

Radioactivity

Lecture 18

Radioactivity in Natural Resources

Natural Resources

Renewables

Sun

Wind

Changeables

Land

Forests

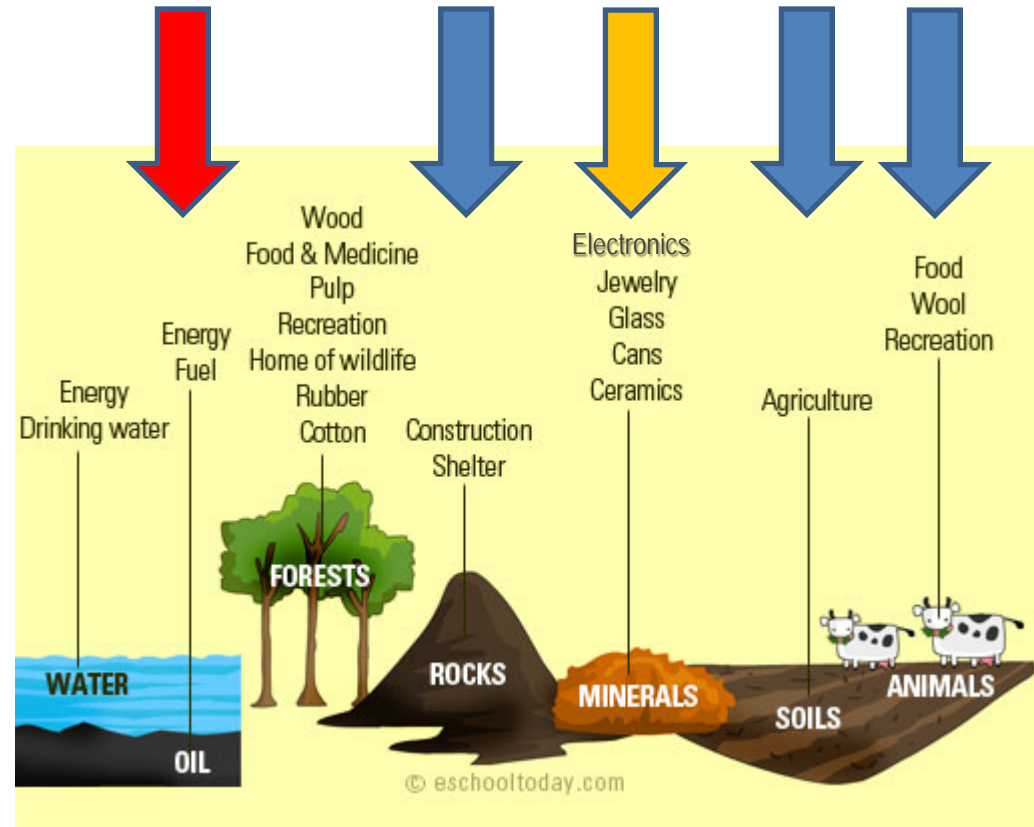
Water

Non-renewables

Materials

Minerals

Energy Fuel



The discussion will concentrate on radioactivity associated with non-renewable minerals and fossil energy fuels (except nuclear energy fuel).

Public Fears

Concerns about CO₂ emission and its impact on climate is complemented by claims of high levels of radioactivity



Coal Ash Is More Radioactive than Nuclear Waste



The waste produced by coal plants is actually more radioactive than that generated by their nuclear counterparts. In fact, the fly ash emitted by a power plant—a by-product from burning coal for electricity—carries into the surrounding environment 100 times more radiation than a nuclear power plant producing the same amount of energy.

Reconsider Nuclear

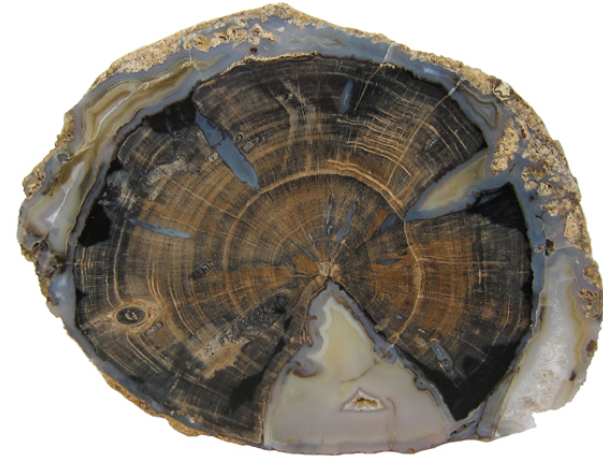
Think.

Radioactive Coal Ash Pollutes Regional Groundwater

Fossil fuels

- Fossil fuel is primarily fossilized organic matter (biomatter). Coal is fossilized wood, depending on age, stone coal or brown coal.
- Oil is un-fossilized, but younger biomaterial liquefied under high pressure.
- Natural Gas are organic gas components that were generated in the fossilization or liquefaction process and trapped in underground caves and/or rock material

Fossilization is a process by which mineral rich water diffuses into the biomaterial, this includes also radioactive ^{238}U , ^{232}Th , and ^{40}K mineral components because of their long lifetime. The fossilization process takes millions of years. The diffusion probability depends on the environmental conditions. On the other hand the original ^{14}C radioactivity in wood disappears because of the relative short half life of 5730 y. Fossil fuel burning does add carbon (in form of CO_2) to the atmosphere but not ^{14}C . The $^{14}\text{C}/\text{C}$ ratio in the atmosphere declines.



Age determination by FUN!
Ratio of fluorine, uranium, and nitrogen content in bones.

Radioactivity in Coal

Coal has a certain amount of inherent radioactivity. The level changes due to the nature of the mineralization process and the mineral content of the environment.

Local radioactivity content of coal in Bq/kg from different locations world-wide.

Origin	U-238	Ra-226	Pb-210	Po-210	Th-232	Ra-228	K-40
Australia	8.5-47	19-24	20-33	16-28	11-69	11-64	23-140
Brazil	72	72	72		62	62	
China	10-25, with local enrichments up to 5600				10-25, with local enrichments up to 29,000		
Germany (brown coal)		10-145, (32)			10-63 (21)		10-700, (225)
Greece	117-390	44-206	59-205			9-41	
Hungary	20-480					12-97	30-384
Poland	10-159 (18)					10-123 (11)	10-785
Romania	1- 415 (80)	10-557 (126)	10-510 (210)	10-580, (262)	10-170 (62)		
Great Britain	7-19	8-22			7-19		55-314
USA	6-73	8.9-59	12-78	3-52	4-21		

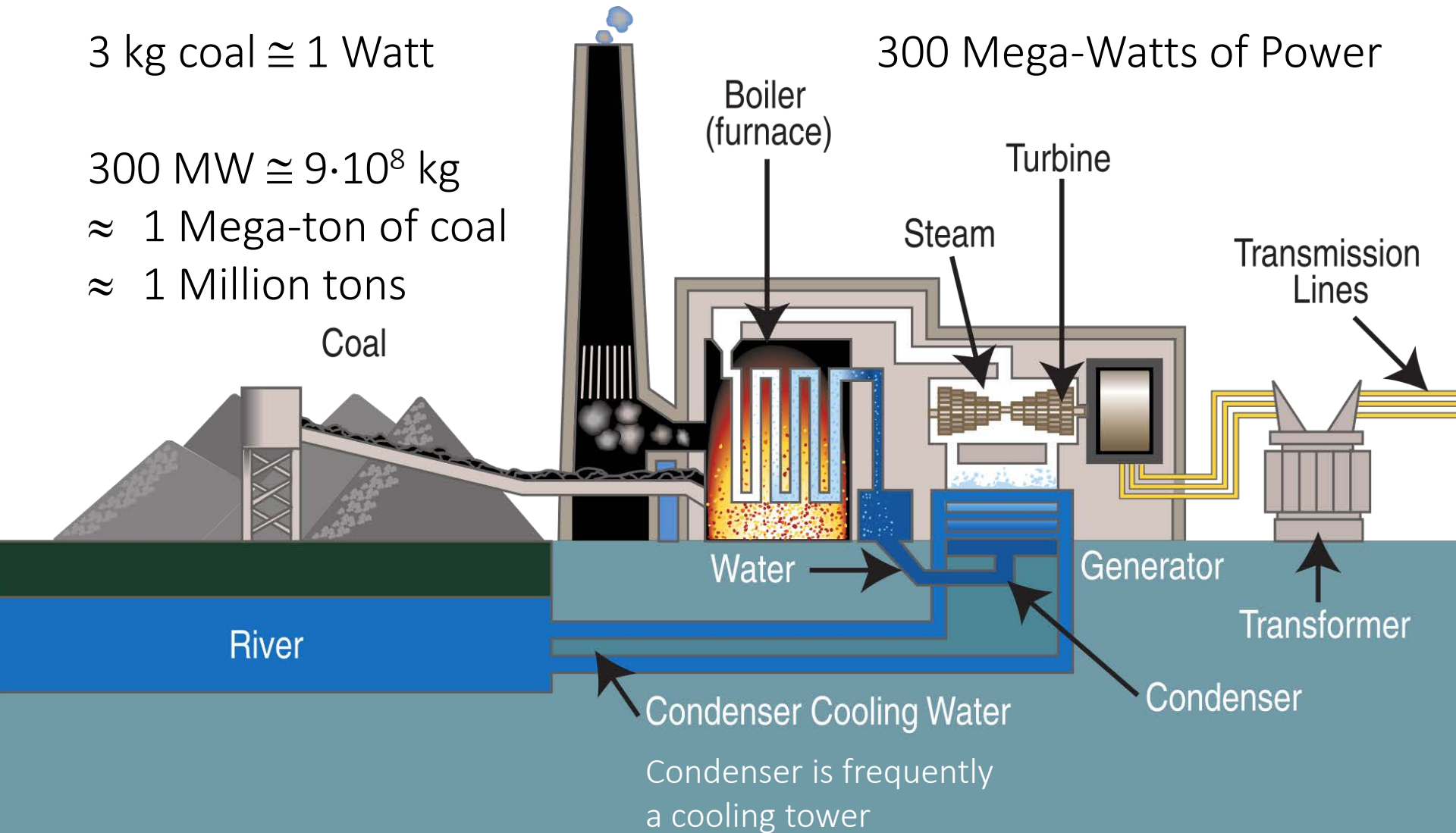
Average total activity of US coal is about 57 Bq/kg which converts to 5.3 kBq/m³.

