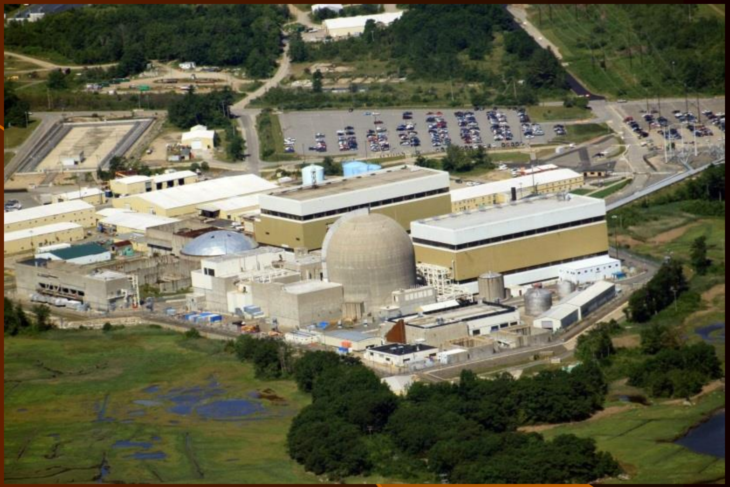
Sequestration of Radioactive Wastes:

The Oklo Experiment (Gabon)

Advantages of Nuclear Power



- At the site of generation, the only major environmental impact is waste heat.
- Given the high energy content per unit mass of uranium, the disturbance of the environment during mining is minimal compared to coal.

Disadvantages of Nuclear Power

- Disposal of high level wastes.
- Vulnerability to attack both at the reactor site and during transportation of radioactive materials.

The ^{235}U Fission-reaction (BWR & PWR)

$$^{235}U + {}^{1}_{0}n \longrightarrow \text{fission fragments} + 2\text{-}3 \text{ neutrons} + \text{energy}$$

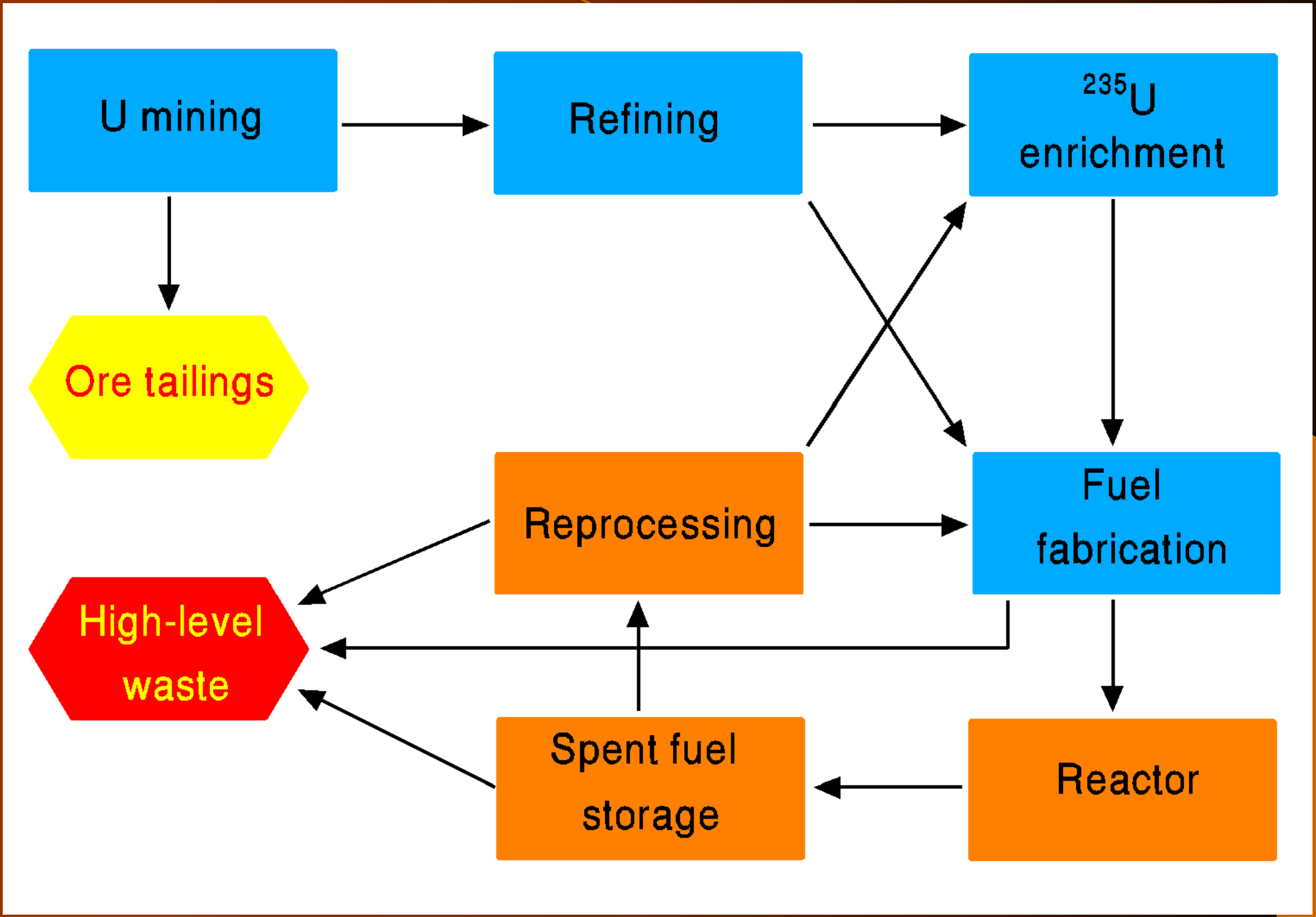
The neutrons released in this reaction are high energy (2MeV) neutrons. They need to be slowed down to thermal velocities to maximize the probability of their reacting with another ^{235}U nucleus. Water plays the role of moderator and also transfers heat from the reactor core to the heat exchanger and cooling loop.

The Pu Breeding-reaction (Breeder reactors)

$$^{238}U + {}^{1}_{0}n \longrightarrow {}^{239}U \longrightarrow {}^{239}Np + \beta^{-} \longrightarrow {}^{239}Pu + \beta^{-}$$

^{239}Pu is a fissionable isotope. To fission this isotope we need to use fast neutrons (>1 MeV). Breeder reactors essentially utilize the other 99.3% of the uranium, a large increase in the availability of nuclear energy. Reactions like those above produce the transuranic nuclides.

The Nuclear Fuel Cycle



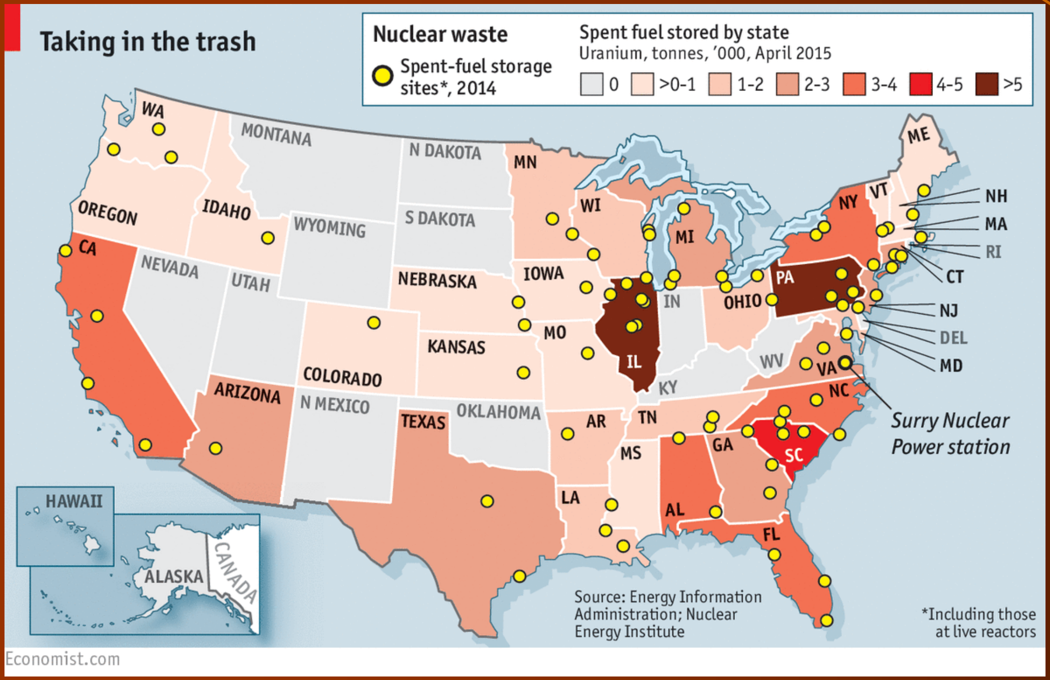
Types of Radioactive Wastes

Low-level radioactive waste has relatively low radioactivity and typically contains radioactive isotopes with short half lives. The volume is large but the total radioactivity is small.

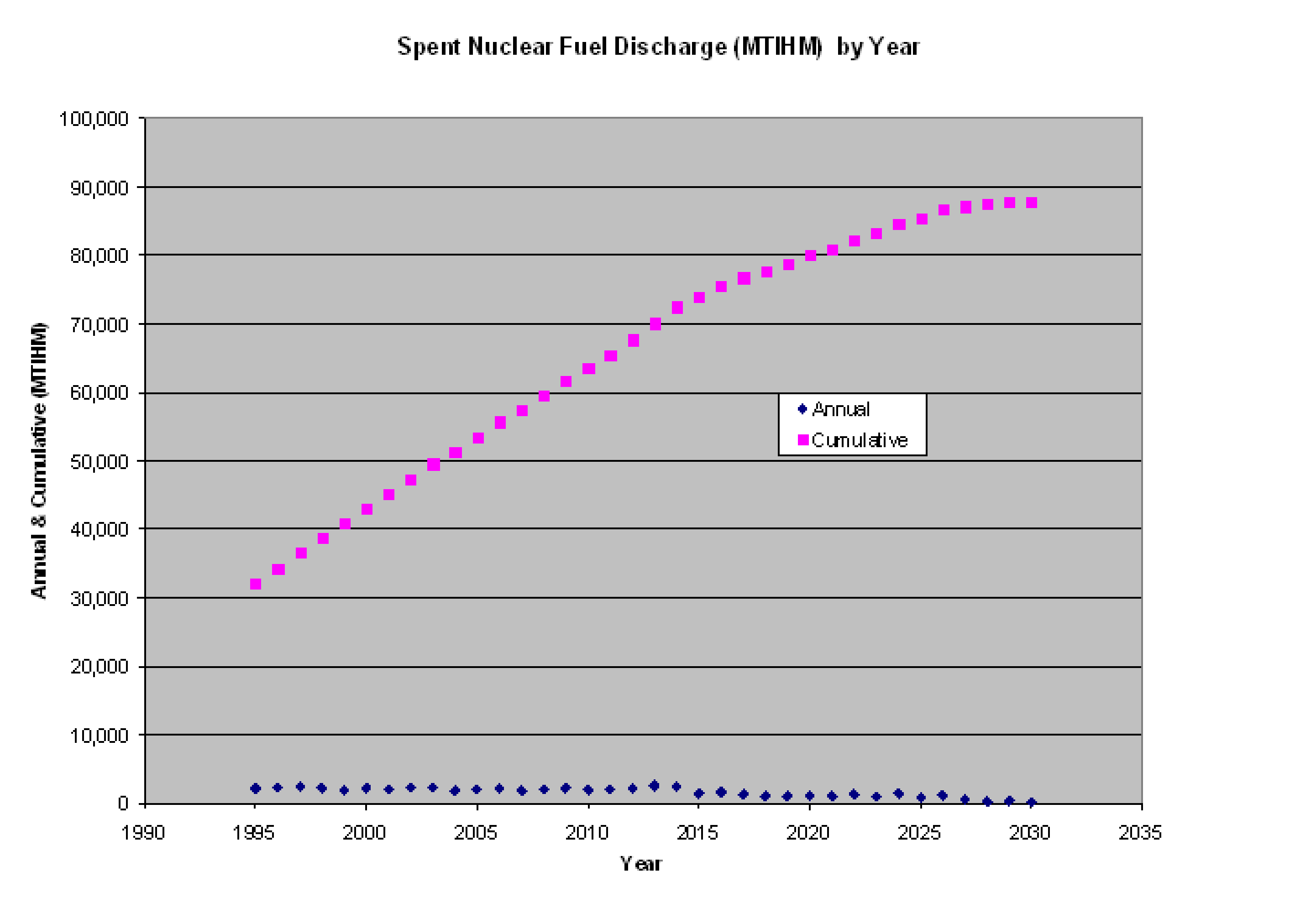
High-level radioactive waste comprises the used fuel elements and the waste generated during the processing of these fuel elements. The volume is small but the total radioactivity is large.

