SUPREME COURT OF THE STATE OF NEW YORK
COUNTY OF STEUBEN

In the Matter of the Application of

SIERRA CLUB, CONCERNED CITIZENS OF ALLEGANY COUNTY, INC., PEOPLE FOR A HEALTHY ENVIRONMENT, INC., JOHN CULVER, AND BRIAN AND MARYALICE LITTLE,

Petitioners,

For a Judgment Pursuant to Article 78 of the Civil Practice Law and Rules,

–against–

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION, BASIL SEGGOS, COMMISSIONER, TOWN OF CAMPBELL AND HAKES C&D DISPOSAL INC.,

Respondents.

AFFIDAVIT OF KATHRYN BARTHOLOMEW IN SUPPORT OF THE VERIFIED PETITION

Index No.

State of New York,
County of Schuyler, ss.:

KATHRYN BARTHOLOMEW, being duly sworn, deposes and says:

I am the chair of the Atlantic Chapter of the Sierra Club, a petitioner in the above-captioned proceeding.

Sierra Club is a not-for-profit corporation organized under the laws of the State of California. It is the oldest and largest grassroots environmental organization in the country. Sierra Club was founded in 1892. Its purposes include practicing and promoting the responsible use of earth's ecosystems and resources, and protecting and restoring the quality of the natural and human environment. The protection of air, soil and water resources is a key aspect of Sierra Club's work. Sierra Club has more than 803,000 members nationwide, including approximately 54,000 members in New York State, many of whom live in the vicinity of the Hakes C&D Landfill ("Hakes Landfill") or downriver from the Village of Bath Wastewater Treatment Plant ("Bath WWTP"), which takes the discharges from the leachate pre-treatment plant at the Steuben County landfill where all of the leachate from the Hakes landfill is sent. The Bath WWTP discharges into the Cohocton River, which is part of the Chesapeake Bay watershed.

The interests of the Club and its members are injured by allowing the proposed expansion of the operations of the Hakes Landfill which will increase the volumes of radioactive leachate entering the Bath WWTP and being discharged into the Cohocton River, increase the volumes of radon and other harmful substances being emitted into the atmosphere at the landfill and increase the adverse health impacts of the landfill.

As described in the petition, Sierra Club and its members participated at every stage in the proceedings conducted by Respondent New York State Department of Environmental Conservation ("Respondent DEC") and Respondent Town of Campbell ("Respondent Town") related to the preparation and issuance of a Final Supplemental Environmental Impact Statement ("FSEIS").

A copy of the Sierra Club comments on the draft scoping document submitted on May 5, 2017, is attached as Exhibit A.

A copy of the comments submitted on the DSEIS on March 19, 2018, by Petitioners Sierra Club, CCAC and PHE is attached as Exhibit B. The affidavits of Dr. David Carpenter, Dr. Raymond Vaughan and Mr. Dustin May and the additional presentation prepared by Dr. Vaughan were provided as exhibits to the comment letter and are included in Exhibit B.

A copy of the January 14, 2019, letter Sierra Club filed a letter with the Town Planning Board asking them to delay consideration of Respondent HCDD's zoning application until Sierra Club could have Dr. Vaughan respond to the May 2018 report by CoPhysics Corporation is attached as Exhibit C.

A copy of the February 21, 2019, letter from Petitioners Sierra Club, CCAC and PHE to Respondent Town enclosing Dr. Vaughan's February 21, 2019, memorandum discussing the reasons why the FSEIS and the CoPhysics Report do not rebut the evidence presented by Petitioners and their experts is attached as Exhibit D.

A copy of the March 6, 2019, letter from Sierra Club to Respondent Town enclosing Dr. Vaughan's March 5, 2019, memorandum describing radon testing that could be conducted of the Hakes landfill gas system and the protocols that need to be following in continuing to test the Hakes landfill leachate for Lead-214 and Bismuth-214 is attached as Exhibit E.

Kathy Bartholom

Sworn to before me this 9th day of April 2019.

Emily Byers
Notary Public

Emily Byers
Notary Public, State of New York
No. 01BY6329883
Qualified in Schuyler County
Commission Expires August 31, 2019

Exhibit A

SIERRA CLUB
FOUNDED 1892
ATLANTIC CHAPTER

May 5, 2017

VIA EMAIL

Kimberly A. Merchant
Deputy Regional Permit Administrator
Division of Environmental Permits
New York State Department of Environmental Conservation, Region 8
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kimberly.merchant@dec.ny.gov

Re: Comments on the Draft Scoping Outline for the Draft Supplemental Environmental Impact Statement (DSEIS) for the proposed expansion of the Hakes Construction and Demolition Landfill, Permit No: 8-4630-00010/00001-0, Facility SW # 51D03

Dear Ms. Merchant:

On behalf of our 51,000 members, the Sierra Club Atlantic Chapter respectfully submits the following comments on the draft scoping outline for the draft supplemental environmental impact statement (DSEIS) on the proposed expansion of the construction and demolition landfill owned and operated by Hakes C&D Disposal Inc. and located at 4376 Manning Ridge Road in the Town of Campbell, Steuben County, New York, Permit No: 8-4630-00010/00001-0, Facility SW # 51D03. The proposed project would add approximately 22.1 acres to the existing 57.9 acres of permitted landfill cell area. It would also add a new on-site soil borrow area of approximately 24.0 acres, from which soils would be excavated for landfill construction and operation.

We applaud DEC's decision as lead agency to issue a positive declaration requiring the preparation of a Supplemental Environmental Impact Statement (SEIS) for the proposed expansion and DEC's decision to conduct scoping for the expansion project. We have reviewed the draft scoping outline issued by DEC on April 3, 2017 and we request that the following issues be added for consideration in the DSEIS:

1. **Consideration of the impacts of radioactivity in the shale gas drilling wastes that have already been placed in the Hakes landfill and consideration of the impacts of adding additional shale gas drilling waste to the landfill.**

It is well-documented that Hakes C&D landfill is taking Marcellus shale drilling waste from Pennsylvania.[1] The impacts of this waste on the landfill have been shown in the radium-

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[1] Records collected by the Pennsylvania Department of Environmental Protection ("PADEP") show that the Hakes Landfill has accepted many tons of shale gas drilling waste from gas wells drilled in Pennsylvania. The most recent reports are posted at http://www.depreportingservices.state.pa.us/ReportServer/Pages/ReportViewer.aspx?%2fOil_Gas%2fOil_Gas_Well_Waste. It is possible that the volumes shown in these reports do not represent the total volumes of shale gas drilling wastes sent to the Hakes landfill in a given year.

226 levels in the landfill's quarterly radiological test results,[2] although it is not apparent that DEC is currently requiring radiological testing of the leachate coming from the landfill.

Given the levels of radioactivity that have been present in the leachate from the Hakes landfill, consideration of the impacts of radioactivity in the shale gas drilling wastes that have already been placed in the Hakes landfill and the impacts of adding additional shale gas drilling waste to the landfill must be added to issues to be addressed in the DSEIS. Testing is needed to determine whether water-soluble radioactive isotopes from these wastes such as isotopes of radium are currently present in the leachate produced by the landfill or in streams near the landfill and whether radioactive gases, such as isotopes of radon are being emitted from the landfill. Consideration of the possibility of radioactive leachate, surface and groundwater contamination and radioactive air emissions from the landfill is necessary to ensure the preparation of an adequate DSEIS. Radioactive discharges and emissions could have serious health consequences on those living downstream or downwind from the landfill.

In conducting these tests, it is essential that appropriate testing methods be used to detect radiation. Gamma radiation can be used to measure Ra-226 and Ra-228 in waste samples, but it may take up to 21 days in the laboratory for gamma radiation to emerge, as shale gas drilling wastes emit alpha and beta radiation much more strongly. If appropriate testing methods are not used, waste samples are not correctly analyzed, radiation concentrations in both waste and landfill leachate—and in turn the potential risks posed to health and the environment—may be underestimated.[3]

The data contained in the report commissioned by PADEP on Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in shale gas drillings indicate significant levels of radioactivity are associated with shale gas drilling wastes, including:[4]

- Samples of produced water from unconventional well sites had concentrations of Ra-226 more than 20 times as high and Ra-228 more than three times as high as those from conventional well sites. Samples of horizontal drill cuttings had Ra-226 levels nearly twice as high as samples of vertical cuttings.

- Surface radioactivity on equipment used to handle and store wastewater was measured above safety guidelines; this level could increase as equipment is reused and pose a risk to the surrounding environment.

- Flowback samples had Ra-226 concentrations about 100-5,000 times higher than the EPA drinking water standard for combined radium (551-

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[2] Radium-226 levels as high as 180 pCi/L were reported in Hakes leachate in 2013, as described in *License to Dump: Despite Ban, New York Permits Pennsylvania to Dump Radioative Fracking Waste Inside Our Borders*, Elizabeth Moran, Environmental Advocates, February 2015, footnote 51. *License to Dump* may be downloaded at http://www.eany.org/our-work/reports/license-dump-february-2015.

[3] This important point is made in *Wasting Away: Four states' failure to manage gas and oil field waste from the Marcellus and Utica Shale*, Nadia Steinzor and Bruce Baizel, Earthworks' Oil & Gas Accountability Project, April 2015, p. 24. https://www.earthworksaction.org/library/detail/wasting_away_full_report.

[4] *Id.* at 23, citing *Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) Study Report*, PermaFix Environmental Services for PADEP, 2015.

25,500 pCi/L); concentrations of Ra-228 were 50-350 times as high (248-1,740 pCi/L).

- Samples of produced water had Ra-226 concentrations 8-5,300 times higher than the EPA drinking water standard for combined radium (40-26,600 pCi/L); Ra-228 concentrations were 5-380 times as high (26-1,900 pCi/L).

- Radiation levels in filter cakes were many times higher than typical background concentrations in soil at publicly owned treatment works (POTWs), centralized/industrial wastewater treatment (CWT) plants, and zero liquid discharge (ZLD) plants that accept Marcellus Shale wastewater.[5]

DEC's own analysis of 13 samples of production brine from 12 gas wells being drilled in the Marcellus Shale in New York show that 11 samples contained levels of radium-226, a derivative of uranium, above the legally allowed amount safe for discharge, which is 60 pCi/L. Seven samples tested above 1,000 pCi/L. Two samples tested at almost 3,000 pCi/L. The DEC's data is reported in Appendix 13 of the revised DSGEIS on hydrofracking, and attached hereto as Exhibit A.

These reports show the importance of addressing radioactivity in discharges and emissions from the Hakes landfill in the DSEIS.

2. A full consideration of the health impacts of living near the Hakes landfill.

The DSEIS must contain a full consideration of the health impacts of living near the landfill and the increased impacts that could result from the proposed expansion in the size of the landfill. There are already anecdotal reports of higher levels of disease and death in the community living near the landfill and a more complete study of the health impacts of living near the landfill is needed. These impacts need to be addressed in the DSGEIS. A number of studies have documented the adverse health impacts of living near waste landfills.

Research published in the *International Journal of Epidemiology* in 2016 showed that health is at risk for those who live within five kilometers of a landfill site. Researchers in Italy evaluated the potential health effects of living near nine different landfills in the Lazio region, and therefore being exposed to air pollutants emitted by the waste treatment plants. 242,409 people were enrolled in the cohort from 1996 to 2008. The results showed a strong association between Hydrogen Sulfide (used as a surrogate for all pollutants co-emitted from the landfills) and deaths caused by lung cancer, as well as deaths and hospitalizations for respiratory diseases. The results were especially prominent in children. The annual average exposure levels of Hydrogen Sulfide was 6.3 ng/m3, compared to people living close to larger landfills in Rome whose levels averaged 45.ng/m3. At the end of the follow-up period there were 18,609 deaths.[6]

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[5] *Id.*

[6] Living near a landfill could damage your health, *Science Daily*, May 24, 2016, https://www.sciencedaily.com/releases/2016/05/160524211817.htm

Dr. David O. Carpenter, director of the Institute for Health and the Environment, at the State University of New York at Albany spoke about his research on landfill toxins and public health at a 2010 program at Hobart and William Smith College in Geneva. As reported in the *Finger Lakes Times*, made the following statements about his research:[7]

> Carpenter said a 1998 series of air sampling of 25 landfills in New York found high concentrations of carcinogenic chemicals that contributed not only to cancer but to neurological and liver diseases as well.
>
> He said data was collected for 10 years on exposure to these chemicals from breathing the air, having it come into contact with skin, eating food grown nearby or drinking groundwater.
>
> "Statistics from reports of illnesses contracted by people living in certain zip codes shows those near landfills have higher birth defects, thyroid disorders, nervous system disorders, immune system diseases and cancer," Carpenter said.
>
> He also said some studies have shown higher levels of hypertension, heart disease, diabetes and cardio-pulmonary disease.
>
> "The facts show you are at higher risk of these diseases if you live near a landfill," Carpenter said.
>
> Later during the panel discussion, he told a questioner that three miles is considered close enough to a landfill to possibly be impacted by the toxins.
>
> "There is reason to be concerned. It's pretty clear that if you live near a waste site, you have increased exposure to toxins and toxins increase your risk of disease," Carpenter said.[8]

These reports show the importance of addressing the health impacts of living near the Hakes landfill in the DSEIS.

* * *

For these reasons, we respectfully request that the draft supplemental environmental impact statement (DSEIS) radiation issues and the health impacts of living near the landfill.

Respectfully,

Roger Downs

Roger Downs
Conservation Director
Sierra Club Atlantic Chapter

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[7] Panelists air landfill concerns: Speaking at HWS, they say health risks to those living nearby are clear, David Shaw, *Finger Lakes Times*, November 9, 2010, http://www.fltimes.com/news/panelists-air-landfill-concerns/article_cce461a9-ec14-5c88-85f2-f6feacb25797.html

[8] Id.

Exhibit B

SIERRA CLUB
ATLANTIC CHAPTER

744 Broadway ● Albany, NY 12207

March 19, 2018

VIA EMAIL

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Re: Comments on the Draft Supplemental Environmental Impact Statement for the proposed expansion of the Hakes Construction and Demolition Landfill

Dear Ms. Merchant and Town of Campbell Planning Board:

On behalf of its 54,000 members, including 230 members in Steuben County, the Sierra Club Atlantic Chapter, together with Concerned Citizens of Allegany County, Inc. and People for a Healthy Environment, Inc. respectfully submit the following comments on the draft supplemental environmental impact statement (DSEIS) on the proposed expansion of the construction and demolition landfill owned and operated by Hakes C&D Disposal Inc. and located at 4376 Manning Ridge Road in the Town of Campbell, Steuben County, New York.

We request that the New York State Department of Environmental Conservation (DEC) and the Town of Campbell Planning Board deny all permit applications made by Hakes C&D Disposal Inc. in connection with the Hakes expansion project. Because the DSEIS is not an adequate review of the impacts of the expansion project in accordance with the requirements of the State Environmental Quality Review Act ("SEQRA"), the permit applications under review must be denied. The DSEIS fails to comply with the requirements of SEQRA because it fails to take a "hard look" at the issue of radioactivity in the landfill or the issue of the health impacts of the landfill or to provide a reasoned elaboration for why increasing the capacity of the landfill will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

Among the specific issues not identified or not adequately addressed in the DSEIS are the following:

1. The DSEIS fails to evaluate the high levels of radioactivity shown in the landfill's leachate test results

The DSEIS asserts that "at no time have any levels" of radioactivity in Hakes leachate "been detected that would indicate any radioactivity beyond those associated with background levels."[1] This statement is contradicted by the landfill's own leachate test results which show high levels of the radionuclides Lead-214 and Bismuth-214 in some of the test samples. The presence of high levels of these short-lived radionuclides in some of the leachate samples demonstrates that high levels of radium and radon are present in the landfill.

The significance of the presence of Lead-214 and Bismuth-214 in the leachate test results is discussed in the affidavits of Dr. Raymond Vaughan, Mr. Dustin May and Dr. David Carpenter, copies of which are attached as Exhibits 1, 2 and 3. These affidavits were originally filed in the case of *Sierra Club v. DEC* in Steuben County Supreme Court on January 19, 2018.[2]

As discussed in the affidavits, the highest levels of lead-214 and bismuth-214 concentrations are shown in samples from landfill cells 3, 4, 5, 6, and 8B, all cells receiving shale gas drilling wastes. The highest observed Lead-214 and Bismuth-214 concentration was approximately 6000 pCi/L from an unfiltered leachate sample taken from Cell 8B in Q2 2017.

The affidavits explain why the presence of high levels of these short-lived radionuclides in some of the leachate samples demonstrates that high levels of radium and radon are present in the landfill. As described in ¶21 of Mr. May's affidavit:

> Of major concern regarding these results are the concentrations of lead-214 and bismuth-214 found in some of the leachate samples analyzed. In 9 of the 79 leachate samples analyzed from 2012 to 2017, lead-214 and bismuth-214 concentrations exceeded 1000 pCi/L and all of these samples showed good agreement between lead-214 and bismuth 214, indicating that these are unlikely to be false positives as lead-214 decays directly to bismuth-214. These elevated lead-214 and bismuth-214 concentrations were found in samples from Cells 3, 4, 5, 6, and 8B, all cells receiving drilling wastes. The highest observed lead-214 and bismuth-214 concentration was approximately 6000 pCi/L from an unfiltered leachate sample taken from Cell 8B take in Q2 2017. Of the 9

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[1] DSEIS, p. 16.

[2] *Sierra Club v. DEC*, Steuben County Supreme Court, Index No. E2017-1384CV, filed November 30, 2017, stipulation of withdrawal filed February 9, 2018. The petitioners in the case are Sierra Club, Concerned Citizens of Allegany County, People for a Healthy Environment, Inc. and three individual neighbors of the landfill, John Culver and Brian and Maryalice Little. The papers filed in the case are posted at http://treichlerlawoffice.com/waste/hakes/index.html .

> samples found to be extremely high in lead-214 and bismuth-214 concentrations, 6 of these samples were filtered in the laboratory prior to analysis. In a number of other samples, a substantial disequilibrium was observed, with the concentration of lead-214 and bismuth-214 greatly exceeding the concentration of radium-226 measured in the samples via EPA 903.1. These results indicate major potential enrichment of leachate with radon-222 gas; the half-lives of lead-214 and bismuth-214, 27 and 20 minutes, respectively, are too short for these radionuclides to exist independently during the time period between collection and analysis, they would have decayed away entirely. Thus, in order for these two radionuclides to be detectable in the samples weeks after collection, they would have to be supported and exist in an equilibrium state with radon-222 gas or radium-226.[3]

Mr. May explains that the variances in the levels of Lead-214 and Bismuth-214 in the test samples indicate that radon gas has escaped from some of the test samples during sample collection and handling of those samples. Mr. May states, "Variability in the measured concentrations between the filtered and unfiltered samples taken at the same time, with one analysis showing extremely high concentrations of lead-214 and bismuth-214 and the other showing much less or no observable lead-214 and bismuth-214, indicates the likely escape of radon-222 gas from the sample."[4]

Dr. Vaughan summarized the significance of the levels of radioactivity in the leachate test results at a presentation at the Campbell high school on February 10, 2018. The slides for Dr. Vaughan's presentation are attached as Exhibit 4. The video of Dr. Vaughan's presentation is posted at https://www.youtube.com/watch?v=8YuC6cHWd3A. In the presentation, Dr. Vaughan explains the scientific principles that show that, in a leachate test sample that contained 6000 pCi/liter of Lead-214 and Bismuth-214 and very little Radium-226 (<10 pCi/liter) 21days after sample collection, there would have had to have been 270,000 pCi/liter of Radon-222 in the sample – and thus in the sampled leachate – at the time of collection.[5] He explains that to have 270,000 pCi/liter of Radon-222 dissolved in the landfill leachate at the time of collection there would have had to have been approximately 1.05 million pCi/liter of Radon-222 in the air of the landfill.[6] Dr. Vaughan points out that the air in the landfill would contain *more* than 1.05 million pCi/liter of Radon-222 if its parent radium remained dry in the landfill, consistent with intended landfill practice, and if the Radon-222 reached the leachate by an air pathway inside the landfill.[7]

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[3] Ex. 2, ¶ 21.

[4] *Id*. ¶ 23-25.

[5] Ex. 4, Presentation on Unresolved Issues for Disposal of Radium-bearing Wastes at Hakes Landfill by Dr. Raymond Vaughan, February 10, 2018, slide 28.

[6] *Id*., slide 30.

[7] See video of Vaughan presentation at https://www.youtube.com/watch?v=8YuC6cHWd3A.

Alternatively, he explained, the air in the landfill could contain less than 1.05 million pCi/liter of Radon-222 if its parent radium remained wet in the landfill, contrary to intended landfill practice, and if the Radon-222 reached the leachate by a water pathway inside the landfill.[8] As he notes, these values apply at a temperature of 20ºC and would be somewhat different at other temperatures.[9]

We are concerned that the report on Hakes' 2017 3rd Quarter Radionuclide Monitoring Results states that "the laboratory indicated that Bismuth-214 and Lead-214 are no longer include [sic] in the gamma list due to the fact that the any [sic] positive results are actually lab created as a result of the ingrowth step in the method."[10] For the reasons stated above and explained in detail in the expert affidavits of Dr. Vaughan, Mr. May and Dr. Carpenter, the Bismuth-214 and Lead-214 results in the leachate test samples are extremely significant information. It would further undermine the ability of DEC and the public to understand and monitor the radionuclides in the landfill if DEC were to allow the landfill to stop collecting this data. As discussed below, the landfill needs to begin testing for a greater number of radionuclides, not eliminate testing of the very radionuclides that have been observed in high levels in the leachate.

The DSEIS fails to address the likely source of the radioactivity demonstrated in the landfill's leachate test results, i.e., the Marcellus shale drill cuttings that have been accepted at the landfill since 2010. According to a 2017 report by Environmental Advocates summarizing data collected by the Pennsylvania Department of Environmental Protection ("PADEP"), the Hakes landfill has taken more tons of solid gas drilling wastes from Pennsylvania than any other landfill in New York other than the Chemung County landfill.[11] According to the EA report, Hakes landfill began accepting Marcellus shale gas drilling wastes from Pennsylvania in 2010. Some of the landfill's annual reports indicate that as much as 45% of the wastes deposited in certain years may have been gas drilling wastes from Pennsylvania.[12]

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[8] *Id.*

[9] *Id.*

[10] 2017 3rd Quarter Radionuclide Monitoring Results, Hakes C&D, Painted Post N.Y., Hakes C&D Landfill Permit No. 8-4630-00010/00001-0, November 14, 2017, released by DEC on February 23, 2018, pursuant to a FOIL request from the Concerned Citizens of Allegany County.

[11] *License to Dump: Addendum*, Environmental Advocates, June 9, 2017, http://www.eany.org/sites/default/files/documents/license_to_dump_addendum_0.pdf , supplementing *License to Dump: Despite Ban New York Permits Pennsylvania to Dump Radioactive Fracking Waste Inside Our Borders*, Environmental Advocates, February 2015, http://www.eany.org/our-work/reports/license-dump-february-2015 . According to the Addendum, Hakes has taken 167,238 tons and 332 barrels of fracking waste between 2010 to 2017.

[12] Hakes' 2011 Annual Report shows that total tonnage of wastes received at the Hakes landfill in 2011 was 376,485.60 tons. Of this amount, the report says 89,837.42 tons came from Bradford County Pennsylvania and 81,121.57 tons came from Tioga County, PA. The combined tonnage from these two counties was 170,958.99 tons, or 45% of the total tonnage received in 2011. The types of wastes received from these counties are not

It has long been recognized that the Marcellus shale contains radioactive materials and that these may have harmful health effects. In its review of DEC's Supplemental Generic Environmental Impact Statement (SGEIS) on high volume hydraulic fracturing, the United States Environmental Protection Agency (EPA) expressed concern about the lack of analysis of radon and other radiation exposure from shale gas drilling wastes. "Who is responsible for addressing the potential health and safety issues and associated monitoring related to external radiation and the inhalation of radon and its decay products?" the EPA asked. "Such potential concerns need to be addressed."[13]

Recent scientific studies raise new questions about the environmental mobility of radionuclides in Marcellus shale drill cuttings deposited in landfills. A team of University of Iowa researchers, including Dustin May whose affidavit is attached as Exhibit 2, released a study in November 2016 which evaluated radioactive materials—uranium, thorium, radium, lead, and polonium isotopes—from drill cutting samples extracted from a single well drilled in northern Pennsylvania. [14] A copy of this study is attached as Exhibit A to Mr. May's affidavit. The Iowa research team found complex patterns of vertical stratification. For example, the deep drill cuttings had significantly more uranium (U) than the cuttings removed from shallow portions of the well. Noting that virtually all drill cutting waste from the Marcellus Shale is deposited in landfills, the authors examined the stability of the various radioactive materials by simulating different conditions of landfill leaching. The results suggest some environmental mobility of radionuclides in drill cuttings. In particular, as acidity increased, radionuclide leaching increased, with Uranium-238 and Uranium-234 being the most leachable radionuclides. The authors conclude, "Although previous studies have suggested that [radioactive materials] in drill cuttings pose a minimal health risk to the general public when deposited in landfills, our results indicate that Marcellus Shale drill cuttings warrant further radiochemical investigation."[15]

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broken down in the annual report, but it is likely that a substantial portion of these wastes were shale gas drilling wastes.

[13] Environmental Protection Agency. (January 11, 2012). EPA comments on revised draft NYSDEC revised dSGEIS for horizontal drilling and high-volume hydraulic fracturing to develop the Marcellus shale and other low-permeability gas reservoirs [Press release]. Retrieved from http://www.epa.gov/region2/newsevents/pdf/EPA%20R2%20Comments%20Revised%20dSGEIS%20Enclosure.pdf .

[14] Eitrheim, E. S., May, D., Forbes, T. Z., & Nelson, A. W. (2016). Disequilibrium of naturally occurring radioactive materials (NORM) in drill cuttings from a horizontal drilling operation. *Environmental Science & Technology Letters* 3, 425-29. doi: 10.1021/acs.estlett.6b00439, described in *Compendium of Scientific, Medical, and Media Findings Demonstrating Risks and Harms of Fracking (Unconventional Gas and Oil Extraction),* Fifth Edition, Concerned Health Professionals of New York and Physicians for Social Responsibility, March 2018 (*Compendium*), p. 93, http://concernedhealthny.org/wp-content/uploads/2018/03/Fracking_Science_Compendium_5FINAL.pdf .

[15] *Id*., p. 428.

In a 2015 study, the same team from the University of Iowa determined that previous testing and study methods likely underestimate radioactivity by focusing only on radium. The researchers developed a new method to accurately predict the concentrations of uranium, thorium, and radium and their alpha-emitting progeny, polonium and lead, in fracking wastewater. They found that, under certain conditions, radioactivity increased over time, due to ingrowth of alpha-emitting radioactive progeny of long-lived parent radionuclides such as radium.[16] The authors warn that these decay products may potentially contaminate recreational, agricultural, and residential areas, and that a more detailed understanding is needed of how radionuclides accumulate in higher organisms.[17]

Because the DSEIS does not discuss the significance of the high levels of the radionuclides Lead-214 and Bismuth-214 in some of the Hakes landfill's leachate test samples or consider the ways in which the testing methodologies used by the labs testing Hakes leachate for radionuclides may have failed to detect radium or allowed radon to escape from the samples, the DSEIS fails to take a "hard look" at the issue of radioactivity in the landfill. The DSEIS also fails to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drilling wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

2. The DSEIS fails to evaluate the adequacy of the landfill's entrance monitors

The DSEIS states, "[a]t no time have any drill cuttings or other wastes from the oil and gas extraction industry set off the detector alarms at the Hakes Landfill. However, the alarms have proven to be effective in detecting several loads of solid waste that did not contain drill cuttings or other wastes from the oil and gas extraction industry, but potentially did contain radioactive wastes. This demonstrates the efficacy of the detection equipment."[18]

However, the efficacy of the landfill's entrance monitors in detecting radioactive waste entering the landfill is discredited by the landfill's leachate radionuclide test results which show that there are significant levels of radium and radon in the landfill.

Given the discrepancy between the landfill's leachate test results and the failure of any of the loads of waste entering the landfill or loads of leachate exiting the landfill to trigger the landfill's entrance monitors, the adequacy of the entrance monitors should have been evaluated in the DSEIS. The DSEIS should have discussed the correlation

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[16] Nelson, A. W., Eitrheim, E. S., Knight, A. W., May, D., Mehrhoff, M. A., Shannon, R., Schultz, M.K. (2015). Understanding the radioactive in growth and decay of naturally occurring radioactive materials in the environment: An analysis of produced fluids from the Marcellus Shale. *Environmental Health Perspectives*, 123(7). doi: 10.1289/ehp.1408855, discussed in *Compendium*, pp. 95-96.

[17] *Id*.

[18] DSEIS pp. 16-17.

reports required by the radiation monitoring protocol in the landfill's operations and maintenance manual (OMM) and what the correlation reports show regarding the ability of the entrance monitors to detect radioactive wastes entering the landfill.[19]

Dr. Vaughan's affidavit addresses problems with the type of entrance monitors used by the landfill. He points out that truckloads carrying identical Radium-226 concentrations can have widely variable levels of gamma radioactivity measurable at the landfill entrance depending on whether radium breakdown products have been allowed to escape from the load or not. He explains that the gamma radiation dose measured outside a truck carrying Lead-214 and Bismuth-214 in secular equilibrium with Radium-226 because the truck is sealed would be about 60 times greater, or almost two orders of magnitude greater, than the gamma radioactivity measured outside an unsealed truck carrying the same amount of Radium-226, from which all breakdown products such as Lead-214 and Bismuth-214 have escaped. Dr. Vaughan states that this renders any "correlation" between the truckload sample analysis and the gamma radioactivity measurements meaningless unless the monitoring procedure at the landfill gate can quantify and control for the concentrations of Lead-214 and Bismuth-214 in the load of waste at the time the truck enters the landfill gate.[20]

Because the DSEIS does not address the adequacy of the landfill's entrance monitors in light of the landfill's leachate radionuclide test results, the DSEIS fails to take a "hard look" at the issue of the adequacy of the landfill's entrance monitors or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take additional truckloads of radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

3. The DSEIS fails to evaluate the presence of radon gas in the landfill's air emissions, gas collection system emissions, and emissions from flaring

The Air and Particulate Items Report and Monitoring Plan, in Appendix H of the DSEIS, shows that the landfill is not testing for emissions of radon gas from the landfill surface or from the landfill's internal gas collection system. The DSEIS assumes that flaring of landfill gases will solve problems with harmful landfill gases, but radon is not flammable and will be collected and released through the landfill's gas collection system

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[19] Page 23 of the OMM provides that "In order to demonstrate a correlation between kcps and pCi/g, the facility will obtain six (6) samples from waste entering the landfill. Three (3) samples will represent loads that have no elevated radiation levels associated with them. The remaining samples will be taken from loads that trigger an investigation level condition. Radiation monitoring information will be obtained for each sample at the time of sampling. The samples will then be sent for radiological analysis by an appropriately certified laboratory. The sample results will be compared to the data collected on-site to further calibrate the monitors. In the event that no loads trigger an investigative level condition, other Department approved methods may be used to demonstrate a correlation."

[20] Ex. 1, ¶¶ 20-22.

during flaring of other landfill gases. This concern is accentuated by the landfill leachate radionuclide test results which indicate that, at least at certain times, there is approximately 1.05 million pCi/liter of Radon-222 in the air within the landfill, as described above.

Because the DSEIS does not address the adequacy of the landfill's air monitoring program in light of the landfill's leachate radionuclide test results, the DSEIS fails to take a "hard look" at the issue of radon gas in the landfill or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

4. The DSEIS fails to evaluate the possible presence of radium, radon and their breakdown products in the landfill's stormwater discharges, groundwater suppression system discharges or liner leakage discharges

The positive declaration issued by DEC for the landfill expansion project states that "[t]he project has the potential for significant impacts to groundwater requiring the design and construction of a landfill liner and leachate collection and leak detection systems. The construction and placement of waste in proximity of the existing water table requires the design and installation of a groundwater suppression system."[21]

DSEIS describes the landfill's groundwater and surface water monitoring program and states that the monitoring program provides the capability of detecting potential impacts to groundwater and surface water quality before they can have a significant adverse impact on the environment. But nowhere in the DSEIS or in the landfill's environmental monitoring protocols is it indicated that testing is conducted for the presence of radium, radon or their breakdown products in surface water and groundwater near the landfill. The DSEIS states that "To date, no groundwater contamination has been detected related to the operation of the lined cells."[22] This statement overlooks the fact that the landfill is not testing for radium, radon and their breakdown products.

Because the DSEIS does not address the adequacy of the landfill's ground and surface water monitoring programs in light of the landfill's leachate radionuclide test results, the DSEIS fails to take a "hard look" at the issue of radium, radon and their breakdown products entering the environment through the landfill's stormwater discharges, groundwater suppression system discharges or liner leakage discharges, or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse

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[21] DSEIS, App. A-1, p. 2.

[22] DSEIS, p. 14.

effect on the environment and the health and safety of the people, animals and plants living near the landfill.

5. The DSEIS fails to evaluate the adequacy of the landfill's liner system and groundwater suppression system to protect against the radium, radon and their breakdown products present in the landfill from entering groundwater and surface water supplies adjoining the landfill

The DSEIS states that the landfill's composite liner system is the main source of protection against groundwater contamination by the landfill. The liner system is described in the DSEIS, but its annual leakage rate is not mentioned. It is widely recognized that every liner system has a leakage rate. It is also well understood that the liner systems used in construction and demolition landfills are much less substantial than the liner systems used in landfills accepting low-level radioactive wastes.

The DSEIS fails to evaluate the expected life of the landfill's liner system and the risks of harmful exposures from failure of the liner. There is no risk analysis and evaluation of the types of environmental hazards the liner system can withstand or the circumstances in which the integrity of the liner system could fail.

The surface water and wetland studies attached as Appendix E and Appendix J to the DSEIS make it apparent that numerous bodies of water and wetlands surround the landfill. As mentioned in the DSEIS, the drainage from landfill is to the Corning aquifer, the primary drinking water supply for the Corning-Painted Post metropolitan area, which is only three miles from the landfill.[23] All of these water bodies would be contaminated if there is leakage from the landfill and the DSEIS should have evaluated the adequacy of the landfill's liner system to protect against the radium, radon and their breakdown products present in the landfill from entering groundwater and surface water supplies adjoining the landfill.

The DSEIS states that Hakes is requesting a variance from the requirement in the solid waste regulations that the base of a landfill disposal cell be at least 10 feet above underlying bedrock.[24] The DSEIS relies upon the effectiveness of a yet-to-be-constructed groundwater suppression system in support of the landfill's request that only a five foot of separation from bedrock be allowed. There is no risk analysis and evaluation of the types of environmental hazards the groundwater suppression system can withstand or the circumstances in which the groundwater suppression system could fail in the DSEIS.

If the liner system or the groundwater suppression system were to fail, those failures might allow radium, radon and their breakdown products trapped in the landfill

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[23] DSEIS, p. 20, Fig. 3-2.

[24] DSEIS pp. 21-23, 26, 70-71, citing 6 NYCRR Part 363-6.4.

to be released into surrounding water bodies and the environment and these risks should have been evaluated in the DSEIS.

Because the DSEIS does not address the adequacy of the landfill's liner system and groundwater suppression system to protect against the release of radium, radon and their breakdown products to the environment, the DSEIS fails to take a "hard look" at the issue of the adequacy of the landfill's liner system and groundwater suppression system or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

6. The DSEIS fails to evaluate the risk that opening up the landfill to tie-in the proposed expansion will create new pathways for radon and radium in the landfill to be released to the environment

The positive declaration issued by DEC for the landfill expansion project states that "[t]he project includes expansion of a solid waste management facility of large magnitude. It may result in the unearthing of C&D material as the new expansion is tied in with the existing facility. Bulk leachate storage and appropriate ongoing measures to prevent releases will be discussed."[25] But there is no discussion in the DSEIS of the process by which the new expansion is proposed to be tied in with the existing landfill.

Opening up the existing landfill to tie in the new expansion will inevitably create additional pathways for radium, radon and their breakdown products in the landfill to be released into the environment. The DSEIS contains no risk analysis and evaluation of the types of environmental exposures that may be entailed by opening up the landfill for the tie-in process.

Because the DSEIS does not evaluate the risk that opening up the landfill to tie-in the proposed expansion will create new pathways for radon and radium in the landfill to be released to the environment, the DSEIS fails to take a "hard look" at the issue of radioactivity in the landfill or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

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[25] DSEIS, App. A-1, p. 2.

7. The DSEIS fails to evaluate the risk that the fires that have been occurring at the landfill have damaged the landfill's liner system, gas collection system or leachate collection system and have created or will create new pathways for radon and radium in the landfill to be released to the environment

The DSEIS acknowledges that the landfill "has experienced both surface and subsurface fires."[26] The DSEIS does not describe how many fires have occurred, in which cells the fires have occurred, how long the fires have lasted, or whether any fires are still burning within the landfill. The DSEIS does not evaluate whether the fires could damage or have already damaged the landfill liner system, gas collection system or leachate collection system and thereby create new pathways for radon and radium in the landfill to be released to the environment. Other than stating that the fires "could impact air resources by the release of smoke and other combustion products,"[27] the DSEIS contains no risk analysis and evaluation of the types of environmental exposures that could result from damages caused to damage the landfill liner, the gas collection system or the leachate collection system by the landfill fires.

Because the DSEIS does not evaluate the risk that the fires that have been occurring at the landfill have damaged the landfill's liner system, gas collection system or leachate collection system and have created or will create new pathways for radon and radium in the landfill to be released to the environment, the DSEIS fails to take a "hard look" at the risk of radioactivity in the landfill, or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

8. The DSEIS fails to evaluate the health impacts of the landfill expansion project

Although the DSEIS states that "[a] major public concern regarding the construction or expansion of any solid waste facility is potential adverse impact on human health and the environment,"[28] the DSEIS fails to provide any meaningful analysis of the potential adverse impact on human health and the environment of the landfill expansion project.

A number of studies have documented the adverse health impacts of living near waste landfills. Research published in the *International Journal of Epidemiology* in 2016 showed that health is at risk for those who live within five kilometres of a landfill site. Researchers in Italy evaluated the potential health effects of living near nine different landfills in the Lazio region, and therefore being exposed to air pollutants emitted by the

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[26] DSEIS, p. 17.

[27] *Id*.

[28] Id., p. xv.

waste treatment plants. 242,409 people were enrolled in the cohort from 1996 to 2008. The results showed a strong association between Hydrogen Sulphide (used as a surrogate for all pollutants co-emitted from the landfills) and deaths caused by lung cancer, as well as deaths and hospitalizations for respiratory diseases. The results were especially prominent in children. The annual average exposure levels of Hydrogen Sulphide was 6.3 ng/m3, compared to people living close to larger landfills in Rome whose levels averaged 45.ng/m3. At the end of the follow-up period there were 18,609 deaths. [29]

Dr. David O. Carpenter, director of the Institute for Health and the Environment, at the State University of New York at Albany spoke about his research on landfill toxins and public health at a 2010 program at Hobart and William Smith College in Geneva. An article in the *Finger Lakes Times*, described his presentation as follows: [30]

> Carpenter said a 1998 series of air sampling of 25 landfills in New York found high concentrations of carcinogenic chemicals that contributed not only to cancer but to neurological and liver diseases as well.
>
> He said data was collected for 10 years on exposure to these chemicals from breathing the air, having it come into contact with skin, eating food grown nearby or drinking groundwater.
>
> "Statistics from reports of illnesses contracted by people living in certain zip codes shows those near landfills have higher birth defects, thyroid disorders, nervous system disorders, immune system diseases and cancer," Carpenter said.
>
> He also said some studies have shown higher levels of hypertension, heart disease, diabetes and cardio-pulmonary disease.
>
> "The facts show you are at higher risk of these diseases if you live near a landfill," Carpenter said.
>
> Later during the panel discussion, he told a questioner that three miles is considered close enough to a landfill to possibly be impacted by the toxins.

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[29] Living near a landfill could damage your health, *Science Daily*, May 24, 2016, https://www.sciencedaily.com/releases/2016/05/160524211817.htm

[30] Panelists air landfill concerns: Speaking at HWS, they say health risks to those living nearby are clear, David Shaw, *Finger Lakes Times*, November 9, 2010, http://www.fltimes.com/news/panelists-air-landfill-concerns/article_cce461a9-ec14-5c88-85f2-f6feacb25797.html

> "There is reason to be concerned. It's pretty clear that if you live near a waste site, you have increased exposure to toxins and toxins increase your risk of disease," Carpenter said.[31]

A key component of a health impact analysis is the study of the health impacts experienced by landfill workers and people living near the landfill, but no such studies are included in the DSEIS. The DSEIS excuses the failure to conduct studies of the health risks of working at the landfill or living near the landfill by stating that, "No specific health studies were included in the Consolidated Scope (Appendix B) prepared for this DSEIS, so none were performed."[32] The DSEIS also states that, "[t]he absence of specific health studies for the Town of Campbell or the landfill area makes it difficult to assess existing health conditions in the vicinity of the Hakes facility."

While it is the case that the scope states that, "[the health issues section of the DSEIS] will be limited to regulatory requirements put in place by state and federal regulations to protect human health, and how the applicant will meet these requirements for the proposed expansion. This will include a discussion of the various air emission and water discharge limits and associated standards that are applicable to a facility of this type and how they were established to be protective of human health,"[33] the DSEIS does not discuss the various air emission and water discharge limits and associated standards that are applicable to the Hakes landfill and how they were established to be protective of human health.

In particular, the DSEIS fails to evaluate the risks of injury to human and environmental health from exposure to the levels of radioactivity shown by the leachate test data to be already present in the landfill or to evaluate the risks of the additional environmental exposures that would result from accepting additional levels of radioactive waste in the landfill.

Given the levels of radionuclides that are present in the landfill's leachate test data, additional tests should have been conducted for the presence of radionuclides in the landfill to try and determine the exposure levels that may be experienced by landfill workers and people living near the landfill now and in the future.

An adequate health impact analysis must look beyond the current health impacts of working at the landfill and living near the landfill and take into account the long-term health and environmental impacts of radium in the landfill. Radium-226 has a half-life of about 1,600 years and bioaccumulates in the food chain. The DSEIS should evaluate the projected health and environmental impacts of radium in the Hakes C&D landfill over the long-term.

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[31] *Id.*

[32] *Id.*, p.63.

[33] DSEIS, App. B, p.35.

New York's low-level radioactive waste disposal facility regulations provide that "[c]oncentrations of radioactive material which may be released to the general environment in groundwater, surface water, air, soil, plants or animals must not result in an annual dose exceeding an equivalent of 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public. Releases of radioactivity in effluents to the general environment must be maintained as low as reasonably achievable." 6 NYCRR 382.11. There is no evaluation of potential exposures to radioactivity from radionuclides in the the Hakes landfill in the DSEIS.

Dr. David Carpenter states in his affidavit attached as Exhbit 3, "[b]ased on the information provided to me and my knowledge of the human health effects arising from exposure to ionizing radiation, I have concluded that: (a) there are substantial and significant risks to human health posed by the current procedures used at the Hakes Landfill and approved by NYSDEC, (b) while the greatest threat to human health comes from inhalation of radon-222, other naturally occurring radioactive material (NORM) and the progeny of these elements pose significant threats to human health, and (c) inhalation is the route of exposure of greatest concern but other routes (ingestion, dermal absorption) are also possible.[34]

Dr. Carpenter concludes that "[t]he net effect of New York accepting drill cuttings and de-watered mud from Pennsylvania fracking sites will be the New Yorkers will have an increased risk of cancer, especially lung and gastrointestinal cancers, an increased risk of birth defects coming from DNA damage and increased risk of a shortened life span."[35]

Because the DSEIS makes no attempt to evaluate the health risks of exposure to the levels of radioactivity shown to be present in the landfill or to evaluate the possibility of additional exposures not identified by the testing methodologies used by the landfill's laboratories, it is apparent that the DSEIS fails to take a "hard look" at the health impacts of the landfill expansion or to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drillings wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill now and in the future.

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[34] Ex. 3, ¶ 6.

[35] Id., ¶ 19.

* * *

For each of the reasons discussed above, we respectfully request that the permit applications made in connection with the Hakes expansion project be denied.

Respectfully,

Roger Downs

Roger Downs, Conservation Director
Sierra Club Atlantic Chapter

Frederick Sinclair, Chairman
Concerned Citizens of Allegany County

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Cc:

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TABLE OF EXHIBITS

Exhibit 1 — Affidavit of Dr. Raymond Vaughan, dated January 18, 2018.

Exhibits to Vaughan affidavit may be downloaded from http://treichlerlawoffice.com/waste/hakes/vaughanexhibits.html.

Exhibit A —	Vaughan CV
Exhibit B —	Truck Monitor Correlation Report, Co-Physics, June 7, 2015
Exhibit C —	Hakes 2Q 2012 Leachate Radionuclide Analytical Results
Exhibit D —	Hakes 4Q 2012 Leachate Radionuclide Analytical Results
Exhibit E —	Hakes 2Q 2013 Leachate Radionuclide Analytical Results
Exhibit F —	Hakes 4Q 2013 Leachate Radionuclide Analytical Results
Exhibit G —	Hakes 2Q 2014 Leachate Radionuclide Analytical Results
Exhibit H —	Hakes 4Q 2014 Leachate Radionuclide Analytical Results
Exhibit I —	Hakes 2Q 2015 Leachate Radionuclide Analytical Results
Exhibit J —	Hakes 4Q 2015 Leachate Radionuclide Analytical Results
Exhibit K —	Hakes 2Q 2016 Leachate Radionuclide Analytical Results
Exhibit L —	Hakes 4Q 2016 Leachate Radionuclide Analytical Results
Exhibit M —	Hakes 2Q 2017 Leachate Radionuclide Analytical Results
Exhibit N —	Chemung Leachate Radionuclide Analytical Results March 2015 through January 2017
Exhibit O —	Answer posted 27 September 2005 by George Chabot, PhD, CHP, providing an expert online answer to the question, What value should be taken as the average gamma energy of 226Ra? at the Health Physics Society website, https://hps.org/publicinformation/ate/q4817.html
Exhibit P —	Uranium-238 decay series
Exhibit Q —	Oak Ridge Institute for Science and Education (ORISE) presentation, Radiological and Chemical Properties of Uranium, available online from the U.S. Nuclear Regulatory Commission website, www.nrc.gov/docs/ML1122/ML11227A233.pdf
Exhibit R —	DEC's September 18, 2015 Program Policy Memorandum on Recommended Permit Modifications and Operating Procedures for Landfills relating to Wastes from Drilling in the Marcellus Shale Formation
Exhibit S —	Graph of time trends for Hakes leachate test results for Bismuth-214
Exhibit T —	Graph of time trends for Hakes leachate test results for Lead-214
Exhibit U —	Graph of time trends for Hakes leachate test results for Radium-226 (tested by Method 901.1)
Exhibit V —	Graph of time trends for Hakes leachate test results for Radium-226 (tested by Method 903.1)
Exhibit W —	Graph of time trends for Chemung leachate test results for Bismuth-214
Exhibit X —	Graph of time trends for Chemung leachate test results for Lead-214
Exhibit Y —	Graph of time trends for Chemung leachate test results for Radium-226 (tested by Method 901.1)

Exhibit Z — Graph of time trends for Chemung leachate test results for Radium-226 (tested by Method 903.1))

Exhibit 2 — Affidavit of Mr. Dustin May, dated January 17, 2018.

Exhibit A — Eitrheim, E. S., May, D., Forbes, T. Z., & Nelson, A. W. (2016). Disequilibrium of naturally occurring radioactive materials (NORM) in drill cuttings from a horizontal drilling operation, Environmental Science & Technology Letters 3, 425-29. doi: 10.1021/acs.estlett.6b00439

Exhibit 3 — Affidavit of Dr. David Carpenter, dated January 17, 2018.

Exhibit 4 — Unresolved Issues for Disposal of Radium-bearing Wastes at Hakes Landfill, slides of presentation by Dr. Raymond C. Vaughan, Sierra Club/CCAC Public Meeting, Campbell, NY, February 10, 2018. Click here for the video of the presentation.

EXHIBIT 1

Affidavit of Dr. Raymond Vaughan

January 18, 2018

SUPREME COURT OF THE STATE OF NEW YORK
COUNTY OF STEUBEN

In the Matter of the Application of

SIERRA CLUB, CONCERNED CITIZENS OF ALLEGANY COUNTY, PEOPLE FOR A HEALTHY ENVIRONMENT, INC., JOHN E. CULVER, AND BRIAN AND MARYALICE LITTLE,

Petitioners,

For a Judgment Pursuant to Article 78 of the Civil Practice Law and Rules,

–against–

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION, BASIL SEGGOS, COMMISSIONER, AND HAKES C&D DISPOSAL INC.,

Respondents.

AFFIDAVIT OF
RAYMOND C. VAUGHAN
IN SUPPORT OF THE
VERIFIED PETITION

Index No. E2017-1384CV

State of New York,
County of Erie, ss.:

RAYMOND C. VAUGHAN, being duly sworn, deposes and says:

1. I am a Professional Geologist (NY license no. 258) and Environmental Scientist with a Ph.D. in Geology from SUNY Buffalo. I am very familiar with radioactive substances or "radionuclides," sometimes also called "radioisotopes." I am familiar with the physical and quantitative properties of these radionuclides and the alpha, beta, and gamma radiation they emit, including quantitative measures such as activity (expressed in Curies, for example), specific activity, and half-life, and also including relationships such as secular equilibrium that may occur when a parent-progeny relationship exists among different radionuclides. Note that a parent-progeny relationship is sometimes called a "parent-daughter" relationship. Note also that the words "activity" and "radioactivity" can be used interchangeably when referring to a quantitative measurement – as expressed, for example, in picocuries per gram (pCi/g) or

picocuries per liter (pCi/L).

2. My familiarity with such properties, measures, and relationships is based partly on work relating to radionuclides that I performed during the twelve years I was employed as an Environmental Scientist at the NYS Attorney General's Office (2000-2012), partly on consulting work done for the Seneca Nation of Indians to review and interpret radiological test results (2016), and partly on technically-oriented volunteer work I have performed for several decades as a member of the Coalition on West Valley Nuclear Wastes (1978-2006) and also as a member of the West Valley Citizen Task Force (1997-present). I was appointed to the West Valley Citizen Task Force (CTF) by the U.S. Department of Energy and the New York State Energy Research and Development Administration (NYSERDA), and on one occasion in 2006 I testified on behalf of the West Valley CTF before the U.S. Nuclear Regulatory Commission in Rockville, MD. I have also spoken on behalf of the West Valley CTF at meetings such as the Council of State Governments/Blue Ribbon Commission public meeting in Boston (2011) and the National Transportation Stakeholders Forum meeting in Buffalo (2013). My CV is attached as Exhibit A.

3. My familiarity with such properties, measures, and relationships is supported in part by my understanding and frequent use of numerical methods and relationships in various fields of science and technology.

Overview of issues and sources

4. I have reviewed reports/information on both Hakes landfill and the Chemung County landfill. The latter is a reasonable proxy for Hakes and is a useful supplement to the limited information that is available for Hakes. In particular I have reviewed:

(a) the current use of radiation monitoring devices to measure and/or detect the radionuclide known as Radium-226 in truckloads of "fracking" waste being delivered to landfill disposal sites, particularly the *Truck Monitor Correlation Report*, CoPhysics Corp., June 7, 2015 (hereinafter "Truck Monitor Correlation Report"), attached as Exhibit B, and

(b) recent measurements of Radium-226 and other radionuclides in leachate from Hakes landfill, particularly the series of reports with the PDF document names *51D03_Hakes_Leachate_Radiological…* for second-quarter (2Q) and fourth-quarter (4Q) testing from 2Q 2012 to 2Q 2017, as submitted to NYSDEC by either Casella Waste Services or On-Site Technical Services, Inc. (hereinafter "Hakes Leachate Test Reports"), attached as Exhibits C-M, and

(c) recent measurements of Radium-226 and other radionuclides in leachate from the Chemung County landfill, particularly *Chemung County Landfill, Elmira, New York – First Quarter 2017 Leachate Radiological Test Results*, On-Site Technical Services, Inc., submitted to NYSDEC April 5, 2017 (hereinafter "Chemung Leachate Test Report"), attached as Exhibit N.

5. Based on my review I conclude that:

a) The current use of radiation monitoring devices to characterize truckloads of "fracking" waste entering landfill disposal sites is not providing a reliable measure of Radium-226. Secular equilibrium between Radium-226 and its progeny (particularly the radionuclides Lead-214 and Bismuth-214) is a crucial factor whose importance has not been sufficiently recognized. This has resulted in substantial uncertainty about the concentrations and quantities of Radium-226 entering landfills such as Hakes and Chemung County.

b) Measurements of leachate from Hakes and Chemung County landfills are not providing a reliable measure of Radium-226 and/or Radon-222. The leachate test reports and their interpretations of the data have failed to assess and determine why Lead-214 and Bismuth-214 test results are orders of magnitude higher than the results for Radium-226.

6. For the current practice of using radiation monitoring devices to characterize truckloads of "fracking" waste, there is no evidence that full secular equilibrium, or a consistent degree of secular equilibrium or disequilibrium, exists between Radium-226 and its progeny such as Lead-214 and Bismuth-214. On the contrary, secular equilibrium is disrupted by escape of the gas-phase radionuclide known as Radon-222, with the loss of radon being highly variable and inconsistent. The effects of this are not acknowledged or addressed. Thus, the Truck Monitor Correlation Report does not provide either a reliable limit or an accurate measure of Radium-226 entering the landfill.

7. For the current practice of measuring radionuclide concentrations in leachate from Hakes and Chemung County landfills, there are two alternative explanations for the mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results. These involve either underreported levels of Radium-226 or high, unreported levels of Radon-222 in the landfill leachate. The latter is more likely, but additional testing is needed to confirm this. Informed decisions could then be made about addressing the high levels of Radon-222 in the leachate and seeking a better understanding of the landfill's inventory of Radium-226 from which the Radon-222 is generated.

Scientific background: Relevant radiological principles

8. A good summary of the relevant science can be found in the following expert explanation which was posted online in response to an online inquiry that was related to, but not

identical to, the points at issue here:

> Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi. These are produced following the decay of 226Ra to 222Rn, which then decays to 214Pb. Since 222Rn is a gas it will escape, to varying degrees, from unsealed sources, and the gamma radiation from the 214Pb and 214Bi may not be significant in such cases. In sealed sources that prevent leakage of 222Rn, the 222Rn, 214Pb, and 214Bi each reach the same activity level as that of the 226Ra within a few weeks of preparation of the source….
>
> (Answer posted 27 September 2005 by George Chabot, PhD, CHP, providing an expert online answer to the question, "What value should be taken as the average gamma energy of 226Ra?" at the Health Physics Society website [https://hps.org/publicinformation/ate/q4817.html]. The web page is attached hereto as Exhibit O.)

Note that the above-quoted explanation by Chabot refers to Radium-226 as 226Ra, Radon-222 as 222Rn, Lead-214 as 214Pb, and Bismuth-214 as 214Bi. He also uses the term "daughter" rather than "progeny."

9. The relevant science and Chabot's explanation are rooted in the fact that Radium-226, Radon-222, Lead-214, and Bismuth-214 are all members of the Uranium-238 decay chain. See Exhibit P, which shows that Uranium-238 decays to Thorium-234, and so on. For the present purpose, the decay chain from Radium-226 onward is most relevant. Radium-226 decays to Radon-222, which in turn decays to Polonium-218, which in turn decays to Lead-214, which in turn decays to Bismuth-214, and so on. Given this relationship, Radium-226 can be called the parent, and Radon-222, Polonium-218, Lead-214, and Bismuth-214 can be called the progeny of Radium-226. In effect, there is a "pipeline" from Radium-226 that delivers the first two generations of progeny (Radon-222 and Polonium-218), from which Lead-214 and Bismuth-214 will in turn be created as the third and fourth generations. However, the "pipeline" can be considered leaky if Radon-222 escapes as a gas, in which case Lead-214 and Bismuth-214 will be created at some downwind location rather than in close proximity to the parent Radium-226. As

stated above by Chabot, “Since 222Rn is a gas it will escape, to varying degrees, from unsealed sources, and the gamma radiation from the 214Pb and 214Bi may not be significant in such cases,” meaning that the gamma radiation from Lead-214 and Bismuth-214 would not contribute significantly in such cases to a gamma radiation measurement of the parent Radium-226. This is important for the reason stated in Chabot’s first sentence: “Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi.”

10. Thus, in measuring gamma radiation from Radium-226, it’s crucial to know whether Lead-214 and Bismuth-214 are present in the sample along with Radium-226. However, even if they’re absent due to a “leaky pipeline,” a sample collected for Radium-226 analysis can be put in a sealed container, thereby allowing “ingrowth” of the progeny. Keeping a sample in a sealed container for about 21 days or more is sufficient to allow ingrowth and thereby to reestablish “secular equilibrium” for the first four generations of progeny, including Radon-222, Polonium-218, Lead-214, and Bismuth-214. As stated by Chabot, “In sealed sources that prevent leakage of 222Rn, the 222Rn, 214Pb, and 214Bi each reach the same activity level as that of the 226Ra within a few weeks of preparation of the source.”