Operation of a Coal Power Plant

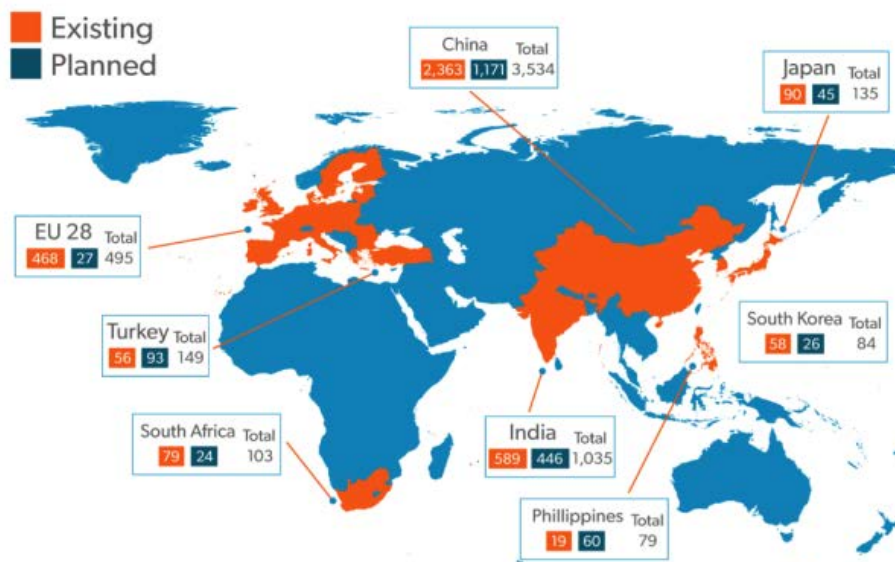
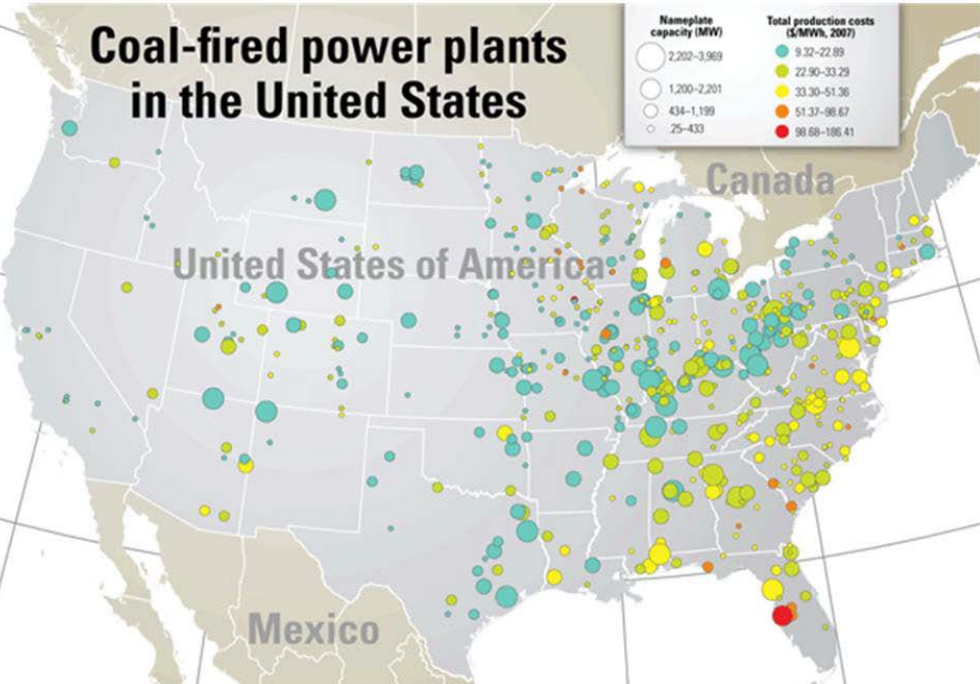
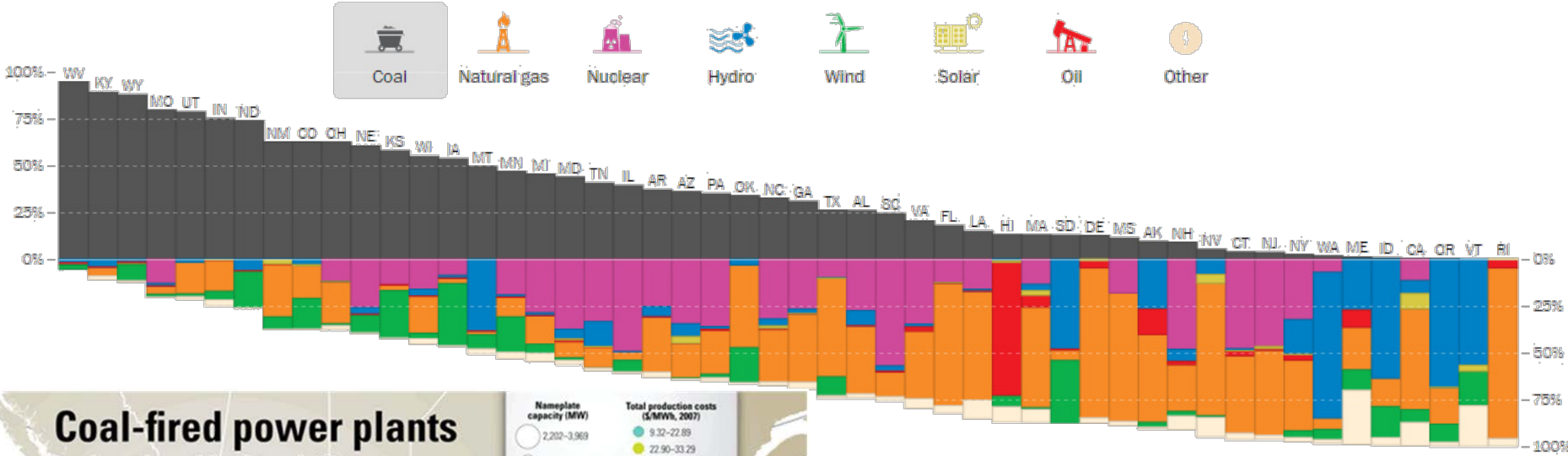
3 kg coal \cong 1 Watt

300 MW \cong $9 \cdot 10^8$ kg
 \approx 1 Mega-ton of coal
 \approx 1 Million tons

300 Mega-Watts of Power



Coal Power Distribution world-wide and in US



Source: Global Coal Plant Tracker; CoalSwarm; Platts WEPF

Annual radioactivity emission by coal power plants

The table provides different estimates of the output on radio-nuclides from coal power plants. The numbers are normalized to a power production of 1000 W. These values can be reduced by implementing efficient filters and absorbers between furnace and chimney exhaust.

