Radioactive Waste
Regulators, Toxicity, and Health

“During the whole of the next day we proceeded on our journey through this interminable gallery, arch after arch, tunnel after tunnel.”

—Henry Lawson in “Journey to the Center of the Earth” by Jules Verne.
Radioactive Waste Regulators

Who’s in Charge?

U.S. Nuclear Regulatory Commission (http://www.nrc.gov/)

*Issues licenses
*Enforces standards
*Regulates possession, transportation, and disposal of commercial radioactive wastes

Chairman Kristine Svinicki
Radioactive Waste Regulators

U.S. Environmental Protection Agency
http://www.epa.gov/radiation/

*Provides radiation protection standards
*Risk assessments of radionuclides in soil, water, air, and food.
*NCR regulates waste disposal using U.S. EPA criteria.
Radioactive Waste Regulators

U.S. Department of Energy (DOE)

* Conducts research and development for waste disposal.
* Assists LLRW disposal programs.
* Manages defense-related (transuranic) wastes
* Slated to manage used nuclear fuel disposal sites when and if they open.
* Plans to manage surplus plutonium.

U.S. Department of Transportation
* Regulates the transport of wastes
Nuclear Regulatory Commission...
- Enforces standards
- Grants or denies licenses
- Inspects
- Regulates shipping casks
- Interacts with State, Tribal, and Local authorities

Department of Energy...
Designs, constructs, and operates a repository

Department of Transportation...
- Regulates transportation of waste
- Enforces requirements on shippers, routing, and vehicles
- Interacts with State, Tribal, and Local authorities

Environmental Protection Agency...
- Establishes standards
Regulation in Illinois

In Illinois, we have the Division of Nuclear Safety (DNS) which is part of the Illinois Emergency Management Agency (IEMA).

* Inspect radioactive material licensees.
* Inspect and register medical radiation equipment.
* Monitor the 11 reactors at the 6 power stations for electricity (such as LLRW).
* Escort used nuclear fuel shipments.

http://www.state.il.us/iema/dns.asp
Radioactive Wastes and Health

Acute radiation symptoms
Nausea, vomiting, diarrhea, bleeding, coma, and then death

Gamma Radiation
Erythema (skin redness)
Ulceration (skin sores than heal slowly if at all)
Tissue necrosis (dead skin)
Uranium is found everywhere in trace amounts.
You always take it into your body from the air, water, food, and soil.
Food and water have small amounts of natural uranium in them.
People eat about 1 to 2 micrograms (0.6 to 1.0 picocuries) of natural uranium every day with their food.
Uranium and Health Issues

People take in about 1.5 µg (0.8 pCi or 29.6 mBq) of natural uranium for every liter of water they drink.

Table 1-3
Summary of the Concentrations of Uranium in Water Samples

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Concentration</th>
<th>Type of water</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium (specific isotope not identified)</td>
<td>&lt;0.1 to 17 µg/L</td>
<td>117 wells in four aquifers in Illinois</td>
<td>Morrow (2001)</td>
</tr>
<tr>
<td></td>
<td>Range: 0.01 to 582 pCi/L Mean: 1.1 pCi/L</td>
<td>35,000 surface water samples in U.S.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Range: 0.01 to 635 pCi/L Mean: 3.2 pCi/L</td>
<td>55,000 groundwater samples</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mean: 1.73 pCi/L</td>
<td>28,000 domestic water supplies</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.4 to 60 pCi/L</td>
<td>Colorado River</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&lt;0.1 to 8.0 pCi/L</td>
<td>Illinois aquifers</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.1 to 40 pCi/L</td>
<td>New Jersey groundwater</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.3 to 44 pCi/L</td>
<td>California groundwater</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.05 to 4.6 pCi/L</td>
<td>U.S. groundwater</td>
<td></td>
</tr>
<tr>
<td>U-234</td>
<td>Mean: 3.3 ± 4.8 pCi/L</td>
<td>U.S. public groundwater supplies</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mean: 0.3 ± 0.0 pCi/L</td>
<td>Ohio public groundwater supplies</td>
<td></td>
</tr>
<tr>
<td>U-238</td>
<td>Mean: 1.7 ± 3.0 pCi/L</td>
<td>U.S. public groundwater supplies</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mean: 0.1 ± 0.0 pCi/L</td>
<td>Ohio public groundwater supplies</td>
<td></td>
</tr>
</tbody>
</table>