11. This statement by Chabot is illustrated graphically by slide 12 of the Oak Ridge Institute for Science and Education (ORISE) presentation, *Radiological and Chemical Properties of Uranium*, available online from the U.S. Nuclear Regulatory Commission website (www.nrc.gov/docs/ML1122/ML11227A233.pdf), hereinafter “ORISE Presentation,” attached hereto as Exhibit Q.

12. An important aspect of the relevant science is the “half-life” of each radionuclide in the decay chain, as shown in Exhibit P. Compared to its progeny, Radium-226 has a long half-life (1600 years). This is much longer than the half-lives of its progeny, including the first four

generations of progeny that are most relevant here: Radon-222 (3.82 days), Polonium-218 (3.1 minutes), Lead-214 (27 minutes), and Bismuth-214 (20 minutes).

13. For the above reasons, Radium-226 in an unsealed container (such as either an uncapped sample vial or the bed of a waste-hauling truck) may be accompanied by relatively little Lead-214 and relatively little Bismuth-214 due to escape of Radon-222 from the container. The loss of Radon-222 will have interrupted the decay chain or the "pipeline" that generates Lead-214 and Bismuth-214. Under these circumstances, the activity or radioactivity of Lead-214 and Bismith-214 in the unsealed container (measured as pCi/g or pCi/L, for example) will tend to be less than the activity of Radium-226 in the container.

14. For the above reasons, Radium-226 which has remained in a sealed container for about 21 days or more will be in secular equilibrium with the first few generations of its progeny, including Lead-214 and Bismuth-214. This means that the activity of Lead-214 and Bismith-214 in the sealed container (measured as pCi/g or pCi/L, for example) will be approximately the same as the activity of Radium-226 in the container. Having Lead-214 and Bismith-214 present will substantially increase the gamma activity as compared to the gamma activity of Radium-226 alone, as discussed below in more detail. As noted by Chabot, "Most of the significant gamma radiation from 226Ra decay comes from the radioactive progeny 214Pb and its daughter, 214Bi."

15. For the above reasons, the activity or radioactivity of Radium-226 which has remained in a sealed container for about 21 days or more cannot be less than the activity of Lead-214 and Bismith-214 in the container. In effect, the "downstream end of the pipeline" cannot generate Lead-214 and Bismuth-214 at a greater rate than is sustained at the "upstream end" of this imaginary pipeline by the decay of Radium-226.

16. For the above reasons, a measurement that shows essentially equal activities for

Lead-214, Bismuth-214, and Radium-226 in a container that has been sealed for about 21 days or more *cannot show whether Lead-214 and Bismuth-214 were present or essentially absent* at the time when the container was sealed. As described on slide 11 of the ORISE Presentation, "Starting with nothing but the parent, the time to reach secular equilibrium is roughly five to ten half-lives of the daughter." In other words, starting with no radionuclides other than Radium-226 in the container when it was initially sealed, ingrowth of progeny will occur as a result of Radium-226 decay. Secular equilibrium with its progeny Radon-222 will be achieved within about 21 days in the sealed container, and secular equilibrium with the next three generations of progeny will likewise be achieved within the same time period. But it cannot be readily determined, based on Lead-214, Bismuth-214, and Radium-226 activity measurements at the end of 21 days or more, whether Lead-214 and Bismuth-214 were essentially absent or already present at some concentration (ranging up to secular-equilibrium concentration) when the container was initially sealed.

17. The general truths expressed above can be applied to the Truck Monitor Correlation Report, Hakes Leachate Test Reports, and Chemung Leachate Test Report.

Unreliability of monitoring trucks for radium concentration at landfill gate

18. The Truck Monitor Correlation Report purports to establish a numerical correlation (i.e., conversion factor) between the concentration of Radium-226 entering a landfill in a waste truckload and the radiation monitor reading at the landfill gate:

> Gamma radiation detectors are routinely used at landfill weighing scales to determine if entering trucks contain unauthorized radioactive materials. However, readings on the monitors cannot easily be related to the concentration of radioactive materials in loads.... Therefore, an actual in-field correlation test was performed to more accurately relate gamma count rate to radionuclide concentration in a load.... A composite sample of the sludge cake was collected from 4 spots near the center of the load (approximately where the detectors were positioned) and was sent to Pace

> Laboratories (NELAP-certified) for gamma spectroscopic analysis after 21-day radon progeny ingrowth.....
>
> (Truck Monitor Correlation Report at 2)
>
> The result of this test of a 30-yard roll-off, filled to near capacity, resulted in a gamma count rate to radium concentration conversion factor or 0.306 KCPS/(pCi/g) over background. For a monitor with a background of 3.6 KCPS (the background occurring during the most recent calibration of the Chemung County Landfill monitor), the count rale corresponding to a 25 pCi/g radium-226 investigation level would be (0.306 x 25) + 3.6 = 11.25 KCPS. Presently the Chemung County monitor's alarm levels arc 10 KCPS sum alarm (sum of both detectors) and a sigma alarm of 110 which equates to approximately 7 KCPS depending on truck speed entering the detection area.... These alarm settings are well within the 11.25 KCPS level corresponding to 25 pCi/g of radium. Therefore, the present alarm settings at the Chemung County Landfill are sufficient to detect a roll-off containing 25 pCi/g or more of radium-226.
>
> (Id. at 5, where KCPS stands for kilocounts per second.)

19. This purported correlation and conversion factor are unreliable because the concentration of relatively strong gamma emitters (such as Lead-214 and Bismuth-214) in the truckload of waste entering the landfill is highly variable and very uncertain for the reasons described above, and because Radium-226 emits only weak gamma radiation in addition to the alpha particles that it emits. As described in an International Atomic Energy Agency report:

> The determination of 226Ra in environmental solids by gamma spectrometry has long been based on the detection of emissions of the radon progeny (222Rn) nuclides, i.e. 214Pb and 214Bi after an ingrowth period of at least 20 days, during which the sample has been hermetically sealed to ensure secular equilibrium between 226Ra and its progeny....
>
> (International Atomic Energy Agency, "Analytical Methodology for the Determination of Radium Isotopes in Environmental Samples," IAEA/AQ/19 (2010) [http://www-pub.iaea.org/MTCD/Publications/PDF/IAEA-AQ-19_web.pdf]).

Since a waste-hauling truck isn't a container that's been sealed for at least 20 or 21 days but may nevertheless contain some quantity of progeny derived from Radon-222 that didn't escape from the truckload of waste, the use of a gamma monitor to infer Radium-226 concentration in the incoming waste has the potential to be wildly inaccurate and cannot be considered reliable.

20. A relatively tightly packed and/or covered truckload of waste, probably including the sludge cake used for the Truck Monitor Correlation Report, will retain higher concentrations of Radon-222 and other progeny than would be found in a less tightly packed and/or uncovered truckload. See, for example, the description of such ingrowth in *Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) Study Repor*t, Rev. 1, 2016, prepared for Pennsylvania Department of Environmental Protection by Perma-Fix Environmental Services, Inc. (hereinafter “Pennsylvania TENORM Report”), § 5.3. The gamma radioactivity of a relatively tightly packed and/or covered truckload would therefore be relatively high compared to the gamma radioactivity of a loosely packed and/or uncovered truckload carrying the same concentration of Radium-226. If the sludge cake used for the Truck Monitor Correlation Report happened to be relatively tightly packed and/or covered, and thus relatively highly radioactive compared to most other truckloads entering the landfill with the same concentration of Radium-226, this would mean that the report’s correlation (indicating that 11.25 kilocounts per second corresponds to 25 pCi/g Radium-226) is unrepresentative and unprotective, allowing truckloads with much more than 25 pCi/g Radium-226 to enter the landfill without exceeding the 11.25 kilocounts per second investigation level. Since the report neither acknowledges this correlation issue nor describes any steps taken to control for the issue, it is difficult to judge from the reported information how high the Radium-226 concentration could be without exceeding the 11.25 kilocounts per second investigation level. In any case, the correlation procedure described in the Truck Monitor Correlation Report is unsupported and cannot be considered reliable based on the information provided in the report.

21. DEC's September 18, 2015 Program Policy Memorandum on "Recommended Permit Modifications and Operating Procedures for Landfills relating to Wastes from Drilling in the Marcellus Shale Formation," attached as Exhibit R, suffers from the same defect of not recognizing the presence/absence/variability of the Lead-214 and Bismuth-214 progeny, and the resulting variability of gamma radioactivity measurable at the landfill gate, in truckloads carrying identical Radium-226 concentrations. See especially §§ 2(b) and 4(b)(v) of DEC's memorandum, which seek to establish or verify a correlation without acknowledging how the Lead-214 and Bismuth-214 progeny affect the intended correlation and without describing any steps needed to control for this issue. DEC's reliance on such a correlation is therefore unfounded.

22. Depending on how much Lead-214 and Bismuth-214 are present in a truckload of Radium-226-bearing waste, the gamma radiation dose measured outside the truck may vary *by almost two orders of magnitude*. In other words, the gamma radioactivity measured outside a truck carrying Lead-214 and Bismuth-214 in secular equilibrium with Radium-226 would be about 60 times greater, or almost two orders of magnitude greater, than the gamma radioactivity measured outside an otherwise equivalent truck carrying no Lead-214 and Bismuth-214. These are the two limiting cases, both of which would show the same radionuclide analysis results for samples collected from within the truckload of waste. The Radium-226 analysis results would be the same because this radionuclide is assumed to be present at the same concentration in both cases. The Lead-214 and Bismuth-214 results would be the same in both cases because of ingrowth and achievement of secular equilibrium during the required sample holding period (at least 21 days). Thus, the initial presence or absence of Lead-214 and Bismuth-214 has no effect on the truckload sample analysis but would have a roughly 60-fold effect on gamma radioactivity

measured outside the truck at the landfill gate. This renders any "correlation" meaningless unless the monitoring procedure at the landfill gate can quantify, and control for, the concentrations of Lead-214 and Bismuth-214 in the load of waste at the time the truck enters the landfill gate.

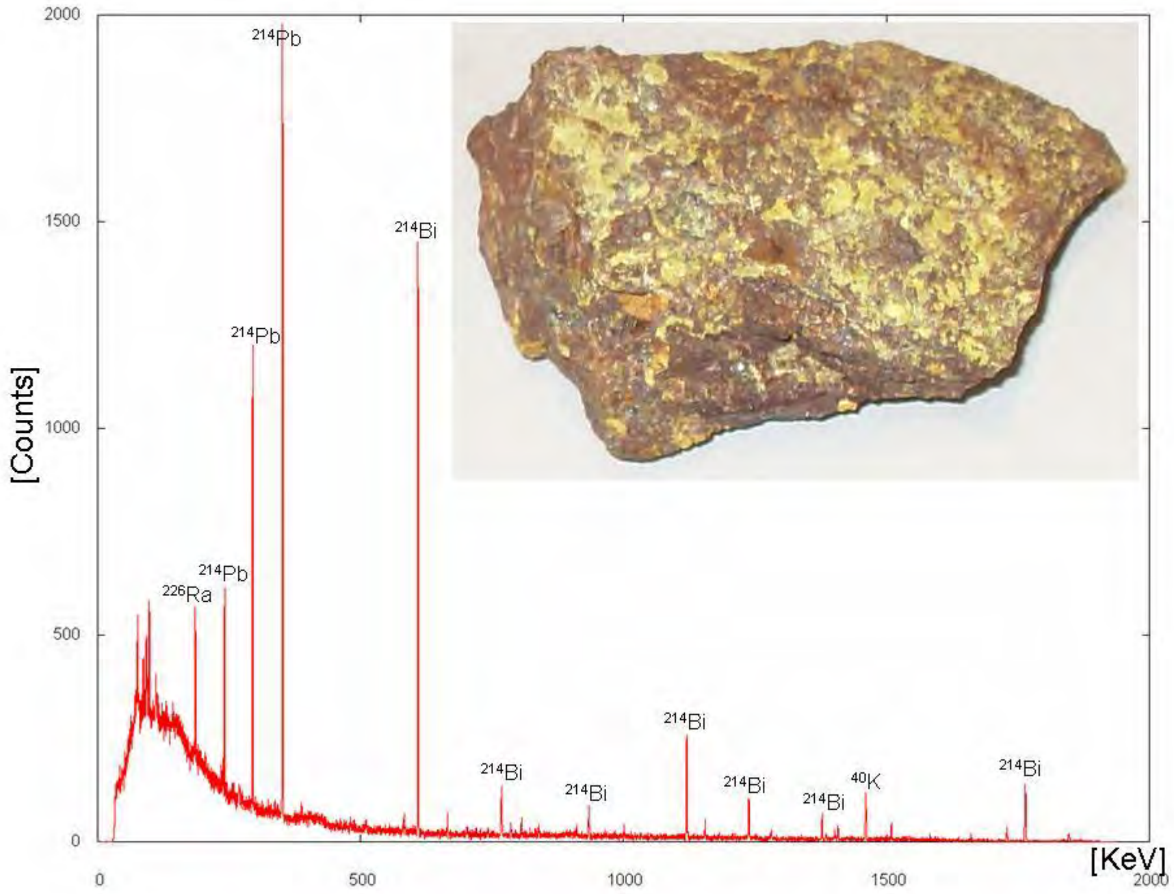
23. The 60-fold variation in the gamma dose measured outside a truck carrying a given concentration of Radium-226 is a combination of two different factors. One factor involves the different gamma yields associated with the three radionuclides; the other is a result of the Lead-214 and Bismuth-214 gamma emissions having greater energy and greater penetrating power than the Radium-226 gamma emissions.

24. The first factor (different gamma yields associated with the three radionuclides) is described in general terms as follows:

> Characteristic gamma rays are emitted in the decay of most radioisotopes; alpha, beta and positron decay and electron capture generally leave the product nucleus in an excited state, which subsequently decays to the ground state with the emission of one or more photons. These gamma rays vary widely in energy and abundance from one isotope to another.
>
> (D. Harris and J. Epstein, eds., *Properties of Selected Radioisotopes*, NASA report SP-7031 (1968), p. 5.)

The "abundance" of gamma rays emitted in the decay of radionuclides can also be called the "gamma yield" of the decay process.

25. The gamma-ray energies and gamma yields associated with Radium-226, Lead-214, and Bismuth-214 decay are available from sources such as D. Delacroix et al., *Radionuclide and Radiation Protection Handbook 2002*; V. Chisté, M.M. Bé, and C. Dulieu, *Evaluation of decay data of radium-226 and its daughters*, International Conference on Nuclear Data for Science and Technology 2007; an online table posted at https://www.cpp.edu/~pbsiegel/bio431/genergies.html; and a graphical representation from https://en.wikipedia.org/wiki/File:Gammaspektrum_Uranerz.jpg which is shown below:



26. As shown in these various sources, decay of Radium-226 is associated with the emission of a relatively weak gamma ray (186 keV), and the gamma yield of this decay process is low, with only about 3.5% of the Radium-226 decay events producing a gamma ray. In other words, each Radium-226 decay emits an alpha particle as the Radium-226 parent atom is transformed into the progeny Radon-222 atom, but gamma rays are emitted in only 3.5% of these transformations or decay events. In other words, there is about a 3.5% probability that a 186 keV gamma ray will be emitted when Radium-226 decays. The 186 keV gamma ray is the only gamma associated with Radium-226 decay. In contrast to this, the decays of Lead-214 and Bismuth-214 may be accompanied by any one of several different gamma energies and yields. Each decay of Lead-214, for example, may be accompanied by emission of a 242 keV gamma ray

(with a gamma yield of about 7%), or by emission of a 295 keV gamma ray (with a gamma yield of about 19%), or by emission of a 352 keV gamma ray (with a gamma yield of about 36%). Each decay of Bismuth-214, for example, may be accompanied by emission of a 609 keV gamma ray (with a gamma yield of about 46%), or by emission of a 768 keV gamma ray (with a gamma yield of about 5%), or by emission of a 934 keV gamma ray (with a gamma yield of about 3%), or by emission of a 1120 keV gamma ray (with a gamma yield of about 16%), or by emission of a 1238 keV gamma ray (with a gamma yield of about 6%), or by emission of a 1378 keV gamma ray (with a gamma yield of about 4%), or by emission of a 1764 keV gamma ray (with a gamma yield of about 16%).

27. It is evident from the foregoing values that the various gamma rays from Lead-214 and Bismuth-214 decay have higher energies than the 186 keV gamma rays from Radium-226 decay. If all three radionuclides are in secular equilibrium, it is also evident that the various gamma rays emitted by Lead-214 and Bismuth-214 will greatly outnumber those emitted by Radium-226, i.e., will be emitted at a substantially higher rate than those emitted by Radium-226. If a monitoring instrument at a landfill gate is counting gamma rays emitted from a truckload of waste (as expressed in kilocounts per second or KCPS), most of the gamma counts will be from Lead-214 and Bismuth-214 rather than Radium-226 if all three radionuclides are in secular equilibrium. Alternatively, if a truckload of waste contains the same concentration of Radium-226 but no Lead-214 or Bismuth-214, the gamma counts will be much lower. The ratio of the gamma counts for these two limiting cases is approximately 40, as can be calculated from the foregoing values. In other words, the gamma count from Radium-226, Lead-214, and Bismuth-214 in a truckload of waste in which all three radionuclides are at secular equilibrium will be roughly 40 times higher than the gamma count from an otherwise equivalent truckload that

contains no Lead-214 or Bismuth-214. This factor of 40 assumes no shielding from the steel walls of the truck. As described below, the shielding of the truck walls will create an even greater difference between these two limiting cases where the Radium-226 concentration remains the same while Lead-214 and Bismuth-214 are either absent, or present at secular equilibrium concentrations.

28. Shielding values for Radium-226, Lead-214, and Bismuth-214 gamma emissions can be calculated from tables of mass attenuation coefficients for iron/steel such as those published by NIST or in the Radiological Health Handbook. A 5-mm thickness of steel, which is an approximate average thickness over all surfaces of a 30-yard roll-off, will attenuate the 186-keV gamma radiation from Radium-226 decay to about half of its unshielded intensity or count rate. The same thickness of steel will attenuate the more energetic gamma radiation from Lead-214 and Bismith-214 decay to about two-thirds of its unshielded intensity or count rate.[1] When these shielding values are combined with the factor of 40 described above, they indicate that the gamma count from Radium-226, Lead-214, and Bismuth-214 in a truckload of waste in which all three radionuclides are at secular equilibrium will be roughly 60 times higher than the gamma count from an otherwise equivalent truckload that contains no Lead-214 or Bismuth-214.

29. Thus, depending on how much Lead-214 and Bismuth-214 are present in a truckload of Radium-226-bearing waste, the gamma radiation dose measured outside the truck may vary by a factor of about 60. This defeats the purpose of using a gamma monitor at the landfill gate and relying on a simple kilocounts-per-second (KCPS) limit. A reliable correlation to a truckload's concentration of Radium-226 cannot be developed unless and until concentrations of Lead-214 and Bismuth-214 are measurable at the moment of entry into the landfill gate. Using

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[1] Attenuation depends partly on load-specific information such as distribution of radionuclides within a waste load – which, where known, would allow more precise calculation that incorporates buildup, etc.

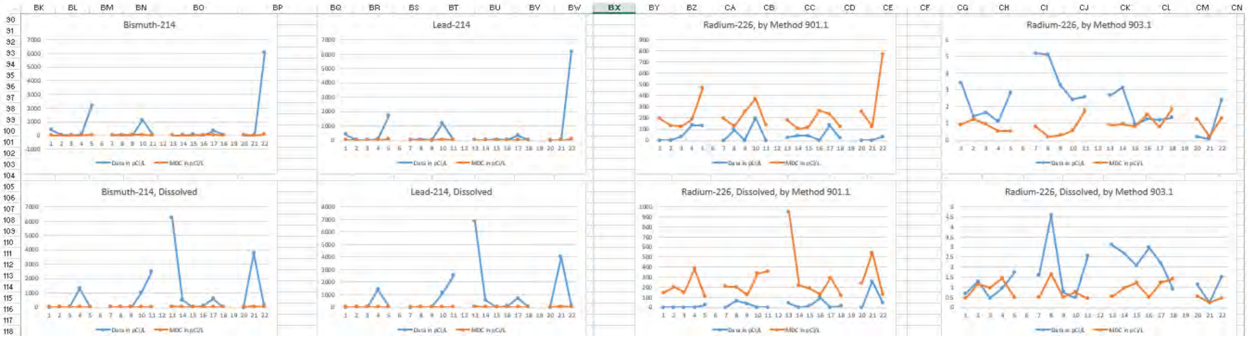
a radioisotope identification device at the landfill gate may be a useful first step toward addressing this problem and developing a reliable correlation, but such a correlation needs to be developed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

Unresolved problems with leachate test results

30. There are significant unresolved problems with the Hakes and Chemung Landfill leachate results reported in the Hakes Leachate Test Reports and Chemung Leachate Test Report. There are at least two problems with these reported results, including (a) high recent radionuclide concentrations and (b) the unrecognized/uninterpreted mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results. See results from the Hakes Leachate Test Reports and Chemung Leachate Test Report plotted below (and in Exhibits S-Z), where the horizontal axis on each graph is time, and the graphs show four different time trends.

For Hakes Landfill:

- 1-5 are the 2015-2017 time trend for Cell 3 Leachate
- 7-11 are the 2015-2017 time trend for Cell 4 Leachate
- 13-18 are the 2014-2017 time trend for Cell 5 Leachate
- 20-22 are the 2016-2017 time trend for Cell 8B Leachate



In the above graphs, the blue lines show time trends for the reported test results, while the orange lines show the detection limit (MDC). See also Exhibits S-V for these graphs in larger format.

For Chemung County Landfill:

- 1-3 are the 2015-2017 time trend for Leachate Pond (Combined Leachate)
- 5-7 are the 2015-2017 time trend for Cells I through III Primary Leachate
- 9-13 are the 2015-2017 time trend for Cell IV Primary Leachate
- 15 is the single data point for the 2017 measurement of Cell V Primary Leachate.

In the above graphs, the blue lines show time trends for the reported test results, while the orange lines show the detection limit (MDC). See also Exhibits W-Z for these graphs in larger format.

31. Despite some variability, the overall trend of high radionuclide concentrations in recent leachate test results is evident in the above graphs.

32. The above graphs also show the mismatch between reported Lead-214 and Bismuth-214 test results and reported Radium-226 test results in leachate from both landfills. The results for Lead-214 and Bismuth-214 are robust and mutually consistent, with recent results ranging up to about 1000 pCi/L for Chemung leachate and about 6000 pCi/L in Hakes leachate. See also the much lower activity reported for Radium-226, consistently less than 10 pCi/L when measured by EPA Method 903.1. Whether these Radium-226 test results are reliable is uncertain and cannot be determined unless/until additional testing is done.

33. The question that needs to be resolved is whether Radium-226 in both landfills' leachate is substantially underreported or whether Radon-222 in both landfills' leachate is high and not recognized as such.[2] One or the other of these possibilities (or some combination of the two possibilities) must be true, given the high activities reported for Lead-214 and Bismuth-214 in both landfills' leachate. Stated another way, either the actual Radium-226 activities have ranged up to ~1000 pCi/L in Chemung leachate and up to ~6000 pCi/L in Hakes leachate, or the actual Radon-222 activities have ranged up to ~45,000 pCi/L in Chemung leachate and up to ~270,000 pCi/L in Hakes leachate. Either of these possibilities – or a combination of the two – warrants followup investigation in order to assess impacts to the environment and public health and safety.

34. Details of these two alternatives are reviewed below along with two other possibilities that can quickly be ruled out. These are the only four possible explanations for how the measured activities of Lead-214 and Bismuth-214 could greatly exceed the measured activity of Radium-226 in samples that have remained sealed for 21 days or more:

- The test measurement of Radium-226 activity is erroneous; the actual activity of Radium-226 in the tested samples was much higher than reported, and in reality was approximately the same as the measured activities of Lead-214 and Bismuth-214. Similar activities for all three of these radionuclides would be expected if all three were in secular equilibrium in the sample. Such an error in the measurement of Radium-226 activity is possible but unlikely based on carrier and tracer yields discussed in the accompanying affidavit submitted by D. May. Details of this alternative are discussed below.
- The test measurements of Lead-214 and Bismuth-214 activities are erroneous; the actual activities of these radionuclides in the tested samples were much lower than reported, and in reality were approximately the same as the measured activity of Radium-226. Similar activities for all three radionuclides would be expected if all

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[2] Hakes and Chemung leachate testing has generally not included tests for Radon-222.

three were in secular equilibrium in the sample. However, such an error in the measurement of Lead-214 and Bismuth-214 activities is highly implausible and can be ruled out here because the measured Lead-214 and Bismuth-214 activities are robust (well above the detection limit) and mutually consistent.

- Lead-214 and Bismuth-214 activities in the sample are much higher than Radium-226 and Radon-222 activities in the sample. In this circumstance, neither Radium-226 nor Radon-222 would be in secular equilibrium with Lead-214 and Bismuth-214, and the decay rate of Radon-222 in the sample would have been much too low to provide Lead-214 and Bismuth-214 ingrowth at the rate needed to maintain their reported activities. In the absence or near-absence of Lead-214 and Bismuth-214 ingrowth, both of these radionuclides must have been present at extremely high levels at the time of sample collection, and the activities of both radionuclides in the sealed sample must have been constantly declining in accordance with their half-lives – but this can be ruled out based on their relatively short half-lives (~20 minutes). Given the number of half-lives that elapsed during the 21-day period that the samples remained sealed before testing, the activities of Lead-214 and Bismuth-214 in leachate at the time of sample collection would have been impossibly high (hundreds of orders of magnitude higher than their reported test results which ranged up to ~6000 pCi/L), thus allowing this possibility to be ruled out.
- Lead-214, Bismuth-214, and Radon-222 activities in the sample are much higher than the Radium-226 activity in the sample, with Radon-222 being at or near secular equilibrium with Lead-214 and Bismuth-214. Decay of Radon-222 within the sample would provide Lead-214 and Bismuth-214 ingrowth at the rate needed to produce their reported activities at the end of the 21-day sample hold period, but the sample would contain far too little Radium-226 to provide Radon-222 ingrowth at the rate needed for the known ingrowth of Lead-214 and Bismuth-214. Given the near-absence of Radon-222 ingrowth, Radon-222 must have been present at a relatively high level at the time of sample collection, and its activity in the sealed sample must have been constantly and exponentially declining in accordance with its 3.82-day half-life. This is possible; it can’t be ruled out based on available

information. Details are discussed below.

35. Thus, there are two alternatives. One is that the actual activity of Radium-226 in the tested samples was much higher than reported due to error(s) in the collection, handling, and/or testing of the samples collected for Radium-226 analysis. The other is that Radon-222 activity (not routinely tested) was high, but Radium-226 activity was not. Distinguishing between these two alternatives could easily be done if samples remained sealed after the original test and could be retested at least several days later. Retesting that showed Lead-214 and Bismith-214 activities similar to the original test results for these two radionuclides would indicate that the first alternative is true or predominant, the reason being that the nearly constant activity of Radium-226, with its 1600-year half-life, would sustain nearly constant rates of Radon-222, Lead-214, and Bismuth-214 regeneration (ingrowth) and decay (activity). Retesting that showed Lead-214 and Bismith-214 activities substantially lower than the original test results for these two radionuclides would indicate that the second alternative is true or predominant, the reason being that the gradually declining activity of Radon-222, with its 3.82-day half-life, would sustain gradually declining rates of Lead-214 and Bismuth-214 regeneration (ingrowth) and decay (activity).

Details of one alternative: Underreported radium concentration in sampled leachate

36. In this alternative, the actual activity of Radium-226 in the tested samples must have been much higher than reported due to error(s) in the collection, handling, and/or testing of the samples collected for Radium-226 analysis. How could such errors arise? Part of the problem may be that an EPA test method for drinking water (Method 903.1) is being used for testing leachate. Leachate samples are aqueous but far less pure than drinking water; they typically contain various dissolved and suspended solids which tend to interfere with sample

preparation. See especially A.W. Nelson, D. May, A.W. Knight, E.S. Eitrheim, M. Mehrhoff, R. Shannon, R. Litman, and M.K. Schultz, "Matrix Complications in the Determination of Radium Levels in Hydraulic Fracturing Flowback Water from Marcellus Shale," *Environ. Sci. Technol. Lett.* **1**, 204−208 (2014). While these authors found radon emanation methods of measuring Radium-226 to be more accurate than wet chemistry methods, the accuracy of an emanation method such as Method 903.1 may be highly dependent on whether Radium-226 will be lost or unrepresentatively partitioned during sample preparation prior to testing. Sample preparation specified in § 8.0 of Method 903.1 includes barium sulfate coprecipitation, which is one of the methods that Nelson et al. found problematic (id. at 206).

37. One possible source of error involves filtration procedures. These cannot be understood in detail from the available information. For example, page 6 of the Chemung Leachate Test Report is a collection record for the C[ell] 5 Primary Leach[ate]; it shows leachate sample collection in a 5-gallon bucket on January 11, 2017. The bottom of the same sheet shows the number of sample containers as 10, evidently indicating that a single bucketful of leachate was poured into ten sample containers, all of which should have contained essentially identical leachate samples. The first of two Chain of Custody forms (id. at 12) shows these ten sample containers being transferred to the ALS Rochester lab along with three other sets of ten from other leachate sample collection points, for a total of 40 containers in all. An instruction printed on the Chain of Custody form says: "Note: Dissolved analysis requires lab filtering."

38. Filtration was evidently performed on half of the 40 containers of samples, and was apparently performed after the ALS Rochester lab transferred the samples to Pace Analytical on January 13, 2017, although no details or specific reference to the filtering process are provided in the report. The second of two Chain of Custody forms (id. at 46-47) shows eight sets of 5 samples

each being transferred to Pace Analytical, half of them marked "dissolved," for a total of 40 containers in all. While the word "dissolved" would tend to indicate that filtration had already been done before ALS Rochester transferred the samples to Pace, this was evidently not the case, as the second Chain of Custody form includes a "Test Comment" that says "Sample Requires In-Lab Filtering" (id. at 46) for the samples designated "Dissolved." Presumably the Pace Analytical lab recognized this as an instruction to filter those samples; however, the uncertainty about filtration is compounded by Pace Analytical's "Sample Condition Upon Receipt" form (id. at 48), which has an ambiguous answer – "N/A" rather than "yes" or "no" – for the line which asks "Filtered volume received for Dissolved tests."

39. While it is possible that filtration was done as a routine procedure within EPA Method 903.1, §§ 8.1 and 8.4, such a procedure needs to be reconciled not only with the findings of Nelson et al. but also with the following observations about filtration in the Pennsylvania TENORM Report:

> Due to high solids content, the samples were not filtered in the field or at the laboratory. The aqueous portion was decanted from 10 of the 51 samples after they had been allowed to settle. The aqueous portion was analyzed for Ra-226 and Ra-228. These results are presented in Table 5-3 along with the original gamma spectroscopy results for the entire sample. The entire sample results include dissolved and undissolved Ra-226 and Ra-228 and are generally one to two orders of magnitude higher than analyses of only the aqueous phase, indicating that the Ra-226 and Ra-228 in these samples were mostly in the form of undissolved solids.
>
> (Pennsylvania TENORM Report at page 5-1.)

Judging from the above observations from the Pennsylvania TENORM Report, the filtration and/or decanting process performed on the Chemung samples was a crucial step, potentially involving a substantial loss of Radium-226, that should have been described in reasonable detail in the Chemung Leachate Test Report.

40. If Radium-226 activities in Chemung and Hakes leachate are indeed in the

neighborhood of 1000 or 6000 pCi/L, they would be much higher than any of the leachate results reported from Pennsylvania:

> Samples of leachate were collected from 51 landfills and analyzed using gamma spectroscopy for Ra-226 and Ra-228. The gamma spectroscopy results are presented in Table 5-1 for the 42 landfills not selected based on volume of O&G waste accepted and Table 5-2 for the nine landfills selected based on the volume O&G waste accepted. Radium was detected above the MDC value in 38 of 51 samples. Sample results from the 42 unselected landfills showed Ra-226 results that ranged from 36.5 to 416 pCi/L with an average of 116 pCi/L. Radium-226 results from the nine selected landfills ranged from 67.0 pCi/L to 378 pCi/L with an average of 125 pCi/L. Radium-228 results ranged from 2.50 to 55.0 pCi/L with an average of 11.9 pCi/L in the 42 unselected landfills. Radium-228 results from the nine selected landfills ranged from 3.00 pCi/L to 84.0 pCi/L with an average of 18.0 pCi/L.
>
> (Pennsylvania TENORM Report at page 5-1.)

41. The above comparison to Pennsylvania test results illustrates the potentially serious nature of errors in Hakes and Chemung sample collection, handling, and/or testing. However, the question of errors in the collection, handling, and/or testing of the Hakes and Chemung samples collected for Radium-226 analysis is entirely speculative in the absence of further testing. The above discussion of possible errors shows ways in which errors might have occurred but offers no clear evidence that errors actually occurred. The important point is that there are only two alternatives – or some combination of the two – that can explain the mismatch between the reported Radium-226 results and the reported Lead-214 and Bismuth-24 test results. Thus, if this alternative isn't true or predominant, the next one must be. As already noted, further testing could readily distinguish between these two alternatives.

Details of the only other alternative: High radon, relatively low radium, in sampled leachate

42. If Radium-226 *wasn't* actually present in the leachate in concentrations ranging up to ~1000 or ~6000 pCi/L, then the leachate must have contained high and apparently unrecognized levels of Radon-222 that were orders of magnitude higher than could be supported by secular

equilibrium with the reported levels of Radium-226. This possibility is plausible, given the high solubility of Radon-222 in water,[3] but further testing is needed to distinguish between this possibility and the possibility of underreported Radium-226. As noted above, further testing could be (or could have been) done in a straightforward manner by retesting existing samples.

43. In this alternative where Radon-222 activity substantially exceeds Radium-226 activity, the concentration and activity of Radon-222 must have been declining exponentially during the sample holding period of about 21 days in accordance with its 3.82-day half-life. During the 21-day holding period[4] – which corresponds to about 5.5 half-lives – the concentration and activity of Radon-222 must have declined by a factor of about 45. Calculating backward from a Radon-222 activity of ~1000 or ~6000 pCi/L at the time of testing (as indicated by the Lead-214 and Bismuth-214 results), the activity of dissolved Radon-222 in the leachate at the time of sample collection must have been ~45 times higher, or ~45,000 or ~270,000 pCi/L. If this alternative is true or predominant, the source and fate of such high levels of Radon-222 need to be investigated. The source is of course Radium-226, but where and how much? Radon-222 is a gas and thus able to migrate away from its parent Radium-226. The short half-life of Radon-222 requires that the parent Radium-226 must be nearby – in other words, within the landfill – so part of what's needed here is an analysis of the pathway within the landfill from the parent radium to its radon progeny. Also needed is a defensible inventory of the amount of Radium-226 within the

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[3] For example, the online *Encyclopaedia Britannica* entry for Noble Gases lists the solubility of Radon-222 as about 230 cm^3 per liter of water at 20º C. This solubility limit can be combined with the specific activity of Radon-222 to express the solubility limit as an activity limit (>300,000 Ci/L) for Radon-222 in water at this temperature. Such a solubility limit, even if somewhat reduced as a result of other solutes present in landfill leachate, is orders of magnitude above the concentrations considered here and would therefore not be a limiting factor for Radon-222 solubility in the Chemung and Hakes leachate samples.

[4] Note that the sample holding time is unavailable for the 4Q 2014 Hakes leachate results because the relevant Hakes Leachate Test Report (Exhibit H) does not include the analytical data sheets that would show the Method 901.1 test dates. This is especially important for the reported 4Q 2014 activities of ~6000 pCi/L for Lead-214 and Bismuth-214. Presumably Pace Analytical and its customers understand the relevance of a 20- or 21-day ingrowth period and specified a sample holding time of least 20 or 21 days.

landfill that would produce enough Radon-222 to account for ~45,000 or ~270,000 pCi/L being present in the leachate at the time of sample collection.

44. Also needed is an analysis of the pathways by which Radon-222 and its progeny may leave the landfill, either escaping through landfill caps, etc., or dissolved in leachate taken offsite for treatment. Part of such analysis would assess the impacts of Radon-222 and its progeny to the environment and public health and safety. The activity of Radon-222 and its ability to flow as a gas would decline relatively quickly in accordance with its 3.82-day half-life, but the fate, transport, and impacts of its longer-lived progeny would need to be addressed.

45. The water-air partition coefficient for radon provides an approximate understanding of Radon-222 concentrations in the air/landfill gas mixtures that are in contact with Hakes and Chemung leachate. The partition coefficient, which can be calculated from the Weigel equation, depends not only on temperature but also on water salinity. See especially E.B. Lieberman, *Radon Solubility in Water as a Function of Salinity and Temperature*, M.S. Thesis, Florida State University (2013). While the coefficient's dependence on other constituents of the aqueous and gas phases has not been fully characterized, the water-air coefficient for radon can be used as an approximation.[5] At a temperature of 20º C, for example, the partition coefficient is about 0.2534. Thus, for air at equilibrium with water in which the dissolved Radon-222 activity is ~45,000 pCi/L (based on Chemung data) or ~270,000 pCi/L (based on Hakes data), the Radon-222 activity in air would be about 45,000/0.2534 = 177,000 pCi/L, or 270,000/0.2534 = 1.05 million pCi/L. Radon-222 concentrations such as 177,000 pCi/L or 1.05 million pCi/L are merely approximations of the levels that would be found in the air/landfill gas mixtures that are in contact with leachate, both within and outside the landfill – but these values provide an idea of the

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[5] Alternatively, the Henry's Law constant for radon (0.0093 mol/L-atm at 25ºC) could be used for this purpose.

non-trivial Radon-222 concentrations that need to be assessed and addressed in the context of this high-radon/low-radium alternative. Within the landfill, one of the main pathways of interest is the aqueous and/or air pathway by which Radon-222 ingrowth from Radium-226 decay reaches the leachate. Other pathways of interest, both outside and inside the landfill, involve plume(s) of Radon-222 that offgas from the leachate.

46. There is currently no proof that this high-radon/low-radium alternative is the true or predominant alternative – but if it's not, then the previous one must be. As already noted, further testing could readily distinguish between these two alternatives.

47. It's important to recognize that the words "high-radon/low-radium" in this alternative refer only to the leachate. A relatively large amount of the parent radium must be present within the landfill even if this radium hasn't migrated into the leachate.[6] Solid materials such as radium and its chemical compounds would not tend to migrate if kept in a relatively dry section of a landfill, but gases such as radon may migrate through interconnected pore spaces within a landfill. Given the high density of radon relative to air or landfill gas, the radon produced from radium decay in a relatively dry upper section of a landfill may migrate downward toward – and into contact with – leachate. Some of the radon gas that comes into contact with leachate will then dissolve into the leachate, as indicated in the above discussion of the water-air partition coefficient for radon. Radon-222 concentrations in the air/landfill gas mixture above the leachate may potentially be as high as 177,000 pCi/L or 1.05 million pCi/L, as discussed above.

48. The radon migration process described above, coupled with radon's solubility in water and in water-based mixtures such as leachate, explains how leachate may become

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[6] The short half-life of Radon-222 dissolved in the leachate requires that the parent radium must be nearby (i.e., within the landfill) in quantities sufficient to generate the high levels of Radon-222 present in the leachate at the time of sample collection (such as ~45,000 or ~270,000 pCi/L of dissolved Radon-222, as discussed above).

"high-radon/low-radium" while most of the parent radium remains in a relatively dry upper section of a landfill. In this manner, radon tends to pose a more immediate risk, but the radium itself will generally be a longer-term risk because of its long half-life. As a general rule, radiological risk from a given radionuclide will decline over time until it becomes negligible after several half-lives, e.g., ten half-lives. For Radium-226, with its 1600-year half-life, the relevant question of long-term risk is whether landfill integrity can be maintained for several half-lives, say 16,000 years.

49. Maintaining landfill integrity for thousands of years against erosion, animal burrows, inadvertent human intrusion, etc., appears unlikely, especially for a landfill not specifically designed for radionuclide disposal. While there are various methods of assessing the long-term risk from radionuclide disposal facilities, one of the crucial missing pieces of information at landfills such as Hakes and Chemung is an accurate inventory of radium contained within the landfill. As discussed above, leachate test results do not directly provide this information. At best, "high-radon/low-radium" leachate test results are proxy measures that show a need for additional testing. Such testing is needed to quantify the radium contained within a landfill that serves as the source of the radon measured in leachate.

Conclusions

50. High radionuclide concentrations in leachate are important not only as current measures of radiological contamination leaving the landfill, but also as proxy measures that may help quantify the source term or inventory of Radium-226 within the landfill. A good understanding of this inventory is needed to assess the landfill's potential long-term impacts and oversight needs. If gamma monitoring at the gate is not providing a reliable measure of Radium-226 entering the landfill, then proxy measures such as Radium-226 and/or Radon-222

concentration in leachate become increasingly important as screening-level indicators of the need for more direct sampling and testing of the contents of the landfill.

51. In assessing offsite radiological impacts to the environment and public health and safety, not only Radium-226 and/or Radon-222 but also the longer-lived progeny such as Lead-210 and Polonium-210 need to be considered.

52. The foregoing issues of high radionuclide concentrations in leachate need to be acknowledged and addressed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

53. Existing methods of monitoring trucks for Radium-226 at the Hakes and Chemung landfill gates are unreliable and unprotective. The so-called correlation between monitor readings and Radium-226 concentrations is flawed, such that waste truckloads containing identical Radium-226 concentrations may exhibit up to 60-fold variations in their monitor readings – and conversely, waste truckloads with up to 60-fold variations in their Radium-226 concentrations may exhibit the same or similar monitor readings. This problem stems from Lead-214 and Bismuth-214 gamma emissions, and from the fact that concentrations of these two radionuclides are not tied to Radium-226 concentrations but may vary widely from truckload to truckload. Using a radioisotope identification device at the landfill gate may help in addressing this problem and developing a reliable correlation. Such a correlation should not be pursued privately or confidentially; it needs to be developed in a transparent, deliberative, and defensible manner, preferably in the context of an EIS process.

54. This report is based on information available to me at this time. Should additional information become available, I reserve the right to determine the impact, if any, of the new information on my opinions and conclusions and to modify or supplement this report if necessary.

Sworn to before me this 18 day of January 2018.

Notary Public, State of New York

CARLYN J. SZAROWICZ
ARY PUBLIC STATE OF NEW YORK
ERIE COUNTY
LIC. #01SZ6320817
COMM. EXP. 3/9/2019

TABLE OF EXHIBITS

May be downloaded from http://treichlerlawoffice.com/waste/hakes/vaughanexhibits.html.

Exhibit A — Vaughan CV

Exhibit B — Truck Monitor Correlation Report, Co-Physics, June 7, 2015

Exhibit C — Hakes 2Q 2012 Leachate Radionuclide Analytical Results

Exhibit D — Hakes 4Q 2012 Leachate Radionuclide Analytical Results

Exhibit E — Hakes 2Q 2013 Leachate Radionuclide Analytical Results

Exhibit F — Hakes 4Q 2013 Leachate Radionuclide Analytical Results

Exhibit G — Hakes 2Q 2014 Leachate Radionuclide Analytical Results

Exhibit H — Hakes 4Q 2014 Leachate Radionuclide Analytical Results

Exhibit I — Hakes 2Q 2015 Leachate Radionuclide Analytical Results

Exhibit J — Hakes 4Q 2015 Leachate Radionuclide Analytical Results

Exhibit K — Hakes 2Q 2016 Leachate Radionuclide Analytical Results

Exhibit L — Hakes 4Q 2016 Leachate Radionuclide Analytical Results

Exhibit M — Hakes 2Q 2017 Leachate Radionuclide Analytical Results

Exhibit N — Chemung Leachate Radionuclide Analytical Results March 2015 through January 2017

Exhibit O — Answer posted 27 September 2005 by George Chabot, PhD, CHP, providing an expert online answer to the question, What value should be taken as the average gamma energy of 226Ra? at the Health Physics Society website, https://hps.org/publicinformation/ate/q4817.html

Exhibit P — Uranium-238 decay series

Exhibit Q — Oak Ridge Institute for Science and Education (ORISE) presentation, Radiological and Chemical Properties of Uranium, available online from the U.S. Nuclear Regulatory Commission website, www.nrc.gov/docs/ML1122/ML11227A233.pdf

Exhibit R — DEC's September 18, 2015 Program Policy Memorandum on Recommended Permit Modifications and Operating Procedures for Landfills relating to Wastes from Drilling in the Marcellus Shale Formation

Exhibit S — Graph of time trends for Hakes leachate test results for Bismuth-214

Exhibit T — Graph of time trends for Hakes leachate test results for Lead-214

Exhibit U — Graph of time trends for Hakes leachate test results for Radium-226 (tested by Method 901.1)

Exhibit V — Graph of time trends for Hakes leachate test results for Radium-226 (tested by Method 903.1)

Exhibit W — Graph of time trends for Chemung leachate test results for Bismuth-214

Exhibit X — Graph of time trends for Chemung leachate test results for Lead-214

Exhibit Y — Graph of time trends for Chemung leachate test results for Radium-226 (tested by Method 901.1)

Exhibit Z — Graph of time trends for Chemung leachate test results for Radium-226 (tested by Method 903.1)

EXHIBIT 2

Affidavit of Mr. Dustin May

January 17, 2018

SUPREME COURT OF THE STATE OF NEW YORK
COUNTY OF STEUBEN

In the Matter of the Application of

SIERRA CLUB, CONCERNED CITIZENS OF ALLEGANY COUNTY, PEOPLE FOR A HEALTHY ENVIRONMENT, INC., JOHN CULVER, and BRIAN and MARYALICE LITTLE,

Petitioners,

For a Judgment Pursuant to Article 78 of the Civil Practice Law and Rules,

–against–

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION, BASIL SEGGOS, COMMISSIONER, AND HAKES C&D DISPOSAL INC.,

Respondents.

AFFIDAVIT OF DUSTIN M. MAY IN SUPPORT OF THE VERIFIED PETITION

Index No. E2017-1384CV

State of Iowa,
County of Johnson, ss.:

DUSTIN M. MAY, being duly sworn, deposes and says:

1. I am a professional chemist and current Ph.D. candidate in Human Toxicology. I received a Bachelor of Science (B.S.) degree in chemistry from the University of Iowa in 2006. I possess substantial experience regarding the analysis of samples for radioactive materials, the properties of radioactive materials (i.e. radioactive decay, radioactive particles, radiation interactions with matter, etc.), and the chemical behavior of radionuclides in the environment. I also possess considerable experience in laboratory operations, accreditation compliance, review and interpretation of laboratory results. I have acquired this knowledge through my employment with the State Hygienic Laboratory at the University of Iowa, the state of Iowa's public health laboratory, in the radiochemistry department for the last eight years, first as an analyst and more recently as department supervisor.

2. Additionally, I, along with colleagues, have published a number of peer-reviewed papers studying drilling wastes, including so-called "hydraulic fracturing flow-black fluids", drilling "bit" cuttings, and environmental impacts downstream from coal-waste processing facilities.

3. All opinions expressed here are solely my own and do not express the views or opinions of my employer or academic institution.

Overview

4. As part of this review I have examined and evaluated several documents related to the Hakes landfill. Specifically, I reviewed the (a) Hakes Landfill Management Plan, (b) Hakes Environmental Monitoring Plan, (c) Hakes Operation and Maintenance Manual, (d) Hakes Landfill Leachate Radiological Analysis Results, and (e) Hakes Annual Operations Reports.

5. Based on this review, I have concluded that: (a) the measured concentrations for radium in the leachate samples are likely correct, and represent the soluble fraction of radium-226 and radium-228 under method conditions, (b) there exists sporadic and extreme disequilibrium between radium-226 and its radioactive daughter radon-222, and (c) radon-222 gas, as well as its radioactive daughters, may be present in and around the leachate at levels of concern.

Background

6. Terrestrial naturally occurring radioactive material (NORM) is ubiquitous in the environment and is primarily the result of two decay series, the so-called Thorium and Uranium Series. Both series begin with long-lived parent radionuclides, thorium-232 and uranium-238, that have been present on Earth since the planet's formation. Isotopes of radium, specifically the long-lived radium-226 and radium-228 have been found of be of particular concern due to their

potentially carcinogenic health impact and their motility in the environment and are thus regulated in drinking water under the Safe Drinking Water Act (SDWA).

7. Analytical methods for the quantification of radium-226 and radium-228 employ principles of radioactive equilibrium. These principles state that when the half-life (i.e. the time taken for half of the atoms of a radioactive isotope to disintegrate/decay) of a parent radionuclide is longer (more than ten times) than the half-life of its radioactive daughter and they not physically separated, the radioactive parent and daughter will reach so-called "secular equilibrium." This means that both the parent and daughter will exist at the same rate of decay and will decay with the half-life of the parent radionuclide.

8. Radium-226 is routinely quantified by measuring the alpha particle emission of its daughter product radon-222 after chemical purification or after sealing by the measurement of the gamma-ray emissions of its more distant decay products, lead-214 and bismuth-214. Both of these measurements require an ingrowth period where the sample is sealed and the radioactive daughters are allowed to reach equilibrium. Radium-228 is also routinely quantified by utilizing its radioactive daughter, actinium-228, either after chemical purification by measuring the beta particle emission or by measuring actinium-228's many gamma-ray emissions.

9. With the regard to drill cuttings and other solid waste materials from the Marcellus Shale, higher concentration of NORM are generally present in these matereials than in typical soil or waste. In a recent study conducted by myself and other co-authors, drill cuttings from Marcellus shale were found to contain between 5 and 8 pCi/g each of thorium-230, uranium-238, uranium-234, radium-226, lead-210, and polonium-210[1]. These concentrations are substantially higher, 2-3 times, than the average concentration found in soil in the eastern United

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[1] Eitrheim, E. S., May, D., Forbes, T. Z., & Nelson, A. W. (2016). Disequilibrium of Naturally Occurring Radioactive Materials (NORM) in Drill Cuttings from a Horizontal Drilling Operation. *Environ Sci Technol Lett, 3*(12), 425-429. A copy of this article is attached as Exhibit A.

States[2]. Sludge from flowback waste impoundment may contain even more NORM, with one recent study showing a "range from 10 pCi/g to several hundred pCi/g depending on age"[3].

Collection and Method Selection

10. Analytical results for leachate provided in reports from On-Site Technical Services, Inc. for gamma emitters, radium-226, radium-228, and uranium on a semi-annual basis from 2012 to 2017. Additionally, leachate characterization results were reviewed from the Hakes C & D, Inc. Annual Operations Reports for the combined leachate. According to the sample collection forms, samples were collected from the cell discharge pipes in 5 gallon buckets. Field measurements of pH, conductivity, turbidity, temperature, and oxygen reducing potential were collected. None of these field measurements appear to be unusual for landfill leachate.

11. Pace Analytical Services LLC performed the analyses for radionuclides. According to Pace Analytical's reports, leachate samples were analyzed for radium-226 EPA 903.1, radium-228 by EPA 904.0, gamma emitters by EPA 901.1, and uranium by EPA 908.0 or ASTM D5174-97. The methods utilized are generally appropriate for aqueous samples, but chemical separations can potentially be susceptible to chemical interferences. According to the provided documents and reports Pace Analytical analyzed samples as received and after filtration in the laboratory. Samples were preserved in accordance with method specifications to a pH of less than 2 with nitric acid.

12. It is important to note that in the process of analysis by both EPA 903.1 and EPA 904.0, solids are removed from the analyzed aliquot by either centrifugation or filtration. This means that only the fractions of radium-226 and radium-228 that are soluble in samples that have

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[2] Shacklette, H.T., Boerngen, J.G. Element Concentrations in Soils and Other Surficial Materials of the Conterminous Untied States. USGS Professional Paper 1270.
[3] Zhang, T., Hammack, R. W., & Vidic, R. D. (2015). Fate of Radium in Marcellus Shale Flowback Water Impoundments and Assessment of Associated Health Risks. *Environmental Science & Technology, 49*(15), 9347-9354.

been preserved to a pH of less than two with nitric acid or in a 10M sodium hydroxide solution (used to dissolve insoluble sulfates) utilized in both methods. It is also important to understand that these methods were developed to analyze drinking water; they may be applied to wastewater, but the initial validations for these methods were focused on drinking water. The leachate samples analyzed are much higher in dissolved solids, with measured conductivities ranging from 2000 to 20,000 µS/cm. These high conductivity readings indicate substantial dissolved solids present in the leachate sample. High concentrations of barium can interfere with these methods, as observed with hydraulic fracturing flowback fluid[4] Characterization results for the combined leachate indicate that these solids are primarily calcium, magnesium, and sodium salts present in the leachate.

Radium Results

13. Across all leachate samples taken from 2012 to 2017, radium-226 and radium-228 concentrations were found to be very low. The concentrations of radium-226 and radium-228 were generally less than 10 pCi/L combined in these samples across all cells. The results reports from Pace Analytical do not contain any notes regarding method deviations or quality control related issues with these analyses. Additionally, the reports state that all quality control parameters meet method-specified limits.

14. Beginning with the 2nd Quarter of 2014 results, Pace began reporting carrier and tracer yields with these results. As part of EPA 903.1 and 904.0 stable barium is utilized to separate radium from the bulk matrix. This known quantity of barium is then measured at the end of the analysis to correct the result for recovery. In EPA 904.0, yttrium is utilized to separate the

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[4] Nelson, A. W., et al. (2014). Matrix Complications in the Determination of Radium Levels in Hydraulic Fracturing Flowback Water from Marcellus Shale. *Environmental Science & Technology Letters, 1*(3), 204-208. doi:10.1021/ez5000379

actinium-228 daughter from the radium-228 parent. This known quantity of yttrium is also measured at the end of analysis to correct the radium-228 result for recovery. The reported carrier yields are generally high and reasonable for all analyses for which they are reported. As neither the samples, nor the general radium concentrations, nor the analysis methods changed during the sampling period, it can be reasonably assumed that the carrier and tracer yields were similar for samples analyzed before these parameters were reported.

15. From review of other stable metal results provided in the Hakes C & D, Inc. Annual Operations Reports, it was also found that concentrations of barium were not found to be substantially elevated in combined leachate samples taken from 2012 through 2017. This indicates that stable barium, a potentially important interferent in both EPA 903.1 and EPA 904.0, is not likely to cause any serious issues with analysis for radium-226 and radium-228 by these methods. Additionally, in review of the reported tracer and carrier yields, no yields greater than 100% were reported. The lack of extremely high carrier yields (>100%) provides additional evidence that stable barium is not present in the leachate samples at concentrations that would cause serious interference with these methods.

16. As a result of the reasoning outlined in this section and the previous section these results likely represent the soluble and leachable fraction of radium-226 and radium-228 present in the leachate samples analyzed. The reported concentrations do not include any contribution from radium-226 and radium-228 that is not soluble in dilute nitric acid, as the preserved samples are stored, nor 10M sodium hydroxide.

Gamma Results

17. Analysis for gamma emissions of naturally occurring radioactive materials (NORM) at low concentrations is generally limited to measuring a small number of radionuclides in the uranium and thorium series and potassium-40. The most useful

radionuclides for these types of measurements are actinium-228, bismuth-212, lead-212 and thallium-208 in the thorium series and lead-214 and bismuth-214 in the uranium series due to their high probability of gamma emission upon decay and their short half-lives, leading to rapid radioactive ingrowth. Under most circumstances, following a period of ingrowth (usually at least 21 days) in a sealed container, actinium-228 is very useful for determining the concentration of radium-228 and lead-214 and bismuth-214 are very useful for determining the concentrations of radium-226 present in a sample as these radioactive daughter products will reach a secular equilibrium state with radium-228 and radium-226 during this period of ingrowth.

18. It should be noted that direct measurement of radium-226, radium-228, uranium-234, and uranium-238 by gamma spectrophotometry is not generally performed in environmental samples. This is due to the lack of useful gamma ray emissions by these radionuclides; gamma rays emitted by these radionuclides are either too rare, occur with energies that are too low to measure, or, as is the case with radium-226, overlap with other radionuclides. As a result of these limitations, the above radionuclides are generally measured by other methods or indirectly by the gamma ray emissions of their daughter products.

19. In some circumstances, secular equilibrium may not be reached in in 21 days. In situations where samples can be substantially enriched in radon-222 gas by physical processes, lead-214 and bismuth-214 may be present at concentrations much greater than that of radium-226. Enrichment can occur in situations where radium-226 is trapped in insoluble solid material, radon-222 escapes as a gas, and dissolves in surrounding water. This is widely seen in groundwater, as shown in the USGS Study, *Trace Elements and Radon in Groundwater Across the United States, 1992–2003,* where radon-222 gas can exist at orders of magnitude higher than radium-226 concentrations would typically exist in groundwater. In these situations, radon-222

gas dissolved in the sample may need to be purged by bubbling air through the sample or boiling the sample to drive out the radon gas prior to sealing to allow for determination of radium-226 concentration by the measurement of lead-214 and bismuth-214.