The Problem

Long term sequestration of highly radioactive wastes



Amount of High Level Waste Through 2030



Total volume in 2030 = 37,100 m^3 = 3,710 SL = 37 KSL

A New SI Unit for Environmental Applications

SL for Volume of Waste

Based on the volume of a Standard Sewage Disposal Truck

SL = 10 m^3

KSL = 10,000 m^3

MSL = 1×10^6 m^3

GSL = 1×10^9 m^3

Representative radioactive isotopes for nuclear wastes

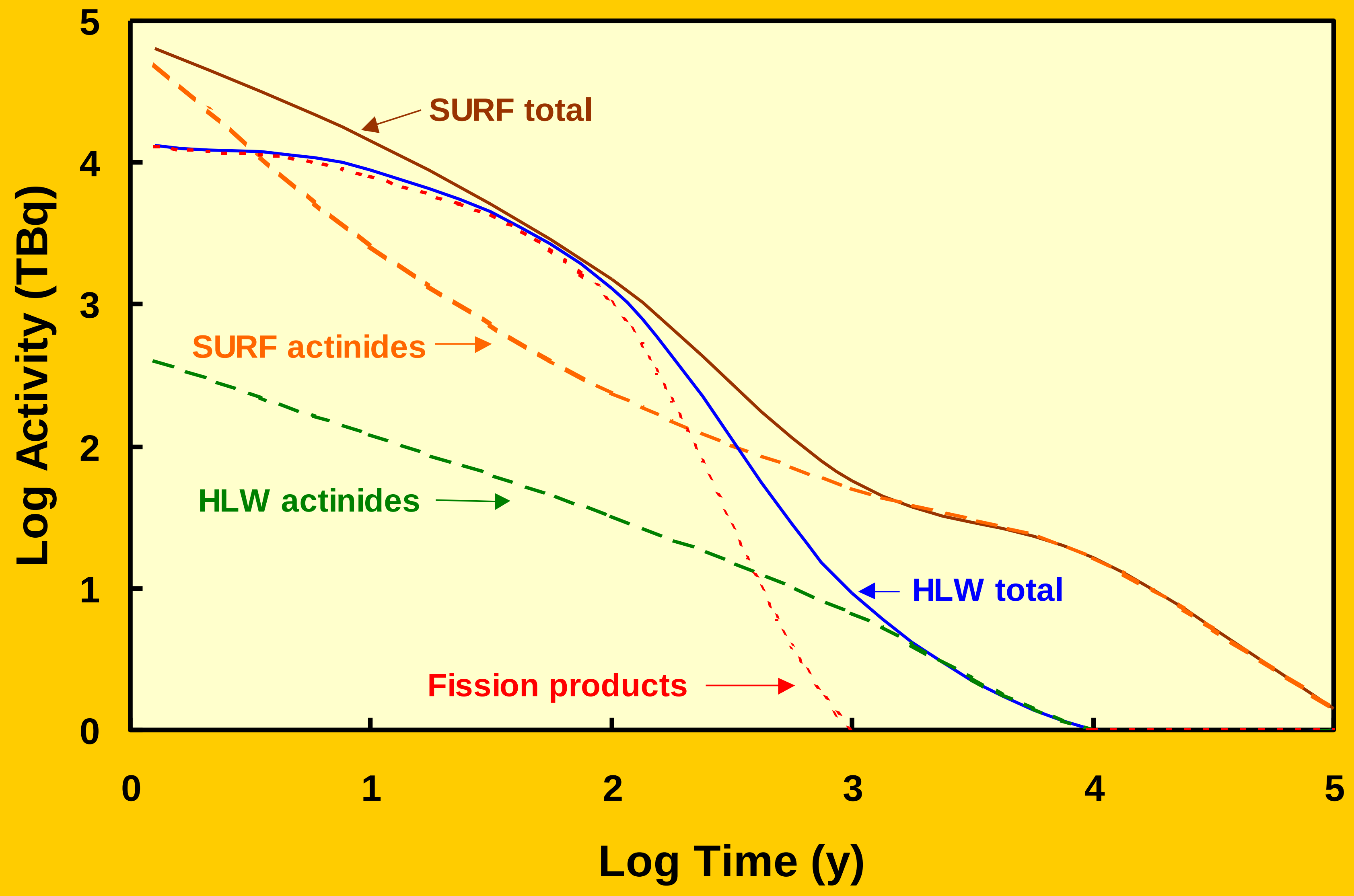
Isotope	Half-life	Decay mode
Fission products		
^{85}Kr	10.8 y	β
^{89}Sr	51 d	β
^{90}Sr	28 y	β
^{95}Zr	64 d	β
^{95}Nb	35 d	β
^{99}Tc	2.1×10^5 y	β
^{106}Ru	1 y	β
^{131}I	8 d	β
^{133}Xe	5.2 d	β

Isotope	Half-life	Decay mode
Fission products		
^{137}Cs	30 y	β
^{141}Ce	33 d	β
^{147}Pm	2.6 y	β
Transuranics		
^{237}Np	2.1×10^6 y	α
^{239}Pu	2.4×10^4 y	α
^{240}Pu	6.6×10^3 y	α
^{241}Am	433 y	α

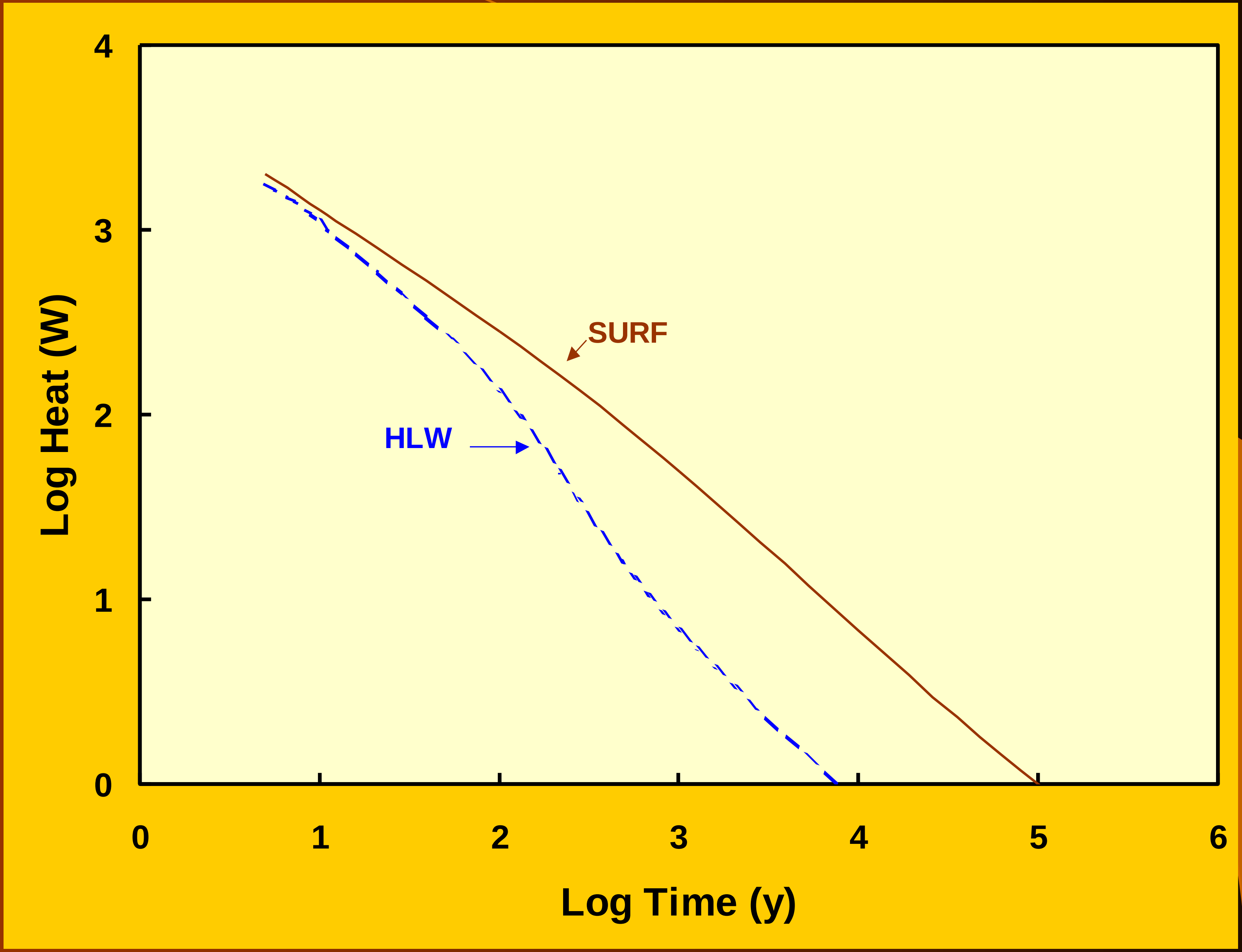
Reprocessing of fuel elements and waste disposal

- If the fuel elements are reprocessed, the waste consists of fission products and transuranic elements (HLW).
- If the fuel elements are not reprocessed, the waste consists of the total inventory of radioactive isotopes; i.e., fission products, uranium, and transuranic elements (SURF).

