

Evidence of High Levels of Radium and Radon in Hakes and Chemung Landfills

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We see strong evidence of high levels of radium and radon in Hakes and Chemung Landfills.

DEC & the landfill operator disagree, *based on a different line of evidence.*

DEC has a process for resolving disputed issues (*Issues Conference & Adjudicatory Hearing*) but hasn't been willing to use it!

Radon testing of landfill gas is a crucial first step toward resolving this. Testing of leachate by EPA Method 901.1 (for Pb-214 and Bi-214) also needs to be resumed.

Our evidence: Radon levels in *landfill leachate* are intermittently very high, indicating that radon levels in *landfill gas* are also high. Radon comes from radium, so radium in the landfill must also be high.

DEC claims their regulatory limit for radium in waste (25 pCi/g) is met and is enforced by radiation monitoring instruments at the landfill gates. We don't find these monitors effective.



Matt Richmond photo, <http://archive.alleghefront.org>

This is the type of radioactivity monitoring used at Hakes landfill gate to check on radium levels in waste loads

Radiation monitoring at Hakes landfill gate is intended to limit incoming waste loads to no more than 25 pCi/gram of radium

- **This type of monitoring cannot serve the intended purpose because highly variable and inconsistent levels of Lead-214 (Pb-214) and Bismuth-214 (Bi-214) interfere with radium monitoring**
- **Waste truckloads with up to 60-fold variations in their radium levels may exhibit the same or similar monitor readings**

Thus: DEC's method of monitoring for radium at the landfill gate is not reliable.

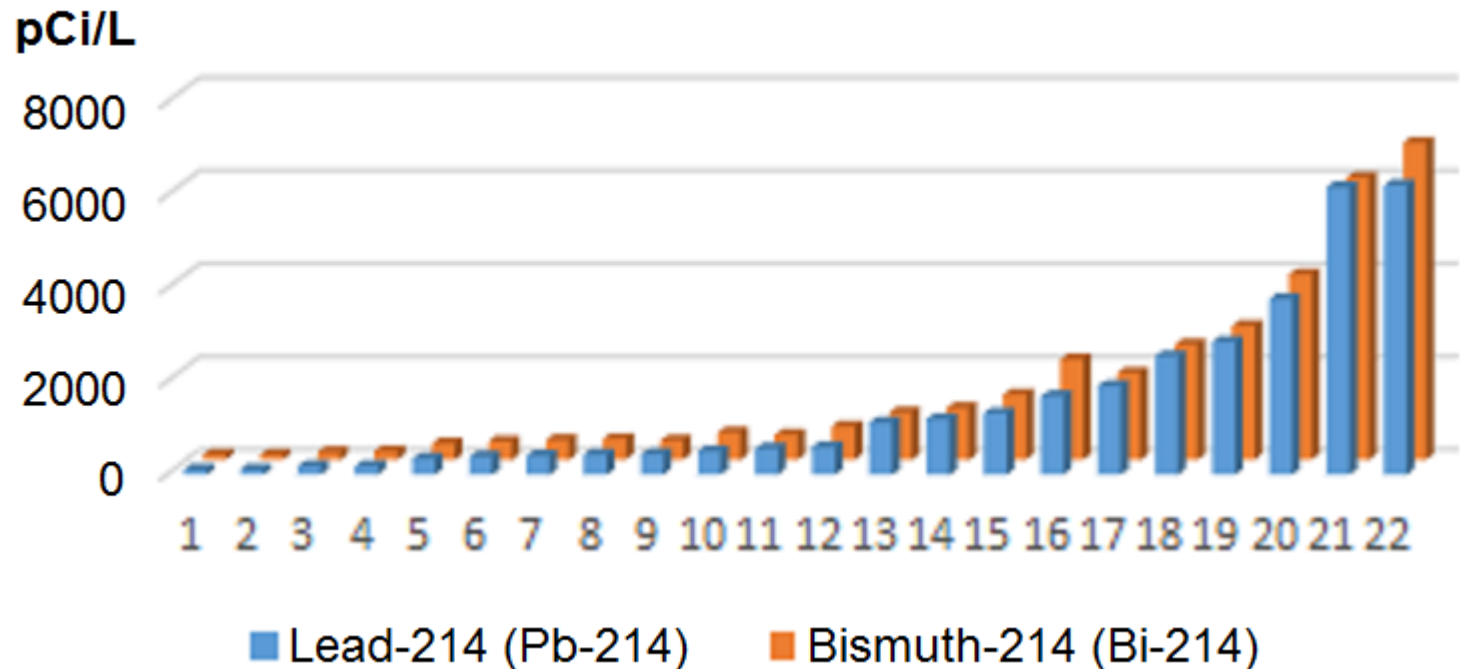
Our evidence, as already noted, indicates more radium than DEC acknowledges:

Radon levels in landfill leachate are intermittently very high, indicating that radon levels in landfill gas are also high. Radon comes from radium, so radium in the landfill must be high.

Most of the data I've reviewed is from Hakes landfill, but Chemung leachate tests also show intermittently high radon.

Test results from 22 Hakes leachate samples (between 2012 and mid-2018) in which Pb-214 and Bi-214 exceeded 100 picocuries per liter

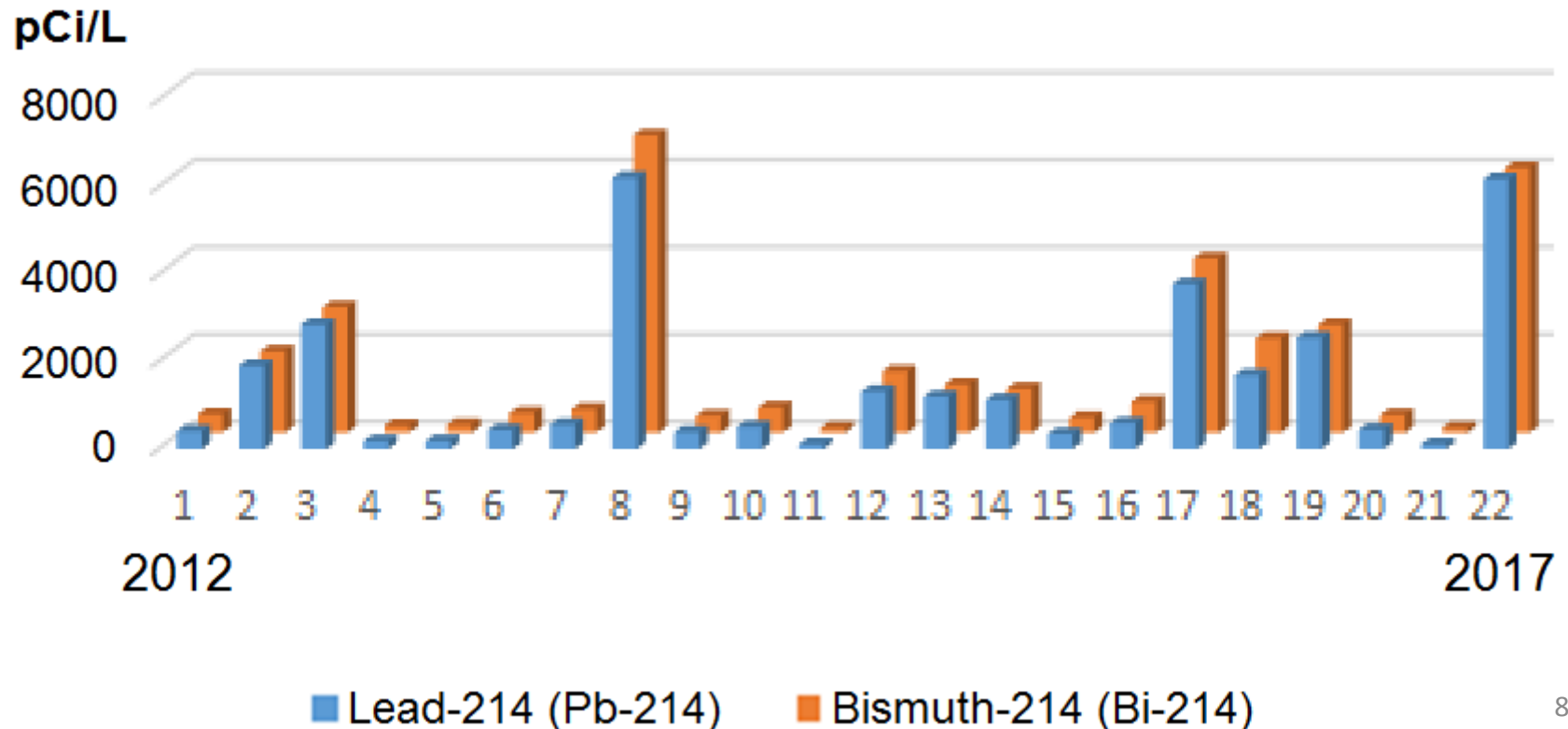
The other 84 Hakes leachate samples tested during this period did *not* exceed 100 pCi/L for either Pb-214 or Bi-214. For the other 84 samples, the average test result for Pb-214 and Bi-214 was about 16 pCi/L. For all 106 samples, the radium test result was very low (just a few pCi/L).