20. In the samples of leachate taken from 2012 to 2017, very few radionuclides were identified above the reported Minimum Detectable Concentrations (MDC), but some results are concerning. In nearly all of these samples, potassium-40 was found above the MDC, but not at particularly high concentrations. This is to be expected considering the detectable concentrations of potassium observed in the leachate characterization results from the Hakes landfill annual reports. Potassium-40 is generally not of major public or environmental health concern due to the tight regulation of potassium concentrations in biological systems[5]; the concentration of potassium-40 is maintained at more or less stable levels regardless of exposure.

21. Of major concern regarding these results are the concentrations of lead-214 and bismuth-214 found in some of the leachate samples analyzed. In 9 of the 79 leachate samples analyzed from 2012 to 2017, lead-214 and bismuth-214 concentrations exceeded 1000 pCi/L and all of these samples showed good agreement between lead-214 and bismuth 214, indicating that these are unlikely to be false positives as lead-214 decays directly to bismuth-214. These elevated lead-214 and bismuth-214 concentrations were found in samples from Cells 3, 4, 5, 6, and 8B, all cells receiving drilling wastes. The highest observed lead-214 and bismuth-214 concentration was approximately 6000 pCi/L from an unfiltered leachate sample taken from Cell 8B take in Q22017. Of the 9 samples found to be extremely high in lead-214 and bismuth-214 concentrations, 6 of these samples were filtered in the laboratory prior to analysis. In a number of other samples, a substantial disequilibrium was observed, with the concentration of lead-214 and

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[5] ORAU Potassium-40 General Information, https://www.orau.org/ptp/collection/consumer%20products/potassiumgeneralinfo.htm

bismuth-214 greatly exceeding the concentration of radium-226 measured in the samples via EPA 903.1. These results indicate major potential enrichment of leachate with radon-222 gas; the half-lives of lead-214 and bismuth-214, 27 and 20 minutes, respectively, are too short for these radionuclides to exist independently during the time period between collection and analysis, they would have decayed away entirely. Thus, in order for these two radionuclides to be detectable in the samples weeks after collection, they would have to be supported and exist in an equilibrium state with radon-222 gas or radium-226.

22. On average, leachate samples were analyzed between two and three weeks following collection. This would allow for 3.6 to 5.5 half-lives of radon-222 to pass between collection and analysis. This period of decay means that approximately 92% to 98% of radon-222 (and due to the properties of radioactive decay, the same activity of lead-214 and bismuth-214) present at collection would have decayed away by the time of analysis. If we back calculate from analysis to collection with the in the sample from Cell 8B mentioned above, this would mean that, at the time of collection, the radon concentration of the sample was approximately 275,000 pCi/L.

23. There is also the potential that the radon-222 concentration in the sample source may be much higher than the concentrations found in the samples. Normally, to analyze for radon-222 in water samples, Standard Methods 7500 Rn-B, a commonly used method for this type of analysis, specifies that samples be taken in glass, septa-sealed vials are filled carefully to avoid loss of radon-222 gas at collection and the elimination of headspace to prevent loss during transport. Variability in the measured concentrations between the filtered and unfiltered samples taken at the same time, with one analysis showing extremely high concentrations of lead-214 and bismuth-214 and the other showing much less or no observable lead-214 and bismuth-214,

indicates the likely escape of radon-222 gas from the sample. This could have occurred at a number of different points between collection and analysis, including the initial filling of the sample bottles at collection or during handling at the laboratory. Vigorous handling of the samples, changes in temperature, and sample containers that do not seal well or have large headspaces could have lead to the escape of radon from the sample container.

24. This variability in lead-214 and bismuth-214 concentrations between filtered and unfiltered samples of the same leachate also indicates that the presence of these radionuclides are not likely the result of radium-226 decay occurring during the period between collection and analysis of the samples. If this were the case, results for the filtered and unfiltered samples would be the same or the unfiltered samples would contain more lead-214 and bismuth-214, as radium-226 is not likely to be lost during collection, handling, or analysis of these samples. In addition, the results of the analyses for radium-226 by EPA 903.1 do not support the extremely high concentrations that would be required for the lead-214 and bismuth-214 concentrations to be the result of radium-226 decay during the period between collection and analysis with these samples.

25. Without careful handling and direct measurement of radon-222 of samples from the leachate source it is impossible to know the true radon-222 concentration present, but it is likely higher than or equal to the measured lead-214 and bismuth-214 concentrations found in the sample already analyzed.

Radon-222 and Lead-210

26. As a result of the extremely high radon-222 concentration likely present in some of these leachate samples, there is also the potential for the long-lived decay products, specifically, lead-210, and polonium-210, to be present as well. In the above mentioned sample from Cell 8B, radon-222 gas present in the leachate at 275,000 pCi/L would result in lead-210 concentrations of approximately 130 pCi/L after 40 days. This radionuclide possesses a half-life

of 22.2 years and decays through bismuth-210 to polonium-210, with a half-life of 130 days. Once formed from radon-222, these radionuclides are persistent and take many years (generally 10 half-lives) to decay away to insignificant concentrations.

27. Additionally, lead-210 and polonium-210 are not regulated nor tested for in wastewater or drinking water and their environmental behavior is not well understood[6]. Complicating the issue further, lead-210 and polonium-210 do not emit any detectable gamma rays, making them difficult to screen for using *in situ* measurement equipment limiting the analysis to measurement via their beta and alpha emissions, respectively.

28. All of these factors indicate that lead-210 and polonium-210 could be present at levels of concern in the leachate from the Hakes landfill and should be characterized to prevent adverse environmental impacts.

Conclusions

29. In review of the provided data regarding the leachate analysis results, issues appear to exist regarding disequilibrium between radium-226 and radon-222. Radium-226 concentrations in the leachate are generally very low and do not appear to be questionable based on the information provided in the reports from Pace Analytical Services. Variable and extremely high concentrations of radon-222 daughter products, especially considering the low concentrations of radium-226 and the two to three week period between collection and counting, are very concerning.

30. The potentially high concentrations of radon-222 in and around the landfill leachate may pose risks to public health and the environment and thus should be characterized to evaluate its potential impact. Additionally, analyses for important radon-222 daughters, lead-210

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[6] Seiler, R. L., and Wiemels, J. L. (2012) Occurrence of (210)Po and Biological Effects of Low-Level Exposure: The Need for Research. *Environ Health Perspect* 120, 1230-1237.

and polonium-210, should also be performed to evaluate the concentrations in the collected leachate solution, especially after the leachate has aged once exiting the landfill. These decay products must be considered when evaluating the potential environment and public health impact of these wastes.

31. Conclusions drawn in this statement are based solely on the information available at this time. If further information becomes available I reserve the right to alter or amend this statement.

Sworn to before me this 17th day of January 2018

Notary Public

NOTARIAL SEAL IOWA
MEGAN FITZGERALD
Commission Number 783103
My Commission Expires
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EXHIBIT A

Environmental Science & Technology LETTERS
Letter
pubs.acs.org/journal/estlcu

Disequilibrium of Naturally Occurring Radioactive Materials (NORM) in Drill Cuttings from a Horizontal Drilling Operation

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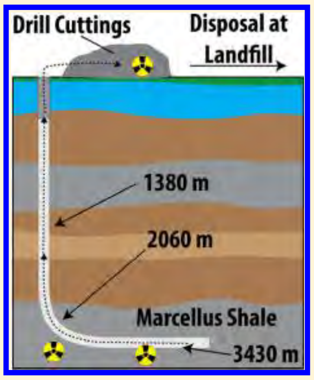
Supporting Information

ABSTRACT: Naturally occurring radioactive materials (NORM) in solid waste or "drill cuttings" produced from unconventional drilling for natural gas extraction wells potentially pose environmental contamination risks; however, the composition and mobility of NORM in these solid wastes are poorly understood. In this study, the composition of NORM, including uranium, thorium, radium, lead, and polonium isotopes, was evaluated in three samples of drill cuttings extracted from a well drilled into the Marcellus Shale formation. Leachability of NORM in drill cuttings was characterized by leaching the solid waste with dilute acetic acid at four different pH values. The uranium-series radionuclides in cuttings and leachate samples displayed isotopic disequilibrium, suggesting some environmental mobility of radionuclides in these shale formations. Our results indicate that isotopic analysis of uranium-series radionuclides is needed for a more complete understanding of the potential environmental contamination risks associated with these solid wastes.



INTRODUCTION

Although there are numerous socioeconomic benefits related to increased domestic energy production, unconventional drilling produces large volumes of waste that may result in long-term undesirable environmental impacts.[1−3] While a range of chemical contaminants have been documented in both solid and liquid wastes, naturally occurring radioactive materials (NORM) from the uranium 238 (^{238}U) and thorium 232 (^{232}Th) series may also be cause for concern.[4,5] Liquid waste (produced fluids and flowback waters) from unconventional wells has received considerable attention from numerous stakeholders because of the large volumes of radium (Ra)-enriched fluids that are produced and the risks these fluids pose to riparian environments.[6−16] Less attention has been paid to the levels of NORM in solid wastes (termed drilling cuttings). Despite the 2.37 million tons of drill cutting extracted in Pennsylvania (PA) in 2011 alone, very little information about the radiochemical profile of these materials is available.[17]

Accurate analysis of NORM in drill cuttings and leachates from drill cuttings requires a multitude of radiochemical techniques that are tailored to the physicochemical and/or radiochemical nature of any given isotope. In 2015, PA and WV (West Virginia) released studies on the NORM content of Marcellus Shale cuttings, indicating that radioactivity levels in cuttings from horizontal portions of an unconventional well were higher than those from vertical portions.[18,19] The studies also concluded that drill cuttings pose minimal risk to the general public. Although this conclusion may be the case, these reports focused on only several long-lived radionuclides from the ^{238}U series, specifically, ^{238}U and ^{226}Ra. Other key ^{238}U-series radionuclides, such as pure α-emitting radionuclides, ^{234}U, thorium 230 (^{230}Th), and polonium 210 (^{210}Po) and the low-energy β-emitter, lead 210 (^{210}Pb), were not explicitly reported. The PA and WV reports also indicated that leachates from landfills accepting drill cuttings contained NORM, and in some cases, the ^{226}Ra concentrations in these samples were above action levels.[18,19] WV and PA did not report levels of ^{238}U in leachates; however, results from sequential leach studies suggest that ^{238}U in Marcellus Shale cuttings is relatively mobile and may contribute to elevated levels of ^{238}U in landfill leachates.[20] While new information about ^{238}U and ^{226}Ra in Marcellus Shale cuttings and leachates is emerging, relative concentrations and mobility of other environmentally persistent radionuclides in the ^{238}U series, including ^{234}U, ^{230}Th, ^{210}Pb, and ^{210}Po, remain unexplored. These radionuclides may provide important information for environmental contamination risk and source apportionment.

Here, we characterized NORM in cuttings associated with horizontal drilling activities in the Marcellus Shale formation. First, using new and modified radiochemical methods, we quantitated and assessed the equilibrium status of environmentally persistent ^{238}U-series radionuclides (^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb, and ^{210}Po) in three solid samples from a horizontal well within the Marcellus Shale formation (Figure 1). Second, the potential for NORM to leach from cuttings extracted from the

Received: November 15, 2016
Revised: November 21, 2016
Accepted: November 23, 2016
Published: November 23, 2016

DOI: 10.1021/acs.estlett.6b00439

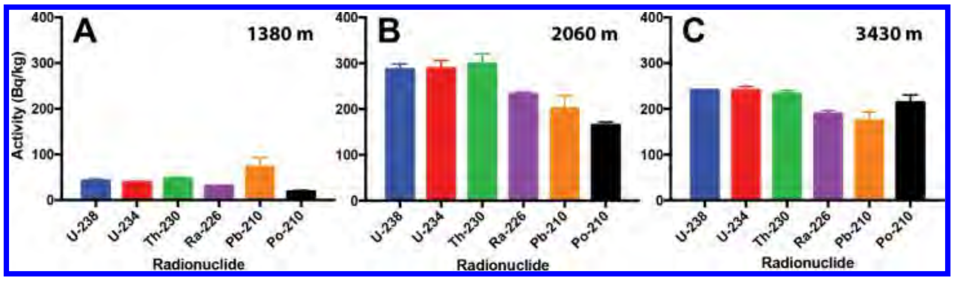


Figure 1. Levels of ^{238}U-series radionuclides in three drill cutting samples from an unconventional drilling operation targeting the Marcellus Shale: (A) sample from a vertical portion of the well at 1380 m, (B) sample from a horizontal drilled portion of the well at 2060 m, and (C) sample from from a horizontal drilled portion of the well at 3430 m.

horizontal portion of the well was assessed using a modified U.S. Environmental Protection Agency (EPA) Toxicity Characteristic Leaching Procedure (TCLP).[21] We hypothesized that ^{238}U-series radionuclides in Marcellus Shale drill cuttings would be in secular equilibrium (steady state) and that ^{238}U-series radionuclides would partition into TCLP leachates with decreasing pH.

■ MATERIALS AND METHODS

General. All chemicals were ACS reagent grade or higher. The University of Iowa State Hygienic Laboratory (SHL) analyzed the solid samples for the content of relevant metals, inorganics, and organics (Table S1 and Figure 2). High-purity germanium (HPGe) γ spectrometry of drill cuttings was performed at the SHL using 500 cm^3 Marinelli beakers on an ORTEC system calibrated to a mixed γ source [Standard Reference Source 101582, Eckert and Ziegler (E&Z), Atlanta, GA] using a previously described methodology.[10,11] Separation and gas flow proportional counting (GFPC) of ^{210}Pb in drill cuttings were performed by PACE Analytical (Greensburg, PA). α spectrometry was performed at the University of Iowa. Radionuclide tracers included ^{232}U standard 92403 (E&Z), ^{229}Th standard 4328C [National Institute of Standards and Technology (NIST), Gaithersburg, MD], ^{209}Po standard 92565 (E&Z), and the cyclotron-produced ^{203}Pb (Lantheus Medical Imaging, Billerica, MA). Emission energies and half-lives were taken from the NUDAT database from the U.S. National Nuclear Data Center (NNDC, Brookhaven National Laboratory, U.S. Department of Energy, Upton, NY).[22] The only exception is for ^{209}Po, for which a half-life of 128.3 years was used, as evidence suggests that its half-life is longer than previously established.[23] All samples were decay corrected to the time of extraction using standard decay correction equations.[24]

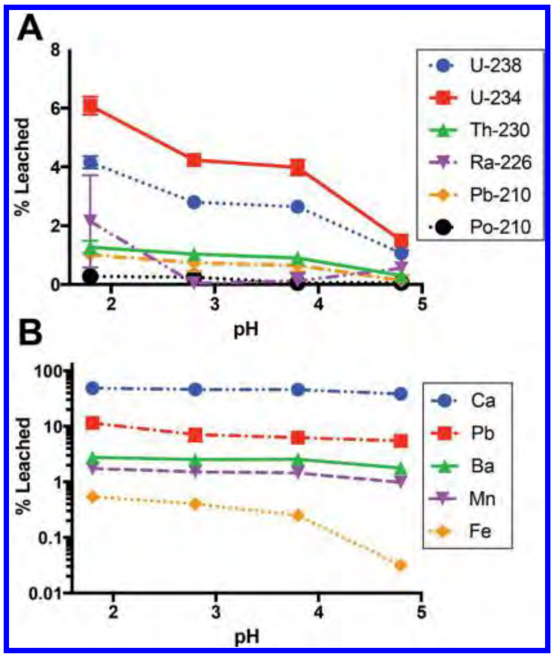


Figure 2. Percentage of (A) radionuclides (Figure 1) and (B) metals leached by acetate buffer at pH 1.8, 2.8, 3.8, and 4.8 from 2060 m drill cuttings (normalized to dry weight).

Drill Cutting Samples. Three solid samples were obtained from a single well in northern PA. The first sample was taken from the vertical, air-drilled section (1380 m) extracted in mid-November 2015, and the two other samples were extracted from the horizontal section (2060 and 3430 m) in mid-December 2015. Samples were received January 8, 2016, and homogenized. Subsamples for U, Th, Ra, and Pb isotopic analysis were dried at 110 °C until a stable weight was achieved. Because of the volatility of ^{210}Po, analysis was performed on the "wet" sample.

Methods of Analysis. *Uranium and Thorium.* ^{238}U, ^{234}U, and ^{230}Th were prepared by a slightly modified rapid method developed for the analysis of actinides in asphalt.[25] First, 50 mBq each of ^{232}U and ^{229}Th tracer were added to the samples (1.0 g), and the dried drill cuttings were ashed in a muffle furnace (600 °C, 1 h) in a platinum crucible. The samples were then rapidly fused in NaOH (10 g, 600 °C, 15 min), before removal from the salt matrix using H_2O. Initially, the samples were precipitated with calcium phosphate [$Ca_3(PO_4)_2$], followed by a second precipitation with cerium fluoride (CeF_3). CeF_3 solids were then dissolved [10 mL, 3 M HNO_3/0.175% boric acid; 10 mL, 3 M HNO_3/1 M $Al(NO_3)_3$] and separated by Eichrom method ACW01.[26] Elemental fractions were prepared for α spectrometry by CeF_3 microprecipitation as previously described.[27]

Radium. Samples (1000 cm^3) for ^{226}Ra analysis were dried, sealed (>21 days, for ^{222}Rn ingrowth), and quantitated by HPGe γ spectrometry (SHL) as previously described.[10,11]

Lead. ^{210}Pb was separated and quantitated by PACE Analytical according to Eichrom method PBS01.[28]

Polonium. ^{209}Po tracer (∼50 mBq) was added to 0.5 g subsamples and then digested with 11 mL of aqua regia overnight on hot plate. Samples were then precipitated [few drops of H_2O_2, $Fe(OH)_3$, manganese dioxide (MnO_2), and NH_4OH], centrifuged, and washed (H_2O) as described for the methodology developed for Marcellus Shale-produced fluids.[11] Pellets were dissolved [20 mL of 0.1 M HCl and 2 mL of 25%

(w/v) hydroxylamine ($NH_2OH{\cdot}Cl$)], autodeposited onto nickel (Ni) disks, and counted by high-resolution α spectrometry.[11]

Leaching Studies. Leaching was performed using a modified EPA TCLP.[21] Centrifugation was used instead of filtration to allow for inclusion of colloid-bound radionuclides. For Pb, Po, U, and Th leaches, 100 mL of acetate buffer adjusted to pH 1.8, 2.8, 3.8, and 4.8 ($n = 4$) was added to 10.0 g of sample. Because of the higher detection limits for ^{226}Ra, samples were increased to 20.0 g of drill cuttings and 200 mL of acetate buffer. All samples were then mixed for 24 h and centrifuged, and the leachates were placed into glass beakers prior to separation and quantitation.

Uranium and Thorium. ^{232}U and ^{229}Th tracers were added, and the sample was subjected to a $Fe(OH)_3$ and $Ca_3(PO_4)_2$ coprecipitation. The resulting solid pellets were dissolved [10 mL, 3 M HNO_3/1 M $Al(NO_3)_3$], separated, and prepared for α spectrometry as described above.

Radium. Leachates were prepared by the EPA 903.0 method by SHL.[29] The only modification to this procedure was that yields were determined by barium 133 (^{133}Ba) using HPGe γ spectrometry en lieu of gravimetric determinations. ^{226}Ra was quantitated by GFPC.

Lead. ^{203}Pb tracer was added, and then the leachates were then subjected to a $Fe(OH)_3$ precipitation. The resultant pellet that was dissolved in 10 mL of 1 M HCl and separated on Pb resin according to Eichrom method PBW01.[30] Yields of ^{203}Pb were determined by sodium iodide (NaI) γ spectrometry.[9] Activities of ^{210}Pb were determined by ingrowth of ^{210}Po via liquid scintillation on an α/β discriminating liquid scintillation counter at SHL.

Polonium. ^{209}Po was added as a tracer, and the leachate was acidified to pH 1−2 using HCl. $NH_2OH{\cdot}Cl$ was added [5 mL, 25% (w/v)], and Po was then autodeposited at 90 °C on a Ni planchet and quantitated by α spectrometry.[11]

Metals. Using the modified EPA TCLP,[21] each acetate buffer (10 mL, pH 1.8, 2.8, 3.8, and 4.8; $n = 3$ for each pH) was added to the drill cuttings (2060 m sample) and homogenized for 24 h. The samples were then centrifuged, decanted, and acidified (100 μL of 16 M HNO_3) prior to analysis by SHL.

■ RESULTS AND DISCUSSION

Chemical Characterization. Cuttings from each location were analyzed for common organic and inorganic constituents that could interfere with radiochemical separations. The shallowest sample (1380 m) was extracted by air drilling from the vertical portion of the well, whereas the two deeper samples from the horizontal section (2060 and 3430 m) were removed by diesel-based drilling techniques. All three samples had high levels of organic fractions, with levels of ethylbenzene (38000−63000 μg/kg), xylenes (270000−450000 μg/kg), diesel fuel (70000−120000 μg/kg), and total extractable hydrocarbons (70000−120000 μg/kg) notably elevated (Table S1). The 1380 m sample is chemically distinct from the deeper Marcellus Shale samples (2060 and 3430 m), which were characteristic of marine black shales as evidenced by enrichment of trace elements cadmium (Cd), copper (Cu), vanadium (V), zinc (Zn), arsenic (As), and selenium (Se).[31]

Characterization of NORM. Environmentally persistent radionuclides from the natural ^{238}U series (^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Pb, and ^{210}Po) were quantified in each drill cutting location (Figure 2A,B and Table S2). On the basis of the chemical analysis and the potential for interference of organic constituents with radiochemical separations, the cutting samples were fired in a muffle furnace. NaOH fusion, based on methods developed for actinide analysis in asphalt, was used to ensure total dissolution of solid material.[25] Drill cuttings are exceptionally challenging samples for radiochemical separations. For example, radiochemical yields for uranium in this study were 40 ± 10%. Previous radiochemical analysis of drill cutting material reported substantially lower radiochemical yields (i.e., ∼2% recovery).[18] These results suggest that new, more robust methods for analysis of drill cutting solid waste are needed. ^{226}Ra analyses are simplified for this complex matrix by elevated abundance and the high-energy γ emissions of ^{226}Ra decay products (^{214}Bi and ^{214}Pb) for HPGe γ spectrometry with little sample preparation (apart from a 30 day hold to allow for decay product ingrowth). The high density of the drill cuttings can interfere with direct measurement of ^{210}Pb (46 keV, 4%) by γ spectrometry;[32,33] therefore, ^{210}Pb was quantitated by methods developed for ^{210}Pb in soil.[28] ^{210}Po was extracted with aqua regia and H_2O_2, because ^{210}Po can adhere to organic matter in the soil[34] and volatilize in dry samples through elevated temperatures (>100 °C)[35] or by biological processes under ambient conditions.[36]

The deep drill cuttings (2060 and 3430 m) have levels of ^{238}U-series radionuclides significantly higher than those of the shallower location (1380 m). These results are consistent with the WV and PA studies that indicate horizontal drill cuttings from the Marcellus Shale have elevated levels of ^{238}U and ^{226}Ra relative to those of the vertical portions.[18,19] The lower level of ^{226}Ra relative to ^{238}U is likely attributable to the partitioning of ^{226}Ra into Marcellus Shale brines that have characteristically elevated levels of ^{226}Ra.[6,8,10,11] As expected, the horizontal portions are also elevated in environmentally persistent radionuclides, ^{234}U, ^{230}Th, ^{210}Pb, and ^{210}Po. Contrary to our hypothesis, the ^{238}U-series radionuclides were not in secular equilibrium. In all samples, ^{226}Ra levels were lower than ^{238}U, ^{234}U, and ^{230}Th levels, which is consistent with other observations.[18,19] Similarly, levels of ^{210}Pb and ^{210}Po were decreased relative to the ^{226}Ra level. This disequilibrium is likely attributable to partitioning of the noble gas, radon 222 (^{222}Rn), in the subsurface as is expected of gaseous hydrocarbons. The disequilibrium between ^{226}Ra and ^{210}Pb is likely explained by partitioning of ^{222}Rn and could be explored as a tool for determining gas migration in the subsurface.[37] These results suggest that radiochemical equilibrium of ^{238}U-series radionuclides cannot be assumed in Marcellus Shale drill cuttings; accurate assessment of environmental contamination risk by ^{238}U-series radionuclides must include detailed radiochemical analyses.

Leaching. Drill cuttings in the Marcellus Shale region are primarily (98.4%) deposited in landfills;[38] however, the stability of NORM in drill cuttings is uncertain. To assess the potential for ^{238}U-series radionuclides to leach from drill cuttings, we employed a simple, acetate buffer leaching protocol based on the EPA TCLP method,[21] which the EPA believes simulates the leaching that occurs in landfills.[39] We chose to analyze the 2060 m sample as it had the highest levels of all ^{238}U-series radionuclides and would allow for shorter counting times and lower detection limits. In general, we observed negative correlations for the percent radionuclide leached with respect to pH for the ^{238}U-series radionuclides tested ($R^2 = -0.96$ for ^{238}U; $R^2 = -0.96$ for ^{234}U; $R^2 = -0.95$ for ^{230}Th; $R^2 = -0.62$ for ^{226}Ra; $R^2 = -0.96$ for ^{210}Pb; $R^2 = -0.91$ for ^{210}Po) (Figure 2A).

DOI: 10.1021/acs.estlett.6b00439
Environ. Sci. Technol. Lett. 2016, 3, 425−429

Fe and Mn displayed similar trends with greater percentages leaching as pH decreased ($R^2 = -0.99$ for Fe; $R^2 = -0.95$ for Mn) (Figure 2B). The partitioning of ^{238}U-series radionuclides into the acetate solution may in part be explained by the desorption from hydrous Fe and Mn oxide minerals, which are well-known to adsorb heavy metals.[40]

Uranium and Thorium. We observed that ^{238}U and ^{234}U were the most leachable radionuclides (4.2 and 6.1% leached at pH 1.8, respectively), which may be explained by the increased solubility of U when it is complexed with the acetate anion.[41] Interestingly, ^{234}U had a mean radioactivity concentration 1.5 ± 0.1 times higher than that of ^{238}U across all leachate samples. This result was unexpected because ^{234}U and ^{238}U were in secular equilibrium in the drill cuttings. However, the isotopic enrichment of ^{234}U is well-known in natural systems because of α recoil enrichment,[42] which occurs when ^{238}U decays and releases high-energy α particles that break chemical bonds. This allows decay products (i.e., ^{234}U) to be forcefully extruded from the crystal lattice and deposits larger amounts of ^{234}U on the outside of the mineral grain, leading to higher leaching rates. ^{230}Th was not leached as readily as U, as expected because of the relatively low solubility of ^{230}Th in environmental systems. Th is particle reactive in most environmental systems and tends to remain adsorbed onto mineral surfaces at pH >2.[43]

Radium, Lead, and Polonium. Interestingly, only 2.1 ± 1.6% of ^{226}Ra leached at pH 1.8, which is consistent with the analogous Ba data, which indicated that the percent leached was 2.76 ± 0.03% . This is surprising, given that high levels of Ba and Ra isotopes were previously reported in Marcellus Shale-produced fluids.[6,10] The amount of ^{210}Pb leached was positively correlated with stable Pb extraction ($R^2 = 0.84$), though in much smaller quantities (1.0 ± 0.1%, vs 11.5 ± 0.6% for ^{210}Pb). The difference in extractability between ^{210}Pb and stable Pb may be a reflection of their geochemical microenvironment, which has been observed for other radionuclides.[44] ^{210}Po was the least soluble radionuclide tested in this system, with only 0.28 ± 0.01% leached at pH 1.8. We expected ^{210}Po to partition into the acetate buffers as ^{210}Po is known to be soluble in acetate,[35] yet the low solubility of ^{210}Po can be explained by its strong particle reactivity and tendency to adhere strongly to organics.[34] Interestingly, recent reports have indicated that ^{210}Po in reduced sediments can be volatilized by aerobic marine microorganisms.[36] Drill cuttings from the Marcellus Shale could serve as an interesting medium for future studies of the volatility of Po from ancient marine sediments.

Although previous studies have suggested that NORM in drill cuttings pose a minimal health risk to the general public when deposited in landfills,[18,19,45] our results indicate that Marcellus Shale drill cuttings warrant further radiochemical investigation. More studies are needed to develop robust, rapid methods that are suitable for a variety of complex matrices typified by drill cuttings. Additionally, field studies are needed to determine the stability of radionuclides in landfills and the potential for NORM from drill cuttings to migrate into landfill leachates. Studies assessing the risks of exposure to NORM should include analysis of pure α emitters (^{234}U, ^{230}Th, and ^{210}Po) and the low-level β emitter (^{210}Pb). For example, the most leachable isotope in these drill cuttings was ^{234}U, which is typically not detected or reported by standard environmental monitoring methods. Accurate assessment of the human health risks associated with drill cuttings should include isotopic analysis of all environmentally persistent radionuclides.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.6b00439.

> Metal, organic, and inorganic composition of drill cuttings (Table S1) and radioactivity concentrations in drill cuttings (Table S2) (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We kindly thank the staff at PACE Analytical Services for contributing time and resources to this study. Funding for these studies was provided by the Center for Global and Regional Environmental Research (CGRER) at the University of Iowa.

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DOI: 10.1021/acs.estlett.6b00439
Environ. Sci. Technol. Lett. 2016, 3, 425−429

EXHIBIT 3

Affidavit of Dr. David Carpenter

January 17, 2018

SUPREME COURT OF THE STATE OF NEW YORK
COUNTY OF STEUBEN

In the Matter of the Application of

SIERRA CLUB, CONCERNED CITIZENS OF ALLEGANY COUNTY, PEOPLE FOR A HEALTHY ENVIRONMENT, INC., JOHN CULVER, AND BRIAN AND MARYALICE LITTLE

Petitioners,

For a Judgment Pursuant to Article 78 of the Civil Practice Law and Rules,

–against–

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION, BASIL SEGGOS, COMMISSIONER, AND HAKES C&D DISPOSAL INC.,

Respondents.

AFFIDAVIT OF
DAVID O. CARPENTER,
MD, IN SUPPORT OF
THE VERIFIED
PETITION

Index No. E2017-1384CV

State of New York,
County of Rensselaer, ss.:

DAVID O. CARPENTER, MD, being duly sworn, deposes and says:

1. I am a public health physician who received his MD degree from Harvard Medical School in 1964. I chose a career of research and public health rather than the practice of medicine. I served as the Chair of the Neurobiology Department of the Armed Forces Radiobiology Research Institute, the research arm of the Defense Nuclear Agency, between 1973-1980 where I directed and performed research on the health effects of ionizing radiation. In great part because of that background and because of the concern of effects of ionizing radiation resulting from the Three Mile Island nuclear plant incident, in 1980 I was appointed as the Director of the Wadsworth Center for Laboratories and Research of the New York State Department of Health, where in addition to my other responsibilities I continued research on

health effects of ionizing radiation, funded in most part by the Defense Nuclear Agency. As a result of my expertise in these areas I have authored numerous scientific research articles, reviews and book chapters on various aspects of human health effects of ionizing radiation.

2. In 1985 I was appointed as Dean of the School of Public Health at the University at Albany, created as a partnership between the New York State Department of Health and the University at Albany. I stepped down from that position in 1998, but continued in my academic appointment as Professor of Environmental Health Sciences and also assumed my current position as Director of the Institute for Health and the Environment at the University at Albany. I continue my research on environmental causes of human disease from exposure to chemicals and various forms of radiation.

3. In 2014 my colleagues and I published a scientific report on air releases around fracking sites in five states, documenting episodic releases of carcinogenic chemicals at levels much above federal and state standards. When New York took action to prevent fracking the Commissioner of Health waved our publication as a basis for that decision. I have remained active in research and study of health effects associated with the oil and gas industry, including a recent report on air releases from natural gas compressor stations in New York but ones that are carrying Pennsylvania natural gas across the state. I have also testified and lectured on health concerns from fracking and also from compressor stations at local and state levels in New York and Pennsylvania.

4. All opinions expressed here are my own and do not necessarily reflect positions of my institution.

Overview:

5. I have been requested to provide an opinion as to whether the procedures regulated by the New York State Department of Environmental Conservation (NYSDEC) regarding drilling wastes from natural gas extraction activities in Pennsylvania are adequately protective to human health. In this regard I have reviewed the expert reports prepared by Dr. Raymond C. Vaughan and Mr. Dustin M May, and the memorandum to Regional Materials Management Engineers issued by the Acting director of the Division of Material Management of NYSDEC regarding drilling wastes from natural gas extraction activities in Pennsylvania coming the landfills in NYS. I have also examined some of the quarterly reports on radionuclide monitoring results of leachate from the Hakes C&D landfill. I have also been provided access to several documents related to the Hakes landfill including the Hakes Landfill Management Plan and the Hakes Environmental Monitoring Plan, although I have not reviewed these in great detail.

6. Based on the information provided to me and my knowledge of the human health effects arising from exposure to ionizing radiation, I have concluded that: (a) there are substantial and significant risks to human health posed by the current procedures used at the Hakes Landfill and approved by NYSDEC, (b) while the greatest threat to human health comes from inhalation of radon-222, other naturally occurring radioactive material (NORM) and the progeny of these elements pose significant threats to human health, and (c) inhalation is the route of exposure of greatest concern but other routes (ingestion, dermal absorption) are also possible.

Background:

7. All forms of ionizing radiation pose threats to human health, and there is no level of exposure that does not increase risk. Radiation is a proven human carcinogen according to all international and national organizations, including the International Agency for Research on Cancer (IARC) of the World Health Organization, the US Environmental Protection Agency and the National Toxicology Program of the National Institutes of Health. Ionizing radiation has sufficient energy to damage DNA, leading to increased risk of cancer and mutations. This results in an increased risk of birth defects and well as mutations that will alter succeeding generations. Ionizing radiation also will damage other cellular components either by direct ionization or through generation of reactive oxygen species (free radicals) that react with DNA and other cellular components to cause damage. It is the policy of the US government that for any mutagenic carcinogen, such as ionizing radiation, there is a linear dose-response relationship between exposure and cancer, which is to say that there is no exposure that does not increase risk of cancer.

8. Ionizing radiation comes in several forms, each with different energies. The most dangerous are alpha particles, which are helium nuclei. These have a positive charge and consist of two protons and two neutrons, so they have a much higher mass than other radioactive emissions, and have high momentum. While they do not penetrate deeply into tissue they are particularly dangerous if inhaled or ingested. Neutrons and protons have only one quarter of the weight of alpha particles, and neutrons have no charge while protons are positively charged. Because of having less weight they penetrate deeper into the skin or body, and neutrons penetrate further than protons because they lack charge. Beta particles are electrons, with a very small mass but a negative charge. Because of their small mass they can penetrate further but their

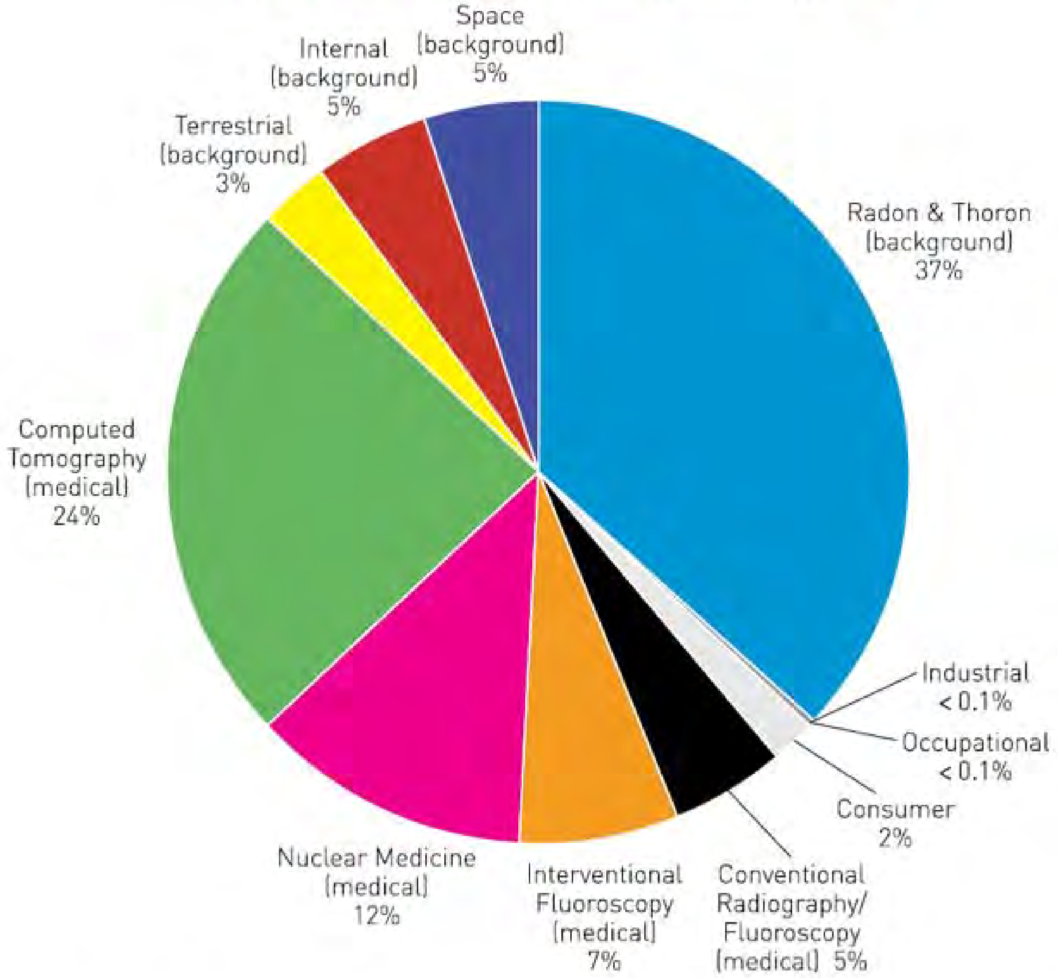
pathway through the body is irregular because of their charge. Gamma rays, x-rays and cosmic rays are electromagnetic forms of energy that do not have any mass. They easily pass through the body but are much less likely to cause harm by colliding with cellular constituents. Depending upon route of exposure the human health hazard coming from ionizing radiation is greatest for alpha particles, followed by neutrons and protons, and much less for electromagnetic radiation.

9. Route of exposure to ionizing radiation is important. Alpha particles do not travel far into the body but because of their mass, they are very dangerous if inhaled or ingested. This is because the lung and the gastrointestinal track are both very sensitive to ionizing radiation, and if an alpha emitter is in either the lung or the gut it can cause significant ionization to the alveoli and bronchioles of the lung or the cells lining of the gastrointestinal track. Neutrons and protons do not have a major role in the concerns over fracking wastes. Gamma rays, because of their low quality factors (a measure of degree of biological damage), are of lesser concern in this case although they are released from the radionuclides found in fracking wastes.

We are all exposed to ionizing radiation. Figure 1 shows the average current sources of exposure. About half comes from medical procedures but the other half comes from radioactive substances found on the earth or coming from cosmic rays. Radon is the single largest source, and the route of exposure is primarily by inhalation. However there are natural radioactive compounds in drinking water and food, including uranium, thorium and radium. These are listed under “consumer products” in Figure 1. There is natural potassium 40 in our bodies, a part of the potassium ion that is the major cation in every cell of the body, identified as “internal background”. There is also some exposure to radioactivity from soil, rocks and other sources under the category of “terrestrial background”. Finally cosmic rays, which are electromagnetic, are listed as "space backgrounds". Thus it is impossible for any person to totally avoid exposure to ionizing radiation. However because any amount of radiation exposure is harmful it is critical that exposure be reduced to the greatest degree possible.

Figure1:

All Exposure Categories
Collective Effective Dose (percent), 2006



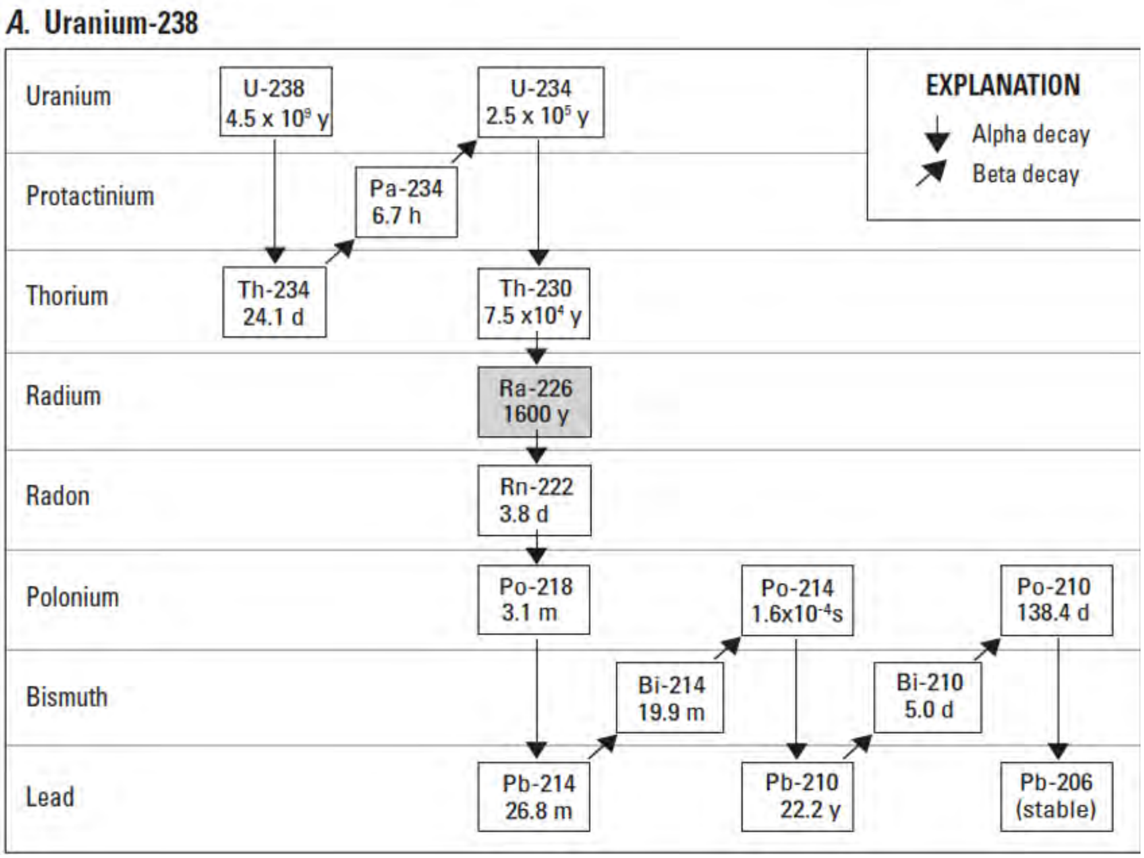
Total collective effective dose (1,870,000 person-sieverts [Sv]) and total effective dose per individual in the U.S. population (6.2 millisieverts [mSv])
Percent values rounded to the nearest 1%, except for those <1%.

Source: National Council on Radiation Protection and Measurement. Ionizing Radiation Exposure of the Population of the United States. Report No. 160, Figure 1-1. Bethesda, MD: NCRP; 2009.

10. The greatest danger to human health comes from alpha particles. This is because of their energy and because the most common routes of exposure are inhalation or ingestion, where the powerful alpha particles have intimate contact with lung or gastrointestinal tissues, leading to

lung or gastrointestinal cancers. Figure 2 shows the decay process of uranium 238. Note that alpha decay is indicated by downward arrows, but beta decay is indicated by upward arrows. Again the greatest risk comes from alpha decay, and most of the progeny of uranium 238 are alpha emitters. Radon-222 is the form of greatest concern because it is a gas. While it has a relatively short half-life of 3.8 days, its decay by alpha emission and its decay products are not gases are also alpha emitters with short half-lives. When radon is inhaled and decays in the lung, its progeny deposit in the lung and undergo further decay, causing damage.

Figure2:



11. Radon-222 is also frequently ingested, most often in drinking water. While uranium and thorium are not very water soluble, radium is quite water soluble, so drinking was can be a source of both radon and radium. Other natural radioactivity substances are also commonly

found in food at low concentrations. When these radioactive elements decay within the gastrointestinal track they damage the cells lining the gut. These cells are particularly sensitive to ionizing radiation because they divide frequently (about every two days), and it is during the process of cell division that ionizing radiation causes the greatest harm. If these radioactive heavy elements are absorbed they tend to deposit in bone, where they increase the risk of leukemia and bone cancer. Thus anything that increases the risk of elevations of radioactive alpha emitters in either water or food poses significant risk to human health. Furthermore lower exposures to ionizing radiation that do not cause specific diseases are known to decrease life span in animals and humans. This is almost certainly due to the generation of the reactive oxygen species mentioned above, as these are known to be the basic cause of the aging process.

12. Drill cuttings and de-watered mud will contain NORM and most of the radioactive elements present are alpha emitters. The multiple leachate radiological analytical results document the presence of uranium 235 and 238, thorium 232 and 234, radium-226 and 228, and lead 212 and 214. For all of these radionuclides there is a distribution between particulate-bound and dissolved concentrations, as expected.

13. When drill cuttings and de-watered mud containing uranium, thorium and radium are placed in the landfill they will continue to emit radiation for a very long time because they have long half-lives. The standard practice in contemporary landfills is to cover them sufficiently to prevent rain water from penetrating and causing greater amounts of leachate. However these covers often erode with time, leading to the strong possibility that 100-plus years from now an increased amount of highly radioactive leachate will be coming from the landfill.

14. As clearly shown in the expert reports of Dr. Vaughan and Mr. May, DEC has grossly underestimated the amount of radium in the landfill because the method used for radium analysis

was not adequate in leachate containing high concentrations of dissolved solids, leading them to conclude that the levels of radium in the leachate were low, and because they ignored the presence of high concentrations of radon. One cannot get high concentrations of radon without having high concentrations of radium. This indicates that there are high concentrations of radium in the landfill that will continue to generate radon and that both will continue to be found in the leachate for centuries, given the half-life of radium being 1600 years.

15. Accepting radioactive fracking waste in the landfills in New York will lead to human exposure to ionizing radiation by various routes. The greatest concern is inhalation of radon. The levels of radon is air above the leachate may potentially be as high as 1.05 million pCi/L, as documented in the report of Dr. Vaughan. This poses a clear hazard to anyone in the vicinity of leachate. Radon will also be released into the air over the landfill. The leachate will migrate into ground water, where radon will be transported and will appear in the drinking water of people on wells and be ingested. A major hazard will come from hot water showers, where the radon is released from the water by the heat and will fill the shower stall and be inhaled. The radon will also migrate up from the ground water in basements of homes, where it will be inhaled by occupants.

16. When ground water is used as drinking water for those persons with wells they will be ingesting radon, radium and lower concentrations of the other less soluble radionuclides that are dissolved in the water as well particulates containing bound radionuclides coming from the fracking drill cuttings and de-watered mud.

17. There is always dust that comes from landfills, and the dust particulates will contain radionuclides that are derived from the progeny of uranium. This dust will get into homes, will be deposited in garden soils and this will be another, albeit less important, route of exposure.

Conclusions:

18. As made clear by the reports of Dr. Vaughan and Mr. May and the leachate analyses, there is a significant amount of radioactivity contained in and coming from the Hakes C&D Disposal Site as the result of the deposits of drill cuttings and de-watered mud coming from fracking sites in Pennsylvania.

19. The net effect of New York accepting drill cuttings and de-watered mud from Pennsylvania fracking sites will be the New Yorkers will have an increased risk of cancer, especially lung and gastrointestinal cancers, an increased risk of birth defects coming from DNA damage and increased risk of a shortened life span.

20. There is reason to believe the DEC is underestimating the amount of radioactivity deposited in and being released from the landfill. The statement made in the Memorandum of 18 September 2015 that "drilling wastes such as drill cuttings do not display elevated radioactivity above naturally occurring background levels" is simply untrue.

21. The peer-reviewed scientific evidence available to anyone indicates that the carcinogenic material found in fracking waste poses a real hazard to human health. Any increase in exposure to ionizing radiation beyond that which is unavoidable should not be tolerated.

Sworn to before me this 17th day of January 2018.

Notary Public

FRANCIS J. ZERONDA
NOTARY PUBLIC, STATE OF NEW YORK
Registration No. 02ZE6185001
Qualified in Albany County
Commission Expires Apr. 14, 2020

EXHIBIT 4

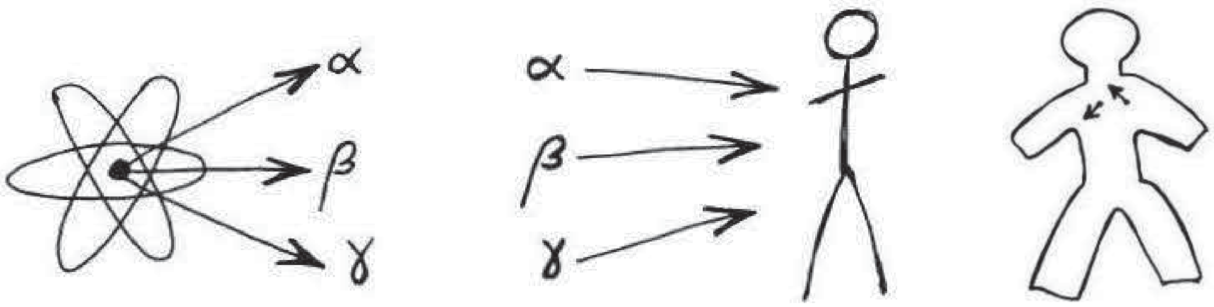
Presentation by Dr. Raymond Vaughan

February 10, 2018

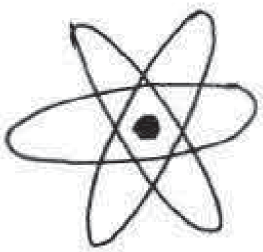
Unresolved Issues for Disposal of Radium-bearing Wastes at Hakes Landfill

Raymond C. Vaughan, Ph.D., P.G.

Sierra Club/CCAC Public Meeting
Campbell, NY
February 10, 2018



Overview of Radioactivity



Some atoms are ***stable:*** for example, **Carbon-12**

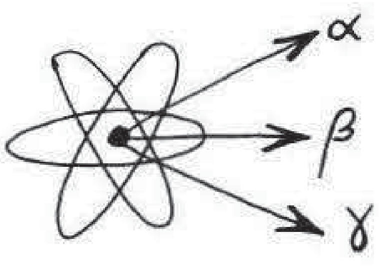
Some atoms are ***unstable*** (**radioactive**):

- Natural: for example, **Carbon-14, Potassium-40, Uranium-238, Radium-226, Radon-222**
- Manmade: for example, **Cesium-137, Plutonium-239**

Radiological Terminology

- **Radioactivity:** the process by which an unstable atom emits radiation and becomes a different atom that is generally more stable
- **Radiation:** energy in the form of waves or particles (*such as alpha or beta particles, or gamma rays*) given off by an unstable atom
- **Radioactive Material:** any substance or material that contains unstable atoms which give off radiation
- **Radioactive Contamination:** radioactive material in an unwanted or undesignated place
- **Decay:** process of radioactive atoms releasing radiation over a period of time and becoming stable (also called disintegration)
- **Half Life:** the time it takes for a group of atoms to decay to half of their original activity

RADIOACTIVE **EMISSION** or **ACTIVITY:**



Alpha particle

Beta particle

Gamma ray

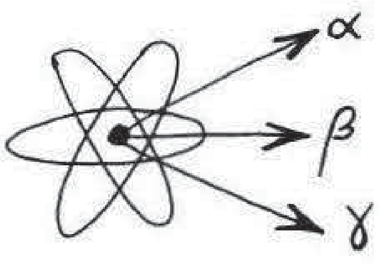
37 billion disintegrations per second = 1 Curie

(in other words, 37 billion unstable atoms per second are decaying and emitting particles/rays)

1/1,000,000,000,000 Curie = 1 picocurie (1 pCi)

1 disintegration every 27 seconds = 1 pCi

RADIOACTIVE **EMISSION** or **ACTIVITY:**



Alpha particle

Beta particle

Gamma ray

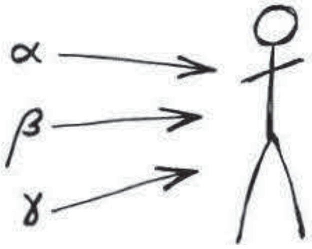
pCi

RADIOACTIVE **EXPOSURE** or **DOSE:**

Alpha particle

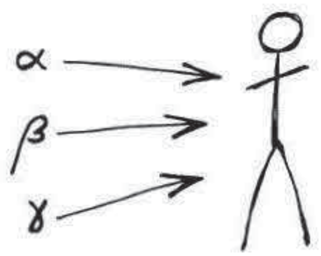
Beta particle

Gamma ray



mrem

RADIOACTIVE **EXPOSURE** or **DOSE:**



Alpha particle

Beta particle

Gamma ray

1 rem = a measure of exposure or dose

1/1000 of 1 rem = 1 millirem (1 mrem)

Radioactive exposure or dose (mrem) can't be correlated with radioactive emission or activity (pCi) without knowing the exposure pathway or pathways. (*How are humans being exposed? Need details*.)

Penetrating power in living tissue:

ALPHA particle: Less than 1 millimeter, so usually can't penetrate human skin or a piece of paper -- but can be very damaging to living tissue if not protected by a barrier such as skin or paper

BETA particle: Can penetrate several millimeters in living tissue

GAMMA ray: Very penetrating (but depends on the energy of the gamma ray)

INTERNAL EXPOSURE if radioactive material is ingested or inhaled

With inhalation or ingestion, there's no protective barrier such as skin.

For example:

- Potassium-40
- Radium
- Radon (**GAS**) and its progeny or "daughters"

If ingested or inhaled, radioactive material can be in direct contact with intestinal lining, lung tissue, and other internal organs

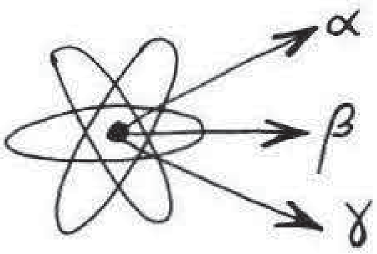
Are there pathways for ingestion or inhalation to occur? If so, the pathways need to be identified by measurement and/or modeling.

Exposure pathways?

