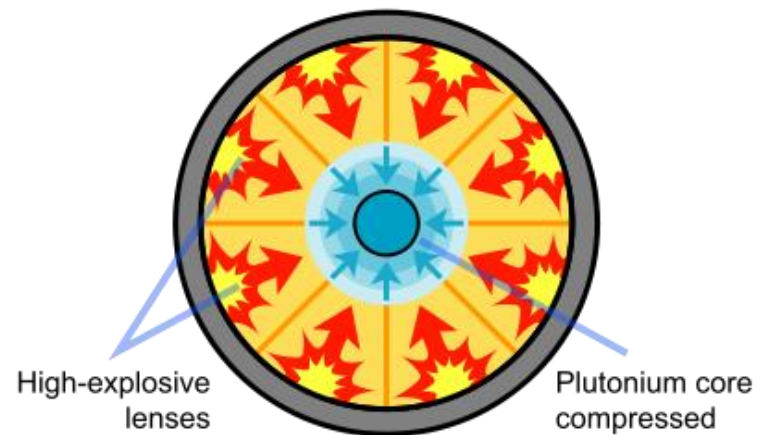
## Approximate Critical Masses of SNM

Material	Bare, Isolated Sphere (kg)	Fully Tamped (Reflected) Sphere (kg)
$^{235}\text{U}$	52	17
$^{239}\text{Pu}$	10-16	4-6
$^{233}\text{U}$	15	6

Conventional chemical explosive      Sub-critical pieces of uranium-235 combined



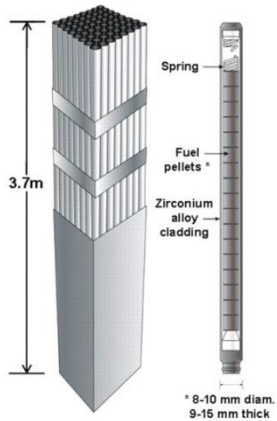
**Gun-type assembly method**



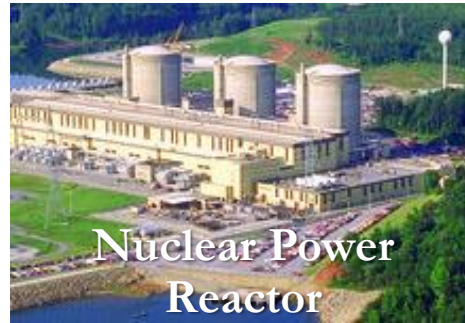
**Implosion assembly method**

# Where does SNM come from?

Fuel Assembly Fuel Rod



Enrichment and Fuel Fabrication

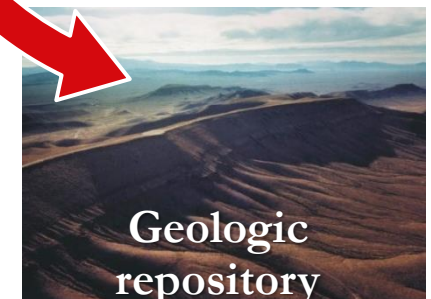
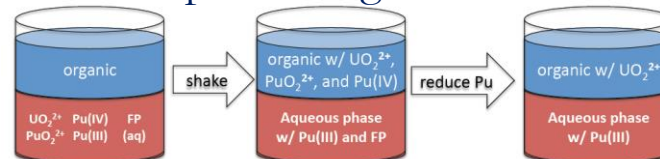


## Nuclear Fuel Cycle

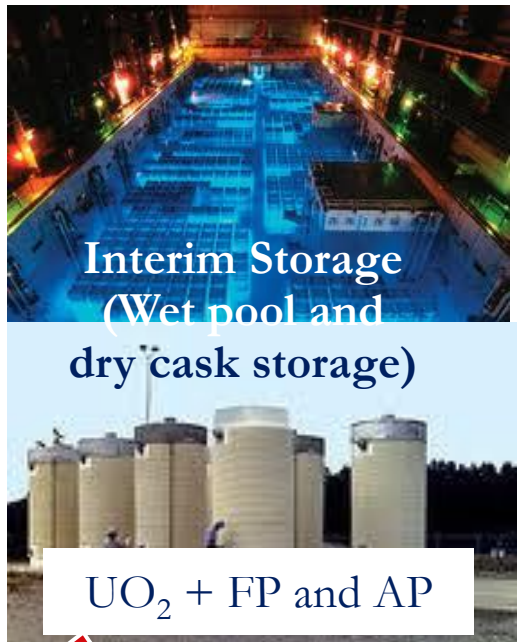
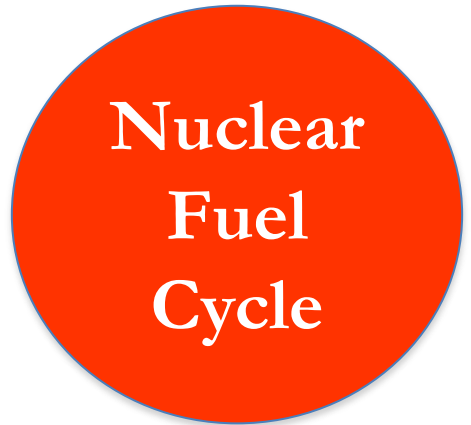
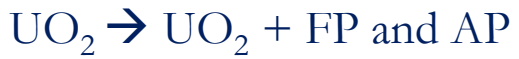
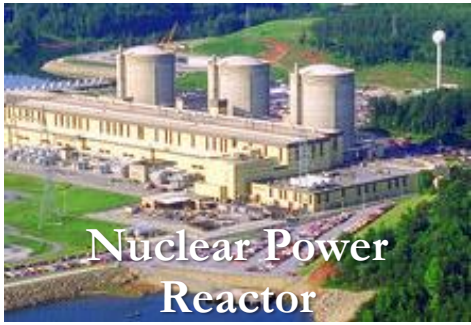
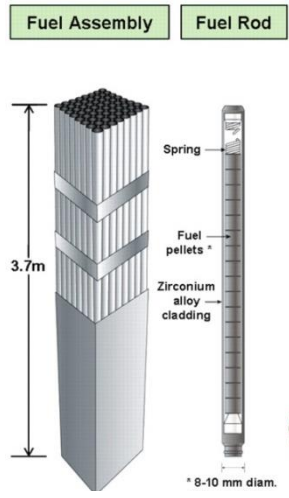


Mining and Milling

### Reprocessing



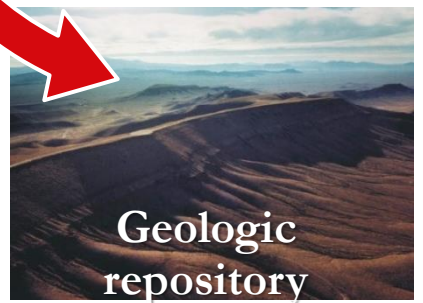
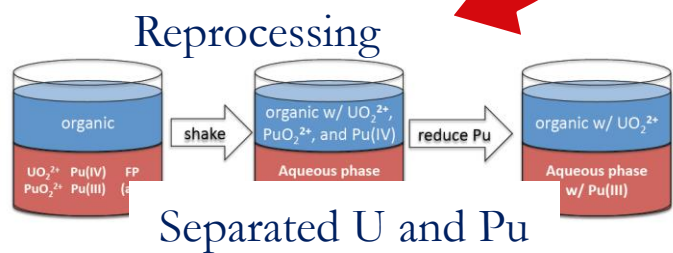
# Where does SNM come from?



$U_3O_8$  to purified  $U_3O_8/UF_6$  to  $UO_2$

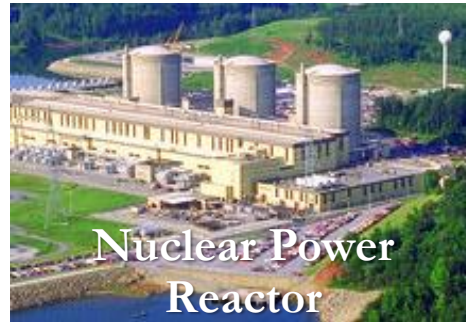
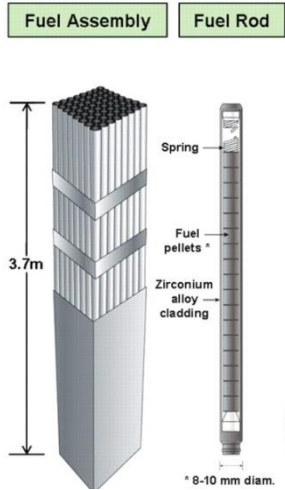


uraninite; betafite; U-rich ore

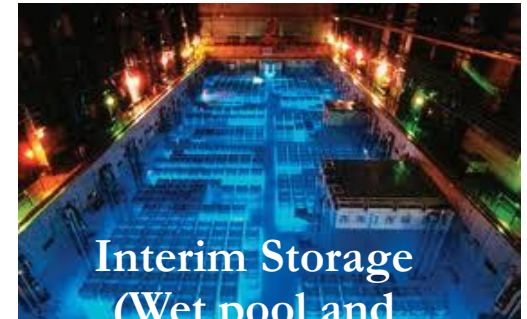




# Where does SNM come from?

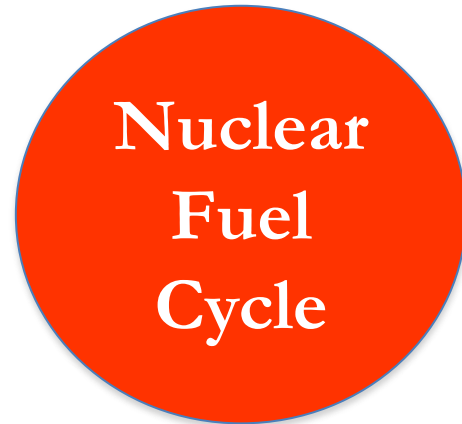


3-5% <sup>235</sup>U



Interim Storage (Wet pool and dry cask storage)

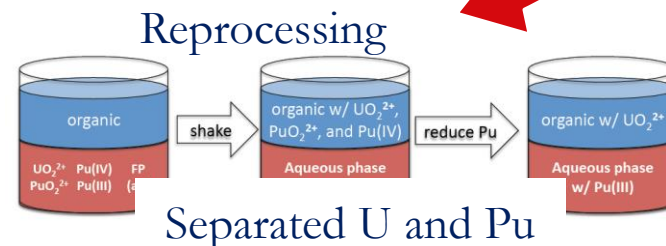
UO<sub>2</sub> + FP and AP



3-5% <sup>235</sup>U



0.72% <sup>235</sup>U



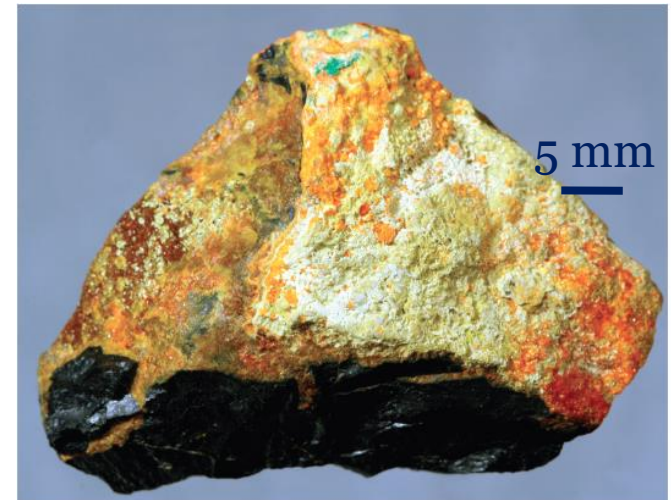
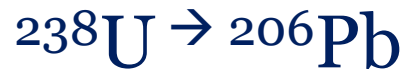
# U and Th mineral evolution