Test results from 22 Hakes leachate samples (between 2012 and mid-2018) in which Pb-214 and Bi-214 exceeded 100 picocuries per liter

Same data, shown here as time trend. Note that testing of Pb-214 and Bi-214 stopped in mid-2018.

For all samples, the radium test result was very low.



Low radium test results in landfill leachate may seem reassuring – but *aren't* because of the high and extremely variable test results for Pb-214 and Bi-214.

These results matter because *Pb-214 and Bi-214 are produced by radioactive decay of radium (Ra-226) and radon (Rn-222).*

All four of these radionuclides are part of the same radioactive “decay chain.”

Uranium-238 (4.5 billion years)



Thorium-234 (24 days)



Protactinium-234m (1.2 minutes)



Uranium-234 (240,000 years)



Thorium-230 (77,000 years)



Radium-226 (1,600 years)



Radon-222 (3.8 days) (**GAS**)



Polonium-218 (3.1 minutes)



Lead-214 (27 minutes)



Bismuth-214 (20 minutes)



Polonium-214 (160 microseconds)



Lead-210 (22 years)



Bismuth-210 (5.0 days)



Polonium-210 (140 days)



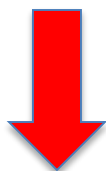
Lead-206 (stable)

Uranium-238 decay series (half-life in parentheses)



**PARENT
RADIONUCLIDE**

**PROGENY
or
DAUGHTER
or
DECAY PRODUCT**



Uranium-238 (4.5 billion years)



Thorium-234 (24 days)



Protactinium-234m (1.2 minutes)



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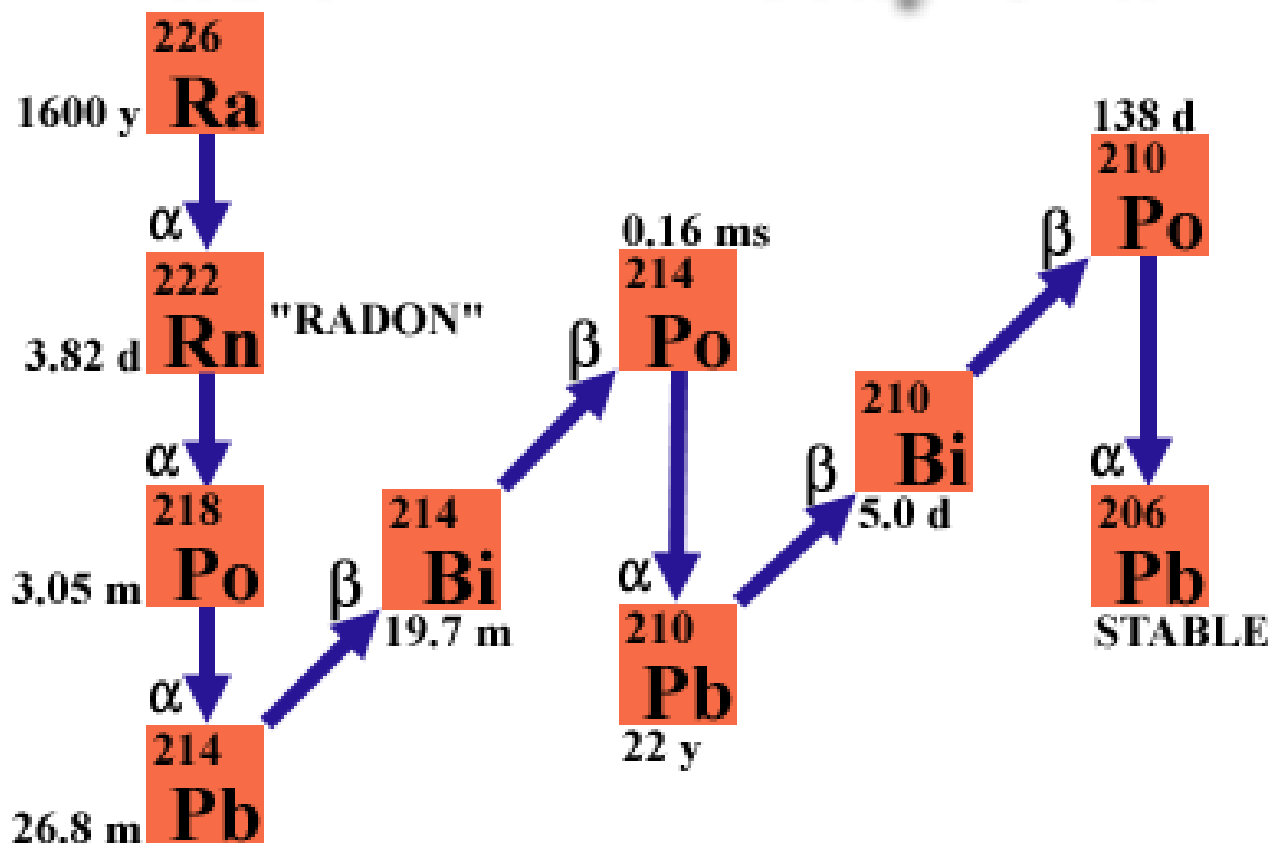
Polonium-210 (140 days)



Lead-206 (stable)

Uranium-238 decay series (half-life in parentheses)

Radium-226 Decay Chain



Pb = Lead

ACCOUNTING FOR RADIOACTIVE ATOMS WITHIN THIS DECAY CHAIN

During radioactive decay, **atoms are transformed into different atoms.** They don't just disappear entirely or appear out of nowhere.

Accounting for these atoms can always be done, at least in principle. It is simplest when the atoms are either trapped in solid rock, or sealed in a sample jar, for long enough to reach “secular equilibrium.”

Secular Equilibrium

Secular equilibrium occurs if/when a relatively long-lived parent radionuclide such as radium is enclosed in a tight geologic matrix (solid rock) *or in a sealed container*, thus keeping progeny such as radon trapped very close to the parent

After a sufficient time interval, the activity of the progeny (in pCi) tends to be the same as the activity of the parent radionuclide. (The progeny stay “in sync” with the decay rate of the parent.)