Release Rates (Bq y^{-1}) of Radionuclides Contained in Flyash, from CFP, Normalized to 1 000 MWe

Reference	Release rates of radionuclides (Bq y^{-1})									
	^{238}U	^{226}Ra	^{210}Pb	^{210}Po	^{222}Rn	^{232}Th	^{228}Ra	^{220}Rn	^{235}U	^{239}Pu
Eisenbud & Petrow (1964)		6.4×10^8					4.0×10^8			
McBride <i>et al.</i> (1977)	3.0×10^8	3.0×10^8	3.0×10^8	3.0×10^8	3.0×10^{10}	1.8×10^8	1.8×10^8	1.5×10^{10}	1.2×10^7	1.2×10^7
Beck & Miller (1979) ^a	10^9	7.8×10^8	2.6×10^9	2.6×10^9	5.5×10^{10}	4.1×10^8	4.1×10^8			
US EPA (1979)—old model ^b	7.8×10^8	7.8×10^8	1.6×10^{10}	1.6×10^{10}	5.3×10^{10}	6.7×10^9	6.7×10^9	4.2×10^{10}	3.7×10^8	2.3×10^9
US EPA (1979)—new model ^a	10^9	7.3×10^8	2.6×10^9	2.6×10^9	6.3×10^{10}	4.4×10^8	6.7×10^8	5.3×10^{10}	5.2×10^7	3.0×10^9
Okamoto (1980)			7.4×10^{10}	7.4×10^{10}						
Camplin & Hallam (1980)	10^9	10^9	10^9	10^9	10^9	10^9	10^9	10^9	3.7×10^7	3.7×10^7
Teknekron (1981)—old model ^b	2.3×10^9		4.8×10^9	4.8×10^9	3.2×10^{10}	1.7×10^9		2.4×10^{10}		
Teknekron (1981)—new model ^a	7.0×10^8	5.2×10^8	1.7×10^9	1.7×10^9	3.6×10^{10}	2.5×10^8	3.8×10^8	2.7×10^{10}		
IAEA (1982) and UNSCEAR (1982) average values	1.5×10^9	1.5×10^9	5.0×10^9	5.0×10^9		1.5×10^9	1.5×10^9			
UNSCEAR (1982)—exceptional values	7.0×10^8 – 1.8×10^{10}	1.8×10^{10}	2.6×10^{10}			2.6×10^{10}				
Corbett (1983)—modern CFP ^a	8.0×10^8	8.0×10^8	8.0×10^9	8.0×10^9		4.0×10^8	4.0×10^8			
Mishra <i>et al.</i> (1984)		4.5×10^8 – 2.6×10^{10}								
De Santis & Longo (1984)	$<2.2 \times 10^8$	$<2.2 \times 10^8$				$<2.2 \times 10^8$	$<2.2 \times 10^8$			
Nakaoka <i>et al.</i> (1984)	7.6×10^7 – 9.0×10^7		3.7×10^9	3.7×10^9		8.1×10^7 – 10^8	8.1×10^7 – 10^8			

^aModern CFP, i.e. equipped with sophisticated devices, from which only about 1% of the ash is released into the atmosphere.

^bCFP which releases about 10% of the ash into the atmosphere.



1800°C – 3272 F
3 kg coal ⇒ 1 Watt

According to UNSCEAR1982, the release is $95 \text{ GBq/y/1000 MW} = 9.5 \cdot 10^{11} \text{ Bq/y/1000 MW}$
The 10 largest US coal power plants generate $200 \text{ TW/y} \Rightarrow \text{US coal releases } 2 \cdot 10^{17} \text{ Bq/y} = 200 \text{ PBq/y}$

Fly Ash from Chimney Emission

Component	Bituminous	Subbituminous	Lignite
SiO ₂ (%)	20-60	40-60	15-45
Al ₂ O ₃ (%)	5-35	20-30	20-25
Fe ₂ O ₃ (%)	10-40	4-10	4-15
CaO (%)	1-12	5-30	15-40
LOI (%)	0-15	0-3	0-5

Uranium and thorium are concentrated at up to 10 times their original levels.

Material	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K	Total
	Bq/kg				
coal	20	25	20	50	115
fly-ash	200	240	70	265	775
slag	70	80	34	320	504

HV 12.50 kV
HFW 398 μm
mag 750 x
WD 8.6 mm
det DualBSD

200 μm
Class C Fly Ash

Releasing and Recycling



Modern filters remove 95-99% of fly ash, activity on Bq/W

CFR#	Fly-ash removal efficiency %		²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
Modern (19)	95 - 99.8	Average	8.4	10.0	3.65	32.2
		Range	0.3-33.1	0.5-36.4	0.14-19.1	1.3-127.1

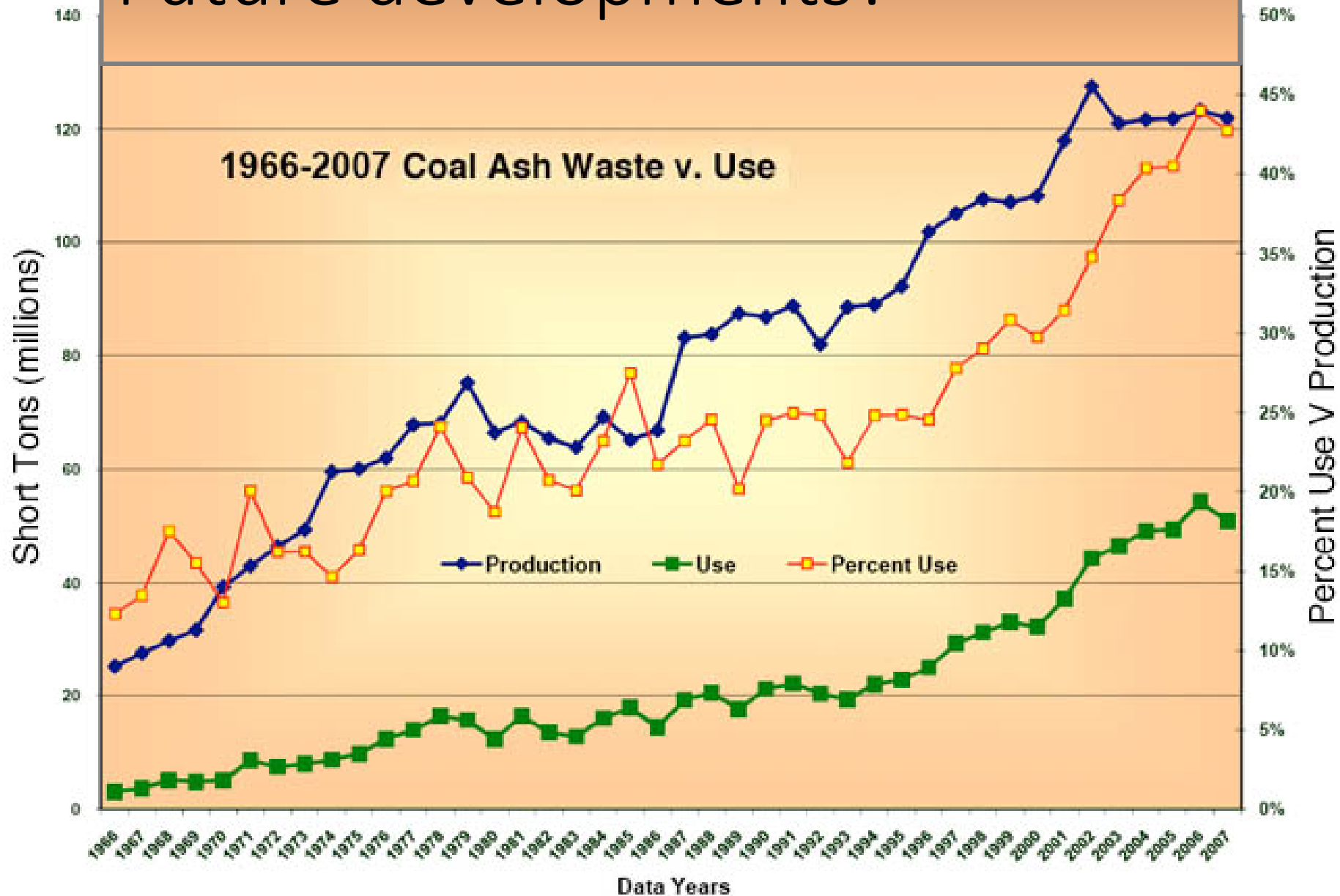
An old 2000 MW coal powered plant released each year 1900 GBq into the atmosphere!
A modern 2000 MW coal powered plant releases each year 109 GBq into the atmosphere!
 The annually released fly ash carries between 1-90 μ Sv with an average value of 12 μ Sv/y
 Filtered fly ash is collected and either goes to landfill or is recycled for applications.
 As of 2005, U.S. coal-fired power plants reported producing 71.1 million tons of fly ash, of which 29.1 million tons were reused.