- 
- I. Phase I – concentration of U and Th from uniform trace distribution to enriched magmatic fluids
  - II. Phase II – detrital Th-bearing uraninite and abiotic alteration

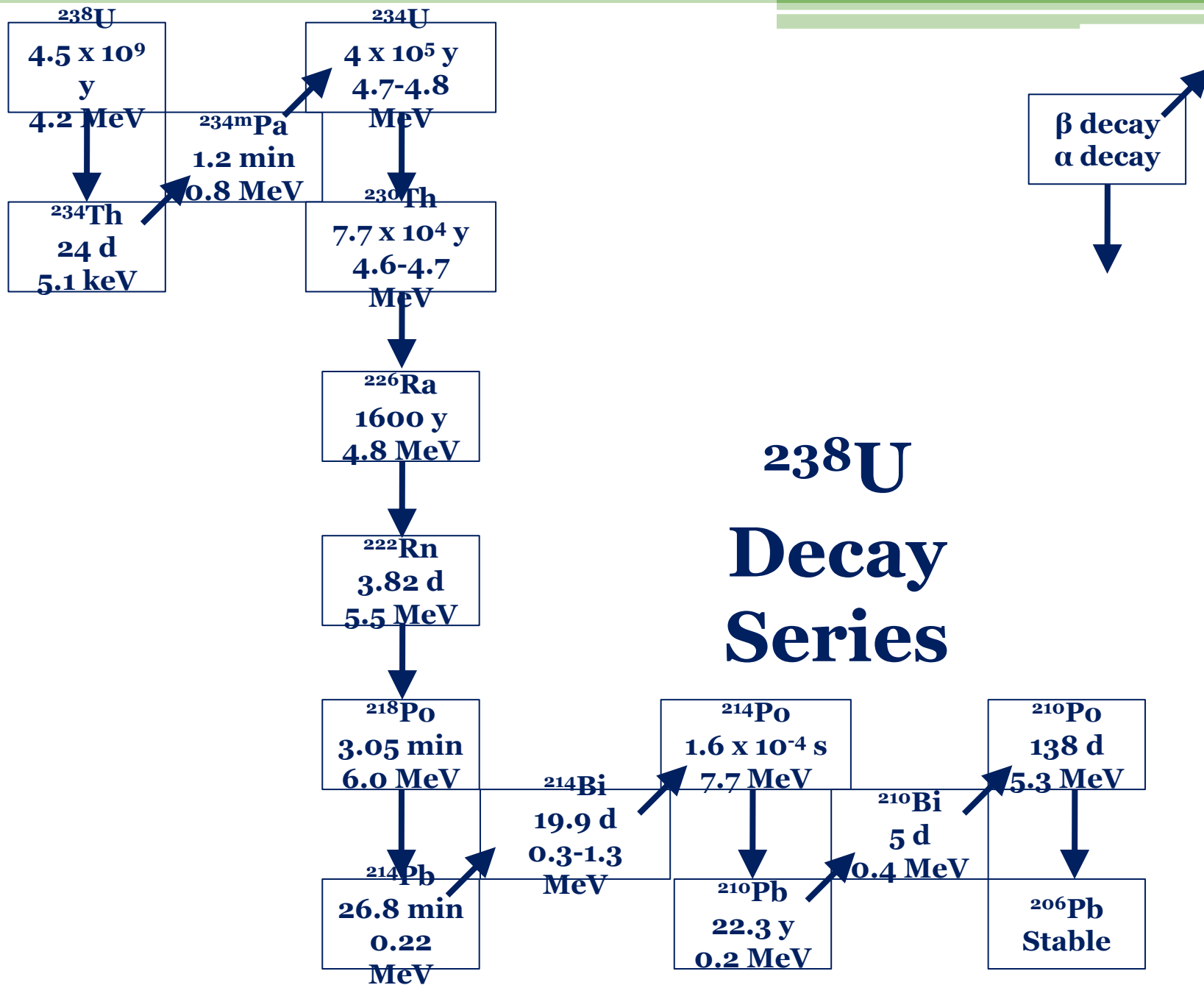
# Phase II Activity - Abiotic Alteration

How might U (and Th) minerals can alter in reducing environments?

1. Radioactive decay



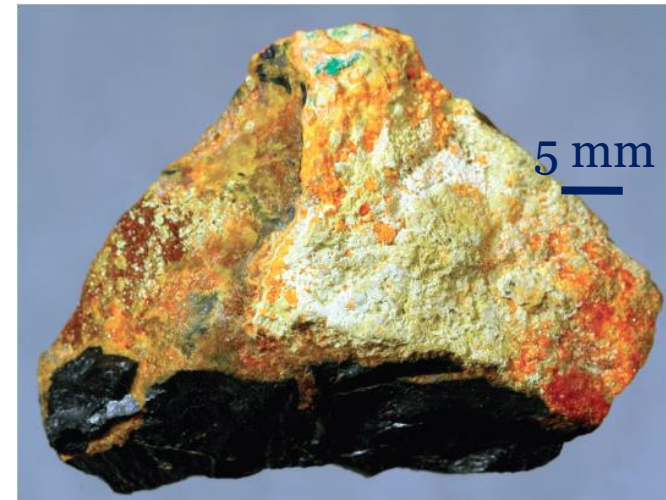
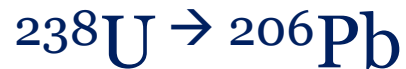
Abiotic alteration leads to formation of Pb-uranyl secondary minerals.



# Phase II Activity - Abiotic Alteration

How might U (and Th) minerals can alter in reducing environments?

## 1. Radioactive decay



Abiotic alteration leads to formation of Pb-uranyl secondary minerals.



## Phase II Activity - Abiotic Alteration

How might U (and Th) minerals can alter in reducing environments?

2. Radiolysis of thin films of water at the mineral surface causes formation of hydrogen peroxide.

uraninite + hydrogen peroxide → studtite



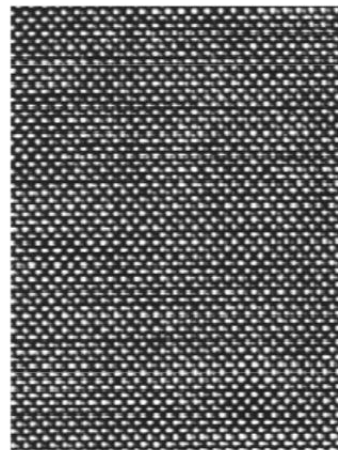
Studtite with fourmarierite on uranophane and uraninite from Shinkolobwe Mine, Katanga, Democratic Republic of Congo

# Phase II Activity - Abiotic Alteration

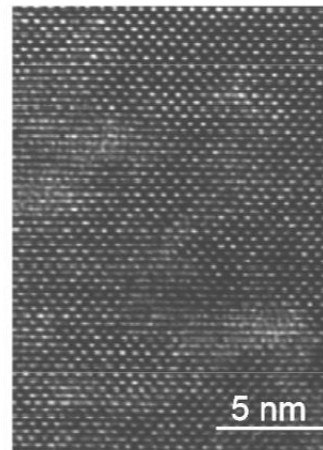
How might U (and Th) minerals can alter in reducing environments?

## 3. Radiation damage from alpha recoil nuclei

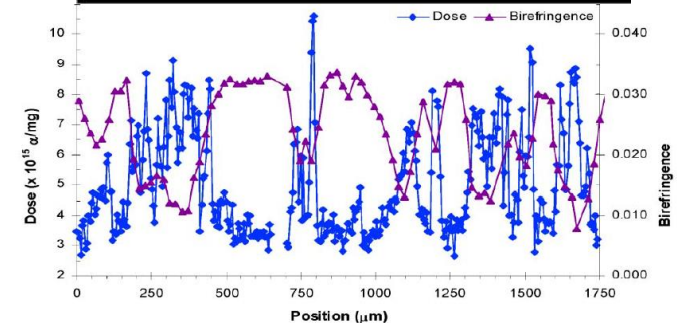
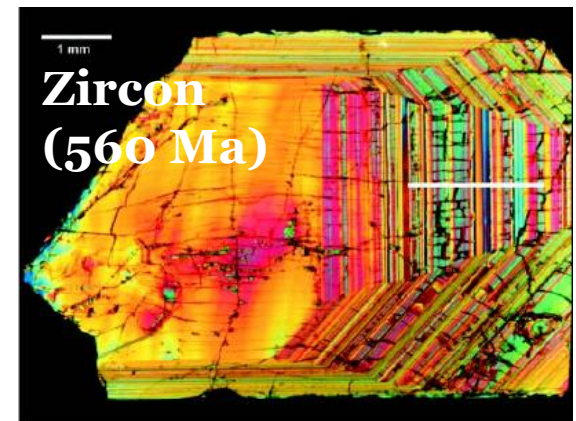
Radiation Damage in Natural Zircon from Alpha Decay



Undamaged  
(no U or Th)




Alpha-Decay Damaged  
Over 550 Million Years  
From U & Th Decay



The inverse relationship  
between dose and birefringence

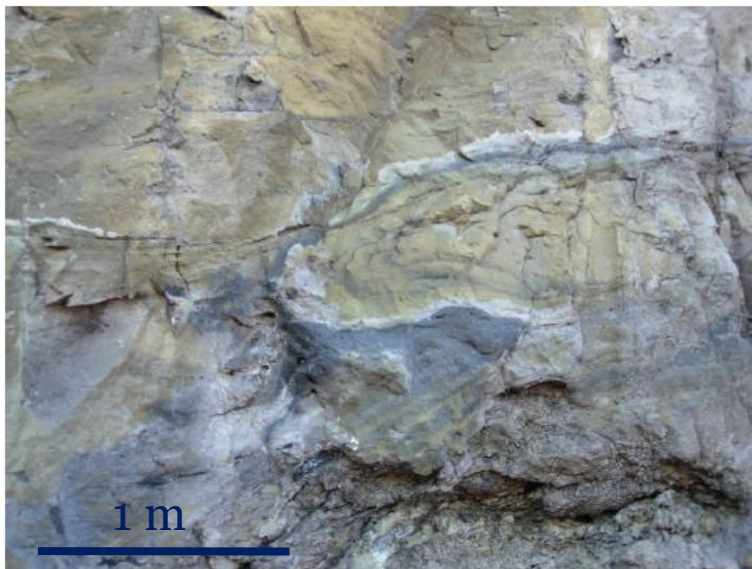
# U and Th mineral evolution

- 
- I. Phase I – concentration of U and Th from uniform trace distribution to enriched magmatic fluids
  - II. Phase II – detrital Th-bearing uraninite and abiotic alteration
  - III. Phase III – result of the Great Oxidation Event (GOE) and biological processes
  - IV. Phase IV – rise of land plants → U(VI) in near-surface waters reduce and precipitate due to organic-rich continental sediments

# U and Th mineralogy - deposits

Decreasing  
Th concentration

- Magmatic (igneous fluids)
- Detrital (sedimentary conglomerates)
- Hydrothermal (e.g., vein-type)



$U^{4+}$  minerals form roll front type ore deposits when they come into contact with reducing, organic material in terrestrial sediments.