ATSDR (1999)
Uranium and Health Issues

We also consume the daughter-decay products:

Uranium and daughter products in water

\[ ^{238}\text{U} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra} \rightarrow ^{222}\text{Rn} \ldots \]

\[ ^{232}\text{Th} \rightarrow ^{228}\text{Ra} \rightarrow ^{228}\text{Th} \rightarrow ^{224}\text{Ra} \rightarrow ^{220}\text{Rn} \ldots \]

Most occur in the pCi/L-level

Back to U—what happens next?
Absorption of U from the intestinal tract may range from < 0.2% to 3%.

The rest of the U leaves the body in feces.

About 90% of the absorbed U will be eliminated by the kidneys while the remainder will accumulate in the skeleton with a biological half-life of about 300 days.
Radioactive Wastes and Health

If you breathe uranium particles, some of it is exhaled and some stays in your lungs.

The smaller particles are inhaled down to the lower part of your lungs. If they do not dissolve easily, they stay there for years and cause most of the radiation dose to the lungs from uranium.

They may gradually dissolve and U is transported to your blood.
Uranium and Health Issues

When you eat foods and drink liquids containing uranium, most of it leaves within a few days in your feces and does not enters the blood.

A small portion will get into the blood and will leave your body through your urine within a few days.

The rest can stay in bones, kidneys, or other soft tissues. A small amount goes to bones and may stay there for years.
Uranium and Health Issues

Most people have a very small amounts of uranium, about 1/5,000th of the weight of an aspirin tablet, in their bodies, mainly in their bones.

From the Agency for Toxic Substances and Disease Registry (ATSDR), an agency of the U.S. Department of Health and Human Services.

http://www.atsdr.cdc.gov/
Uranium and Health Issues

Case studies

Acute U overdoses resulted in renal failure in animal studies, but mostly with water-soluble forms of U like uranyl nitrate hexahydrate, uranium hexafluoride, uranyl fluoride, uranium tetrachloride, and uranium pentachloride.

For chronic exposure at small U levels, no permanent damage has been reported.
Uranium and Health Issues

No increase in leukemia or other cancers has been linked to exposure to uranium.

Exposure to enriched uranium, used as uranium fuel in nuclear energy production, may present a radiological health hazard.

Exposure to highly enriched uranium may be able to produce bone sarcomas in humans.
Uranium and Health Issues

“Evidence from animal studies suggests that high radiation doses associated with large intakes of $^{234}\text{U}$ and $^{235}\text{U}$-enriched uranium compounds can be hazardous. Adverse effects reported from such exposures include damage to the lungs (fibrosis) and cardiovascular abnormalities” (ATSDR, 1999).”
“However, access to $^{235}$U-enriched or other high specific-activity uranium is strictly regulated by the NRC and the U.S. Department of Energy. Therefore, the potential for human exposure to this level of radioactivity is limited to rare accidental releases in the workplace.” (ATSDR, 1999).
No definitive evidence has been found that links human deaths to uranium exposure.

Among uranium miners, death rates from diseases of the cardiovascular system and the urogenital system were decreased when compared to other populations.
Occupational Exposure

Uranium miners have higher-than-expected rates of death from lung cancer; however, attributed to the radiological effects of radon and its decay products, which are progeny of uranium and, therefore, present in uranium mines.
Radium and Health Issues

Radiotoxicity of radium

$^{226}\text{Ra}$ yields 1 Ci per gram of metal

$^{228}\text{Ra}$ yields 280 Ci per gram of metal

99% of all radium is $^{226}\text{Ra}$ which emits 4.8 MeV energy of alpha particles. $^{228}\text{Ra}$ emits only about 0.021 MeV of beta radiation.
Radium and Health Issues

About 80% of ingested Ra will leave the body in feces. The remaining 20% will enter the bloodstream. Like calcium, a large fraction is preferentially deposited in bone and teeth. Release from bones in human takes years, if not a lifetime.

