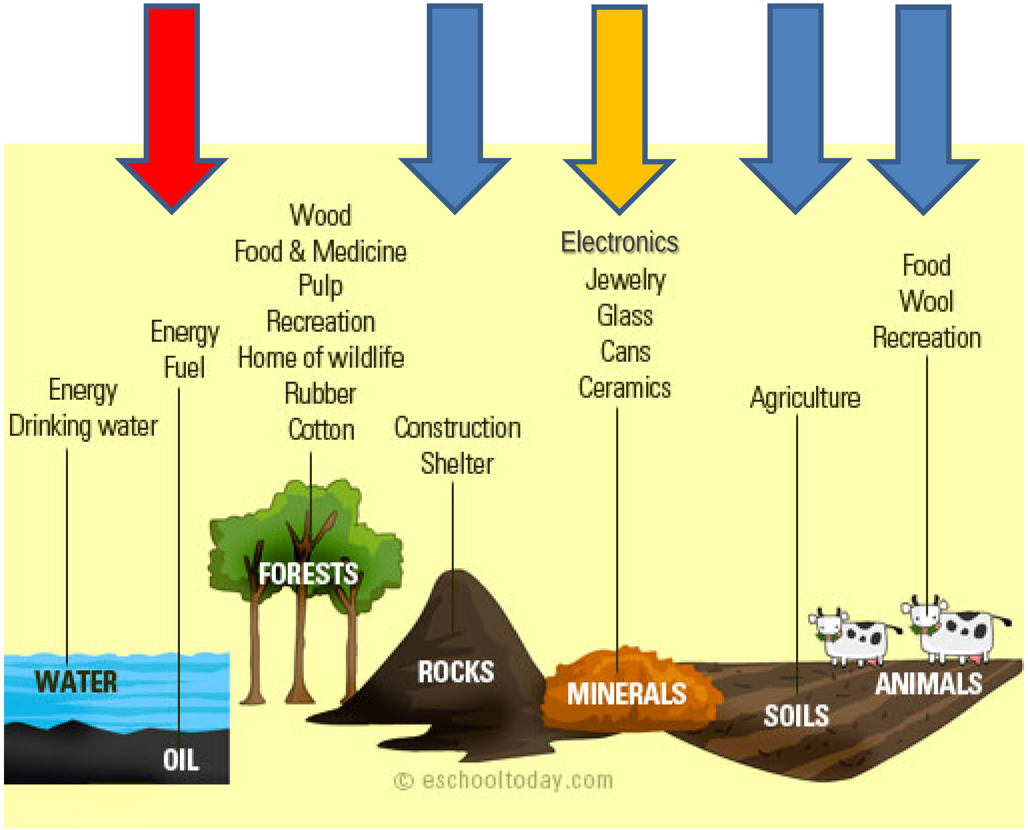
Radioactivity

Lecture 18

Radioactivity in Natural Resources

Natural Resources

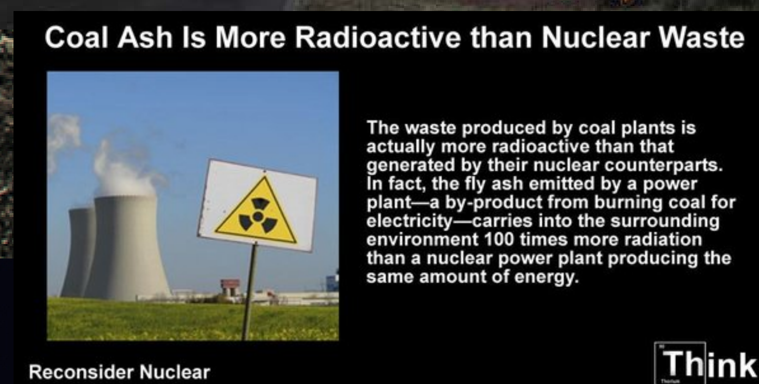
Renevables	Sun
	Wind
Changables	Land
	Forests
	Water
Non-renewables	Materials
	Minerals
	Energy Fuel



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

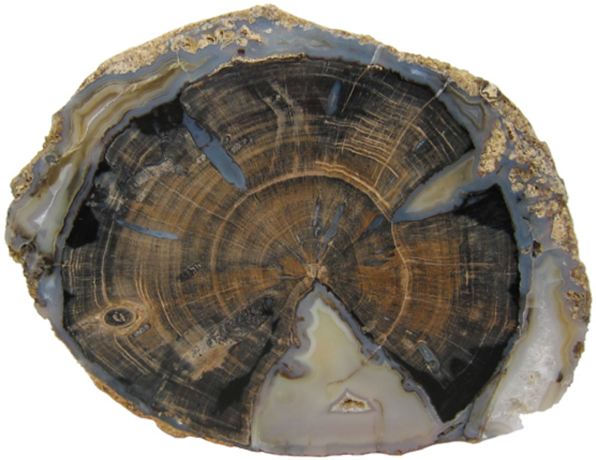
Public Fears

Concerns about CO_2 emission and its impact on climate is complemented by claims of high levels of radioactivity



Fossil fuels

- Fossil fuel is primarily fossilized organic matter (bio-matter). 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 $^{14}C/C$ ratio in the atmosphere declines.

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

Radioactivity in Coal

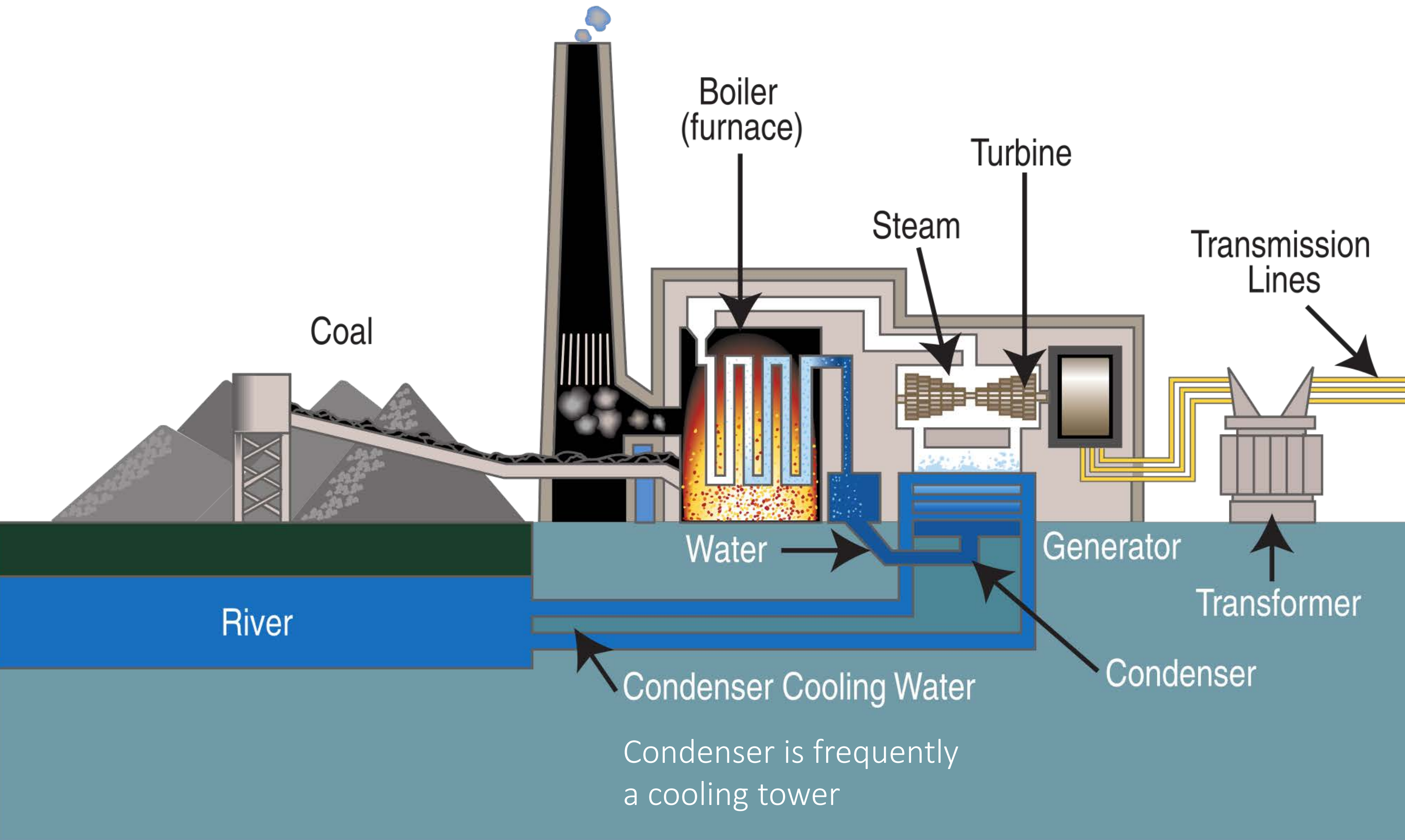
Coal has a certain amount of inherent radioactivity. The level changes due to the nature of the mineralization process and the mineral content of the environment.

