$^{210}$Po in drinking water, the MCL, and health effects of low-level exposure.

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***The opinions expressed here are mine and are not those of the USGS or US Government***
Polonium and me

Between 1997 and 2001, 16 cases of acute childhood leukemia were diagnosed in Fallon, Nev. Nine cases were diagnosed in 2000 alone.

I was in charge of the investigation of ground-water quality USGS did to support the CDC/ATSDR and State investigations of the cluster.
Excess radioactivity

What would be expected from uranium

Intermediate aquifer
Basalt aquifer
Shallow aquifer

MCL
What is the source of the excess radioactivity?

226Ra $t_{1/2} = 1601$ yr
228Th $t_{1/2} = 1.90$ yr
210Po $t_{1/2} = 138.4$ d
224Ra $t_{1/2} = 3.64$ d
Fallon $^{210}$Po investigation

- Initial reconnaissance in 2007 funded by USEPA. Further work funded by Churchill County and USGS.

- From 2007-2010, USGS analyzed 86 wells in Fallon area

  - $^{210}$Po levels ranged from $<1$ mBq/L to 6.6 Bq/L.
  - $^{210}$Po exceeded 555 mBq/L (the MCL) in 30% of the domestic wells.
$^{210}$Po at dairies

- $^{210}$Po exceeding 2 Bq/L was found in stockwater at two dairies.
Fallout from release of the highly radioactive data

Study finds elevated levels of polonium-210 in rural wells

Two local dairies forced to dump thousands of gallons of milk

By VIKTORIA PEARSON

Fallon is the most important dairy area in northern Nevada.

FDA measured $^{210}\text{Po}$ in the milk and reported it was safe.
Chemistry of contaminated areas

- Processes mobilizing $^{210}$Po from Fallon sediments.
- Compared Fallon water with contaminated waters from
  - Florida
  - Maryland
  - Finland
In Fallon, radon is low and $^{210}\text{Pb}$ is less than the $^{210}\text{Po}$, indicating $^{210}\text{Po}$ is being mobilized from the sediments and not from decay of radon in the water.

$^{210}\text{Po}$ mobilization in the Nevada and Florida wells involves $\text{SO}_4^-$ reducing bacteria.

The Maryland well is probably similar, but $\text{H}_2\text{S}$ and DO analyses were not available to confirm it.
The average $^{238}$U concentration in US sediments is 2.7 mg/kg.

The $^{210}$Po in secular equilibrium with that amount of $^{238}$U would be about 33 Bq/kg.

Assuming typical sediment porosity and density, there is sufficient $^{210}$Po present in most US sediments that processes that mobilize only 1% of it could yield water with $>1$ Bq/L.
Finnish wells

- The Finnish wells are drilled in granite bedrock. They have very high radon concentrations and $^{210}\text{Pb}$ is greater than $^{210}\text{Po}$.

- These data mean $^{210}\text{Po}$ in Finnish wells is coming from decay of radon. The $^{210}\text{Pb}$ and $^{210}\text{Po}$ adsorb to colloids in the water and do not settle out.
Exposure can be extremely high

- A public-supply well with 1.7 Bq/L was found servicing a subdivision in Charles County Maryland.
- A public-supply well with 22 Bq/L was found servicing the Veteran’s Center in Alexandria Louisiana.
- Contaminated private wells have been found:
  - in Fallon Nevada, maximum 6.6 Bq/L.
  - in central Florida, maximum 26 Bq/L.
  - in Finland, maximum 14 Bq/L.
But in typical US groundwater

- $^{210}\text{Po}$ activity in groundwater is usually $<5$ mBq/L.
- In the US fewer than 100 wells have been reported with $>555$ mBq/L.
- Even studies that specifically went looking for $^{210}\text{Po}$ have rarely found it.
  - Ruberu et al. (2007): 3/26 $> 150$ mBq/L; median 3 mBq/L
  - Arndt (2010): 6/79 $> 150$ mBq/L; median 4 mBq/L
Three reasons $^{210}$Po is rare

1. Because of its chemical properties.
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2. Because nobody samples for it because it is extremely rare, so it is never found and therefore is extremely rare.
Three reasons $^{210}\text{Po}$ is rare

1. Because of its chemical properties.

2. Because nobody samples for it because it is extremely rare, so it is never found and therefore is extremely rare.

3. Because the Gross Alpha MCL is seriously flawed if used to monitor for $^{210}\text{Po}$. 
Let’s talk about the MCL

- The US does not have a drinking-water standard specific for $^{210}$Po. The only applicable standard in the US is Gross Alpha Radioactivity.