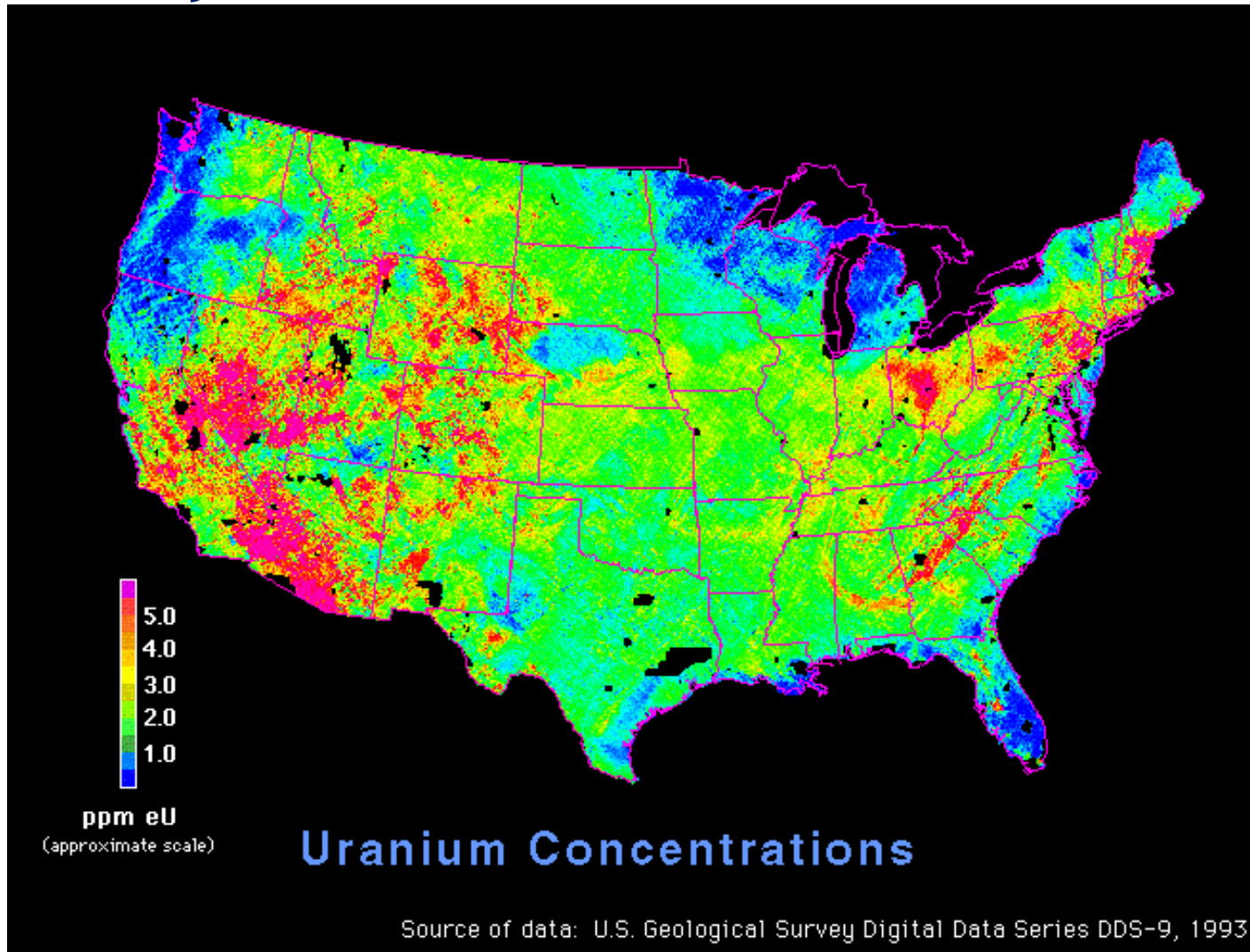
Uraninite crystal with feldspar from the Swamp Mine, Maine.



# Ubiquitous Uranium

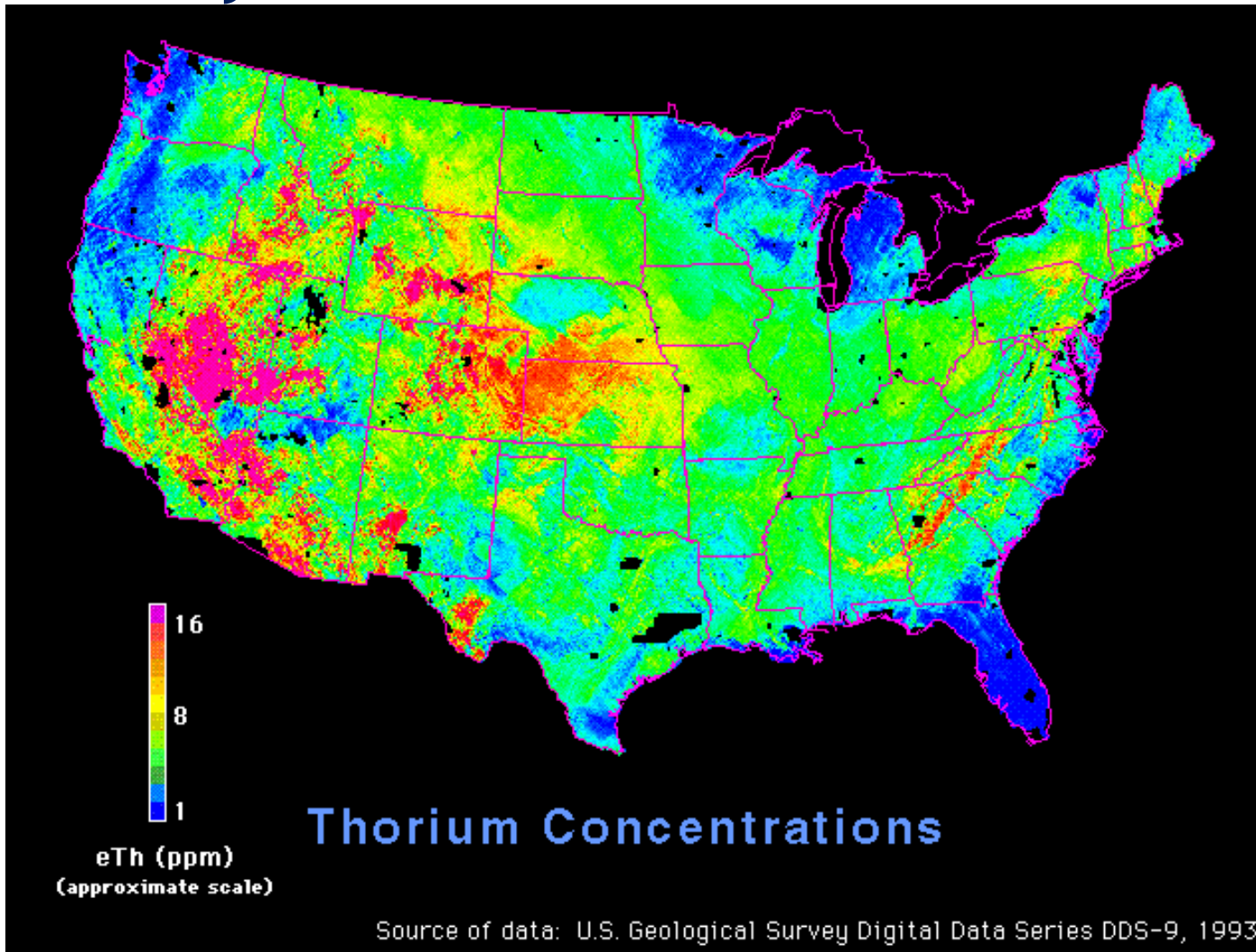
- High Grade Ore >1 wt%
  - Zaire/D.R. of Congo and Canada (12%)
  - Uraninite  $U_3O_8$  and higher oxides, pitchblende
- Medium Grade Ore 0.2-1.0 wt %
  - U.S. (CO, NM, UT, WY), Australia, Czech Republic
  - Carotite, thorianite, phosphate minerals, carbonate minerals
- Low Grade Ore < 0.2%
  - Economical to mine because it is a “byproduct” of another mining operation
  - phosphate in FL and ID
  - gold in South Africa

# Uranium Concentrations from Aerial Gamma-Ray Data



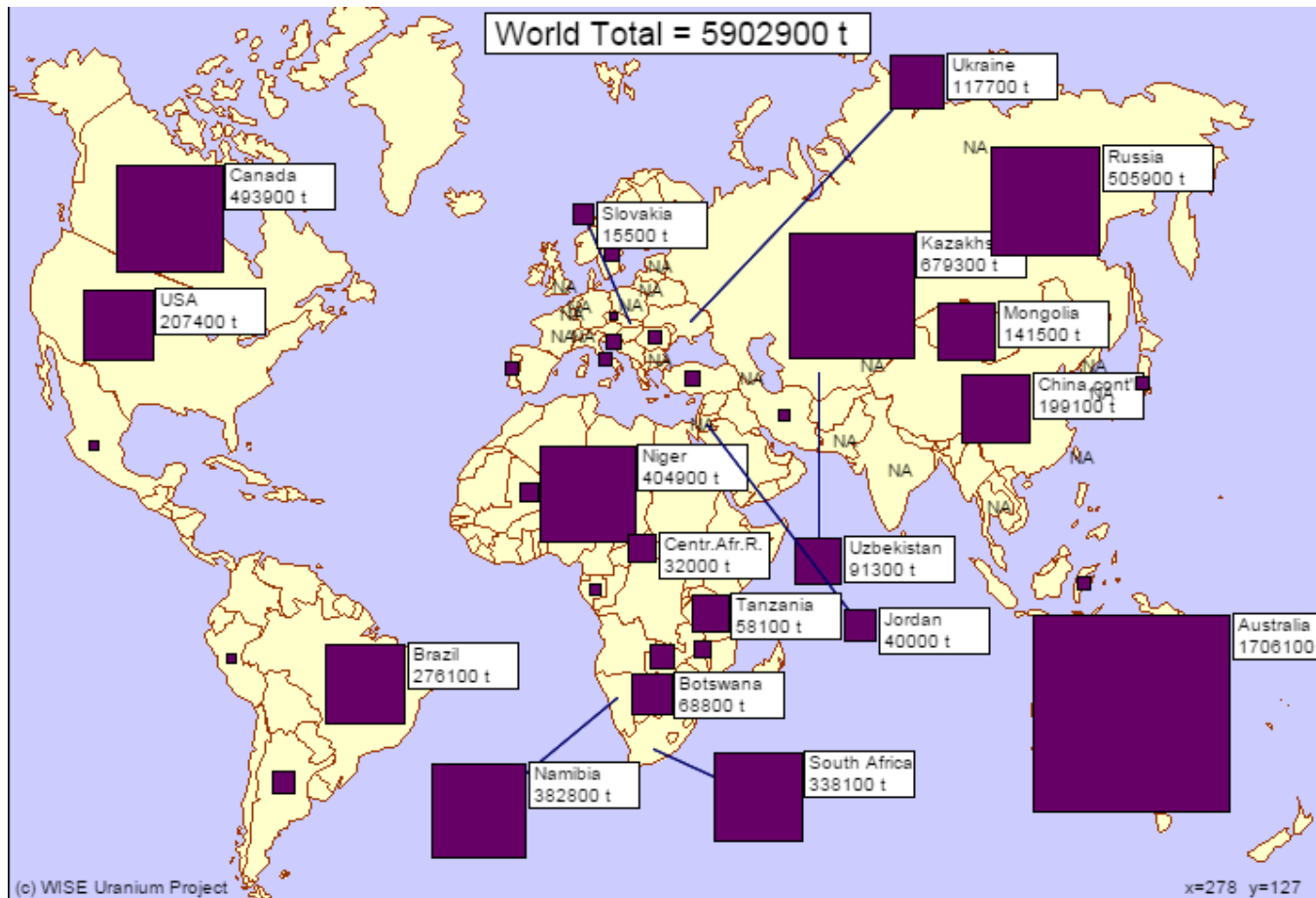
<http://energy.cr.usgs.gov/radon/dds-9.html>