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

<table>
<tr><th>Origin</th><th>U-238</th><th>Ra-226</th><th>Pb-210</th><th>Po-210</th><th>Th-232</th><th>Ra-228</th><th>K-40</th></tr>
<tr><td>Australia</td><td>8.5-47</td><td>19-24</td><td>20-33</td><td>16-28</td><td>11-69</td><td>11-64</td><td>23-140</td></tr>
<tr><td>Brasilia</td><td>72</td><td>72</td><td>72</td><td></td><td>62</td><td>62</td><td></td></tr>
<tr><td>China</td><td colspan="4">10-25, with local enrichments up to 5600</td><td colspan="2">10-25, with local enrichments up to 29,000</td><td></td></tr>
<tr><td>Germany</td><td></td><td>10-145, (32)</td><td></td><td></td><td>10-63 (21)</td><td></td><td>10-700, (225)</td></tr>
<tr><td>(brown coal)</td><td></td><td>0-58</td><td></td><td></td><td>0-58</td><td></td><td>4-220</td></tr>
<tr><td>Greece</td><td>117-390</td><td>44-206</td><td>59-205</td><td></td><td></td><td>9-41</td><td></td></tr>
<tr><td>Hungary</td><td>20-480</td><td></td><td></td><td></td><td></td><td>12-97</td><td>30-384</td></tr>
<tr><td>Poland</td><td>10-159 (18)</td><td></td><td></td><td></td><td></td><td>10-123 (11)</td><td>10-785</td></tr>
<tr><td>Romania</td><td>1- 415 (80)</td><td>10-557 (126)</td><td>10-510 (210)</td><td>10-580, (262)</td><td>10-170 (62)</td><td></td><td></td></tr>
<tr><td>Great Britain</td><td>7-19</td><td>8-22</td><td></td><td></td><td>7-19</td><td></td><td>55-314</td></tr>
<tr><td>USA</td><td>6-73</td><td>8.9-59</td><td>12-78</td><td>3-52</td><td>4-21</td><td></td><td></td></tr>
</table>

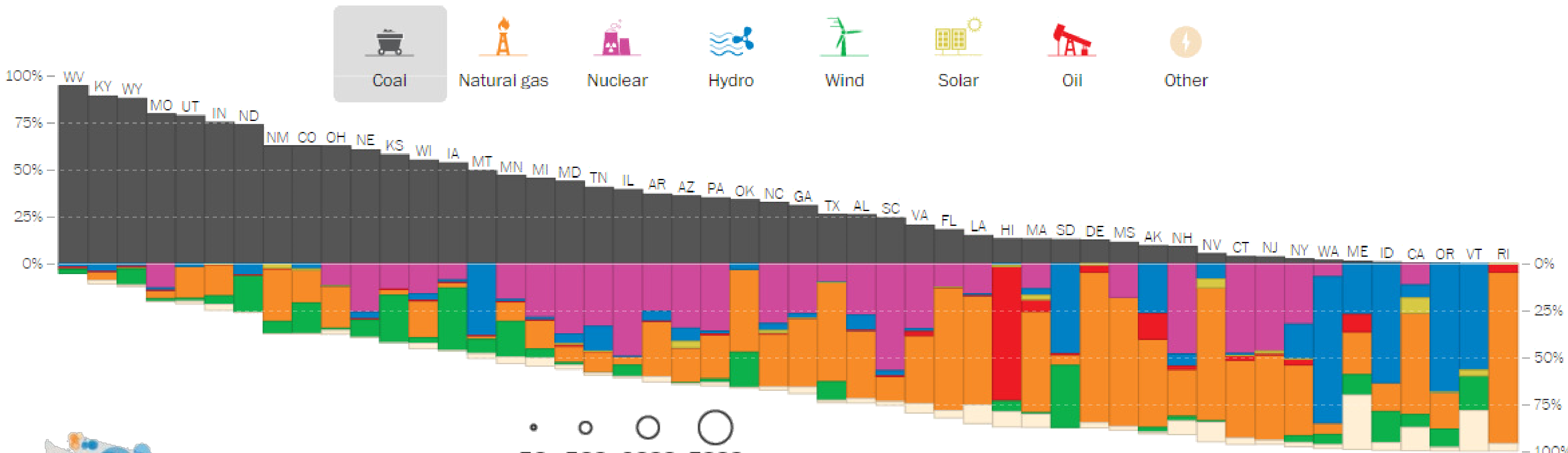
Average total activity of US coal is about 57 Bq/kg which converts to 5.3 kBq/m^3.