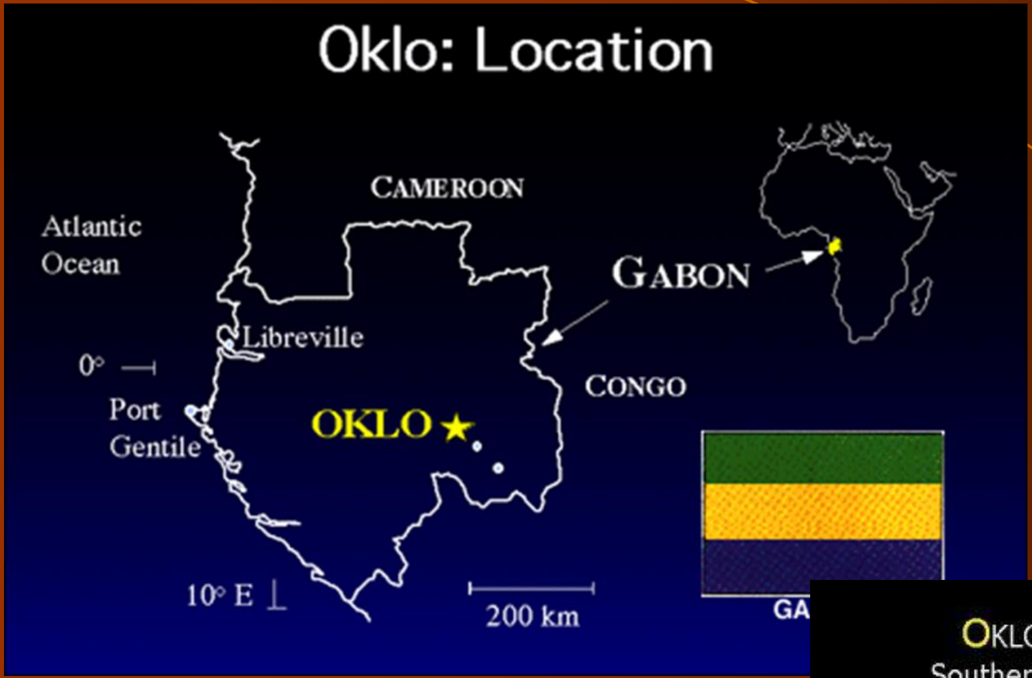
Radioactivity as a function of time



Heat production as a function of time

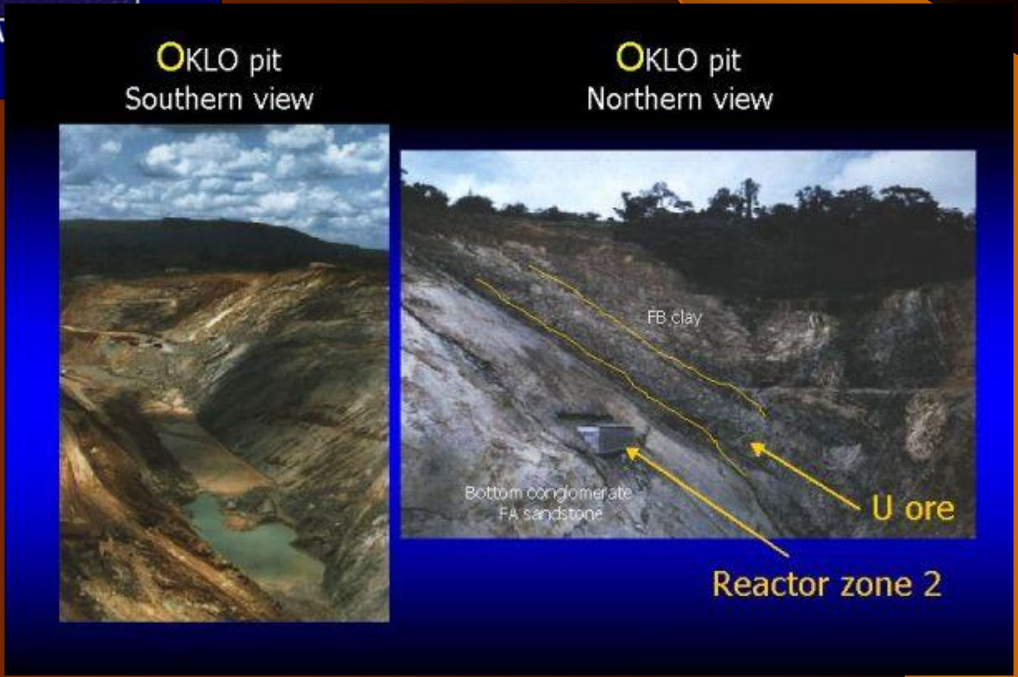


The Oklo Experiment



In the 1970s the French nuclear industry began mining uranium ore deposits in Gabon.

The Oklo mine



The Mystery

- In present-day uranium ore deposits ^{235}U constitutes 0.72% of the total uranium. This isotope must be enriched to 3 - 4% for use in power reactors.
- After enrichment at the processing plant in France ^{235}U enrichment was only 0.5 - 1%.
- An investigation revealed that the uranium ore from Gabon contained 0.559 to 0.698 atom-% ^{235}U instead of 0.719 atom-% ^{235}U. The ore was depleted in ^{235}U.
- Where was the rest of the ^{235}U? Diversion for terrorist activities? Natural phenomena?

Formation of U ore deposits

- Oxidizing environment required for the transport of U in the +6 state.
- Reducing environment required for the precipitation of U in the +4 state [uraninite (UO_2)] or pitchblende (massive, amorphous, cryptocrystalline with some attached waters).
- Organic matter and sulfur reducing bacteria play a key role in establishing the reducing environment.

Cross-section of a typical “roll-front” uranium ore deposit

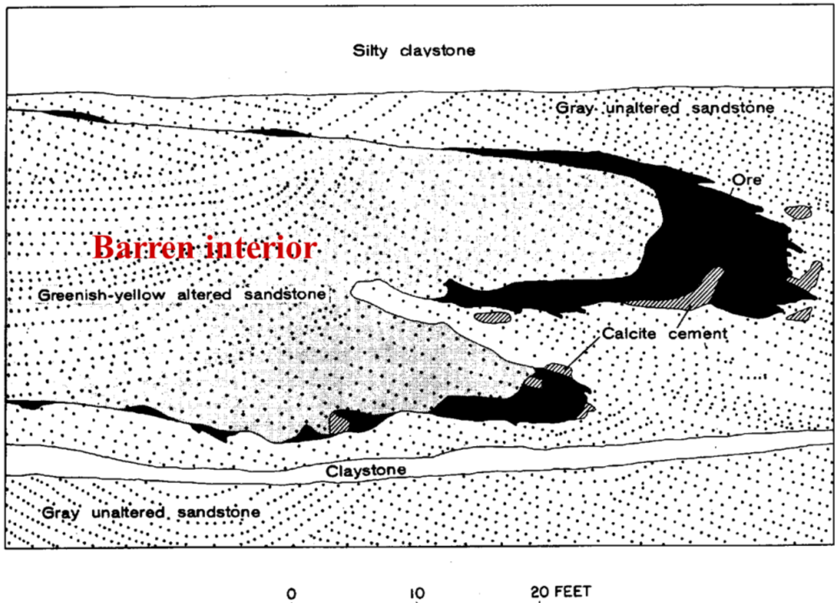
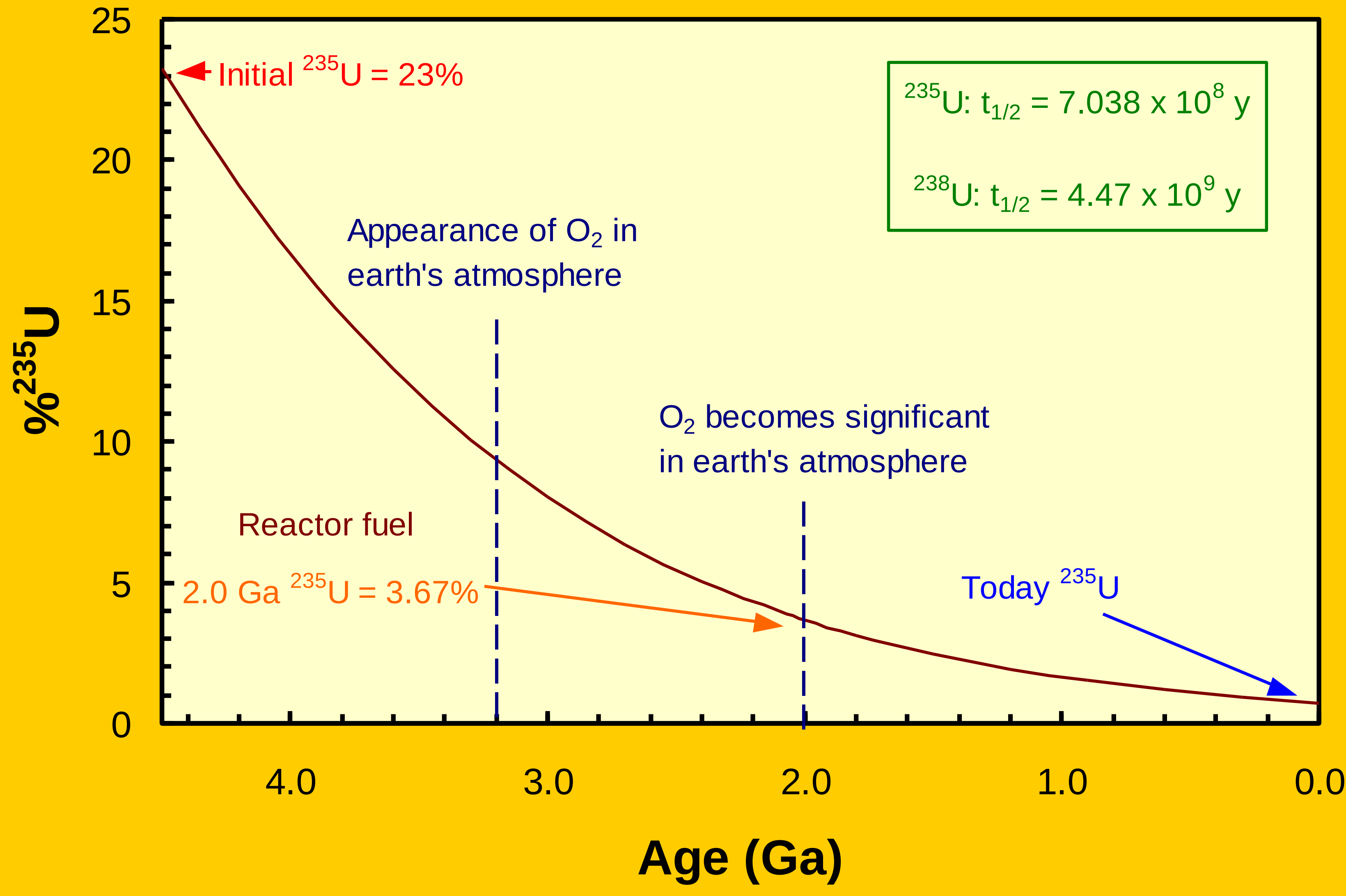


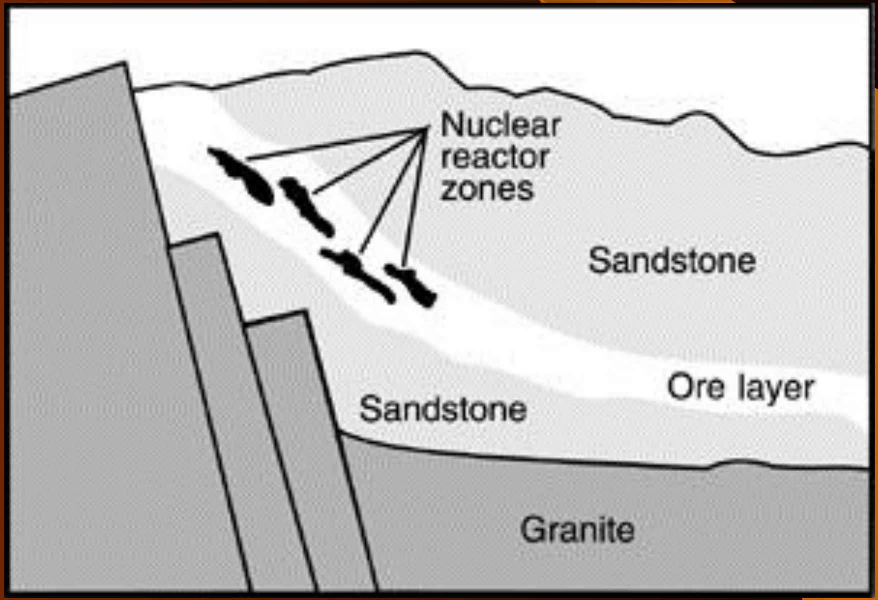
Figure 10-5 Section normal to west edge of altered sandstone tongue, showing the relation of ore to calcite cement to altered sand. (From J. Ridge, ed., *Ore Deposit of the United States*, 1968.)