ASH		Activity concentration (Bq kg ⁻¹)			
(no of sample)		²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
Collected fly-ash (27)	Average	100.0	129.3	49.6	421
	Range	25-223	50-221	3-86	96-885
Disposed ash (27)	Average	88.6	107.7	41.9	403
	Range	17-230	21-227	5-86	83-1,097
Slag (20)	Average	68.5	79.0	33.7	317
	Range	6-161	15-159	3-84	71-852



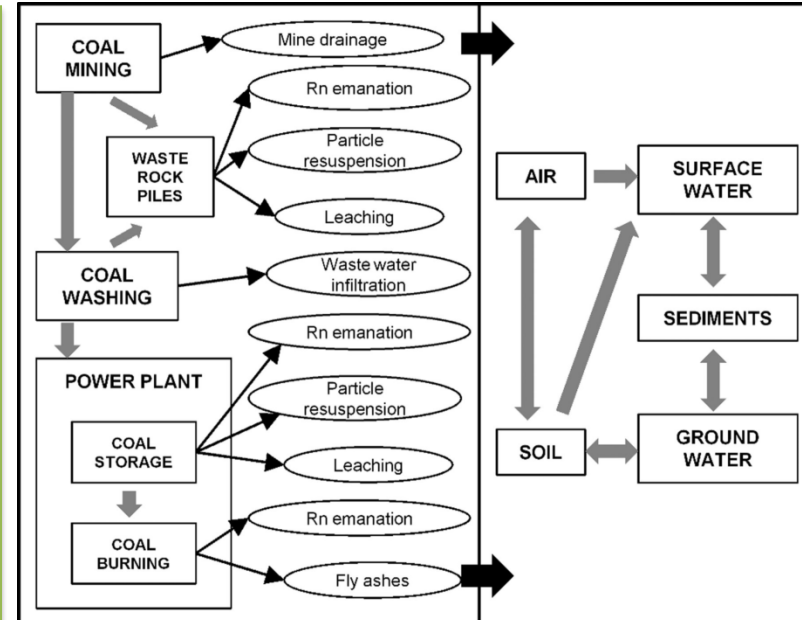
Applications are: cement and concrete production, road bedding and substrates, land stabilization. These materials carry a dose level of up to 250 μ Sv/y.

Future developments?



Environmental measurements of natural radionuclides from a coal-fired power plant

Sample	^{238}U	^{226}Ra	^{224}Ra	^{210}Pb	^{232}Th	^{40}K	D
NW-1	64±5	82±6	60±4	89±7	50±4	590±40	84
NW-2	64±5	80±6	78±6	96±7	66±5	800±60	103
NEW _{average}	64	81	69	92	58	695	94
OW-1	28±2	42±3	58±4	49±4	50±4	660±60	71
OW-2	30±2	39±3	64±5	44±4	53±4	750±50	77
OW-3	33±3	41±3	71±5	72±6	61±4	860±60	84
OLD _{average}	30	41	64	55	55	757	77
NEW/OLD	2.1	2.0	1.1	1.7	1.1	0.9	—
ROM Coal	32	40	36	51	30	330	47



Activity concentrations (in Bq kg^{-1}), activity ratios and air absorbed dose rate D (nGy h^{-1}) for waste samples and ROM coal (estimated values).

Coal Power Plant



US coal power plant in Kentucky



US coal power plant in Virginia

CO₂ is emitted, but also a fair fraction of unfiltered coal ash (depends on quality of filter). It is estimated that coal power plants world-wide are releasing 800.000 tons of Uranium and 2 Million tons of Thorium between 1940 and 2040.

This in turn translates to an emission of $1.4 \cdot 10^{17}$ Bq or 140 PBq for the uranium decay chain and $8 \cdot 10^{16}$ Bq or 80 PBq for the thorium decay chain into the atmosphere which eventually has settled down as dust in the local neighborhoods.

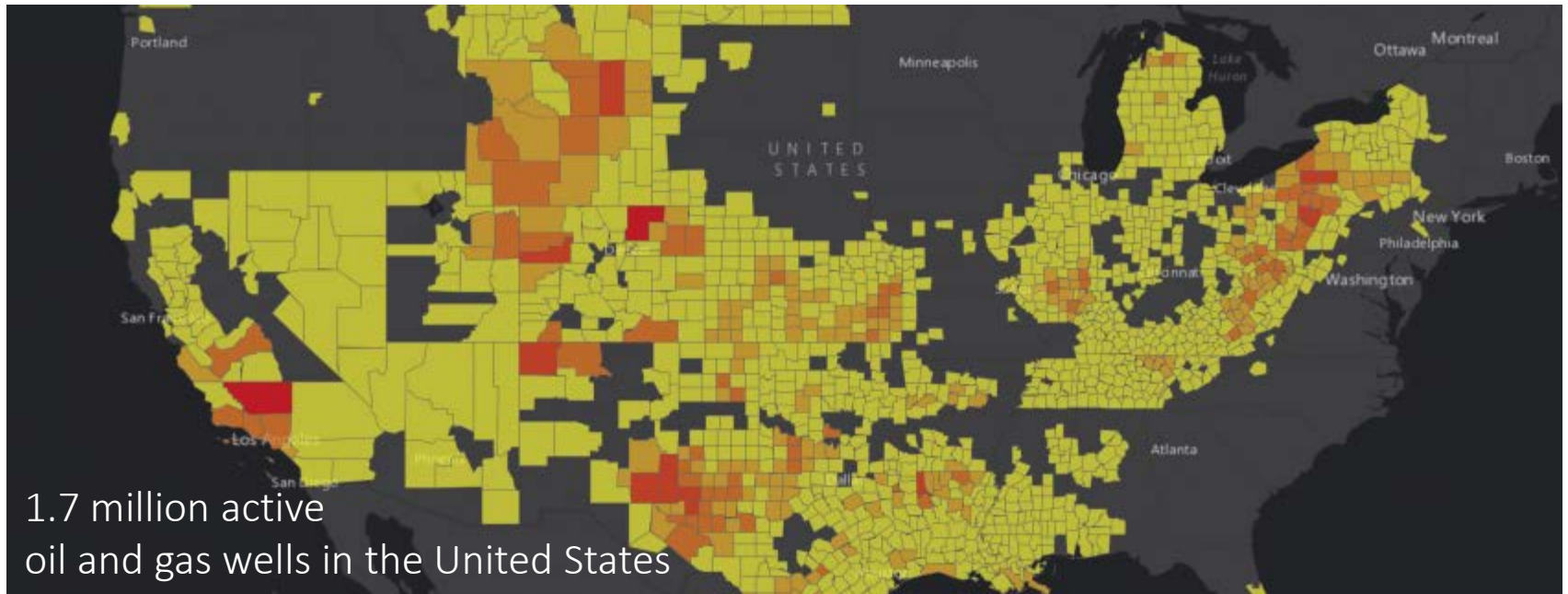
Oil and Gas



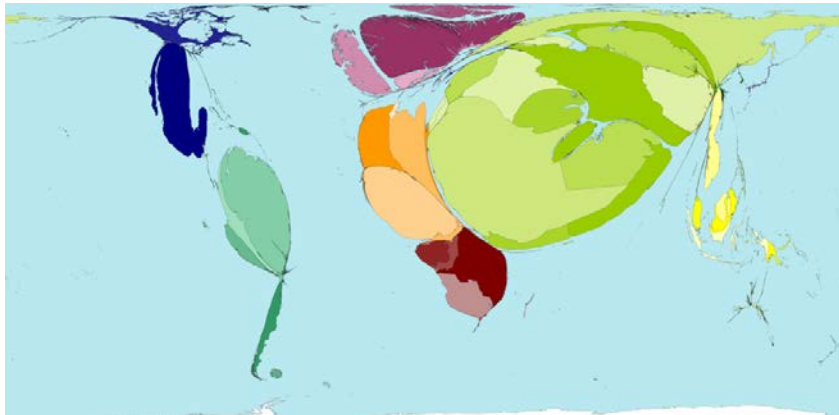
Oil is liquified biomaterial, compressed, complex organic hydrocarbon materials. The radioactivity is initial leftover from bio materials or migrated from surrounding rock material. The activity values for ^{238}U vary between 13 and 67 Bq/kg, similarly the values for ^{232}Th between 10 and 55 Bq/kg, the ^{40}K levels range from 5 to 600 Bq/kg.

According to UNSCEAR 2000 the average value for all oil fields is 33 Bq/kg for ^{238}U , 45 Bq/kg for ^{232}Th und 127 Bq/kg for ^{40}K .