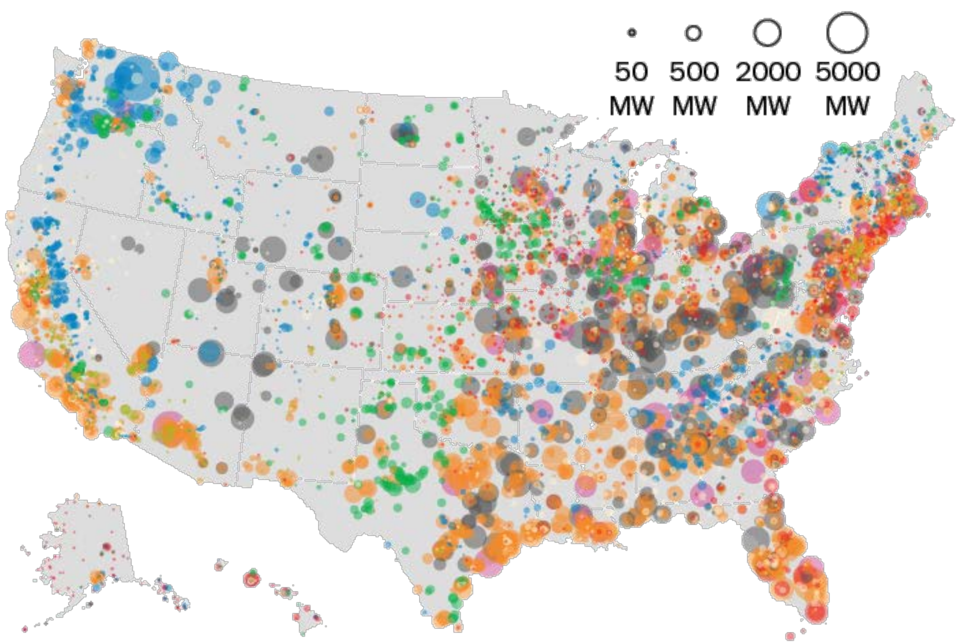
Operation of a Coal Power Plant

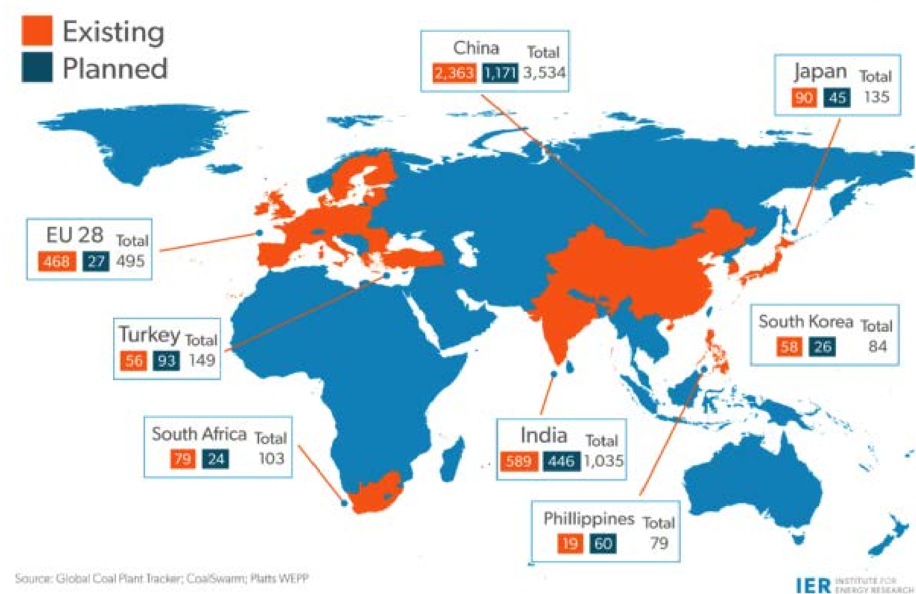


Condenser is frequently
a cooling tower

Coal Power Distribution world-wide and in US







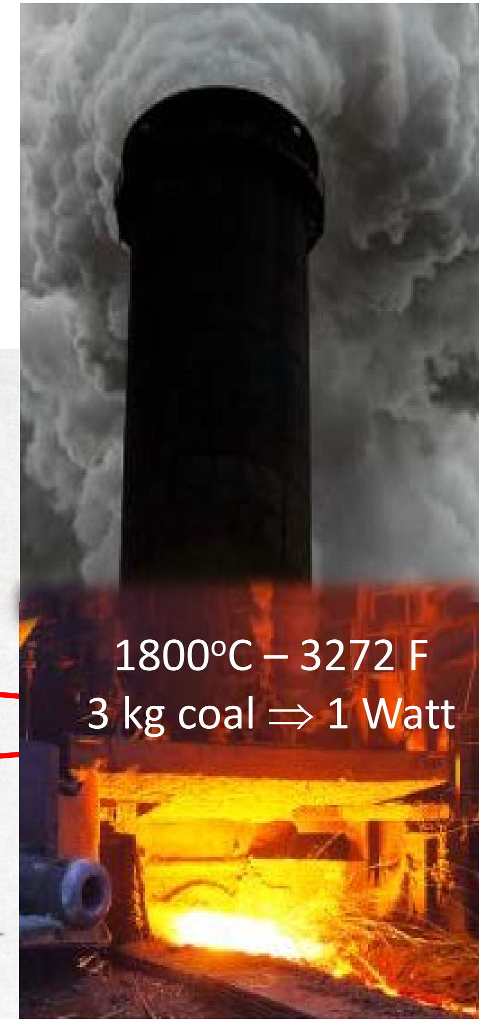
Annual radioactivity emission by coal power plants

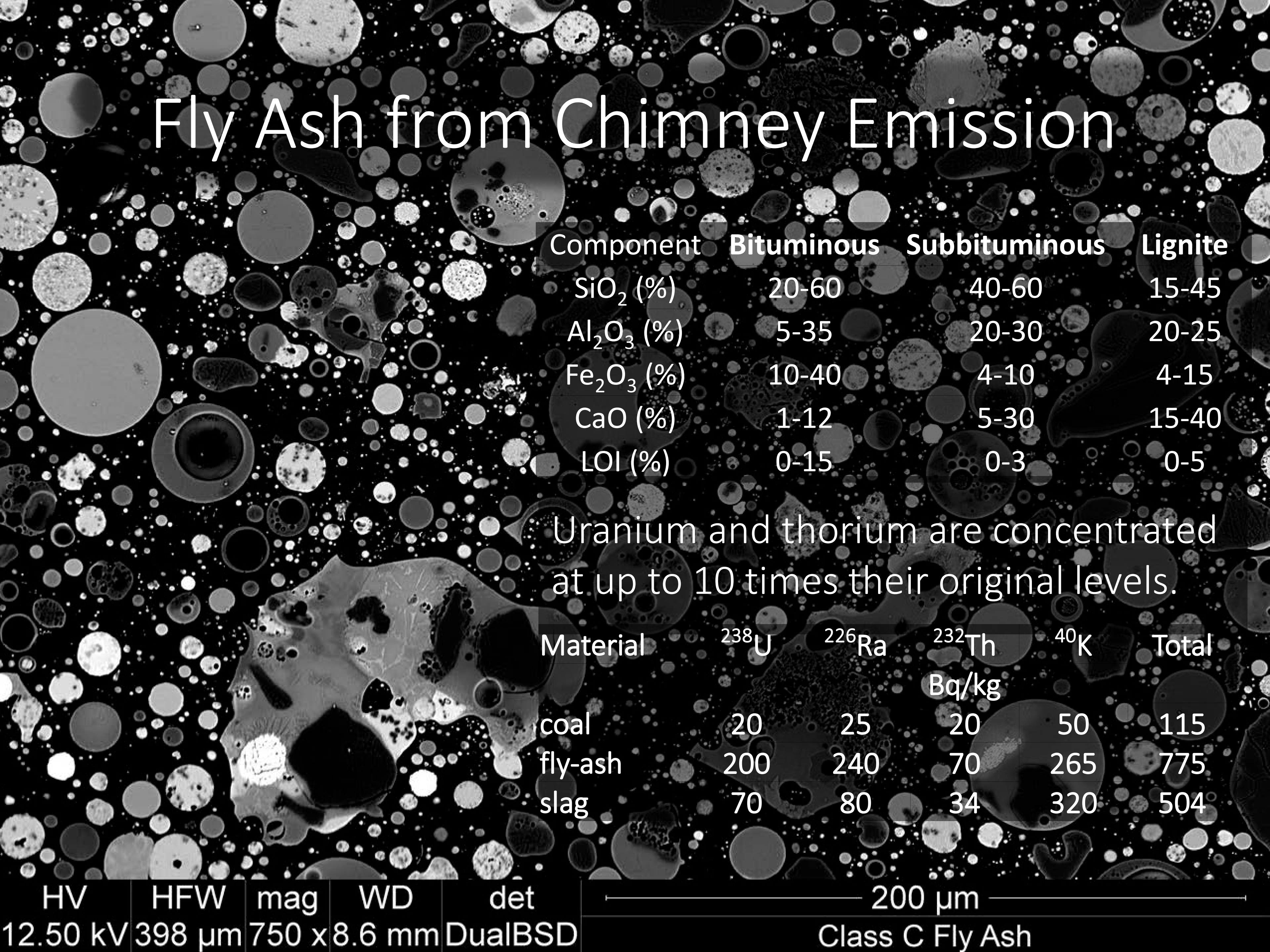
The table provides different estimates of the output on radio-nuclides from coal power plants. The numbers are normalized to a power production of 1000 W. These values can be reduced by implementing efficient filters and absorbers between furnace and chimney exhaust.