Variations in ^{235}U and ^{238}U as a function of time



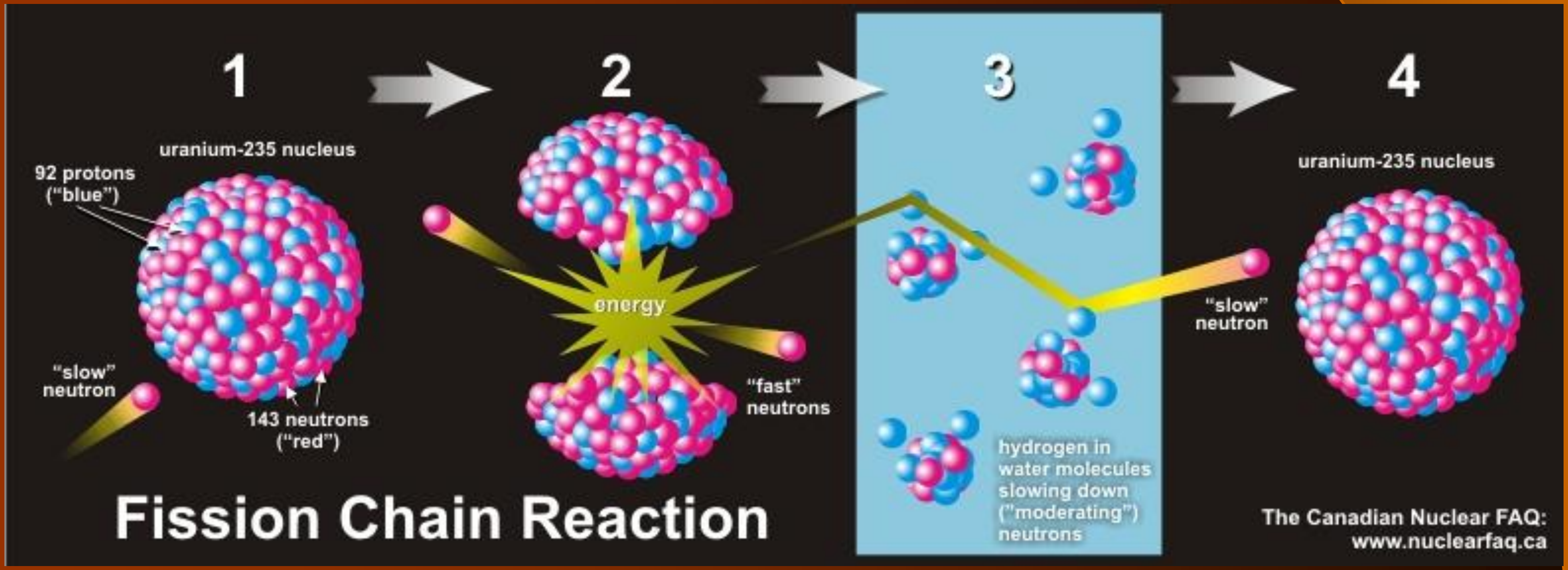
Geology of the Oklo ore deposits

- Located in the Francevillian, a Lower Proterozoic series of unmetamorphosed volcanic and sedimentary rocks.
- The basal section consists of ~900 m of fluviatile sandstone and conglomerate with argillaceous and carbonate cement.
- U mineralization occurred at the top of this sequence in a 5-6 m thick layer.
- The overlying sequence consists of black pelites, dolomite, and Mn-rich carbonates.
- At the mine site the strata dip to the northeast at ~45º.
- The ore zones consists of cycles of conglomerate grading upwards to sandstone.
- In the ore zone U occurs as pitchblende with an average value of 0.2%, locally reaching 25%.
- In the reactor zones U occurs as uraninite with U concentrations of 25-60%.

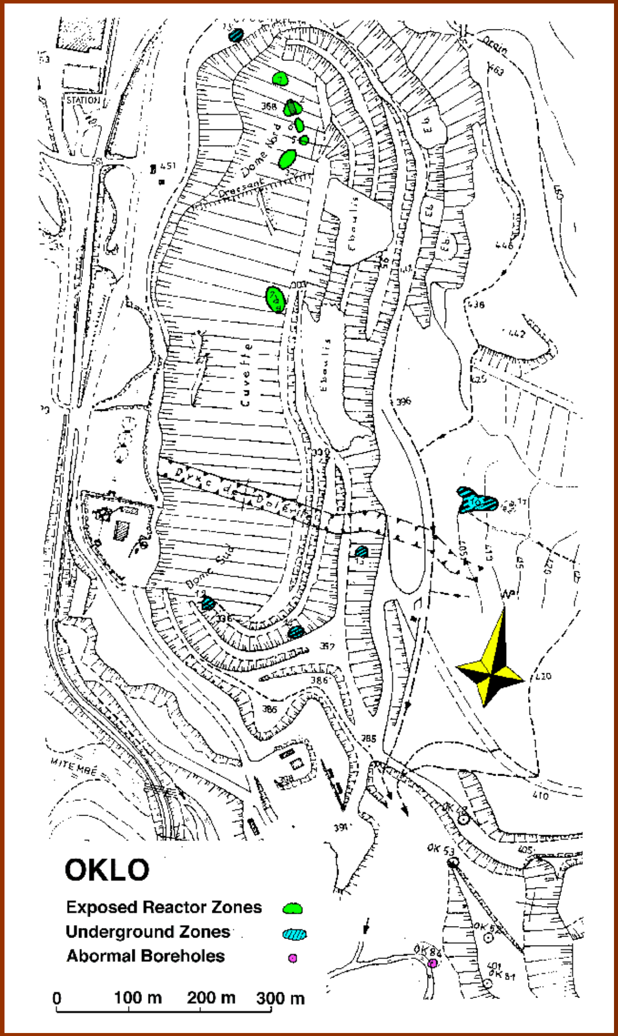


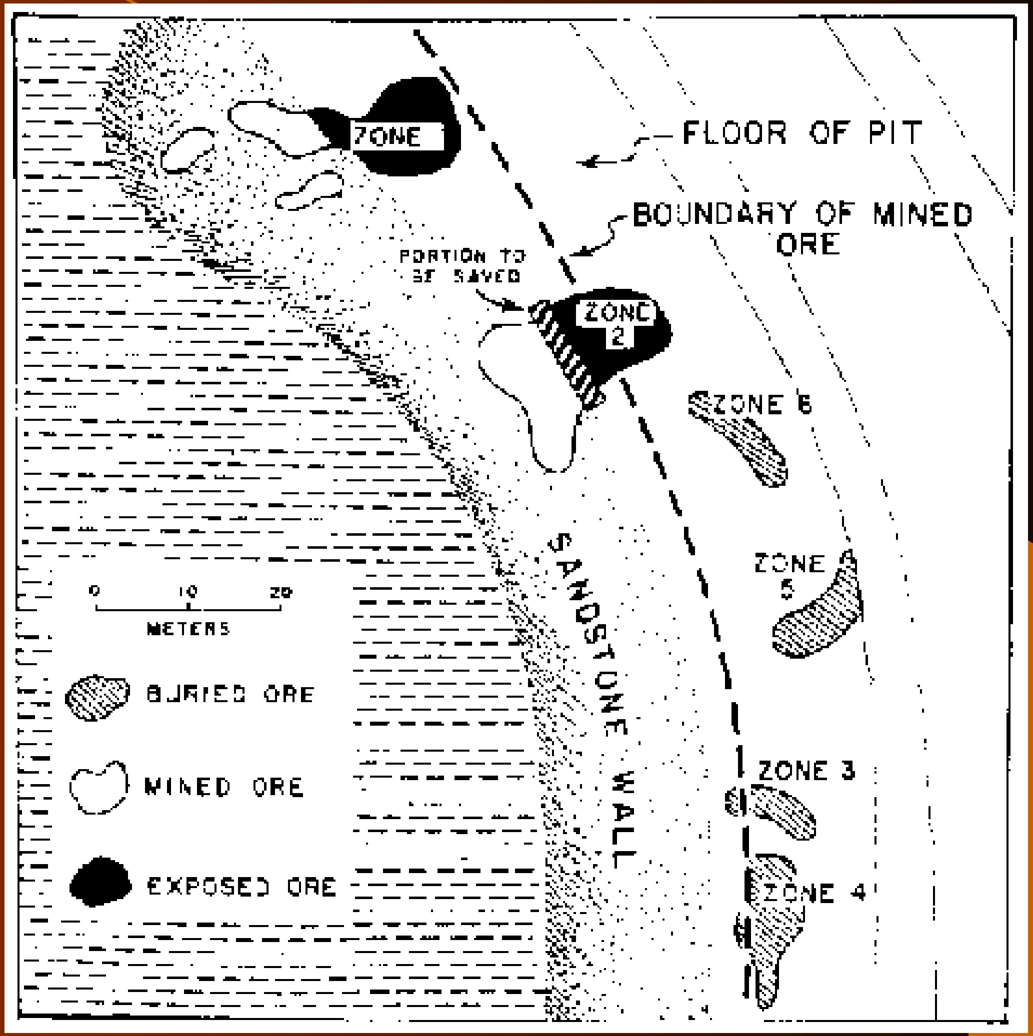
Conditions for a natural chain reaction

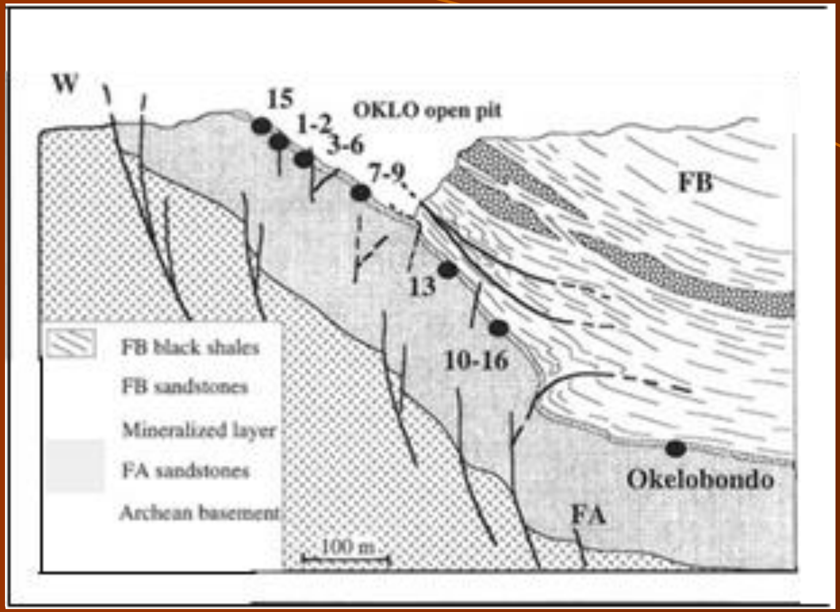
- High concentrations of U. Oklo is a secondary ore deposit with U-rich lenses containing >50% uranium.
- High U-235 abundances (~3.7% at 2.0 Ga).
- High water concentrations. Water acts as a moderator.
- No neutron poisoning. 100 ppm Li; 20 - 250 ppm B; 200 - 800 ppm V; 5 - 100 ppm Cr, Ni, Cu; just a few ppm Cd; 100 ppm REE; <20 ppm Hf.



Location of the ore deposits



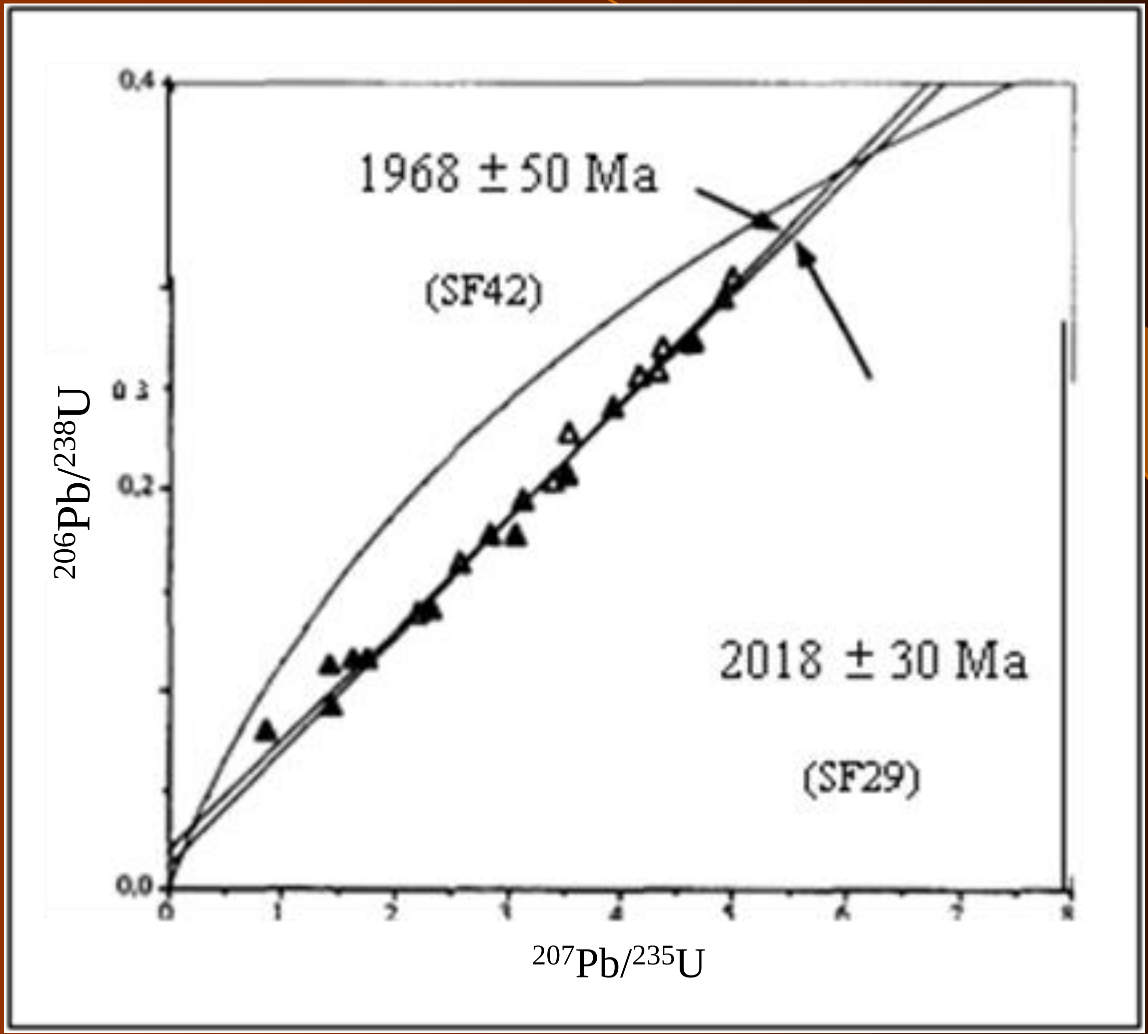



W
15
1-2
3-6
OKLO open pit
7-9
FB
13
10-16
FB black shales
FB sandstones
Mineralized layer
FA sandstones
Archean basement
Okelobondo
FA
100 m