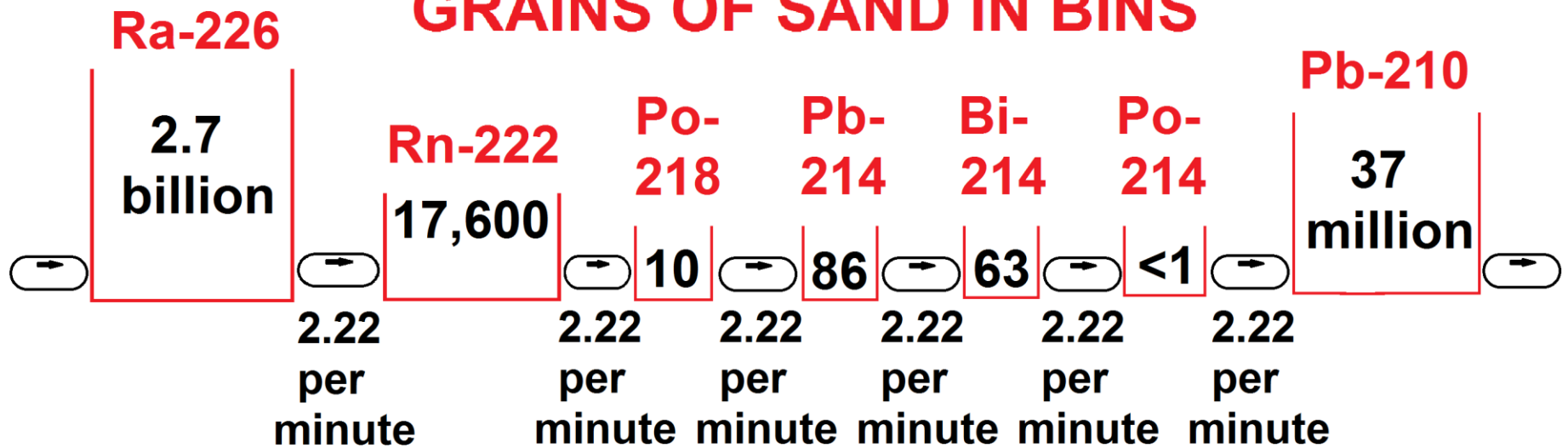
And even if the progeny are initially absent, they’ll be generated and “catch up” if the parent is put into a sealed container.

Starting with radium but none of its progeny in a sealed container, a period of about **21 days** is long enough to (re)establish secular equilibrium from radium on down to Bi-214. (In pCi, this means that $\text{Ra-226} = \text{Rn-222} = \text{Pb-214} = \text{Bi-214}$.)

Starting with radon but none of its progeny in a sealed container, a period of about **5 hours** is long enough to (re)establish secular equilibrium from radon on down to Bi-214. (In pCi, this means that $\text{Rn-222} = \text{Pb-214} = \text{Bi-214}$.)

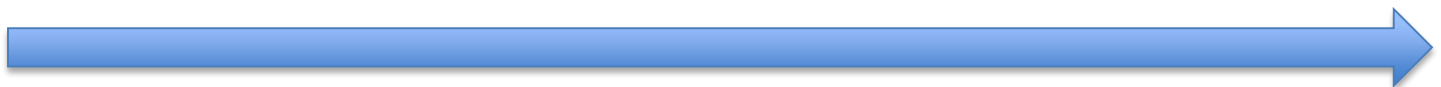
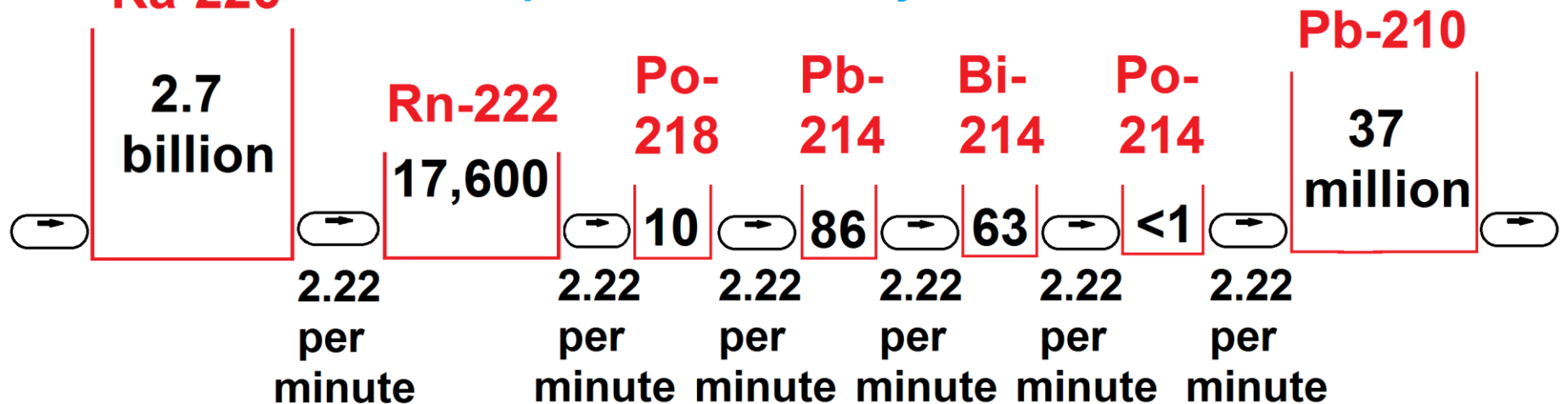
Pb-214 and Bi-214 have such short half-lives that **they'll be gone within ~5 hours** if not constantly replenished by radon decay. Any found in a sample must be less than about 5 hours old.

GRAINS OF SAND IN BINS



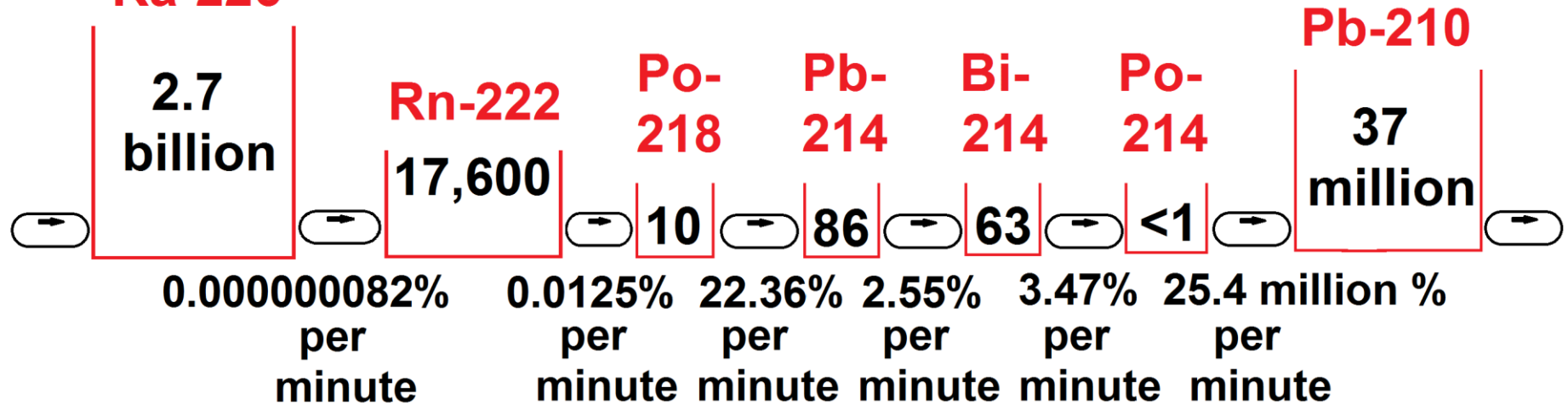
ATOMS IN DECAY CHAIN (1 pCi OF EACH RADIONUCLIDE)