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

Reference	^{238}U	^{226}Ra	^{210}Pb	^{210}Po	^{222}Rn	^{232}Th	^{228}Ra	^{220}Rn	^{235}U	^{219}Rn
	Release rates of radionuclides (Bq y^{-1})									
Eisenbud & Petrow (1964)		6.4×10^{8}					4.0×10^{8}			
McBride *et al.* (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^{9}	7.8×10^{9}	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}	2.8×10^{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^{9}–1.8×10^{10}	1.8×10^{10}	2.6×10^{10}			2.6×10^{10}				
Corbett (1983)—modern CFP[a]	8.0×10[illegible]	8.0×10[illegible]	8.0×10[illegible]	8.0×10[illegible]		4.0×10^{8}	4.0×10^{8}			
Mishra *et al.* (1984)		4.5×10^{9}–2.6×10^{10}								
De Santis & Longo (1984)	$<2.2\times10^{8}$	$<2.2\times10^{8}$				$<2.2\times10^{8}$	$<2.2\times10^{8}$			
Nakaoka *et al.* (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}			

[a]Modern CFP, i.e. equipped with sophisticated devices, from which only abcut 1% of the ash is released into the atmosphere.
[b]CFP which releases about 10% of the ash into the atmosphere.





Fly Ash from Chimney Emission

Component	**Bituminous**	**Subbituminous**	**Lignite**
SiO_2 (%)	20-60	40-60	15-45
Al_2O_3 (%)	5-35	20-30	20-25
Fe_2O_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

Releasing and Recycling

Modern filters remove 95-99% of fly ash

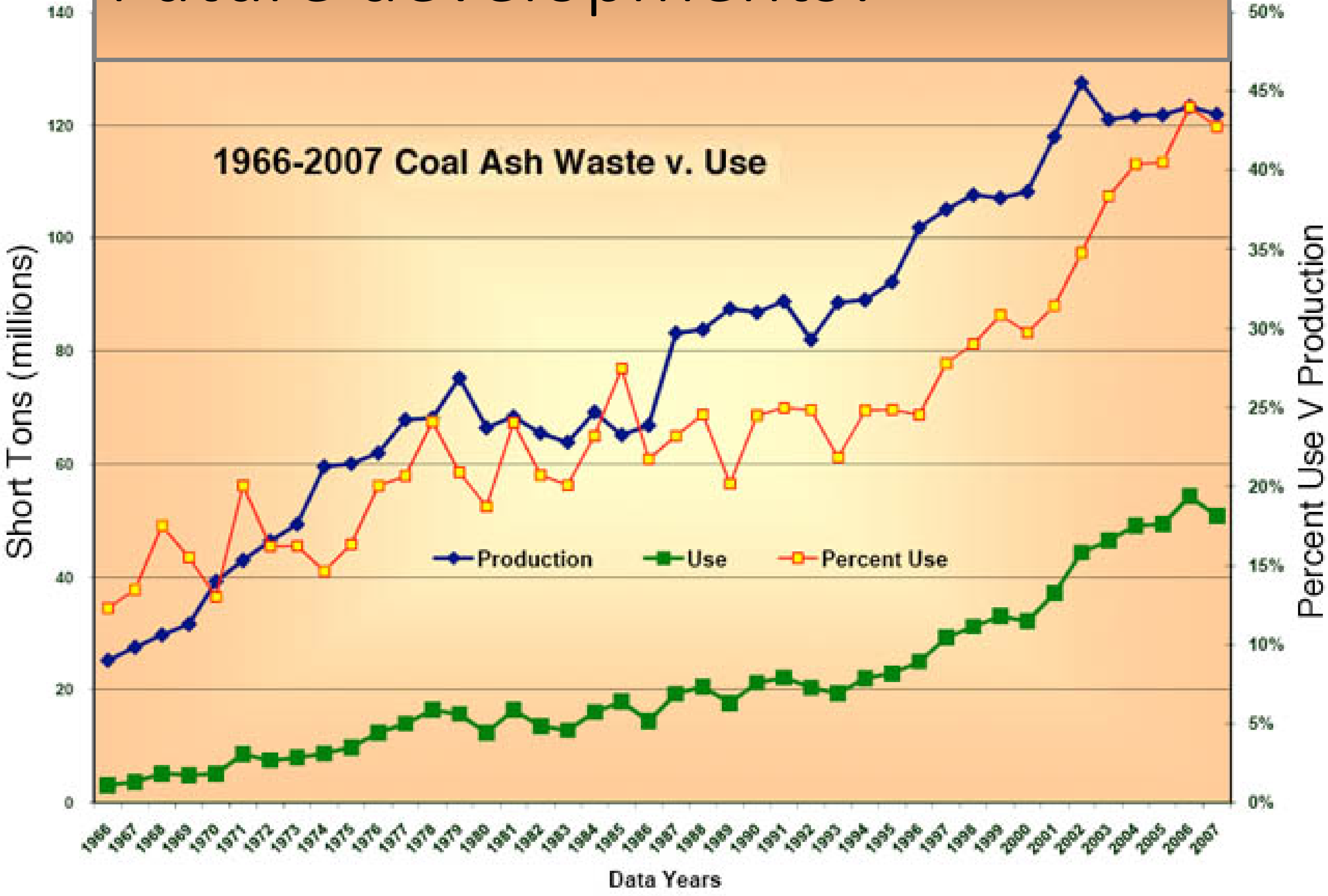
CFPs	Fly-ash removal efficiency %		^{238}U	^{226}Ra	^{232}Th	^{40}K
Modern (19)	95 - 99.8	Average	8.4	10.0	3.65	32.2
		Range	0.3-33.1	0.5-36.4	0.14-19.1	1.3-127.1

The annually released fly ash carries between 1-90 μSv with an average value of 12 μSv/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 (no of sample)		Activity concentration ($Bq\ kg^{-1}$) ^{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 μSv/y.

Future developments?



Coal Power Plant

US coal power plant in Kentucky

US coal power plant in Virginia

CO_2 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.

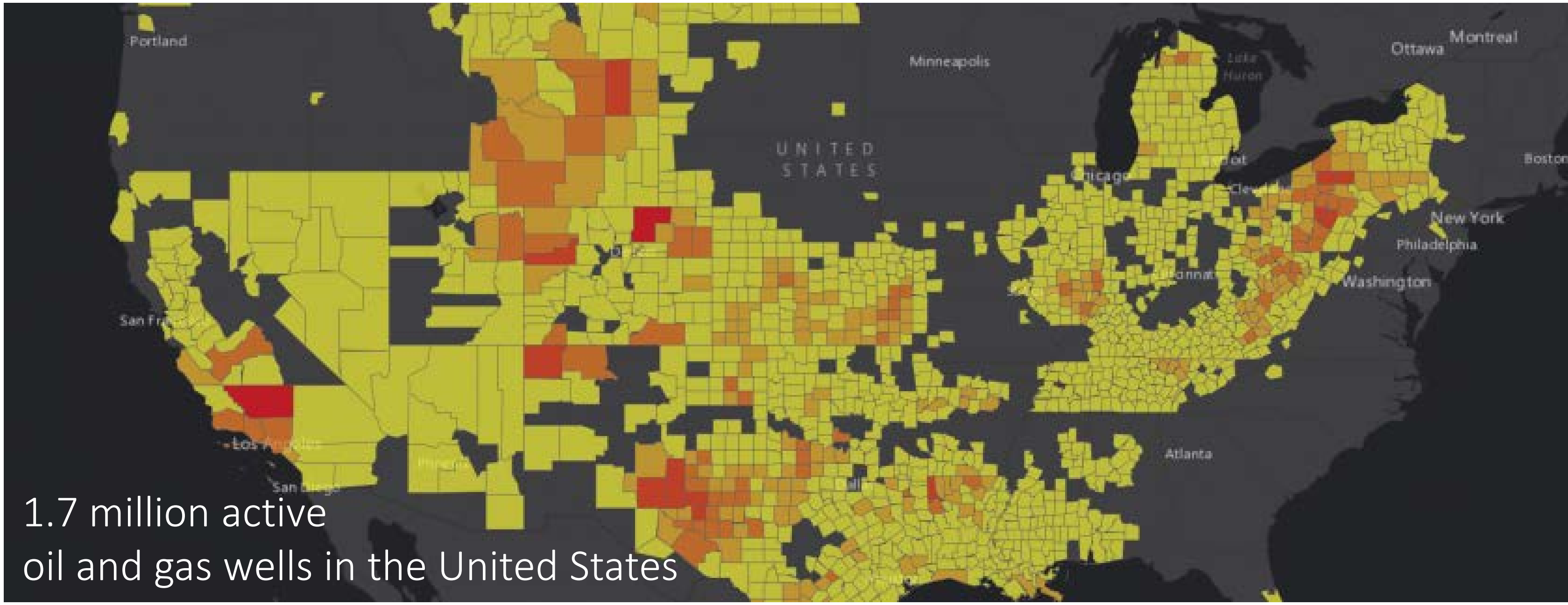
This in turn translates to an emission of $1.4 \cdot 10^{17}$ Bq or 140 PBq for the uranium decay chain and $8 \cdot 10^{16}$ Bq or 80 PBq for the thorium decay chain into the atmosphere eventually settling down as dust in the local neighborhood.