Remains of zone 2

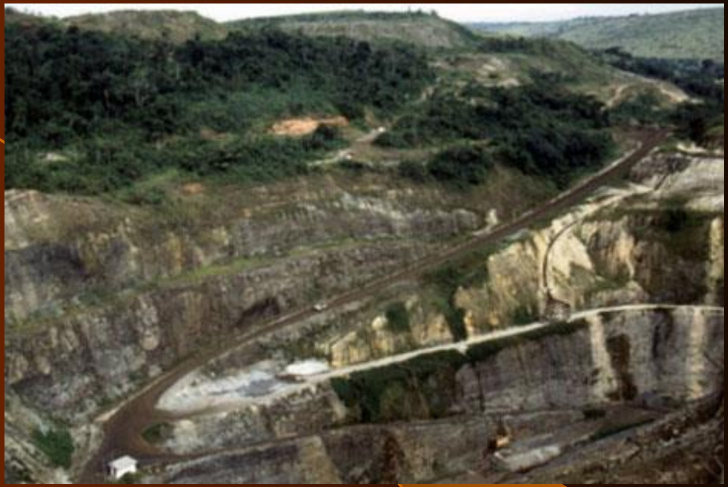

Entrance to Zone 15
Remains of Zone 2

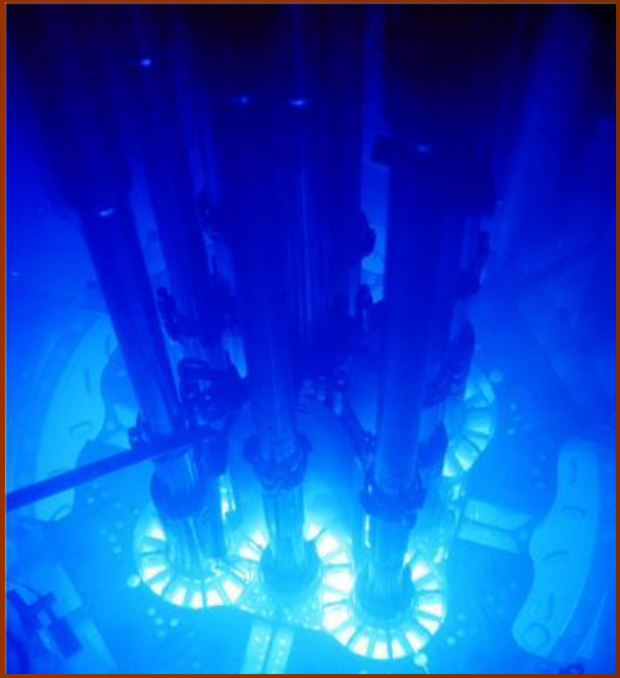
Concordia plot showing U/Pb isotope analyses by SIMS of uraninite grains from reactor 10.



Summary of the Oklo fission event

- Age - +/- 2.0 Ga
- Duration - 200 - 800 ka
- Fluence - mean 3.6 x 10^{20} n cm^{-2}
- Flux density - 10^7 - 10^8 n cm^{-2} s^{-1}
- Burn-up - 10,000 - 25,000 MW d tU^{-1}





Comparison to modern power reactor

- Burn-up - 33,000 MW d tU^{-1}
- Flux density - 10^{13} - 10^{14} n cm^{-2} s^{-1}

Effect of the fission reactions on the surrounding rock

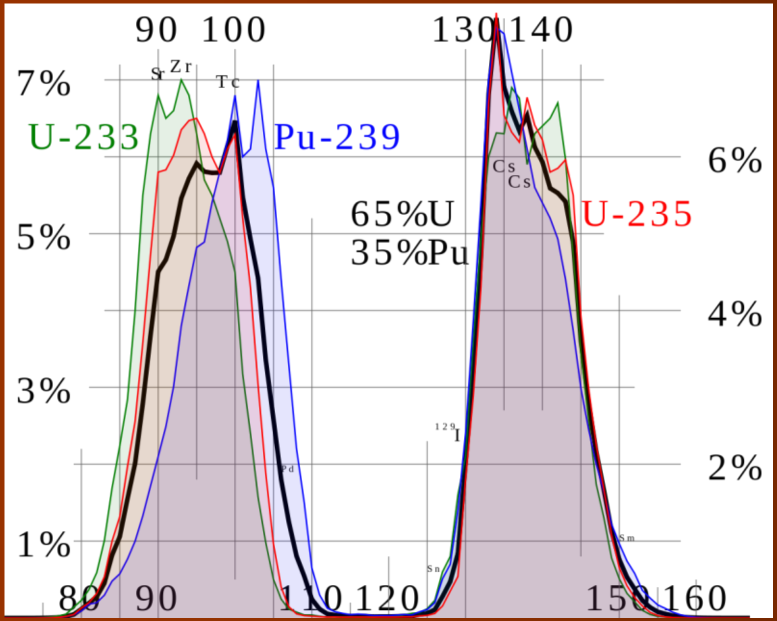
- Uraninite (UO_2) was essentially unaltered during the fission events and has suffered little alteration during the last 2 Ga.
- The normal sandstones (90% quartz and 10% clays) have been altered by the hot waters associated with the fission reactions. This alteration has led to desilification (essentially 100% removal of quartz from the rock) and its replacement by chlorite and various polymorphs of illite (a clay mineral).

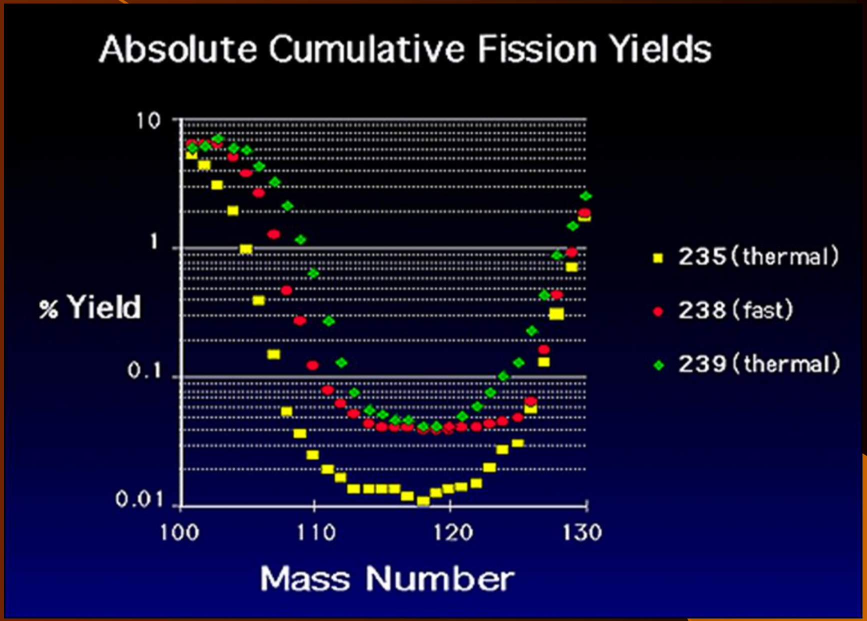
What has happened to the various radionuclides formed during the fission events?

- Retained at reactor site
- Migrated from reactor site but were retained in the immediate geologic environment
- Totally removed from reactor site and immediate geologic environment

The analysis - let the good times begin!

Fission products are concentrated in the mass range 80-110 and 130-150. Fission yields depend on the reaction path. For example, use the abundance of various Pd isotopes to estimate the relative importance of each path.





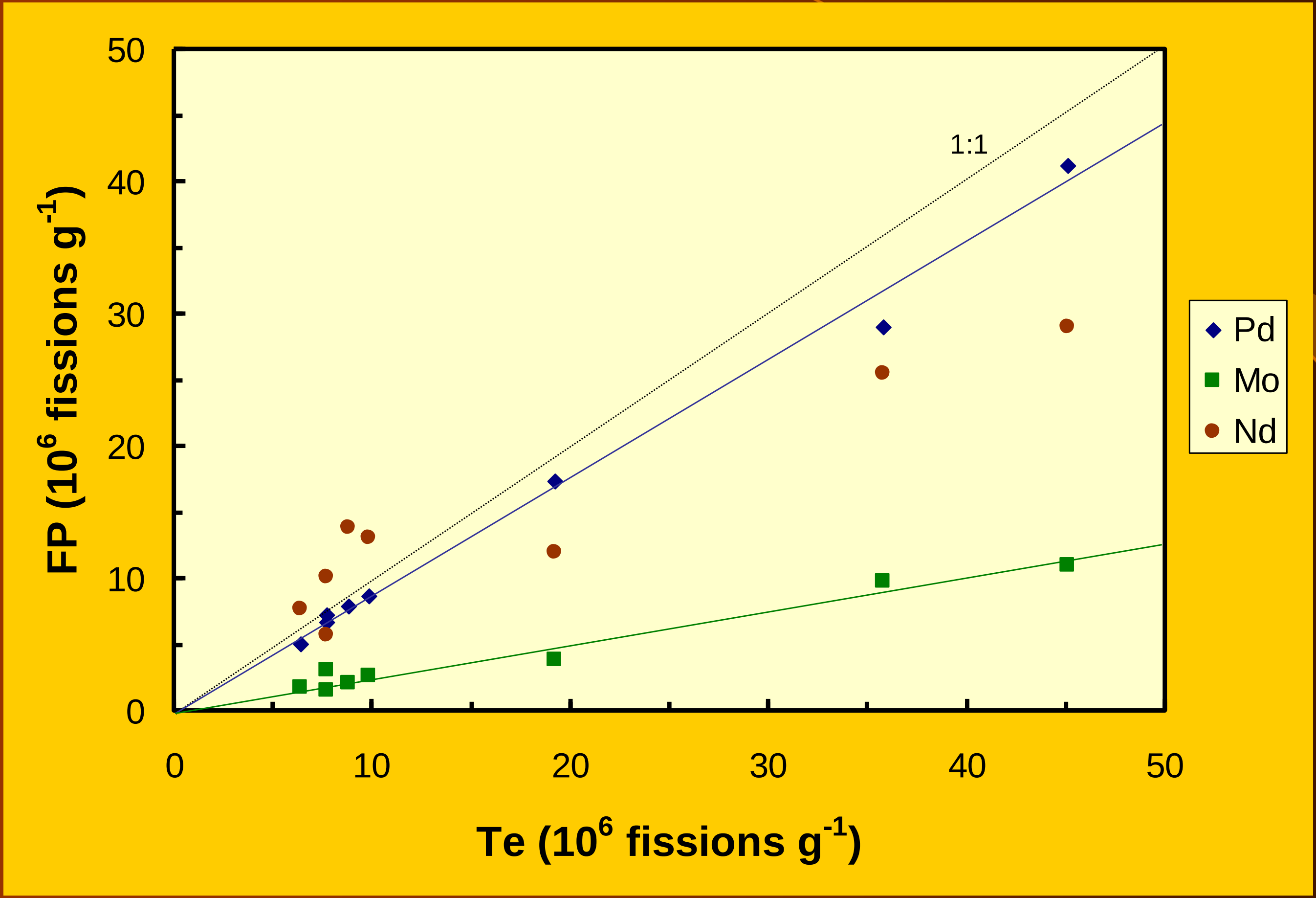
Fission Pathway	
Isotope	%
^{235}U	87.7
^{238}U	7.8
^{239}Pu	4.5