Ra-226 Secular equilibrium: "Conveyors" are unbroken



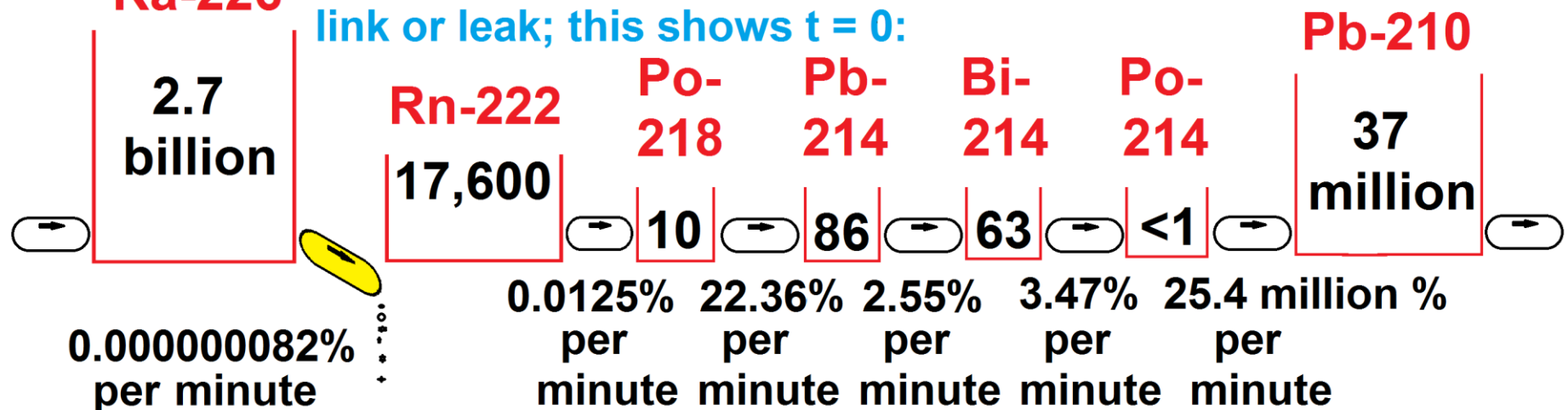
ATOMS IN DECAY CHAIN (1 pCi OF EACH RADIONUCLIDE)

Ra-226 Secular equilibrium: "Conveyors" are unbroken



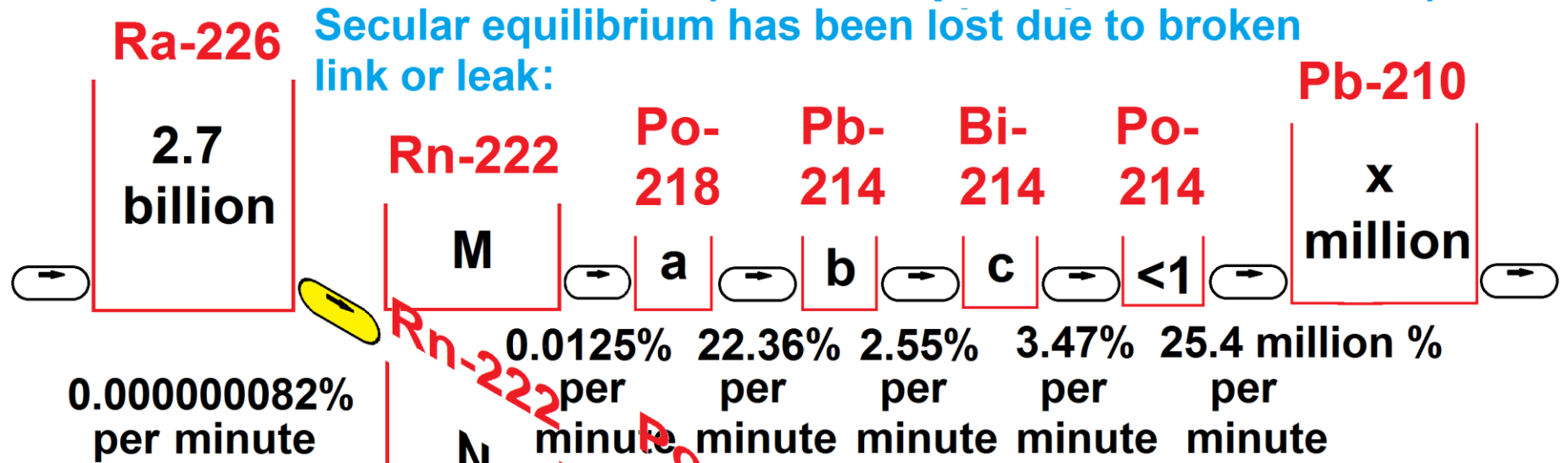
ATOMS IN DECAY CHAIN (1 pCi OF EACH RADIONUCLIDE)

Ra-226 Secular equilibrium has just been lost due to broken link or leak; this shows $t = 0$:



...but the "bins" from Rn-222 onward will be gradually depleted without ongoing input from Ra-226

ATOMS IN DECAY CHAIN (FROM 1 pCi OF RADIUM-226)



$$M + N = 17,600$$

$$a + d = 10$$

etc.

NOTE:

When there's a leak or other broken link in the decay chain, *the decay chain hasn't stopped.*

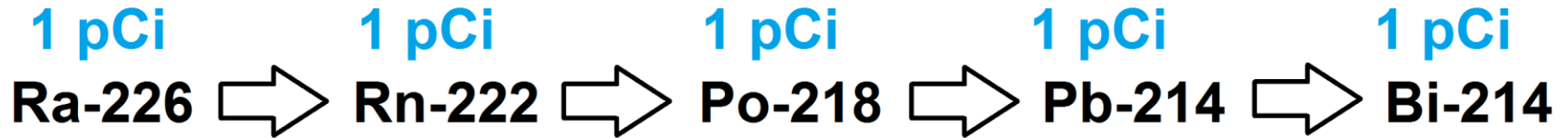
It has shifted to a new location (e.g., because the wind carries radon gas in a new direction).

The bin and conveyor-belt analogies on the preceding slides are for just *one picocurie* (1 pCi)

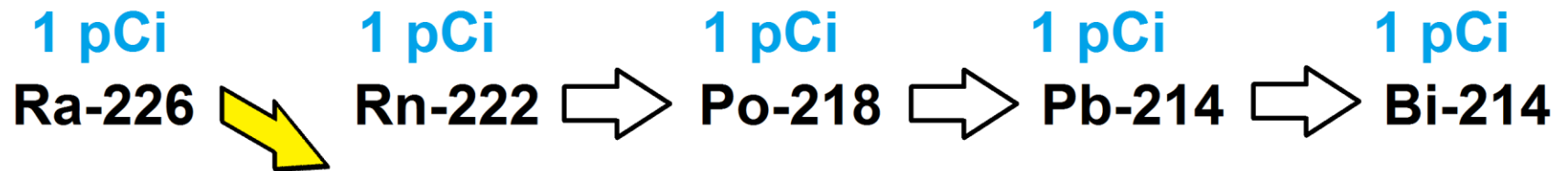
Multiply all the bin quantities by a higher number (such as 6000) when dealing with a higher number of picocuries – as in some of the Hakes landfill leachate samples where thousands of picocuries are present in each liter

(Quantities in bins *will always be proportional* to the bin quantities shown on preceding slides.)