- The EU standard for $^{210}$Po is 100 mBq/L and the Canadian standard is 200 mBq/L.

- USEPA (2000) concluded a MCL specific for $^{210}$Po would be 41 mBq/L based on a risk level of 1:10000 for lifetime total cancer risk.

- Thus, the MCL is 13 times greater than a health based standard for $^{210}$Po would be.
Developing the MCL

In 2000 when the US standard was promulgated, EPA believed $^{210}$Po was extraordinarily rare:

- USEPA relied heavily on a USGS reconnaissance of Public Water Supply wells from 27 States that found $^{210}$Po activity >41 mBq/L in only 2 of 95 wells.
- The only high levels known in the US during their data analysis were the Florida wells. They missed the Louisiana well because the reference was very obscure.
Developing the MCL

- EPA believed $^{210}$Po would only occur in unusual geochemical environments that would not be developed as water supplies:
  - High $^{210}$Po was only known from shallow, phosphate-mineralized deposits.
  - $^{210}$Po was only known from very acidic water (pH < 6) and investigators thought it would precipitate out at higher pH.
My opinion about the MCL

- Gross Alpha is a terrible way to regulate $^{210}\text{Po}$. 
- $^{210}\text{Po}$ decays during permitted holding times.
  - The holding-time permitted used to be a year (84% $^{210}\text{Po}$ loss). It is now 6 months (60% $^{210}\text{Po}$ loss).
  - Quarterly sample compositing is permitted (64% $^{210}\text{Po}$ loss).
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- Po may be volatile and lost during sample preparation.
  - 50% of $^{210}\text{Po}$ in a Florida well was lost during sample bubbling. Like radon, volatile $^{210}\text{Po}$ species will be lost during sample drying.
  - Inorganic Po may be driven off during sample flaming done to remove hygroscopic salts.
But mostly it’s terrible because

- If there is a Gross Alpha exceedance, regulators can request that it be fixed but they can’t request that it be explained.
- Blending water could reduce exposure to $^{210}$Po, but the water could still have $^{210}$Po levels $\gg 41$ mBq/L.
- The current rule adds no new occurrence data.
Changing topics
Effects of low-level $^{210}$Po exposure

Review

Occurrence of $^{210}$Po and Biological Effects of Low-Level Exposure: The Need for Research

Ralph L. Seiler$^1$ and Joseph L. Wiemels$^2$

$^1$Environmental Sciences Graduate Program, University of Nevada, Reno, Reno, Nevada, USA; $^2$Laboratory for Molecular Epidemiology, University of California at San Francisco Helen Diller Comprehensive Cancer Center, San Francisco, California, USA

BACKGROUND: Polonium-210 ($^{210}$Po) concentrations that exceed 1 Bq/L in drinking-water supplies have been reported from four widely separated U.S. states where exposure to it went unnoticed for decades. The radionuclide grandparent of $^{210}$Po are common in sediments, and segments of the isotope in the $^{238}$U decay series and is present in the environment wherever $^{238}$U or any other members of the $^{238}$U decay series, such as $^{226}$Ra, $^{222}$Rn, and $^{210}$Po, occur. The average annual

**Basic Toxicology:**

- $^{210}$Po is a Group 1 human carcinogen and numerous animal studies have demonstrated its carcinogenicity.
- $^{210}$Po accumulates in soft tissues.
- $^{210}$Po is only harmful if it is ingested because alpha particles are stopped by the skin.
α-particle irradiation

- Substantial doses of radiation (~0.5 Gray) are delivered to individual cells traversed by a single alpha particle.
- On average, an alpha particle traversing a nucleus will produce several hundred single strand breaks and about 10 double-strand breaks.
- DSBs can cause apoptosis.
- Incorrectly repaired DSBs can cause deletions, translocations, and fusions in DNA.
The bystander effect

- After α-particle irradiation, irradiated cells produce chemicals that can cause apoptosis and chromosomal instability in un-irradiated cells.