# Thorium Concentrations from Aerial Gamma-Ray Data



<http://energy.cr.usgs.gov/radon/dds-9.html>

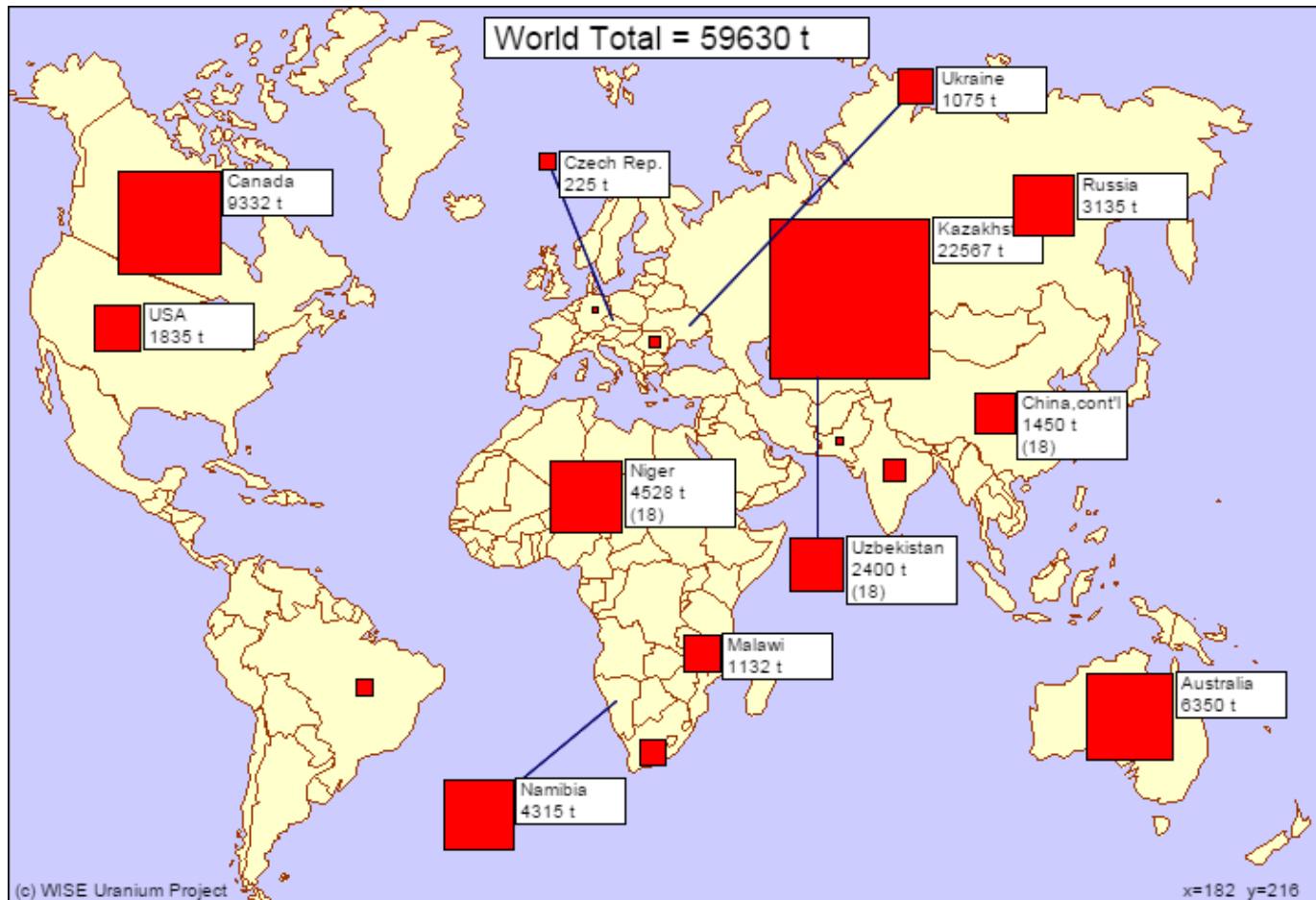
# Identified Uranium Resources (t U)



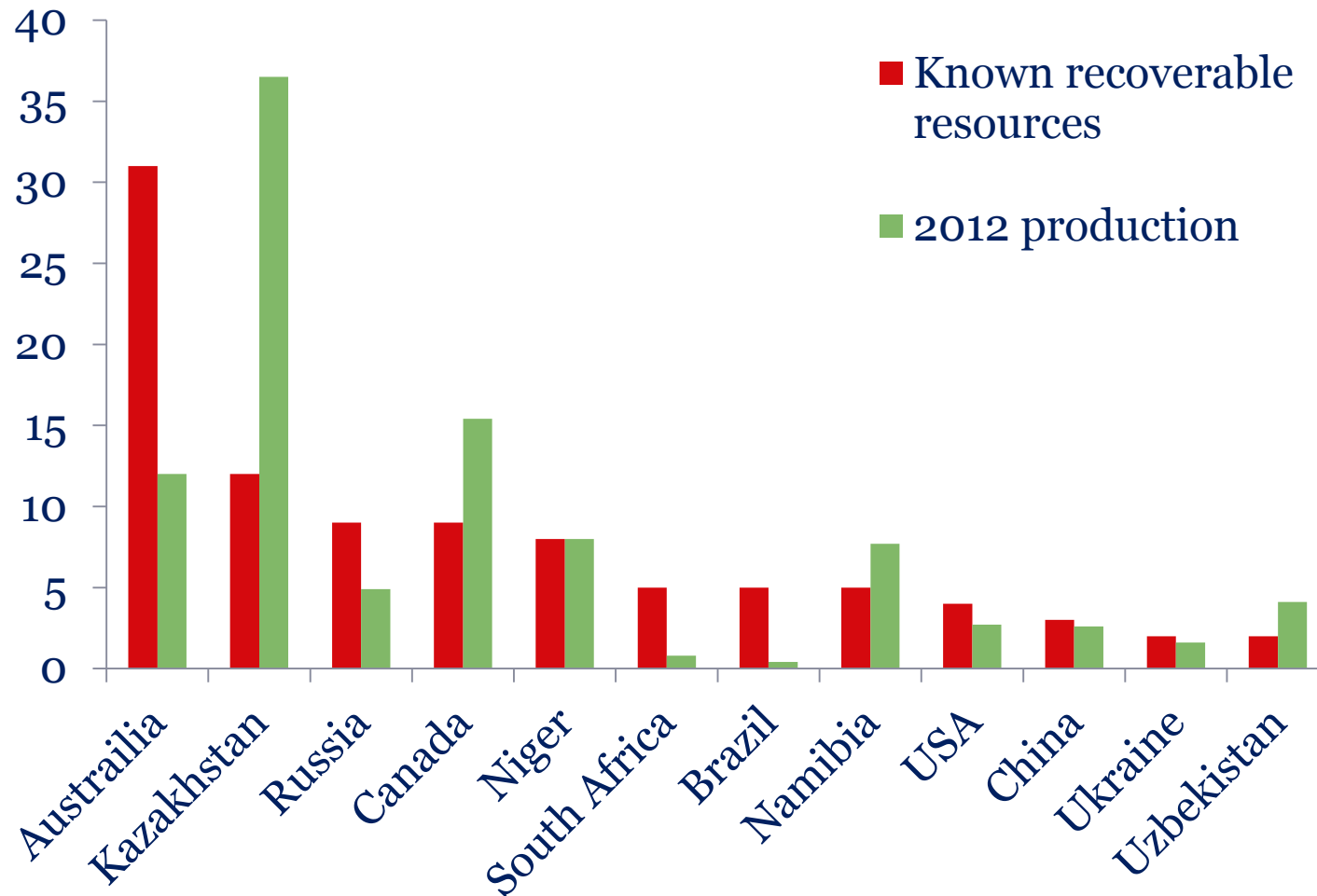
RAR+Inferred, Cost range < \$130/kg U, 1/1/2013 (OECD 2014)



# Annual Uranium Production (t U)



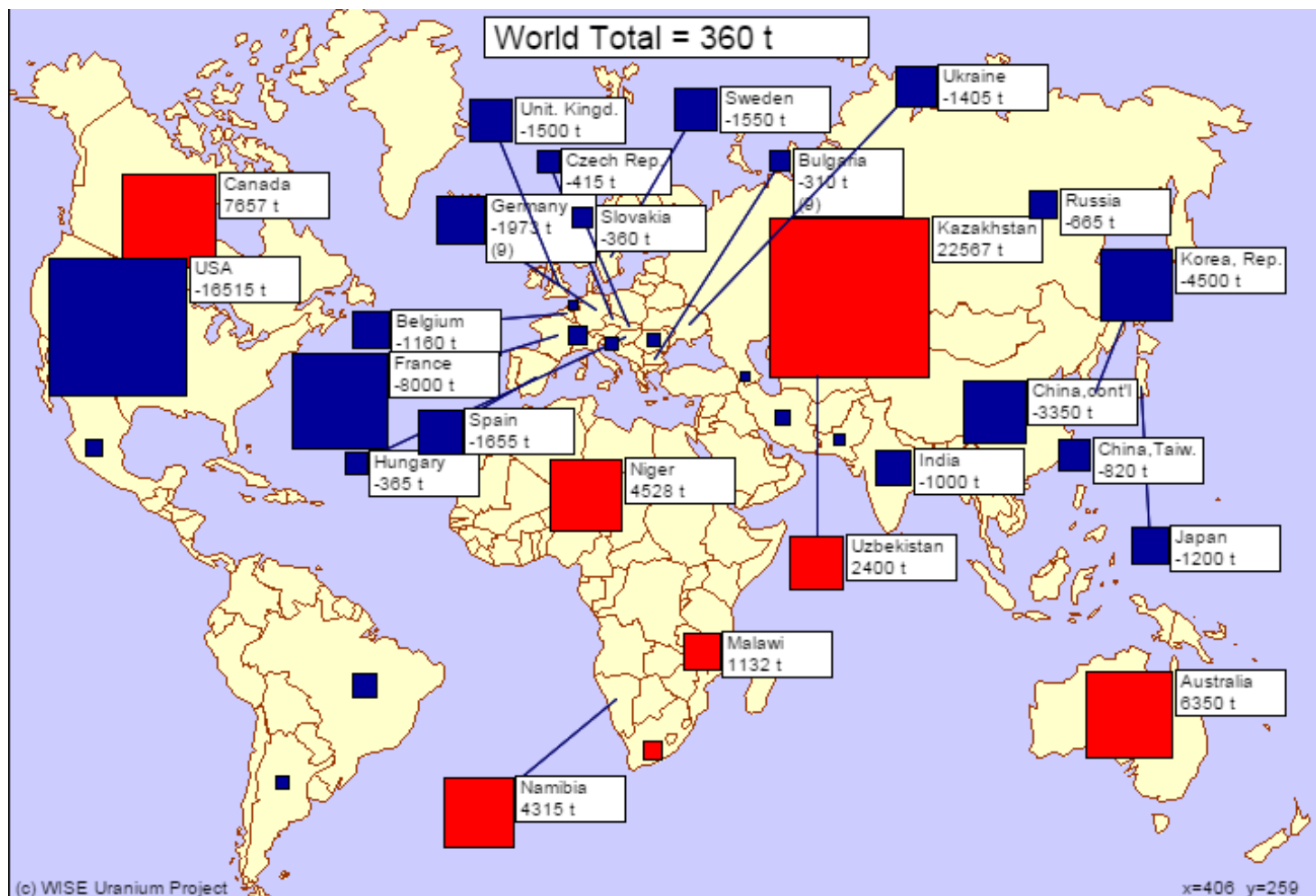
2013 Production (WNA Dec. 2014)



Reasonably Assured Resources plus Inferred Resources, to US\$ 130/kg U, 1/1/07, from OECD NEA & IAEA, *Uranium 2011: Resources, Production and Demand* ("Red Book").