During the early part of the 20th century in the U.S., radium was thought to be a cure for “high blood pressure, cancer, goiter, stomach trouble, arthritis, female troubles, rheumatism, kidney trouble, constipation . . .”
“Radium cures (c. 1928)"
“Radium cures (c. 1928)”

Contained 1 mCi of $^{226}$Ra and $^{228}$Ra per 0.5 ounce.

Consumed by Eben Byers, owner of a steel company.

He consumed 3 bottles a day for three years.

Teeth fell out. Holes appeared in his skull. Died at the age of 51 in 1932.
Other radium cures (c. 1915 to 1935)
The story of the “Radium Girls”

The Radium Girls were a group of female factory workers who ingested radium from painting watch dials with glow-in-the-dark paint.

Two examples:

The United States Radium factory in New Jersey from 1917 to 1926.

The Radium Dial Company in Ottawa, Illinois.
The story of the “Radium Girls”

Early 1920s: dentists observed increased occurrences of jaw necrosis; bone decay.

1922-1924: Four dial painters died of jaw necrosis.

1925: Possibly first suggestion that jaw decay linked to dial painting.

U.S. Radium Corporation hired a fake doctor.
The story of the “Radium Girls”

Five of the women challenged the system in a court case that established the right of individual workers who contract occupational diseases to sue their employers. All died by the 1930s.

Body content measurements from 27 dial painters were made. This information was used in 1941 by the National Bureau of Standards to establish a tolerance level for radium as 0.1 μCi (3.7 kBq).
Radium girls in Ottawa Illinois

Radium Dial, Inc. began operation in 1916. Moved to a second building in 1930 and changed their name to Luminous Processes, Inc. During World War II, the company prepared luminous dials for the military. Again, the workers were not told about potential health affects (suspected by 1925).
The end of the Ottawa Radium girls

By 1934, seven women were called the “Ottawa Society of the Living Dead.” More joined with time.

The radium-contaminated buildings in Ottawa were demolished in 1969 and 1984, but radium contamination was detected at 16 locations in Ottawa. 13 have been remediated.

Plutonium and Health Issues

Radiotoxicity of Plutonium

$^{241}\text{Pu}$ yields 104 Ci per gram of metal

$^{238}\text{Pu}$ yields 17.3 Ci per gram of metal

Most plutonium isotopes emit a high-energy (generally > 5 MeV) alpha particles and low-energy (< 20 keV) gamma and x-rays as they transform into uranium. The others ($^{241}\text{Pu}$ and $^{243}\text{Pu}$) undergo beta decay and transform into isotopes of americium.
Plutonium and Health Issues

If ingested, very little Pu (≈ 0.05%) is absorbed from the gastrointestinal tract. Very little Pu enters the body by dermal contact (unbroken skin).

Breathing Pu-contaminated air is the greatest threat to human health.

$^{239}\text{PuO}_2$, is only moderately soluble in water, which results in long-term retention in the lung following inhalation exposure.
Plutonium and Health Issues

Some Pu will remain in the lungs and will eventually migrate to bones and the liver.

Pu leaves the body slowly via urine and feces. If Pu enters your lungs today, a portion of it will still be present in the body 30 to 50 years later.
Plutonium and Health Issues

Pu is carcinogenic:
Cancers of the lungs, bone and liver
Pu impairs the immune system.
Birth defects? No clear evidence.
“In the 1950, Queen Elizabeth II was visiting the Harwell [Laboratory], and was handed a lump of plutonium in a plastic bag and invited to feel how warm it was” (from http://www.eoearth.org/article/Plutonium)
Plutonium and Health Issues

Case studies

Lab studies with animals: exposures to “high levels of Pu” can result in
- Decreased life spans
- Diseases of the respiratory tract
- Cancer

Few studies available on humans exposed to smaller levels of plutonium.
Plutonium and Health Issues

Decreased survival has been observed in rats, mice, hamsters, and baboons exposed to $^{239}$PuO$_2$.

In these animal species, death was usually caused by radiation pneumonitis accompanied by edema, fibrosis, and other signs of respiratory damage.