Oil and Gas

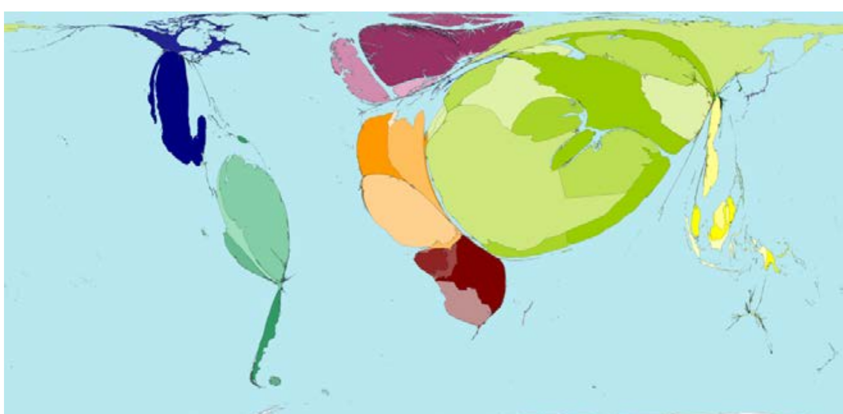
Oil is liquified biomaterial, compressed, complex organic hydrocarbon materials. The radioactivity is initial leftover from bio materials or migrated from surrounding rock material. The activity values for ^{238}U vary between 13 and 67 Bq/kg, similarly the values for ^{232}Th between 10 and 55 Bq/kg,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

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

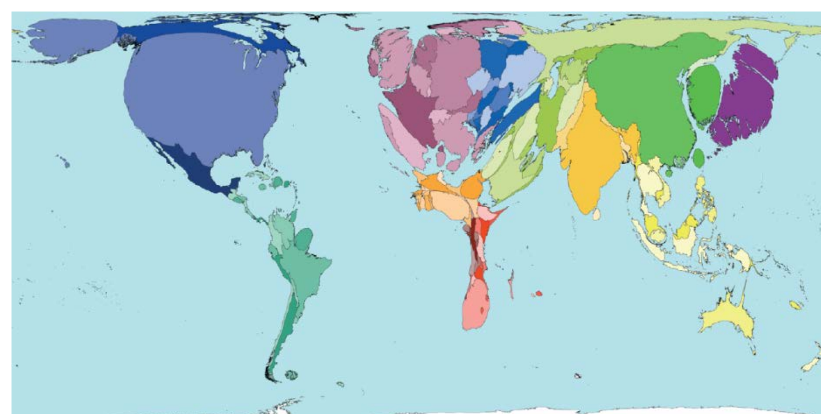
Active Oil and Gas fields in the United States



Oil producing countries

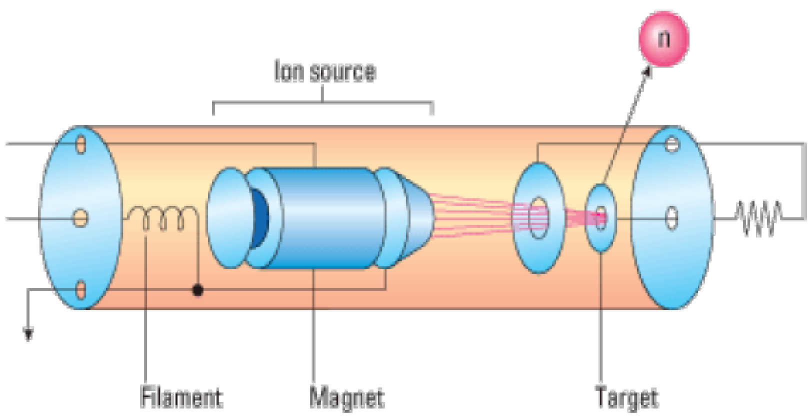


Oil using countries

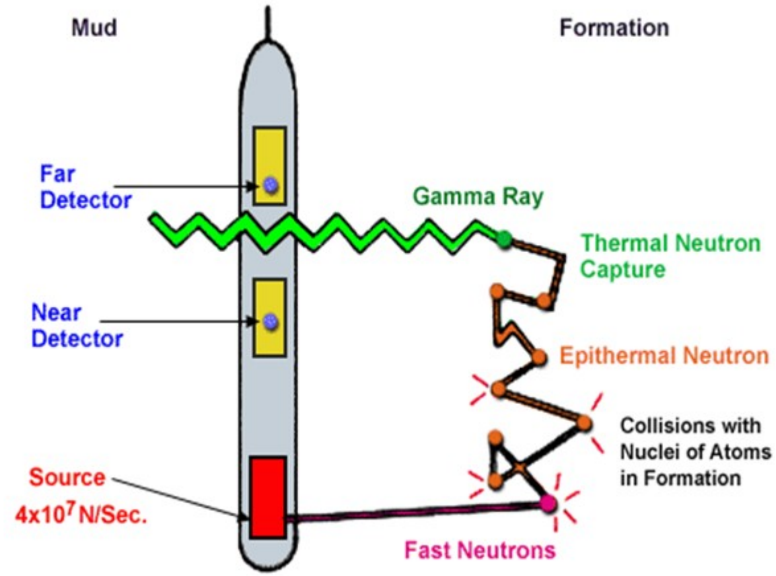


Size of the country scales with its role in producing oil and using oil based products