Exposure pathways from Hakes landfill to humans have not been clearly identified or adequately investigated – but high levels of radon within the landfill and its leachate may cause *some* level of human exposure at downwind locations

Radium may also pose *some* level of long-term health risk for thousands of years if landfill integrity can't be guaranteed

Exposure pathways can/should be identified and quantified by testing and modeling – *preferably within an EIS process*



The Uranium-238 Decay Series (decay chain) and radionuclides such as radium, radon, lead, and bismuth in that 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)

↑

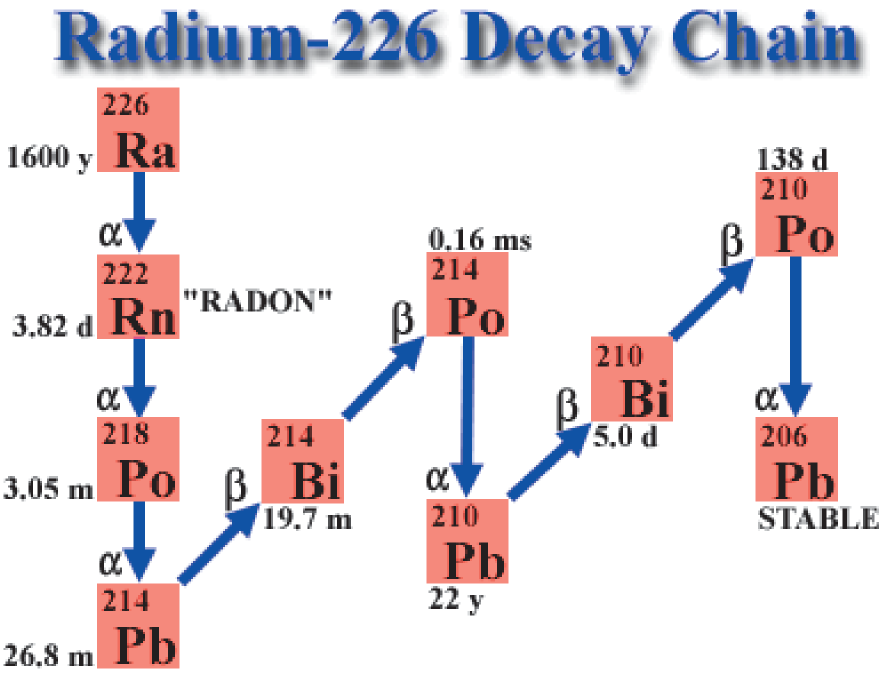
PARENT RADIONUCLIDE

PROGENY
or
DAUGHTER
or
DECAY PRODUCT

↓

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

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 µsec)
↓
Lead-210 (22 years)
↓
Bismuth-210 (5.0 days)
↓
Polonium-210 (140 days)
↓
Lead-206 (stable)



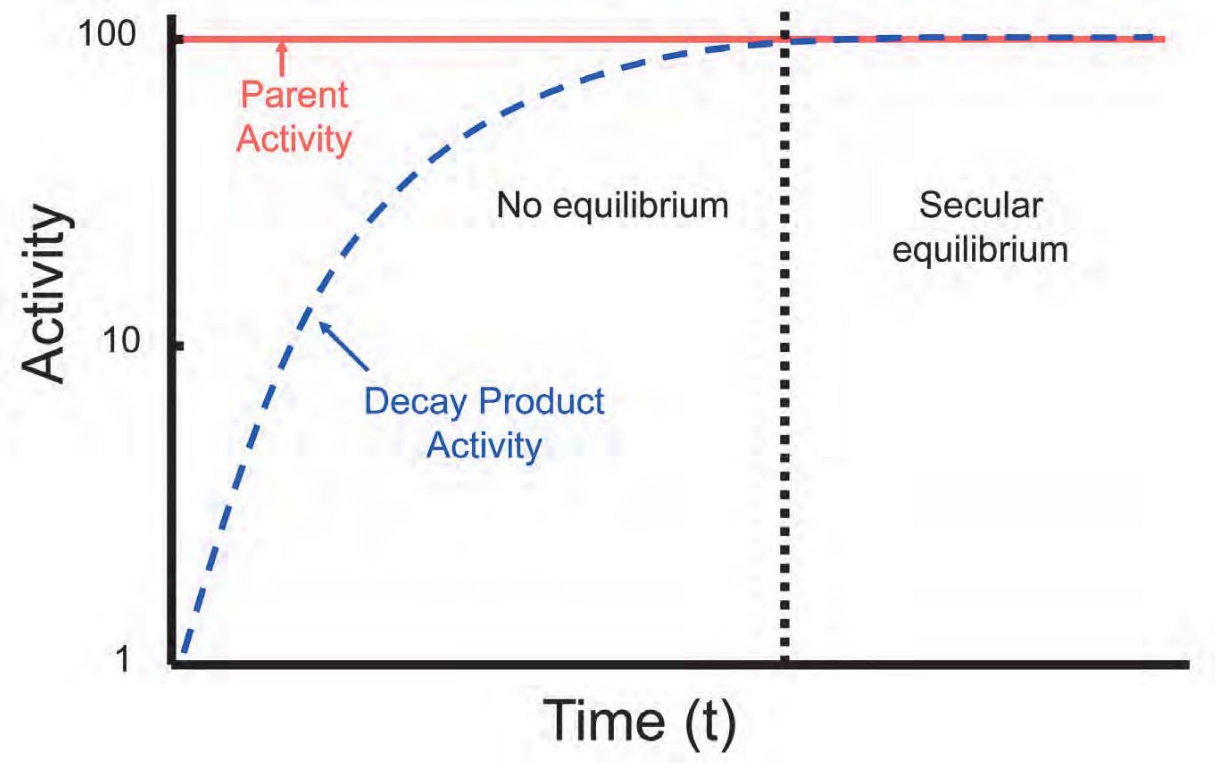
Secular Equilibrium

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

In this circumstance, 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.)

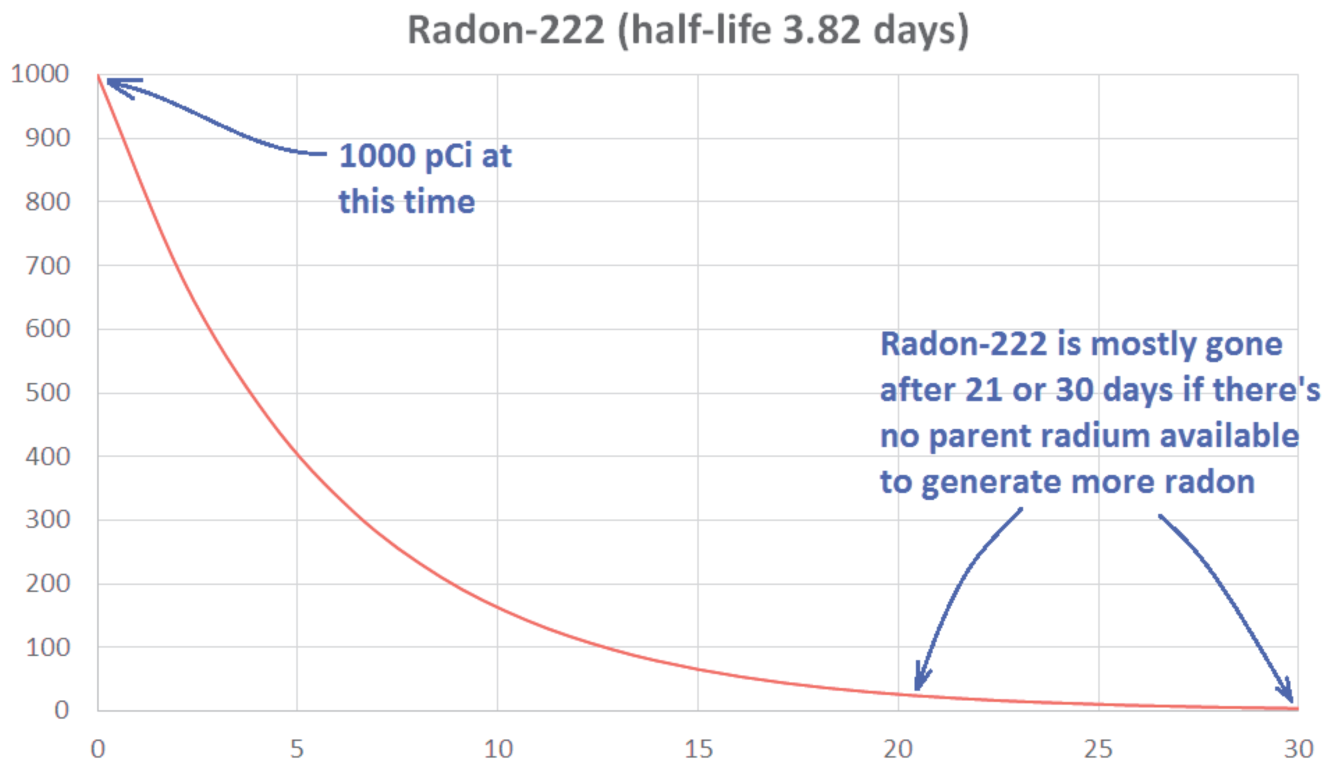
And even if the progeny are initially absent, they'll be generated and "catch up" if the parent is put into a sealed container.

Secular Equilibrium – “catching up”



Source: ORISE

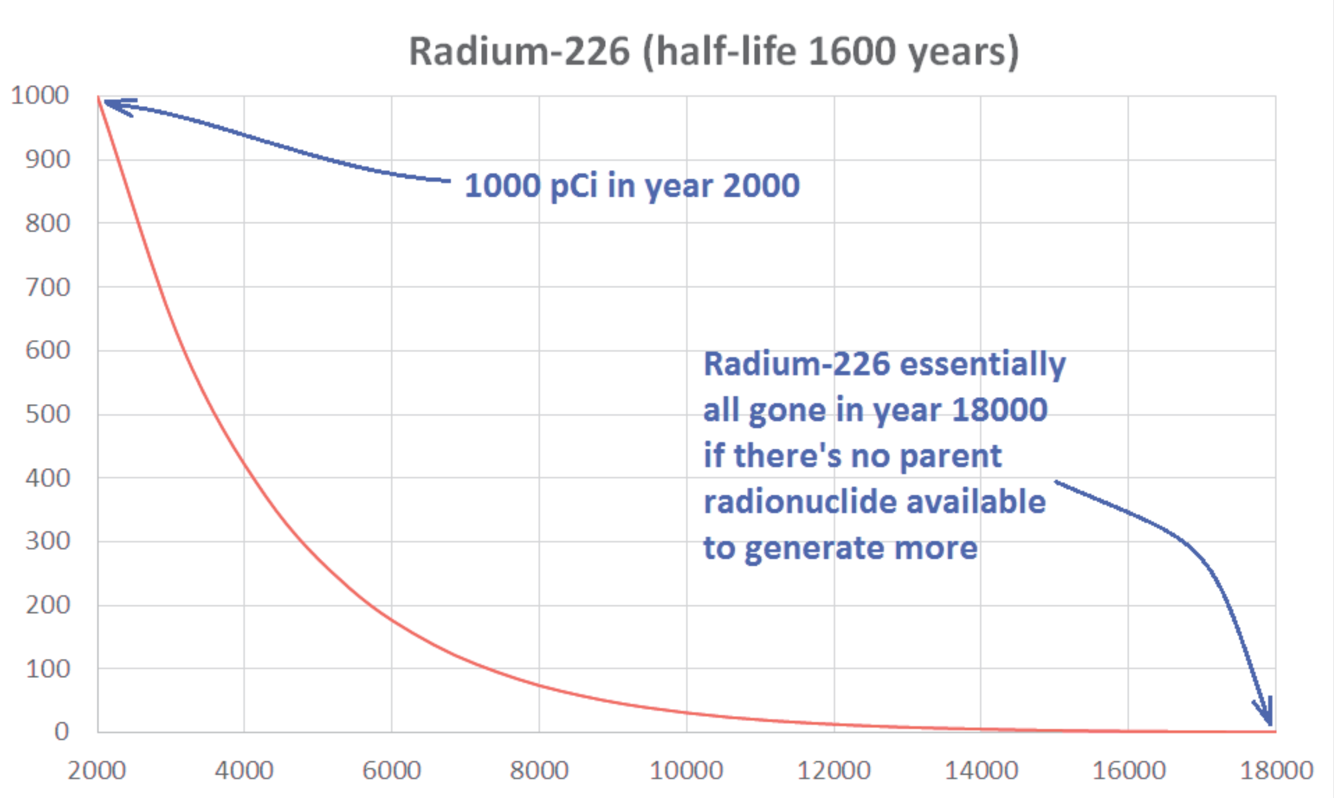
Radioactive decay without secular equilibrium with the parent



1000 pCi at this time

Radon-222 is mostly gone after 21 or 30 days if there's no parent radium available to generate more radon

Radioactive decay without secular equilibrium with the parent

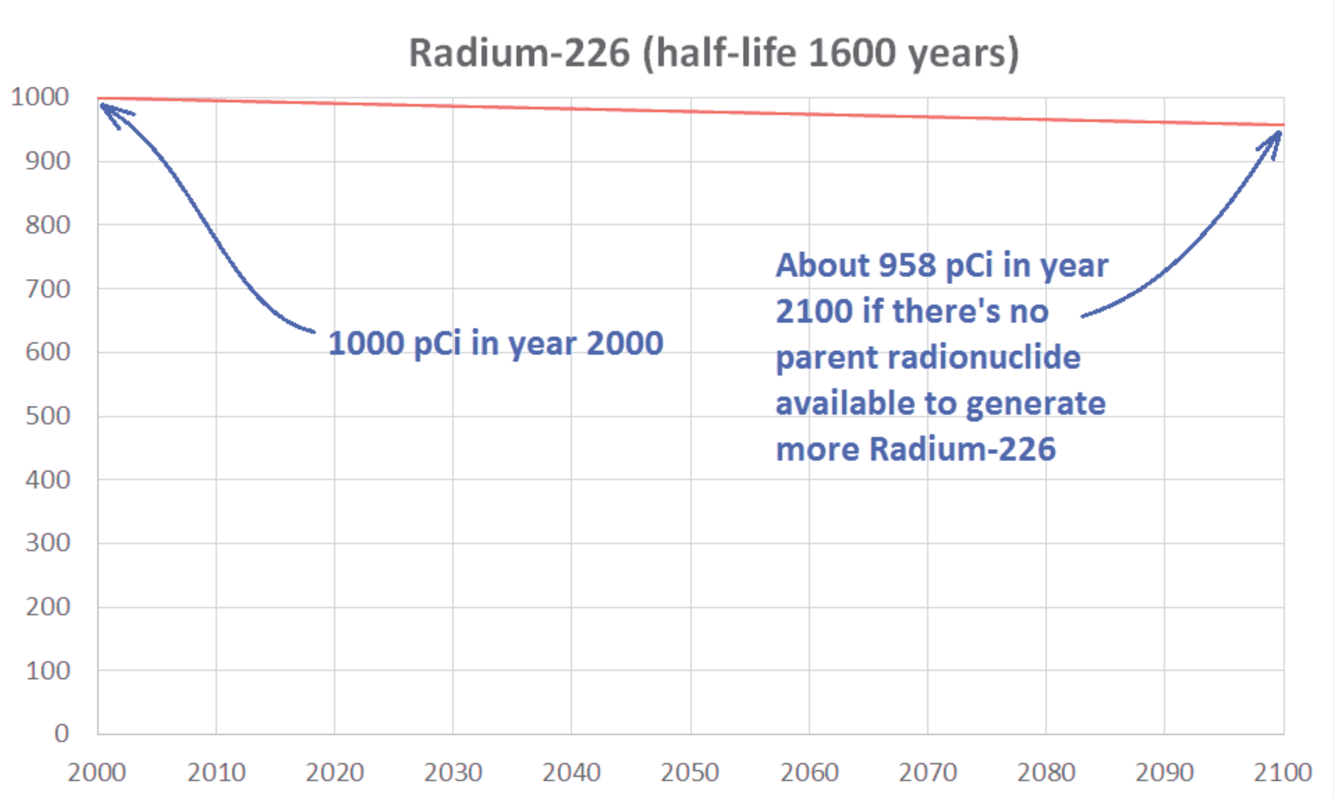


Radium-226 (half-life 1600 years)

1000 pCi in year 2000

Radium-226 essentially all gone in year 18000 if there's no parent radionuclide available to generate more

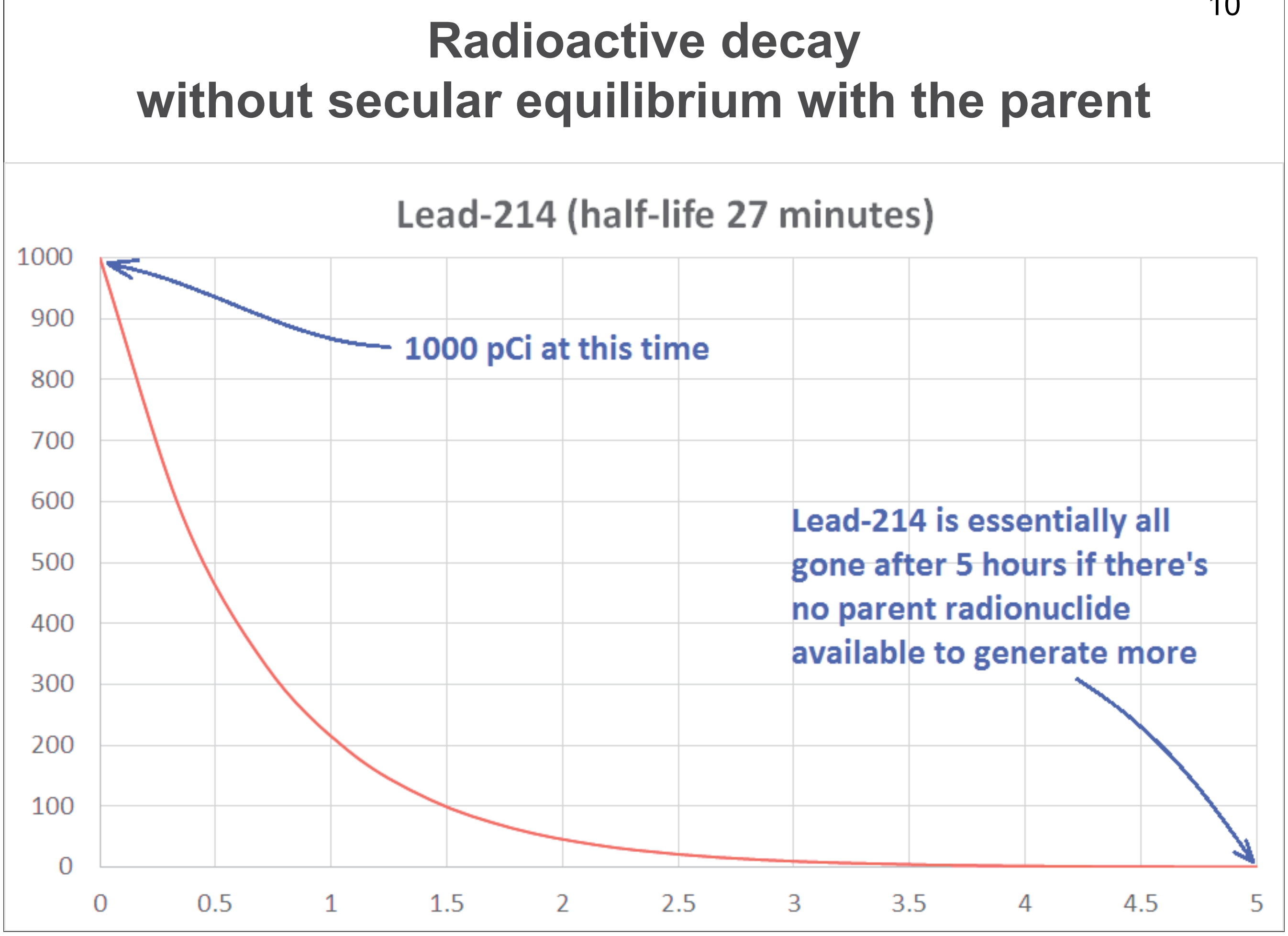
Radioactive decay without secular equilibrium with the parent



Radium-226 (half-life 1600 years)

1000 pCi in year 2000

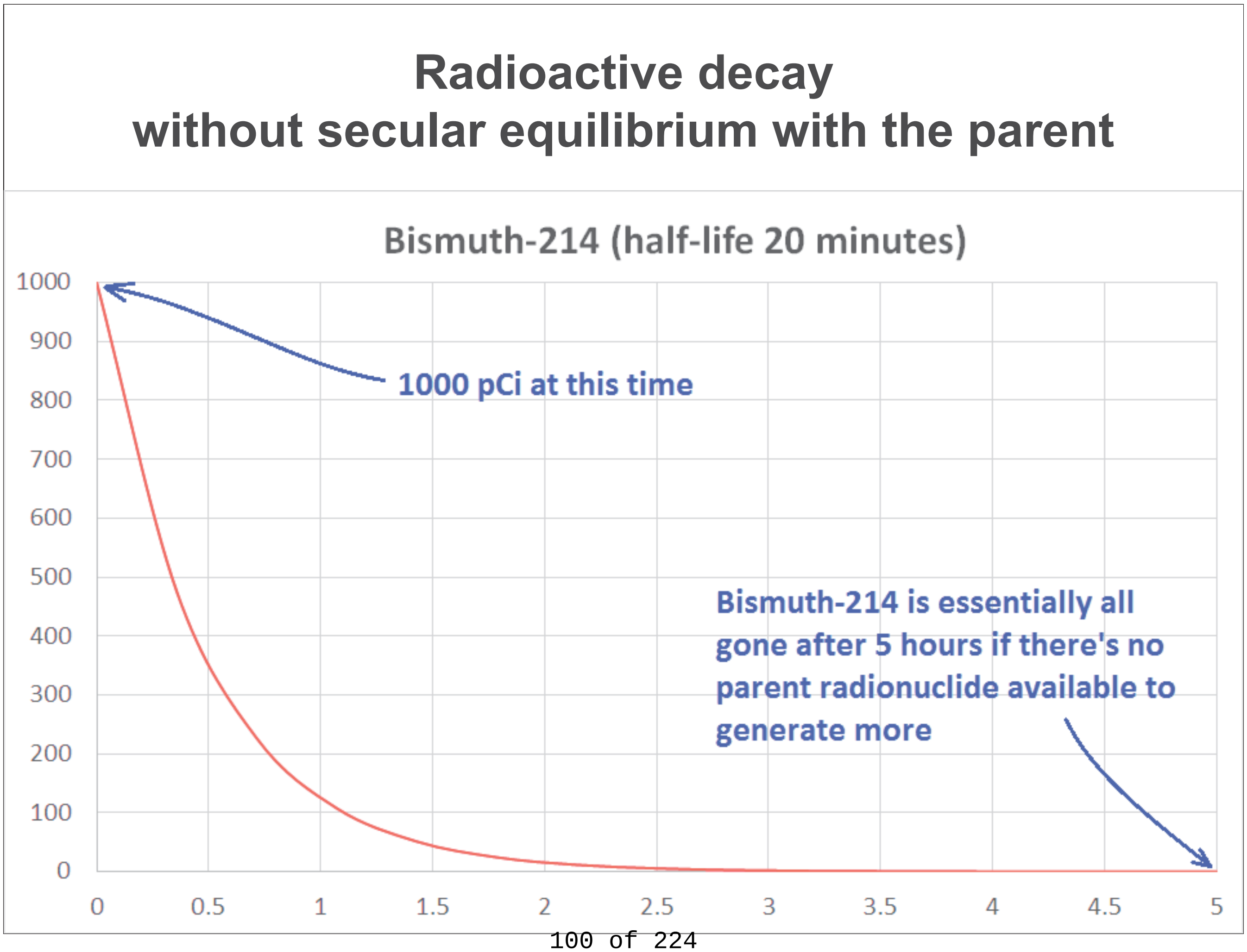
About 958 pCi in year 2100 if there's no parent radionuclide available to generate more Radium-226



Radioactive decay
without secular equilibrium with the parent

1000 pCi at this time

Lead-214 is essentially all gone after 5 hours if there's no parent radionuclide available to generate more



Radioactive decay
without secular equilibrium with the parent

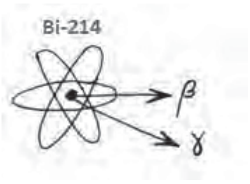
1000 pCi at this time

Bismuth-214 is essentially all gone after 5 hours if there's no parent radionuclide available to generate more

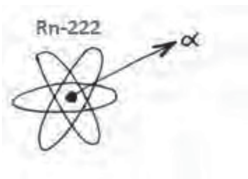
Lead-214 and Bismuth-214

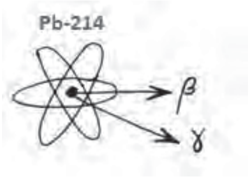
Both of these radionuclides come from radon decay; both have such short half-lives that they'll be essentially gone within 5 hours if not constantly regenerated by radon decay. THUS:

- **Any Lead-214 or Bismuth-214 measured in a sample must be less than about 5 hours old…**
- **Indicating approx. secular equilibrium among Lead-214, Bismuth-214, and parent Radon-222**
- **Meaning that *Radon-222 must be present in a sample at approximately the same activity* (in pCi) *as Lead-214 and Bismuth-214* if the sample is more than about 5 hours old**



***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…**



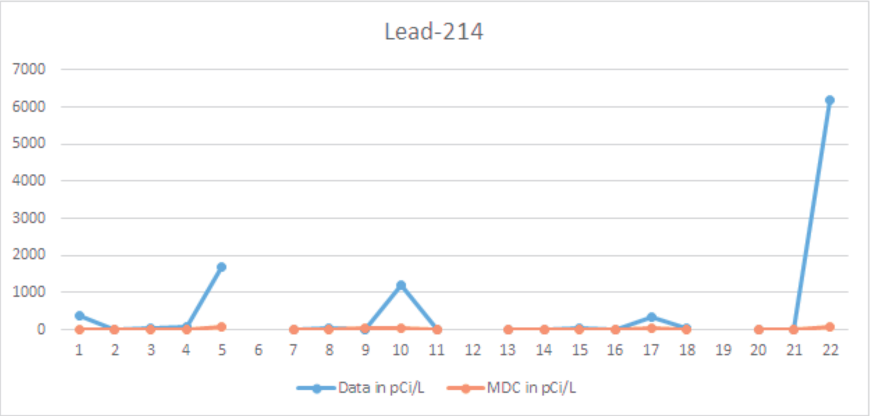


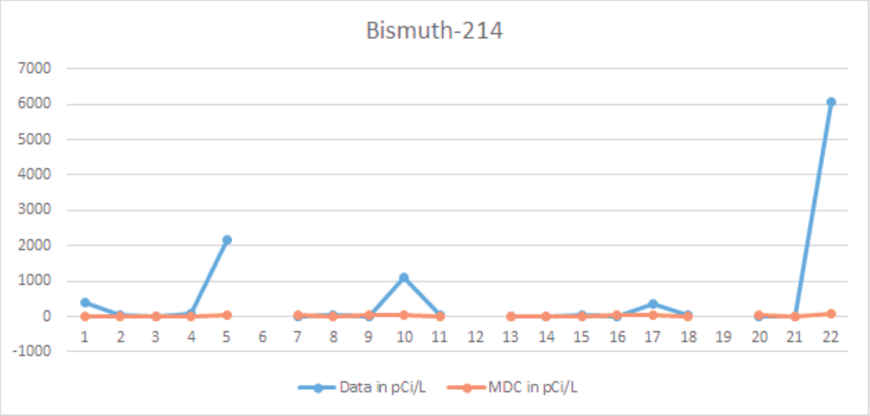
Applying these radiological principles to Hakes leachate test results

Lead-214, Bismuth-214, and Radon-222

Hakes leachate samples are generally not tested for Radon-222, but some of the test results show high levels of Lead-214 and Bismuth-214 (~6000 pCi/liter) in Hakes leachate samples at time of testing

Since these samples were held ~21 days before testing, the Lead-214 and Bismuth-214 results indicate that ~6000 pCi/liter Radon-222 was present in these Hakes leachate samples *at time of testing*

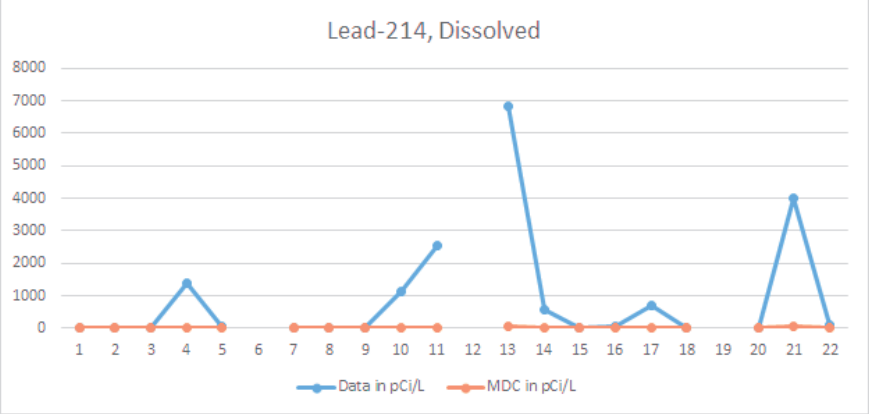


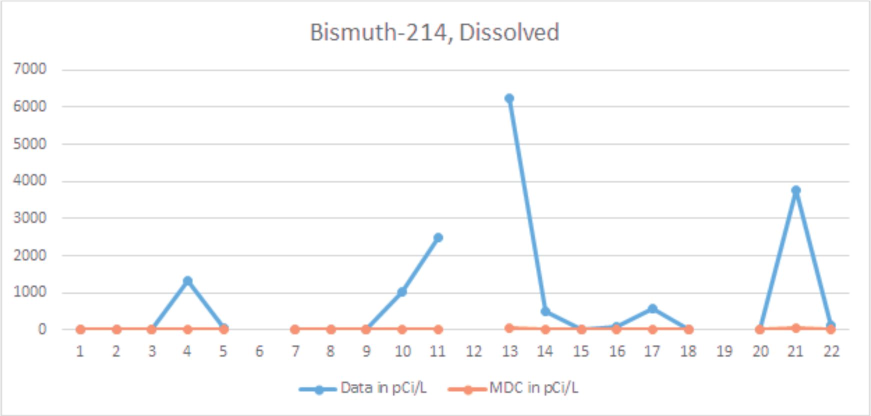


Hakes leachate test results (*in blue*) **and detection limits** (*orange*)

The horizontal axis on each graph is time, and the graphs show four different time trends:

- **1-5 are the 2015-17 time trend for Cell 3 Leachate**
- **7-11 are the 2015-17 time trend for Cell 4 Leachate**
- **13-18 are the 2014-17 time trend for Cell 5 Leachate**
- **20-22 are the 2016-17 time trend for Cell 8B Leachate**





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- **13-18 are the 2014-17 time trend for Cell 5 Leachate**
- **20-22 are the 2016-17 time trend for Cell 8B Leachate**

Lead-214, Bismuth-214, and Radon-222

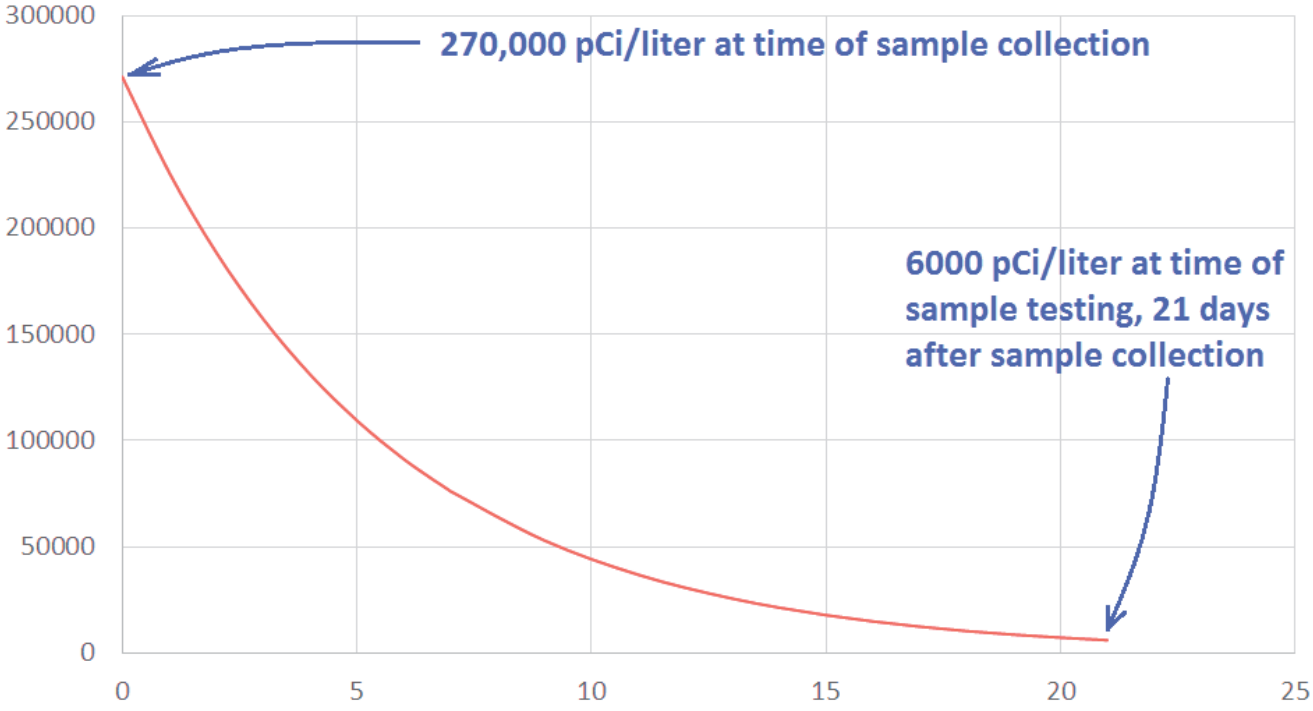
Hakes leachate tests show ~6000 pCi/liter Lead-214 and Bismuth-214, indicating ~6000 pCi/liter Radon-222 in leachate *at time of testing* – but not all samples show such high test results. WHY??

- **Radon level in leachate varies over time??**
- **Or radon level in leachate was relatively high when most/all samples were collected, but radon leaked out of many sample containers during the sample holding period of about 21 days?? (*This possibility is discussed in affidavit by our expert Dustin May*)**

Important points….

- If radon leaked out of some sample containers, does this mean that Hakes leachate *usually* (not just occasionally) contains high levels of *radon*?? Can't know without additional testing
- Tests show low levels (less than 10 pCi/liter) of *Radium-226* in Hakes leachate:
  - This *doesn't* show or mean that radium levels in landfill are low
  - This *does* show that the radium in leachate can't generate much radon (<10 pCi/liter)
- *Radon* activity in some samples was ~6000 pCi/liter *at time of testing – but much higher in leachate from which samples were collected*

21-day decay curve for Radon-222 (half-life 3.82 days) in Hakes leachate without secular equilibrium with parent radium



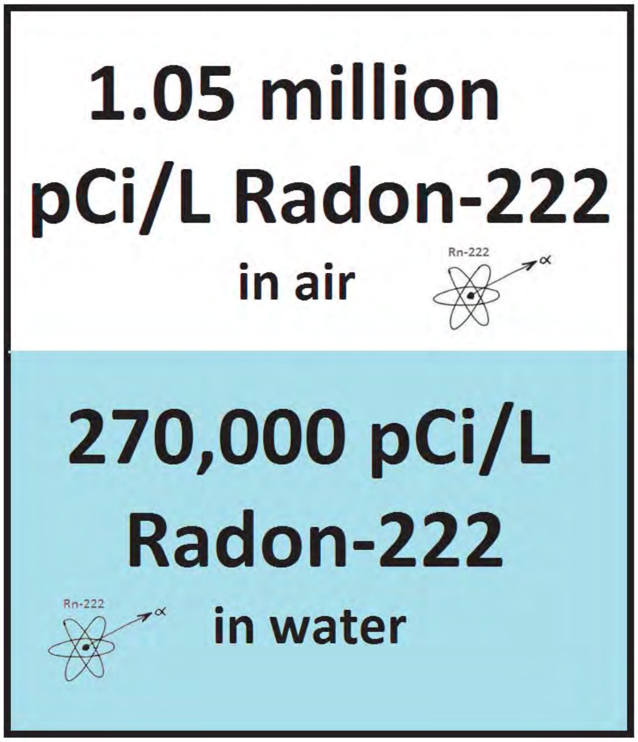
270,000 pCi/liter at time of sample collection

6000 pCi/liter at time of sample testing, 21 days after sample collection

Important points….

- Radon is a radioactive gas which, like other gases, can mix with air and can also dissolve in water and water-based mixtures such as leachate
- Radon activity in Hakes leachate from which samples were collected was (sometimes) ~270,000 pCi/liter
- 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 20ºC

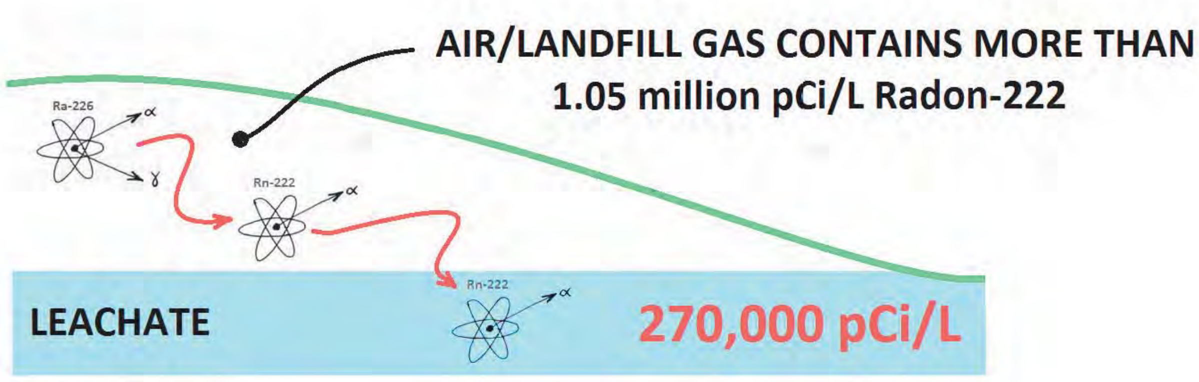


1.05 million pCi/L Radon-222 in air

270,000 pCi/L Radon-222 in water

Not at full equilibrium in an imperfectly sealed landfill, at ~20ºC

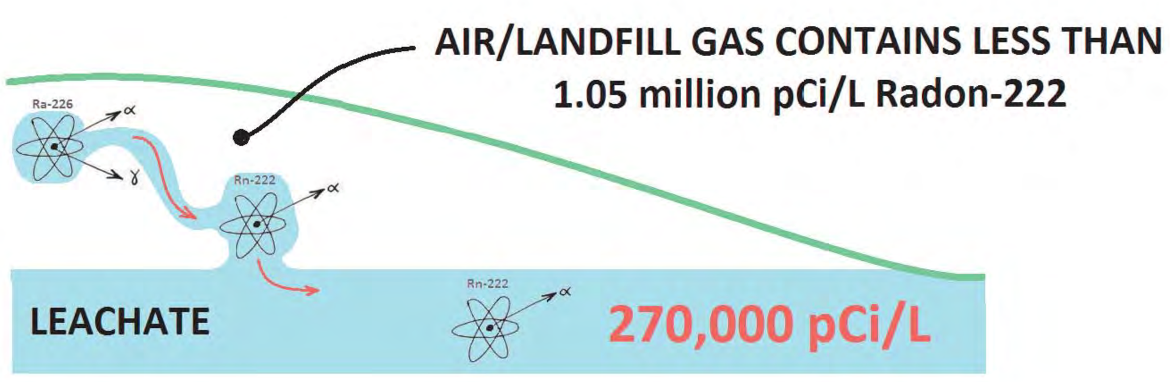
Likely: *Radium* remains dry
Radon reaches leachate by an air pathway
>1.05 million pCi/L radon in air/landfill gas



AIR/LANDFILL GAS CONTAINS MORE THAN
1.05 million pCi/L Radon-222

Not at full equilibrium in an imperfectly sealed landfill, at ~20ºC

Unlikely: *Radium* is immersed in water
Radon reaches leachate by water pathway
<1.05 million pCi/L radon in air/landfill gas



AIR/LANDFILL GAS CONTAINS LESS THAN
1.05 million pCi/L Radon-222

Important points….

- Radon activity in air/landfill gas within Hakes landfill may exceed ~1 million pCi/liter, either most of the time or part of the time. How much escapes through cap? *Testing needed.*
- Testing and air dispersion modeling need to be conducted to determine radon activity at downwind locations and *ensure that landfill radon emissions don't cause exceedances of residential indoor-air guidance* (4 pCi/liter)
- *Radon in leachate* also needs attention/testing
- Radon comes from radium decay – *how much radium is in Hakes landfill?*

Three Widely Used Air Dispersion Models

AERMOD ISCST3 CALPUFF	See also papers on radon dispersion modeling by Dong Xie (U. South China), K.J. Kearfott (U. Mich.) & others

===

"Most of the Atmospheric Background Radiation is caused by radon and its decay products…. The atmospheric background varies greatly with wind direction and meteorological conditions. Radon also can be released from the ground in bursts and then form 'radon clouds' capable of traveling tens of kilometers."

Gamma Spectrometry: Gamma Radionuclides and X Ray Spectrometry, Theremino System, Rev.2; http://www.theremino.com/wp-content/uploads/files/GammaSpec_ENG.pdf

Matt Richmond photo, http://archive.alleghenyfront.org

Applying similar radiological principles to truck monitoring at Hakes landfill gate

Radiation monitoring at Hakes landfill gate is intended to limit incoming waste loads to no more than 25 pCi/gram Radium-226

- As described in my affidavit, this type of monitoring cannot serve the intended purpose because highly variable and unknown levels of Lead-214 and Bismuth-214 interfere with Radium-226 monitoring

- Waste truckloads with up to 60-fold variations in their Radium-226 concentrations (activities) may exhibit the same or similar monitor readings

Radiation monitoring at landfill gate cannot reliably limit incoming waste loads to no more than 25 pCi/gram Radium-226

- **Even if waste truckloads with Radium-226 concentrations (activities) that are only *eight times* the 25 pCi/gram limit were able to enter the Hakes gate without triggering the monitor, this would still allow disposal of unprotectively high levels (200 pCi/gram) of Radium-226**
- **Compare this to the radium background level in typical local soil (about 1 pCi/gram)**
- **ALSO: the radium limit in soil for home & other building sites (5 pCi/gram above background)**

Conclusions

Many unknowns – testing needed

Exposure pathways from Hakes landfill to humans have not been clearly identified or adequately investigated – but high levels of radon within the landfill and its leachate may cause some level of human exposure at downwind locations. *What exposure level?*

Radium may also pose some level of long-term health risk for thousands of years if landfill integrity can't be guaranteed. *What risk level?*

Current unknowns and exposure pathways can and should be identified & quantified by testing and modeling – *preferably within an EIS process*

Questions?

Exhibit C

SIERRA
CLUB
ATLANTIC CHAPTER

January 14, 2019
VIA EMAIL
Town of Campbell
Town Board and Town Planning Board
8529 Main St
Campbell, New York 14821
E-mail: townclerk@campbellny.com

Re: Request for Opportunity to Respond to CoPhysics Report

Dear Town of Campbell Town Board and Town Planning Board:

I am writing to request that the Town of Campbell Planning Board postpone the vote on the application of the Hakes Landfill to establish a Non-Residential Planned Development District that has been scheduled for their meeting this Wednesday.

The Sierra Club applauds the Town for seeking more information regarding radioactivity issues than was provided in the Draft Supplemental Environmental Impact Statement on the proposed expansion of the landfill (DSEIS). We do not believe, however, that the May 2018 CoPhysics Report provided to the Town by Casella Waste Management satisfactorily addresses the concerns outlined in the comments we submitted on the DSEIS on March 19, 2018.

We have engaged Dr. Raymond Vaughan to respond to the CoPhysics Report. As you know, Dr. Vaughan prepared an expert affidavit on the radioactivity issues raised by the landfill's leachate test results that was provided to the Town and to DEC as an exhibit to our comments on the DSEIS. The CoPhysics Report is in large part a response to Dr. Vaughan's affidavit and the other expert affidavits we provided.

In the short time that has elapsed since the CoPhysics Report was made available to the public as an appendix to the Final Supplemental Environmental Impact Statement on December 5, 2018, a period that was filled with major holidays, Dr. Vaughan has not yet been able to complete his response. We urge the Town to take the time to understand our concerns with the CoPhysics Report and we respectfully request that you give us a month for Dr. Vaughan to complete his response so that we can submit it for your consideration before you vote on the Hakes rezoning application.

Respectfully yours,

Kate Bartholomew

Kate Bartholomew, Chair
Sierra Club Atlantic Chapter
ecogreenwolf@gmail.com

Exhibit D

SIERRA CLUB
ATLANTIC CHAPTER
744 Broadway ● Albany, NY 12207

February 21, 2019

VIA EMAIL

Jeffrey Horton, Supervisor
and Town Board
Town of Campbell
8529 Main Street
Campbell, New York 14821
E-mail: jjphorton@gmail.com, deputysupervisor@campbellny.com, tewheat1@yahoo.com, and townclerk@campbellny.com

Re: Comments on the Hakes Landfill Rezoning Application and FSEIS

Dear Supervisor Horton and Members of the Campbell Town Board:

The Sierra Club Atlantic Chapter, the Concerned Citizens of Allegany County, Inc. and People for a Healthy Environment, Inc. have reviewed the application of Hakes C&D Disposal Inc. to establish the site of its Hakes C&D Landfill located at 4376 Manning Ridge Road in the Town of Campbell as a Non-Residential Planned Development District (NRPDD) under the Town of Campbell Zoning Law and the Final Supplemental Environmental Impact Statement prepared for the expansion project (FSEIS) and we appreciate the opportunity to comment on these documents. We represent hundreds of residents living near and downstream from the Hakes landfill, the Chemung County landfill and the Hyland landfill in Allegany County.

The proposal to expand the size of the Hakes landfill brings to the forefront concerns that our members have about the environmental and health effects of radioactive shale gas drilling wastes going into solid waste and C&D landfills in New York. Our concerns are reflected in the thousands of comment letters our members and colleagues have emailed to Governor Cuomo and DEC Commissioners Seggos and Martens over the last six years asking DEC to ban radioactive gas drilling waste in New York landfills. At least fourteen county legislatures in New York have banned fracking wastes in landfills in their counties, in wastewater treatment facilities in their counties or in road-spreading in their counties. These counties include: Albany, Cayuga, Clinton, Erie, Nassau, Onondaga, Oneida, Orange, Putnam, Rockland, Suffolk, Tompkins, Ulster and Westchester. The Clinton County ban enacted in 2014 prohibits landfills and wastewater treatment plants in the county from accepting fracking waste, and bans the spreading of fracking waste on all roads within the county.[1]

You know the hazards of radioactive waste because your host agreement with the landfill bans the deposition of radioactive waste in the landfill. For this reason, we urge you not to proceed to approve Hakes' expansion plans until further study has been made of the evidence

---

[1] "Clinton County Passes New York's Most Comprehensive Fracking Waste Ban," Kathryn Rumbles, *Riverkeeper Blog,* August. 4, 2014, https://www.riverkeeper.org/blogs/dont-frack-with-new-york/clinton-county-passes-new-yorks-most-comprehensive-fracking-waste-ban/

showing extremely high levels of radon gas in the Hakes landfill leachate test results intermittently over time.

The Town has plenty of time to study the radioactivity issues presented by the landfill's leachate test results before the expansion project can proceed because DEC, the lead agency on the project, says it will delay issuing its findings statement on the FSEIS until it issues the necessary permits and it has not yet received the permit applications. This means that there is no urgency to the requested zoning approval.

We urge you to take advantage of this time, deny the rezoning application at this time and insist on more testing and analysis of radioactivity in the landfill before approving the NRPDD or issuing a findings statement on the FSEIS.

The landfill's leachate test results provide strong evidence of extremely high levels of radon gas in the Hakes landfill. Although the high levels appear intermittently in the leachate test results, for them to appear at all indicates that high levels of radon are being generated in the landfill at all times. In the exhibits to the Sierra Club's comment letter on the DSEIS, the Club's experts explain why the presence of high levels of the radionuclides Lead-214 and Bismuth-214 in the some of the test results demonstrates the presence of high levels of radium and radon in the landfill.[2]

The leachate evidence shows that the restrictions that have been put in place in the host agreement and the applicable DEC regulations have not been sufficient to limit the acceptance of radioactive waste at the landfill. Nor have the landfill's entrance monitors prevented significant amounts of radioactive waste from being deposited into the landfill.

The FSEIS and the CoPhysics report appended to the FSEIS fail to rebut the significance of the evidence of intermittently high levels of radon in the leachate test results. The fact that the levels of radon are intermittently high does not diminish the significance of these results. It may be evidence that some of the test samples were not properly sealed and radon gas was allowed to escape from the test samples. Or it may mean that the levels of radon gas in the landfill vary over time depending upon the opening and closing of various pathways of dispersal to the atmosphere. The test methodology used by the landfill's laboratory to test the leachate samples for Lead-214 and Bismuth-214 is a well-recognized testing methodology and provides reliable results. The fact that levels of Lead-110 were not tested does not diminish the significance of the levels of Lead-214 and Bismuth-214 that were tested. The fact that the geology of this area results in radon levels in home in the range of 4 pCi/liter does not plausibly explain levels of up to 1.05 million pCi/liter of radon in the air of the landfill as indicated by the landfill's leachate test results. A major problem with the entrance monitors is that loads of waste can be manipulated by allowing radon gas to escape before the loads pass through the monitors.

The deficiencies in the reasoning offered in the FSEIS and the CoPhysics report are addressed in detail in a memorandum prepared by Dr. Raymond Vaughan dated February 21, 2019. A copy of this memorandum is attached.

---

[2] See exhibits attached to the Sierra Club's comment letter on the DSEIS, dated March 19, 2018, a copy of which was submitted to the Town of Campbell.

As Dr. Vaughan explains, similar evidence of intermittently high levels of radon gas is shown in the leachate test results of the Chemung County landfill. It is imperative that further investigation be undertaken as to why the leachate test results of the two landfills in New York taking the greatest volume of drill cuttings from gas drilling operations in the Marcellus shale in Pennsylvania show intermittently high levels of radon breakdown products.

We urge you to insist on more testing and evaluation of radioactivity in the Hakes landfill before approving the NRPDD or issuing a findings statement on the FSEIS.

Respectfully,

Kate Bartholomew

Kate Bartholomew, Chair
Sierra Club Atlantic Chapter

Karen G Ash

Karen Ash, Chair
Concerned Citizens of Allegany County

Gary McCaslin

Gary McCaslin, President
People for a Healthy Environment, Inc.

Attachment

Cc:

Basil Seggos
Commissioner
New York State Department of Environmental Conservation
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Albany, NY 12233-1011

Dr. Howard Zucker
Commissioner
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Division of Environmental Permits
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RAYMOND C. VAUGHAN, PH.D., P.G.
Professional Geologist / Environmental Scientist
534 Delaware Ave., Suite 302, Buffalo, NY 14202
(716) 332-7113 • rcvaughan9@gmail.com

From: Raymond C. Vaughan, Ph.D., P.G.

To: Sierra Club Atlantic Chapter

Subject: Hakes FSEIS does not rebut the evidence presented by Sierra Club

Date: February 21, 2019

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Introduction

This memorandum addresses the points made regarding radioactivity issues in the responses to public comments by both the NYS Department of Environmental Conservation (DEC) and the Town of Campbell in the Final Supplemental Environmental Impact Statement for the Hakes C&D Disposal - Landfill Expansion Project, dated December 5, 2018 (the "FSEIS"). The responses of the Town of Campbell include a report prepared for the town by Theodore E. Rahon, Ph.D., Certified Health Physicist, CoPhysics Corporation, titled *Report: A Review of Drill Cuttings Disposal at the Hakes C&D Landfill and Response to Public Comment*, dated May 16, 2018, and attached to the Town of Campbell response to comments, Appendix 5 to the FSEIS (the "CoPhysics Report"). Responses from both DEC and CoPhysics are addressed here. The Town of Campbell has not provided independent responses on radioactivity issues but, instead, has referred to the DEC and CoPhysics responses.

In particular, this memorandum addresses whether DEC and CoPhysics satisfactorily respond to the points made in my affidavit of January 18, 2018, my presentation of February 10, 2018, and the comments on the Draft Supplemental Environmental Impact Statement ("DSEIS") that the Sierra Club Atlantic Chapter submitted to DEC and the Town of Campbell on March 19, 2018 (the "Sierra Club comment letter"). My affidavit and presentation were attached as exhibits to the Sierra Club comment letter.

I. The evidence shows that Hakes Landfill contains radioactive material, and that this radioactive material is poorly characterized

The evidence of radioactive material in the landfill comes from the landfill's leachate test results which show intermittently high levels of certain radionuclides (known as "radium progeny") that are produced by the radioactive decay of radium. The overarching issue is whether the leachate test results combined with the unreliability of the entrance monitors indicate that the Hakes landfill contains unacknowledged radioactive waste (particularly radium) that substantially exceeds the landfill's regulatory limit of 25 picocuries per gram (pCi/g). If so, the long-term health issues from such disposal need to be addressed.

Evidence that the radioactive material in the landfill is poorly characterized includes a major discrepancy between a) the radium levels detected by the landfill's entrance monitor which my January 18, 2018 affidavit shows is unreliable and b) the high levels of radium progeny in the leachate and landfill gas. Additional evidence that the radioactive material in the landfill is poorly characterized is provided by the fact that the levels of radium progeny in leachate are *intermittently* very high, varying by orders of magnitude from one test to another, for reasons that are neither explained nor understood. The issues that must be resolved are:

- why the radon levels are intermittently very high,
- the radium source for the intermittently high radon levels, and
- the effects on human health from the presence and dispersal of intermittently high radon levels, and also from the radium itself.

DEC acknowledges in the FSEIS that *there is a major discrepancy between the radium that DEC and the landfill operator can account for and the high levels of radium progeny in the leachate and landfill gas*. In responding to a comment on the high levels of radium progeny in the leachate and landfill gas, DEC has said:

> Considering the limited amount of drill cuttings that have been accepted to date at the landfill, and the minimal values of Ra-226 present in those cuttings, there is no plausible manner in which such radon values in air or leachate can be caused by the drill cuttings present.

FSEIS at 26.

This is exactly the point at issue. DEC has attempted to dismiss the intermittently high radon levels by referencing the landfill's regulatory limit of 25 pCi/g and the ability of the landfill's entrance monitors to detect waste loads above the regulatory limit. DEC also asserts that there are "minimal values" of Radium-226 in the drilling wastes accepted for disposal. On the basis of these three points, DEC claims the test results showing high levels of radium progeny in the leachate and landfill gas must be either wrong or not attributable to the drilling wastes. However, the available scientific evidence shows that the test results are *not* wrong and not readily attributable to any other source than the wastes accepted from drilling operations, as explained in detail in my affidavit of January 18, 2018, my presentation of February 10, 2018, the Sierra Club comment letter dated March 19, 2018, and this memo. Additional testing and modeling must be conducted to adequately rebut/resolve the leachate test results.

A major component of DEC's argument is that the gamma-detector entrance monitors used at Hakes will reliably detect radioactive materials coming into the landfill. To the contrary, as explained in my affidavit, these entrance monitors cannot reliably detect and prevent entry of waste loads carrying more than 25 pCi/g radium due to poorly constrained disequilibrium between radium and radon.

DEC argues unsuccessfully against the landfill's leachate tests results and what they show. Levels of radium progeny Lead-214, Bismuth-214, and Radon-222 in landfill leachate are intermittently very high (with radon ranging up to about 270,000 picocuries per liter [pCi/L]), as known from strong supporting evidence and lack of contrary evidence. Recent Lead-210 tests are said to contradict the intermittently high results but are in fact irrelevant; they fall in the category of "lack of contrary evidence." The strong supporting evidence includes valid and internally consistent results from gamma spectroscopy/spectrometry leachate testing, combined with recognized decay-correction procedures, with error bounds for such test results and procedures being well-established. These well-supported results show intermittently high radon levels in landfill gas that may range up to about 1.05 million pCi/L or more. Such intermittently high radon levels are likely to have onsite and offsite effects, exposing landfill workers and downwind residents to some level of radiological dose and risk. Additional testing and modeling are needed to a) characterize and quantify such dose and risk, b) identify and characterize the flow pathways for radon and the extreme variations in test results for its progeny, and c) locate, characterize, and quantify the radium that is generating the intermittently high levels of radon and other progeny. As indicated below, the intermittently high results are problematic regardless of whether the radon is from naturally occurring onsite radium or from radium-bearing waste brought into the landfill.

A. Summary of evidence previously presented by Sierra Club of intermittently high levels of radioactivity in landfill's leachate test results[1]

The annual reports of the Hakes C&D landfill in the Town of Campbell, Steuben County, NY, show that the landfill, which began operation in 1989, has accepted certain drilling-related wastes from Pennsylvania oil & gas operations since about 2010. As explained in documents

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[1] See also section IV of this memo for a comment-by-comment review of the Sierra Club comment letter.

such as my January 18, 2018 affidavit on behalf of Sierra Club, *a major concern is that a substantial amount of the drilling-related waste in Hakes landfill exceeds the 25 pCi/g regulatory limit*. Construction and demolition debris (C&D) landfills in New York are not allowed to accept drilling-related waste that contains more than 25 pCi/g radium.

Hakes has been required to submit leachate samples for semiannual testing of radium and certain radium progeny, including Lead-214 and Bismuth-214, with results reported in pCi/L. Test results from *most* of the leachate samples have shown relatively low levels of these radionuclides, typically less than about 50 pCi/L for both Lead-214 and Bismuth-214 and less than about 4 pCi/L for Radium-226.

If all of the Hakes leachate test results were similarly low, there would be no reason to suspect that a substantial amount of the radium-bearing waste brought into the landfill exceeds the 25 pCi/g regulatory limit. In other words, there would be little or no scientific basis for such a concern if the leachate test results *always* showed less than about 50 pCi/L for both Lead-214 and Bismuth-214.

In fact, the Hakes leachate test results for Lead-214 and Bismuth-214 are *intermittently very high*, ranging far beyond 50 pCi/L to about 6000 pCi/L. These strange and unexplained test results create a justified concern that substantial amounts of the radium-bearing waste brought into the landfill have exceeded the 25 pCi/g regulatory limit. The scientific basis for this concern has been set forth in my affidavit of January 18, 2018, my presentation of February 10, 2018, and the Sierra Club comment letter dated March 19, 2018.

The same sources, as discussed herein, show two further implications of such leachate test results ranging up to ~6000 pCi/L. First, the evidence shows that intermittently high levels of radon have been present in Hakes leachate, ranging up to ~270,000 pCi/L at the time the leachate samples were collected. Second, it is likely that continually high or intermittently high levels of radon are/have been present in landfill gas at levels ranging up to about 1 million pCi/L.