Three monkeys died at 155, 188, and 718 days, respectively, after aerosol exposure to $^{239}$Pu(NO$_3$)$_4$ at levels projected to produce an initial total lung burden of 40 kBq (1.1 μCi); each was diagnosed with radiation pneumonitis.
“The Plutonium Files”  
by E. Welsome

Human experiments with Pu on humans from 1940s to the 1960s.

Crude experiments to use Pu as a “magic bullet” to cure cancer and other diseases.

Sponsored by the Atomic Energy Commission and the Department of Defense (Cold War Era).

Often conducted in secret.

Patients were not always informed as to what was being given to them or why.
Simeon Shaw, "CAL-2," an Australian boy suffering from an osteogenic sarcoma, arrives in San Francisco with his mother, Freda. He was injected with plutonium and two other radioisotopes on April 26, 1946, when he was two months shy of his fifth birthday. He died eight months later. (Courtesy of Joshua Shaw)

William Purcell, "HP-2," a forty-eight-year-old hemophiliac who owned a cigar store, was injected on October 23, 1945, during his thirty-eighth trip to the hospital. He died two and a half years later. (Reprinted with permission of the Democrat and Chronicle, Rochester, NY)

Arthur Hubbard, "CHI-1," was a sixty-eight-year-old businessman whose love of baseball dated from the years he played on his college team. He was injected on April 26, 1945, at Billings Hospital in Chicago, where he had gone for treatment for mouth cancer. He died five months later. (Courtesy of Rita Delmar)

Una Macke, "CHI-2," was injected on December 27, 1945, when she was fifty-six and suffering from two kinds of cancer. The dose she received was equivalent to nearly one hundred times what scientists then believed could be tolerated without harm. She died seventeen days later. (Courtesy of Marcia Sullivan)
Plutonium and Health Issues

The National Academy of Sciences recommended a model in which there is a 15-year latent period following inhalation exposure of Pu in which there are no effects, followed by a 30-year period in which there is a constant risk of:

1.3 chance per million per year per rem for lung cancer.
1.0 chance per million per year per rem for bone cancer.
Plutonium and Health Issues

0.3 chance per million per year per rem for GI and liver cancer from Pu.

If one inhales 10 μg of Pu-239, you may have a 1 in 130 chance of developing cancer as a result.

No studies are known regarding death or lifespan shortening in humans after oral exposure to plutonium.
Possible associations between exposure to plutonium and mortality from diseases of the gastrointestinal tract have been examined in studies of workers at plutonium production and/or used fuel reprocessing facilities in the United Kingdom.

No statistically significant associations between mortality rates from diseases of the digestive tract and exposure to plutonium among workers at these facilities.
Occupational Exposure

Possible associations between exposure to plutonium and mortality from hematopoietic* diseases have been examined in studies of workers at plutonium production and/or processing facilities in the United States (Rocky Flats).

*Hematopoiesis: The production of all types of blood cells
The Rocky Flats Plant was a nuclear weapons-production facility near Denver, Colorado that operated from 1952 to 1988. In 1967, 3,500 barrels of Pu-contaminated oils and solvents were stored; some leaked.

Pu-contaminated soil. Poor management of liquid wastes.
Occupational Exposure

No statistically significant associations between mortality rates from diseases of blood or blood-forming organs and exposure to plutonium among workers at these facilities.
Hanford Site in Washington

By 1963, nine nuclear reactors and five reprocessing plants.

177 underground waste tanks were built.

Peak production from 1956 to 1965.

Produced about 63 short tons of Pu, supplying the majority of the 60,000 weapons in the U.S. arsenal.
No statistically significant associations between mortality rates for bone cancer, musculoskeletal disease, and liver cancer and exposure to Pu among workers at these facilities.
Occupational Exposure

The Mayak (Russia) studies provide evidence for an association between cancer mortality and exposure to plutonium.

Built in 1945-48 to make Pu for weapons.

Five nuclear reactors.

Later reprocessing Pu from decommissioned weapons, and SNF.
The Mayak Site

Data difficult to interpret because:

Multiple accidents and releases of radionuclides.