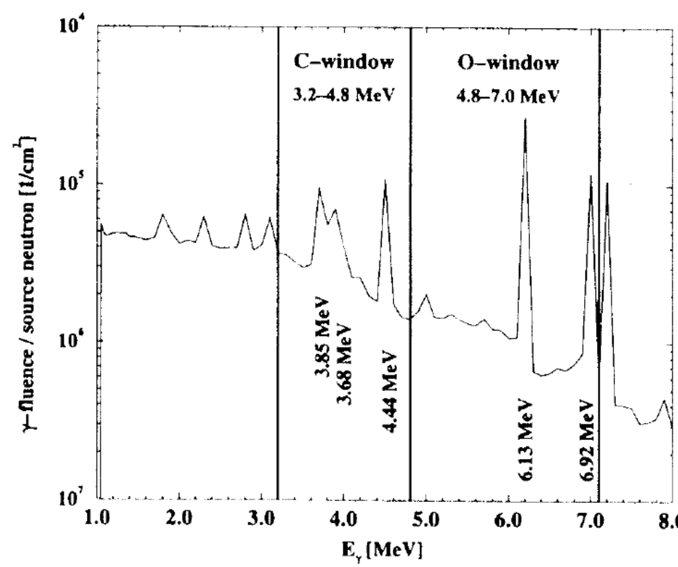
Oil Searching with Neutrons



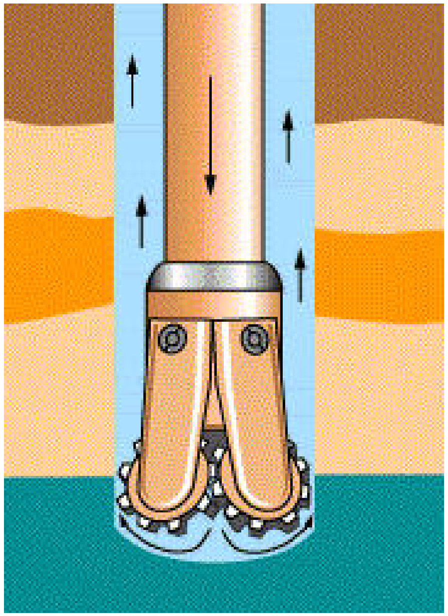
Neutron generator: A downhole accelerator used to emit pulses of high-energy neutrons into the formation.



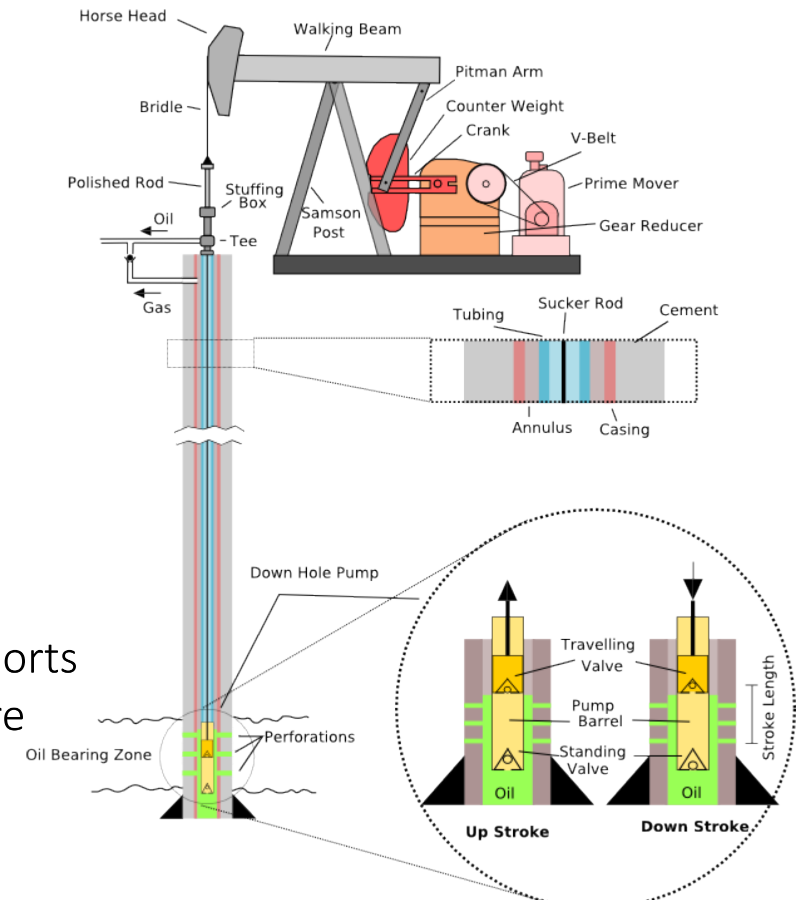
Using the d+t nuclear fusion reaction produces a high flux of neutrons with 14 MeV energy. The neutrons loose energy by inelastic scattering and produce characteristic gamma radiation. Decay energy of excited ^{12}C nuclei give 4.4 MeV indicating carbo-hydrates (oil) and decay energy of ^{16}O at 6.1 MeV and 6.9 MeV points to water content. The gamma radiation is detected with scintillator detectors.

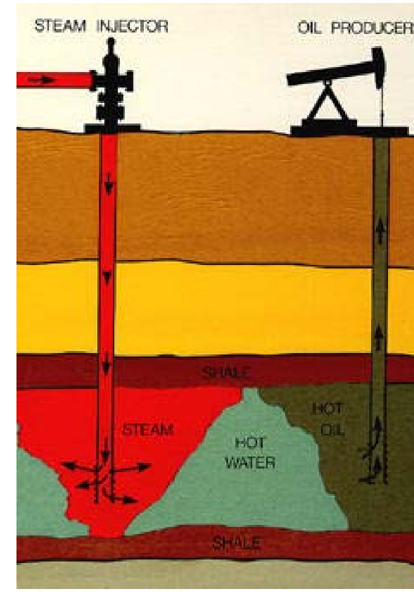


Oil Drilling and Completion



Water cooled drill bit transports slush through water pressure towards surface

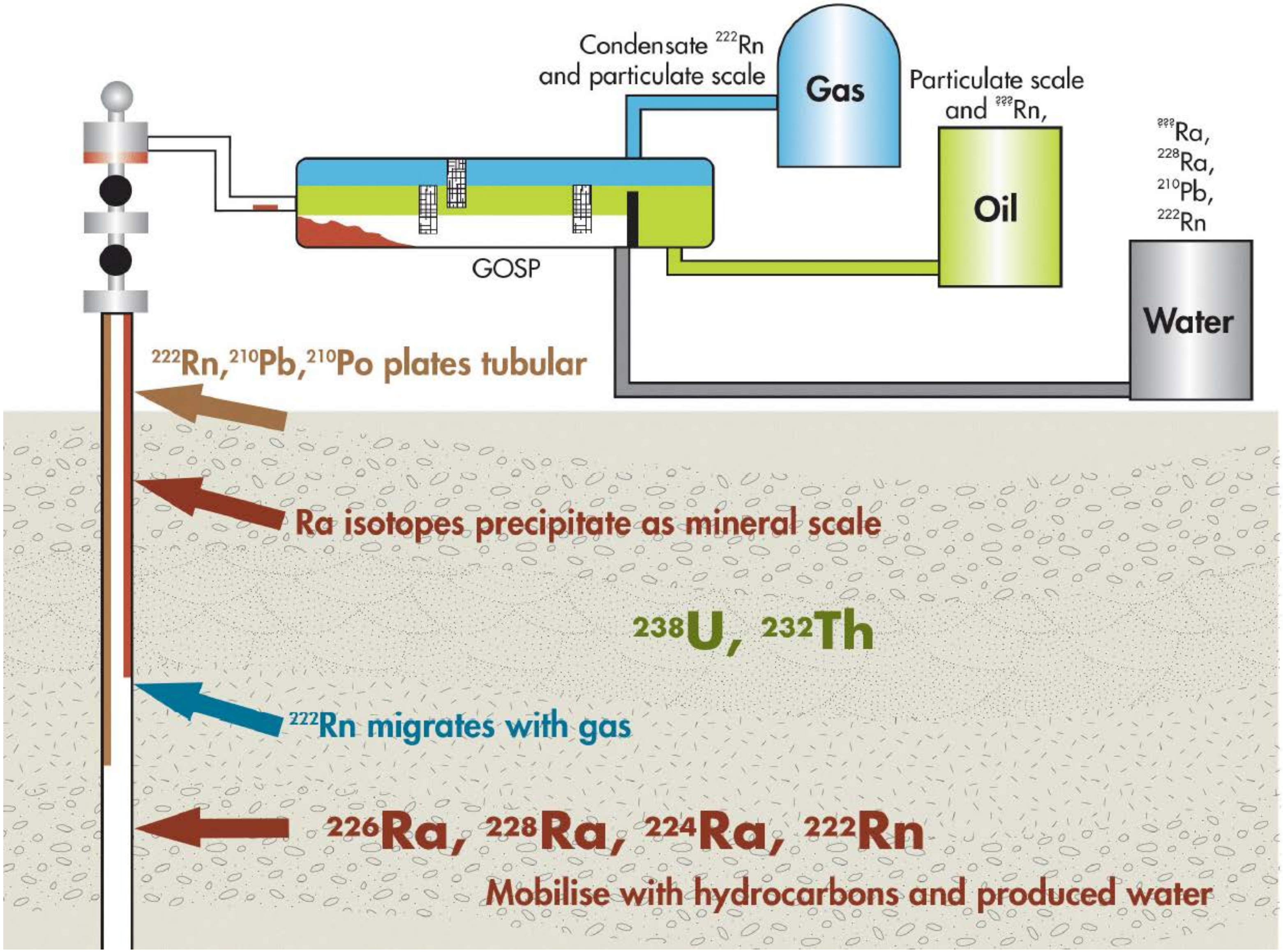




Production enhancement through steam or hot water injection.