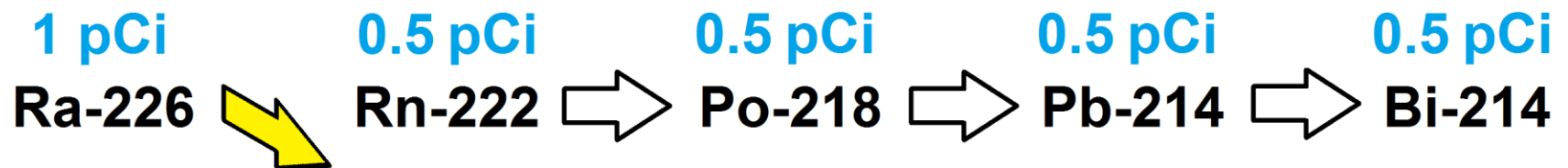
1) AT SECULAR EQUILIBRIUM:



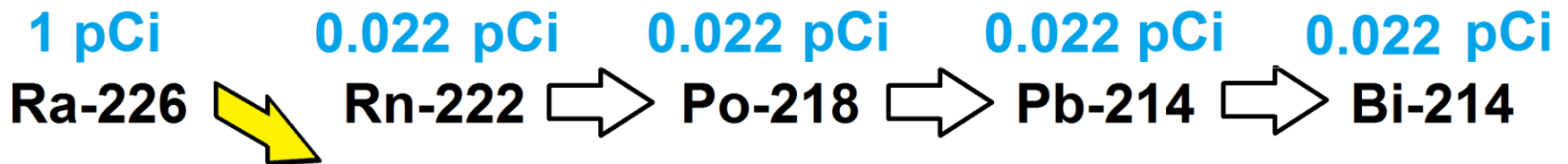
2) SECULAR EQUILIBRIUM LOST; t = 0:

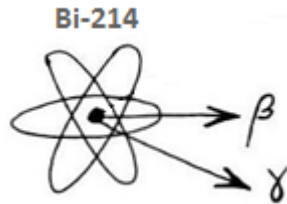


3) SECULAR EQUILIBRIUM LOST; t = 3.82 days:

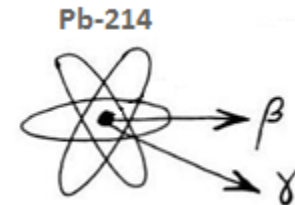
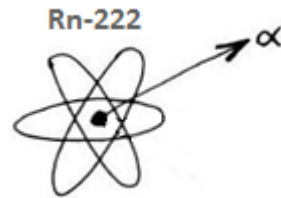


4) SECULAR EQUILIBRIUM LOST; t = 21 days:



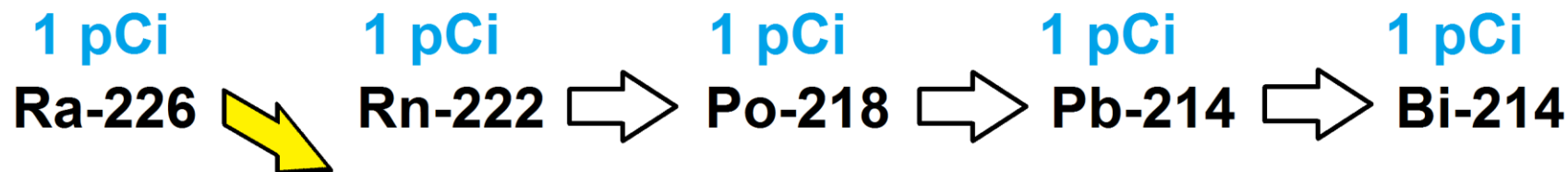


Radon-222 must be present in a sample at approximately the same activity as Lead-214 and Bismuth-214 if the sample is more than about 5 hours old...

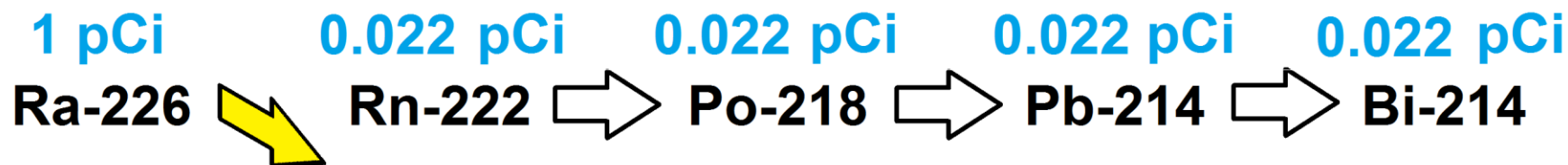


**Applying these
radiological principles
to Hakes leachate test results**

EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 0$:

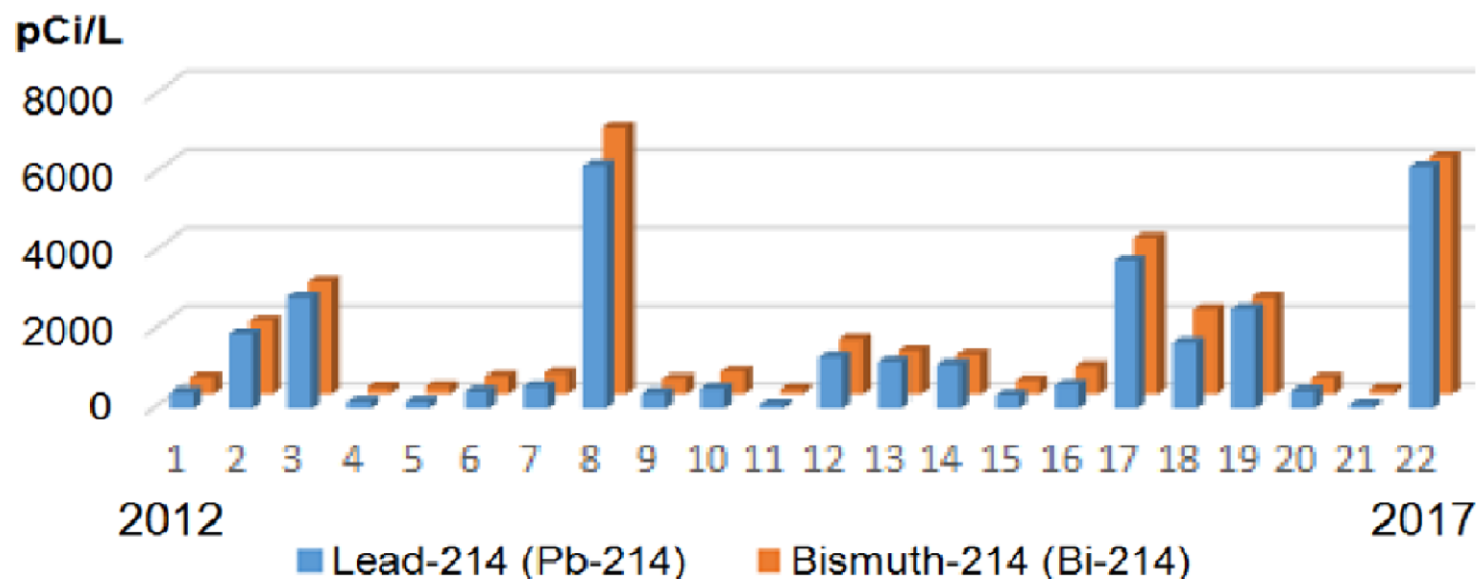


EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 21$ days:



Test results from 22 Hakes leachate samples

...in which radium test results remain very low (typically 2-3 pCi/L)



EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 0$:


1 pCi
Ra-226

1 pCi
Rn-222

1 pCi
Po-218

1 pCi
Pb-214

1 pCi
Bi-214



EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 21$ days:

1 pCi
Ra-226

0.022 pCi
Rn-222

0.022 pCi
Po-218

0.022 pCi
Pb-214

0.022 pCi
Bi-214

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graph LR; Ra226[1 pCi Ra-226] -- yellow arrow --> Rn222[0.022 pCi Rn-222]; Rn222 -- white arrow --> Po218[0.022 pCi Po-218]; Po218 -- white arrow --> Pb214[0.022 pCi Pb-214]; Pb214 -- white arrow --> Bi214[0.022 pCi Bi-214];
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LEACHATE SAMPLE at time of sample collection; $t = 0$:

Ra-226 Rn-222 \Rightarrow Po-218 \Rightarrow Pb-214 \Rightarrow Bi-214

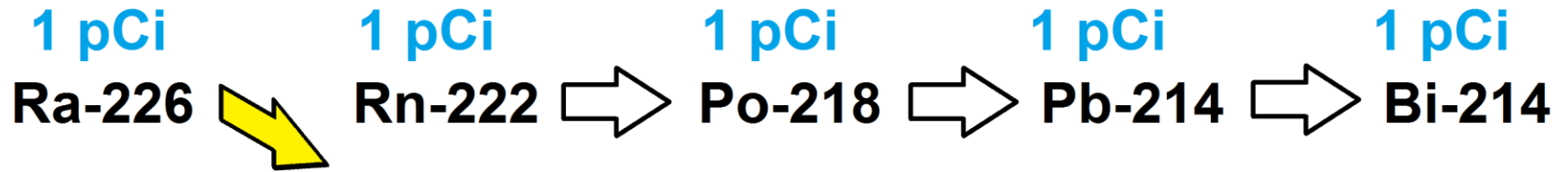
LEACHATE SAMPLE at time of sample testing; t = 21 days:

3 pCi/L
Ra-226

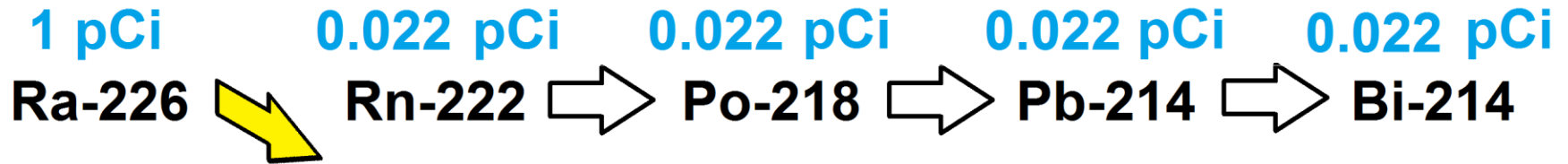
Rn-222 \Rightarrow **Po-218** \Rightarrow **Pb-214** \Rightarrow **Bi-214**

6000 pCi/L **6000 pCi/L**

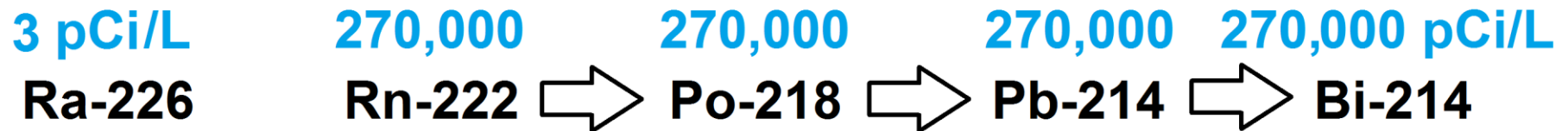
EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 0$:



EXAMPLE: SECULAR EQUILIBRIUM LOST; $t = 21$ days:



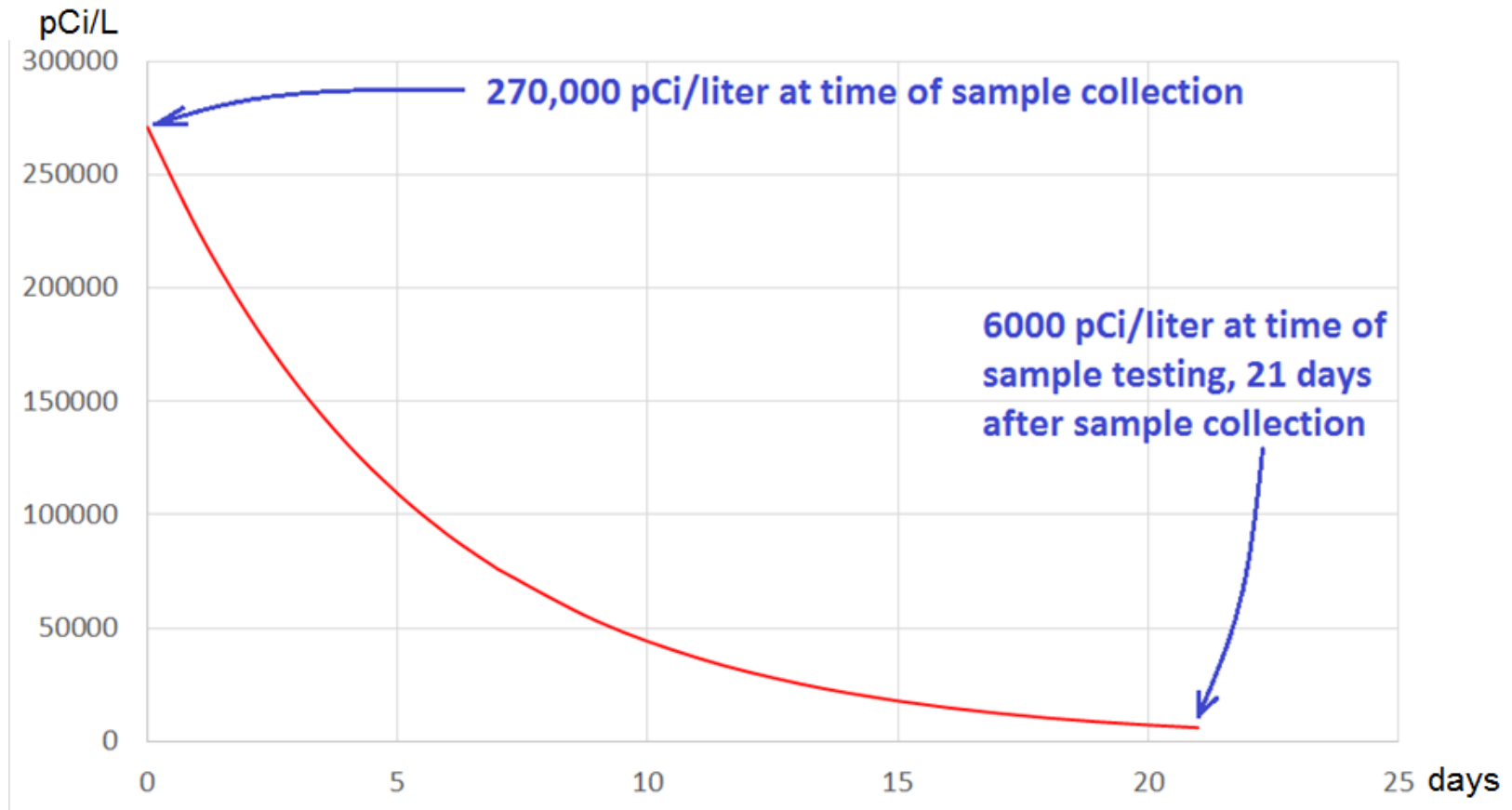
LEACHATE SAMPLE at time of sample collection; $t = 0$:



LEACHATE SAMPLE at time of sample testing; $t = 21$ days:



THUS: Radon activity in some Hakes leachate samples ranges up to ~270,000 pCi/L



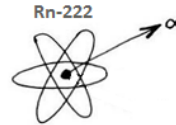
This is a 21-day decay curve for Radon-222 (half-life 3.82 days) in leachate, without secular equilibrium with parent radium, i.e., with essentially no replenishment by parent radium.