The following figures from my January 18, 2018 affidavit show the intermittently high levels of Lead-214 and Bismuth-214 in the leachate, ranging up to ~6000 pCi/L, and the substantially lower levels of Radium-226 in the leachate, as reported in the semiannual test results for these three radionuclides.[2] Similar results, ranging up to ~1000 pCi/L, are shown by the data from the Chemung County landfill leachate tests. These results are included here because they illustrate that another landfill that has taken high levels of drill cuttings and other gas drilling wastes also manifests intermittently high levels of Lead-214 and Bismuth-214 in leachate.

Results from the Hakes and Chemung County leachate test reports are plotted below, where the horizontal axis on each graph is time, and the graphs show four different time trends. The blue

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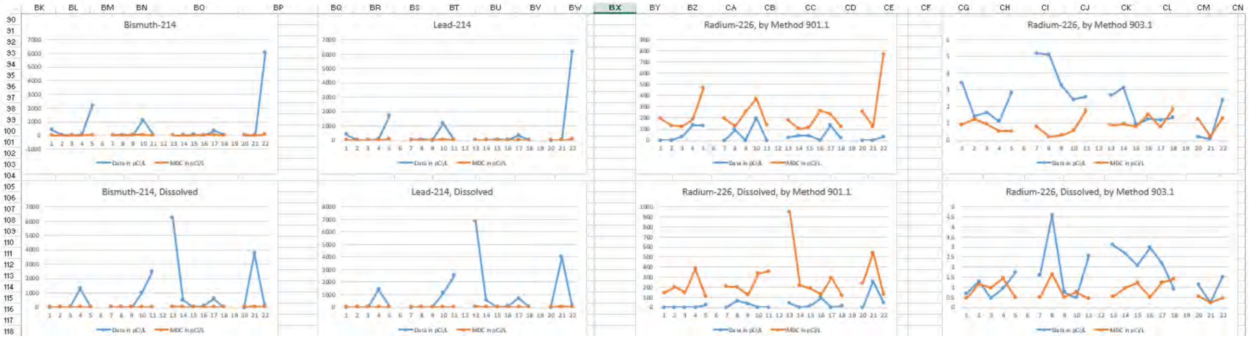
[2] Note that Radon-222 has not been routinely tested in these semiannual samples, but its concentration in a given sample can be determined from its parent-progeny relationships to Lead-214 and Bismuth-214. Nor was Lead-210 routinely tested prior to 2018, as discussed below.

lines show the reported test results, while the orange lines show the detection limit (MDC) for each test. See Exhibits S-Z of my January 18, 2018 affidavit for these graphs in larger format.

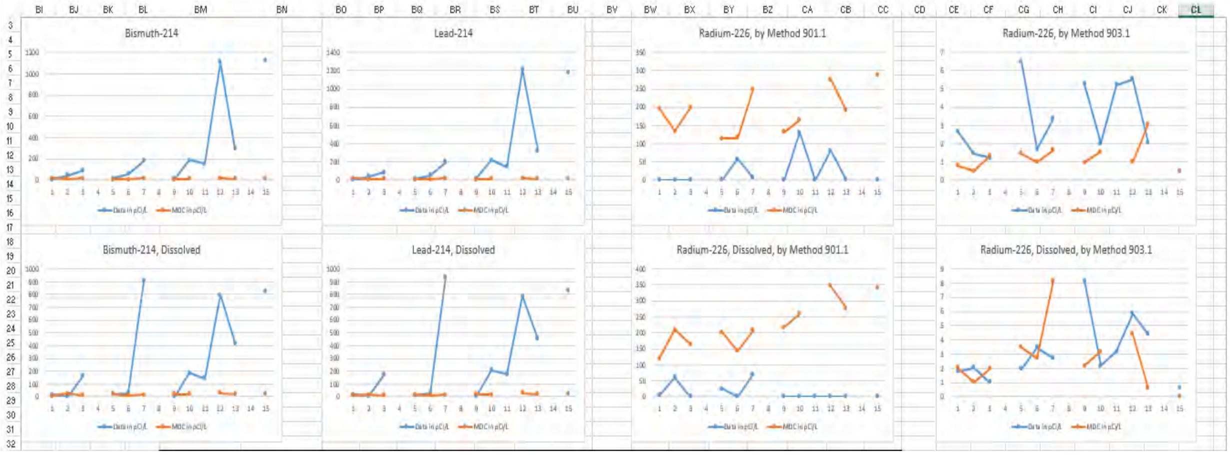
For Hakes Landfill:

- 1-5 are the 2015-2017 time trend for Cell 3 Leachate
- 7-11 are the 2015-2017 time trend for Cell 4 Leachate
- 13-18 are the 2014-2017 time trend for Cell 5 Leachate
- 20-22 are the 2016-2017 time trend for Cell 8B Leachate



For Chemung County Landfill:

- 1-3 are the 2015-2017 time trend for Leachate Pond (Combined Leachate)
- 5-7 are the 2015-2017 time trend for Cells I through III Primary Leachate
- 9-13 are the 2015-2017 time trend for Cell IV Primary Leachate
- 15 is the single data point for the 2017 measurement of Cell V Primary Leachate.



In summary, the above graphs show that the test results (blue lines) for Bismuth-214 and Lead-214 are intermittently very high, ranging up to about 6000 pCi/L in Hakes leachate and 1000 pCi/L in Chemung leachate, while the test results (blue lines) for radium in Hakes and Chemung leachate remain much lower.

B. Significance of this evidence

The evidence of intermittently high levels of radon in the landfill's leachate test results is significant because of two unresolved issues involving radon gas. One issue is the *effects* of radon in landfill gas, particularly human health effects, which have not been addressed. The other issue is the *source* of this radon. The source must be radium, but the unaddressed/unresolved issue is the quantity, origin, and location of radium within the landfill.

1. EFFECTS OF RADON: Part of the significance of the evidence is the likelihood of continually high or intermittently high radon in landfill gas, ranging up to ~1 million pCi/L, escaping into the atmosphere to an unknown extent and exposing humans and the environment to currently unmeasured radiological dose and thus risk.

2. SOURCE OF RADON: Part of the significance of the evidence is that the amount and location of *radium* capable of producing intermittently high radon levels within the landfill remain unknown. The existing evidence cannot resolve the question of whether such radium is mostly:

a) naturally occurring onsite radium (but if so, why are radon levels so intermittently high, and how do such large quantities of radon pass through the landfill liner into the leachate?), or

b) offsite radium brought onsite in radium-bearing wastes such as drill cuttings that *do not* exceed the 25 pCi/g limit (but if so, why are radon levels so intermittently high?), or

c) offsite radium brought onsite in radium-bearing wastes that *exceed* the 25 pCi/g limit.

Note that the effects and source may be interrelated. If the source is (c), offsite radium brought onsite in radium-bearing wastes that exceed 25 pCi/g, then *long-term health effects from such radium disposal* become increasingly significant.

II. DEC and CoPhysics do not rebut the evidence presented by Sierra Club

This section provides a detailed point-by-point review of the evidence and lack of substantive and credible rebuttal. See also section IV below for a comment-by-comment overview of the Sierra Club comment letter.

A. Recent Lead-210 test results do not rebut the earlier test results

The recent Lead-210 test results do not rebut the earlier test results showing intermittently high levels of radioactive material in the landfill. The recent Lead-210 tests cited by CoPhysics were performed on leachate samples in which Lead-214, Bismuth-214, and radon were *not* intermittently high. Such results cannot rebut earlier test results from leachate samples that were not tested for Lead-210 but showed high Lead-214, Bismuth-214, and radon.

This is one of the instances where CoPhysics and DEC have set up flawed arguments by which they seek to dismiss any concerns about the high radionuclide levels. In this instance the flaw is a failure to distinguish between *continuously high* and *intermittently high* levels of radium progeny such as Lead-214, Bismuth-214, and Radon-222. The CoPhysics argument relies on recent test data showing relatively low levels of Lead-210, Lead-214, and Bismuth-214. Based on the low test results for Lead-210, CoPhysics concludes that high levels of radium progeny such as Lead-214, Bismuth-214, and Radon-222, *if continuously high*, are impossible. Such a conclusion is irrelevant and invalid for the intermittently high levels that have been documented at both Hakes and Chemung landfills.

It is unfortunate that Lead-210 testing was not done on the earlier samples that contained high levels of Lead-214 and Bismuth-214. Such testing would have resolved most of the questions at issue here. The Lead-210 level in a given sample is necessarily correlated with the Lead-214 and Bismuth-214 levels through the parent-progeny relationships among these radionuclides.

B. The landfill’s own tests used a valid method to measure radioactivity in leachate samples that showed high results

EPA Method 901.1 is valid. Questions in the FSEIS about its validity are improper and misleading. In questioning the method, DEC and CoPhysics cast unfounded doubt on the validity of a well-known gamma spectroscopy/spectrometry test method that has been routinely used for radiological analysis of the leachate samples collected semiannually at Hakes and other landfills.

DEC and CoPhysics claim that gamma spectroscopy/spectrometry results obtained with EPA Method 901.1 are invalid or untrustworthy because the uncertainty associated with this method is too high. I have researched the documents summarized in Exhibit A in an effort to find any support for these claims. DEC’s recent revision of 6 NYCRR Parts 360-363 provides a few poorly explained clues and noticeable gaps (see Exhibit A), but nothing resembling a rational basis for distrusting Method 901.1 due to its alleged uncertainty.

DEC’s and CoPhysics’ complaints about “uncertainty” boil down to a simple fact that is well-known to testing labs and those who submit samples and review the results. Quite simply, when the activity (radioactivity) of a given radionuclide in a given sample is *higher* than the Minimum Detectable Concentration (MDC), then Method 901.1 is reliable and useful. When the activity of a given radionuclide in a given sample is *below* the Minimum Detectable Concentration, then Method 901.1 does not provide reliable or useful results because, in effect, “noise” overwhelms the signal. This is simple common sense that should not be distorted into a broad suspicion about the “uncertainty” of Method 901.1.

Note that "uncertainty" in this context doesn't have its everyday meaning of generalized doubt or unpredictability. "Uncertainty" is a well-defined numerical measure that is reported along with test results. It's simply the "plus or minus" value that accompanies many different types of measures, representing the outer limits of the likely true value of the measurement. Exhibit B, excerpted from a set of Hakes leachate results in which Lead-214 and Bismuth-214 are high (6/6/2017 Cell 8B), provides some examples. As seen in Exhibit B, results for Bismuth-214 and Lead-214 are both in the neighborhood of 6000 pCi/L, which is well above the Minimum Detectable Concentration (the MDC is in the neighborhood of 70 or 84). The uncertainty is high (plus or minus about 660 pCi/L), indicating that it's highly likely that the real result for either Bismuth-214 and Lead-214 is about 660 pCi/L higher than 6000 pCi/L, or about 660 pCi/L lower than 6000 pCi/L, or anywhere in between.[3] There would be no rational basis for rejecting these reported values (consisting of the combination of measured activity, uncertainty, and MDC) as "uncertain."

Similarly, the 11/18/2016 Hakes leachate results shown in Exhibit B for dissolved Bismuth-214 and dissolved Lead-214 are both in the neighborhood of 3900 pCi/L, which is well above the Minimum Detectable Concentration (the MDC is in the neighborhood of 49 or 59). The uncertainty is high (plus or minus about 420 pCi/L), so it's highly likely that the real result for either radionuclide is about 420 pCi/L higher than 3900 pCi/L, or about 420 pCi/L lower than 3900 pCi/L, or anywhere in between.[4] Here again, unless there is some unreported complication, these reported values (consisting of the combination of measured activity, uncertainty, and MDC) must be regarded as valid.

The other Method 901.1 test results shown in Exhibit B have measured activities that are variously reported as being above, about equal to, or below the MDC. Where the uncertainty is greater than the measured activity, the lower limit for the likely real activity will be a negative number – which isn't physically possible but reflects the difficulty (impossibility) of getting a realistic test result when the activity is below the MDC. As can be seen in Exhibit B, Method 903.1 provides somewhat better (i.e., more valid or trustworthy) results for the relatively low levels of radium found in the leachate samples. This is true simply because its MDC tends to be lower than the MDC for Method 901.1, making Method 903.1 a more suitable radium test at these low radium levels. However, if a substantially higher level of radium were present in one of these leachate samples, it would exceed the MDC and should show up "loud and clear" in the Method 901.1 test result. In this sense, all of the current Method 901.1 test results for radium in Hakes leachate provide a redundant safeguard (redundant with Method 903.1) that confirms the relatively low radium levels.

Returning to the DEC/CoPhysics allegations about uncertainty associated with Method 901.1, and likewise to the cloudy logic about Method 901.1 in DEC's recent revision of Parts 360-363 (see Exhibit A), the entire concern seems to revolve around test protocols for samples in which *radium* levels are below the Method 901.1 MDC. Granted, radium levels have tended to be

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[3] These values are expressed approximately for the sake of discussion. See Exhibit B and its source for the actual numbers under discussion here.

[4] See footnote 3.

below the Method 901.1 MDC, but this fact provides no sound basis for thinking that the test is dispensable because levels of radium – and other radionuclides – would always remain below the MDC. The excerpted test results in Exhibit B show why this is so. In a competent regulatory regime it's important to know when radionuclide levels in a sample are low, and also when they're high.

As noted above, there's always a remote possibility of an unreported complication that could render reported test results unreliable – but in a competent regulatory regime there's no reasonable basis for suspecting unreported complications. Laboratory certification programs such as the Environmental Laboratory Approval Program (ELAP) take proactive steps to avoid such complications. ELAP is New York's own codification of laboratory standards based on the National Environmental Laboratory Accreditation Conference (NELAC) standard.[5] It specifies that laboratory test results shall be reported accurately, clearly, unambiguously and objectively; that uncertainties shall be identified and reported; that periodic audits must evaluate whether there are any findings that cast doubt on the effectiveness of the operations or on the correctness or validity of the laboratory's environmental test results; and that a laboratory shall take timely corrective action and shall notify clients if investigations show that the laboratory results may have been affected. Relevant portions of the ELAP standards are quoted below:

> 3.10.1 General
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> The results of each test, or series of environmental tests carried out by the laboratory shall be reported accurately, clearly, unambiguously and objectively, and in accordance with any specific instructions in the environmental test....

ELAP Certification Manual (https://www.wadsworth.org/sites/default/files/WebDoc/1076921392/210.pdf), 5/6/08, page 40 of 69.

> 3.4.6 Estimation of Uncertainty of Measurement
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> 3.4.6.1 Environmental testing laboratories shall have and shall apply procedures for estimating uncertainty of measurement. In certain cases the nature of the test method may preclude rigorous, metrologically and statistically valid, calculation of uncertainty of measurement. In these cases the laboratory shall at least attempt to identify all the components of uncertainty and make a reasonable estimation, and shall ensure that the form of reporting of the result does not give a wrong impression of the uncertainty.

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[5] See the Environmental Laboratory Approval Program (ELAP) Certification Manual, online at https://www.wadsworth.org/sites/default/files/WebDoc/1076921392/210.pdf, which states that "The New York State Department of Health, Wadsworth Center, Environmental Laboratory Approval Program (ELAP) has adopted as its Quality System Standard the current version of the National Environmental Laboratory Accreditation Conference (NELAC) standard. This is Chapter 5 of the 2003 NELAC standards, and it is reproduced herein in an edited form." ELAP Certification Manual, 5/6/08, page 1 of 69.

> Reasonable estimation shall be based on knowledge of the performance of the method and on the measurement scope and shall make use of, for example, previous experience and validation data.
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> In those cases where a well-recognized test method specifies limits to the values of the major sources of uncertainty of measurement and specifies the form of presentation of calculated results, the laboratory is considered to have satisfied this clause by following the test method and reporting instructions (see 3.10).
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> 3.4.6.2 When estimating the uncertainty of measurement, all uncertainty components which are of importance in the given situation shall be taken into account using appropriate methods of analysis.

Id., page 26 of 69.

> 2.13 Internal Audits
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> 2.13.1 The laboratory shall periodically, in accordance with a predetermined schedule and procedure, and at least annually, conduct internal audits of its activities to verify that its operations continue to comply with the requirements of the quality system and this Standard. The internal audit program shall address all elements of the quality system, including the environmental testing activities. It is the responsibility of the quality manager to plan and organize audits as required by the schedule and requested by management. Such audits shall be carried out by trained and qualified personnel who are, wherever resources permit, independent of the activity to be audited. Personnel shall not audit their own activities, except when it can be demonstrated that an effective audit will be carried out.
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> 2.13.2 When audit findings cast doubt on the effectiveness of the operations or on the correctness or validity of the laboratory's environmental test results, the laboratory shall take timely corrective action and shall notify clients, in writing, if the investigations show that the laboratory results may have been affected.
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> The laboratory shall notify clients promptly, in writing, of any event such as the identification of defective measuring or test equipment that casts doubt on the validity of results given in any test report or test certificate or amendment to a report or certificate.

Id., page 17 of 69.

Under this protocol, test results and associated uncertainties must be clearly identified and accurately reported. If there is doubt about the validity of results, test labs are required to investigate, take corrective action, and provide notification of any laboratory results that may have been affected. The applicability of this protocol to landfills in New York is expressed in 6 NYCRR 363-4.6(g)(4)(i), which requires that "Laboratory analyses must be performed by a laboratory currently certified under the appropriate approval categories by the New York State Department of Health's Environmental Laboratory Approval Program (ELAP)." There is no place within such a protocol for vague, undocumented assertions about "uncertainty" of laboratory test results.

Another misstatement by CoPhysics is that "The analysis method (EPA 901.1) used for leachate analysis in the past (and for the lead-214 and bismuth-214 values that are at issue here) is a soil analysis method and, when used to analyze a water sample, produces very inconsistent and possibly erroneous results." (CoPhysics Report at 30.) This is an incorrect statement. EPA Method 901.1 is clearly an analysis method for water. See pp. 21-25 of the EPA report entitled *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, EPA-600/4-80-032, August 1980.[6] This report is cited as an authoritative source in current NYS landfill regulations (6 NYCRR 360.3(b)(5) and footnote 14 of 6 NYCRR 363-4.6); hence DEC should be well aware of the report and the fact that Method 901.1 is described therein as an analysis method for water.

C. A valid method of back-calculation (decay-correction) has been used by Sierra Club's expert(s) to determine that the radon level in leachate has been intermittently as high as approx. 270,000 or 275,000 pCi/L radon

The method of calculating that radon in Hakes leachate has intermittently been as high as 270,000 or 275,000 pCi/L is valid and correct,[7] and the uncertainty associated with the method is very low and well-bounded. CoPhysics acknowledges that:

> It is true that two leachate samples collected from Hakes Cell #5 on 11/11/14 and Hakes Cell #8 on 6/6/17, measuring approximately 6000 pCi/L of Bismuth-214 and Lead-214 (radon progeny), are unusually high relative to other leachate samples. Taking the higher #5 values and back-calculating (decay-correcting) from the analysis time to the time of collection results in an approximate Bismuth-214, Lead-214, and Radon-222 concentration of 275,000 pCi/L, which sounds like a very high value to a layman....

CoPhysics Report at 29.

CoPhysics proceeds to make various explanations that are incorrect, misleading, and/or irrelevant.

First, CoPhysics claims that "Past leachate sampling and analysis methods were never designed to be used for radon assessment." (Id.) This claim about the purpose of past samples ("never designed to be used for radon assessment") is not supported by the Project Narrative from one of the past tests. The Project Narrative uses the word "ingrowth," referring to creation of Lead-214 and Bismuth-214 from radon decay within the sample.[8] In any case, it does not matter whether

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[6] The report is posted on the EPA website and may be accessed at https://tinyurl.com/ydg2rtqg or https://nepis.epa.gov/Exe/ZyPDF.cgi/30000QHM.PDF?Dockey=30000QHM.PDF.

[7] My January 18, 2018 affidavit used 270,000 pCi/L as an approximation. CoPhysics uses 275,000 pCi/L as an approximation. Either value serves the purpose of discussion.

[8] The word "ingrowth," referring to creation of Lead-214 and Bismuth-214 from radon decay, was applied to at least one set of past leachate samples submitted for analysis. The Pace Analytical "Project Narrative" sheet dated Nov. 25, 2014 for Method 901.1 analysis of the Hakes leachate samples collected

past samples were collected and analyzed for the specific purpose of assessing radon; they are well-suited for this purpose. Prior to 2018, the Hakes leachate sampling and analysis methods did not test for either radon or Lead-210. Despite the absence of radon data*, it is possible and scientifically valid* (see further discussion below) *to back-calculate or decay-correct from the Lead-214 and Bismuth-214 data to determine the Radon-222 concentration in leachate at the time of sample collection*. The absence of Lead-210 data is unfortunate because such data would have been very useful for double-checking the back-calculated (decay-corrected) radon concentrations such as 270,000 or 275,000 pCi/L – but the back-calculation/decay-correction method is entirely correct regardless of whether it can be double-checked by Lead-210 data. At best, the claim that the leachate test results were "never designed" for radon assessment by back-calculation/decay-correction is irrelevant and misleading.

Next, CoPhysics claims (id.) to "have discussed these unusual results [back-calculation/decay-correction of past leachate sampling and analysis methods for the purpose of radon assessment] with the manager of the analysis laboratory. He believes there is so much uncertainty in this type of analysis that, to make a decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels. So, the 275,000 pCi/L calculation cannot be relied upon as an accurate estimation of radon and progeny in the original on-site samples."

The claims that "there is so much uncertainty in this type of analysis that, to make a decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels" and that "the 275,000 pCi/L calculation cannot be relied upon as an accurate estimation of radon and progeny in the original on-site samples" are both incorrect, as explained below.

Here is why these claims are incorrect. As a preliminary matter, it is reasonable to make three assumptions. These assumptions, apparently undisputed, are a necessary part of the foundation for back-calculation (decay-correction) of radon activity within the sealed sample container:

- First, the test results reported semiannually through 2017 for the Hakes leachate samples, including the radium test results[9] and the ~6000 pCi/L results for Bismuth-214 and Lead-214 in two samples (Hakes Cell #5 on 11/11/14 and Hakes Cell #8 on 6/6/17), are *reliable measures within the uncertainty values reported by the lab*. One indication of

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Nov. 11, 2014, refers to the samples as "901.1 Gamma Spec INGROWTH." This terminology implies that the purpose was to understand radionuclide ingrowth during the sample holding period. Such a purpose is inseparable from radon assessment during the sample holding period, including the endpoints of that period (the sample collection date and testing date).

[9] My January 18, 2018 affidavit at ¶¶ 36-41 expresses the possibility that the radium measurements in leachate samples may be in error. While that possibility needs to be recognized as part of the logic presented there, the possibility appears remote in view of the consistently low radium measurements by both Method 901.1 and 903.1. Thus, the most likely possibility is that the reported results are accurate within the reported uncertainties; that the radium in leachate is low, as reported; that the Lead-214 and Bismuth-214 in leachate are intermittently high, as reported; and that radium elsewhere in the landfill (not in the leachate) remains unmeasured and unknown. This last conclusion ("unmeasured and unknown") is discussed and supported in this memo, in my January 2018 affidavit, etc.

the reliability of these high Lead-214 and Bismuth-214 results is the absence of any cautionary "flag" attached to these values in the analytical lab's report. Another is the mutual corroboration of the independent measurements of Lead-214 and Bismuth-214 in a given sample. When one of these radionuclides is high, the other is also high, as would be expected due to secular equilibrium in samples that had been sealed for 21 days.

- Second, in accordance with secular equilibrium, the activities of Radon-222, Lead-214, and Bismuth-214 are all approximately the same after 21 days in a sealed container. Consequently, the reported activities and associated uncertainties for Lead-214 and/or Bismuth-214 in a given sample can be used as reasonable approximations of the activity and associated uncertainty of Radon-222 in the same sample.

- Third, the activity and associated uncertainty for a given radionuclide can be converted with negligible error from pCi to other units such as mass or moles or number of atoms (nuclei) of that radionuclide, or can be converted back to pCi, in accordance with the radionuclide's specific activity and Avogadro's number. See especially the USGS report attached as Exhibit C, at 1.

In summary, the Lead-214 and Bismuth-214 lab results (~6000 pCi/L) and associated uncertainty values can be considered accurate, within the accepted meaning of "accurate," at the time of sample analysis.[10] In turn, these results and associated uncertainty values provide a very good approximation of the activity (alternatively expressed as mass, moles, or number of atoms) and the associated uncertainty of Radon-222, per unit volume of sample, *at the time of sample analysis*.

The next step (looking backward 21 days) is to determine the activity and associated uncertainty of Radon-222, per unit volume of sample, *at the time of sample collection*. In this process of back-calculating (decay-correcting) Radon-222 in a sealed container, there are no uncertainties due to sample counting and other measurement procedures. Such measurement-based uncertainties *have already been accounted for* in the uncertainty values that the lab reported along with the ~6000 pCi/L test results. Hence, for a sample held in a sealed container for 21 days, the only uncertainties in back-calculating/decay-correcting are 1) the purely statistical uncertainty of radioactive decay, based entirely on well-established equations, and 2) the uncertainty in the half-life of Radon-222 based on historic (not current test-specific) measurements. As explained below, both of these uncertainties are extremely small, well understood, and well-bounded. Thus, in the context of the Hakes samples, it is incorrect to say that "decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels" and that the resulting calculation "cannot be relied upon as an accurate estimation of radon and progeny..."

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[10] For an overview of what's meant by "accurate within" the reported uncertainty, see USGS report, attached as Exhibit C, at 3-4.

The well-known statistical uncertainty of radioactive decay in a sealed container can be understood from the radioactive decay law:

$$N = N_0 \exp\left(\frac{-\ln(2)\,\Delta t}{T_{½}}\right) \qquad (1)$$

$$S.D. = \sqrt{N} \qquad (2)$$

where N_0 is the number of atoms (nuclei) of Radon-222 initially present at $t = 0$; N is the mean number of atoms (nuclei) of Radon-222 present after an interval Δt; and $T_{½}$ is the half-life of Radon-222. The standard deviation of N, designated *S.D.* in Equation (2), is the square root of N in accordance with the well-known relationship (variance = mean) in a Poisson process such as radioactive decay. The statistical uncertainty of radioactive decay of Radon-222 in a Hakes leachate sample that remains sealed for $\Delta t = 21$ days is thus expressed by *S.D.* in Equation (2), and this uncertainty, which incorporates the very small uncertainty in the half-life of Radon-222 ($T_{½} = 3.8232 \pm 0.0008$ days[11]), constitutes the only substantial uncertainty in back-calculating (decay-correcting) for $\Delta t = 21$ days. Ingrowth of radon from radium in the sample might also be accounted for – but, while this could readily be included in the present calculations, it is omitted here as a reasonable simplification because the test results consistently show a relatively small quantity of radium in the leachate samples, including those collected from Hakes Cell #5 on 11/11/14 and Hakes Cell #8 on 6/6/17.

The only remaining question in this uncertainty analysis is the number of Radon-222 atoms (nuclei) that were present in the Hakes leachate sample container, either at $t = 0$ or $t = 21$ days. Knowing either the value of N_0 or the value of N will serve essentially the same purpose since the two values are interrelated by Equation (1). The size of the sample container is not clearly specified in the Hakes leachate analytical reports but is apparently in the range of 250 mL (one-quarter liter) to one liter.[12] The sample container size is a variable that needs to be quantified (the values of N and N_0 depend on it), but, as will be shown below in Tables 2-3, the uncertainty in the back-calculation/decay-correction process is very low regardless of whether the sample container size is a fraction of a liter or a full liter.

Table 1 provides a simplified illustration of these relationships, using the approximate values from my January 18, 2018 affidavit (~270,000 pCi/L Radon-222 at time of sample collection in samples that contained ~6000 pCi/L Lead-214 and Bismuth-214 at time of sample testing), but note that Table 1 does not yet show any of the uncertainties. (Table 2 will address the uncertainties in detail.) Table 1 shows N and N_0 values (based on 6000 pCi/L and Equation (1)) for either 250-mL or 1-L sample containers. Tables 2-3 will provide numerical details for $\sqrt{N}$,

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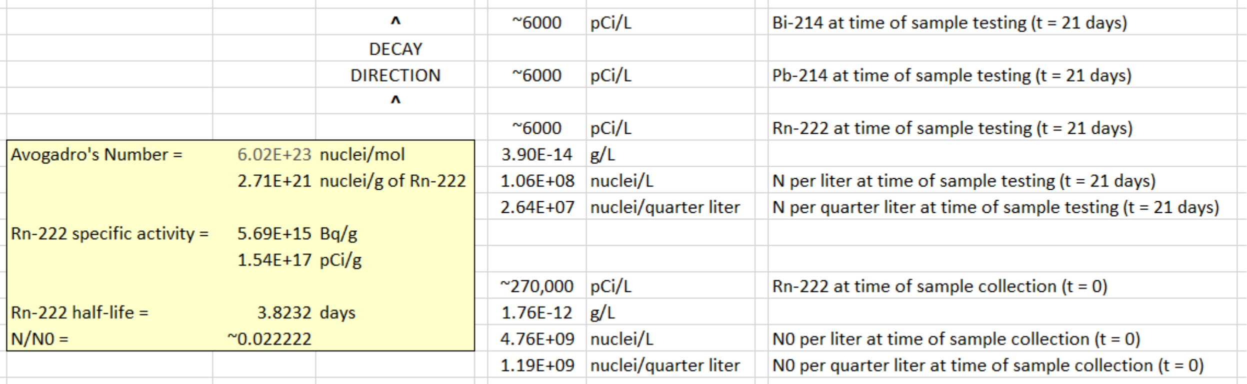
[11] Source: http://www.nucleide.org/DDEP_WG/Nuclides/Rn-222.lara.txt.

[12] In the analytical results reported for 4Q 2014 Hakes leachate, page 2 of the Pace Analytical "Sample Condition Upon Receipt" form indicates "Glass Jar," with 120 [mL] and 250 [mL] circled. The analytical results reported for 2Q 2017 Hakes leachate do not appear to identify sample volume but say that "12 mls of nitric acid were added to the sample to meet the sample preservation requirement of pH <2 for radiochemistry analysis" for the Cell 8B samples tested by Method 901.1 (and similarly for other 2Q 2017 samples), implying a sample volume that is a substantial fraction of a liter, based on an accepted practice of adding 15 mL of nitric acid per liter of sample.

the standard deviation of a radioactive decay process as defined in Equation (2), where $\sqrt{N}$ expresses the only substantial uncertainty in back-calculating/decay-correcting the activity of Radon-222 in a sealed sample container for $\Delta t = 21$ days.

Table 1: Approximate numerical relationships in samples with high Pb-214 and Bi-214



		^	~6000	pCi/L	Bi-214 at time of sample testing (t = 21 days)
		DECAY			
		DIRECTION	~6000	pCi/L	Pb-214 at time of sample testing (t = 21 days)
		^			
			~6000	pCi/L	Rn-222 at time of sample testing (t = 21 days)
Avogadro's Number =	6.02E+23	nuclei/mol	3.90E-14	g/L	
	2.71E+21	nuclei/g of Rn-222	1.06E+08	nuclei/L	N per liter at time of sample testing (t = 21 days)
			2.64E+07	nuclei/quarter liter	N per quarter liter at time of sample testing (t = 21 days)
Rn-222 specific activity =	5.69E+15	Bq/g			
	1.54E+17	pCi/g			
			~270,000	pCi/L	Rn-222 at time of sample collection (t = 0)
Rn-222 half-life =	3.8232	days	1.76E-12	g/L	
N/N0 =	~0.022222		4.76E+09	nuclei/L	N0 per liter at time of sample collection (t = 0)
			1.19E+09	nuclei/quarter liter	N0 per quarter liter at time of sample collection (t = 0)

Table 2 shows Pb-214 and Bi-214 results and associated uncertainties reported for Hakes Cell #5 leachate on 11/11/14 and Hakes Cell #8 leachate on 6/6/17. The Bi-214 values are shown only for comparison, while the Pb-214 values are used as the basis for the Radon-222 values.

In Table 2, the last four blocks of values show the decay relationships for various initial Radon-222 activities ranging from 248,183 to 340,665 pCi/L. These various initial values, representing Radon-222 activity in leachate at the time of sample collection ($t = 0$), are chosen such that their 21-day decay-corrected values (in bold) match the values (also in bold) that are known from the Lead-214 data. The decays calculated for the various initial values in Table 2 incorporate the quantified uncertainties in half-life and container size, so that these uncertainties are accounted in combination with the statistical decay-process uncertainty and with the analytical-test uncertainties shown at the top of the table. Table 2 thus shows the upper and lower bounds for Radon-222 activity in leachate at the time of sample collection (248,183 to 340,665 pCi/L overall, encompassing both test dates), based on the reported data and the four types of uncertainties that are accounted for in the table.

Table 2 is detailed and is the source of the values presented in Table 3.

Table 2: Numerical relationships based on reported Pb-214 data

11/11/2014			6/6/2017				
6221.7	±	670.09	6067.2	±	653.72	pCi/L	Bi-214 at time of sample testing (t = 21 days), as reported
6819.9	±	738.23	6183.9	±	666.77	pCi/L	Pb-214 at time of sample testing (t = 21 days), as reported
6081.67	to	7558.13	5517.13	to	6850.67	pCi/L	Pb-214 at time of sample testing (t = 21 days), range
6081.67	to	**7558.13**	**5517.13**	to	**6850.67**	pCi/L	Rn-222 at time of sample testing (t = 21 days), range based on Pb-214
3.95E-14	to	4.91E-14	3.59E-14	to	4.45E-14	g/L	
1.07E+08	to	1.33E+08	9.73E+07	to	1.21E+08	nuclei/L	N per liter at time of sample testing (t = 21 days)
2.68E+07	to	3.33E+07	2.43E+07	to	3.02E+07	nuclei/quarter liter	N per quarter liter at time of sample testing (t = 21 days)
							Rn-222 half-life = 3.8232 ± 0.0008 days
							USING 3.8240-day Rn-222 HALF-LIFE, HENCE N/N0 = 0.022226:
273,607	to	340,093	248,208	to	308,261	pCi/L	Rn-222 at time of sample collection (t = 0)
1.78E-12	to	2.21E-12	1.61E-12	to	2.00E-12	g/L	
4.82E+09	to	6.00E+09	4.38E+09	to	5.44E+09	nuclei/L	N0 per liter at time of sample collection (t = 0)
1.07E+08	to	1.33E+08	9.73E+07	to	1.21E+08	nuclei/L	Calculated N per liter at time of sample testing (t = 21 days)
1.04E+04	to	1.15E+04	9.86E+03	to	1.10E+04	nuclei/L	S.D. of N per liter at time of sample testing (t = 21 days)*
1.07E+08		1.33E+08	9.73E+07		1.21E+08	nuclei/L	N ± S.D. per liter at time of sample testing (t = 21 days)**
3.95E-14		4.91E-14	3.59E-14		4.45E-14	g/L	Ditto expressed as g/L
6081.67		**7558.13**	**5517.13**		**6850.67**	pCi/L	Ditto expressed as pCi/L
273,581	to	340,123	248,183	to	308,289	pCi/L	Rn-222 at time of sample collection (t = 0)
1.78E-12	to	2.21E-12	1.61E-12	to	2.00E-12	g/L	
4.82E+09	to	6.00E+09	4.38E+09	to	5.44E+09	nuclei/L	N0 per liter at time of sample collection (t = 0)
1.21E+09	to	1.50E+09	1.09E+09	to	1.36E+09	nuclei/quarter liter	N0 per quarter liter at time of sample collection (t = 0)
2.68E+07	to	3.33E+07	2.43E+07	to	3.02E+07	nuclei/quarter liter	Calculated N per quarter liter at time of sample testing (t = 21 days)
5.18E+03	to	5.77E+03	4.93E+03	to	5.50E+03	nuclei/quarter liter	S.D. of N per quarter liter at time of sample testing (t = 21 days)*
2.68E+07		3.33E+07	2.43E+07		3.02E+07	nuclei/quarter liter	N ± S.D. per quarter liter at time of sample testing (t = 21 days)**
9.88E-15		1.23E-14	8.97E-15		1.11E-14	g/quarter liter	Ditto expressed as g per quarter liter
3.95E-14		4.91E-14	3.59E-14		4.45E-14	g/L	Ditto expressed as g/L
6081.67		**7558.13**	**5517.13**		**6850.67**	pCi/L	Ditto expressed as pCi/L
							USING 3.8224-day Rn-222 HALF-LIFE, HENCE N/N0 = 0.022190:
274,043	to	340,636	248,604	to	308,753	pCi/L	Rn-222 at time of sample collection (t = 0)
1.78E-12	to	2.21E-12	1.62E-12	to	2.01E-12	g/L	
4.83E+09	to	6.01E+09	4.38E+09	to	5.44E+09	nuclei/L	N0 per liter at time of sample collection (t = 0)
1.07E+08	to	1.33E+08	9.73E+07	to	1.21E+08	nuclei/L	Calculated N per liter at time of sample testing (t = 21 days)
1.04E+04	to	1.15E+04	9.86E+03	to	1.10E+04	nuclei/L	S.D. of N per liter at time of sample testing (t = 21 days)*
1.07E+08		1.33E+08	9.73E+07		1.21E+08	nuclei/L	N ± S.D. per liter at time of sample testing (t = 21 days)**
3.95E-14		4.91E-14	3.59E-14		4.45E-14	g/L	Ditto expressed as g/L
6081.67		**7558.13**	**5517.13**		**6850.67**	pCi/L	Ditto expressed as pCi/L
274,017	to	340,665	248,578	to	308,781	pCi/L	Rn-222 at time of sample collection (t = 0)
1.78E-12	to	2.21E-12	1.62E-12	to	2.01E-12	g/L	
4.83E+09	to	6.01E+09	4.38E+09	to	5.44E+09	nuclei/L	N0 per liter at time of sample collection (t = 0)
1.21E+09	to	1.50E+09	1.10E+09	to	1.36E+09	nuclei/quarter liter	N0 per quarter liter at time of sample collection (t = 0)
2.68E+07	to	3.33E+07	2.43E+07	to	3.02E+07	nuclei/quarter liter	Calculated N per quarter liter at time of sample testing (t = 21 days)
5.18E+03	to	5.77E+03	4.93E+03	to	5.50E+03	nuclei/quarter liter	S.D. of N per quarter liter at time of sample testing (t = 21 days)*
2.68E+07		3.33E+07	2.43E+07		3.02E+07	nuclei/quarter liter	N ± S.D. per quarter liter at time of sample testing (t = 21 days)**
9.88E-15		1.23E-14	8.97E-15		1.11E-14	g/quarter liter	Ditto expressed as g per quarter liter
3.95E-14		4.91E-14	3.59E-14		4.45E-14	g/L	Ditto expressed as g/L
6081.67		**7558.13**	**5517.13**		**6850.67**	pCi/L	Ditto expressed as pCi/L

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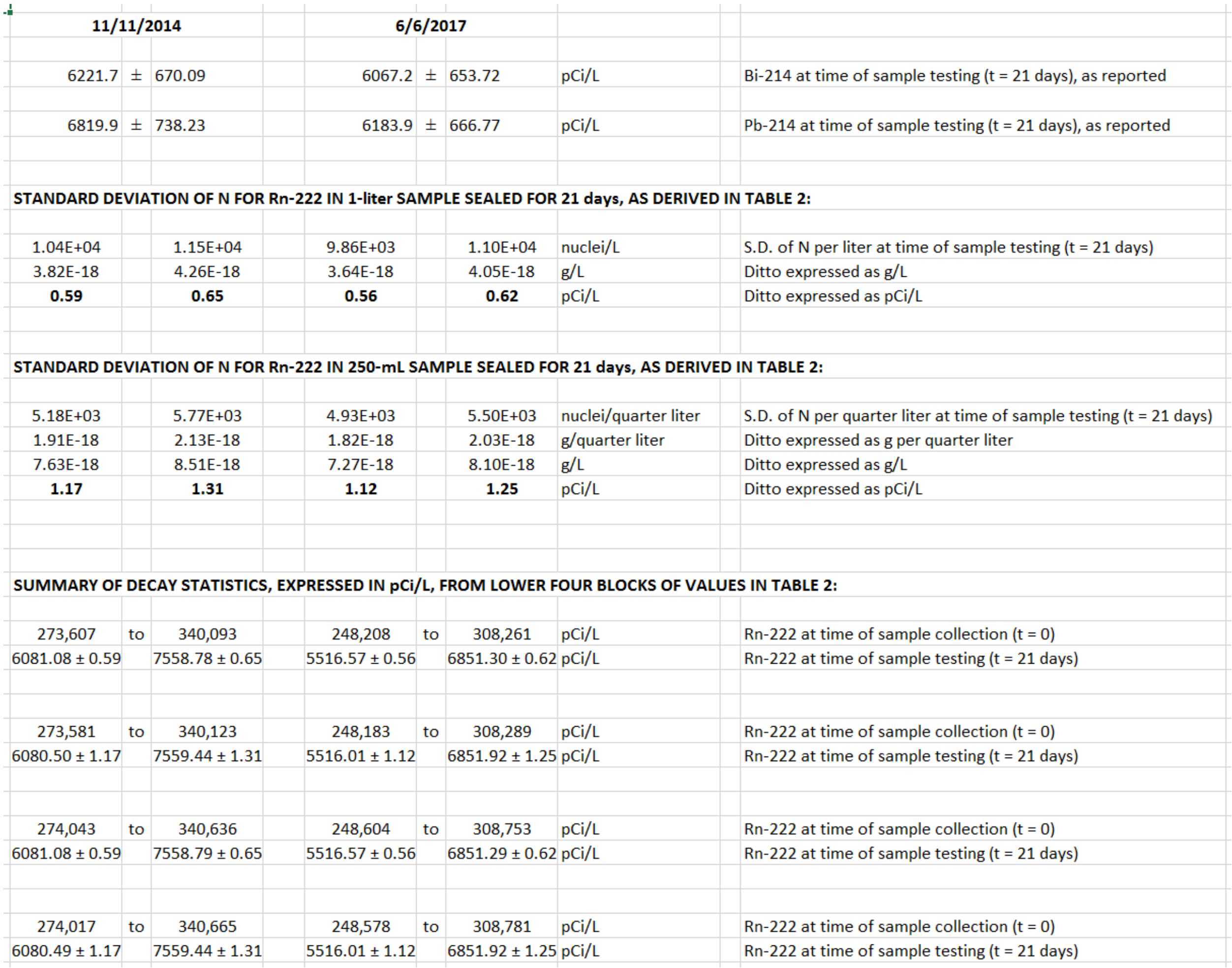
* Standard deviation (S.D.) of N is square root of N, in accordance with Equation (2)

** Values of N plus or minus S.D. are calculated as either plus or minus, whichever maximizes the range or spread of Rn-222 at time of sample collection

Table 3: Summary of 21-day decay statistics for Radon-222



11/11/2014			6/6/2017				
6221.7	±	670.09	6067.2	±	653.72	pCi/L	Bi-214 at time of sample testing (t = 21 days), as reported
6819.9	±	738.23	6183.9	±	666.77	pCi/L	Pb-214 at time of sample testing (t = 21 days), as reported
STANDARD DEVIATION OF N FOR Rn-222 IN 1-liter SAMPLE SEALED FOR 21 days, AS DERIVED IN TABLE 2:							
1.04E+04		1.15E+04	9.86E+03		1.10E+04	nuclei/L	S.D. of N per liter at time of sample testing (t = 21 days)
3.82E-18		4.26E-18	3.64E-18		4.05E-18	g/L	Ditto expressed as g/L
0.59		**0.65**	**0.56**		**0.62**	pCi/L	Ditto expressed as pCi/L
STANDARD DEVIATION OF N FOR Rn-222 IN 250-mL SAMPLE SEALED FOR 21 days, AS DERIVED IN TABLE 2:							
5.18E+03		5.77E+03	4.93E+03		5.50E+03	nuclei/quarter liter	S.D. of N per quarter liter at time of sample testing (t = 21 days)
1.91E-18		2.13E-18	1.82E-18		2.03E-18	g/quarter liter	Ditto expressed as g per quarter liter
7.63E-18		8.51E-18	7.27E-18		8.10E-18	g/L	Ditto expressed as g/L
1.17		**1.31**	**1.12**		**1.25**	pCi/L	Ditto expressed as pCi/L
SUMMARY OF DECAY STATISTICS, EXPRESSED IN pCi/L, FROM LOWER FOUR BLOCKS OF VALUES IN TABLE 2:							
273,607	to	340,093	248,208	to	308,261	pCi/L	Rn-222 at time of sample collection (t = 0)
6081.08 ± 0.59		7558.78 ± 0.65	5516.57 ± 0.56		6851.30 ± 0.62	pCi/L	Rn-222 at time of sample testing (t = 21 days)
273,581	to	340,123	248,183	to	308,289	pCi/L	Rn-222 at time of sample collection (t = 0)
6080.50 ± 1.17		7559.44 ± 1.31	5516.01 ± 1.12		6851.92 ± 1.25	pCi/L	Rn-222 at time of sample testing (t = 21 days)
274,043	to	340,636	248,604	to	308,753	pCi/L	Rn-222 at time of sample collection (t = 0)
6081.08 ± 0.59		7558.79 ± 0.65	5516.57 ± 0.56		6851.29 ± 0.62	pCi/L	Rn-222 at time of sample testing (t = 21 days)
274,017	to	340,665	248,578	to	308,781	pCi/L	Rn-222 at time of sample collection (t = 0)
6080.49 ± 1.17		7559.44 ± 1.31	5516.01 ± 1.12		6851.92 ± 1.25	pCi/L	Rn-222 at time of sample testing (t = 21 days)

Table 3 shows that the uncertainty (standard deviation) for the decay of Radon-222 in a sealed sample container for 21 days is on the order of 1 pCi/L – which is essentially negligible in this context. This statistical uncertainty is a consequence of the decay relationships expressed in Equations (1) and (2) which are well-established – beyond any reasonable doubt – for any such Poisson process that exhibits exponential decay. Thus, any claim that “decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels” or that the resulting calculation “cannot be relied upon as an accurate estimation of radon and progeny...” is entirely incorrect.

As can be seen from Tables 2 and 3, the simplified version of decay correction presented in my January 18, 2018 affidavit (see Table 1 above) is an *understatement* of the Radon-222 activity in the 11/11/14 Cell #5 and 6/6/17 Cell #8 leachate samples at the time of sample collection. As shown in Table 2, *the Radon-222 activity in these leachate samples may have been as high as 340,665 pCi/L or as low as 248,183 pCi/L*, depending on the uncertainties that are taken into account. There is no other credible interpretation of the reported Pb-214 and Bi-214 data for 11/11/14 Cell #5 and 6/6/17 Cell #8 leachate.

Tables 2 and 3 could be recalculated, as needed, with uncertainties representing greater than 1 sigma (e.g., 1.96 sigma). If this were done, it would expand the bounds for Radon-222 activity in leachate at the time of sample collection (currently 248,183 to 340,665 pCi/L overall, encompassing both test dates), thereby lowering the lower bound and raising the upper bound, while leaving the mean or central value essentially unchanged.

Uncertainties in the 21-day back-calculation/decay-correction process have been worked out numerically in the above paragraphs and Tables 2-3. The same result could be obtained from the principles of error propagation by applying the standard equation for a function R of variables X, Y, etc., where the uncertainty δR in R can be calculated from the uncertainties δX, δY,… in X, Y,…, respectively:

$$R = R(X, Y, \dots) \tag{3}$$

$$\delta R = \sqrt{\left(\frac{\partial R}{\partial X}\delta X\right)^2 + \left(\frac{\partial R}{\partial Y}\delta Y\right)^2 + \cdots} \tag{4}$$

The results of such error-propagation calculation would be the same as the numerically-derived results presented above.

In summary, ~270,000 pCi/L Radon-222 remains a very good and entirely supported approximation for Hakes Cell #5 leachate on 11/11/14 and Cell #8 leachate on 6/6/17. There are at least two reasons why such high radon activity in leachate can't be dismissed as a fluke or artifact. One is simply the mutual corroboration of Lead-214 and Bismuth-214 results: when one is high, the other is also high. The other reason is the relatively frequent occurrence of these sporadic "highs." The highest examples are of course from Cell #5 leachate on 11/11/14 and Cell #8 leachate on 6/6/17, but other mysteriously high test results for Lead-214 and Bismuth-214 include:

- ~3900 pCi/L from Cell #8B on 11/18/16 (implying ~175,000 pCi/L radon in leachate),
- ~2500 pCi/L from Cell #4 on 6/6/17 (implying ~112,000 pCi/L radon in leachate),
- ~1800 pCi/L from Cell #3 on 6/6/17 (implying ~81,000 pCi/L radon in leachate),
- and two examples of ~1000 pCi/L from *Chemung* landfill, as seen above on page 6.

All of these are far above the results normally reported for Lead-214 and Bismuth-214 in Hakes leachate samples. There is, however, a reasonable question of whether the normally low results for Lead-214 and Bismuth-214 are valid. The normally low results may not be valid (i.e., may not be representative of the sampled leachate and its 21-day ingrowth and decay) if the sample containers are not well sealed, allowing radon to leak out of some of the sample jars.

D. A valid method of calculation has been used by Sierra Club's expert(s) to determine that the radon level in landfill gas has likely been as high as approx. 1.05 million pCi/L radon

Here again, *the method is valid*. Specifically, the calculation that radon in landfill gas has likely been as high as ~1.05 million pCi/L is valid and correct.

As outlined in my January 18, 2018 affidavit, water that contains ~270,000 pCi/L dissolved radon is at equilibrium with an overlying air-radon mixture that contains ~1.05 million pCi/L radon. If leachate and landfill gas are reasonable analogs of water and air, then leachate that contains ~270,000 pCi/L dissolved radon is approximately at equilibrium with an overlying landfill gas mixture that contains ~1.05 million pCi/L radon.

If the radon in leachate is only intermittently high, then an equilibrium relationship cannot provide a simple answer about radon activity in overlying landfill gas and how it may vary over time – yet the equilibrium relationship does contribute to an important general truth about the concentration gradient needed to transfer radon across an interface between landfill gas and leachate.

The question is how a sufficient quantity of radon was able to dissolve in leachate to produce a concentration of ~270,000 pCi/L radon in the leachate.[13] This is a crucial question regardless of whether the level of dissolved radon remains continually high or is only intermittently at a level of ~270,000 pCi/L. There are apparently only two possible answers.

The most likely explanation is that the parent radium remains relatively "high and dry," immersed primarily in landfill gas rather than any hydrologically connected pool or stream of leachate, such that the ingrowth of radon occurs mainly within the landfill gas. In this case, radon must migrate across the landfill gas/leachate interface in order to dissolve into the leachate and reach a concentration of ~270,000 pCi/L. Such migration will occur only if there's a sufficient concentration gradient across the interface to make it happen, which means that the landfill gas at the interface must at least briefly contain more than about 1.05 million pCi/L radon in order to carry enough radon into the leachate to reach ~270,000 pCi/L dissolved radon. This must be the case, given the contradictions to the other answer. If so, there is a need for additional testing to characterize the source and migration of the radon, including tests to identify *how much radium is in the landfill* and *what the radon flow pathways are*.

The less likely explanation is that the parent radium is immersed in the leachate, such that the ingrowth of radon occurs within the leachate. This would not require any radon to migrate across an interface from landfill gas to leachate. Some radon would presumably migrate in the

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[13] As noted above, the activity of a given radionuclide can be converted from pCi to other units such as mass or moles or number of atoms (nuclei) of that radionuclide, or can be converted back to pCi, in accordance with the radionuclide's specific activity and Avogadro's number. In this manner, activity per unit volume can be readily converted to concentration and vice versa.

opposite direction by offgassing from the leachate into the landfill gas – at least intermittently when the leachate ranged up to ~270,000 pCi/L dissolved radon – resulting in some concentration of radon in the landfill gas, probably much less than ~1.05 million pCi/L. However, there are two factors that disfavor or contradict this possibility. One is that immersion of radium-bearing waste in leachate is contrary to good landfill practice; any radium should be relatively "high and dry" in the landfill.[14] The other contradiction is that leachate test results consistently show low levels of radium, strongly implying that the contact between leachate and radium (and/or its soluble compounds) is minimal, i.e., too low to account for radon levels ranging up to ~270,000 pCi/L in leachate. Arguments could conceivably be made that radon generated from small quantities of radium is somehow concentrating itself at certain points in space and time, but such arguments are usually not thermodynamically plausible.

E. The significance or physical interpretation of the fact that leachate test results are only intermittently high

There are three possible explanations for why the high levels of Lead-214, Bismuth-214, and radon are only intermittently high:

- Radon is continually high in Hakes leachate but is lost from poorly sealed sample jars,
- Radon in leachate is intermittently high due to *fluctuations in the radon flow path from radium to leachate* within the landfill, or
- Radon in leachate is intermittently high due to *variations in the tightness of the landfill cap*, allowing radon to escape most of the time but causing radon to accumulate when the cap is tight.

The first of these possible explanations (radon in leachate is continually high but lost from poorly sealed sample jars) is unlikely, based partly on the reasonable presumption that sample collection has been done professionally and partly on the recent Lead-210 results reported by CoPhysics. The recent Lead-210 results, while uninformative about prior leachate samples showing intermittently high Lead-214 and Bismuth-214, do show that radon cannot have been continually high in the leachate (because otherwise Lead-210 would be detected at a higher level than in the recent Lead-210 results reported by CoPhysics).

In the second of these possible explanations, there is continual radon ingrowth from radium within the landfill, resulting in localized mixing of radon with landfill gas, but there is no continuously open circulation within the landfill that would allow such localized pockets of radon-landfill gas to come into contact with the leachate. Most of the time, flow paths within the landfill would be sufficiently constricted that most of the radon ingrowth would decay to its solid (less mobile) progeny before reaching the leachate – but at other times, as shown by the

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[14] See also 6 NYCRR 363-7.1(a)(3), which requires that "Drilling and production waste may not be placed within 6 feet of the leachate collection and removal system or within 10 feet of any final cover."

intermittently high readings in the test data, flow paths within the landfill are able to convey relatively large quantities of the radon toward and into the leachate. Given the magnitude of the intermittently high readings and the currently uncharacterized impacts, there is a need for additional testing to characterize the source and migration of the radon, including tests to identify how much radium is in the landfill and what the radon flow pathways are.

In the third of these possible explanations, there is continual radon ingrowth from radium within the landfill, resulting in a mixture of radon with landfill gas that periodically comes into contact with the leachate – but the radon pathway leading outward from the parent radium is highly variable. Most of the time, a radon-landfill gas mixture is *escaping freely into the atmosphere and effectively bypassing the leachate* in this scenario – but at other times, as shown by the intermittently high readings in the test data, the landfill cap or other barrier is confining the radon-landfill gas mixture in proximity to the leachate, allowing and causing substantial quantities of radon to dissolve into the leachate. Given the high activity (~1 million pCi/L) of the plume of radon-landfill gas leaking from the landfill most of the time under this scenario, there is a need for additional testing to characterize the source, migration, and impacts of the radon, including tests to identify how much radium is in the landfill and what the radon flow pathways are.

For any of these possible interpretations of the intermittently high results, it may be useful to determine whether the parent radium is mostly naturally occurring or mostly from radium-bearing waste disposal. However, neither outcome can ignore the question of health effects, nor can either outcome allow the intermittently high radionuclide levels to be dismissed as inconsequential. If naturally occurring, the intermittently high results raise a fundamental question about whether the landfill can be reasonably characterized, modeled, analyzed, and monitored. If mostly from radium-bearing waste disposal, the intermittently high results need regulatory attention and resolution.

There may be other explanations of the intermittently high results. Testing is the avenue that must be undertaken to properly characterize and understand the sources and consequences of radioactivity at the landfill.

F. Evidence of radioactivity in the leachate test results is not rebutted by the fact that all waste entering the landfill has passed through entrance monitors

The leachate test results are *not* rebutted by the fact that waste entering the landfill has passed through radiation-detecting monitors. For this type of monitoring to be effective, landfills would need entrance monitors that cannot be "gamed" by methods such as *deliberately manipulating truckloads of radium-bearing waste* in order to reduce the amount of radon in the incoming load. "Gaming" may be done by simple methods such as aerating and/or flushing and/or suction (e.g., drawing a partial vacuum within a waste load by covering the load with a tarp and applying suction from an ordinary shop-vac type of vacuum cleaner). DEC has not recognized that "gaming" is a problem and does not impose requirements to guard against "gamed" loads of waste entering landfills. Consequently, DEC's claims that wastes with high levels of radium and radon could not have passed through the gamma-detector entrance monitors at the Hakes landfill without triggering the monitors are unwarranted.

I identified this problem in my January 18, 2018 affidavit and showed how gamma-detector entrance monitors at Hakes and other landfills cannot reliably detect and prevent entry of waste loads carrying more than 25 pCi/g radium if precautions are not taken to prevent manipulation of loads of wastes entering landfills.

The issue of gamma-emission variability within waste loads that contain identical amounts of radium was identified as a study topic for the Pennsylvania DEP TENORM Study Report. In the scope of work for the report, the study topic was identified as follows:

> An assessment of secular equilibrium for the full uranium and thorium natural decay series as well as the Ra-226 and short-lived progeny sub-series, including the rapid buildup of radon and progeny in samples/waste streams impacted with radium. The evaluation of waste containing Ra-226 is subject to the buildup of radon gas and the other short-lived progeny of Ra-226, *complicating any decision made to transport or dispose of such materials based on an exposure rate survey of the container. The exposure rate is directly proportional to the degree of secular equilibrium and NOT proportional to the activity concentration of Ra-226* (remains the same as radon and other progeny buildup).