Safety and health ignored.

Information suppressed or not documented.
Occupational Exposure

No conclusive evidence that Pu produces genetic damage in humans (alteration or mutation of reproductive cells).

Some studies suggest evidence of dose-related increases in chromosomal damage in Pu workers who have with measurable levels of Pu in their bodies.
Cesium and Health

$^{137}\text{Cs}$ decays by beta radiation to $^{137}\text{Ba}$ with a half-life of $30.23 \pm 0.16$ years.

Low toxicity to animals: acute oral $LD_{50}$ for mice and rats range from 800 to 2,000 mg Cs/kg of the animal.

Cesium and Health

Soluble forms of Cs are absorbed by the GI tract. Absorbed Cs behaves like K.

$^{137}\text{Cs}$ is absorbed by dermal contact. If inhaled, $^{137}\text{Cs}$ will become rapidly distributed throughout the body via blood circulation.
Cesium and Health

In a study in which 10 healthy volunteers were fed $^{134}\text{Cs}$- and $^{137}\text{Cs}$-contaminated food: 6% eliminated with a half-life of 0.3 day, 95% eliminated with a half-life of 90 days.

Once cesium enters the body, kidneys begin to remove it from the blood; some cesium is quickly released in the urine. A small portion is also released in the feces.
Cesium and Health

Depending on the dose, radioactive Cs can cause nausea, vomiting, diarrhea, bleeding, coma, and even death.

There are no known studies that link cancer to oral doses of stable Cs.
Cesium and Health

Levels of $^{137}\text{Cs}$ were less than detection limits for all foods analyzed for in the U.S. Food and Drug Administration (FDA) Total Diet Study in 1991–1996 with the exception of honey (ATSDR, 1999) which contained 181 pCi/kg.

FDA limit for accidently contaminated human food as 1,360 Bq/kg food (36,720 pCi/kg).
Cesium in the environment

The maximum concentration of $^{137}$Cs in pasteurized milk from 65 cities in the United States was 14 pCi/L in May 1989

($Chernobyl$ was in 1986)

NRC effluent concentrations (limits) for $^{137}$Cs is 1,000 pCi/L in water and 0.2 pCi/L of air
Tritium and Health

Toxicity of Tritium ($^3\text{H}$)

One proton and two neutrons

Decays as $^3\text{H} \rightarrow ^3\text{He} + \beta^- + \text{anti-}\eta$

Half-life of 12.26 years

By-product of neutron absorption by boron.

Occurs naturally in surface water; 10 to 30 pCi/L
Tritium and Health

$^3$H$_2$O is indistinguishable from ordinary water, and it moves like water in the environment.

$^3$H can enter the body by drinking water or breathing air containing $^3$H gas or as $^3$H$_2$O vapor. Will be completely absorbed into the bloodstream.
Tritium and Health

Become uniformly distributed throughout all biological fluids within one to two hours

Has a biological half-life of about 10 days (can be shortened to 4 to 8 hours using dialysis machines)

As with all beta sources, the major health concern is cell damage caused by ionizing radiation with a potential for future cancer growth.
Tritium and Health

The US EPA decided that the Maximum Contaminant Level (MCL) of $^3$H in drinking water is 20,000 pCi/L.

The life-time cancer mortality risk factor for ingesting $^3$H is $4.4 \times 10^{-14}$/pCi. What does this mean?

Suppose that you drank well water containing the MCL of $^3$H during your entire life. Would this harm you?
If you consumed water with tritium . . .

20,000 pCi/L \times 2\text{L/day} \times 365\text{days/year} \times \frac{78.2\text{years/life-time}}{} = 1.1417 \times 10^{9} \text{pCi}

1.1417 \times 10^{9} \text{pCi} \times 4.4 \times 10^{-14}/\!	ext{pCi} = 5.0236 \times 10^{-5} \quad \text{or 1 chance in 19,906 of death by cancer because of consuming this much tritium. Is this risk acceptable?}
If you consumed water with tritium . . .

Another example,

If a residential drinking water well contained 1,600 pCi/L of $^{3}$H$_{2}$O, the radiation dose would be about 0.3 mrem per year. This dose is about 1,000 times less than background levels (620 mrem).