- These chemicals can migrate substantial distances (>0.5 mm) in tissue.
The ovary may be the critical organ for determining the lowest injurious dose to $^{210}\text{Po}$ (Finkel et al., 1953).

$^{210}\text{Po}$ in the ovary is located principally in the follicles where oocytes develop (Samuels, 1966).

Significant killing of oocytes occurred at exposures of 40 mBq/g body weight in mice (Samuels, 1966).
How $^{210}$Po could affect fertility

- A woman is born with a fixed number of egg cells, \(~300,000\). This is the ovarian reserve.
- A tropism of $^{210}$Po to follicle cells would expose immature egg cells to alpha radiation.
- The sensitivity of the egg cells to $\alpha$-particles may be caused by their sensitivity to bystander signals.
- Egg cells that die will never be replaced.
- Exposure as a fetus, infant, child, or adult could all affect a woman’s fertility.
Lung Cancer: Is the Increasing Incidence Due to Radioactive Polonium in Cigarettes?*

JEROME MARMORSTEIN, MD, Santa Barbara, Calif

ABSTRACT: This paper presents clinical, experimental, and epidemiologic evidence to help explain the rapidly increasing incidence of primary lung cancer, with recently observed reversal in leading cell type from squamous cell to adenocarcinoma. It postulates that this may be due to changes in modern cigarettes, with or without filters, which allow inhalation of increased amounts of radioactive lead and polonium and decreased amounts of benzopyrene. This hypothesis is based upon measurements of increased concentrations.

- \(^{210}\text{Po}\) is present in cigarettes, probably from radioactive contaminants in phosphate fertilizers.
- A smoker is exposed to from 13 to 590 mBq/d of \(^{210}\text{Po}\).
- A person drinking \(^{210}\text{Po}\)-contaminated drinking water that meets the MCL could be exposed to almost twice that much \(^{210}\text{Po}\).
Smoking and female infertility

There is a significantly increased risk of infertility in women who smoke.

Cigarette smoke depletes the ovarian follicle reserve.
Could the effects of cigarette smoking on female fertility be explained by $^{210}\text{Po}$ exposure?

- PAHs in cigarettes (e.g. Benzo[a]pyrene) can also affect ovarian reserve.

If $^{210}\text{Po}$ is the link between smoking and infertility, then $^{210}\text{Po}$ doses of 13-590 mBq/d may be sufficient to affect fertility regardless of the source.
Conclusions

- $^{210}$Po is common in the environment but exposure to it in drinking water is very rare.
- The Gross Alpha MCL is seriously flawed if the objective is to protect people from $^{210}$Po in drinking water.
- $^{210}$Po accumulates in the ovary and the ovary may be the critical organ in determining safe levels of exposure.
- Exposure to $^{210}$Po may kill or damage critical reproductive cells.
Basic health research needs

- Epidemiological research is needed in areas of known $^{210}$Po contamination.

- Do contaminated stockwater wells lead to contaminated food chains?

- New research is needed on the toxicology of $^{210}$Po because much of the existing toxicological research was done in the 1950’s and 1960’s.
  - Accumulation and distribution of $^{210}$Po in the ovary and fetus.
  - Sensitivity of follicle to bystander signals.
Next time USEPA considers the rules

- ‘Fixing’ a Gross Alpha exceedance should require identifying its cause.
- Every PWS should have 1-2 direct measurements for Ra and $^{210}$Po. Subsequent GA analyses can be used to monitor for changes.
- Samples should be analyzed within 1 week of sample collection.
- Sample compositing should not be permitted.
More unwanted suggestions

- Fund State and National reconnaissances for $^{210}\text{Po}$ in drinking water.