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

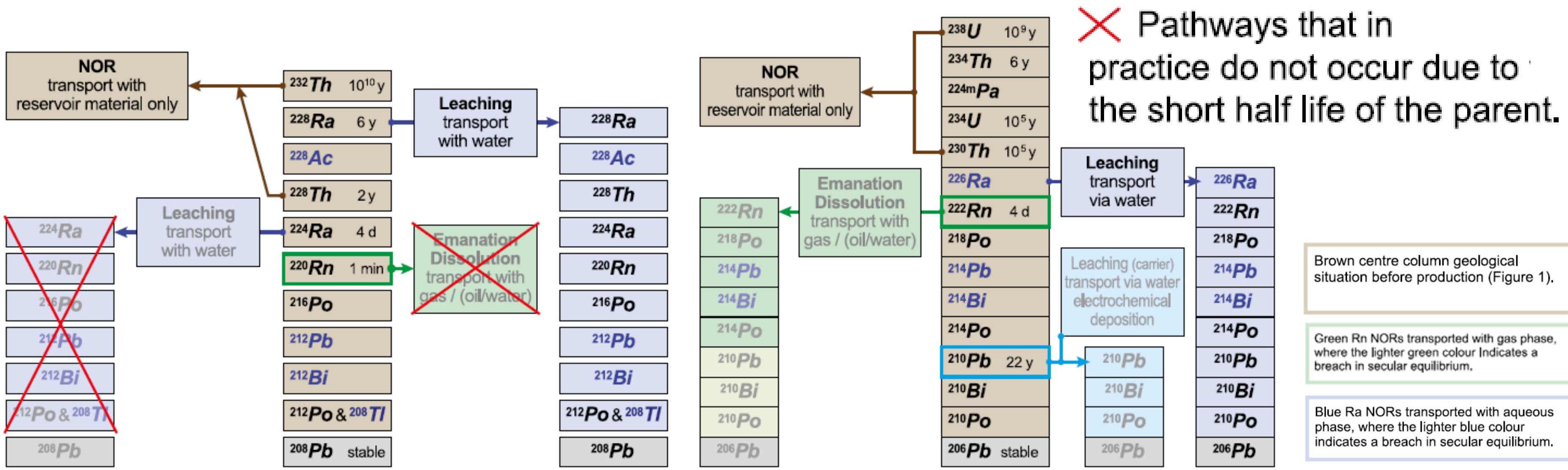


Detailed Numbers

Sedimentary Rock Class	potassium (K)			thorium (Th)			uranium (U)		
	%			(ppm)			(ppm)		
	mean	range	Bq(^{40}K)/g mean	mean	range	Bq(^{232}Th)/g mean	mean	range	Bq(^{238}U)/g mean
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

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.



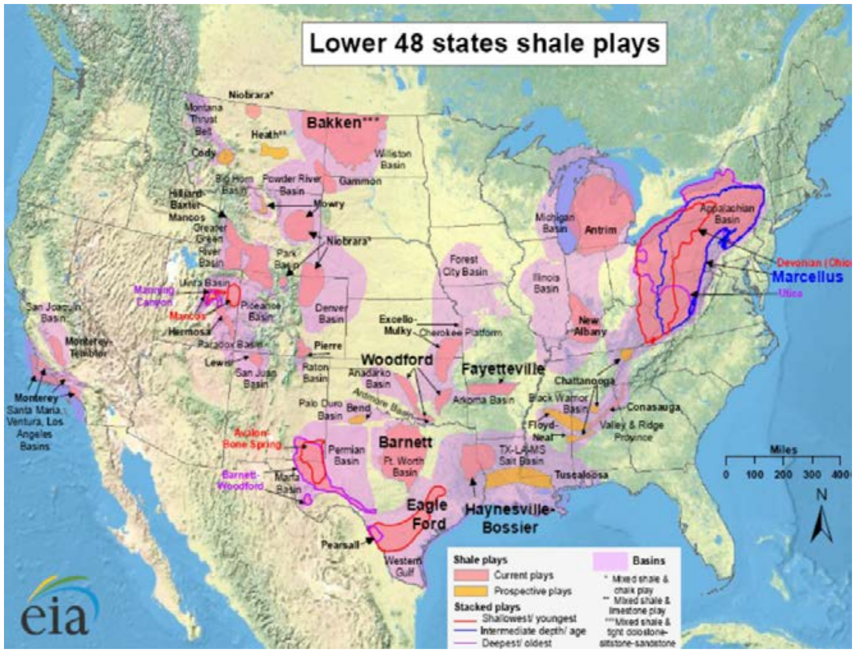
Ra: the partitioning of Ra (^{228}Ra, ^{226}Ra, ^{224}Ra) is special as they are formed by α-decay from their immediate parent and may be injected from rock/fluid interface into well fluids by so-called α-recoil. In addition, Ra prefers the aqueous phase, leading to somewhat naturally enhanced concentrations.