Nuclear power plants routinely release tritiated water. The NRC ALARA release limit of $^{3}$H$_{2}$O is 3 mrem per year.
Lifetime risk of death by cancer from ingesting contaminated water

Estimated mortality = concentration in water (pCi/L) \times 2\text{L/day} \times 365.4\text{ days/year} \times 78.2\text{ years} \times \text{risk coefficient} \times \text{the number of people}.

Risk Coefficients. “The U.S. EPA has developed mortality risk coefficients for nearly all radionuclides to estimate the lifetime risk of incurring by a fatal cancer from environmental exposures.”
U.S. EPA Risk coefficients

“These coefficients have been calculated by state-of-the-art methods and computer models that averaged over age, gender dependence of intake, metabolism, and radiogenic risk, as well as competing causes of death, to estimate health risks from internal and external exposures.”

Example: $^{241}\text{Am} = 9.5 \times 10^{-11}/\text{pCi}$
Lifetime risk of death by cancer from ingesting contaminated water

Let the concentration = 1 pCi/L. A person drinks 2L per day for his or her entire life.

$^{241}\text{Am} = 5$ deaths in $1,000,000$ more than background.

$^{36}\text{Cl} = 2$ deaths in $10,000,000$

$^{99}\text{Tc, }^{129}\text{I} = 1$ death in $1,000,000$

$^{242}\text{Pu} = 7$ deaths in $1,000,000$

$^{226}\text{Ra} = 166$ deaths in $1,000,000$
Linear relationship between predicted mortality and concentration
Lifetime risk of death by cancer from inhaling contaminated air

Estimated mortality = concentration in air \((\text{pCi/m}^3)\) \(\times\) 20 \(\text{m}^3/\text{day}\) \(\times\) 365.4 days/year \(\times\) 78.2 years \(\times\) risk coefficient \(\times\) the number of people.

Inhalation risk coefficients

Example: \(^{242}\text{Pu} = 2.8 \times 10^{-8}/\text{pCi}\)
Inhaling Plutonium dust

Let the concentration of particulate matter = 0.01 \( \eta g/m^3 \)

\( 10^{-11} g \times 0.004 \text{ Ci/g (specific activity)} = 0.04 \text{ pCi/m}^3 \)

\( (0.04 \text{ pCi/m}^3) \times (20 \text{ m}^3/\text{day}) \times 365.4 \text{ days/year} \times 78.2 \text{ years} \times 2.8 \times 10^{-8}/\text{pCi} \times 10^6 \text{ people} = 640 \) cases of fatal cancer greater than background.

Unrealistic scenario.
Occupational exposure to Pu

\[(0.04 \text{ pCi/m}^3) \times (60 \text{ m}^3/\text{work day}) \times 300 \text{ days} \times 20 \text{ years} \times 2.8 \times 10^{-8}/\text{pCi} \times \ldots \]

1,000 people at a plant: < 1 person

10,000 people at a plant: 1 person
How toxic is Pu?

Plutonium is constantly referred to by the news media as “the most toxic substance known to mankind.” (Ralph Nader, activist and lawyer)

Is this true?

Radiological toxicity versus chemical toxicity
Long-term carcinogen because of its radiotoxicity but $^{226}\text{Ra}$ is about 200 times more radiotoxic than plutonium.
How toxic is plutonium?

No evidence that Pu is acutely toxic in short-term, oral exposures.
The “poison is in the dose”, but arsenic, parathion (a banned insecticide), HCN, sarin (a banned nerve gas) and botulinum toxin are more acutely toxic than Pu.

Botulinum (baht-chu-line-um) toxin is protein produced by bacteria (Clostridium botulinum), and is thought to be the most toxic substance known with an human LD$_{50}$ of roughly 0.005 to 0.05 µg/kg.
How toxic is plutonium?

If we gave 100 people who weighed 60 kg, a dose of about 1.7 ug of botulinum toxin given to each person, it would kill 50 people in a matter of days (nerve paralysis).

Needless to say, the claims about plutonium toxicity have been exaggerated.