Important points: Leachate and Landfill gas

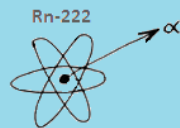
- **Radon activity in Hakes leachate from which samples were collected is intermittently very high, ranging up to ~270,000 pCi/liter**
- **Radon is a radioactive gas. Like other gases, it mixes with air (or landfill gas) and dissolves in water & water-based mixtures such as leachate.**
- **Radon's equilibrium concentration (or activity) in *air* is related to its concentration (or activity) in *water* through known principles of physical chemistry involving *partition coefficient* and/or *Henry's Law*. (Provides a good approximation for water-based mixtures such as leachate.)**

**At equilibrium in a sealed container,
at 20°C**

**1.05 million
pCi/L Radon-222
in air**



**270,000 pCi/L
Radon-222**

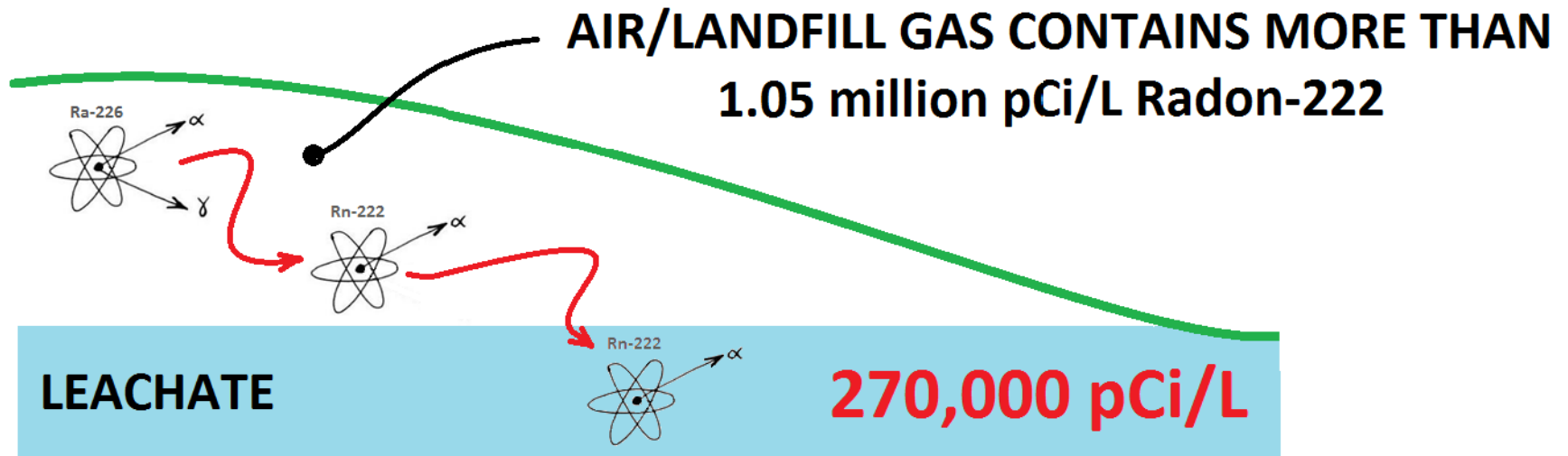


in water

**Not at full equilibrium in an
imperfectly sealed landfill, at ~20°C**

**Likely explanation: *Radium* remains above the
leachate; it's “high and dry” in the landfill.**

***Radon* reaches leachate by a gas/air pathway.
Radon in landfill gas exceeds 1 million pCi/liter.**

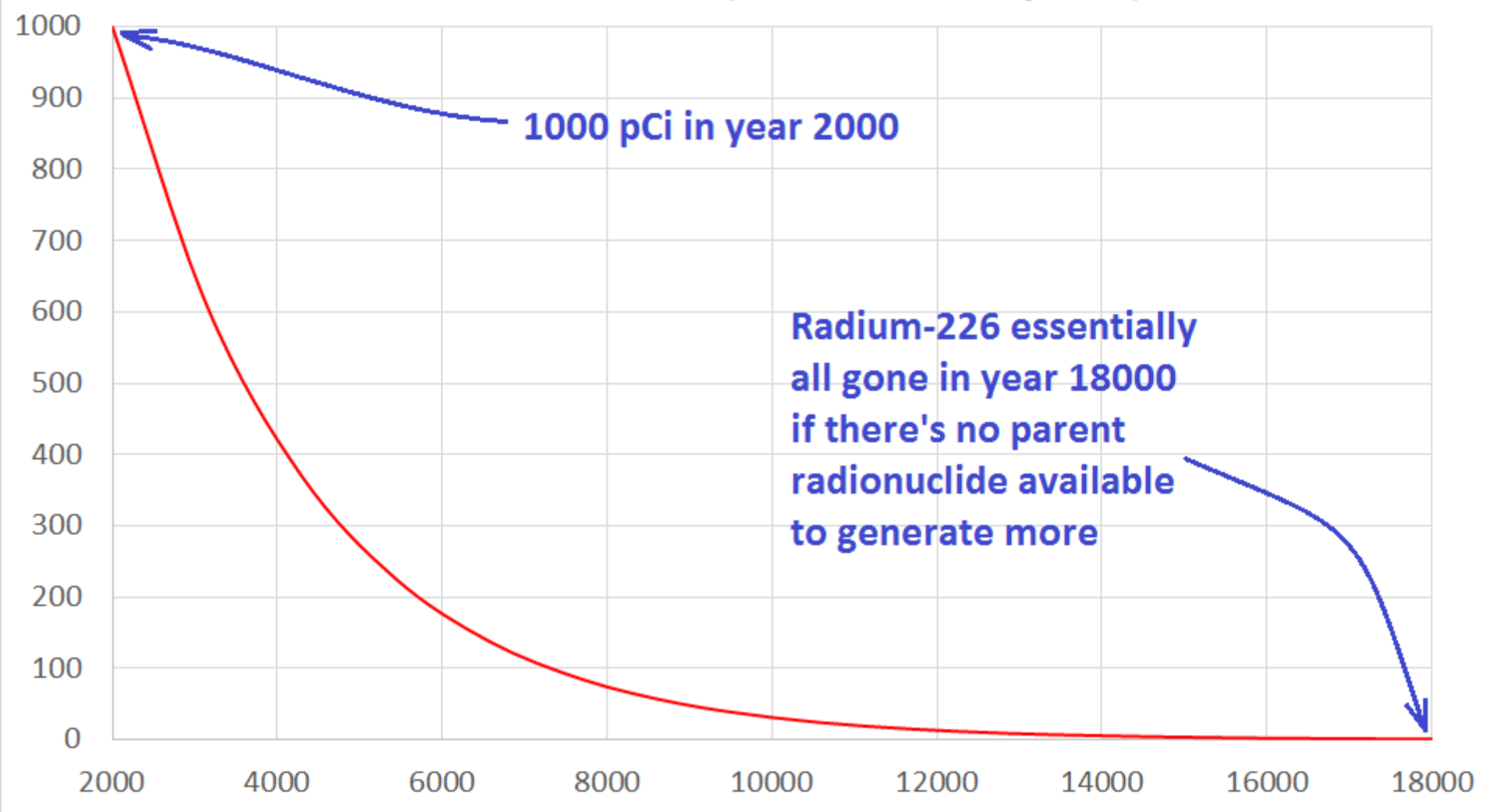


Important points: Landfill gas and Radium

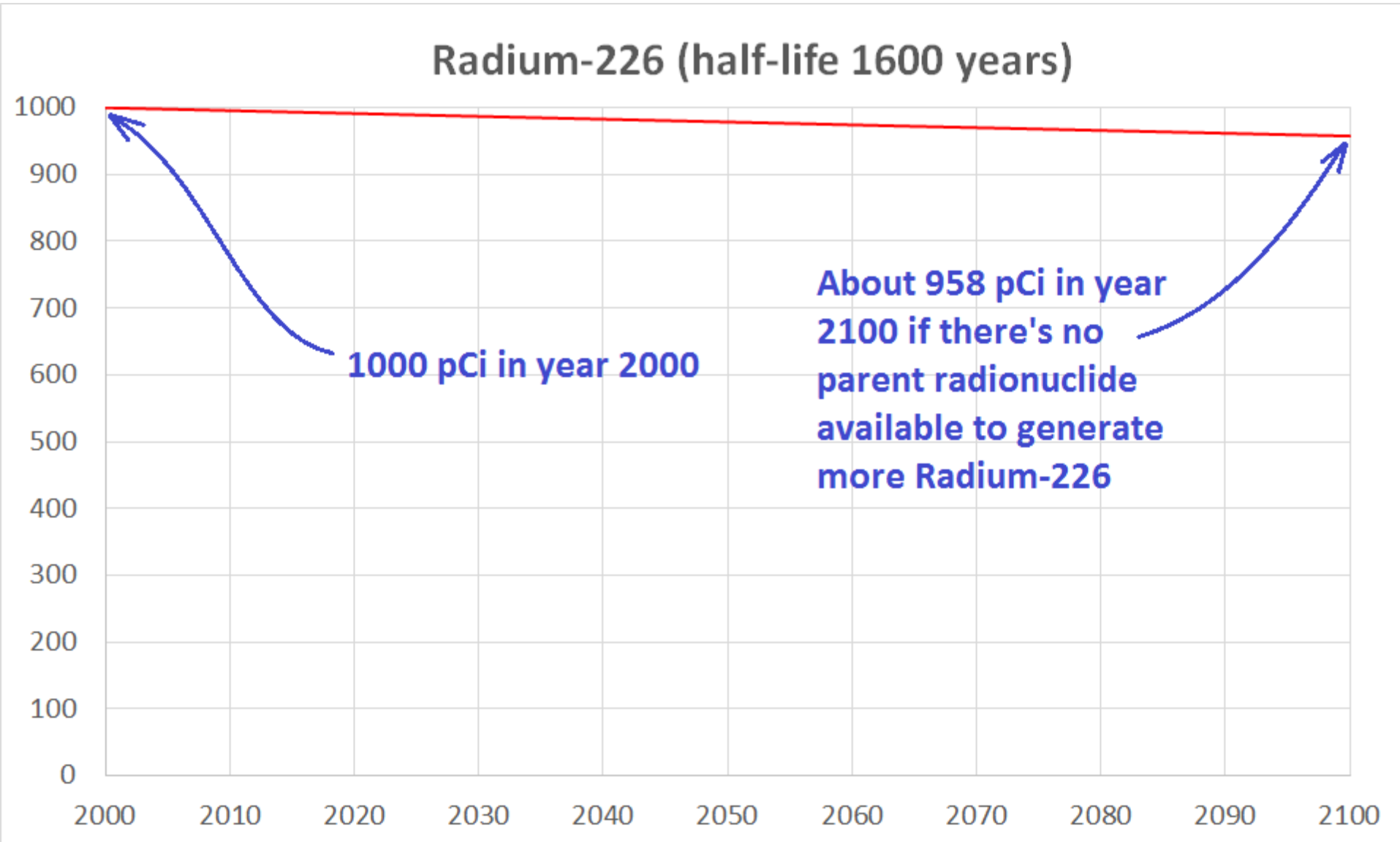
- Radon activity in landfill gas within the landfill likely exceeds ~1 million pCi/L, either most of the time or part of the time. How much escapes through cap? How much is released from the landfill-gas flare? *Testing is needed.*
- Testing and air dispersion modeling are needed to determine radon levels and health risks at downwind locations
- Radon comes from radium decay – *how much radium is in Hakes landfill?*
- Radium is a long-term issue: The “conveyor belts” keep running for thousands of years

Radioactive decay without secular equilibrium with the parent

Radium-226 (half-life 1600 years)



Radioactive decay without secular equilibrium with the parent



Very little info is available on radon in landfill gas or on radon emission rates through landfill caps and vents – but there's a useful landfill modeling study by Walter et al. (2012)* that looks at radon emissions from a modeled landfill that contains more drilling waste and more radium (?) than is present in Hakes landfill:

	Hakes	Walter et al.
Landfill gas emission rate (scfm)**	~250-750	424
Gas development waste (tons)***	~77,000	~165,000
Radium in waste (pCi/g)	<25	50

*G.R. Walter, R.R. Benke, and D.A. Pickett, "Effect of biogas generation on radon emissions from landfills receiving radium-bearing waste from shale gas development," *J. of Air & Waste Management Association* **62**, 1040-1049 (2012).

**Hakes current (~250 scfm) and expanded (~750 scfm) rates from DSEIS, Appendix H, at 7. Walter et al. rate reported as 0.2 std. m³/sec, equivalent to 424 scfm.

***Hakes drilling waste reported by PA DEP for 2009-2019 period. Walter et al. assume 200 x 200 x 2.5 = 100,000 m³ TENORM waste volume with 1500 kg/m³ bulk density.

Radon emission should be *roughly* proportional to radium in landfill waste, all else being equal

That's not the case here where the landfill that nominally accepts less radium (25 pCi/g limit) has far more radon in its landfill-gas emissions.

As already shown, radon activity in landfill gas within Hakes landfill *likely exceeds ~1 million pCi/L*, either most of the time or part of the time.