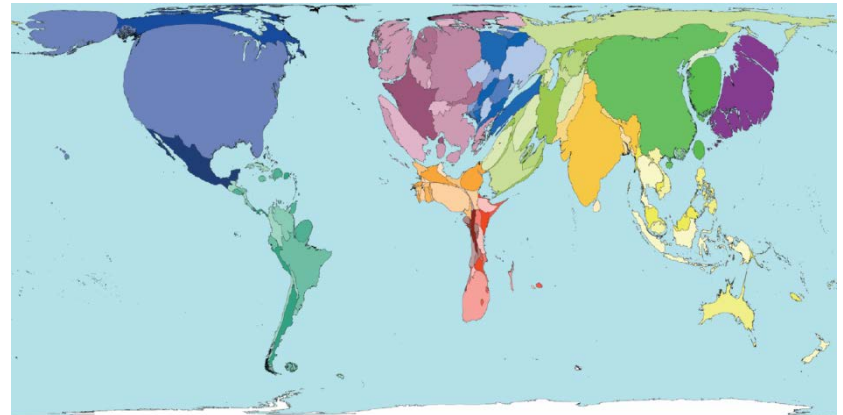
Active Oil and Gas fields in the United States



Oil producing countries



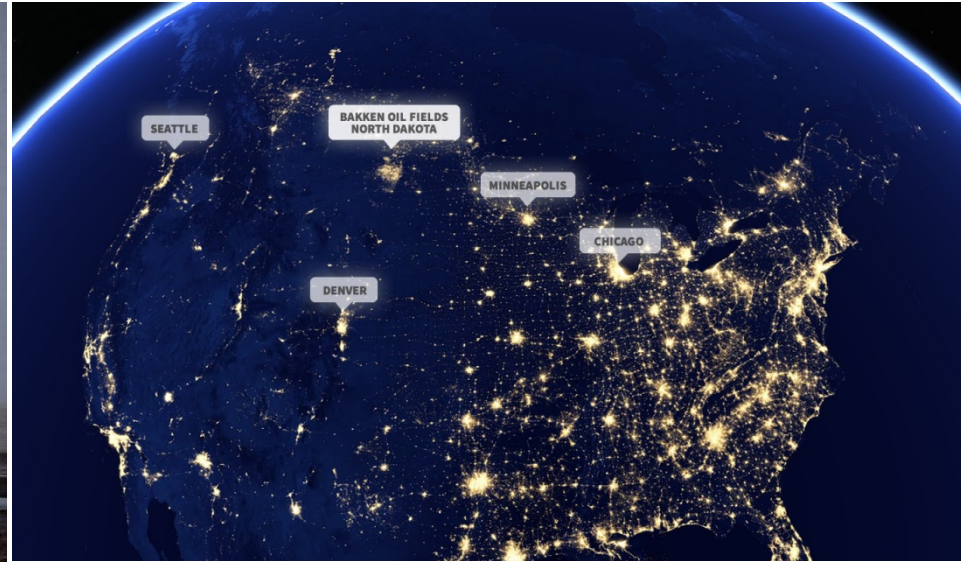
Oil using countries



Size of the country scales with its role in producing oil and using oil based products

US based Oil fields

Bakken Field, North Dakota



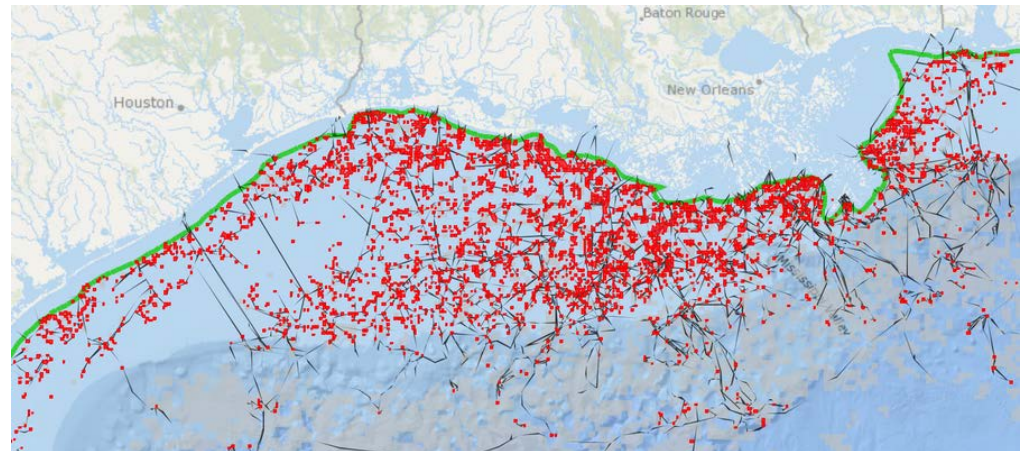
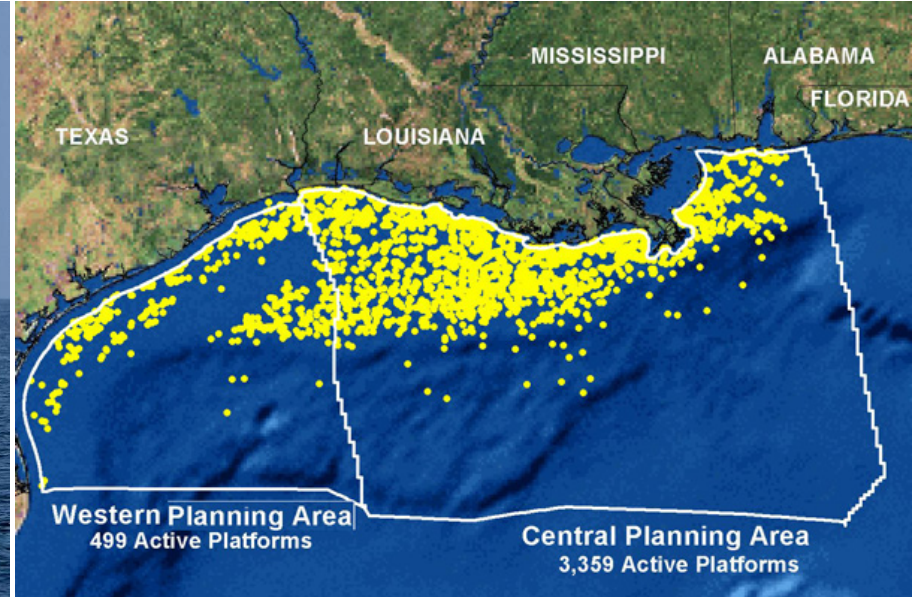
Bakersfield, California