Pennsylvania Dept. of Environmental Protection (DEP), TENORM Study Scope of Work, p. 8, http://files.dep.state.pa.us/OilGas/BOGM/BOGMPortalFiles/RadiationProtection/TENORM-Study_SoW_04_03_2013_FINAL.pdf; emphasis added; word "NOT" capitalized in original.

The results of this assessment are presented and discussed in the Pennsylvania DEP TENORM Study Report, Rev. 1, May 2016. The results, while expressed for sludge or filter cake rather than drill cuttings, apply to any such radium-bearing waste. As stated in section 5.3 of the Report, "During handling and/or transport, the sludge or filter cake may be disturbed and some of the Rn gas may escape, *greatly reducing the gamma-emitting progeny* that follow Rn-222 in the natural decay series." (TENORM Study Report, Rev. 1, pp. 5-3 and 5-4, "Radon Ingrowth Within Filter Cake from WWTP to Landfills," emphasis added.) Modeling by Pennsylvania DEP examined the different gamma exposure rates measured 6 inches from the surface of the waste containers and found substantial variation in the gamma emission, depending on how much of the radon progeny remained in a given waste load along with its parent radium.[15]

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[15] The Pennsylvania DEP TENORM Study Report, Rev. 1 (May 2016), refers to a six-fold difference, as compared to the 60-fold difference presented and discussed in my January 18, 2018 affidavit. The discrepancy is due to a minor error by the TENORM report authors in describing their own results. Their Figure 5.1, depicting the results of their MicroShield modeling, shows 3.94 for the lowest exposure rate and 24.1 for the highest exposure rate. The ratio of 24.1 to 3.94 is 6.12, which can be rounded off to "six times" or "six-fold." However, the same page of the report says "*Starting from zero Rn progeny* to full

In summary, the entrance monitors at Hakes and other landfills cannot reliably detect and prevent entry of waste loads carrying more than 25 pCi/g radium if precautions are not taken to prevent manipulation of loads of wastes entering landfills. Reliance on these monitors by DEC and CoPhysics is unfounded; hence there is no basis for DEC's argument in the FSEIS (at 26) that "there is no plausible manner in which such radon values in air or leachate can be caused by the drill cuttings present."

G. 1.05 million pCi/L radon in landfill gas exceeds radon levels found or reported in other landfills and landfill models – and also in uranium mines

It would be useful to assess Hakes landfill against either landfill models or actual landfills that have roughly comparable radon levels in landfill gas, but levels as high as 1 million pCi/L have not been found or reported for other landfills. Another potentially useful comparison would be to radon levels in uranium mines, even in older mines that took fewer protective measures, but here again the available data show substantially less than 1 million pCi/L radon in mine air. The lack of comparative examples in such subterranean spaces adds to the uncertainty about whether radium-bearing waste brought into Hakes has been limited to 25 pCi/g.

Available information on landfills provides little or no discussion or quantification of radon in landfill gas or its emission rate through the landfill cap.[16] Such information is likewise sparse in

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equilibrium after 21 days, the exposure rate measured 6 inches from the outside of the roll-off container increased six-fold. Based on the MicroShield® modeling results, there may be an increase of six times the gamma exposure rate measured 6 inches from the surface of the roll-off container during the first 21 days after a wastewater treatment sludge is generated. This is a theoretical curve and assumes all of the Rn is removed when the sludge is formed at time zero." (emphasis added) Contrary to the emphasized words in this quote, the TENORM report's graph (Fig. 5.1) and its interpretation of the modeling results fail to include the "zero Rn progeny" point. See the list of five data points at the top of page 5-4, starting with point "a" which is described as "0-day ingrowth (13.4 pCi/g of Ra-226 only)." In fact, this point is omitted from further consideration; only the last four data points are included in the graph (Fig. 5.1), despite what the authors say in the above-quoted words. Hence, the first of five data points, representing the gamma exposure rate from time zero (gamma from Ra-226 only, with no progeny) needs to be put into Fig. 5.1 before the full exposure trend can be seen. Just by eyeballing the existing curve – projecting it downward and to the left toward time zero – it is evident that the curve will intersect the vertical axis slightly above zero exposure rate. It can't be as low as zero exposure because that would mean that Ra-226 emits no gamma at all (untrue), but it may be as low as 0.5 or 0.4. The ratio of 24.1 to 0.5 is 48.2 (or 48-fold); the ratio of 24.1 to 0.4 is 60.25 (or 60-fold). While eyeballing like this won't provide a precise answer, it's obvious that the MicroShield results, if correctly reported, would show the full range of gamma-emission variation from a given quantity of radium as being close to the 60-fold value presented and discussed in my January 18, 2018 affidavit. And regardless of whether it's six-fold or 60-fold, this is an excessive and unacknowledged uncertainty in the landfill entrance monitoring procedure.

[16] For example, no substantive information on landfill radon is found on U.S. EPA websites such as https://www.epa.gov/landfills/industrial-and-construction-and-demolition-cd-landfills and https://www.epa.gov/lmop/basic-information-about-landfill-gas; NYS Dept. of Health website, https://www.health.ny.gov/environmental/outdoors/air/landfill_gas.htm; Illinois Dept. of Public Health

the available literature on landfill modeling studies. DEC's response to the Sierra Club comment letter relies substantially on two Argonne National Laboratory reports, by Smith et al. (1999)[17] and Harto et al. (2014),[18] both of which find acceptable human exposures from modeled landfills in which radium-bearing waste is limited to 50 pCi/g. These modeled results, which DEC considers strong evidence of minimal health impacts, beg the crucial question of whether radium-bearing waste brought into Hakes is as low as claimed (<25 pCi/g) or is in fact much higher. For the purpose of comparison, it is unfortunate that neither of these Argonne landfill models provides any quantitative details about radon flux through the modeled landfill cap, and neither model considers downwind radon transport and inhalation exposure to an offsite resident.[19] Fortunately, a 2012 study by Walter et al.[20] uses a roughly similar methodology, including the 50 pCi/g limit assumed by the Argonne authors, and also provides both landfill-gas and radon emission rates through a modeled 200 m x 200 m landfill cap.

For landfill-gas emission through the cap, Walter et al. show a rate of 0.2 m^3/sec, equivalent to roughly 8.5 moles/second of landfill-gas emission.[21] For radon emission through various cap systems of their modeled landfill, they show a range of about 6E+4 to 4E+6 pCi/sec, equivalent to roughly 10^{-13} to 10^{-15} moles/second of radon emission. In combination, these values indicate that the landfill gas emerging from the cap and vents of their modeled landfill *contains radon ranging from about 300 to about 20,000 pCi/L* on a steady-state basis, with the radon mole fraction being on the order of 10^{-14} to 10^{-16}. Such radon activities of 300 to 20,000 pCi/g are far less than 1 million pCi/L; hence, even allowing for some variation among the models by Smith,

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fact sheet, http://www.idph.state.il.us/envhealth/factsheets/landfillgas.htm; etc. *This is a serious omission if high levels of radon in landfill gas could indeed be produced by either local geology or typical C&D wastes, as DEC suggests may be happening at Hakes landfill*. Information on radon levels in landfill gas and radon emission rates through landfill caps is generally not included in otherwise useful literature on sites known to contain radium-bearing wastes (e.g., USGS, *Background Groundwater Quality, Review of 2012-14 Groundwater Data, and Potential Origin of Radium at the West Lake Landfill Site, St. Louis County, Missouri*, Dec. 17, 2014; revised June 10, 2015; "Missouri Department of Natural Resources to Transition Air Monitoring at Bridgeton Landfill Responsible Parties to Continue Air Monitoring," https://dnr.mo.gov/bridgeton/documents/airmonitoringstepdownannouncementwithcharts_000.pdf).

[17] K.P. Smith, D.L. Blunt, G.P. Williams, J.J. Arnish, M. Pfingston, J. Herbert, and R. Haffenden, *An Assessment of the Disposal of Petroleum Industry NORM in Nonhazardous Landfills*, DOE/BC/W-31-109-ENG-38-8, prepared by Argonne National Laboratory for U.S. Department of Energy, National Petroleum Technology Office, Tulsa, OK (1999).

[18] C.B. Harto, K.P. Smith, S. Kamboj, and J.J. Quinn, *Radiological Dose and Risk Assessment of Landfill Disposal of Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in North Dakota*, ANL/EVS-14/13, prepared by Argonne National Laboratory (November 2014).

[19] Both focus more on *groundwater* pathways for radon exposure to an offsite resident. Thus, both acknowledge the possibility of radon inhalation by an offsite resident during showering, due to volatilization of radon from well water into the shower stall, but dismiss this inhalation risk and do not acknowledge or consider direct airborne impacts from radon flux through the landfill cap. See Smith et al., op. cit., at 34; Harto et al., op. cit., at 21.

[20] 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," *Journal of the Air & Waste Management Association* **62**, 1040-1049 (2012).

[21] Hakes landfill-gas emission, mostly collected and flared, is a *roughly* similar rate. DSEIS, Appendix H.

Harto, Walter, and others, the radium-bearing waste which is limited to 50 pCi/g in these modeled landfills cannot account for 1 million pCi/L radon in landfill gas.

The inferred 1 million pCi/L in Hakes landfill gas may be either continual or intermittent, as discussed above. However, even if intermittent, there is no clear explanation of how an intermittently high level of about 1 million pCi/L could be generated and remain present in Hakes landfill gas for a long enough time to infuse the leachate with 270,000 pCi/L radon. This is yet another indication that measurement and modeling are needed to characterize the source and flux of radon within the Hakes landfill.

A corrective action report for the Blue Ridge Landfill in Kentucky shows two different modeled rates of radon emission through the cap of that landfill (0.0749 pCi/sec and 2.37 pCi/sec, per square meter of cap area), with the difference between these two values due to different modeled thicknesses of the radium-bearing waste layer and the overlying "clean" layer of municipal waste.[22] These radon emission rates correspond to about 2E-21 and 7E-20 moles/second per square meter of cap area, respectively. These emission rates are *roughly* comparable to the radon emission rates modeled by Walter et al.[23] The modeling done for the Blue Ridge Landfill assumed about 27 pCi/g radium in the radium-bearing waste layer, which is close to the 25 pCi/g nominal limit for Hakes, so it is unfortunate that the Blue Ridge corrective action report does not specify a landfill-gas emission rate for purposes of comparison.

Turning now to uranium mines in which both radium and radon are typically present at high levels, the U.S. National Academies' authoritative report, *Health Effects of Exposure to Radon*,[24] shows that mine radon levels are substantially lower than 1 million pCi/L, even in older mines that took fewer precautions to protect miners. Average radon activities for some of the highest-exposure uranium mines are reported as 1.6 WL or about 400 pCi/L (New Mexico cohort of miners), 4.9 WL or about 1230 pCi/L (Newfoundland cohort), 11.7 WL or about 2940 pCi/L (Colorado cohort), and 14.9 WL or about 3740 pCi/L (Port Radium cohort in the Northwest Territories of Canada).[25] *All of these radon levels are less than 1% of 1 million pCi/L*. Granted, these numbers don't express the higher and lower radon values that average out to the mean exposures, but another table from *Health Effects of Exposure to Radon* provides some of these higher values from the Ambrosia Lake uranium mines in New Mexico from the 1960-1961

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[22] Gradient Corp., *Corrective Action Plan (CAP) for the Blue Ridge Landfill, Estill County, Kentucky* (May 3, 2017), page 55 of Attachment A1 and page 20 of Attachment A2.

[23] The range of radon emission rates modeled by Walter et al., op. cit. (about 10^{-13} to 10^{-15} moles/second) is about 3E-18 to 4E-20 moles/second *per square meter of cap area*, thus *roughly* comparable to the modeled Blue Ridge Landfill emission rates.

[24] National Research Council, Committee on Health Risks of Exposure to Radon, *Health Effects of Exposure to Radon*, commonly called the "BEIR VI" report (Washington, DC: The National Academies Press, 1999).

[25] Id., Table D-12 (p. 270), where radon and radon-progeny exposure is reported as Mean Working Level (WL), weighted by person-years, including 5-year lag interval; with correlation between WL and pCi/L taken from Table ES-1 (p. 12). Note that exposures reported as "Weighted Mean WL" in Table D-12 are somewhat higher, sometimes by a factor of 2 or 3 or slightly more. See also pp. 254-269 for additional detail.

period, including measurements of 20 WL (about 5000 pCi/L) and 37 WL (about 9300 pCi/L).[26] But even these radon levels are less than 1% of 1 million pCi/L.

The above comparisons serve as a reminder that radon levels in enclosed underground spaces where radium is present, whether landfills or mines, are limited by such factors as the radium level (e.g., 25 pCi/g), the available space that the radon can occupy, and the pathway(s) that the radon can migrate along. It is instructive to consider a one-cubic-meter sealed container that contains, for example, compacted radium-bearing waste or soil with 25 pCi/g activity, 2 g/cm^3 bulk density, and 24.5% porosity. Let all the radium be Radium-226; the radon will then be Radon-222. The pore space can be considered occupied by air and/or landfill gas, and, after a few weeks, the pore space will also be occupied by an essentially constant level of radon that is at secular equilibrium with the radium in the sealed container. The total radium activity in the sealed container is 50 million pCi, and, after a few weeks, the total radon activity in the container will also be 50 million pCi. The volume of the pore space is 245 liters (i.e., 24.5% of one cubic meter), and the radon activity[27] in the air or landfill gas in the pore space is therefore 204,082 pCi/L. This radon level in the sealed container is about 20% of 1 million pCi/L. It will never exceed 204,082 pCi/L but will remain almost constant at that level, declining very slightly over time as its parent radium decays.

Further calculation with this example will show that the mole fraction of radon in the pore space is on the order of 10^{-13}, indicating that the radon atoms are dispersed among much larger numbers of molecules such as nitrogen, oxygen, and methane. Changing the radium activity in this example from 25 pCi/g to a higher or lower value would make a proportional change in the essentially constant radon level in the sealed container. Changing it to 125 pCi/g would increase the long-term radon level in the sealed container to about 1 million pCi/L. Changing it to a typical level for Steuben County soils would reduce the long-term radon level in the sealed container to much less than 200,000 pCi/g.

Alternatively, let the one-cubic-meter sealed container contain radium-bearing waste or soil with 25 pCi/g activity, 1.855 g/cm^3 bulk density, and 30% porosity. The total radium activity in the sealed container is 46.375 million pCi, and, after a few weeks, the total radon activity in the container will also be 46.375 million pCi. The volume of the pore space is 300 liters (i.e., 30% of one cubic meter), and the radon activity in the air or landfill gas in the pore space is therefore 154,583 pCi/L. The mole fraction of radon in the pore space is slightly smaller than in the previous example, but still on the order of 10^{-13}. Note that increasing the porosity and pore space does not increase the radon activity in pCi/L; it reduces it.

The interconnected pathways in underground spaces cannot be characterized with the simple type of calculation performed above for sealed one-cubic-meter containers. Characterization must usually be based on a combination of measurement and modeling – but it needs to be more than mere speculation about how radon might migrate. There are thermodynamic constraints on whether and how radon can become increasingly concentrated as it moves from a quasi-sealed

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[26] Id., Table E-1 (p. 294), with correlation between WL and pCi/L again taken from Table ES-1 (p. 12).

[27] Assuming 1.54E+17 pCi/g specific activity of Radon-222.

environment (such as the above container or a tightly packed landfill space) into a series of pathways where it mixes – but is less likely to unmix! – with other gases. The very low mole fraction of radon is an important factor. Different gas solubilities may also play a role, as would need to be determined by the necessary combination of measurement and modeling. In the end, such characterization must explain not only the radon levels in landfill gas but also the intermittent process by which Hakes leachate has been infused with ~270,000 pCi/L radon. Such characterization, when done, will provide a meaningful basis for assessing health effects.

H. Could the leachate test results be measuring radiation coming from area geology?

While the possibility cannot be ruled out that the intermittently high radon comes mainly from naturally occurring onsite radium, the available evidence does not provide good support for this possibility. It is undisputed that many Steuben County homes have >4 pCi/L naturally occurring radon, but this fact does not translate into an explanation of the intermittently high radon at Hakes. The problems with attributing intermittently high radon to the local geology include 1) explaining how a sufficient amount of radon gas, or groundwater carrying dissolved radon, could migrate upward through the landfill liner to raise the radon concentration in Hakes leachate to ~270,000 pCi/L, at least intermittently, and 2) explaining how such upward migration through the liner could be so highly intermittent.

Before natural geology can be claimed as an explanation, it needs to be explained. Well-designed and properly executed testing would be needed to show that the intermittently high radon comes mainly from naturally occurring onsite radium. For example, while leakage through landfill liners to certainly known to exist,[28] it cannot be proffered as a vague, unquantified explanation of how radon could leak upward into leachate, especially because a) leachate typically flows *downward* through liner leaks, and b) liner leaks are recognized as serious problems in landfill operation and can't just be offered as a casual guess about how underlying radon might enter the leachate.

Furthermore, any finding that the intermittently high radon comes mainly from naturally occurring onsite radium would raise several new questions for regulators and policymakers.

It should be obvious that levels such as 1.05 million pCi/L radon in landfill gas, *if predominantly from radium-bearing waste*, warrant investigation of both the radium (how much? where in the landfill?) and the resulting radon, particularly the radon flow pathways and any offsite fate, transport, and associated risk. But even if the intermittently high radon is shown to be mainly from naturally occurring onsite radium, the above questions about radon flow pathways and offsite fate, transport, and associated risk do not automatically disappear! The landfill's role in concentrating the "natural" radon to such unusually high levels would need to be assessed and understood, and questions of onsite and downwind health impacts would still need to be assessed and resolved. Furthermore, if the intermittently high radon is mainly from naturally occurring

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[28] Smith et al., op. cit., at 43 and 67.

onsite radium, an important question of site suitability arises for both the landfill and its proposed expansion.

As an example, the site would fail one of the U.S. Nuclear Regulatory Commission's disposal site suitability requirements for land disposal of radioactive wastes:

> The disposal site shall be capable of being characterized, modeled, analyzed and monitored.

10 CFR 61.50(a)(2).

This requirement, while not directly applicable to C&D landfills in New York, is eminently sensible. A site that undergoes wild swings in its naturally occurring radon cannot be reasonably characterized, modeled, analyzed, and monitored. Its unpredictable swings in natural radon would "mask" any migration of radium progeny and thwart any meaningful monitoring program. This principle is also expressed in the NYS siting requirements for radioactive waste disposal:

> The site must not be located where currently existing radioactive material, including but not limited to naturally occurring radioactive material, may mask the monitoring program.

6 NYCRR 382.21(a)(7).

6 NYCRR 363-7.1(a)(5)(iii), which *is* applicable to C&D landfills, requires that "Background radiation readings at the facility must be measured and recorded at least daily." Such a requirement would be meaningless if measurements of natural "background radiation" ranged from a few pCi/L to more than 1 million pCi/L at slightly different locations "at the facility."

Generally speaking, disposal-site performance needs to be understandable in some reasonable fashion. Wild swings in naturally occurring radon cannot reasonably be given a "free pass" that would exempt their causes and effects from being characterized and understood.

III. What might be the health effects of the levels of radioactivity shown?

A. What are the radiation dose, the applicable standard, and the associated risk?

The public health and occupational health risks associated with Hakes leachate, containing relatively low radium levels but intermittently very high levels of radon, have not been assessed and are currently unknown.[29] These risks need to be determined by a combination of testing and modeling, especially in view of the evidence that the landfill gas at least occasionally contains more than 1.05 million pCi/L radon. If there were either steady-state emissions or occasional

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[29] See above and below for discussion of Argonne reports by Smith et al., op. cit., and Harto et al., op. cit.

"puffs" of such landfill gas, how often would downwind receptors be unduly exposed? What levels would landfill workers be exposed to? The risks cannot reasonably be assumed trivial, especially since landfill-gas emissions from the expanded landfill may triple from their current rate (see DSEIS, Appendix H, at 7). A publicly transparent program of air monitoring for radon and its progeny, combined with well-constructed air dispersion modeling, is needed. Dispersion modeling based on monitored pollution data is a well-known technique[30] for generating air-pollution maps and should be conducted at the Hakes landfill and surrounding area.[31]

Such testing and modeling are needed *regardless of whether such high levels of radon and its progeny are from radium-bearing waste or from naturally occurring onsite radium*. It is undisputed that naturally occurring radon causes exceedances of the 4 pCi/L health guidance level in many Steuben County homes, but a radon level such as 1.05 million pCi/L in landfill gas is so vastly different from 4 pCi/L that it requires testing and resolution. Does 1.05 million pCi/L landfill gas move offsite, and, if so, how much dilution and dispersion occur? How much do dilution and dispersion depend on weather conditions? These are among the questions that need to be answered to determine health impacts.

In addition, if further investigation of the disequilibrium between low radium and intermittently high radon in leachate shows substantially higher levels of radium in the landfill than currently acknowledged, then the *long-term health impacts* of such radium would need to be assessed and addressed.

When health effects are assessed, EPA's 4 pCi/L "action level" for indoor radon should *not* be used or adopted as the applicable radon exposure standard. The applicable exposure standard *must represent a level that has been recognized as reasonably safe*. Such a level is roughly an order of magnitude lower, i.e., about 0.4 or 0.5 pCi/L. EPA's 4 pCi/L action level is too high for this purpose; it corresponds to a Relative Risk of about 1.14 (i.e., about 14% excess risk)[32] and, according to EPA's estimates, about 7 in 1000 nonsmokers and about 62 in 1000 smokers may get lung cancer from lifetime exposure to radon at 4 pCi/L.[33] As enacted in U.S. law, the "national long-term goal of the United States with respect to radon levels in buildings is that the air within buildings in the United States should be as free of radon as the ambient air outside of

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[30] Models such as ISCST3 and AERMOD typically represent pollutant emissions as point sources, but cf. S.G. Perry et al., "AERMOD: A Dispersion Model for Industrial Source Applications, Part II: Model Performance against 17 Field Study Databases," *Journal of Applied Meteorology* **44**, 694-708 (2005), at 703-04, for references to modeling involving multiple near-surface emission sources. Whether radon emission from Hakes landfill is best represented as a point source or multiple sources is unknown pending site-specific monitoring.

[31] DEC's SCREEN3 modeling in DSEIS Appendix H could provide a first step toward such modeling. This modeling was for hydrogen sulfide but would serve as a screening-level tool for radon if emission rates were either known or estimated over a range of possible values.

[32] National Research Council, BEIR VI, op. cit., 11, 17, 85, 115, etc.

[33] U.S. Agency for Toxic Substances and Disease Registry, *ATSDR Case Studies in Environmental Medicine: Radon Toxicity* (2010), https://www.atsdr.cdc.gov/csem/radon/radon.pdf, Fig. 2, at 31-32.

buildings,"[34] which is about 0.4 pCi/L.[35] Another standard in the same range, although not directly applicable, is the 0.5 pCi/L specified for emissions from the residual radioactive materials or tailings at inactive uranium processing sites. According to 40 CFR § 192.02(b)(2), "Control of residual radioactive materials and their listed constituents shall be designed to… [p]rovide reasonable assurance that releases of radon-222 from residual radioactive material to the atmosphere will not… [i]ncrease the annual average concentration of radon-222 in air at or above any location outside the disposal site by more than one-half picocurie per liter."

Walter et al., in their study of municipal solid waste landfills accepting up to 50 pCi/g radium-bearing waste, find that in some of the disposal scenarios they considered, "the radon flux from the landfill and off-site atmospheric activities exceed levels that would be allowed for radon emissions from uranium mill tailings" and that "...the simulated radon fluxes for the scenarios analyzed exceed the uranium mill tailings regulatory flux limits for the cases without a geomembrane cover, and exceed the off-site radon activity limit in all cases."[36] The similarity of the Walter et al. study to those cited by DEC (Smith et al. and Harto et al.), and the fact that the 50 pCi/g waste acceptance limit in all three is only twice the nominal 25 pCi/g limit at Hakes, serve as a reminder that health effects need site-specific investigation at Hakes. This is true regardless of whether the nominal 25 pCi/g limit at Hakes is shown to be credible or not.

B. Is the relationship between dose and risk *linear* down to very low doses, with *no threshold* below which there is no risk?

The U.S. National Academy of Sciences' Committees on the Biological Effects of Ionizing Radiation ("BEIR") have produced two comprehensive reports in recent decades; these are typically referred to as the BEIR V report and the BEIR VII report. I own a copy of the BEIR V report (it is a book of about 400 pages, entitled *Health Effects of Exposure to Low Levels of Ionizing Radiation*, published in 1990 by the National Academy Press). I have periodically consulted but do not own a hard copy of the BEIR VII report (*Health Risks from Exposure to Low Levels of Ionizing Radiation*, 2006, online at https://www.nap.edu/catalog/11340/health-risks-from-exposure-to-low-levels-of-ionizing-radiation) which provides a scientific update of BEIR V.[37] Both BEIR V and BEIR VII find that, *according to the preponderance of the scientific evidence*, there is a "linear, no-threshold" (LNT) relationship between dose and health risk, extending down to very low doses.[38] Both BEIR V and BEIR VII acknowledge the difficulty of proving this relationship at very low doses yet find that this LNT relationship, frequently called the LNT hypothesis, is the best fit of the available data.

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[34] 15 USC § 2661, Indoor Radon Abatement Act of 1988.
[35] U.S. EPA, *A Citizen's Guide to Radon*, EPA402/K-12/002 (2016), at 7.
[36] Walter et al., op. cit., at 1040 and 1048.
[37] Other BEIR Committees convened by the National Academies have produced more specialized reports such as National Research Council, BEIR VI, *Health Effects of Exposure to Radon*, op. cit.
[38] For example, BEIR V at 4 and 171-81; also Dr. Arthur C. Upton, pers. comm.; BEIR VII at 245-46 and discussion thereof in K.D. Crowley et al., *Radiation Research* **183**, 476-481 (2015).

I am not an expert in health physics but have been aware for the past few decades of ongoing scientific controversy about the LNT hypothesis. I have observed that scientists who challenge the LNT hypothesis often have a vested interest in nuclear technology, e.g., careers in a field such nuclear research, nuclear medicine, or the nuclear power industry.

This ongoing controversy should be recognized as relevant background information for interpreting the last part of the CoPhysics Report, where Dr. Rahon argues against the LNT hypothesis in his "Key Issue" #6, and where his Exhibit I is a published paper by Calabrese and O'Connor that is offered as a refutation of the LNT hypothesis. It should be noted that this Calabrese and O'Connor paper is not the "last word" on the topic, as both a response and a counter-response have been published.

The response is by K.D. Crowley et al., "Comments on Estimating Risks of Low Radiation Doses—A Critical Review of the BEIR VII Report and Its Use of the Linear No-Threshold (LNT) Hypothesis by Edward J. Calabrese and Michael K. O'Connor," *Radiation Research* **183**, 476-481 (2015). These authors quote from the BEIR VII report and discuss the BEIR VII Committee's reasoning for its conclusion:

> …the balance of scientific evidence at low doses tends to weigh in favor of a simple proportionate relationship between radiation dose and cancer risk.

BEIR VII at 246, quoted and discussed by Crowley et al. at 476.

The counter-response which continues to criticize the LNT hypothesis is by E.J. Calabrese and M.K. O'Connor, "Response to Comments on 'Estimating Risks of Low Radiation Doses—A Critical Review of the BEIR VII Report and Its Use of the Linear No-Threshold (LNT) Hypothesis'," *Radiation Research* **183**, 481-484 (2015).

Those assessing the scientific points at issue in the Hakes landfill expansion will need to draw their own conclusions about the merits of these differing views of the LNT hypothesis. My own best understanding is that K.D. Crowley et al. and the BEIR VII Committee exhibit the better scholarship on this subject, and that a) the evidence for understanding the dose-response relationship at low doses is difficult to assemble and interpret, yet b) when done carefully, the LNT hypothesis remains the best-supported scientific understanding of this relationship.

IV. DEC and CoPhysics have not provided substantive/credible responses to the Sierra Club comments

On a variety of points, DEC and CoPhysics have not provided substantive/credible responses. Most of these points have been discussed above in section II but are summarized in this section for each of the eight Sierra Club comments, listed below as A through H.

In general, DEC argues that the wastes accepted for disposal at the Hakes facility

> are not proposed to change. As explained in Section 2.4.9 of the DSEIS, these wastes have historically included drill cuttings and, therefore, the scope of the DSEIS was properly developed. Nevertheless, while the scope of the action before the Department remains the expansion of the landfill (including the borrow area) and does not include any approval related to a change in the acceptable waste streams, the Department has provided responses to all substantive comments related to radioactivity…

FSEIS at 14.

In fact, this claim that "the Department has provided responses to all substantive comments related to radioactivity" does not hold up to scrutiny. The responses provided by DEC and CoPhysics are not substantive and credible, as explained more fully below.

A. The DSEIS fails to evaluate the high levels of radioactivity shown in the landfill's leachate test results

The Sierra Club comment letter at 2-6 notes the contradiction between the landfill's own leachate test results (showing high levels of Lead-214 and Bismuth-214 in some of the samples) and the DSEIS claim that "at no time have any levels" of radioactivity in Hakes leachate "been detected that would indicate any radioactivity beyond those associated with background levels." The Sierra Club comment discusses the likely escape of radon from certain samples and the implications and significance of the high levels of Lead-214 and Bismuth-214 in certain other samples, with the latter result indicating about 270,000 pCi/L radon in the sample – and thus in the sampled leachate – at the time of collection, and further indicating about 1.05 million pCi/L radon in the air of the landfill, depending partly on whether or not the parent radium was immersed in leachate. The Sierra Club comment expressed a further concern about the reported discontinuation of Bismuth-214 and Lead-214 testing in leachate samples, the concern being that the loss of such data would undermine the ability of DEC and the public to understand and monitor the radionuclides in the landfill. Indeed, the comment recommended testing for a *greater* number of radionuclides rather than elimination of testing of the very radionuclides that have been observed in high levels in the leachate. Overall, by failing to address the likely source of the radioactivity demonstrated in the landfill's leachate test results, i.e., the Marcellus shale drill cuttings that have been accepted at the landfill since 2010, the DSEIS has not adequately assessed the issue of radioactivity in the landfill. The DSEIS has thus failed to provide a reasoned elaboration for why increasing the capacity of the landfill and allowing it to take more radioactive shale gas drilling wastes will not have an adverse effect on the environment and the health and safety of the people, animals and plants living near the landfill.

DEC's response (FSEIS at 18) refers to this Sierra Club comment as "B-1" and refers readers to its responses to comments B-10 and B-13.

DEC's response to comment B-10 (FSEIS at 21) does not address or acknowledge the issue raised in Sierra Club's comment about high levels of radioactivity shown in the landfill's leachate test results. DEC's first two points of response allege that categories of drilling wastes with high radioactivity would not be allowed for disposal at Hakes, and, furthermore, that "The radiation detector would allow Hakes to prevent any wastes higher than 25 pCi/g from being accepted by the landfill." This last statement is incorrect; the entrance monitors (radiation detectors) cannot reliably prevent entry of waste loads exceeding 25 pCi/g. DEC's claims about waste categories that are not allowed for disposal at Hakes do not address or acknowledge the high levels of radioactivity that have been shown in the landfill's leachate test results. DEC refers to a truckload carrying radium-bearing waste (a radio-luminous ship deck marker) that tripped the Hakes entrance monitor in 2016. The description of this incident, while interesting with respect to wastes that were excluded from the landfill, is not informative in detail (it does not quantify either the amount of radium or the concentration of its progeny in the truckload), nor does it address or acknowledge the point raised in this comment about high levels of radioactivity shown in the landfill's leachate test results.

DEC's response to comment B-13 (FSEIS at 22-23), while it acknowledges the issue raised in Sierra Club's comment about high levels of radioactivity in the leachate test results, *provides no meaningful response*. DEC states that radium levels in the *leachate* are relatively low, which is generally not at issue.[39] DEC acknowledges that "Radon values can be inferred from the Pb-214 and Bi-214 gamma spectrometry results" but alleges "inaccuracies with results obtained for leachate and other liquid samples using the method that the comment is based on, which is EPA 901.1 gamma spectrometry." Such claims about "inaccuracies" are incorrect and misleading with respect to the intermittently high test results for Pb-214 and Bi-214 in Hakes leachate, as discussed above (in section II) in detail. Thus, no meaningful response has been provided to Sierra Club's comment about high levels of radioactivity shown in the landfill's leachate test results. DEC's claim that "any elevated radon and subsequent radon progeny in leachate is likely related to the local geologic sources" fails to recognize or address the distinction between radon levels ranging up to 1.05 million pCi/L in the landfill and the "indoor radon issues" in which household radon levels often exceed 4 pCi/L due to the natural geology of Steuben County. Any meaningful response by DEC would need to address the vast difference between 1.05 million pCi/L and 4 pCi/L (the former remains unexplained; the latter is readily explained by local geology), and would also need to address the extreme variability of the landfill radioactivity measurements (they're *intermittently* very high). DEC's response addresses neither of these. DEC's discussion of allowable radionuclide concentrations in sewer discharges is interesting but does not provide the type of evaluation indicated as necessary in the Sierra Club comment. The high radon levels, in particular, need to be – but have not been – evaluated with respect to their source and their impacts. Evaluation of their source would, in turn, address the fundamental issue of whether the landfill contains unacknowledged radioactive waste that substantially exceeds the landfill's nominal limit of 25 pCi/g.

DEC's response does not acknowledge or address the concern expressed in the Sierra Club comment about the reported discontinuation of Bismuth-214 and Lead-214 testing in leachate samples. As noted in the comment, the loss of such data would undermine the ability of DEC and the public to understand and monitor the radionuclides in the landfill. If any change is made, a *greater* number of radionuclides should be tested (including Lead-210). There would be no reasonable basis for eliminating the testing of the radionuclides that have been present in intermittently high levels in the leachate.

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[39] The unknown radium levels in the *landfill* are a central issue, as discussed above. Test results indicate that radium levels in the *leachate* remain relatively low, as discussed above.

Part of DEC's response to comment B-9 (FSEIS at 20), proffered in response to a Concerned Citizens of Allegany County comment, is also noteworthy. DEC claims that "The two isotopes referred to in the comment are Pb-214 and Bi-214, which have half-lives of 26.9 and 19.9 minutes respectively. These isotopes will decay to stable elements before they could reach any potential receptors." The second sentence of this DEC claim is incorrect. The Pb-214 and Bi-214 radionuclides (isotopes) will in fact decay to the radionuclides Pb-210, Bi-210, and Po-210, which are widely recognized as health risks if ingested or inhaled,[40] before decaying to stable elements.

Part of DEC's response to comment B-11 (FSEIS at 22) is likewise noteworthy for its misleading statement that "Po-210 and Pb-210 break secular equilibrium, which in simple terms means that the resulting amount of these isotopes is a very small fraction of the initial Rn-222 value." This statement depends on what is meant by "amount" and on the time frame during which the health risk is considered. When Pb-210 and Po-210 are compared to their parent radon ("the initial Rn-222 value"), it is true that their activity in curies or pCi will be lower at the outset, but the fact remains that every Radon-222 atom will decay to Lead-210, Bismuth-210, and Polonium-210 before decaying to a stable Lead-206 atom.[41] Thus, compared to radon, the health risk window for Pb-210, Bi-210, and Po-210 is attenuated and prolonged, but not reduced overall. DEC's claim in the same response that "there is no realistic potential for buildup or accumulation resulting in impacts" depends entirely on fate, transport, and exposure scenarios, *which have not been addressed or assessed*.

The CoPhysics Report (at 18) argues that "levels of radioactivity in the leachate remain far below allowable Part 380 discharge limits (i.e., be it as effluent discharge or sewer discharge)" and that *radium* levels in leachate are low. These claims miss the point of the *radon* levels that, as discussed above in section II, are intermittently very high in leachate and apparently in landfill gas as well, resulting in a host of unaddressed/unanswered questions about:

- the parent radium source (how much? where? is it "high and dry"? naturally occurring or from radium-bearing waste?),
- the fluctuating pathways and possible barriers responsible for the intermittent nature of the high radionuclide readings,
- the nature and magnitude of radon emission from the landfill, whether in puffs or a more constant plume, and
- impacts to downwind residents and landfill workers.

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[40] For example, see https://www.ncbi.nlm.nih.gov/pubmed/4003271 (primarily about radon progeny in liquified petrolem gas [LPG]); https://www.ncbi.nlm.nih.gov/pmc/articles/PMC2477690/ (Po-210 dose to Alexander Litvinenko); https://ehs.stanford.edu/reference/pb-210bi-210po210-radionuclide-fact-sheet (Stanford campus fact sheet). As usual with radioactivity and radioactive materials, the health risk is understood to be proportional to the exposure or dose.

[41] There is a negligible branching ratio from Pb-210 that may bypass Bi-210, etc.

B. The DSEIS fails to evaluate the adequacy of the landfill's entrance monitors

The Sierra Club comment letter at 6-7 notes that the DSEIS fails to evaluate the adequacy of the entrance monitors and points out the following claims that were made in the Hakes DSEIS:

> [a]t no time have any drill cuttings or other wastes from the oil and gas extraction industry set off the detector alarms at the Hakes Landfill. However, the alarms have proven to be effective in detecting several loads of solid waste that did not contain drill cuttings or other wastes from the oil and gas extraction industry, but potentially did contain radioactive wastes. This demonstrates the efficacy of the detection equipment.

As the Sierra Club comment letter points out, the logic of these claims in the DSEIS is faulty. With support from my January 2018 affidavit, the comment letter explains that the claimed efficacy of the detection equipment (the landfill's entrance monitors) in detecting radioactive waste entering the landfill is discredited by the landfill's leachate test results which show that there are significant levels of radium and radon in the landfill. My affidavit pointed out that truckloads carrying identical Radium-226 concentrations can have widely variable levels of gamma radioactivity measurable at the landfill entrance, depending on whether radium breakdown products have been allowed to escape from the load or not. This renders any "correlation" between the truckload sample analysis and the gamma radioactivity measurements meaningless unless the monitoring procedure at the landfill gate can quantify and control for the concentrations of radium progeny such as Lead-214 and Bismuth-214 in the load of waste at the time the truck enters the landfill gate.

DEC's main response (response B-2, FSEIS at 18) states that the landfill entrance monitors or "radiation portal monitors" are an established part of the landfill's permit requirements and "will continue to be operated to ensure compliance with any applicable waste acceptance limitations." This statement is merely conclusory and does not address the point at issue. DEC's response also claims that Radium-226 decaying within the matrix of drill cuttings "will retain most of the Rn-222 within the rock matrix, therefore the other progeny will remain within the matrix as well." As the basis for this claim, DEC refers to the behavior of "many natural materials including shale." This fails to address the point at issue, namely waste truckloads being *deliberately manipulated to remove radon gas* in order to pass through the entrance monitor while carrying substantially more than 25 pCi/g of radium-bearing waste. DEC's response also cites its 2015 High Volume Hydraulic Fracturing Response to Comments, page RTC-103 and its current Hakes FSEIS response to Comment B-10.

DEC's response to Comment B-10 (FSEIS at 21) does not acknowledge the point at issue, namely, that the entrance monitors detect only gamma radiation, most of which is from radium progeny rather than radium itself. DEC's first two points of response allege that categories of drilling wastes with high radioactivity would not be allowed for disposal at Hakes, and, furthermore, that "The radiation detector would allow Hakes to prevent any wastes higher than 25 pCi/g from being accepted by the landfill." This last statement is unsupported and merely conclusory. The claims about waste categories that are not allowed for disposal at Hakes do not address the efficacy of the entrance monitor. DEC's most informative response to comment B-

10 involves a truckload carrying a manufactured object containing radium (a radio-luminous ship deck marker) that tripped the Hakes entrance monitor in 2016. However, while interesting, DEC's description of this incident does not quantify either the amount of radium or the concentration of its progeny. It thus fails to address the point at issue here, namely, whether/how radium can be detected and properly quantified if accompanied by variable concentrations of its strongly gamma-emitting progeny.

DEC's 2015 High Volume Hydraulic Fracturing Response to Comments, p. RTC-103, defends gamma spectroscopy as a broadly recognized, efficient, and accurate method for quantifying radium in environmental samples. This is undisputed; it adds nothing to the point at issue here.

The CoPhysics Report (at 25-27, in its "Key Issue #3", and also at 17-18) recognizes the issue, namely, that the entrance monitors detect only gamma radiation, most of which is from radium progeny rather than radium itself, which "could result in truckloads of drill cuttings carrying the same amount of radium to have a 60-fold difference in radium-226 measurements at the Landfill gate." Id. at 25. Despite this recognition, the CoPhysics report claims that "The truck monitoring technique is highly proven and reliable" (id. at 17). The CoPhysics report's defense of the entrance monitors relies mainly on an exaggerated depiction of the issue ("the gaseous decay product [progeny] of radium, namely radon-222, emanates in vast amounts out of open-top truckloads," id. at 25) and on the professional experience of Dr. Rahon of CoPhysics:

> I can assert with a high degree of professional confidence that there are easily detectable and quantifiable gamma rays emitted from soil even while it has been exposed to air over a long time. Id.
>
> Considering that a truckload of drill cuttings is not spread out and mixed, I estimate that greater than 90% of the gamma-emitting progeny of radium is present in a load (that is, less than 10% of the radon in a load is lost to the atmosphere). Id. at 26.
>
> Based on my extensive degree of professional experience in these matters, I find the commentators' contentions to be unfounded. In my professional opinion, (1) the use of gamma radiation detectors is an entirely appropriate method to screen incoming waste loads (and, in fact, is the state-of-the-art industry standard that has worked in practice at numerous landfills, including Casella's facilities), and (2) application of the conversion factor in the CoPhysics Truck Monitor Correlation Study is valid and results in fully accounting for the amount of radium in the incoming truckload. Id. at 27.

All of these defenses of the efficacy of entrance monitors miss the point of *deliberately manipulated* truckloads of waste. The CoPhysics report also says that

> While it is true that 50% or more of the radon and its gamma-emitting decay products can be released from a ground-up, well-aerated and oven-dried soil sample, as described above, truckloads of drilled rock certainly are not aerated and oven dried.

Id. at 26.

Again, these arguments about "not aerated and oven dried" miss the point of waste truckloads being *deliberately manipulated to remove radon* in order to pass through the landfill entrance monitor while carrying substantially more than 25 pCi/g of radium-bearing waste. As noted above, an efficient way to remove radon would not be aeration or oven-drying, but, instead, covering the waste load with a tarp and applying a partial vacuum with suction from a readily available shop-vac type of vacuum cleaner.

Another CoPhysics response on the entrance monitors involves the many truckloads of drilling waste that have *not* triggered the entrance monitors and the incorrect/misleading claim that "…Casella records show direct proof that the monitors work." Id. at 17. The alleged proof is that the entrance alarms at three Casella landfills *have* sounded a total of six times since being installed in 2010, with all six incidents traceable to nuclear medicine procedures or waste materials from such procedures. This information, if true,[42] is irrelevant to the entrance monitors' ability to detect and quantify radium. The sensitivity of the entrance monitors is not the main issue here. The issue is radium and whether/how it can be detected and properly quantified if accompanied by *widely variable concentrations* of its progeny which are strong gamma emitters.

CoPhysics also makes the following claims about the entrance monitors:

> As to the underlying premise that drill cuttings contain low levels of radioactivity, further review of Casella records shows that since January 1, 2011, more than 500,000 tons of waste from the oil and gas industry have been accepted collectively at the Hakes, Hyland and Chemung Landfills. None of these waste loads have triggered the alarms. The fact (1) is consistent with the findings in initial radioactivity studies that there are low radium levels in drill cuttings (i.e., at worst, just slightly above background); and (2) further demonstrates that the industry has been effective in keeping unauthorized drilling wastes (e.g., sludges, scales, etc.) out of their drill cutting trucks.
>
> Leachate monitoring redundantly confirms that drill cuttings (including drill cuttings from Marcellus shale at Hyland and Chemung) do not result in significant radium levels in leachate and disposal of these materials in Part 360 landfills does not create any public health risk....

CoPhysics Report at 17-18.

These claims are misleading and fail to address the relevant questions. Whether waste loads have triggered the alarms, and whether unauthorized drilling wastes have been kept out, is dependent on whether the radiation monitors at the gates are capable of detecting exceedances of the 25 pCi/g limit. This has been discussed above. Leachate monitoring can't "redundantly" confirm anything useful as long as the intermittently high radionuclide levels remain

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[42] The CoPhysics description – stating that nuclear medicine isotopes rather than radium were involved in all six instances of entrance monitor alarms being triggered – is contradicted by DEC's description of the 2016 incident at Hakes where radium in a radio-luminous ship deck marker triggered the alarm.

unaddressed and unexplained. Most importantly, leachate monitoring can't confirm that there's no public health risk from drill-cutting disposal in Part 360 landfills until public-health (and occupational-health) risks from radon and its progeny have been satisfactorily identified and addressed.

C. The DSEIS fails to evaluate the presence of radon gas in the landfill's air emissions, gas collection system emissions, and emissions from flaring

The Sierra Club comment letter at 7-8 notes that the landfill is not testing for emissions of radon gas from the landfill surface or from the landfill's internal gas collection system; that radon is not flammable and will be released through the landfill's gas collection system during flaring of other landfill gases; that this concern about adverse effects on the environment and the health and safety of people, animals and plants living near the landfill is accentuated by the landfill leachate radionuclide test results which indicate that, at least at certain times, there is approximately 1.05 million pCi/liter of Radon-222 in the air within the landfill.

DEC's response (responses B-3a and B-3b, FSEIS at 18-19) argues that, while there is no standard for radon emissions from landfills, potential radon emissions from drilling-related wastes were part of the studies done by Argonne National Laboratory which showed that modeled radon exposures (assuming Radium-226 activity/concentrations of 50 pCi/g in the landfill waste mass) were below allowable limits. This fails to address the site-specific comment about ~1.05 million pCi/L radon in the air within the landfill. DEC's response thus fails to address the adverse effects of such radon, which cannot plausibly be assessed without testing and modeling. See also discussion above about the shortcomings of the Argonne studies by Smith et al. and Harto et al. DEC also claims that "Radioactive waste is not accepted at the landfill," but this begs the question of whether the Hakes landfill contains unacknowledged radioactive material or waste that substantially exceeds 25 pCi/L.

The CoPhysics Report (at 28-33, in its "Key Issue #5") acknowledges that some of the submitted comments express a concern about high levels of Radon-222, including radon in the air/landfill gas mixture that could have been as high as 1.05 million pCi/L:

> Commentators assert that there is a "trend" of increasing radioactivity in leachate from the Hakes (and Chemung) Landfills, with particular concern being high levels of radon-222. They point to a sample from cell #5 taken in 2014 and another sample from cell #8 taken in 2017, which measured approximately 6000 pCi/L of bismuth-214 and lead-214 (which are progeny of radon-222). Since, after collection, the samples had been sealed for 21 days to reach equilibrium, commentators assert that given the short half-life of parent radon-222 (3.8 days), there had to be levels of radon-222 much higher than 6000+ pCi/L when the samples were collected. Back-calculating to account for the decay of radon-222 (from the time of sample collection to 21 days thereafter), commentators assert that the radon level at the time of sampling was 275,000 pCi/L. They also assert that this means that radon in the air/landfill gas mixture could have been as high as 1.05 million pCi/L.

> With respect to radon, commentators express environmental/health concerns regarding the effects of high levels of radon in leachate and airborne emissions (flaring, vents, downwind effects, "nuclear fallout"). Specifically, they assert that radon-222 associated with the disposal of drill cuttings at the Hakes and Chemung Landfills (airborne and in leachate) presents an increased environmental risk and enhanced cancer risk to the public, as well as an increased risk of birth defects and a shortened life span (for example, due to inhalation, migration through groundwater, drinking water wells, taking hot showers, migration into basements, and dust particles).
>
> In addition, some commentators express concern that the longer-lived progeny of radon-222, lead-210 and polonium-210, in leachate from the Hakes and Chemung Landfills present health concerns and should be characterized to prevent adverse environmental impacts. And, commentators attribute the asserted elevated levels of radioactivity to the disposal of drill cuttings.

The CoPhysics Report responds by acknowledging the high levels of Bismuth-214 and Lead-214 in leachate samples collected from Hakes Cell #5 on 11/11/14 and Hakes Cell #8 on 6/6/17, and by recognizing that back-calculating (decay-correcting) from the analysis time to the time of sample collection would, if valid, imply an approximate Bismuth-214, Lead-214 and Radon-222 concentration of 275,000 pCi/L. However, the CoPhysics Report proceeds to make various arguments that have already been refuted in this memo (such as the alleged unreliability of EPA Method 901.1 for water analysis, the claim that "leachate sampling and analysis methods were never designed to be used for radon assessment," and the claim of "so much uncertainty in this type of analysis that, to make a decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels"). The CoPhysics Report thereby reaches an incorrect conclusion that "the 275,000 pCi/L calculation cannot be relied upon as an accurate estimation of radon and progeny in the original on-site samples." After comparing "the 275,000 pCi/L calculation" to allowable sewer discharge levels (not directly relevant to the issue of 1.05 million pCi/L radon *in landfill gas*), the CoPhysics Report acknowledges that "The unusual results are certainly reason to conduct further investigation of the issue" but immediately follows this acknowledgment with the incorrect claim (refuted above) that Lead-210 analysis has already demonstrated "that the back-calculated radon result (275,000 pCi/L) is unreliable."

After further discussion of allowable releases to sewers, the CoPhysics Report returns briefly to radon in soil gas and notes that 63% of the homes in Steuben County "have natural radon concentrations exceeding the USEPA guideline." The CoPhysics Report then says (at 31-32) that

> …Part 380 further confirms that the radon at issue here is NORM, part of natural background and not subject to regulation. 6 NYCRR 380-l.2(c) & (e); 6 NYCRR 380-2.l(a)(8).
>
> As for commentators' concerns regarding airborne releases of radon from the Landfill, allegations of downwind "nuclear fallout" effects, and claims of radon migration and infiltration into people's homes from the Landfill, the physical properties of radon and

> controls/procedures in place at the Landfill belie any such impacts. Regarding dust-related and other airborne emissions of radon, landfills (including the Hakes Landfill) are not significant sources of downwind airborne radon. Soil cover used at the Landfill is clean soil and also has nothing to do with the drill cuttings, muds, and C&D waste being accepted for disposal. Airborne emission of radon-222 from the Landfill is simply a natural occurrence from the native, local soils used as a cap, just as would happen in an open field. However, there is a confounding feature of landfills that does not exist in open fields: namely, landfill gas vents and flares. While there certainly would be higher concentrations of radon in the gas released from vents and flares, the volume of gas released is very small. These small but concentrated discharge points when averaged over the area of the Landfill do not pose a downwind hazard due to rapid mixing and dilution in the atmosphere. This point source dilution is the same technique used for residential radon mitigation systems where high levels of radon from the sub-foundation are discharged through a pipe above the home's roof line. Relative to the radon exposure occurring in homes with or without mitigation systems, downwind radon exposures from landfills are extremely minor.
>
> Indeed, given radon’s physical properties (for example, short half-life, rapid mixing/dilution in the atmosphere), to the extent local residents' homes were found to have enhanced levels of radon, such would result from radon emanating from soils on the homeowner's own property, and not the migration of radon from the Landfill hundreds of yards away. And, as already noted, per Part 380, radon is NORM, part of natural background, and not subject to regulation as a radioactive material under Part 380. 6 NYCRR 380-l.2(c) & (e); 6 NYCRR 380-2.l(a)(8); Part 380 Public Comment Assessment, Response 17-5 (which states that "[t]he constraint on radioactive emissions in Part 380 [radiation dose constraint for airborne emissions] does not include NORM, such as radon"); Responses 14-1, 15-4, 15-5, 15-6, 15-17 (all of which state that Part 380 regulates TENORM, not NORM).

These comparisons between Part 380 and Parts 360-363, and between their respective requirements for TENORM and NORM, beg the fundamental issue raised in Sierra Club’s comments, namely, whether the leachate test results combined with the unreliability of the entrance monitors indicate that the Hakes landfill contains unacknowledged TENORM and/or NORM that substantially exceeds 25 pCi/L.

The above-quoted text from the CoPhysics Report (at 31-32) also makes several claims that are entirely conclusory:

- “...landfills (including the Hakes Landfill) are not significant sources of downwind airborne radon”
- “While there certainly would be higher concentrations of radon in the gas released from vents and flares, the volume of gas released is very small.”
- “These small but concentrated discharge points when averaged over the area of the Landfill do not pose a downwind hazard due to rapid mixing and dilution in the atmosphere.”

- "Relative to the radon exposure occurring in homes with or without mitigation systems, downwind radon exposures from landfills are extremely minor."

These assertions do not resolve the significant points at issue, especially since landfill-gas emissions from an expanded Hakes landfill may triple from their current rate, from ~250 scfm to ~750 scfm. (See DSEIS, Appendix H, at 7.) As indicated in the submitted comments, all of the above points need to be resolved by testing and modeling, not by mere assertion.

Finally, the CoPhysics Report returns to "potential impacts from radon's longer lived progeny, lead-210 and polonium-210" but does not address those impacts. Instead, the CoPhysics Report makes the incorrect argument (refuted above) that the "the relatively low lead-210 results serve to prove that the actual radon levels in the leachate are far less than the very rough calculational level of 275,000 pCi/L, and the results of EPA 901.1 should be rejected."

The CoPhysics Report concludes its response to the issue of intermittently high radon in landfill gas as follows:

> There are certainly elevated levels of radon in leachate as there are in any groundwater sample, but not to the extent suggested by the commentators. Finally, in my professional opinion, which I can assert to a reasonable degree of scientific certainty, there is "no cause and effect" between the disposal of drill cuttings and radon-222 levels in leachate. Rather, radon is a natural occurrence and not the result of drill cuttings being disposed there. The drill cuttings in the Hakes Landfill account for less than 11 percent of the waste and overburden deposited to date at the facility and, at worst, have only slightly higher radium levels than background; thus, drill cuttings are only a small portion of the source of the radon. To the extent that radon-222 exists in the leachate, this is a natural occurrence due to both the native, local soils, clay and gravel that are used to construct the Landfill and the C&D materials deposited in the Landfill (such as brick, sheetrock, concrete block, wood ash, coal ash, etc.), which are also sources of radon. The radon in leachate comes from all of these Landfill constituents which have radium concentrations on the order of 1 to 10 pCi/g. And, we know from the NYS Department of Health's published data, this whole region is prone to elevated radon levels. Also relative to the Hakes Landfill, the concentration of radon in leachate may be further enhanced by the fact that the Landfill materials have more air spaces than does undisturbed soil. Natural radon would build up in these spaces, and this is so whether the surrounding material is drill cuttings, C&D waste, gravel or fluffed native soil. As rainwater infiltrates through these spaces, radon dissolves into the water more so than rainwater infiltrating through native, settled undisturbed soil. Therefore, some level of radon concentration in Landfill leachate should be expected.
>
> In the end, it is important to reiterate that the naturally-occurring levels of radioactivity in the leachate are due to all of the materials in the Landfill, including the indigenous soil and rock from the Steuben County area and C&D materials such as brick, sheetrock, concrete block, ash, drill cuttings, etc. These are all sources of radium and radon

> contributing to that found in the leachate, and there is no scientific basis to conclude that the measured levels are a result of solely drill cutting disposal.

This CoPhysics response to the issue of intermittently high radon in landfill gas is primarily a mixture of conclusory statements ("not to the extent suggested by the commentators" and "no cause and effect") and unsupported claims about the radon in question being "a natural occurrence." The more substantive CoPhysics claims that "the concentration of radon in leachate may be further enhanced by the fact that the Landfill materials have more air spaces than does undisturbed soil" and that "Natural radon would build up in these spaces" cannot explain high radon ranging up to ~1.05 million pCi/L. The accumulation of natural radon in air spaces is bounded by both theoretical limits and the empirical evidence from uranium mines, as discussed above in section II.G. It is well-known that radon builds up in air spaces at levels exceeding 4 pCi/L, but not at levels approaching 1 million pCi/L unless the surrounding soil and/or wastes contain far more than 25 pCi/g of radium.

Last but not least, it is important to recall the siting criteria for radioactive waste disposal which disallow disposal sites where existing conditions (in this case, high radon ranging up to ~1.05 million pCi/L) would mask any meaningful monitoring program. Meaningful monitoring should, for example, be able to distinguish whether Part 380 wastes are being improperly placed in Part 360-363 landfills. And even if further testing shows that C&D materials deposited in the landfill (brick, sheetrock, concrete block, wood ash, coal ash, etc.) are the main sources of radon within the landfill, such a finding could not reasonably be deemed acceptable without additional testing and modeling to determine fate, transport, and downwind impacts from radon levels that are either intermittently or continually as high as 1.05 million pCi/L.