Landfill-gas emission rate of Walter et al. (about 0.2 m³/sec) combined with their range of radon emission rates (about 6E4 to about 4E6 pCi/sec) indicates *that the landfill gas emerging from their modeled landfill contains radon ranging from about 300 to about 20,000 pCi/L. Big difference!*

Radon emission should be *roughly* proportional to radium in landfill waste, all else being equal

Does radium in Hakes waste exceed 25 pCi/g limit?

Range of ~300 to ~20,000 pCi/L radon in modeled landfill gas may imply ~2500 to ~175,000 pCi/g radium in Hakes waste??

	(At limit) Hakes	(Proportional) Walter et al.	Hakes
Radon in landfill gas (pCi/L)	1 million	300	1 million
Radium in waste (pCi/g)	<25	50	~175,000
Radon in landfill gas (pCi/L)	1 million	20,000	1 million
Radium in waste (pCi/g)	<25	50	~2500

=====

This comparison assumes “all else being equal” (*not likely*) but illustrates the difficulty of reconciling 1 million pCi/L radon in landfill gas with 25 pCi/g radium limit

Important points: Leachate and Landfill gas

- **Radon activity in Hakes leachate from which samples were collected is intermittently very high, ranging up to ~270,000 pCi/liter**
- **Radon activity in landfill gas within the landfill likely exceeds ~1 million pCi/L, either most of the time or part of the time. How much escapes through cap? How much is released from the landfill-gas flare? *Testing is needed.***
- **1 million pCi/L radon in landfill gas cannot be easily reconciled with 25 pCi/g radium limit – indicating far more than 25 pCi/g radium??**
- **Radium is an issue for thousands of years**

Why are Lead-214 and Bismuth-214 *only intermittently high* in leachate test results?

- 1) Some of the test samples were ***apparently not properly sealed***, allowing radon gas to escape from the samples before testing, and/or
- 2) Levels of radon gas contained in the landfill ***vary over time***, depending upon the opening and closing of various pathways that allow radon to reach the leachate and escape to the atmosphere.

But even if radon levels vary over time, remember that the “conveyor belts” are constantly running. Radon always needs a radium source. Radium keeps producing radon and its other progeny.

Many unknowns – testing needed

Radon exposure pathways from Hakes landfill to humans have not been clearly identified or adequately investigated – but cause downwind human exposure. *What exposure and risk?*

Radium poses a long-term risk for thousands of years if landfill integrity can't be guaranteed.

Where is the radium that generates the radon?

What are the levels of radium in the waste?

What long-term exposure and risk?

What are the **landfill-gas flow pathways** through which radon reaches the leachate and escapes to the atmosphere?

Many unknowns – testing needed

Specific testing and modeling needs:

- Radon level (emission rate) at landfill flare
- Modeling of downwind dispersion
- Resumption of gamma spectroscopy testing of Pb-214 and Bi-214 in leachate (EPA Method 901.1). This testing has not been done since mid-2018, so DEC and the public have lost the ability to detect radium decay products in the landfill leachate. This has created a “blind spot” in radiological monitoring.
- Testing/modeling at Chemung Landfill as well as Hakes Landfill

Questions?