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

Radioactivity contamination in oil drill and fracking sludge

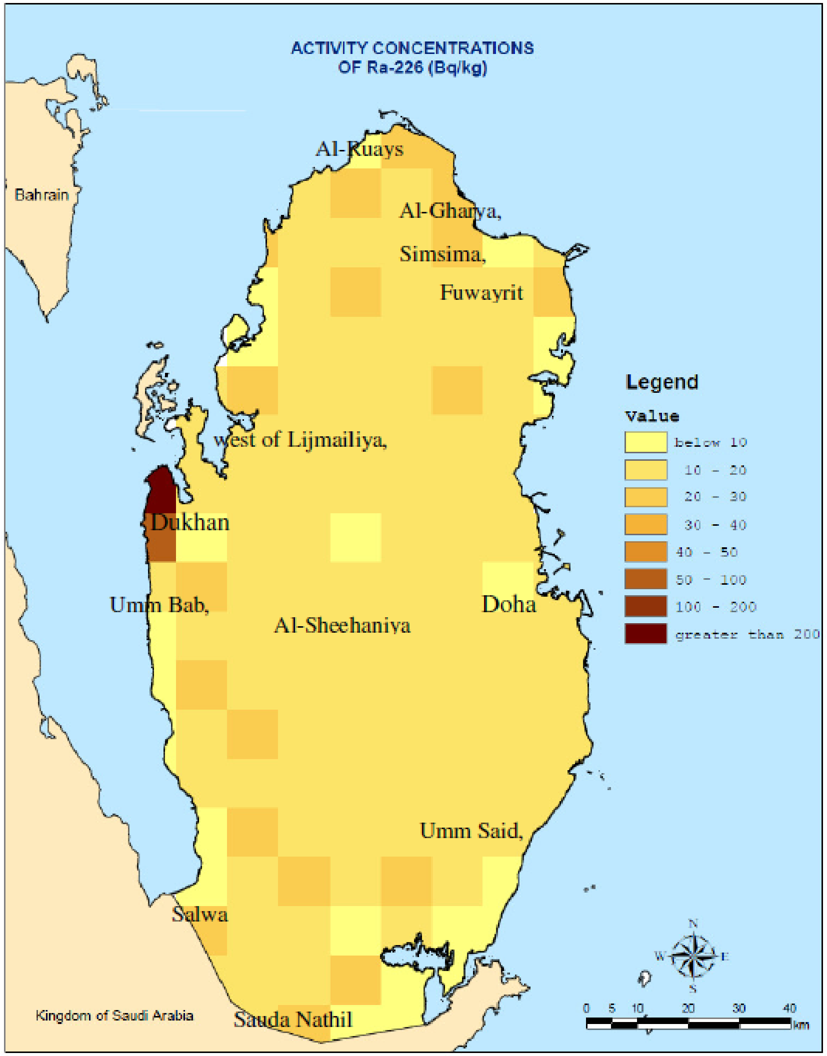


- **Treated frac H20 sludge: 6 - 250 μR/h =0.06-2.5μGy/h**
- **Treated frac H20 sludge: BG - 3,000 pCi/g=10,000 Bq/kg**
- **Gas Well Environs, 45 PA Sites, CY08-09**
  - Background: 9.0 μR/h
  - Well Pads: 8.7 μR/h
  - Well Pits: 9.3 μR/h [some 15-20 μR/h]
- **Radon in Natural Gas: 37 pCi/L [1-79 pCi/L] =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
	$^{228}Ra_{eq}$	0.05 < > 2800	0.05 - 300
^{238}U series	$^{238}U_{eq}$	0.001 < > 0.5	0.001 - 0.05
	$^{226}Ra_{eq}$	0.1 < > 15,000	0.8 - 400
	$^{210}Pb_{eq}$	0.02 < > 75	0.05 < >2000

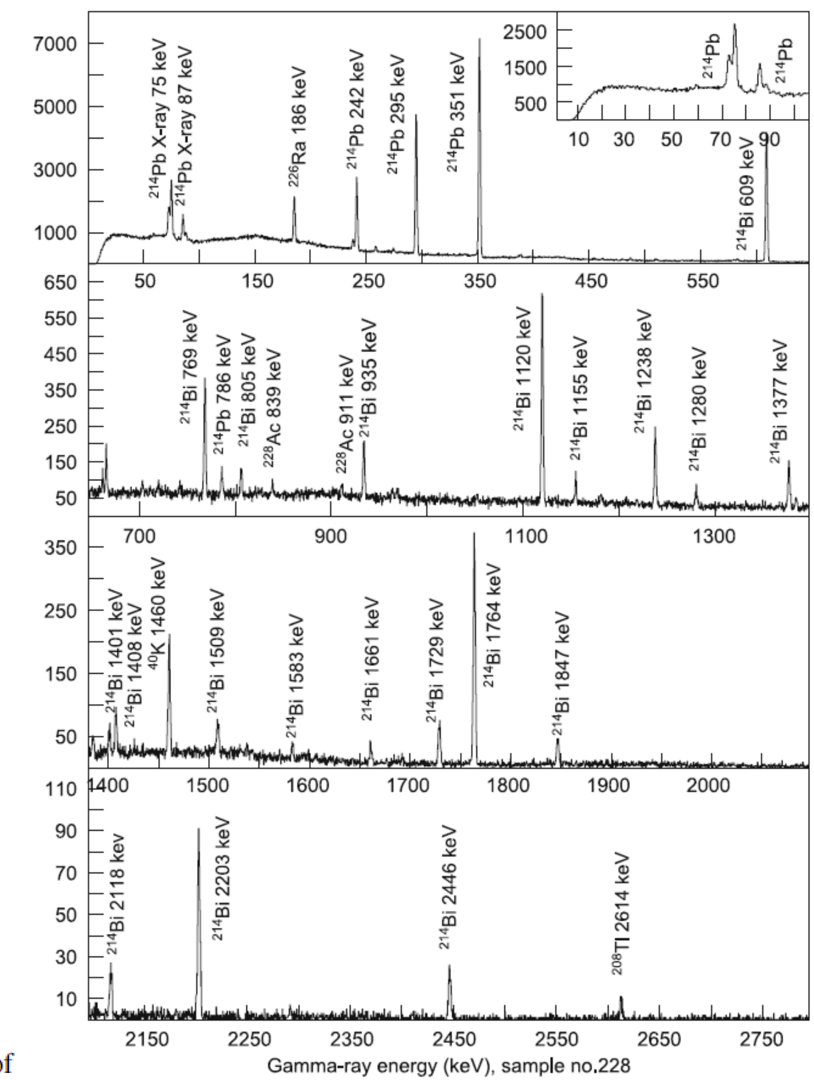
	Radon-222 Concentration (pCi/L)	
Area	Average	Range
Chicago	14.4	2.3-31.3
New York City	1.5	0.5-3.8
Denver	50.5	1.2-119
West Coast	15	1-100
Colorado	25	6.5-43
Nevada	8	5.8-10.4
New Mexico	45	10-53
Houston	8	1.4-14.3
Overall	23	

Oil drilling waste in Qatar



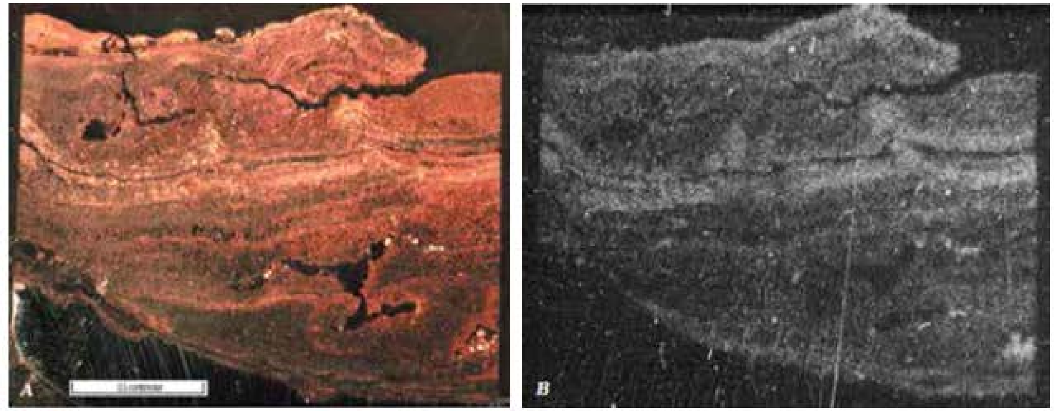
A radiological map of Qatar showing the distribution of the weighted means of activity concentrations of ^{226}Ra.

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

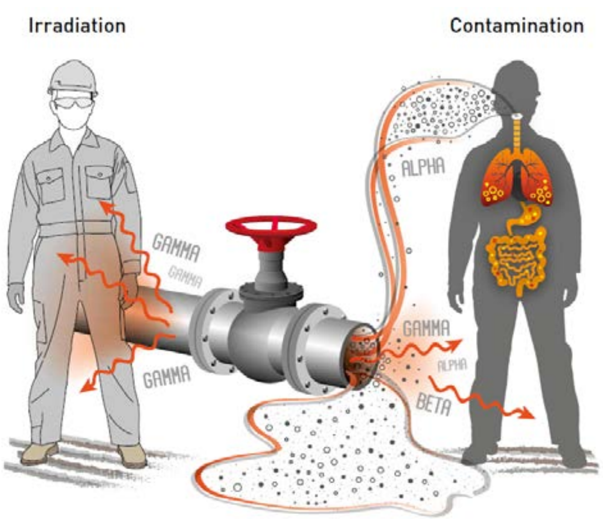


Radioactive Depositions

Scale build up
within a pipe (8 µSv/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

Activity in Bq/kg

Material	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?