Sun Field, Alaska

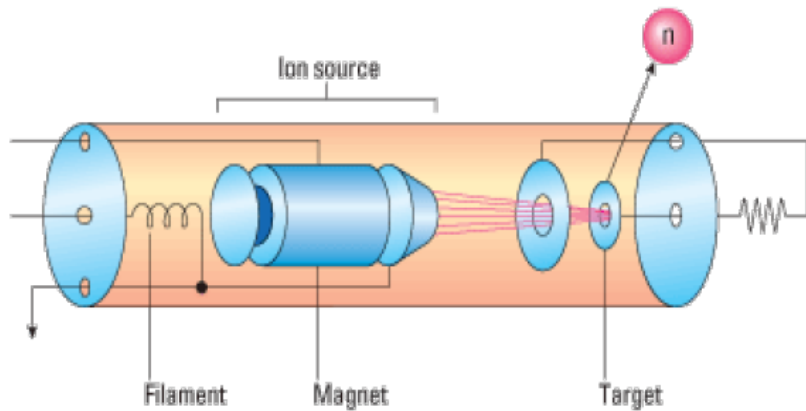


Off-Shore Drilling

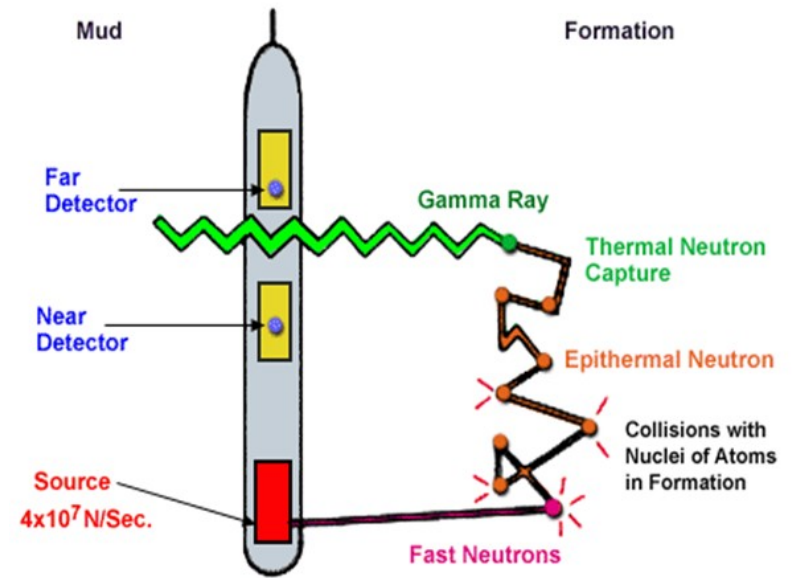


Map of incidents in the Gulf of Mexico fields

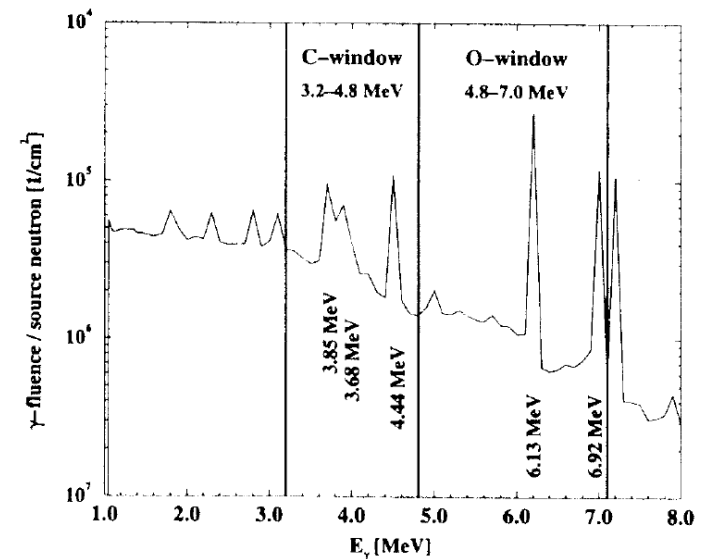
Oil Searching with Neutrons



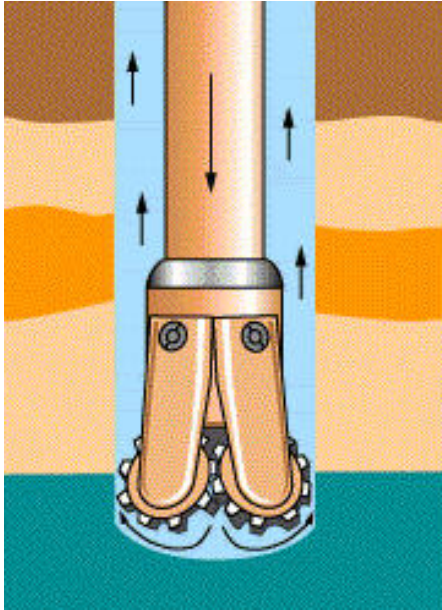
Neutron generator: A downhole accelerator used to emit pulses of high-energy neutrons into the formation.



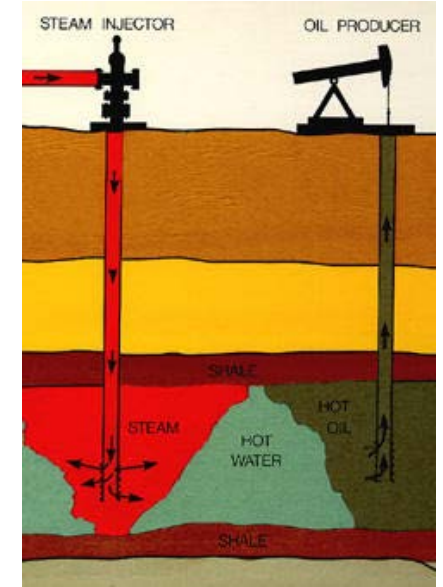
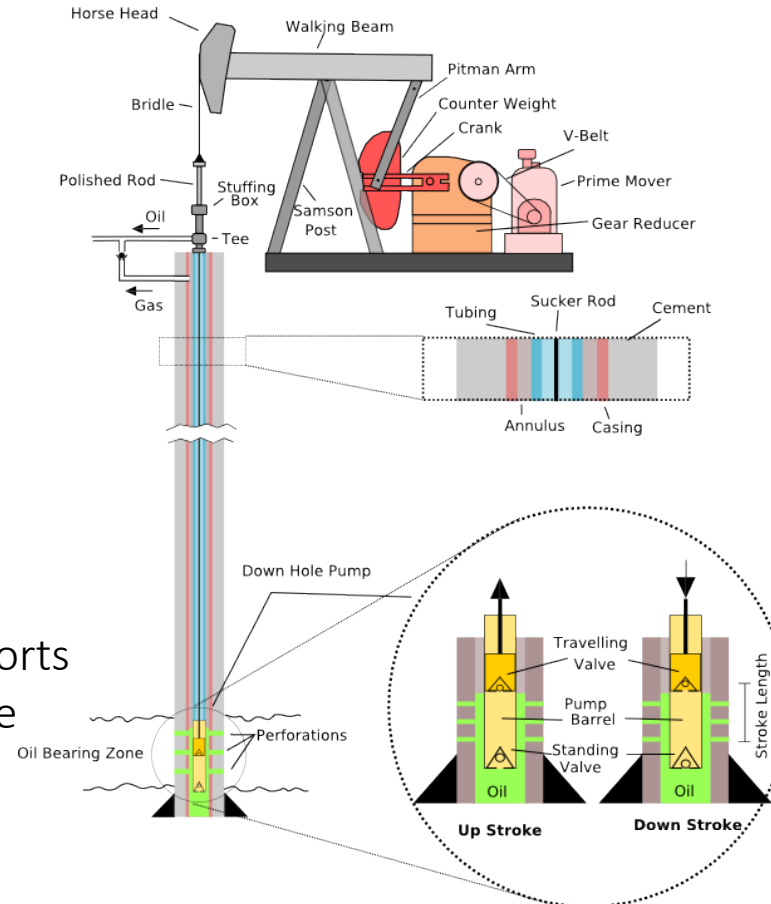
Using the $d+t$ nuclear fusion reaction produces a high flux of neutrons with 14 MeV energy. The neutrons lose energy by inelastic scattering and produce characteristic gamma radiation for the elements in soil, rock or oil. Decay energy of excited ^{12}C nuclei give 4.4 MeV indicating carbohydrates (oil) and decay energy of ^{16}O at 6.1 MeV and 6.9 MeV points to water content. The gamma radiation is detected with scintillator or semiconductor detectors.



Oil Drilling and Completion



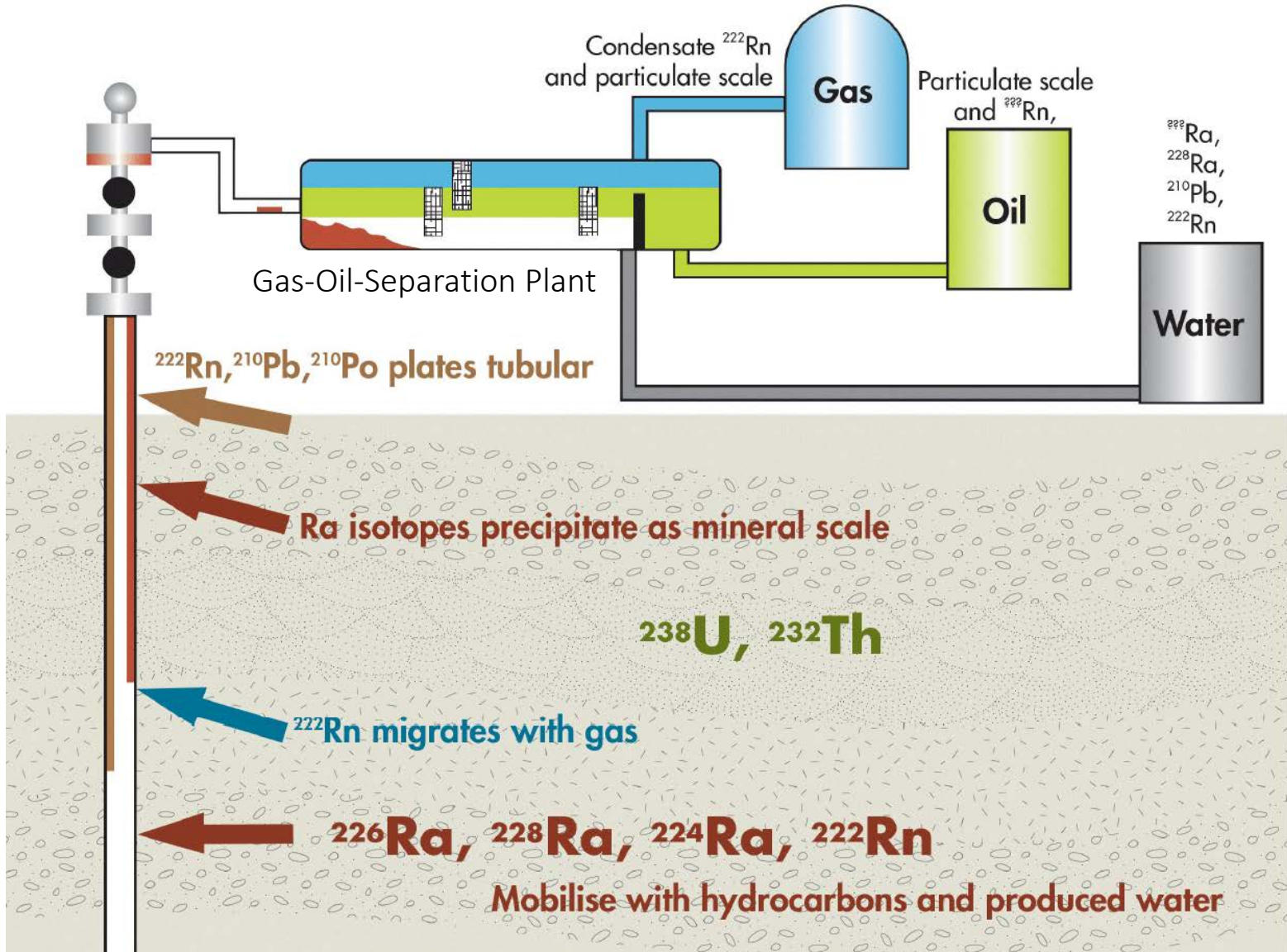
Water cooled drill bit transports slush through water pressure towards surface



Production enhancement through steam or hot water injection.