Palladium isotopic compositions (atom%)

	^{102}Pd	^{104}Pd	^{105}Pd	^{106}Pd	^{108}Pd	^{110}Pd
Primordial						
	1.04	11.3	22.5	27.4	26.3	11.5
Fissiogenic						
^{235}U			68.2	28.4	3.66	1.8
^{239}Pu			44.2	34.2	16.6	4.9
^{238}U			55.8	33.9	8.4	1.9

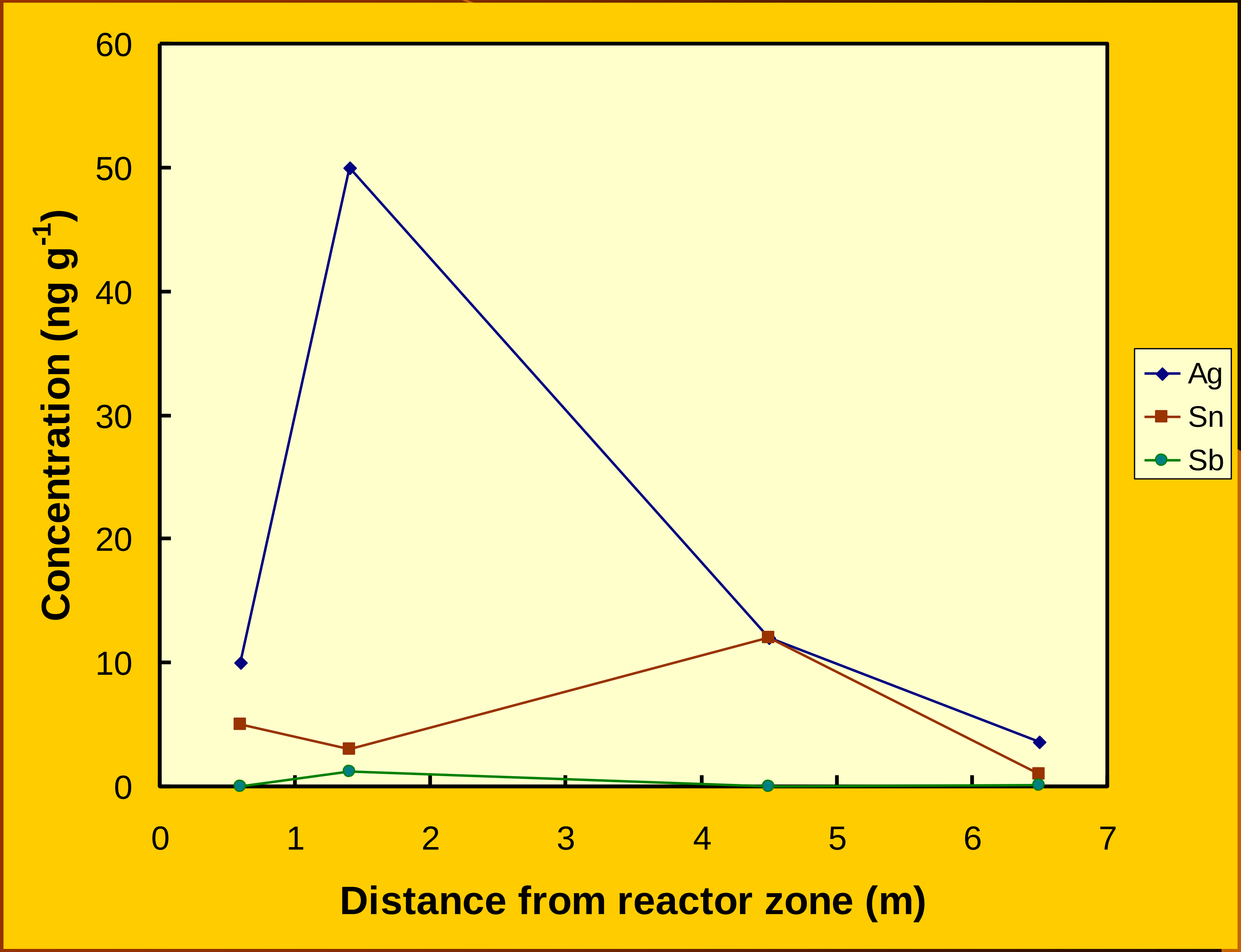
- Determine primordial amount by normalizing to isotopic ratios in primordial material
- Difference is the amount produced by the fission process
- Normalize this amount to the fission yield

Retention of fission products in reactor zones



Fission Product Retention in Reactor Zone	
FP	Retention
Te	1
Ru	0.91
Pd	0.88
Nd	0.83
Ag	<0.34
Mo	0.26
Sn	<0.26
Cd	0.002

Distribution of fission products around reactor zone



Estimates of solubilities (mg L^{-1}) of important radioisotopes at 25°C and 1 atm*

	Reducing conditions		Oxidizing conditions	
	Eh = -0.2 V		Eh = +0.2 V	
Element	pH 9	pH 6	pH 9	pH 6
Sr	0.6	high	0.6	high
Cs	high	high	high	high
Tc	10^{-10}	high	high	high
I	high	high	high	high
U	10^{-3}	10^{-6}	high	high
Np	10^{-4}	10^{-4}	10^{-2}	10^{-1}
Pu	10^{-5}	10^{-4}	10^{-5}	10^{-3}
Am	10^{-8}	10^{-5}	10^{-8}	10^{-5}
Ra	10^{-3}	10^{-1}	10^{-3}	10^{-1}
Pb	10^{-1}	1	10^{-1}	1

*From Krauskopf (1986)

Retardation factors for radioactive isotopes in various geomedia*

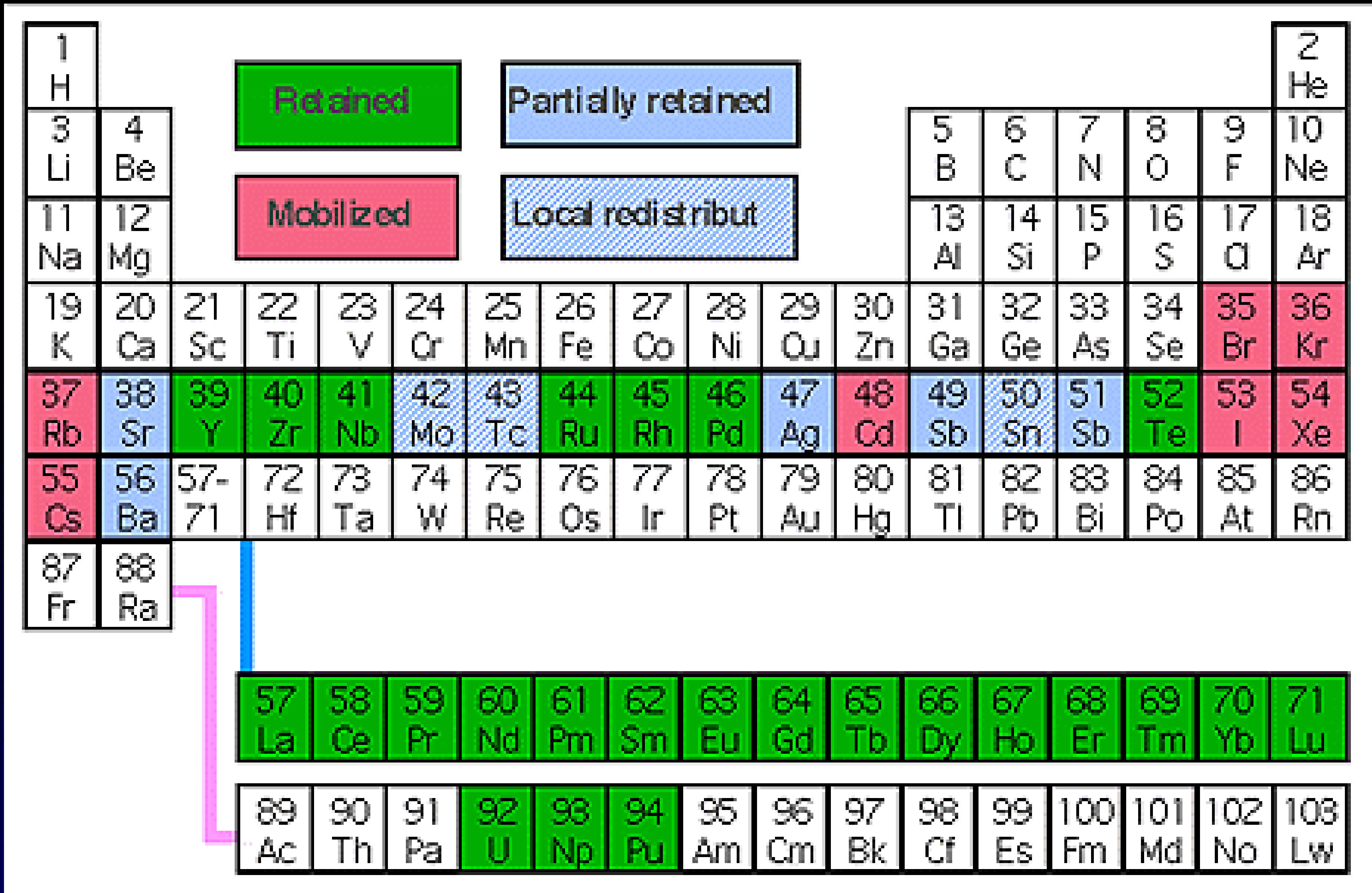
Element	Granite	Basalt	Volcanic ash	Shale (or clay)
Sr	20 - 4000	50 - 3000	100 - 100000	100 - 100000
Cs	200 - 100000	200 - 100000	500 - 100000	200 - 100000
Tc	1 - 40	1 - 100	1 - 100	1 - 40
I	1	1	1	1
U	40 - 500	100 - 500	40 - 400	100 - 2000
Np	20 - 500	20 - 200	20 - 200	50 - 1000
Pu	20 - 2000	20 - 10000	20 - 5000	50 - 100000
Am	500 - 10000	100 - 1000	100 - 1000	500 - 100000
Ra	50 - 500	50 - 500	100 - 1000	100 - 200
Pb	20 - 50	20 - 100	20 - 100	20 - 100

*From Krauskopf (1986)

$R_d = x_{non\text{-}absorbed}/x_{absorbed}$ $v_{contaminant} = v_{non\text{-}absorbed}/R_d$

$v_{Sr} = 100\ m\ y^{-1}/100 = 1\ m\ y^{-1}$

Retention of Fission Products at OKLO



Factors that favor retention of radionuclides in the natural environment

- Radionuclides contained in a refractory mineral(s). Uraninite in the case of Oklo
- Presence of clay minerals which adsorb radionuclides
- Natural materials that have a low permeability
- Absence of faulting
- Geologically stable region
- Low redox potential and high pH environment

The proposed (but no longer funded) waste repository at Yucca Mountain

How well does it fit the geologic criteria?

Volcanic activity has occurred more or less continuously in this region from about 15 Ma to the present.