D. The DSEIS fails to evaluate the possible presence of radium, radon and their breakdown products in the landfill's stormwater discharges, groundwater suppression system discharges or liner leakage discharges

The Sierra Club comment letter at 8-9 questions whether the DSEIS claim of "no groundwater contamination…" can be considered meaningful since neither the DSEIS nor the landfill's environmental monitoring protocols indicate any intention of testing for radium, radon or their breakdown products in ground- and surface water near the landfill. The comment notes that the positive declaration issued by DEC for the landfill expansion project acknowledged the potential for "significant impacts to groundwater requiring the design and construction of a landfill liner and leachate collection and leak detection systems," and acknowledged that the resulting construction and placement of waste in proximity to the existing water table requires that a groundwater suppression system be designed and installed. The Sierra Club comment then refers to the DSEIS which describes the landfill's groundwater and surface water monitoring program, states that the monitoring program is capable of detecting potential impacts to ground- and surface water before they can have a significant adverse impact on the environment, and claims that "To date, no groundwater contamination has been detected related to the operation of the lined cells." In light of the landfill's leachate test results, it is a serious omission for the DSEIS not to address the adequacy of the landfill's ground- and surface water monitoring programs with respect to the radionuclides that are known to be present.

DEC's response (response B-4, FSEIS at 19) claims that:

> The acceptance of drill cuttings, the waste that prompted this comment, is not a new waste stream for the Hakes landfill. There is no change as part of this action related to waste types being accepted for disposal. Therefore, the type of waste being accepted and their potential impacts are outside of the scope of this EIS.
>
> The current Part 360 Hakes landfill permit already requires semi-annual leachate monitoring for radiological content, specifically for radium-226. If the landfill liner has a leak, it would be identified by the presence of Part 360 regulated landfill constituents in groundwater /or surface water samples which are obtained and analyzed quarterly in accordance with the EMP. Monitoring to date has not identified exceedances of groundwater standards for landfill constituents. If there was a buildup of NORM constituents in the landfill, it would be observed in leachate well before it was observed in the groundwater samples.
>
> Stormwater discharges from the facility do not pass through the waste mass, but are instead generated from waters diverted around the site, surface areas already completed with final cover, and other facility areas (e.g., roads). As such, it is not necessary to monitor stormwater runoff for radiological content. Such discharges are subject to the requirements of the SPDES Multi-Sector General Permit for Stormwater Discharges associated with Industrial Activity, Sector L (GP-0-17-004).

There are several problems with this response, starting with the above claim that "it is not necessary to monitor stormwater runoff for radiological content." This claim that monitoring "is not necessary" is contradicted by DEC's response B-7 (FSEIS at 20) which says that, "In the unlikely event of a leak in the liner system, leachate would ultimately migrate to the storm water system or groundwater collection system, which are both monitored."

Other problems include DEC's claim that any liner leak "would be identified by the presence of Part 360 regulated landfill constituents in groundwater /or surface water samples which are obtained and analyzed quarterly." This response does not address the portion of Sierra Club's comment about radiological groundwater contamination – and whether it can be detected, given the fact that "neither the DSEIS nor the landfill's environmental monitoring protocols indicate any intention of testing for radium, radon or their breakdown products in ground- and surface water near the landfill." If DEC's logic is that groundwater testing for chemical substances would reveal liner leakage even if no radionuclide testing is done, then this rationale should be clearly stated. Such rationale needs to include a quantitative sensitivity analysis to rule out the possibility that combined leakage of radionuclides and non-radioactive chemical substances may occur without exceeding thresholds for the tested chemicals. Radiological testing may be the more sensitive test, capable of detecting combined leakage at levels that may not exceed chemical test thresholds yet may be problematic with respect to radionuclide leakage.

DEC's claim that the acceptance of drill cuttings "is not a new waste stream for the Hakes landfill" does not address the concern that the 25 pCi/g waste acceptance limit may not have been honored. This concern, discussed at length above, is based partly on the intermittently high leachate results that remain unexplained. The concern originates from wastes that have already been landfilled but also applies to wastes that would be accommodated in the future by landfill expansion.

E. The DSEIS fails to evaluate the adequacy of the landfill's liner system and groundwater suppression system to protect against the radium, radon and their breakdown products present in the landfill from entering groundwater and surface water supplies adjoining the landfill

The Sierra Club comment letter at 9-10 refers to the DSEIS which states that the landfill's composite liner system is the main source of protection against groundwater contamination by the landfill but which fails to mention the liner's annual leakage rate, its expected life, and the risks of harmful exposures from its failure. The comment notes that C&D landfill liner systems are much less substantial than those used in landfills accepting low-level radioactive wastes, and that the DSEIS provides no risk analysis nor any evaluation of the types of environmental hazards the liner system can withstand or the circumstances in which the integrity of the liner system could fail. Given the numerous bodies of water and wetlands surrounding the landfill, and given the DSEIS's acknowledgment of landfill drainage to the Corning aquifer – the primary drinking water supply for the Corning-Painted Post metropolitan area – the DSEIS should have evaluated the adequacy of the landfill's liner system to protect these ground- and surface water resources from the radium, radon and their breakdown products present in the landfill.

Furthermore, the Sierra Club comment notes that, according to the DSEIS, Hakes is requesting a variance from the requirement in the solid waste regulations that the base of a landfill disposal cell be at least 10 feet above underlying bedrock. Given the reliance of the DSEIS on the effectiveness of a yet-to-be-constructed groundwater suppression system (which would nominally support the landfill's request that only a five foot of separation from bedrock be allowed), the Sierra Club comment expresses concern about the absence of any risk analysis and/or other evaluation in the DSEIS of the types of environmental hazards the groundwater suppression system can withstand or the circumstances in which the groundwater suppression system could fail. As noted in the comment, if the liner system or the groundwater suppression system were to fail, those failures may allow radium, radon and their breakdown products to be released from the landfill into surrounding water bodies and the environment. Such risks should have been, but were not, evaluated in the DSEIS.

DEC's response (response B-5, FSEIS at 19) simply refers readers to its previous B-4 response which is quoted above in relation to the preceding comment. There are several additional problems with this response with respect to the present Sierra Club comment, including DEC's failure to acknowledge Sierra Club's comments about:

- the liner's annual leakage rate which has not been identified (quantified) by DEC,
- the expected life of the liner,

- the risks of harmful exposures from liner failure,
- the lack of risk analysis or other evaluation of the types of environmental hazards the liner system can withstand or the circumstances in which the integrity of the liner system could fail,
- associated impacts to ground- and surface water resources, including wetlands, and
- the requested variance from the regulatory requirement that the base of a landfill disposal cell be at least 10 feet above underlying bedrock.

These issues have not been adequately addressed.

F. The DSEIS fails to evaluate the risk that opening up the landfill to tie-in the proposed expansion will create new pathways for radon and radium in the landfill to be released to the environment

The Sierra Club comment letter at 10 notes that the positive declaration issued by DEC for the landfill expansion project acknowledged that "[t]he project includes expansion of a solid waste management facility of large magnitude. It may result in the unearthing of C&D material as the new expansion is tied in with the existing facility. Bulk leachate storage and appropriate ongoing measures to prevent releases will be discussed." Despite this acknowledgment, there is no discussion in the DSEIS of the process by which the new expansion is proposed to be tied in with the existing landfill. DEC's comment notes that, although the process of opening up the existing landfill to tie in the new expansion will inevitably create additional pathways for radium, radon and their breakdown products in the landfill to be released into the environment, the DSEIS contains no risk analysis and/or other evaluation of the types of environmental exposures that may result from opening up the landfill for the tie-in process.

DEC's response (response B-6, FSEIS at 19-20) simply says that, "Based on an analysis performed by Argonne National Laboratory, even at radioactive concentrations of 50 pCi/g (twice the allowable acceptance limit set by the Department), potential public doses from all pathways were still below allowable dose limits," and also refers readers to its response B-3a.

The main problem with these DEC responses is their unfounded reliance on the Argonne studies by Smith et al. and Harto et al. See above discussion of the shortcomings of these two studies, including the fact that they do not acknowledge or consider direct airborne impacts from radon flux through the landfill cap.[43] See also the above-quoted findings by Walter et al. that in some of the relevant disposal scenarios, "the radon flux from the landfill and off-site atmospheric activities exceed levels that would be allowed for radon emissions from uranium mill tailings" and that "...the simulated radon fluxes for the scenarios analyzed exceed the uranium mill tailings regulatory flux limits for the cases without a geomembrane cover, and exceed the off-site radon activity limit in all cases."[44] Given the similarity of the Walter et al. study to the two Argonne studies, and given the fact that the 50 pCi/g waste acceptance limit in all three studies was only twice the nominal 25 pCi/g limit at Hakes, and especially given the various unanswered

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[43] Smith et al., op. cit., at 34; Harto et al., op. cit., at 21.

[44] Walter et al., op. cit., at 1040 and 1048.

questions at Hakes, DEC's claim that "potential public doses from all pathways were still below allowable dose limits" cannot be considered protective and applicable. The unanswered questions at Hakes are very interrelated with radon pathways, which in turn are dependent on openings in the landfill cap. DEC's generalized response hasn't addressed such openings. They need to be addressed in a site-specific manner as part of the measurement-and-modeling-based assessment of radon pathways and health effects.

G. The DSEIS fails to evaluate the risk that the fires that have been occurring at the landfill have damaged the landfill's liner system, gas collection system or leachate collection system and have created or will create new pathways for radon and radium in the landfill to be released to the environment

The Sierra Club comment letter at 11 notes that:

> The DSEIS acknowledges that the landfill "has experienced both surface and subsurface fires." The DSEIS does not describe how many fires have occurred, in which cells the fires have occurred, how long the fires have lasted, or whether any fires are still burning within the landfill. The DSEIS does not evaluate whether the fires could damage or have already damaged the landfill liner system, gas collection system or leachate collection system and thereby create new pathways for radon and radium in the landfill to be released to the environment. Other than stating that the fires "could impact air resources by the release of smoke and other combustion products," the DSEIS contains no risk analysis and evaluation of the types of environmental exposures that could result from damages caused to damage the landfill liner, the gas collection system or the leachate collection system by the landfill fires….

DEC's response (response B-7, FSEIS at 20) simply says that, "In the unlikely event of a leak in the liner system, leachate would ultimately migrate to the storm water system or groundwater collection system, which are both monitored." This statement appears inconsistent with DEC's response B-4 (FSEIS at 19) which states that "it is not necessary to monitor stormwater runoff for radiological content" and is also misleading because leaks in the liner system are not "unlikely" (see overview of leaks in the Argonne report by Smith et al.[45] that is cited by DEC in the FSEIS). If the single composite liner system at the base of the Hakes landfill, consisting of a 60-mil textured HDPE geomembrane underlain by a compacted clay liner at least 2 feet thick, is substantially less prone to leakage than the liners described in the Argonne report by Smith et al. that DEC relies on, then DEC's response should address this in more specific and quantitative language than "unlikely."

DEC's response also refers readers to its responses C-1 through C-4 for information pertaining to landfill fires and the integrity of the landfill liner system, and to B-4 (already discussed in the preceding paragraph) for potential liner leaks. These referenced responses refer, in turn, to other

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[45] Smith et al., op. cit., at 43 and 67.

responses such as G-5 for airborne resources impacts due to fires. Responses B-11 and B-15 are also cited for radiation-related issues (but their deficiencies have already been reviewed above). In these various responses, DEC acknowledges that landfill fires are "not an acceptable condition" under NYS solid waste management regulations, particularly 6 NYCRR § 215.2 and the conditions of a facility's Air State Facility permit. DEC refers to both surface and subsurface fires, saying that surface fires are promptly fought and extinguished, while subsurface fires (or "thermal oxidation") are monitored closely. If subsurface fires persist for more than a few days, DEC's response G-5 says that "injection wells/monitoring points are installed throughout the area. Water or leachate is introduced into the buried waste to cool, contain and extinguish any thermal oxidation." FSEIS at 43. DEC also says in response C-1 that:

> Once the temperature and the CO [carbon monoxide] return to safe levels for operation, the water or leachate introduction is suspended. Leachate is then collected and sent to a wastewater treatment plant. The integrity of the liner is evaluated based on the monitoring of the groundwater and stormwater at the site.

FSEIS at 31.

Whether the integrity of the liner can be evaluated based on groundwater and stormwater monitoring depends on two points noted above: **A**) Whether radiological testing is a more sensitive test than chemical analysis for a given mixture of radionuclides and non-radioactive substances, and thus more capable of detecting leakage through a damaged liner. This point has not been addressed but needs to be. **B**) Whether groundwater and stormwater are, or are not, tested for radionuclides. DEC's responses B-4 and B-7 appear inconsistent on this point.

DEC's response C-1 continues, and responds to part of Sierra Club's comment, as follows:

> At this point, there have been no leachate indicators leaving the site, indicating that no damage to the liner has occurred from a fire. This monitoring will continue during operation as outlined in the facility's Environmental Monitoring Plan.
>
> In 2014, concerns regarding potential damage to the liner system after a subsurface fire prompted an investigation into the liner system integrity. The landfill was excavated in an area where a subsurface fire was suspected of being in contact with the top of the leachate collection layer. The waste and leachate collection drainage layers were removed exposing the top of the geomembrane liner. There were no signs of distress (i.e., holes or obvious defects) in the geomembrane liner indicating that the leachate collection stone provided protection to the underlying layers. The rest of the isolated fires were elevated in the waste mass away from the landfill liner system.
>
> The landfill liner system and gas collection system are periodically monitored to evaluate their integrity. Monitoring includes the following activities;

- The gas collection system is monitored quarterly for several parameters including; temperature, oxygen, carbon dioxide and methane. These parameters can provide early warning signs that conditions are changing that may impact the gas collection system. To date, there have been no signs that the gas collection system has been impacted by the fires.

- The groundwater collection system beneath the base of the liner system is sampled and tested quarterly. The groundwater collection system analytical results would provide an early indication if the liner system was not operating effectively. To date, there has been no indication that the liner system has been compromised by the fires.

- The leachate collection system is cleaned annually. A high-pressure nozzle and hose are passed through each leachate pipe as water is used to flush and clean the pipes. There have been no blockages or signs that the pipes have been impacted by the fires.

Id. at 31-32.

One of the main points raised in the Sierra Club comment letter (that fires "have created or will create new pathways for radon and radium in the landfill to be released to the environment") remains unaddressed pending the measurement and monitoring that need to be done to characterize the landfill pathways through which radon moves. DEC's response G-5, while acknowledging the possibility of fires "which could impact air resources by the release of smoke and other combustion products," reaches a faulty conclusion that there would be "no significant release of radioactive air emissions due to fire" based on the logic that drill cuttings are not combustible. FSEIS at 42-43. The problem with this logic is the continually or intermittently high level of radon in Hakes landfill gas, ranging up to ~1 million pCi/L, which has not been addressed by DEC. While the methane present in landfill gas[46] will tend to burn and be consumed during a surface or subsurface landfill fire, the commingled radon in the landfill gas will continue through the combustion zone and be part of the visible smoke or any less visible plume of combustion products. The same is true, of course, for the routine flaring of landfill gas. Radon in the landfill gas continues through the flare and moves downwind with the combustion products.

H. The DSEIS fails to evaluate the health impacts of the landfill expansion project

The Sierra Club comment letter at 12-14 notes that, although the DSEIS states that "[a] major public concern regarding the construction or expansion of any solid waste facility is potential adverse impact on human health and the environment," it fails to provide any meaningful analysis of the potential adverse impact on human health and the environment of the landfill expansion project. In particular, it fails to evaluate human and environmental health risks from exposure to the levels of radioactivity shown by the leachate test data to be already present in the landfill or to evaluate the risks of the additional environmental exposures that would result from accepting additional levels of radioactive waste in the landfill. As a basis for comparison, the comment quotes New York's low-level radioactive waste disposal facility regulations which provide that "[c]oncentrations of radioactive material which may be released to the general

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[46] The landfill gas contains at about ~31% methane, according to the DSEIS at 36.

environment in groundwater, surface water, air, soil, plants or animals must not result in an annual dose exceeding an equivalent of 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public. Releases of radioactivity in effluents to the general environment must be maintained as low as reasonably achievable." 6 NYCRR 382.11. The comment notes that the DSEIS provides no evaluation of potential exposures to radioactivity from radionuclides in the Hakes landfill. This omission is serious, as indicated by Dr. David Carpenter in the following portion of his affidavit that was quoted in the Sierra Club comment:

> [b]ased on the information provided to me and my knowledge of the human health effects arising from exposure to ionizing radiation, I have concluded that: (a) there are substantial and significant risks to human health posed by the current procedures used at the Hakes Landfill and approved by NYSDEC, (b) while the greatest threat to human health comes from inhalation of radon-222, other naturally occurring radioactive material (NORM) and the progeny of these elements pose significant threats to human health, and (c) inhalation is the route of exposure of greatest concern but other routes (ingestion, dermal absorption) are also possible.

For these reasons, the DSEIS should have provided an evaluation of potential exposures to radioactivity from the Hakes landfill and its proposed expansion. *The FSEIS does not remedy this defect*. Its argument on pages 55-56 that expansion would not increase the landfill's daily waste tonnages fails to acknowledge that landfill-gas emissions may triple from the current rate, from ~250 scfm to ~750 scfm (DSEIS, Appendix H, at 7), and misses the point that every additional amount of radium introduced to the landfill will add to the quantity already there.

The study by Walter et al. discussed above provides further support for the need for site-specific investigation of health effects, especially in view of the fact that the study's waste acceptance limit is only twice the nominal 25 pCi/g limit at Hakes. Walter et al., in their modeling of municipal solid waste landfills that accepted up to 50 pCi/g radium-bearing waste, found for some of their disposal scenarios that "the radon flux from the landfill and off-site atmospheric activities exceed levels that would be allowed for radon emissions from uranium mill tailings" and that "...the simulated radon fluxes for the scenarios analyzed exceed the uranium mill tailings regulatory flux limits for the cases without a geomembrane cover, and exceed the off-site radon activity limit in all cases."[47] Given these findings by Walter et al. in combination with the Hakes leachate test results, it is essential to characterize the landfill's radon impacts – and also its long-term radium impacts – or to engage NYS Dept. of Health to do so as a cooperating agency.

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[47] Walter et al., op. cit., at 1040 and 1048.

Exhibit List

A) Excerpts from documents relating to EPA Method 901.1 uncertainty (with RV comments).

B) Excerpts from 6/6/17 Cell 8B leachate data.

C) D.E. McCurdy, J.R. Garbarino, and A.H. Mullin, *Interpreting and Reporting Radiological Water-Quality Data*, Techniques and Methods book 5, chapter B6, USGS Office of Water Quality, National Water Quality Laboratory (2008).

Exhibit A

DEC received more than 13,000 public comments on its High-Volume Hydraulic Fracturing Draft Supplemental Generic Environmental Impact Statement (Draft SGEIS) issued in September 2009 (ftp://ftp.dec.state.ny.us/dmn/download/OGdSGEISFull.pdf).

In response to issues raised, DEC prepared and released for public review a Revised Draft SGEIS on September 7, 2011 (http://www.dec.ny.gov/data/dmn/rdsgeisfull0911.pdf). On page 6-65, this RDSGEIS says: "However, based on the analytical results from field-screening and gamma ray spectroscopy performed on samples of Marcellus Shale, NORM levels in cuttings are not likely to pose a problem because – as set forth in Section 5.2.4.2 – the levels are similar to those naturally encountered in the surrounding environment." DEC held four additional public hearings around the state and received another 67,000 comments.

After release of this second draft, DEC proposed regulations to supplement and reinforce the proposed permit conditions and received 180,000 public comments. In all, DEC received 260,000 public comments on the SGEIS and the proposed regs. (The proposed regs have lapsed.)

DEC FSGEIS on the Oil, Gas and Solution Mining Regulatory Program [*a.k.a. High-Volume Hydraulic Fracturing Environmental Impact Statement*], Vol. 1, May 2015 (https://www.dec.ny.gov/docs/materials_minerals_pdf/fsgeis2015.pdf) contains no mention of EPA Method 901.1 or measurement of radionuclides in landfill leachate within its 1448 pages. It notes that analytical results from gamma ray spectroscopy are useful for determining radionuclide levels in Marcellus Shale samples from drill cuttings (see page 16 of Exec. Summary; also page 5-29 and Tables 5.2(a) and 5.3) but otherwise contains no discussion of the uses and/or limitations of gamma ray spectroscopy. The word "leachate" appears only once, in the context of whether tanks or impoundments are preferred for storage of flowback water, and there is no mention or discussion of analytical methods applicable to leachate. [*Note by RV: These conclusions are based on searches for "901" and "spectro" and "leachate." The absence of any discussion of radionuclide analytical methods applicable to landfill leachate is not surprising, given the fact that this FSGEIS was directed to the then-pending decision of whether High-Volume Hydraulic Fracturing would be allowed within New York State. Impacts of landfill disposal of drill cuttings were not a central issue in this FSGEIS.*]

DEC FSGEIS on the Oil, Gas and Solution Mining Regulatory Program [*a.k.a. High-Volume Hydraulic Fracturing Environmental Impact Statement*], Vol. 2, Response to Comments, May 2015 (https://www.dec.ny.gov/docs/materials_minerals_pdf/fsgeis2015rtc.pdf), page RTC-103, says: "The Department does not agree that gamma spectroscopy is an insufficient method by which to determine NORM concentrations in various media. Gamma spectroscopy is broadly recognized as an efficient and accurate method to quantify radium in environmental samples. Radiological analytical laboratories have been quantifying environmental levels of radium and other NORM constituents successfully using gamma spectroscopy for decades. Claims that alpha spectroscopy or delayed-neutron analysis is required to adequately quantify NORM content are based on a misunderstanding of the difference between a radioactive isotope such as radium-226 and the subatomic radioactive particles it emits. The isotope radium-226 gives off gamma rays, beta particles and alpha particles as it decays. However, none of this emitted radiation is present without the actual radium itself. Such radiation exists for infinitesimally short periods of time before their energies are absorbed into surrounding materials. Therefore, although alpha radiation poses the greatest risk of the various subatomic emissions from radium, the simpler, less costly and faster gamma spectroscopy method is adequate to determine the overall concentration of radium present, and thus the risk posed by all types of radioactive emissions, including alpha particles. Moreover, the discussion of analytical methods in the SGEIS is accurate and sufficient as it employs generally accepted analytical methods for NORM."

Id., Vol. 2 (https://www.dec.ny.gov/docs/materials_minerals_pdf/fsgeis2015rtcfull.pdf), Appendix A, includes a list and summary of the "Supplemental Literature" considered by NYS Dept. of Health for its Public Health Review. One of the documents listed and summarized by NYS DOH, on page RTC A-165 ff. of this FSGEIS appendix, is an EPA report (B. Schumacher et al., *Development of Rapid Radiochemical Method for Gross Alpha and Gross Beta Activity Concentration in Flowback and Produced Waters from Hydraulic Fracturing Operations*, EPA/600/R-14/107, July 2014) wherein coauthors Marinea Mehrhoff (State Hygienic Laboratory at the University of Iowa) and Michael Schultz and Andrew Nelson (University of Iowa) conducted the method development studies. This EPA report (as quoted or paraphrased by NYS DOH on page RTC A-167) includes comments such as the following that are directed at gamma spectroscopy as a method of radium-226 measurement for flowback and produced waters (hence a somewhat different context from measuring radium-226 progeny in landfill leachate):

> Although the development process detected bias in the gamma spectrometry measurements at some levels, the magnitude of the bias is lower than that observed for the alpha and there is no need for concern about the ruggedness of the non-destructive measurement technique since there are no variables such chemical separations that will introduce variable levels of bias into the method. Section 11 [of the EPA report] suggests the possibility of future work to improve the sensitivity of the gamma spectrometry measurement. Due to the physics of the measurement technologies, radionuclide determinations performed by gamma spectrometry are generally less sensitive and have higher uncertainty that [sic] those performed by the liquid scintillation counting [LSC]. This complicates the reporting process, the determination of uncertainty, and prevents calculation of a single meaningful value for gross alpha detection capability. Section 11 recommends that measurements of gross alpha by LSC and of 226Ra be reported and interpreted separately and suggests the possibility of future work that would improve the sensitivity of the gamma spectrometry measurement thereby minimize the disparity in the sensitivity of the two techniques.

DEC policy memo on fracking wastes, issued September 18, 2015 by Robert Phaneuf, p. 5, says "For a normal round of sampling, radionuclide analytes should include:

Radium-226 per EPA 903.1
Radium-228 per EPA 904.0
Total Uranium per EPA 908.0
Gamma Spectrum per EPA 901.1"

Draft revision of Part 360-363 regulations was published for public review and comment in 2016, with the public comment period ending on September 13, 2016. Public hearings were held on June 2, 6, 7, and 9, 2016. The draft revision [*not yet seen by RV*] apparently included the gamma spectrum analytical requirement.

DEC Initial Assessment of Public Comment on draft revision of Part 360-363, dated June 2017 (https://www.dec.ny.gov/docs/materials_minerals_pdf/assesspubcom.pdf), p. 310, says "Due to potential for confusion between gamma spectrum and radium testing and in order to reduce costs on the landfills, the gamma spectrum analytical requirement has been removed from the regulation."

Revised Draft Regulations for Part 360-363 were issued by DEC in July 2017.

DEC Supplemental Assessment of Public Comment ("Addressing the Revised Draft Regulations Issued in July 2017"), dated August 2017 (https://www.dec.ny.gov/docs/materials_minerals_pdf/supressummary.pdf), says falsely on p. 139 that "No changes have been made to the final regulations" (this was in response to a comment saying "Table 3A: should include Radon, Polonium 210 and Lead 210. This tracks the full progeny of Radium 226."); and has no response (a blank response) on p. 140 to a comment advocating Method 901.1 M testing; and says vaguely on p. 141 that "the proposed regulations include radionuclide testing in the landfills environmental monitoring plan."

DEC's Revised Consolidated Regulatory Impact Statement for revision of Part 360-363 (https://www.dec.ny.gov/docs/materials_minerals_pdf/part360finalris.pdf), p. 35, says that "Radionuclides, including radium-226, radium-228, and total uranium, as well as gamma spectrum analysis were added to the water quality analysis tables for expanded parameters. Based on further consideration and false positives that are typical from the analysis, the gamma spectrum analysis was removed from the regulation."

The Hakes expansion FSEIS, December 5, 2018 (https://drive.google.com/file/d/1gHhCDlW_odGPmhGTyvCLfG9g8DBmrFXm/view), pp. 16-17, quotes the above-quoted RTC-103 paragraph and appends a footnote #2 that claims that "This paragraph indicates that gamma spectroscopy is an accurate and efficient method to identify radium in environmental samples. It should be noted that the method is accurate and efficient for solid matrix samples. It is not as good a method for liquids, like groundwater and leachate."

Id., p. 22, acknowledges that "Radon values can be inferred from the Pb-214 and Bi-214 gamma spectroscopy results" but claims that "there are inaccuracies with results obtained from leachate and other liquid samples using the method that the comment is based on, which is EPA 901.1 gamma spectrometry. EPA Method 901.1 is a standardized method for soils and other solid matrices and is not appropriate for measuring radiological content in leachate or liquid samples. For these reasons, this methodology was excluded from the High Volume Hydraulic Fracturing (HVHF) Environmental Impact Statement. The revised Part 360 regulations [at 6 NYCRR §

363-4.6(h)] now specify the use of EPA method 903.1 for solid waste leachate testing for radium-226 (and EPA method 904.0 for radium 228)." [*Note by RV: I find no evidence that the 901.1 gamma spectroscopy methodology "was excluded from" the HVHF EIS. See above.*]

Id., p. 23, acknowledges that "The maximum observed Bi-214 leachate result was 6067 pCi/l.... The maximum observed Pb-214 leachate result was 6183 pCi/l...."

The CoPhysics report entitled *Report: A Review of Drill Cuttings Disposal at the Hakes C&D Landfill and Response to Public Comments*, May 16, 2018 (included in Appendix 5 of the Hakes expansion FSEIS, starting at page 10 of Appendix 5, at https://drive.google.com/file/d/1CNSR9nIxkznMA6EJPAQ1hqEDJoAuzErz/view), pp. 11-12, refers to the radioactivity impact study in which CoPhysics was engaged in 2010 and says that radium-226 is used in their risk assessment "as the main radionuclide of interest because it and its progeny cause the majority of the radiation dose to humans in the North American natural environment…. Therefore, radium-226 and its progeny (radon-222, polonium-218, lead-214, bismuth-214, polonium-214, lead-210, bismuth-210 and polonium-210) are specifically assessed."

Id., p. 29, referring to the two leachate samples collected from Hakes Cell #5 on 11/11/14 and Hakes Cell #8 on 6/6/17 which measured approximately 6000 pCi/L of bismuth-214 and lead-214, acknowledges that "Taking the higher cell #5 values and back-calculating (decay-correcting) from the analysis time to the time of collection results in an approximate bismuth-214, lead-214 and radon-222 concentration of 275,000 pCi/L, which sounds like a very high value to a layman. However, these discrete values need to be put in perspective…. Past leachate sampling and analysis methods were never designed to be used for radon assessment. I have discussed these unusual results with the manager of the analysis laboratory. He believes there is so much uncertainty in this type of analysis that, to make a decay correction of several orders of magnitude would result in a multiplication of the uncertainties to unreliable levels. So, the 275,000 pCi/L calculation cannot be relied upon as an accurate estimation of radon and progeny in the original on-site samples…. Even if the 275,000 pCi/L calculation were accurate, however, it would not present health or regulatory problems since this level would be only 9% of the bismuth-214 limit and only 28% of the lead-214 limit for discharges to sewers." [*Note by RV: This last sentence about 275,000 pCi/L not presenting health or regulatory problems focuses on bismuth-214 and lead-214; it misses the point of, and fails to consider, the issue of impacts and health problems associated with 275,000 pCi/L of radon-222.*]

Id., p. 30, says that "The unusual results are certainly reason to conduct further investigation of the issue, including analysis for lead-210 which is the longer-lived decay product of radon-222. As discussed below, however, that investigation has been performed and reveals very low levels of lead-210, further demonstrating that the back-calculated radon result (275,000 pCi/L) is unreliable."

Id., p. 30, continues by claiming that "The analysis method (EPA 901.1) used for leachate analysis in the past (and for the lead-214 and bismuth-214 values that are at issue here) is a soil analysis method and, when used to analyze a water sample, produces very inconsistent and possibly erroneous results. In fact, the NYSDEC has recognized the problems with the EPA 901.1 method for water analysis as reflected by their removal of the requirement for its use in the recent (September, 2017) modification of its landfill regulation (6 NYCRR Parts 360 & 363). The radiochemical and emanation methods for radium analysis (EPA 903.1 and 904.0) are far more sensitive and accurate and will continue to be used. If the NYSDEC wishes radon in water to be analyzed, then a radon-specific test method should be used, such as ASTM D5072. In other words, the 6000 pCi/L values for bismuth-214 and lead-214 obtained utilizing EPA 901.1 are unreliable and do not provide an accurate measure of actual activity levels."

Some of the above sources acknowledge the validity and usefulness of Method 901.1.

Some of the above sources suggest possible inaccuracy and unreliability of Method 901.1 *under certain circumstances*. These circumstances are *mostly not discussed in detail nor linked to a citation that could provide substantive detail*, but are said to be involve issues such as the following:

1. Potential for confusion between gamma spectrum and radium testing

2. Reducing costs on the landfills

3. False positives that are typical from the analysis

4. Not appropriate for measuring radiological content in leachate or liquid samples

5. Is a soil analysis method that, when used to analyze a water sample, produces very inconsistent and possibly erroneous results

6. Manager of analysis laboratory believes "there is so much uncertainty in this type of analysis"

7. Bias in the gamma spectrometry measurements

8. Radionuclide determinations performed by gamma spectrometry are generally less sensitive and have higher uncertainty than those performed by the liquid scintillation counting

RV, 2/16/19

Exhibit B

Excerpt from 51D03_Hakes_Leachate_Radiological_2Q2017_Report_2017-08-29, Table 1, Leachate Radionuclide Analytical Results, Second Quarter 2015 through Second Quarter 2017, Hakes C and D Landfill, Painted Post, New York

Hakes Cell 8B Leachate test results, reported in picocuries per liter

		11/18/2016				5/4/2016				6/6/2017			
		Activity		Uncertainty	(MDC)	Activity		Uncertainty	(MDC)	Activity		Uncertainty	(MDC)
Bismuth-214	(EPA 901.1)	10.19	±	11.736	(12.56)	0	±	12.206	(26.3)	6067.2	±	653.720	(70.33)
Bismuth-214, Dissolved	(EPA 901.1)	3779.3	±	407.970	(48.82)	0	±	9.677	(28.89)	113.23	±	18.794	(12.43)
Lead-214	(EPA 901.1)	11.641	±	9.172	(10.6)	0	±	9.755	(21)	6183.9	±	666.770	(83.91)
Lead-214, Dissolved	(EPA 901.1)	3990.1	±	429.570	(59.09)	0	±	11.947	(22.78)	113.2	±	16.304	(11.11)
Radium-226	(EPA 901.1)	0	±	38.887	(122.7)	0	±	102.250	(256.4)	32.493	±	635.900	(770)
Radium-226	(EPA 903.1)	0.0769	±	0.351	(0.208)	0.211	±	0.774	(1.27)	2.43	±	2.24	(1.32)
Radium-226, Dissolved	(EPA 901.1)	254.23	±	451.840	(540.6)	0	±	110.980	(240.1)	44.007	±	103.980	(128.6)
Radium-226, Dissolved	(EPA 903.1)	0.233	±	0.355	(0.21)	1.13	±	0.622	(0.554)	1.51	±	1.00	(0.455)

MDC = Minimum Detectable Concentration

Note that the columns are correctly dated in the yellow headers but are out of chronological order, as in the cited source. Thus, the columns under the 11/18/2016 header are at the left; those under the 5/4/2016 header are in the middle; and those under the 6/6/2017 header are at the right. Results are shown to varying numbers of decimal places, as in the cited source.

RV 12/28/18

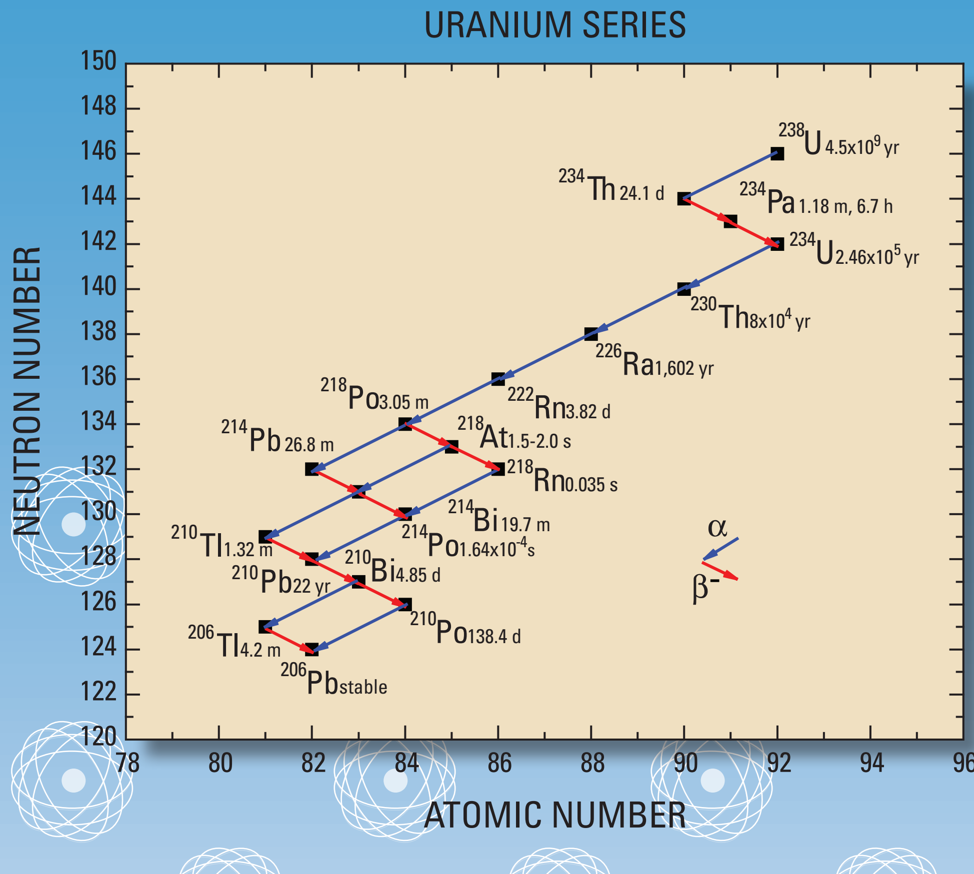
Exhibit C

USGS
science for a changing world

Prepared by the U.S. Geological Survey Office of Water Quality, National Water Quality Laboratory

Interpreting and Reporting Radiological Water-Quality Data

Book 5, Laboratory Analysis
Section B, Methods of the National Water Quality Laboratory
Chapter 6



Techniques and Methods 5–B6

U.S. Department of the Interior
U.S. Geological Survey

Interpreting and Reporting Radiological Water-Quality Data

By David E. McCurdy, John R. Garbarino, and Ann H. Mullin

Book 5, Laboratory Analysis
Section B, Methods of the National Water Quality Laboratory
Chapter 6

Techniques and Methods 5–B6

U.S. Department of the Interior
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Suggested citation:

McCurdy, D.E., Garbarino, J.R., and Mullin, A.H., 2008, Interpreting and reporting radiological water-quality data: U.S. Geological Survey Techniques and Methods, book 5, chap. B6, 33 p.

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Conversion Factors

To Convert	To	Multiply by	To Convert	To	Multiply by
years (y)	seconds (s)	3.16×10^{7}	s	y	3.17×10^{-8}
	minutes (min)	5.26×10^{5}	min		1.90×10^{-6}
	hours (h)	8.77×10^{3}	h		1.14×10^{-4}
disintegrations per second (dps)	becquerels (Bq)	1.0	Bq	dps	1.0
Bq	picocuries (pCi)	27.0	pCi	Bq	3.7×10^{-2}
Bq/kg	pCi/g	2.70×10^{-2}	pCi/g	Bq/kg	37
Bq/m^{3}	pCi/L	2.70×10^{-2}	pCi/L	Bq/m^{3}	37
Bq/m^{3}	Bq/L	10^{3}	Bq/L	Bq/m^{3}	10^{-3}
microcuries per milliliter (μCi/mL)	pCi/L	10^{9}	pCi/L	μCi/mL	10^{-9}
disintegrations per minute (dpm)	μCi	4.50×10^{-7}	mCi	dpm	2.22×10^{6}
	pCi	4.50×10^{-1}	pCi		2.22
Tritium Unit (TU)	Bq/L	0.118	Bq/L	TU	8.47
	dpm/L	7.08	dpm/L		0.141
	pCi/L	3.19	pCi/L		0.313
cubic feet (ft^{3})	cubic meters (m^{3})	2.832×10^{-2}	cubic meters (m^{3})	cubic feet (ft^{3})	35.31
gallons (gal)	liters (L)	3.78	liters	gallons	0.265
gram (g)	ounce, avoirdupois (oz)	0.03527	ounce, avoirdupois (oz)	gram (g)	28.35
kilogram (kg)	pound (lb)	2.205	pound (lb)	kilogram (kg)	0.454

Acronyms and Abbreviations

α	probability of a Type I error
β	probability of a Type II error
λ	decay constant
CSU	combined standard uncertainty (1-sigma)
DQO	data quality objective
USEPA	U.S. Environmental Protection Agency
L	liter
L_C	critical level
MARLAP	Multi-Agency Radiological Laboratory Analytical Protocols Manual
MDC	minimum detectable concentration
MQO	measurement quality objective
NWIS	National Water Information System
NWQL	National Water Quality Laboratory
pCi	picocurie
PWS	performance work statement
QC	quality control
SI	International System of units
ssL_C	sample-specific critical level
ssMDC	sample-specific minimum detectable concentration
USGS	U.S. Geological Survey
WSC	Water Science Center

Interpreting and Reporting Radiological Water-Quality Data

By David E. McCurdy,[1] John R. Garbarino,[2] and Ann H. Mullin[2]

Abstract

This document provides information to U.S. Geological Survey (USGS) Water Science Centers on interpreting and reporting radiological results for samples of environmental matrices, most notably water. The information provided is intended to be broadly useful throughout the United States, but it is recommended that scientists who work at sites containing radioactive hazardous wastes need to consult additional sources for more detailed information. The document is largely based on recognized national standards and guidance documents for radioanalytical sample processing, most notably the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP), and on documents published by the U.S. Environmental Protection Agency and the American National Standards Institute. It does not include discussion of standard USGS practices including field quality-control sample analysis, interpretive report policies, and related issues, all of which shall always be included in any effort by the Water Science Centers. The use of “shall” in this report signifies a policy requirement of the USGS Office of Water Quality.

Introduction

Interpreting and reporting radiological results requires a full understanding of the concepts of detectability and quantification unique to radiochemistry and radiation emissions. Radioactivity describes a group of processes by which matter and energy are released from the nucleus of atoms as an alpha particle ($^4He^+$ nucleus), a beta particle (equivalent to an electron), or a gamma ray (photon or energy wave). The residual nucleus usually is transformed to a different element, and for alpha- and gamma-emitting nuclides, the energy of the emission(s) can be measured to identify the source radionuclide. Natural and anthropogenic radionuclides occur widely in the hydrologic environment (Hem, 1985, p. 146–151; Drever, 1988, p. 379–381; Lieser, 2001). The rate of decay of a given quantity of radioactive atoms ($-dN/dt$) is proportional to the amount of atoms present (N) as shown in equation 1 where λ is the decay constant.

$$-\frac{dN}{dt} = \lambda N \qquad (1)$$

The USGS currently (2008) reports the activity of a radionuclide in curies (Ci). The corresponding International System (SI) unit is the becquerel (Bq), and one curie equals 3.7×10^{10} Bq. The rate of decay for environmental samples is commonly measured in picocuries (pCi = 10^{-12} Ci). One curie is defined as 3.7×10^{10} disintegrations per second, the approximate rate of alpha radiation from one gram of radium. One pCi is thus 0.037 disintegration per second or 2.22 disintegrations per minute (dpm). Radiological analysis in essence involves detecting and counting individual decay-product emissions from a sample for enough time to compute an average rate. The decay constant λ is inversely proportional to the half-life of the radionuclide ($T_{1/2}$), which is the time required for half of the original amount to decay as shown in equation 2.

$$\lambda = \ln 2 / T_{1/2} \qquad (2)$$

A radionuclide concentration reported in picocuries per unit of measure can be converted to specific activity (SA) per moles or grams by using the half-life of the radionuclide and Avogradro’s number using equation 3.

$$SA = \left(\lambda \times N_o\right) / 3.7 \times 10^{-2} = (N_o \times 18.73) / T_{1/2} \qquad (3)$$

where N_o is Avogradro’s number of atoms (6.023×10^{23}) per gram mole of the radionuclide, λ is the decay constant (unit of 1 per second), and $T_{1/2}$ is half-life of the radionuclide in units of seconds. By incorporating Avogradro’s number into equation 3, a simpler equation 4 can be derived:

$$SA = (1.13\times10^{25}) / (AW \times T_{1/2}) \qquad (4)$$

where AW is the atomic weight of the radionuclide. The following table provides similar equations for a specific activity (but in units of picocuries per gram, pCi/g) for radionuclides that have half-lives in years, days, hours,

---

[1] McCurdy Associates, Northboro, Mass.

[2] U.S. Geological Survey National Water Quality Laboratory, Denver, Colo.

$T_{1/2}$	Equation (pCi/g)	Equation (g/pCi)
Years	$SA = (3.58 \times 10^{17})/(AW \times T_{1/2})$	$AW \times T_{1/2} \times 2.80 \times 10^{-18}$
Days	$SA = (1.31 \times 10^{20})/(AW \times T_{1/2})$	$AW \times T_{1/2} \times 7.63 \times 10^{-21}$
Hours	$SA = (3.14 \times 10^{21})/(AW \times T_{1/2})$	$AW \times T_{1/2} \times 3.18 \times 10^{-22}$
Minutes	$SA = (1.88 \times 10^{23})/(AW \times T_{1/2})$	$AW \times T_{1/2} \times 5.32 \times 10^{-24}$
Seconds	$SA = (1.13 \times 10^{25})/(AW \times T_{1/2})$	$AW \times T_{1/2} \times 8.85 \times 10^{-26}$

minutes, and seconds. For example, by using the equation above corresponding to $T_{1/2}$ in years, 1 pCi of ^{238}U with a half-life of 4.46×10^9 years corresponds to a mass of 2.97×10^{-6} grams ($238 \times 4.46 \times 10^9$ years $\times 2.80 \times 10^{-18}$). A more complete introduction to environmental radiochemistry can be found in Eisenbud and Gesell (1997).

Some radiochemical analyses involve separation or isolation steps followed by a quantitation step typically based on some radiation emission measurement. The uncertainty of a radiological result is affected by the activity in the sample, the duration of the measurement, and other various factors that can be controlled for that single measurement. Supplemental information and guidance relative to these concepts have been provided in an Appendix.

Analytical laboratories that provide chemical, radiochemical, and biological analyses to the U.S. Geological Survey (USGS) shall be evaluated relative to the objectives of a project requiring analyses and approved for use for that specific project. Analysis of performance-testing samples will provide the basis for the initial laboratory approval, and an approved laboratory must continue to provide acceptable performance-testing sample results during the life of the project. The National Water Quality Laboratory (NWQL) submits performance-testing samples before the award of all radioanalytical contracts it administers. Review the Policy for the Evaluation and Approval of Analytical Laboratories before submitting samples for analysis (U.S. Geological Survey Office of Water Quality Technical Memorandum No. 2007.01, 2007). Additional information on the laboratory evaluation process can be found at *http://qadata.cr.usgs.gov/lep* (accessed 2008).

USGS National Programs and Water Science Centers (WSCs) conduct research studies and monitoring programs that focus on the detection and quantification of radiological constituents in various environmental matrices, most notably water. In order to support these studies and programs, the USGS NWQL maintains memoranda of understanding and contracts with various USGS and commercial laboratories to process and analyze samples collected by the WSCs. Within the memoranda of understanding and contracts are generic performance work statements (PWS) that apply to all USGS projects. Once the analyses have been completed, the contract laboratories provide NWQL with the results for each radiological constituent and sample matrix combination. The NWQL staff evaluates the reported data and information for technical issues and compliance to specifications stated in the PWS. The acceptable data are transferred to the USGS National Water Information System (NWIS) database through which they are provided to WSCs for further analysis, interpretation, and publication. The ancillary data stored in NWIS include all the parameters needed to publish the data in a report or on NWISWeb including the result, its associated combined standard uncertainty, the sample-specific critical level, and appropriate remark and value-qualifier codes. An additional data package sent by the NWQL to the WSCs provides radiological results with their associated combined standard uncertainty and other parameters and data, which provide ancillary measurement information including quality-control sample results. These ancillary data are used by NWQL to make decisions about detection, uncertainty of results, and reporting of data. Definitions and use of these ancillary data and information are detailed elsewhere in this document.

The laboratories used by NWQL analyze environmental samples according to the PWS, including the measurement quality objective of the *a priori* minimum detectable concentration (*a priori* MDC) for a radiological constituent and sample matrix combination. Unacceptable Type II errors (false nondetection) are avoided through the establishment of the *a priori* MDC and the method selected by the laboratory. Limiting unacceptable Type I errors (false detection) is assured by verifying that each reported result exceeds its respective sample-specific critical level (ssL_C). Specifications for the Data Quality Objective (DQO) required detection levels for certain radiological constituents relative to the USEPA Safe Drinking Water Act (SDWA) also are included in the PWS. In this report, DQO is used in the same manner as the term Measurement Quality Objective used in the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP). The *a priori* MDC is the only DQO specified by NWQL in the PWS. The PWS also will specify the approved radiological method that must be used for the analysis (U.S. Environmental Protection Agency, 1980a). Other important information specified in the PWS include the calculation of the ssL_C, sample-specific Minimum Detectable Concentration (ssMDC), and Combined Standard Uncertainty (CSU) of the measured result. The reported laboratory values for these measurement parameters are used by NWQL personnel to verify contractual compliance with PWS specifications, to determine detection of radiological constituents, to interpret the quality of the result, and to support decisions related to data usability and reporting of data in reports.

Appendix sections A3 and A4 contain the technical basis for the ssL_C and the ssMDC as defined in chapter 20 of the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP, 2004) and a discussion on the effect of sample size and instrument sample processing time on achieving a specified minimum detectable concentration. Terms are defined in a Glossary at the back of the report.

1. Analytical Information Reported by the Contract Laboratory

Various information related to the sampling and radioanalytical processes are reported by the contract laboratories to NWQL and subsequently to USGS WSCs. An Electronic Data Deliverable (EDD) is provided to NWQL by the contract laboratory that contains the unrounded result, CSU, ssL_C, and ssMDC. The information to be reported by the contract laboratory is defined within the PWS and includes sampling and laboratory processing parameters. The analytical information includes:

- client sample identification (ID) code
- sample collection date
- sample matrix
- sample size received
- special instructions

The contract laboratory also reports the following information with the analytical results:

- laboratory identification number cross-referenced to client sample ID
- sample receipt date
- analyte (radiological constituent)
- analysis date
- result value (positive, negative, or zero)
- combined standard uncertainty (CSU; 1-sigma uncertainty)
- sample-specific minimum detectable concentration (ssMDC; *a posteriori* MDC)
- contract required minimum detectable concentration (MDC; *a priori* MDC)
- sample-specific critical level (ssL_C)
- chemical yield (percent) for radiochemical processing
- aliquant size processed
- tracer used

The contract laboratory provides additional information in the form of narrative comments for use in evaluating results for data usability. Also, the method used and its reference designation are provided. All of this information is included on the compact disk data package that is sent to the contact person listed on the Analytical Services Request form (see section 3.6).

2. Definitions of Important Analytical Parameters

2.1 Combined Standard Uncertainty

The Combined Standard Uncertainty (CSU) can be viewed as the statistical standard deviation of an individual radiological result. The concentration of a radiological constituent in a sample is typically calculated using a mathematical equation that includes such parameters as the measured signal response of a radiation detector (events per time unit), the detector background signal response, the detector efficiency for the radiation emission producing the response, sample aliquant size processed, chemical yield of the radiochemical process, and decay and ingrowth factors based on the half-life of the radionuclide or its decay product. Each measurement parameter in the equation has its own uncertainty defined as a standard uncertainty. The CSU of the final result is determined using the common statistical approach that the variance (squared CSU) of a function of several variables can be approximated by applying the function to the variance of each variable component (for example, Benjamin and Cornell, 1970, p. 180–186; MARLAP, 2004, chapter 19). Using this logic, the CSU of a radiological result is the square root of a sum of variances. The Appendix provides an example of a generic equation for calculating concentration (section A1) and the propagation of the standard uncertainties to derive the combined standard uncertainty (section A2).

The statistical normal distribution describes the uncertainty of most contributing variables in a radiological analysis, but the measured number of counts for an analysis follows the Poisson distribution. Poisson variables have a lower bound (zero for radiation counts) and thus have a positive skew. For sufficiently large counts, however, the Poisson distribution can reasonably be approximated as a normal distribution. All the statistical calculations in this report treat the uncertainty of a radiological result as a normal distribution.

When a concentration and its associated CSU are reported, a confidence interval can be calculated that defines the range of concentration (the lower and upper concentration) for the "true concentration" with a certain confidence. Contract laboratories calculate and report the CSU at the 68-percent or 1-sigma (1σ) confidence level (analogous to the standard confidence level used when reporting the standard deviation for other water-quality results). The confidence level that is used when interpreting or publishing radiological results is dependent on the DQOs of the project. Reporting the concentration with its corresponding CSU (as provided in the NWIS database) provides the 68-percent confidence interval. The WSC shall always state the level of confidence of the CSU that is reported; for example, 1.25 ± 0.25 picocuries per liter (pCi/L) at 1σ or 1.25 ± 0.25 pCi/L at the 68-percent confidence level. The corresponding 68-percent confidence interval would be 1.00 to 1.50 pCi/L; or in other words, there is a 68-percent chance that the true value is between 1.00 and 1.50 pCi/L.

For most radionuclide concentrations reported by NWQL, the principal contributor to the CSU is the standard uncertainty of the net count rate. The relation between the CSU and the calculated activity for two radioanalytical measurement techniques is shown in figures A5a, A5b, A6a, and A6b in section A4.5 of the Appendix.

2.2 Sample-Specific Critical Level (ssL_C)

The critical level (L_C) is the smallest measured concentration that is statistically different from the instrument background or analytical blank, and it serves as the detection threshold for deciding whether the radionuclide is present in a sample. The L_C is calculated from measurements obtained using nominal or typical analytical parameter values, whereas the sample-specific critical level (ssL_C) is calculated from measurements obtained using the same analytical parameter values that were used during the analysis of a sample. USGS PWSs require the routine calculation of the ssL_C for each sample, using parameter values that were actually measured during the generation of the sample result. The null hypothesis for establishing the critical level is that "the sample activity level is the same as the measured instrument background or blank sample value." The maximum acceptable probability α of false detection (significance level), together with the standard deviation of the net blank sample distribution having a mean value of zero, forms the basis for the critical level upon which detection decisions may be made (Currie, 1968). For analysis of USGS radiological samples, a false detection rate of 5 percent (α_0=0.05) is used. This hypothesis test strives to limit false detection (known as Type I error). Figure A1, section A3.1 in the Appendix graphically illustrates the critical level concept. Note that the critical level concept as applied to radionuclide detection is based on a "one-sided" hypothesis test that considers only the upper-tail probabilities of the null distribution and is different from a two-sided test that would consider both the upper- and lower-tail probabilities.

A detection decision is based on comparison of the sample result with the ssL_C. Because the ssL_C is a hypothesis-testing concept based on a preestablished probability of false detection and the standard deviation of the net background distribution, the combination of the result and the ssL_C and not the measurement CSU (and resultant symmetrical confidence interval) is used for detection decisions. Whenever the concentration of a radiological constituent is greater than the ssL_C, it shall be considered detected; that is, the reported concentration is positive and greater than the measurement (instrument) background or the radiological constituent's concentration in a blank sample. When the concentration is greater than the ssL_C, the decision "detected" shall be reported, and a symmetrical confidence interval shall be given "after the detection decision is made" (Currie, 1968).

Scientists can evaluate the reported sample data set to determine if the reported ssL_C has been calculated properly. Section A3.3 of the Appendix provides a practical approach for verifying the ssL_C for most radioanalytical methods using the reported CSU. This guidance is not definitive but may be used to determine whether or not the relation of reported ssL_C and its uncertainty is reasonable.

2.3 Minimum Detectable Concentration

The critical level concept discussed in section 2.2 addresses Type I error (false detection), but it does not consider Type II error (false nondetection). If the true concentration of a radionuclide were exactly equal to the critical level, the inherent uncertainty of the measurement would produce larger (detected) results for some samples and smaller (not detected) results for others. The Minimum Detectable Concentration (MDC) concept addresses Type II error. The MDC can be calculated *a priori*, using nominal or typical analytical parameter values, or *a posteriori* for a specific sample, using the ssL_c and parameter values for an individual sample.

2.3.1 *a priori* Minimum Detectable Concentration (*a priori* MDC)

Consideration of both Type I and II errors is the basis of the *a priori* MDC concept. The critical level incorporated in the expression for the *a priori* MDC (see equation A9 of section A4.1 of the Appendix) is typically calculated using nominal or typical parameter values such as detector efficiency, chemical yield, and sample aliquant processed. The *a priori* MDC for a radioanalytical method is a laboratory analytical-method performance characteristic.

The *a priori* MDC is an *a priori* (before the sample measurement) concept that is only used to facilitate comparisons of the relative detection capabilities of measurement systems or radiological methods. It is defined as the lowest true concentration that gives a specified probability that the measured concentration will exceed its critical level concentration (Currie, 1968; MARLAP, 2004, chapter 20). The *a priori* MDC satisfies the hypothesis testing the probability (β) at the 0.05 or 95-percent confidence level that the true result is greater than the L_C, given that the analytical result equals the *a priori* MDC and assuming a normal distribution. As such, its definition may be restated as the lowest concentration for which there is a 95-percent probability of producing a result greater than the critical level and a 5-percent probability of falsely concluding that a blank measurement represents a positive measurement (above the critical level). Figure A2, section A4.1 in the Appendix, graphically illustrates the *a priori* MDC concept and shows a distribution of measurement results from a set of samples having a radiological constituent concentration at the MDC.