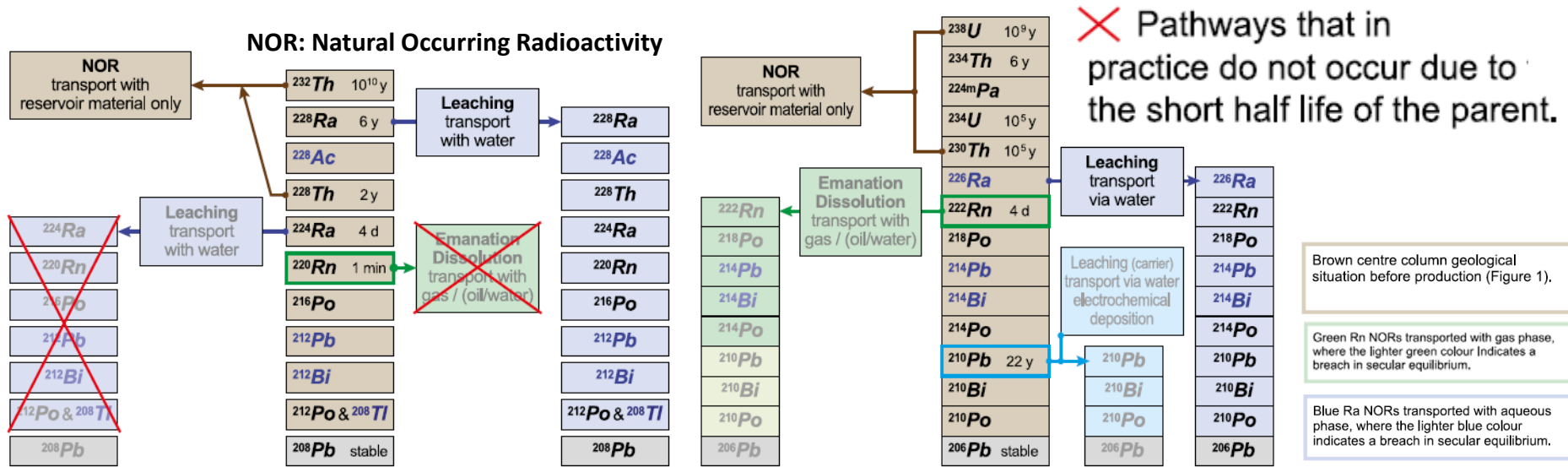
Depressurizing the tube, allowing the oil to enter through tube perforations, being pumped upwards. Besides oil, sand, rocks and stones, the so-called **drill cuttings** are being transferred to surface.

Up-take of radioactive isotopes



Separation in Filtering Process

Th & U: as a gas/oil reservoir is a reducing environment both Th and U prefer the solid rock phase and do not dissolve in the aqueous or oily phase. Consequently the top of both series remains with reservoir rock and can only appear in natural concentrations at the surface during drilling operations.



Ra: the partitioning of Ra (^{228}Ra , ^{226}Ra , ^{224}Ra) is special as they are formed by α -decay from their immediate parent and may be injected from rock/fluid interface into well fluids by so-called α -recoil. In addition, Ra prefers the aqueous phase, leading to somewhat naturally enhanced concentrations.

Drill Cuttings and Sludge

Radium-226 concentrations were detected in produced water samples ranging from 40.5 – 26,600 pCi/L. Radium-228 concentrations were also detected ranging from 26.0 – 1,900 pCi/L.

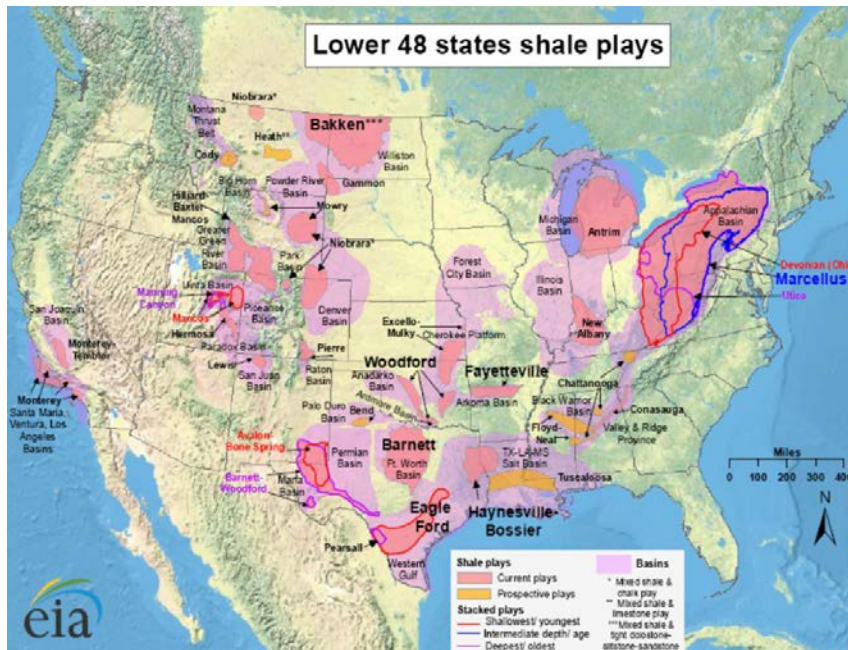


Sample of drill cuttings under a 10x microscope

Detailed Numbers

Sedimentary Rock Class	potassium (K)			thorium (Th)			uranium (U)		
	%		Bq(⁴⁰ K)/g mean	(ppm)		Bq(²³² Th)/g mean	(ppm)		Bq(²³⁸ U)/g mean
	mean	range		mean	range		mean	range	
DETRITAL (mineral rich sediments)	2.1	0.01-9.7	7	12.4	0.2-362	0.05	4.8	0.1-80	0.06
Sandstone & Conglomerate Orthoquartzites Arkoses	1.2	0.1-8.5	0.4	9.7	0.7-227	0.04	4.1	0.1-62	0.05
	2		0.06	1.5		0.0006	0.5	0.5-3	0.005
				5		0.02	1.5		0.02
Shale grey/green back	3.5	0.9-8.5	1.1	16.3	5.3-39	0.07	5.9	0.9-80	0.07
	3		0.9	13		0.05	3	3-4	0.04
								8-20	
Clay	0.6	0.1-2.6	0.2	8.6	1.9-55	0.03	4	1.1-16	0.05
CHEMICAL	0.6	0.02-8.4	0.2	14.9	0.03-132	0.06	3.6	0.03-27	0.04
Carbonates Limestone	0.3	0.01-3.5	0.09	1.8	0-11	0.007	2	0.03-18	0.02
			0.2	3		0.01	13		0.16
Evaporites							<0.1		<0.001

Radioactivity contamination in oil drill and fracking sludge



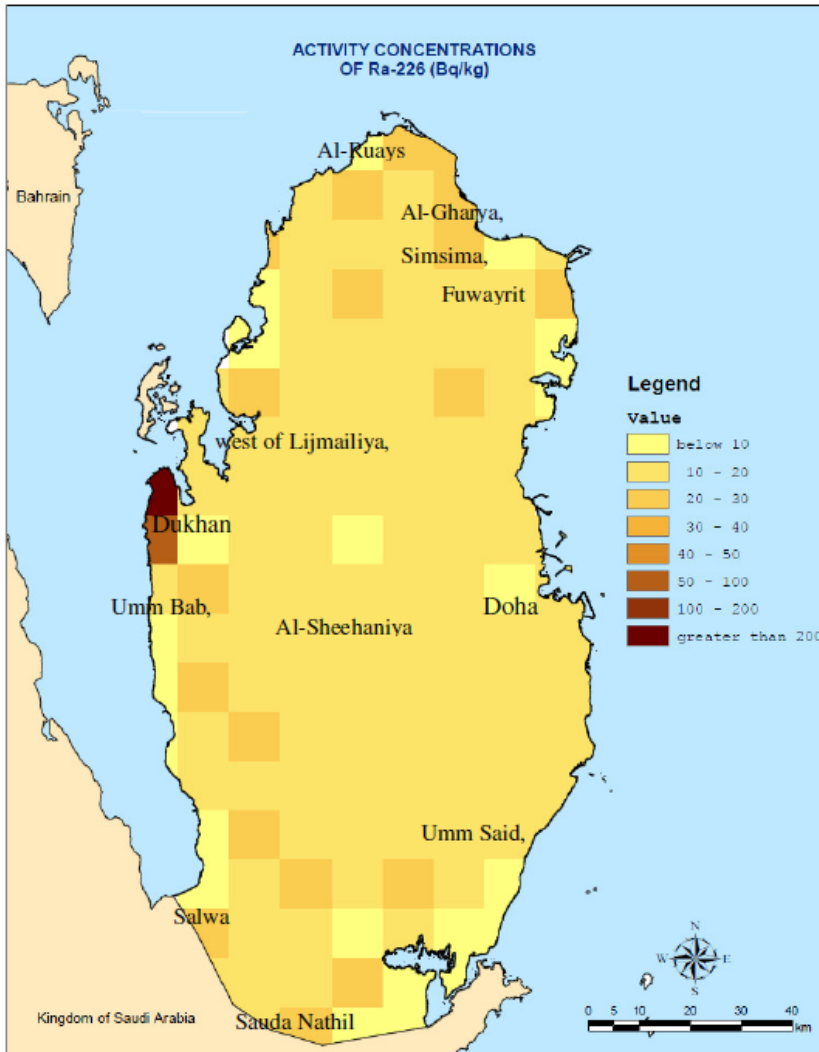
- ❖ Treated frac H2O sludge: 6 - 250 $\mu\text{R/h}$ = 0.06-2.5 $\mu\text{Gy/h}$
- ❖ Treated frac H2O sludge: BG - 3,000 pCi/g = 10,000 Bq/kg
- ❖ Gas Well Environs, 45 PA Sites, CY08-09
 - Background: 9.0 $\mu\text{R/h}$
 - Well Pads: 8.7 $\mu\text{R/h}$
 - Well Pits: 9.3 $\mu\text{R/h}$ [some 15-20 $\mu\text{R/h}$]
- ❖ Radon in Natural Gas: 37 pCi/L [1-79 pCi/L] = 1.4 Bq/l