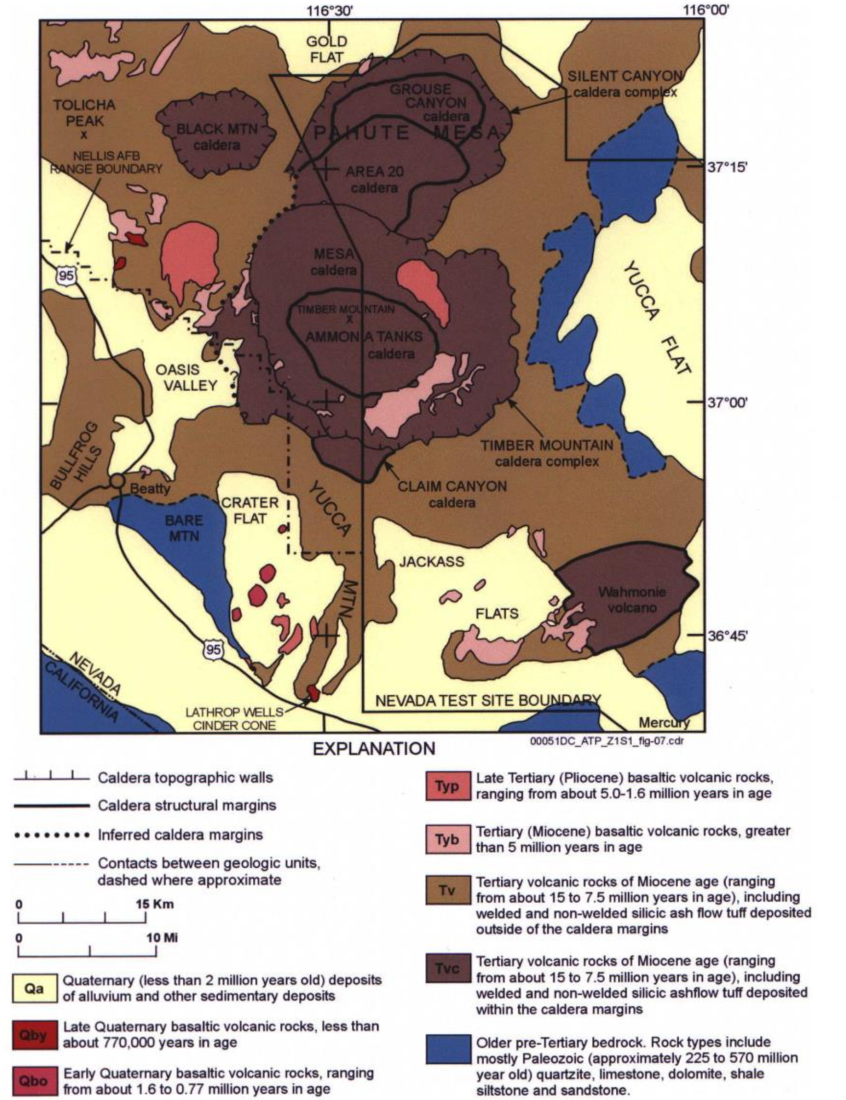


Figure 1-8. Simplified Geologic Map Showing the Location of Yucca Mountain in Relation to the Southwestern Nevada Volcanic Field

The map shows the location of (1) pre-Tertiary basement rocks (mostly Paleozoic sedimentary rocks ranging from 225 to 570 million years in age); (2) Tertiary volcanic rocks of Miocene age (i.e., 15 to 7.5 million years old) deposited during very large, caldera-forming eruptions; (3) Tertiary and Quaternary basaltic rocks of the southwestern Nevada volcanic field formed during much smaller volcanic eruptions, with ages ranging from more than 5 million years to about 77,000 years; and (4) Quaternary deposits of alluvium and other sedimentary deposits less than 2 million years old.

Faults occur throughout the area and in many places extend to basement cutting across all the volcanic units.

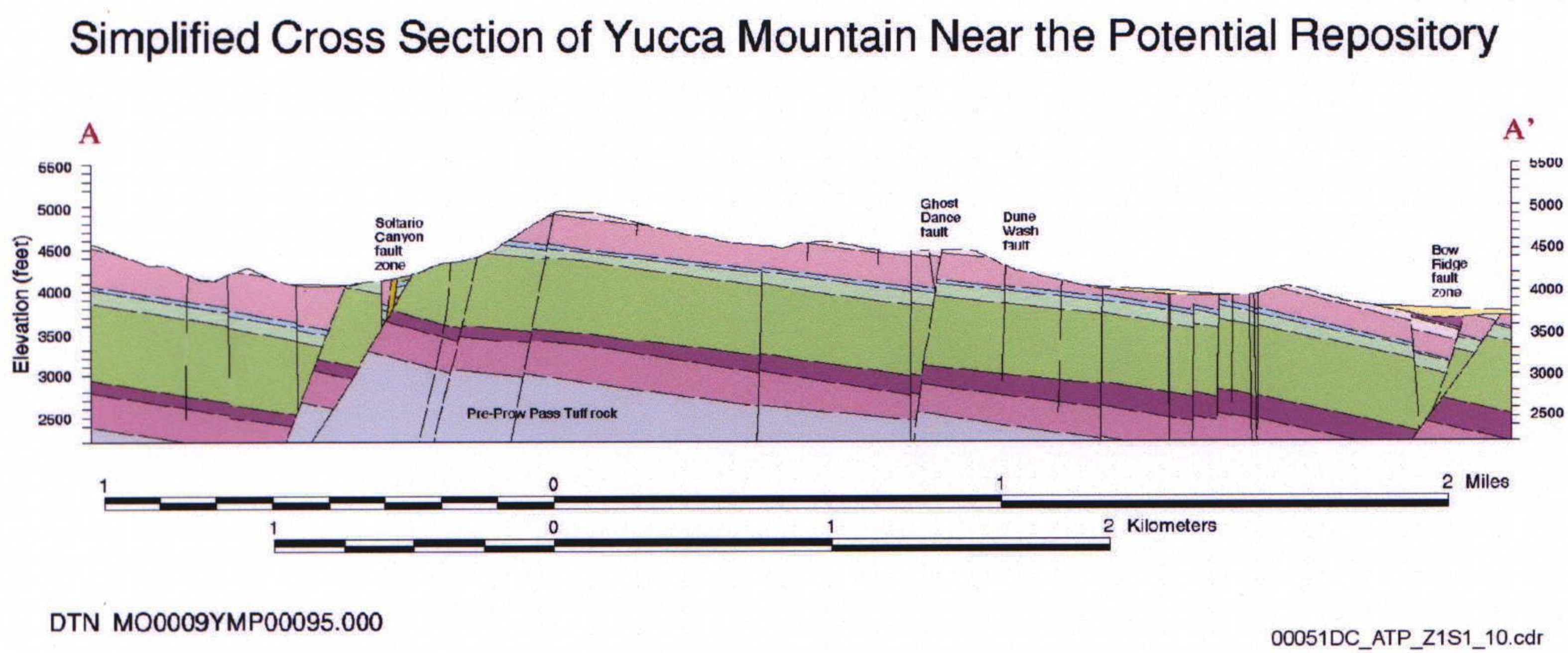


Figure 1-10. Simplified Cross Section of Yucca Mountain Near the Potential Repository
The cross section shows the distribution of the eastward dipping volcanic rocks that form Yucca Mountain, and the locations of selected faults. The location of this cross section, A–A', is indicated in Figure 1-9, and the legend in Figure 1-9 corresponds to this geologic cross section. Source: Modified from Day, Dickerson et al. 1998.

Strike-slip, reverse, and normal faults are abundant throughout the region and many show recent seismicity.

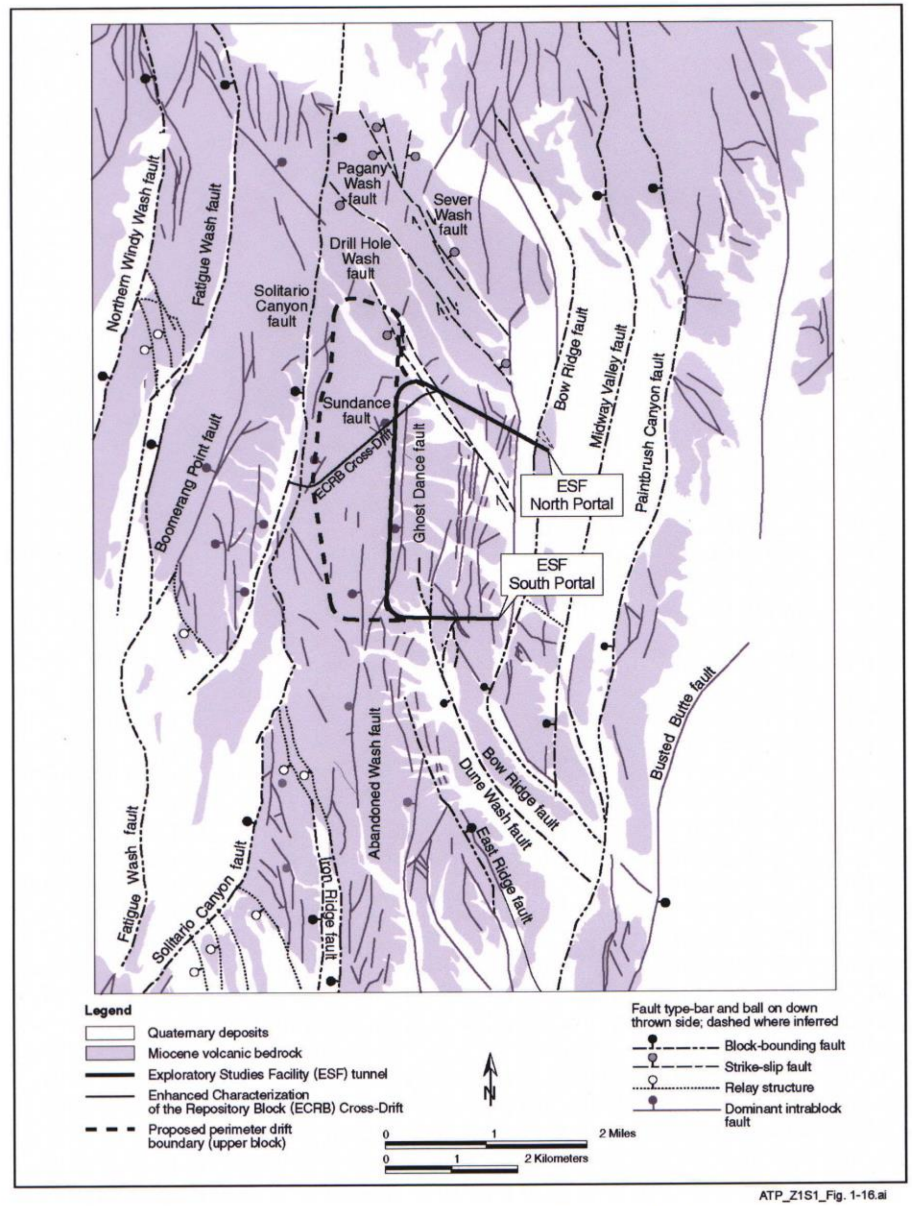


Figure 1-14. Mapped Faults at Yucca Mountain and in the Yucca Mountain Vicinity
This map shows the location of various types of faults, as described in Section 4.6.4 of the *Yucca Mountain Site Description* (CRWMS M&O 2000c).