<http://www.world-nuclear.org/info/inf23.html>

# Requirements Balance [t U] (WNA 2014, OECD 2014)

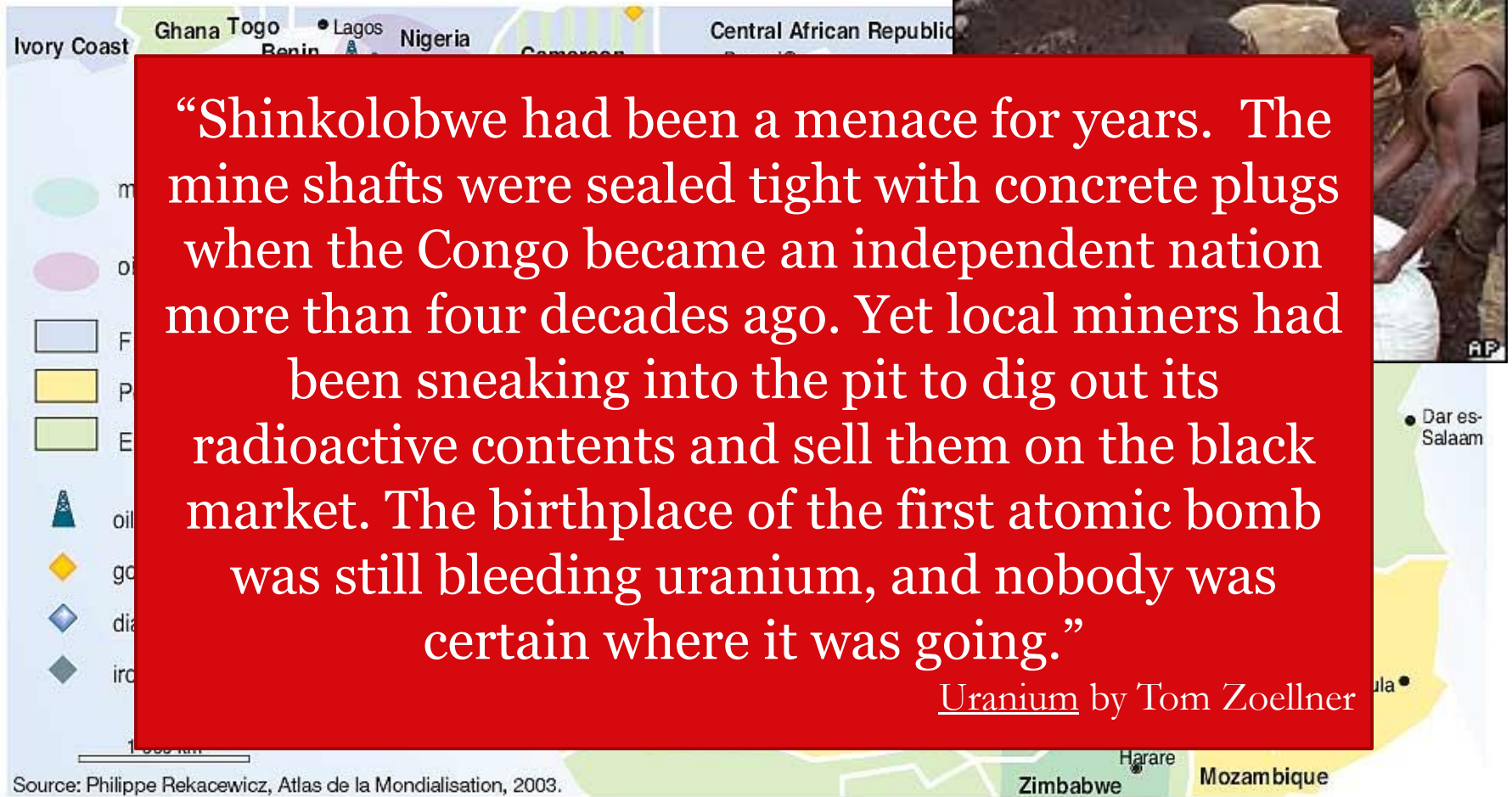


# Ubiquitous Uranium



Shinkolobwe – Midnight Miners

# Ubiquitous Uranium



“Shinkolobwe had been a menace for years. The mine shafts were sealed tight with concrete plugs when the Congo became an independent nation more than four decades ago. Yet local miners had been sneaking into the pit to dig out its radioactive contents and sell them on the black market. The birthplace of the first atomic bomb was still bleeding uranium, and nobody was certain where it was going.”

Uranium by Tom Zoellner

Source: Philippe Rekacewicz, Atlas de la Mondialisation, 2003.

Shinkolobwe – Midnight Miners

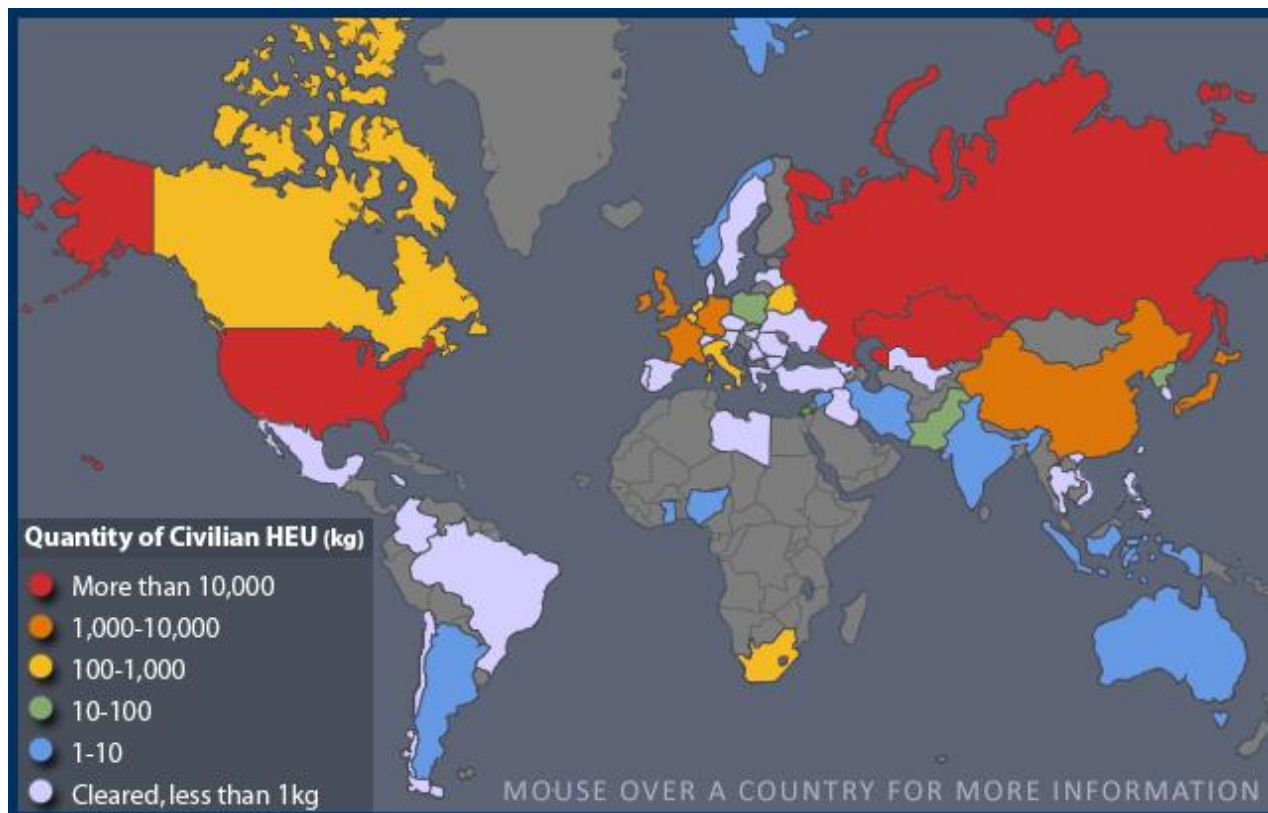
# For what purpose is the U being mined?

- Energy
  - What type of reactor?
    - CANDU – natural U (0.711% U-235)
    - LWR – low enriched U (3-5% U-235)
- Research
  - What type of reactor?
    - Fuel/targets range from LEU (< 20% U-235) to HEU > 90% U-235
- Weapons
  - What type of weapon?
    - U weapon – weapons grade (> 90% U-235)
    - Pu weapon – ranges (natural is best), Pu produced in a reactor from U fuel



# Pathways to HEU

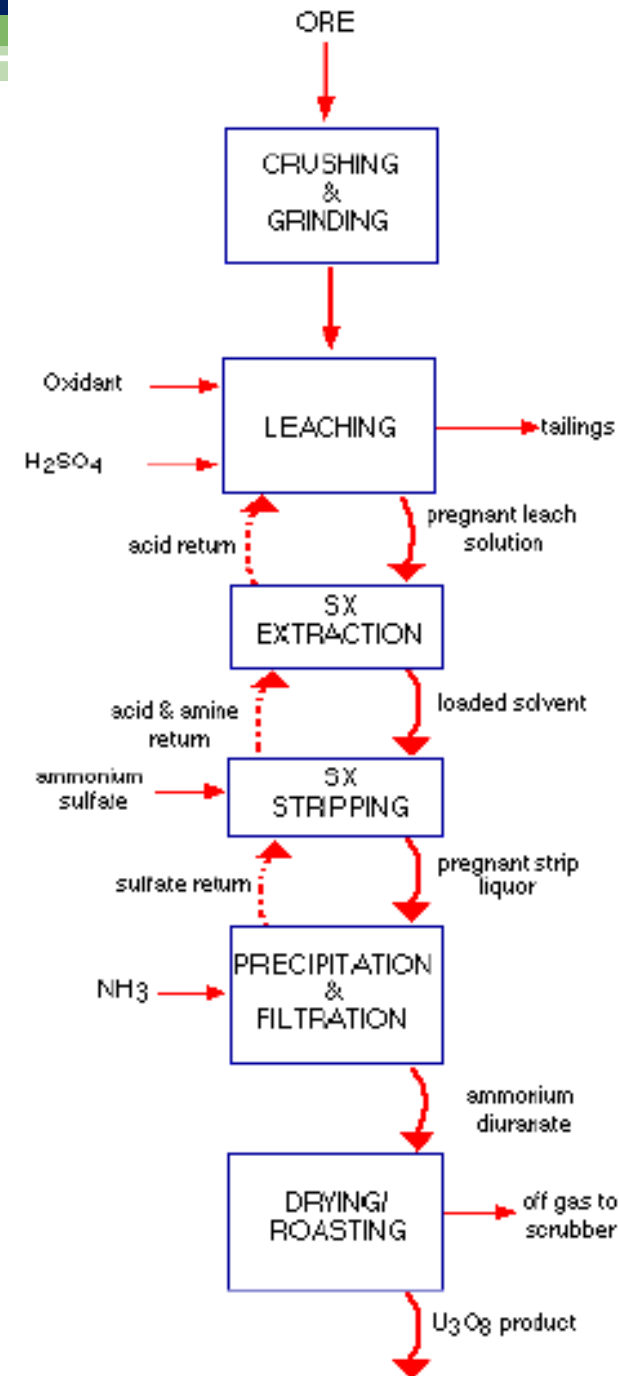
- Enrich natural or LEU to HEU
- Global inventory of HEU



[http://www.nti.org/gmap/other\\_maps/heu/#](http://www.nti.org/gmap/other_maps/heu/#)

# Yellow Cake

- Milling process leaves 70-90%  $U_3O_8$  with impurities (e.g., B, Cd, Cl, REE)
- Yellow cake may be in the form of ammonium-diuranate or sodium-diuranate





# Yellowcake in the news...

## Yellowcake Uranium Trucks "Can Go Wherever They Want" After Recent State Decision

The Colorado Independent | By David O. Williams

Posted: 11/21/2009 5:12 am EST | Updated: 05/25/2011 2:05 pm EDT

**MONTROSE** -- Opponents of a proposed uranium mill in southwestern Colorado near the Utah state line may be relieved to hear that state officials in charge of overseeing the transport of incoming ore and outgoing yellowcake don't actually consider such things "nuclear materials."