	NOR	Bq[NOR]/g[scale]	Bq[NOR]/g[deposit]
^{232}Th series	^{232}Th	0.001 < > 0.002	0.001 - 0.07
	$^{228}\text{Ra}_{\text{eq}}$	0.05 < > 2800	0.05 - 300
^{238}U series	$^{238}\text{U}_{\text{eq}}$	0.001 < > 0.5	0.001 - 0.05
	$^{226}\text{Ra}_{\text{eq}}$	0.1 < > 15,000	0.8 - 400
	$^{210}\text{Pb}_{\text{eq}}$	0.02 < > 75	0.05 < > 2000

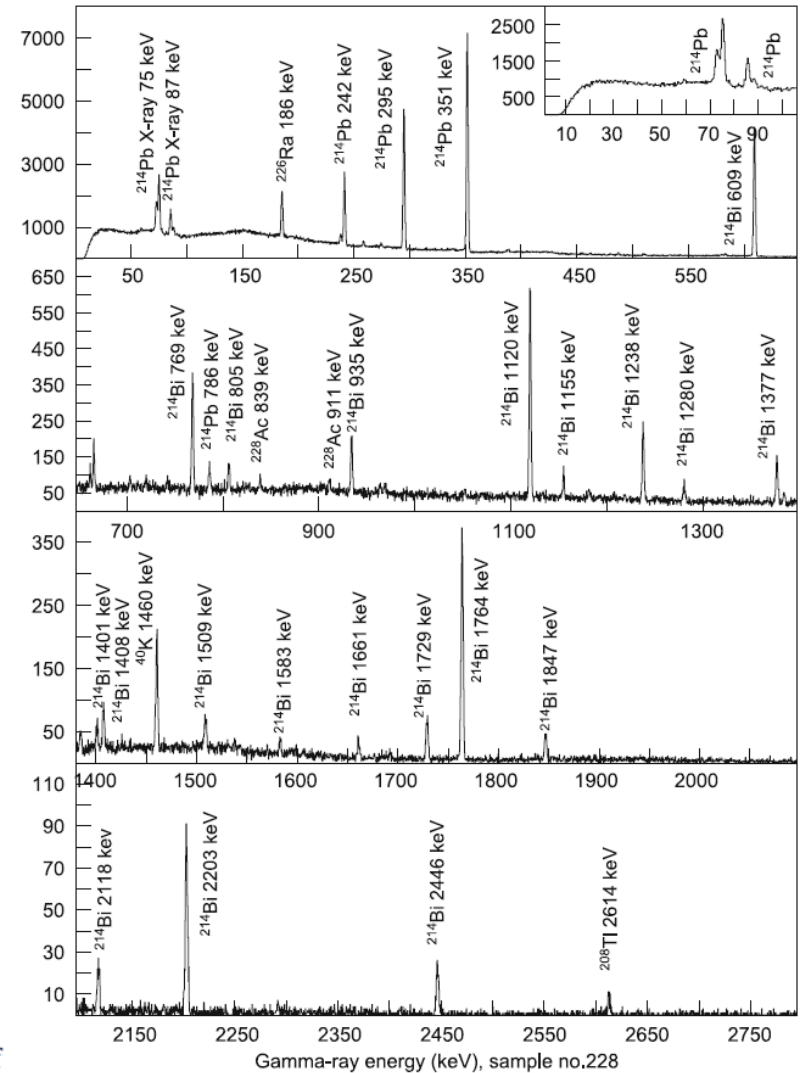
Area	Average	Range
Chicago	14.4	2.3-31.3
New York City	1.5	0.5-3.8
Denver	50.5	1.2-119
West Coast	15	1-100
Colorado	25	6.5-43
Nevada	8	5.8-10.4
New Mexico	45	10-53
Houston	8	1.4-14.3
Overall	23	= 0.9 Bq/l

Oil drilling waste in Qatar

H. Al-Sulaiti et al. / Nuclear Instruments and Methods in Physics Research A 619 (2010) 427–431



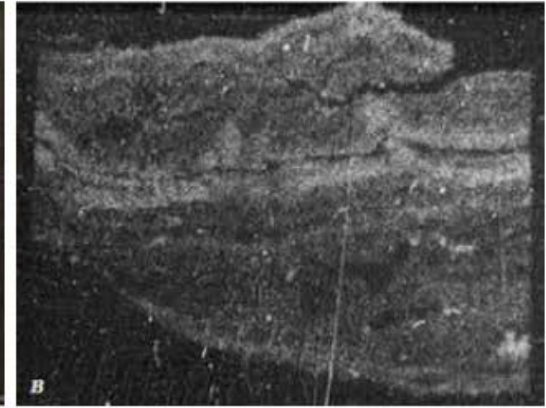
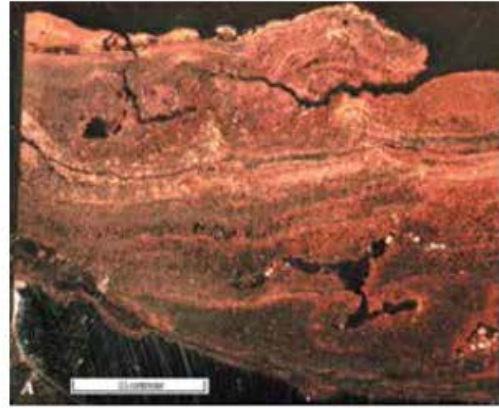
A radiological map of Qatar showing the distribution of the weighted means of activity concentrations of ^{226}Ra .



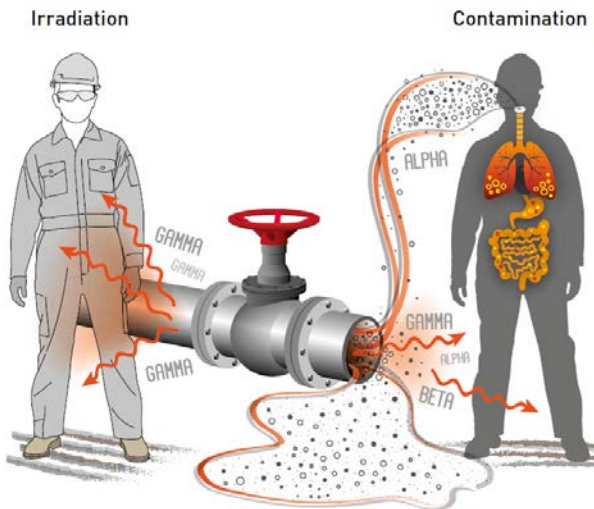
Radioactive Depositions



Scale build up within a pipe (8 $\mu\text{Sv/h}$ dose rate at pipe exterior). Picture courtesy of ExxonMobil



Radioactive scale deposits inside oil-field pipe (A) and the distribution of alpha-particle emitting radium and radium decay products in the same sample (B)



Exposure minimal, far below the permissible limits. High contaminated materials are being transported to storage site.



Comparison of fossil energy sources with typical activity levels

- Emission of Coal Power Plants
- Coal ashes
- Oil sludge
- Soil
- Human

Material	Activity in Bq/kg				
	Uranium	Thorium	Radium	^{40}K	Total
Fly ash	100	50	130	421	701
Coal ashes	90	40	110	403	643
Oil sludge	1 - 50	1-70	800		800-900
soil	25	40	48	400	513
Human	0.016	0.0016	0.016	63	63.03

Do we have to think about alternative energy sources?