Geology of the repository

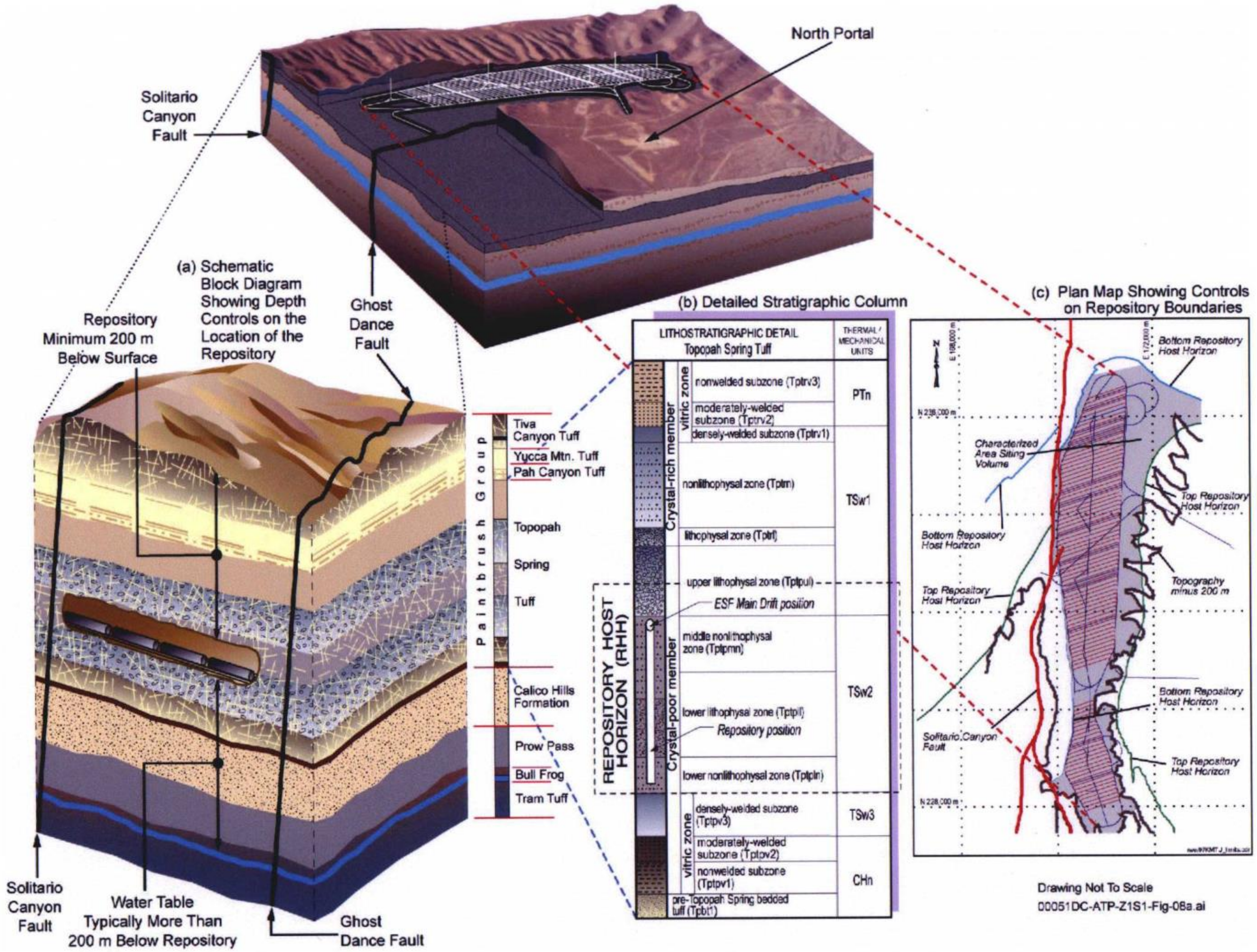


Figure 1-11. Layout and Boundaries of the Potential Repository
(a) Block diagram showing the location of the potential repository with respect to depth below the surface, the water table, and nearby faults. The scale of the emplacement drifts is exaggerated to show schematic waste packages. (b) Stratigraphic section showing the rock units in the Topopah Spring welded tuff that could host emplacement drifts. (c) Plan map showing how the lateral boundaries of the potential repository were identified based on the combined constraints of all of the factors. ESF = Exploratory Studies Facility.

Contours of surface of regional water table. The water table slopes to the NW indicating the direction of groundwater flow and is on average about 200 m below the repository.

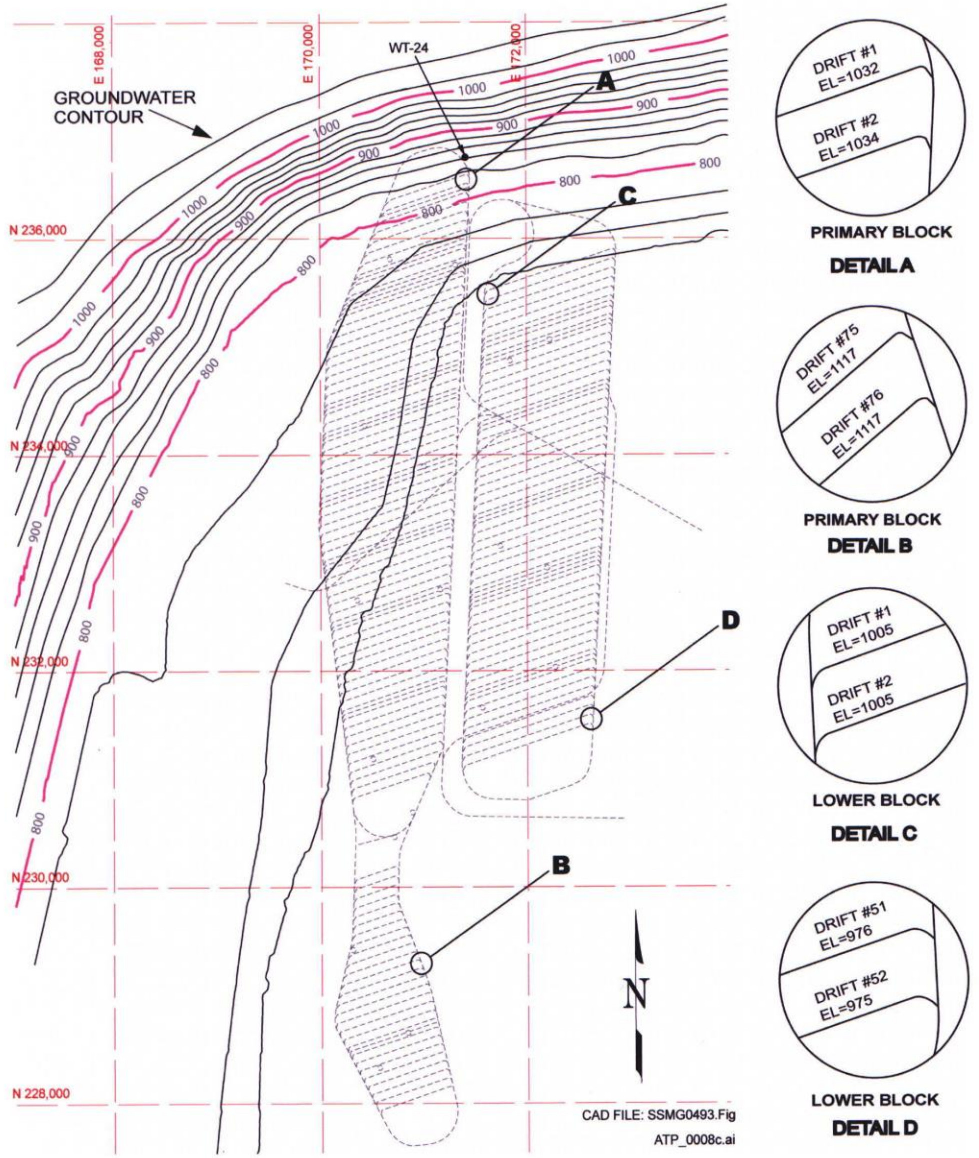
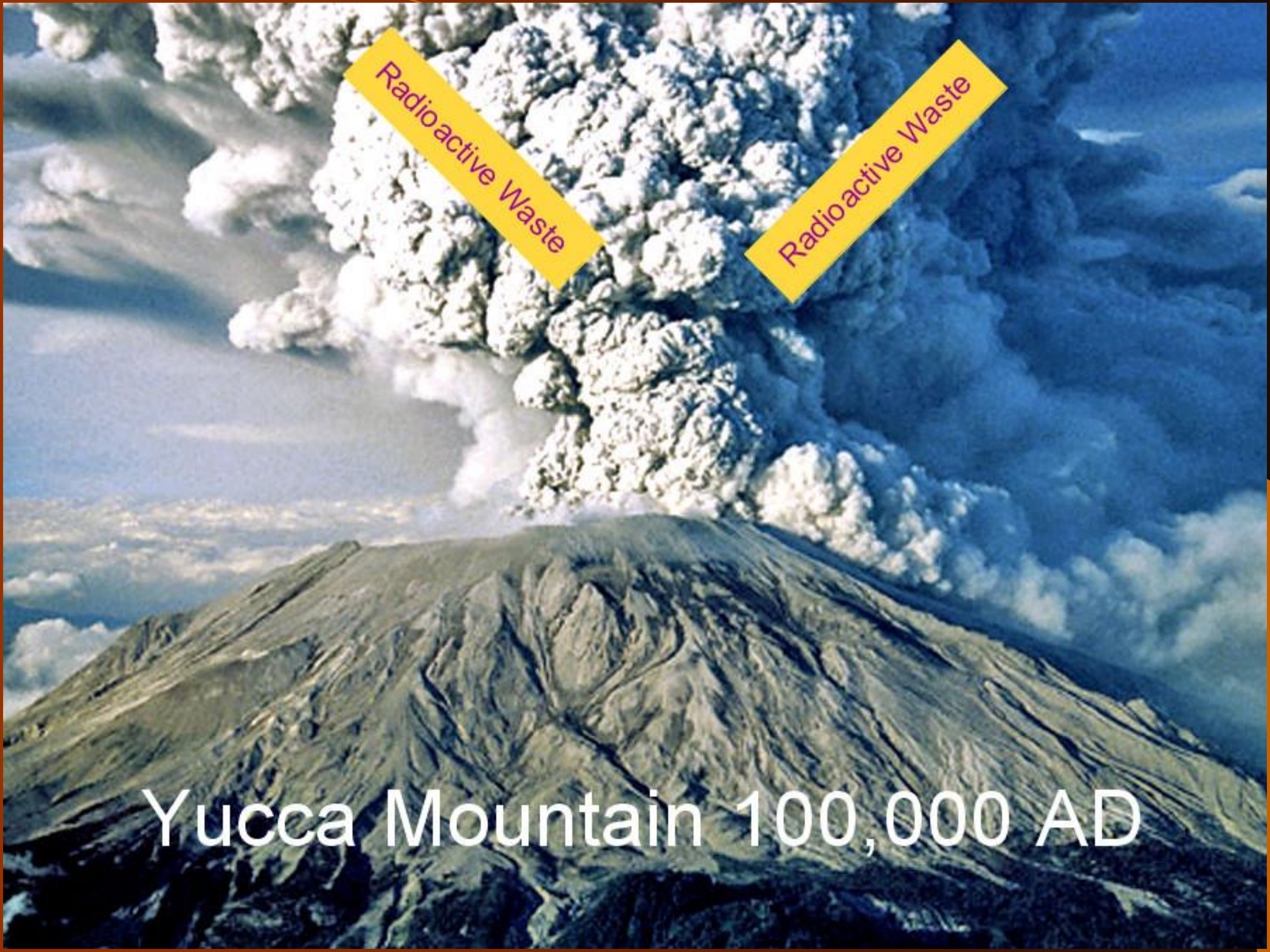


Figure 1-13. Groundwater Elevation Contours, with Their Relationship to a Conceptual Repository Layout
The water table in the vicinity of Yucca Mountain shows a steeper gradient toward the north and northwest. The northernmost emplacement drifts in the primary block area would be closest to the water table, the closest emplacement drift being approximately 210 m (690 ft) above it. The areas where the emplacement drifts in the primary and lower blocks would be closest to (locations A and C, respectively) or farthest from (locations B and D, respectively) the water table have been identified in the figure. EL = elevation. Elevations are given in meters above sea level. Groundwater contours are shown at intervals of 20 m (66 ft). Source: BSC 2001a.

Comparison of the Yucca Mountain site to factors that favor retention of radionuclides

- Radionuclides contained in a refractory mineral(s). Uraninite in the case of Oklo (Y)
- Presence of clay minerals which adsorb radionuclides (N)
- Natural materials that have a low permeability (N)
- Absence of faulting (N)
- Geologically stable region (N)
- Low redox potential and high pH environment (N)


Radioactive Waste
Radioactive Waste
Yucca Mountain 100,000 AD

So where and how should we sequester reactor wastes?

- A third world country that's so desperate for hard currency that they'll take anything. (N)
- A stable continental interior. (Y)
- Old terranes often have a high fault/fracture density because of multiple tectonic cycles, but careful field geology can demonstrate whether or not any of these faults have been active in the last 10s of millions of years. (Y)
- Crystalline rocks in general (granites, gneisses) would be excellent choices for a waste site. (Y)
- In terms of engineering solutions, encapsulation of the waste (we really do know how to do this) plus multiple barriers within the repository (such as smectite clay layers) will ensure long term storage. (Y)