By state statute, uranium ore and processed yellowcake, used to make fuel rods for nuclear reactors, are considered mere hazardous materials and therefore not limited to transportation along the state's designated nuclear materials routes.

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South Africa Spy cables

South Africa monitored Iranian activity under US pressure, spy cables show

A Mossad document, dated 28 September 2000, shows that the South African government was monitoring Iranian activity under US pressure, according to spy cables that were recently declassified.

**POLITICO**

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### Video

Benjamin Netanyahu: Iran's 'Rouhani thinks he can have yellowcake and eat it too'



**THE WORLD POST**

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**Knut Royce** Become a fan

Author, 'The Italian Letter: The Forgery That Started The Iraq War'



## Remembering the Hoax That Helped Launch the U.S. Invasion, and Later Disintegration, of Iraq

Posted: 07/10/2014 10:58 am EDT | Updated: 09/09/2014 5:59 am EDT

The documents were crafted by rogue Italian intelligence officers who wanted to peddle them to unsuspecting countries, including Britain and the U.S. -- or anyone else with cash. The centerpiece, or The Italian Letter, was a July 27, 2000, letter purportedly written to Saddam by the president of Niger, an impoverished African country. It allegedly formalized an agreement reached by representatives of both countries three weeks earlier for the supply of 500 tons of uranium ore, also known as yellow cake. The Italian intelligence service, SISMI, first alerted the CIA to the alleged transaction on Oct. 15, 2001, when America was still reeling from al Qaeda's attacks. But it gave no details, such as the tonnage being purchased, and provided no documentation.

# World Primary Conversion capacity

World Nuclear Association *Nuclear Fuel Report 2013 & 2015*

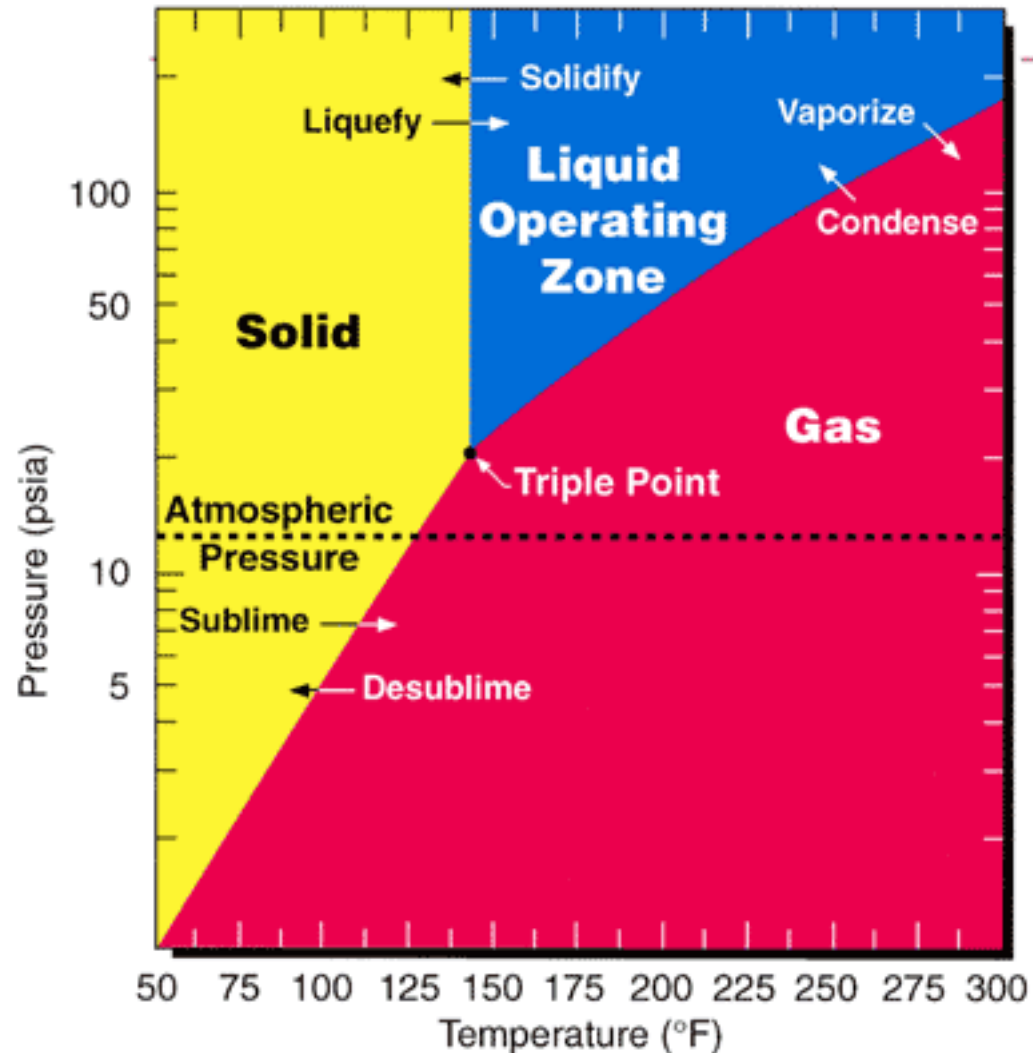
Company	Nameplate capacity (tonnes U/yr as UF <sub>6</sub> )	Approx capacity utilisation 2015	Capacity utilisation 2015, tU/yr
Cameco, Port Hope, Ont, Canada	12,500	70%	8750
Springfields Fuels, UK	(closed August 2014)	0%	0
TVEL at Siberian Chemical Combine, Seversk, Russia	12,500	100% assumed	12,500
Comurhex (Areva), Malvesi (UF <sub>4</sub> ) & Tricastin (UF <sub>6</sub> ), France	15,000	70%	10,500
Converdyn, Metropolis, USA	15,000	70%	10,500
CNNC, Lanzhou, China	4000	unknown	4000
IPEN, Brazil	100	70%	70
<b>World Total</b>	<b>59,100</b>		<b>46,320</b>

# Conversion to UF<sub>6</sub>: Why UF<sub>6</sub>?

- Only <sup>19</sup>F isotope
- Gas at moderate T
- Handled at reasonable T and P as a solid
- Water soluble

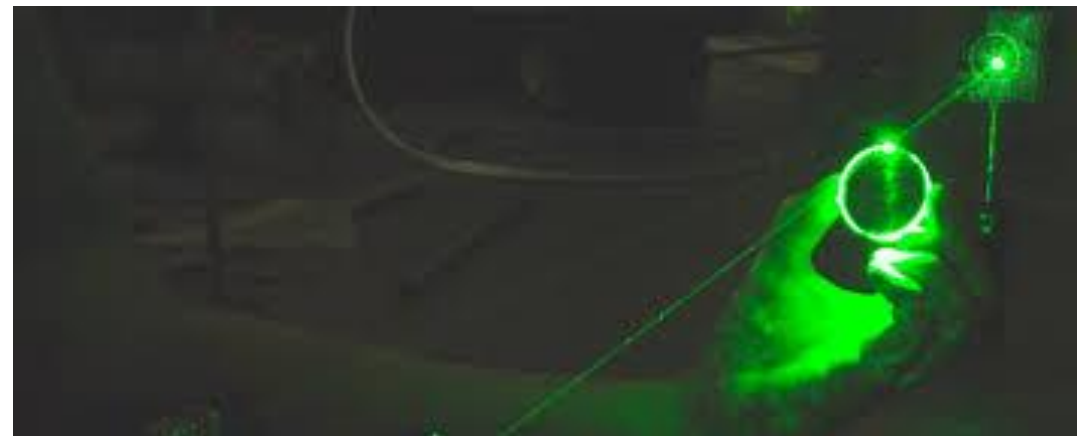
Methods for conversion:

- Wet solvent extraction process
- Dry hydrofluor process





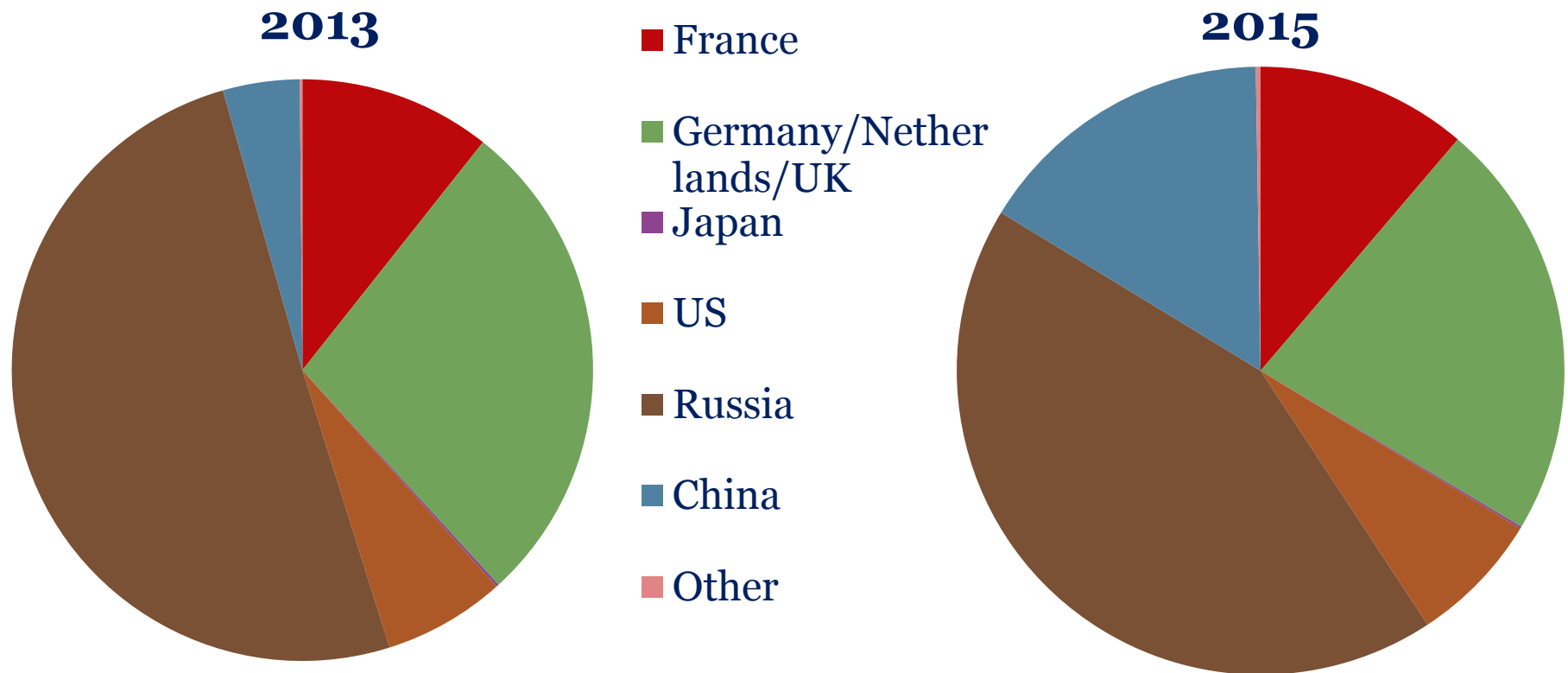
# Enrichment



A calutron operator at her control panel in the Y12 plant.