- A good starting place would be a National reconnaissance of public-supply wells where existing Gross Alpha data show $^{210}\text{Po}$ may be present.
### Undiscovered contamination?

#### National Drinking Water Database

**Skyview Park - Gastonia, NC**

Serves 170 people - Test data available: 2004-2009

#### Contaminants Exceeding Health Guidelines

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Average/Maximum Result</th>
<th>Health Limit Exceeded</th>
<th>Legal Limit Exceeded</th>
<th>04</th>
<th>05</th>
<th>06</th>
<th>07</th>
<th>08</th>
<th>09</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alpha particle activity (excl radon and uranium)</td>
<td>27.55 pCi/L 29 pCi/L</td>
<td>Yes MCLG: 0 pCi/L</td>
<td>Yes 15 pCi/L</td>
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<td>Manganese</td>
<td>645 ppb 645 ppb</td>
<td>Yes 50 ppb</td>
<td>Yes 50 ppb</td>
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<tr>
<td>Combined Radium (-226 &amp; -228)</td>
<td>1.59 pCi/L 2.55 pCi/L</td>
<td>Yes MCLG: 0 pCi/L</td>
<td>No 5 pCi/L</td>
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<tr>
<td>Radium-226</td>
<td>0.64 pCi/L 0.96 pCi/L</td>
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<tr>
<td>Radium-228</td>
<td>0.48 pCi/L 0.65 pCi/L</td>
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#### Other Detected Contaminants

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<tr>
<th>Contaminant</th>
<th>Average/Maximum Result</th>
<th>Health Limit Exceeded</th>
<th>Legal Limit Exceeded</th>
<th>04</th>
<th>05</th>
<th>06</th>
<th>07</th>
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<th>09</th>
</tr>
</thead>
<tbody>
<tr>
<td>Combined Uranium (pCi/L)</td>
<td>2 pCi/L 3.1 pCi/L</td>
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<td>No 15 pCi/L</td>
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</tbody>
</table>
### National Drinking Water Database

#### Muirfield Village - Portland, ME
- Serves 33 people - Test data available: 2004-2007

This drinking water quality report shows results of tests conducted by the water utility and provided to the Environmental Working Group.

### Contaminants Exceeding Health Guidelines

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Average/Maximum Result</th>
<th>Health Limit Exceeded</th>
<th>Legal Limit Exceeded</th>
<th>Testing History</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium-238</td>
<td>0.03 pCi/L 0.03 pCi/L</td>
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<td>Yes 0.03 pCi/L</td>
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<tr>
<td>Alpha particle activity (excl. radon and uranium)</td>
<td>22.45 pCi/L 24 pCi/L</td>
<td>Yes MCLG: 0 pCi/L</td>
<td>Yes 15 pCi/L</td>
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<tr>
<td>Radon</td>
<td>20265.3 pCi/L 29500 pCi/L</td>
<td>Yes 1.5 pCi/L</td>
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<tr>
<td>Radium-226</td>
<td>0.4 pCi/L 0.4 pCi/L</td>
<td>Yes MCLG: 0 pCi/L</td>
<td>No 5 pCi/L</td>
<td></td>
</tr>
</tbody>
</table>

**NOTE:** Each dot in the above graph represents one month.
More unwanted suggestions

- Fund State and National reconnaissances for $^{210}\text{Po}$ in drinking water.
  - A good starting place would be a National reconnaissance of public-supply wells where existing Gross Alpha data show $^{210}\text{Po}$ may be present.

- If Congress doubles or triples USEPA funding:
  - Private domestic wells that tap the same aquifers as contaminated Public Supply Wells should be tested.
  - Geochemical investigations of contaminated wells to identify processes mobilizing $^{210}\text{Po}$ should be done.
Thank You

- Citations for data presented here are in:

- Available on ResearchGate

- Contact me rlseiler@juno.com