Photo by Ed Westcott, official US Army photographer for the Manhattan Project  
American Museum of Science and Energy, Oak Ridge, TN • [www.amse.org](http://www.amse.org)

# World enrichment capacity - operational and planned (thousand SWU/yr)



Source: World Nuclear Association *Nuclear Fuel Report 2013 & 2105*, information paper on [China's Nuclear Fuel Cycle](#), Areva [2014 Reference Document](#) for most 2013 figures.

'Other' includes Resende in Brazil, Rattehallib in India and Natanz in Iran. At end of 2012 Iran had about 9000 SWU/yr capacity operating, according to ISIS and other estimates.

The Euratom Supply Agency [Annual Report 2014](#) estimated world nameplate capacity at 56 million SWU, Russia 28 million SWU, Urenco 18.1 million SWU and Areva 7.5 million SWU.

# Enrichment calculations

- Iran's Fordow enrichment facility in Qom has been quoted to contain 3000 P2 centrifuges with an annual SWU capacity ranging from 2-5 SWU per centrifuge.
- Prior to the Iran Nuclear Deal, it was possible for LEU from the Natanz enrichment facility to be further enriched at the Fordow facility.
- Using the SWU calculator, we can see how long it would take to convert 3.5% enriched  $UF_6$  to 90%  $UF_6$  depending on the quality of the centrifuges used.

# SWU Calculator

$$\frac{SWU}{T} = PV(x_p) + WW(x_w) - FV(x_f)$$

kg-SWU/yr to express **capacity of an enrichment plant**

T = time

V(x<sub>i</sub>) = separation potential  $V(x_i) = (2x_i - 1) \ln \left( \frac{x_i}{1 - x_i} \right)$

P = product

W = waste

F = feed

Typically...

x<sub>f</sub> = 0.00711 (natural U)

x<sub>p</sub> = 0.035 (LWR fuel) – 0.95 (weapons U)

x<sub>w</sub> = 0.002-0.004 (depleted U)

# SWU Calculator

What do we know?

- Starting enrichment ( $x_f$ ) = 3.5%
- Final enrichment ( $x_p$ ) = 90%
- Assume depletion ( $x_w$ ) = 1%
- Final product (P) = 29.7 kg of  $UF_6$  (20 kg U)
  
- Basic equations:
- Feed (F) = Product (P) + Waste (W)
- $x_f F = x_p P + x_w W$



# SWU Calculator

F	1057.3
P	29.7
W	1027.6
$x_f$	0.035
$x_p$	0.90
$x_w$	0.01
SWU	1418.4
Facility	3000
yr	0.47
weeks	24.59
d	172.69

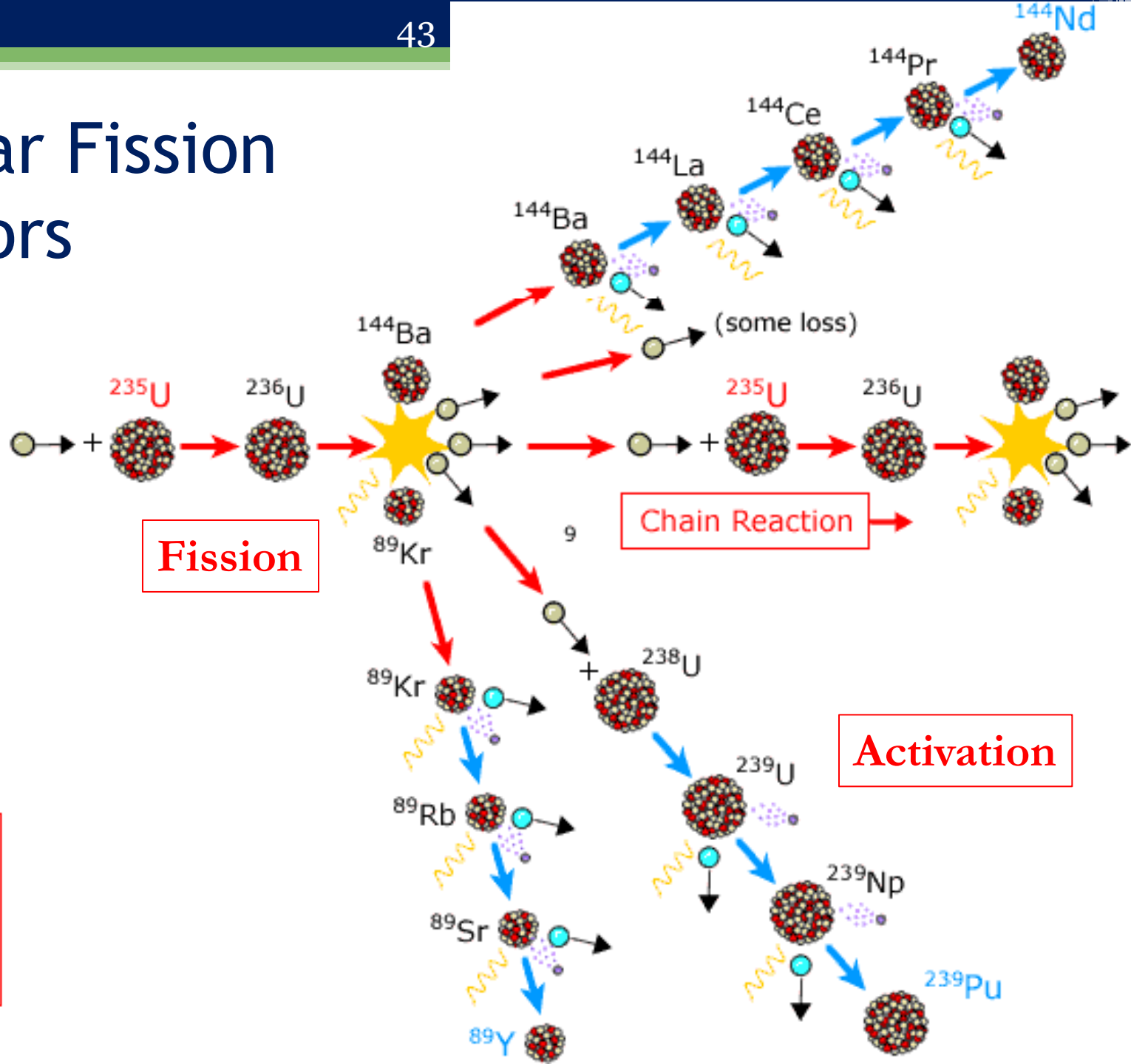
If centrifuges operate at:

- 0.5 SWU/yr,  $t = 345.38$  d
- 3.0 SWU/yr,  $t = 57.56$  d

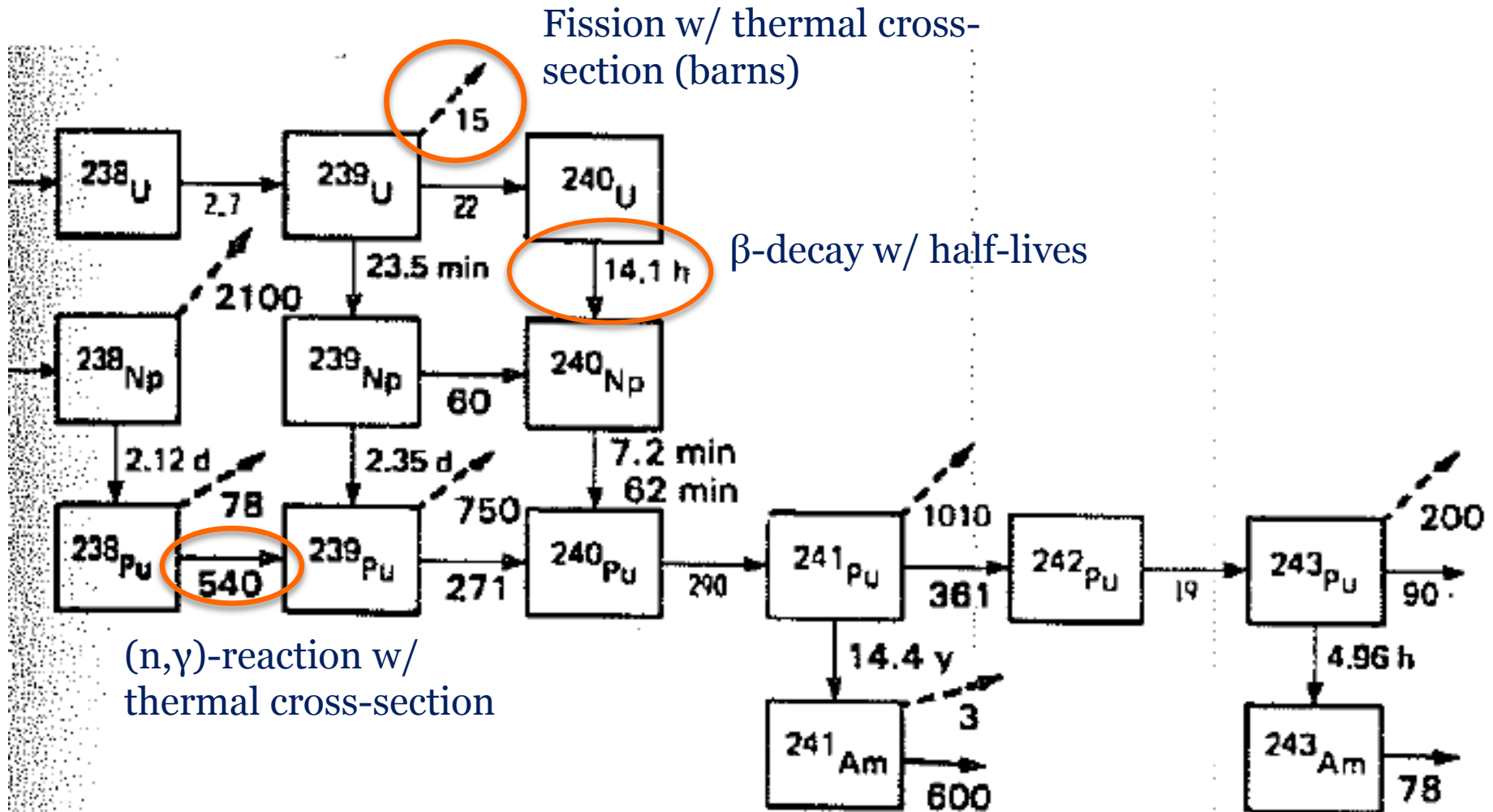
# A few interesting articles...

- K. Yost, “Opinion: The Real Danger of Qom” The Tech Online Edition 129(44) 10/13/2009.  
<http://tech.mit.edu/V129/N44/yost.html>
- J. Bernstein, “The Simple Math of an Iranian Nuclear Bomb” Tablet 10/2/2014.  
<http://www.tabletmag.com/jewish-news-and-politics/183851/nuclear-iran-bernstein>

# Nuclear Fission Reactors



# $^{238}\text{U}$ Fuel Cycle $\rightarrow$ Pu-isotopes



# How much Pu is produced in a reactor?

Rate of radionuclide production = Production by fission + absorption + decay of parent - Loss by absorption + decay + escape

$$\frac{dn_i}{dt} = \gamma_i n_i \sigma_f \phi + n_{ji} \sigma_{a,ji} \phi + \lambda_k n_k - n_i \sigma_{a,i} \phi - \lambda_i n_i - E$$

$n_i$  = atom density of nuclide i

t = time

$\gamma_i$  = fission yield for nuclide i

$\sigma_f$  = fission cross-section

$\sigma_a$  = absorption/capture cross-section

$\Phi$  = neutron flux

$\lambda$  = decay constant

E = escape term

i = nuclide of interest

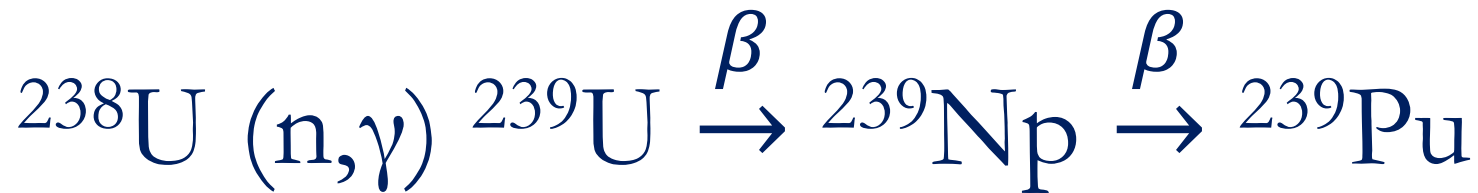
j = nuclide that can transmute to nuclide i

k = nuclide that can decay to nuclide i

# How much Pu is produced in a reactor?

Rate of radionuclide production = Production by fission + absorption + decay of parent - Loss by absorption + decay + escape

$$\frac{dn_i}{dt} = \gamma_i n_i \sigma_f \phi + n_{ji} \sigma_{a,ji} \phi + \lambda_k n_k - n_i \sigma_{a,i} \phi - \lambda_i n_i - E$$





# How much Pu is produced in a reactor?

## Some assumptions

- Neglect  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  decay
- After 2-3 weeks for 1 year irradiation, rate of production/loss of  $^{239}\text{U}$  and  $^{239}\text{Np}$  are equal.
- Neglect  $^{238}\text{U}$  burn-up.

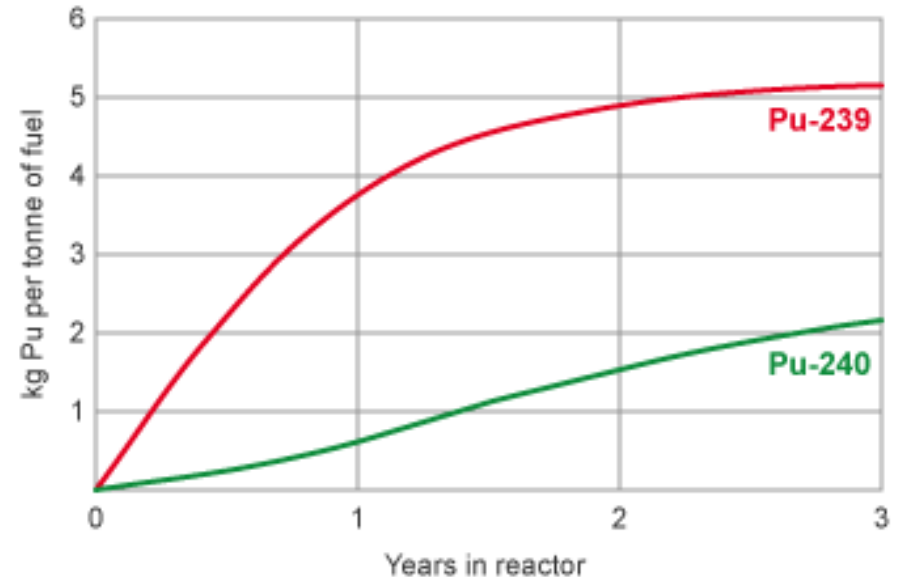
$$\frac{dn_{^{239}\text{Pu}}}{dt} = n_{^{238}\text{U}} \sigma_{c,^{238}\text{U}} \phi - n_{^{239}\text{Pu}} \sigma_{a,^{239}\text{Pu}} \phi$$

## Some parameters

- Irradiation time,  $t = 1 \text{ yr}$
- Neutron flux,  $\Phi = 4.0 \times 10^{13} \text{ neutrons/cm}^2\text{s}$
- Capture cross section,  $\sigma_{c,^{238}\text{U}} = 2.1 \text{ barns}$
- Absorption cross section,  $\sigma_{a,^{239}\text{Pu}} = 600 \text{ barns}$
- 90 tons of uranium (3 wt. %  $^{235}\text{U}$ )

# How much Pu is produced in a reactor?

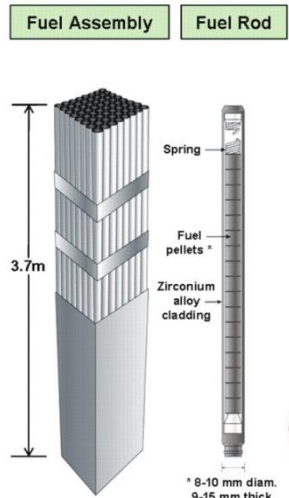
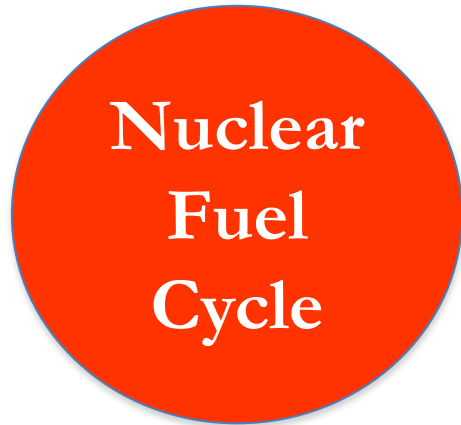
$$n_{239\text{Pu}} = 148 \text{ kg}$$



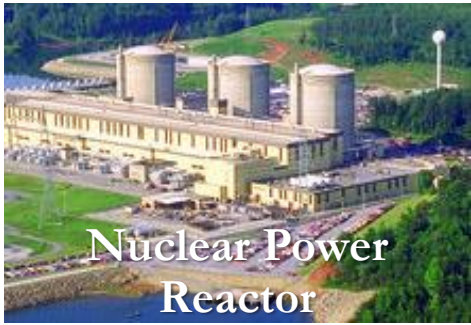
<http://www.world-nuclear.org/info/nuclear-fuel-cycle/fuel-recycling/plutonium/#Notes>

- 70 t Pu produced globally/yr

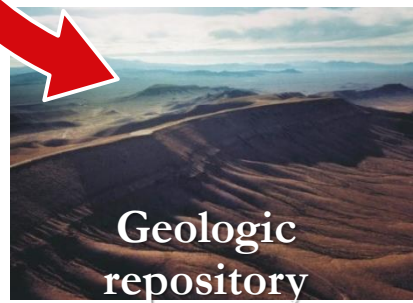
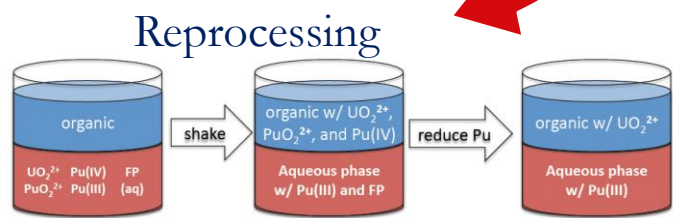
# Where does SNM come from?



Enrichment and Fuel Fabrication



Mining and Milling



# Uranium Forensics Case Study

- June 2003

- ITU  
from

- All

– C

– D

• Are these pellets  $\text{UO}_2$ ?

• What enrichment?

• Where are they from?

Macroscopic

Pellet no.

1				
2	14.7014	14.20	11.44	2.1
3	15.3979	14.91	11.45	2.1
4	14.8626	14.46	11.45	2.1

# Uranium Forensics Case Study

- Pellet dimensions
- 2%  $^{235}\text{U}$  enrichment
- $\text{UO}_2$
- $12.6 \pm 0.8$  years (as of 16.06.2003)
  - Produced in end of 1990
  - $^{234}\text{U}/^{230}\text{Th}$  chronometer

Uranium content in mass%

Technique	U-content
HKED	$87.43 \pm 0.32$
Titration	$87.90 \pm 0.13$
IDMS	$87.99 \pm 0.24$

Impurities in  $\mu\text{g/g}$  U by SF-ICP-MS

Element	Concentration
Al	$6.08 \pm 0.73$
Ca	$18.4 \pm 2.2$
Cr	$6.12 \pm 0.73$
Cu	$1.80 \pm 0.22$
Fe	$91.9 \pm 7.4$
K	$44.7 \pm 3.6$
Mg	$4.71 \pm 0.57$
Mn	$1.13 \pm 0.14$
Na	$17.9 \pm 2.1$
Ni	$5.14 \pm 0.62$
Zn	$3.40 \pm 0.41$

Isotopic composition of uranium by mass spectrometry in mass%

Technique	$^{234}\text{U}$	$^{235}\text{U}$	$^{236}\text{U}$	$^{238}\text{U}$
TIMS	$0.0147 \pm 0.0010$	$2.0005 \pm 0.0010$	$0.0071 \pm 0.0067$	$97.9778 \pm 0.0019$
MC-ICP-MS	$0.0142 \pm 0.0002$	$2.0005 \pm 0.0001$	$0.0071 \pm 0.0000$	$97.9782 \pm 0.0010$

# Uranium Forensics Case Study

- Pellet dimensions
  - 2%  $^{235}\text{U}$  enrichment
- } **RBMK-1500** (Russian type water-cooled, graphite-moderated reactor)
- $12.6 \pm 0.8$  years  
(as of 16.06.2003)
    - Produced in end of 1990
    - $^{234}\text{U}/^{230}\text{Th}$  chronometer
  - Impurities

**RBMK-1500** (Russian type water-cooled, graphite-moderated reactor)

**Ignalina Unit 2** (Lithuania)  
Started Aug. 1987

**MZ Electrostal**  
(Moscow, Russia)

Elem.	Conc. ( $\mu\text{g/g U}$ )	Elem.	Conc. ( $\mu\text{g/g U}$ )
Al	6.08 + 0.73	Mg	4.71 + 0.57
Ca	18.4 + 2.2	Mn	1.13 + 0.14
Cr	6.12 + 0.73	Na	17.9 + 2.1
Cu	1.80 + 0.22	Ni	5.14 + 0.62
Fe	91.9 + 7.4	Zn	3.40 + 0.41
K	44.7 + 3.6		



# References

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# Upcoming Webinars

- Nuclear Materials Analysis — Physical and Spectroscopic Methods
- Sample Matrices and Collection, Sample Preparation
- Nuclear Materials Analysis — Physical and Spectroscopic Methods

NAMP website: [www.wipp.energy.gov/namp](http://www.wipp.energy.gov/namp)