Based on USGS program needs and state-of-the art radioanalytical methods, standardized *a priori* MDC DQOs for various radiological constituent/matrix combinations have been established by NWQL. The *a priori* MDC for a radiological analyte is in the NWQL Catalog (see the reporting level entry at *http://nwql.cr.usgs.gov/usgs/catalog/index.cfm*). The established *a priori* MDC DQO specification is based on need or the expected detection capability of a method, or both. For example, an *a priori* MDC contractual specification of 1 pCi/L for tritium (^{3}H) in water has been chosen for certain research studies. These standardized *a priori* MDC concentrations and matrix combinations become laboratory contract specifications as defined in the PWS. By establishing the *a priori* MDC, an acceptable Type II error is defined. When the laboratory selects a method to meet the *a priori* MDC, unacceptable Type II errors are limited.

The contract laboratory uses the *a priori* MDC requirements to select appropriate methods and method parameters to meet the contract specifications. Nominal or typical parameter values (detector efficiency, chemical yield, sample aliquant processed, and so forth) of the radioanalytical method are generally chosen by the contract laboratory when calculating the *a priori* MDC for a given method and radiological constituent.

2.3.2 Sample-Specific Minimum Detectable Concentration (ssMDC)

NWQL contracts also require calculation and reporting of an *a posteriori* (after the measurement) or sample-specific Minimum Detectable Concentration (ssMDC) in association with each radiological result reported. The contract laboratory uses the ssL_C as the basis for establishing the ssMDC as evaluated in context with the *a priori* MDC on each individual measurement to establish the Type II error. The ssMDC is used by the NWQL to verify that the *a priori* MDC DQO has been met. Because the ssMDC is calculated with actual parameter values used during the analysis of the sample in question, the ssMDC, in most cases, may tend to be below the *a priori* MDC, which uses more conservative nominal method parameter values. In most cases, the actual method parameter values used for the ssMDC calculation do not substantially change from the nominal values used in the *a priori* MDC calculation unless certain circumstances have occurred; for example, smaller sample size processed or lower chemical yields may lead to longer counting times. Occasionally, if the laboratory does not adjust certain method parameters, the *a priori* MDC DQO may not be met because of possible unexpected chemical and instrumental interferences and small sample sizes. In such cases, the ssMDC will be greater than the *a priori* MDC DQO.

The MDC (*a priori* MDC or *a posteriori* ssMDC) shall never be applied to make decisions about whether a radiological constituent has been detected in a sample; rather, the ssL_C shall be used for defining when a concentration is different from zero with a specified probability (5 percent for most cases) of false detection. Section A4.2 of the Appendix provides additional information and the typical equations used by a laboratory to calculate the ssMDC. Practical approaches for determining whether a reported ssMDC has been calculated properly (section A4.3), and the effects of sample volume and counting time on the magnitude of the ssMDC (section A4.4) are provided in the Appendix.

2.4 Comparison of Radiological, Inorganic, and Organic Detection Levels

The concepts on radiological detectability as presented in section 2.2 on the ssL_C and section 2.3.2 on the ssMDC (also sections A3 and A4 in the Appendix) are similar to those presented for organic and inorganic analytes in the U.S. Geological Survey Open-File Report 99–193 (Childress and others, 1999) for the long-term method detection level (LT–MDL) and the laboratory reporting level (LRL), respectively. The LT-MDL is based on a modification of the U.S. Environmental Protection Agency's method detection limit (MDL) procedure and relies on several of its key assumptions (Childress and others, 1999). The primary difference between the MDL and the LT–MDL is that the LT–MDL is designed to measure more sources of variability and therefore is expected to be higher than the MDL. The MDL uses the standard deviation of spiked samples based on a minimum number of seven spiked samples (a snapshot or single set of measurements made at between 1 and 5 times the estimated MDL concentration), whereas the LT–MDL uses the standard deviation based on a much larger set of spiked samples (at least 24 per year) collected over an extended period of time, typically 6 to 12 months.

The basic concepts presented for radiological, organic, and inorganic analytes assume a normal distribution for the blank and spike sample measurements and use the standard deviations of these distributions and defined error rates for false detection and false nondetection. The equations for the critical level (L_C and ssL_C) and the long-term method detection level (LT–MDL) are basically identical except that the critical level equations use a 5-percent α error rate for false detection compared with 1-percent α error rate for false detection used to calculate the long-term method detection level. For both applications, the critical level and LT–MDL are incorporated into the determination of the MDC and the LRL, respectively. Similar to the L_C and the LT–MDL, the error rates for the MDC and LRL differ; the MDC equation uses a 5-percent β error rate for false nondetection, whereas a 1-percent β error rate for false nondetection is used for the LRL.

Another basic difference in calculating the L_C and the LT–MDL is that the L_C uses a standard deviation for a distribution of blank sample results whereas, for organic analytes and some inorganic analytes, the LT–MDL uses a standard deviation of a distribution of results from samples spiked near the estimated detection level. In both cases, the calculations assume the standard deviation in blanks and the standard deviation near the critical or detection level are equal. A comparison of the basic differences between detection-level terms and error rates and basic assumptions, detection decisions, and results reporting for radiological, organic, and inorganic constituents is presented in tables 1 and 2, respectively.

3. National Water Quality Laboratory Evaluations of Contract Laboratory Results

Initial evaluation of the quality of contract laboratory radiological data is conducted by NWQL before the data are sent to the NWIS database. This initial evaluation is done largely to determine contractual compliance and overall quality of the data. The data are then transferred to the NWIS database through which they can be accessed by USGS WSCs.

3.1 Initial Evaluation Criteria

As a standard practice, NWQL evaluates a contract laboratory's reported radiological data for each sample for contractual compliance for technical items such as, but not limited to:

- reporting of sample parameters and information according to specifications (see section 1);
- contractual MDC specification (by comparing the ssMDC value to the contractual *a priori* MDC);
- radiological hold time (by comparing sample collection and analysis dates);
- processing turnaround time (by comparing sample receipt and analysis report dates);
- insufficient sample size for analysis;
- yield for certain radiological constituents; and
- batch Quality Control (QC) sample results related to method bias (laboratory control and matrix spike samples), excess uncertainty or imprecision (split or duplicate sample analyses), and false positive and negative (blank samples).

The typical processes that NWQL uses to evaluate contract laboratory results are listed in table 3. Several examples of the most common and relatively unambiguous situations and those that occur less frequently and require deeper scrutiny are discussed.

3.2 Rounding Results

The NWQL Laboratory Information Management System (LIMS) rounds contract laboratory results received through the EDD by using the American National Standards Institute procedure N42.23 (American National Standards Institute, 2003). The CSU shall be rounded to two significant figures, and both the radiological concentration and CSU shall be reported to the same number of decimal places. Proper rounding conventions notwithstanding, one must always remember that the CSU reported in association with the sample concentration, and not the base-10 rounding of results, establishes the number of significant digits in a radiological result. Examples are provided in table 3 showing how contract laboratory radionuclide concentrations and their CSUs are rounded before they are sent to the NWIS database.

Table 1. Equations and error rates used for calculating the critical level and long-term method detection level.

[L_C, critical level; LT–MDL, long-term method detection level; MDC, minimum detectable concentration; LRL, laboratory reporting level; NA, not applicable; s_{blanks}, standard deviation for blanks; s_{LT-MDL}, standard deviation for spikes at 1 to 5 times the estimated detection level; k_β, statistical factor; $t_{(n-1,1-\alpha=0.99)}$, statistical factor; *n*, number of samples; α, probability of false detection; β, probability of false nondetection]

	L_C[a]	**LT–MDL**[b]	***a priori* MDC**[c]	**LRL**
Basic practical equation	$sblanks \times k_\alpha$	$s_{LT-MDL} \times t_{(n-1,1-\alpha=0.99)}$	$L_C + k_\beta \times s_{blanks}$	$2 \times s_{LT-MDL} \times t_{(n-1,1-\alpha=0.99)}$
Specific equation	$s_{blanks} \times 1.645$	$s_{LT-MDL} \times 2.50$	$2.71 + 3.29 \times s_{blanks}$	2 × LT–MDL
False detection error rate	0.05	0.01	0.05	0.01
False nondetection error rate	NA	NA	0.05	0.01

[a] When $\alpha = 0.05$, $k_\alpha = z(_{1-\alpha}) = 1.645$; where $z_{(1-\alpha)}$ denotes the $(1-\alpha)$ quantile of the standard normal distribution.

[b] The basic equation is described by Childress and others (1999). The Student's *t* statistic for 23 degrees of freedom is equal to 2.50 for $\alpha = 0.01$.

[c] For practical purposes, the basic assumption is that s_{blanks} is approximately equal to $s_{spikes-MDC}$ (the standard deviation of the distribution of a sample spiked at the MDC). The equation simplifies by assuming $\beta = 0.05$ and $k_\alpha\beta = z_{(1-\beta)} = 1.645$; where $z_{(1-\beta)}$ denotes the $(1-\beta)$ quantile of the standard normal distribution.

Table 2. Basic assumptions, detection decisions, and results reporting for radiological, organic, and inorganic methods.

[L_C, critical level; ssL_C, sample-specific critical level; *a priori* MDC, method-specific *a priori* minimum detectable concentration; ssMDC, sample-specific MDC; LT–MDL, long-term method detection level; LRL, laboratory reporting level; PWS, performance work statement for contract laboratory; NA, not applicable; lc std, lowest calibration standard; ≥, greater than or equal to; <, less than]

Radiological methods	**L_C**	**ssL_C**
Basic assumptions	Typical blank or instrument background distribution and sample parameters; can be instrument specific or average value for all instruments using method	Instrument-specific background distribution and sample-specific parameters
Detection decisions	Not used	Result ≥ ssL_C
Reporting results	Not reported	Is always reported with the result (negative, zero, or positive) and the combined standard uncertainty (CSU)
Organic and inorganic methods	**LT–MDL**	**No corresponding term**
Basic assumptions	Multiple instrument and multiple analysts; analysis of samples spiked at 1 to 5 times the estimated detection level; spike distribution of ≥ 24 samples over 6 to 12 months; and constant sample parameters used	NA
Detection decisions	Result ≥ LT–MDL	NA
Reporting results	Result concentrations ≥ LT–MDL are reported with a qualifier when the concentration is less than the LRL or lc std, whichever is greater (ideally, the lc std is equal to the LRL); when the result < LT–MDL, then < LRL is reported; for information-rich organic methods, qualitative results are reported with a qualifier when a result is < LT–MDL[b]	NA
Radiological methods	***a priori* MDC**	**ssMDC**
Basic assumptions	Typical blank or instrument background distribution and sample parameters; can be instrument specific or average value for all instruments using method	Instrument-specific background distribution and sample-specific parameters
Detection decisions	Not used	Not used
Reporting results	Available from NWQL Catalog[a]	Not generally reported; it is used only to evaluate contractual requirements of the PWS
Organic and inorganic methods	**LRL**	**No corresponding term**
Basic assumptions	Multiple instrument and multiple analysts; analysis of samples spiked at 1 to 5 times the estimated detection level; spike distribution of ≥ 24 samples over 6 to 12 months; and constant sample parameters used	NA
Detection decisions	Not used	NA
Reporting results	All results greater than the LRL or lc std, whichever is greater, are reported without a qualifier[b]	NA

[a] The *a priori* MDC is listed in the NWQL Catalog under the reporting level entry; see *http://nwql.cr.usgs.gov/usgs/catalog/index.cfm*.

[b] Refer to figure 10 in U.S Geological Survey Open-File Report 99–193 (Childress and others, 1999) for details.

Table 3. Examples showing the processes that are used by the National Water Quality Laboratory to review radiological results.

[All numbers are in units of picocuries per liter; CSU, 1-sigma Combined Standard Uncertainty; ssL_C, sample-specific critical level sent to the National Water Information System (NWIS) database; *a priori* MDC, contractual *a priori* Minimum Detectable Concentration; ssMDC, sample-specific Minimum Detectable Concentration; Code, remark or value-qualifier code(s) sent to the NWIS database; R, nondetect, result less than sample-specific critical level;), ssMDC exceeded the *a priori* MDC; =, negative result may indicate potential negative bias; NWQL, National Water Quality Laboratory; ±, plus or minus; <, less than; >, greater than; WSC, Water Science Center]

Unrounded concentration	Unrounded CSU	ssL_C	ssMDC	*a priori* MDC	Code	Decision processes	Rounded concentration and (CSU) sent to the NWIS database
2.346	0.542	0.93	2.3	3.0		Result > ssL_C; reasonable relation between CSU, ssL_C, and ssMDC; ssMDC < *a priori* MDC.	2.35 (0.54)
0.534	0.542	0.93	2.3	3.0	R	Result < ssL_C; reasonable relation between CSU, ssL_C, and ssMDC; ssMDC < *a priori* MDC. Remark code sent to NWIS.	0.53 (0.54)
6.636	1.542	3.2	6.5	3.0	)	Result > ssL_C, reasonable relation between CSU, ssL_C, and ssMDC. However, the ssMDC > contractual *a priori* MDC. NWQL will try to determine the cause by looking at other data and information and consulting with the contract laboratory. Reanalysis may be requested. Value-qualifier code sent to NWIS.	6.6 (1.5)
0.534	0.742	1.6	3.2	3.0	R)	Result < ssL_C; reasonable relation between CSU, ssL_C, and ssMDC. However, the ssMDC > contractual *a priori* MDC. NWQL will try to determine the cause by looking at other data and information and consulting with the contract laboratory. Reanalysis may be requested. Remark and value-qualifier codes sent to NWIS.	0.53 (0.74)
–1.525	0.972	0.93	2.7	3.0	R	Result < ssL_C; reasonable relation between CSU, ssL_C, and ssMDC; ssMDC < *a priori* MDC; and no negative bias (negative concentration < 1.65 × CSU). Remark code sent to NWIS.	–1.52 (0.97)
–0.504	0.735	0.22	0.70	0.90		The ssMDC < *a priori* MDC and there is no negative bias (negative concentration < 1.65 CSU). However, the result is unacceptable because the ssL_C and ssMDC are too small relative to the CSU. NWQL may request a recalculation or reanalysis.	Results are not sent to the NWIS database unless the problem is resolved.

Table 3. Examples showing the processes that are used by National Water Quality Laboratory to review radiological results.—Continued

[All numbers are in units of picocuries per liter; CSU, 1-sigma Combined Standard Uncertainty; ssL_C, sample-specific critical level sent to the National Water Information System (NWIS) database; *a priori* MDC, contractual *a priori* Minimum Detectable Concentration; ssMDC, sample-specific Minimum Detectable Concentration; Code, remark or value-qualifier code(s) sent to the NWIS database; R, nondetect, result less than sample-specific critical level;), ssMDC exceeded the *a priori* MDC; =, negative result may indicate potential negative bias; NWQL, National Water Quality Laboratory; D, detection; ±, plus or minus; <, less than; >, greater than; WSC, Water Science Center]

Unrounded concentration	Unrounded CSU	ssL_C	ssMDC	*a priori* MDC	Code	Decision processes	Rounded concentration and (CSU) sent to the NWIS database
–2.523	0.731	0.93	2.7	3.0	R =	Result < ssL_C; reasonable relation between CSU, ssL_C, and ssMDC; ssMDC < *a priori* MDC. However, the result is unacceptable or requires careful qualification because the negative concentration > 1.65 × CSU. Remark and value-qualifier codes sent to NWIS. WSC scientists should search for patterns among any samples in this category.	–2.52 (0.73)
0.636	2.542	3.2	2.7	3.0		Result < ssL_{Cc} and ssMDC < *a priori* MDC. However, the result is unacceptable because of the unusually high CSU and because the ssL_C and ssMDC are too small relative to the CSU. NWQL may not accept this result for technical reasons and may request a reanalysis of this sample. NWQL will try to determine the cause of error by looking at other data and information.	Results are not sent to the NWIS database unless the problem is resolved
1.005	0.544	0.53	2.7	3.0		Result > ssL_C and ssMDC < a priori MDC. However, the result is unacceptable because the ssL_C is too small relative to the CSU. NWQL may not accept this result for technical reasons and may request a reanalysis of this sample. NWQL will try to determine the cause of error by looking at other data and information.	Results are not sent to the NWIS database unless the problem is resolved
10.783	4.204	8.9	19.7	1.0	)	Result > ssL_C and there is a reasonable relation between CSU, ssL_c, and ssMDC. However, the ssMDC > a priori MDC. Upon further review, NWQL determined that a small sample volume was used. Therefore, because the result is positive and reasonable for sample size and reanalysis is not possible because of the lack of sample, the result is recorded in the NWIS database. Value-qualifier code sent to NWIS.	10.8 (4.2)

3.3 Review of Negative Results

Analysis of a radiological sample produces a gross signal response that is related to the quantity of the radionuclide present. However, random measurement uncertainties will cause this signal to vary somewhat if the measurement is repeated. A nonzero signal may be produced even when no radionuclide is present. For this reason, the contract laboratory analyzes an instrument background or a blank sample (discrete from the blank used for quality-control purposes) and subtracts its signal from the gross signal to obtain the net signal. If the measurement process is under control (free from systematic bias) and a series of blanks were analyzed and the background signal subtracted from each measurement, the results should be evenly distributed above and below a zero concentration, with negative values in approximately one-half of the blanks (see fig. A1 in the Appendix). Therefore, negative results are possible due to the randomness of the measurement process. Nevertheless, this does not imply that there is negative radioactivity. Each calculated result will have an associated CSU, and thus a confidence interval can be calculated and interpreted. Sometimes the lower end of the confidence interval may be negative, meaning that the true concentration may not be different from zero.

In order to determine if a negative result is valid, it is compared to the lower 95 percent one-sided confidence interval. A negative result is considered valid if the magnitude of the negative result is ≤ 1.65 times the reported CSU (1.65 is the 95th percentile of the standard normal distribution). When the magnitude of the negative result is greater than 1.65 times the reported CSU, the result may be considered invalid because there is less than 5-percent probability that the result is from a blank or instrument background distribution (for example, with a zero mean value), indicating that the measurement process may not be in control (see examples in table 3). Typical reasons for invalid negative results include a nonrepresentative background or blank signal or an inaccurate determination of radionuclide interferences. An invalid negative result can be reported with the corresponding value-qualifier code (see section 3.4). A valid negative result can be reported as a nondetect concentration.

3.4 Assigning Remark and Value-Qualifier Codes

Remark and value-qualifier codes are assigned by NWQL and are included with results whenever additional information is needed for interpretation. The following remark and value-qualifier codes with their explanations can be used with radiological results. Only one remark code can be included with a radiological result, whereas up to three value-qualifier codes can be used. Remark and value-qualifier codes are assigned to the results during evaluation by the NWQL (see examples in table 3). Contractual acceptance criteria associated with the remark and value-qualifier codes can be found at *http://wwwnwql.cr.usgs.gov/USGS/acu_contracts.html*.

Remark code	**Explanation**
R	Nondetect, result below sample-specific critical level (ssL_C)

Value-qualifier	**Explanation**
(	Blank greater than the sample-specific critical level (ssL_C)
)	Sample-specific Minimum Detectable Concentration (ssMDC) is above the contractual *a priori* MDC
/	Matrix Spike (MS) recovery is outside of contractual acceptable range (see Glossary for definition of recovery)
@	Exceeded sample holding time
\	Laboratory Control Sample (LCS) recovery is outside of contractual acceptable range
~	Duplicates are not within the contractual acceptance limits
=	Negative result may indicate potential negative bias
^	Yield is outside of contractual acceptable range (see glossary for definition of yield)

3.5 Information Sent to the National Water Information System (NWIS) Database

The following information is sent to the NWIS database. The concentration, CSU, and ssL_C are reported in either pCi/L or pCi/g.

- Site-agency code
- Station-identification number
- Sample-collection date
- Sample-collection time
- Sample-collection end date
- Sample-collection end time
- Sample-medium code
- Parameter Code
- Rounded concentration
- Remark and value-qualifier code(s)
- Rounded Combined Standard Uncertainty (1-sigma)

- Sample-specific critical level

The NWIS database information also is transferred to NWISWeb to provide electronic access to radiological and other water-quality information through website *http://waterdata.usgs.gov/nwis*.

3.6 Information in Detailed Data Packages

Additional information associated with the sample analysis is provided in a compact disk data package sent to the WSC to assist with the review of the laboratory and field QA sample results. Some of the information in the data package is not recorded in the NWIS database. The data package includes a narrative, data, sample information, and laboratory information sections as shown in the following list. The data section provides the result, CSU (1-sigma), ssMDC, *a priori* MDC, ssL_C, percent yield, aliquant size, and results for laboratory quality-control samples.

- Data report narrative (additional details specific to the analyses; for example, relative percent difference for duplicate samples)
- Data section
  - Sample summaries (client sample ID, location, matrix, laboratory sample ID, chain of custody, sample date and time, amount of sample received, and the WSC contact
  - Sample batch QC summary (number of blanks, laboratory control samples, and duplicates)
  - Work summary (date collected, date received, date analyzed, date reviewed)
  - Method blank results
  - Laboratory control sample results
  - Matrix spike results
  - Duplicate results
  - Results by sample and method
- Analytical Services Request (ASR) form for each sample
- Radiological login sheet

4. Water Science Center Reviews of National Water Information System (NWIS) Database Results

Specific information from NWIS is needed to complete a thorough review of radiological results. For the radiological data corresponding to samples analyzed after March 1, 2003, the following list of alpha parameters should be retrieved from NWIS into a "by-result" table for review.

PCODE – Parameter code

PSNAM – Parameter abbreviated name

REMRK – Remark code; this will include any remark code stored with the result

VALUE – Result value; if retrieved with the no-rounding option, this will be the laboratory result

UNITS – Result unit of measure

QUAL1 – First value-qualifier code stored with the result

QUAL2 – Second value-qualifier code stored with the result

QUAL3 – Third value-qualifier code stored with the result

LSDEV – Laboratory standard deviation; this field is where the Combined Standard Uncertainty (1σ CSU) for the result is stored

RLTYP – Report level type; for radiological samples analyzed after March 1, 2003, this field always equals "ssL_C"

RPLEV – Report level; this field is where the sample-specific critical level (ssL_C) for the result is stored

RCMLB – Result-level laboratory comment; this field will provide any additional information stored with the result

Results should be retrieved using the unrounded option because radiological results stored in NWIS are already rounded. The VALUE, LSDEV, and RPLEV are reported in the same UNITS.

For radiological samples analyzed before March 1, 2003, sample-specific critical levels (ssL_C) were not reported, and 2-sigma precision estimates or 2SPE (equivalent to 2 sigma Combined Standard Uncertainty) were reported under separate parameter codes. For tritium and radon samples analyzed prior to August 1, 2008, the ssL_C was not reported and the 2SPE was reported under a separate parameter code. For tritium and radon samples submitted after August 1, 2008, the ssL_C and 1σ CSU will be reported. More details about the retrieving results from the NWIS database can be found at *http://wwwnwql.cr.usgs.gov/USGS/rapi-note/05-019.html* and in section 3.4.6 of Web page *http://wwwnwis.er.usgs.gov/currentdocs/qw/QW.user.book.html*.

Much of the review by the WSC is focused on data interpretation. The WSC shall review the radiological results with respect to historical data from the collection site. Results obtained for QC samples, such as matrix-spike samples, can be reviewed to identify quality problems in laboratory analytical performance, sample matrix effects, and field sample collection. Matrix spike results can be used to establish bias, whereas laboratory-duplicate results can be used to establish subsampling and method variability. Duplicate field samples can be used to establish sample-collection variability. Remark and value-qualifier codes should be reviewed in order to evaluate their effect on interpretation and for providing descriptive information presented in publications.

5. Publishing Results

5.1 Technical Reports

The USGS conventions for publishing radiological results as outlined in this report follow the practices of the U.S. Environmental Protection Agency (U.S. Environmental Protection Agency, 1980b), American National Standards Institute N42.23 (American National Standards Institute, 2003), and Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP, 2004, chapter 16). These national standards and guidance documents state that reported radiological data should always consist of two numbers, the measured concentration (or activity) and the associated measurement uncertainty (CSU at a stated level of confidence). Therefore, after radiological results have been reviewed by the WSC, the minimal acceptable information to be published shall include the:

- Result (positive, negative, or zero)
- CSU (1σ)
- Reference radionuclide for gross alpha and beta analyses

The concentration (or activity) and CSU should not be interpreted as a single point, but as a confidence interval about the measured concentration in which one has a high statistical probability of finding the true concentration of the sample (approximately 68 percent at 1-sigma). The practice of not including the CSU is ill advised as it withholds critical information associated with the result that could lead to misinterpretation or even critical misapplication of the data. Although the measurement uncertainty is not used in determining compliance with the Safe Drinking Water Act (SDWA), it will be needed for data evaluation of other studies.

The concentration, including zero and negative results, and the CSU shall be recorded in the same units (for example, picocuries per liter). In addition, the CSU shall never be stated as a relative percentage or fraction of the result because as a result approaches zero the relative uncertainty becomes exceedingly large and does not lend itself to meaningful interpretation. When nondetect results are published, it is strongly recommended that the report include a “detection indicator” for clarification. For example, the result and CSU are reported and flagged with a nondetect identifier whose definition is provided. Results shall not be reported as <ssMDC or <ssL_C.

Radiological results should be reported according to conventions that establish and preserve their technical defensibility. Results should be identified in a manner that permits them to be connected unambiguously to a sampling event and to the radioanalytical measurements used to generate the results. Individual results are best reported in association with a unique identifier that is, or can be, associated with a project, location, date, time, and record of collection. If groups of data are being averaged, it may not be feasible to reference each unique identifier, but the descriptor associated with the averaged data point should always accurately and unambiguously characterize the group of data in question. Results should always be presented in association with the name of the analyte, the measured concentration (inclusive of all positive, zero, or negative values) and associated CSU, the level of significance for the confidence interval reported, the ssL_C, and an activity reference date for shorter lived radionuclides or mixtures of radionuclides. In the case of nonradionuclide-specific measurements, one should include the measurement parameter, such as gross alpha or total uranium, as well as any applicable assumptions underlying the gross measurement. For example, for gross alpha, the WSC should specify the reference nuclide used for calibration of the instrument that is “gross alpha (referenced to ^{230}Th).”

Oftentimes, the activity reference date and time are overlooked by investigators who are unfamiliar with radioanalytical measurements. The measured activity reported for a sample is only valid for a specified point in time because the radioactivity of a sample changes over time, depending on the half-life of the supporting radionuclide. Failure to specify the activity reference date and time, especially with short-lived radionuclides, can render published results useless. If the holding time for a sample analyzed for a short-lived radionuclide is exceeded, the published result shall include a statement that the holding time was exceeded with the specific time interval beyond the holding-time limit.

Table 4 shows an example of the types of information that should be included when publishing radiological results, such as those discussed in table 3. NWIS remark and value-qualifier codes can be translated to convey additional interpretive information for the data presented.

5.2 Nontechnical Reports

Presenting radiological data in a technically defensible, yet understandable manner is a challenge to any investigator who prepares reports for issuance to the general public. Clearly, WSCs must always ensure that data are presented in a manner that addresses the subject clearly without compromising technical accuracy or validity. While attempting to prevent or minimize confusion among the lay reader, the WSC may decrease the level of detail of the data provided or simplify the complexity of concepts presented to a level appropriate for the purpose and the perceived background of the audience. The WSC shall ensure that reports are presented clearly and that the depth and limitations of the presentation are clear to any reader, ranging from the layperson to the expert. Although authors cannot foresee every use or interpretation of their published data, it is important that they remain mindful that

Table 4. An example of typical information that should be provided when publishing radiological results.

[Result, radiological concentration plus or minus the 1-sigma combined standard uncertainty; ssL$_C$, sample-specific critical level; GA, gross alpha; 72h, sample analyzed for GA concentration at approximately 72 hours after sample collection as referenced to a detector calibrated using ^{230}Th; 30d, sample used for the 72-hour GA analysis is counted a second time approximately 30 days after the initial count as referenced to a detector calibrated using ^{230}Th; pCi/L; picocurie per liter; D, analyte detected; ND, analyte not detected, concentration is less than the sample-specific critical level; a, 72-hour sample holding time was exceeded; b, negative result may indicate potential negative bias; c, sample-specific Method Detectable Concentration (ssMDC) exceeded the *a priori* MDC; d, ssMDC exceeded the *a priori* MDC but was reasonable for the sample volume processed; ±, plus or minus; EPA, U.S. Environmental Protection Agency]

Sampling site	Sample collection date and time	Analysis date and time	Radiological constituent	Result	ssL$_C$	Remark	Units	Sample type	Analytical method
Well 5	9/18/2006 14:10	9/25/2006 8:35	GA (72h)	17 ± 12	23	ND, a	pCi/L	Unfiltered	EPA 900.0
Well 6	9/21/2006 8:37	9/23/2006 9:40	GA (72h)	5.1 ± 1.9	2.5	D	pCi/L	Filtered	EPA 900.0
Well 5	9/18/2006 14:10	10/20/2006 15:11	GA (30d)	23 ± 14	28	ND	pCi/L	Unfiltered	EPA 900.0
Well 6	9/21/2006 8:37	10/27/2006 8:25	GA (30d)	5.8 ± 2.7	4.2	D	pCi/L	Filtered	EPA 900.0
Well 5	10/7/2006 9:11	10/9/2006 8:11	^{228}Ra	2.35 ± 0.54	0.93	D	pCi/L	Filtered	EPA 904.0
Well 5	10/14/2006 9:15	11/16/2006 13:25	^{228}Ra	0.53 ± 0.54	0.93	ND	pCi/L	Filtered	EPA 904.0
Well 5	11/20/2006 9:04	11/25/2006 10:04	^{228}Ra	–2.52 ± 0.73	0.93	ND, b	pCi/L	Filtered	EPA 904.0
Well 6	10/22/2006 9:45	10/25/2006 14:45	^{228}Ra	6.6 ± 1.5	3.2	D, c	pCi/L	Filtered	EPA 904.0
Well 6	10/31/2006 9:30	11/2/2006 10:30	^{228}Ra	0.53 ± 0.74	1.6	ND, c	pCi/L	Filtered	EPA 904.0
Well 6	11/20/2006 9:10	11/25/2007 11:04	^{228}Ra	7.8 ± 2.2	3.9	D, d	pCi/L	Filtered	EPA 904.0

data may lose validity when it is taken out of the context or presented in an otherwise incomplete manner. WSCs shall always attempt to minimize the probability that results could be misinterpreted or misconstrued.

USGS WSCs conduct research studies and monitoring programs that focus on the detection and quantification of radiological constituents in various environmental matrices at substantially lower concentrations than those associated with regulatory action levels (AL), Safe Drinking Water Act Maximum Contaminant Levels (MCL), or other health benchmark levels. Surface water and ground water may reasonably be expected to have at least small amounts of some radionuclides. However, their presence does not necessarily indicate the water poses a health risk. Therefore, providing a comparison of WSC results to AL or MCL should be considered to ensure the lay public interprets the radiological concentrations from a relevant perspective. In addition, it also should be emphasized that nondetection does not imply that the radiological constituent is not present; rather, its concentration is below the level that can be measured. Table 5 shows an example of the types of information that should be included when publishing radiological results in nontechnical reports.

6. Interpretation and Reporting of Results from an Aggregated Dataset

As discussed in section 5, reporting of radiological results can be either simple and straightforward or challenging. Therefore, the aggregation of individual results into a single dataset for graphical presentation, summarization, or other purposes must be considered carefully within the limitations of individual results. For example, it is not uncommon to have an aggregated dataset that includes positive, negative, and zero results. The WSC should exercise caution when summarizing large-scale multisite, single-measurement datasets that have a large percentage of data below detection because such data could impart substantial weight to the overall statistical computation, depending on the treatment used. Appropriate statistical tools for the analysis of such datasets are presented by Helsel (2005) and Taylor (1990). Many of the same treatments that are used with other aggregated water-quality datasets are appropriate for aggregated radiological data as long as the implications and limitations cited in this document are clearly accounted for.

Table 5. An example of typical information that could be provided when publishing radiological results in a nontechnical report.

[Filtered water samples were collected from wells and analyzed for gross alpha, radium-226, and radium-228 using U.S. Environmental Protection Agency methods EPA 900.0, EPA 903.1 and EPA 904.0, respectively; gross alpha analysis is referenced to a detector calibrated using 230Th]

Sampling site	Contaminant, units[1]	MCL[1]	Number of samples[1]	Average concentration[1]	Number of results greater than the critical level[1]	Range of concentrations[1]
Well 5	Gross alpha, pCi/L	15	5	5.86	4	ND to 9.71
Well 6	Gross alpha, pCi/L	15	7	9.2	7	6.5 to 11
Well 5	^{226}Ra + ^{228}Ra, pCi/L	5	5	2.97	5	2.63 to 3.31
Well 6	^{226}Ra + ^{228}Ra, pCi/L	5	7	0.65	3	ND to 1.05

[1] Definitions:

The average contaminant concentration, uncertainty, and critical level for a radiological constituent(s) is given in picocuries per liter (pCi/L).

The average is calculated by adding together all the individual results from a sampling site and dividing the sum by the number of individual results.

The Safe Drinking Water Act's Maximum Contaminant Level (MCL) is the highest concentration of a contaminant that is allowed in drinking water.

The number of samples corresponds to the number of samples analyzed from the location.

The uncertainty characterizes the range of the concentrations, low to high limit, which could reasonably be attributed to the radiological measurement.

The critical level is the concentration below which results are considered to be nondetections with a 5-percent probability of false detection.

A radiological contaminant is not detected (ND) when its concentration is less than the critical level.

The combined standard uncertainty (CSU) may be used when interpreting radiological results. For example, a graphic display of the sample result and CSU for four samples collected from the same location is provided in figure 1. The CSU provides an upper and lower limit to the range in which the true sample result lies; the bar chart shows the relation between the activities measured in individual samples.

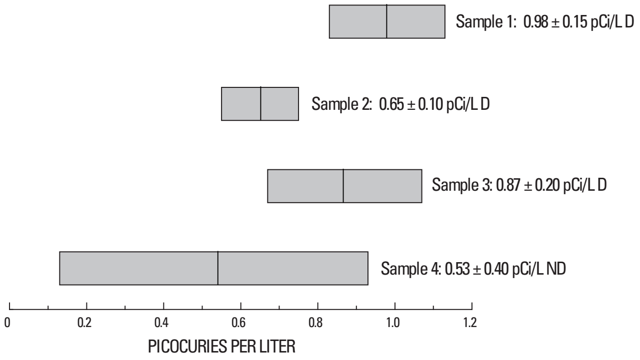

Figure 1. Graphical interpretation of radiological results. The detection (D) and nondetection (ND) values are shown, and the 68-percent confidence level or 1-sigma Combined Standard Uncertainty (1σ CSU) are identified by the shaded areas. Units are picocuries per liter (pCi/L).

The results and associated CSUs from two samples collected at the same site and time (or duplicate samples in a laboratory's batch QC) can be evaluated to determine whether they are statistically the same or different. An example of a simple equation that may be used to determine if two results (R_1 and R_2) with their associated CSUs (CSU_1 and CSU_2) are different is provided in equation 5. This equation is taken from the concept of normalized absolute difference (Paar and Porterfield, 1997), which tests the null hypothesis that the results do not differ significantly when compared to their respective CSUs. When the normalized absolute difference expression exceeds the z value, the results may be considered to be different on the basis of a defined significance level. It is common to use a z value of 2 or 3 (corresponding to 5 and about 0.3 percent significance levels, respectively).

$$| R_1 - R_2 | / \sqrt{\left(CSU_1^{\,2} + CSU_2^{\,2}\right)} > z \qquad (5)$$

Other possible approaches for interpreting or presenting aggregated radiological results, such as a statistical summary or graphical illustration, are provided herein as examples. These examples are not meant to be all-inclusive nor are they the only viable approaches. However, they serve to provide a perspective on aggregating and displaying radiological data. As with any interpretation or presentation of data, any approach should be reproducible and documented.

For certain projects, a WSC may want to use statistical analysis to summarize radiological results from samples collected from the same location at different times or from samples collected at different locations. Results obtained from the NWIS database should be reviewed and noted for acceptability before aggregating the data. When results are determined to be acceptable, the statistical analysis shall include the concentration and CSU (include the ssL_C for graphical representations) no matter if the result is negative, zero, or determined to be detected or nondetected. Excluding any positive, negative, or zero result from a dataset will bias the statistical evaluation and lead to possible erroneous conclusions.

Basic statistical terms such as the mean and standard error of the mean can be used to summarize aggregated measurements and are presented here. Other statistical approaches also can be used, but their description and use are beyond the scope of this report. Information on other statistical treatments can be found in Bevington and Robinson (1992). The average (x) of multiple laboratory measurements $x_1, x_2, \ldots, x_N$ of the same sample or of samples collected at the same location at the same time can be calculated using equation 6, where N is the number of measurements.

$$\overline{x} = \frac{x_1 + x_2 + \cdots + x_N}{N} \qquad (6)$$

The corresponding standard uncertainty of $\overline{x}$, based on the variance of the measurement $u^2(x_N)$, can be calculated using either equation 7 or 8, depending on how strongly correlated the measurements are with each other.

$$\frac{1}{N}\sqrt{u^2(x_1) + \cdots + u^2(x_N)} \qquad (7)$$

$$\frac{u(x_1) + \cdots + u(x_N)}{N} \qquad (8)$$

If all the measurement errors are essentially independent, the standard uncertainty is calculated using equation 7. If all the measurement errors are very strongly correlated, the standard uncertainty is calculated using equation 8. Equation 7 reduces the uncertainty roughly by a factor of $1/\sqrt{N}$, whereas equation 8 does not reduce the uncertainty at all.

For single measurements on samples collected at different locations and (or) times, it is not appropriate to propagate the uncertainties for the individual measurements when calculating the average because of the variability in sample collection. Sampling variability is usually assumed to be much larger than laboratory measurement variability. Therefore, in this case, the standard error of the mean is the best estimate of uncertainty for the average measurement and is calculated using equation 9.

$$s(\overline{x}) = \sqrt{\frac{1}{N(N-1)} \sum_{i=1}^{N} (x_i - \overline{x})^2} \qquad (9)$$

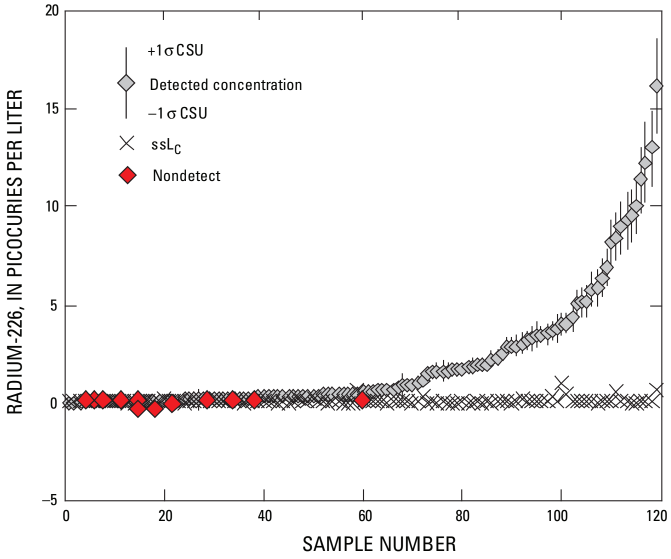

Figure 2. Graphical presentation of radiological results with pertinent supporting information.

All the required radiological parameters also can be presented graphically. Figure 2 illustrates ^{226}Ra results for 120 water samples. The figure includes reported results, their corresponding CSU and ssL$_C$, and nondetections. The dataset also included a few negative results that could be presented as reported and without manipulation or substitution. This simple illustration provides perspective on the overall dataset, including the range of detections and their concentrations. Using a graphical approach to presentation of data may facilitate interpretations not immediately evident in a data table by expanding the perspective on a large radiological dataset without censoring or manipulating the data.

Many USGS water-quality projects transcend the basic reporting of occurrence data described previously and require more detailed assessments of fate and transport or other process-oriented interpretations, such as comparisons of aggregated datasets between different environmental sites. It is beyond the scope of this report to provide details on all such activities. However, the WSC may build on the basic concepts of data reporting described in this report to reliably design and carry out its technically sound projects. Most of the potentially limiting factors are centered on the pitfalls that could arise if interpretations are made without consideration of the uncertainties in each individual result or aggregate of results. For example, in order to assess the fate of a naturally occurring radiological constituent, the WSC may need to distinguish between low levels of true environmental concentrations and the uncertainties due to laboratory analyses. In some cases, the uncertainties will be large, thereby making calculations of concentration gradients or differences between sites impossible or unreliable (for example, the data from sample numbers 1–40 in figure 2 are all of similar magnitude and associated uncertainty). On the other hand, in many cases, the differences between measurements will exceed the associated uncertainties by large margins and therefore could reliably be compared in these types of analyses (for example, in figure 2, the data point for sample number 80 could reliably be compared to the data point for sample number 100). In addition, statistical analysis of individual or aggregated data also should consider the associated uncertainties as part of the interpretation.

Acknowledgments

Anita Bhatt (U.S. Department of Energy-Idaho National Laboratory Radiological and Environmental Sciences Laboratory), Richard Graham (U.S. Environmental Protection Agency –Region VIII), John Griggs and Keith McCroan (U.S. Environmental Protection Agency –National Air and Radiation Environmental Laboratory), Rodney Melgard (Eberline Services Analytical Laboratory), Donovan Porterfield (Los Alamos National Laboratory), Robert Shannon (Quality Radioanalytical Support, LLC), and Myint Thein (U.S. Department of Energy, Oak Ridge National Laboratory) are thanked for graciously reviewing this report. The authors also thank James Eychaner, Michael Focazio, Dorrie Gellenbeck, Jeanne Hatcher, Gregory Mohrman, Lisa Olsen, Lisa Senior, Sylvia Stork, and Zoltan Szabo of the U.S. Geological Survey for their thorough reviews, which greatly improved the report.

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Glossary

Many of the definitions in the glossary are taken from MARLAP (2004).

A

activity: Mean rate of nuclear decay occurring in a given quantity of material. The “Curie” unit for activity is currently (2008) used by the USGS. The SI unit of activity is the becquerel (Bq), which equals one nuclear transformation per second. One curie equals 3.7×10^{10} Bq.

activity reference date: The date that is synonymous with the activity (concentration) on the day of sample collection.

aliquant: A representative portion of a homogeneous sample removed for the purpose of analysis or other chemical treatment. The quantity removed is not an evenly divisible part of the whole sample. By contrast, an aliquot is an evenly divisible part of the whole.

B

background (instrument): Radiation detected by an instrument when no source is present. The background radiation that is detected may come from radionuclides in the materials of construction of the detector, its housing, its electronics, and the building as well as the environment and natural radiation.

bias (of a measurement process): A persistent deviation of the mean measured result from the true or accepted reference value of the quantity being measured, which does not vary if a measurement is repeated.

blank (analytical or method): A sample that is assumed to be essentially free of the radionuclide that is carried through the radiochemical preparation, analysis, mounting, and measurement process in the same manner as a routine sample of a given matrix.

C

calibration: The set of operations that establish, under specified conditions, the relation between values indicated by a measuring instrument or measuring system, or values represented by a material measure, and the corresponding known value of a parameter of interest.

combined standard uncertainty (CSU): Standard uncertainty of an output estimate calculated by combining the standard uncertainties of the input estimates. The combined standard uncertainty of y is denoted by $u_C(y)$. The CSU is reported at the 68 percent or 1-sigma (1σ) confidence level.

critical level (L_C): In the context of analyte detection, critical level means the minimum measured value (for example, of the instrument signal or the radionuclide concentration) that indicates a positive (nonzero) amount of a radionuclide is present in the material within a specified probable error. The critical level is sometimes called the critical value or decision level. The general use of the term “critical level” is for a method wherein nominal measurement parameters are used in the calculation. Contrast this use to the sample-specific critical level (ssL_C) defined herein.

D

decay factor: The fractional amount of the original radionuclide activity in a sample that remains after decay in the time interval between sample collection and sample analysis.

duplicate sample: Two equal-sized samples of the material being analyzed, prepared, and analyzed separately as part of the same batch, used in the laboratory to measure the overall precision of the sample-measurement process beginning with laboratory subsampling of the field sample.

E

emission probability per decay event: The fraction of total decay events for which a particular particle or photon is emitted. The emission probability per decay event is also known as the branching fraction or branching ratio.

I

ingrowth factor: The activity of a supported radionuclide progeny at a specific time after chemical separation, expressed as a fraction of the amount of radioactivity at full ingrowth.

L

laboratory control sample: A standard material of known composition or an artificial sample (created by fortification of a clean material similar in nature to the sample), which is prepared and analyzed in the same manner as the sample. In an ideal situation, the result of an analysis of the laboratory control sample should be equivalent to (give 100 percent of) the target analyte concentration or activity known to be present in the fortified sample or standard material. The result normally is expressed as percent recovery.

M

matrix spike sample: An aliquant of a sample prepared by adding a known quantity of target analytes to a specified amount of matrix and subjected to the entire analytical procedure to establish if the method or procedure is appropriate for the analysis of the particular matrix.

maximum contaminant level (MCL): A regulatory limit established by the U.S. Environmental Protection Agency (USEPA) for the concentration of certain radionuclides in drinking water. The highest level (concentration) of a contaminant that is allowed in drinking water distributed to the public. MCLs are set as close as feasible to the level believed to cause no human health effects, while using the best available treatment technology and taking cost into consideration. MCLs are enforceable standards.

measurement quality objective (MQO): The analytical data requirements of the data-quality objectives, which are project- or program-specific and can be quantitative or qualitative. These analytical data requirements serve as performance measurement criteria or objectives of the analytical process. Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP) refer to these performance objectives as MQOs. Examples of quantitative MQOs include statements of required radionuclide detectability and the uncertainty of the analytical protocol at a specified radionuclide concentration, such as an action level. Examples of qualitative MQOs include statements of the required specificity of the analytical protocol (for example, the ability to analyze for the radionuclide of interest, given the presence of interferences).

method blank: A sample assumed to be essentially target analyte-free that is carried through the radiochemical preparation, analysis, mounting, and measurement process in the same manner as a routine sample of a given matrix.

minimum detectable concentration (MDC): The minimum detectable concentration of the analyte in a sample. The smallest (true) radionuclide concentration that gives a specified probability (Type II - β) that the value of the measured radionuclide will exceed its critical level concentration (that is, that the material analyzed is not "blank" or free of analyte) (Currie, 1968; MARLAP, 2004, chapter 20). The general use of the term "MDC" or "*a priori* MDC" is for a method wherein nominal measurement parameters are used in the calculation. Contrast this use to the sample-specific MDC (ssMDC) defined herein.

N

nominal value: A value related to a designated or theoretical size that may vary from the actual.

null hypothesis (H_0): One of two mutually exclusive statements tested in a statistical hypothesis test (compare with alternative hypothesis). The null hypothesis is presumed to be true unless the test provides sufficient evidence to the contrary, in which case the null hypothesis is rejected and the alternative hypothesis (H_a) is accepted.

P

Poisson distribution: A random variable X has the Poisson distribution (Pr) with parameter λ if for any nonnegative integer k, $\Pr[X = k] = (\lambda^k e^{-\lambda})/k!$. In this case, both the mean and variance of X are numerically equal to λ. The Poisson distribution is often used as a model for the result of a nuclear counting measurement.

precision: The closeness of agreement between independent test results obtained by applying the experimental procedure under stipulated conditions. Precision may be expressed as the standard deviation. Conversely, imprecision is the variation of the results in a set of replicate measurements.

Q

quality control (QC): The overall system of technical activities that measures the attributes and performance of a process, item, or service against defined standards to verify that they meet the stated requirements established by the project; operational techniques and activities that are used to fulfill requirements for quality. This system of activities and checks is used to ensure that measurement systems are maintained within prescribed limits, providing protection against out-of-control conditions and ensuring that the results are of acceptable quality.

quantile: A p-quantile of a random variable X is any value x_p such that the probability that $X < x_p$ is at most p and the probability that $X \leq x_p$ is at least p.

R

radioactivity: The property possessed by some elements or isotopes of spontaneously emitting energetic particles (electrons or alpha particles) by the disintegration of their atomic nuclei.

radioanalytical analysis: A general term used to denote the analysis of a sample for a specific radionuclide, group of radionuclides, or gross screening of radioactivity. The term may be used for a single radionuclide analysis or to denote a collection of analyses that may include gamma-ray spectrometric analyses, gross alpha and beta analyses, and specific radionuclide analyses that require chemical separations such as isotopic uranium, ^{226}Ra, and ^{90}Sr.

radiochemical analysis: A term used to denote the analysis of a radionuclide in a sample that requires chemical processes to isolate the radionuclide in the sample. Isotopic uranium, ^{226}Ra, and ^{90}Sr in a sample require radiochemical analyses.

radiological: An adjective relating to nuclear radiation.

radiological hold time: Refers to the time differential between the sample collection date and the final sample counting (analysis) date.

radionuclide: A nuclide that is radioactive (capable of undergoing radioactive decay).

recovery: The ratio of the amount of analyte measured in a spiked or laboratory control sample, to the amount of analyte added, usually expressed as a percentage. For a matrix spike, the measured amount of analyte is first decreased by the measured amount of analyte in the sample that was present before spiking. Contrast this to yield defined herein.

relative standard uncertainty: The ratio of the standard uncertainty of a measured result to the result itself. The relative standard uncertainty of x may be denoted by $u_r(x)$.

S

sample: A portion of material selected from a larger quantity of material or a set of individual samples or measurements drawn from a population whose properties are studied to gain information about the entire population.

sample-specific critical level (ssL$_C$): The sample-specific critical level is calculated using the parameter values measured during the generation of the sample result. This is different than the critical level for a method wherein nominal measurement parameters are used in the calculation (see critical level). Concentrations below the ssL$_C$ are considered nondetections.

sample-specific minimum detectable concentration (ssMDC): The sample-specific minimum detectable concentration is calculated using the parameter values measured during the generation of the sample result (see minimum detectable concentration).

significance level: The risk (probability) of making a Type I error (α) is traditionally called the level of significance of the test.

standard uncertainty: The uncertainty of a measured value expressed as an estimated standard deviation, often call a “1-sigma” (1-σ) uncertainty. The standard uncertainty of a value x is denoted by $u(x)$.

T

Type I decision error: In a hypothesis test, the error made by rejecting the null hypothesis when it is true. A Type I decision error is sometimes called a “false detection” or a “false positive.”

Type II decision error: In a hypothesis test, the error made by failing to reject the null hypothesis when it is false. A Type II decision error is sometimes called a “false nondetection” or a “false negative.”

U

uncertainty: A parameter, usually associated with the result of a measurement, that characterizes the dispersion of the values that could reasonably be attributed to the measurement of interest.

Y

yield: The ratio of the amount of radiotracer or carrier determined in a sample analysis to the amount of radiotracer or carrier originally added to a sample. The yield is an estimate of the analyte during analytical processing. It is used as a correction factor to determine the amount of radionuclide (analyte) originally present in the sample. Yield is typically measured gravimetrically (through a carrier) or radiometrically (through a radiotracer).

Appendix: Typical Equations for Calculating Radiological Parameters

The information presented in this appendix conforms to the formulations and concepts presented in the Multi-Agency Radiological Laboratory Analytical Protocols, commonly referred to as MARLAP (2004), the International Organization for Standardization (1995), and International Union of Pure and Applied Chemistry (1995).

A1. Calculating the Concentration

The general equation used to calculate concentration takes on the following form (MARLAP, 2004, chapter 19):

$$\mathit{Concentration} = \frac{(\mathit{Gross\ Instrument\ Signal}) - (\mathit{Blank\ Signal} + \mathit{Estimated\ Interferences})}{(\mathit{Sensitivity\ Factor})} \tag{A1}$$

where:

Concentration is in pCi/L;

Gross Instrument Signal is detector response in units of detector events registered per unit time from all constituents in the sample (includes detector and process-related background events);

Blank Signal is detector events registered per unit time from analyzing a blank sample having no target constituent;

Estimated Interferences is detector counts registered per unit time from nontarget constituents in the sample; and

Sensitivity Factor is a combination of multiplicative parameters, such as sample size, detector efficiency for radiation emitted, chemical yield of process, decay factor, ingrowth factor, and unit conversion factor.

The complexity of the equation used to calculate the concentration varies substantially and usually depends on the type of radiation emitted during radioactive decay (α, β, γ) and the radiochemical method chosen. The basic equation that has all possible parameters incorporated in a complex equation, such as for ^{228}Ra or ^{90}Sr by the analysis of their decay products ^{228}Ac and ^{90}Y, has the form taken from "Inventory of Radiological Methodologies for Sites Contaminated with Radioactive Material" (U.S. Environmental Protection Agency, 2006).

$$\mathit{Concentration}\ \left(pCi/L\right) = \frac{\left(\frac{N_s}{t_s} - \frac{N_B}{t_B}\right) \times \lambda_2 t_S}{CF \times B \times Y \times \varepsilon \times V \times I \times DF_P \times DF_{DP} \times \left(1 - e^{-\lambda_2 t_S}\right)} \tag{A2}$$

where:

N_S	is the number of accumulated detector events (counts) for the decay product;
N_B	is the number of background detector events (counts) for the equivalent sample count interval;
t	is the counting interval for sample (t_S) and background (t_B);
CF	is the factor used for converting to desired reporting units, typically 2.22 disintegrations per picocurie (pCi);
V	is the sample size (mass or volume);
B	is the branching fraction of the particle emission being counted. This is the fraction of all decays that result in an emission of the characteristic radiation (alpha, beta, or gamma);
Y	is the chemical yield of the analysis;
ε	is the detector efficiency for the particular emission of the radionuclide;
λ	is the decay constant of the radionuclide ($\lambda = 0.69315/t_{1/2}$); and $t_{1/2}$ is the half-life of the radionuclide (λ_1 for parent and λ_2 for the decay product);
DF_P or $\left(e^{-\lambda_1 T_1}\right)$	is the decay correction factor for the parent from sample collection to second chemical separation;
DF_{DP} or $\left(e^{-\lambda_2 T_3}\right)$	is the decay correction factor for decay product from second chemical separation to start of counting;
I or $\left(1-e^{-\lambda_2 T_2}\right)$	is the ingrowth correction factor for the ingrowth of the decay product used to calculate the activity of a parent;
T_1	is the time interval between sampling and beginning the sample count;
T_2	is the time interval between first and second decay product chemical separations;
T_3	is the time interval between second decay product chemical separation and count;
$\lambda_2 t_s/(1-e^{-\lambda_2 t_s})$	is the correction factor for radioactive decay of the decay product during the counting interval.

Note: Because measurement parameters are never truly known exactly, each factor has an associated standard uncertainty.

For a simple analysis, such as gross alpha and beta analysis, the following equation is used:

$$"c_\alpha" \; Concentration \; (\mathrm{pCi/L}) = \frac{\left(\frac{N_S}{t_S} - \frac{N_B}{t_B}\right)}{CF \times \varepsilon \times V} \tag{A3}$$

A2. Calculating the Combined Standard Uncertainty

As presented in section A1, each factor in the equation used to calculate the concentration has an associated uncertainty called the standard uncertainty. By convention, a standard uncertainty is quoted at the 68-percent or 1-sigma confidence level. For example, the determination of the detector efficiency is not exact because of the uncertainty in the radioactive source used to determine the detector efficiency and the Poisson random counting uncertainty when counting the radioactive source.

The uncertainty for the stated concentration is calculated by combining (or propagating) these standard uncertainties into an overall uncertainty called the combined standard uncertainty. MARLAP (2004, chapter 19) uses a first-order uncertainty propagation formula shown in equation A4 to propagate the standard uncertainties. The general formula uses the partial differential of each factor (x_i) to calculate the concentration y. Equation A4 is used to calculate the variance $u_c^2(y)$ of the concentration based on the uncertainty propagation with uncorrelated inputs.

$$u_c^2(y) = \sum_{i=1}^{N} \left(\frac{\partial f}{\partial x_i}\right)^2 u^2(x_i) \tag{A4}$$

The combined standard uncertainty $u_c(y)$ is calculated by taking the square root of the variance $u_c^2(y)$.

For the simple concentration equation A3, the resulting equation (MARLAP, 2004, chapter 19) used to calculate the variance is the following:

$$u_c^2(c_\alpha) = \left(\frac{\partial c_\alpha}{\partial N_S}\right)^2 u^2(N_S) + \left(\frac{\partial c_\alpha}{\partial N_B}\right)^2 u^2(N_B) + \left(\frac{\partial c_\alpha}{\partial \varepsilon}\right)^2 u^2(\varepsilon) + \left(\frac{\partial c_\alpha}{\partial V}\right)^2 u^2(V) \tag{A5}$$

where the uncertainties in the time variables are assumed to be negligible compared to the uncertainties of the other terms. Equation A5 reduces to equation A6 using the same parameters defined for equation A2.

$$u_c^2(c_\alpha) = \frac{\left(N_S \times t_B^2 - N_B \times t_S^2\right)}{t_S^2 \times t_B^2 \times \varepsilon^2 \times V^2} + \frac{\left(\frac{N_S}{t_S} - \frac{N_B}{t_B}\right)}{\varepsilon^2 \times V^2} \times \left[\frac{u^2(\varepsilon)}{\varepsilon^2} + \frac{u^2(V)}{V^2}\right] \tag{A6}$$

The combined standard uncertainty $u_c(c_\alpha)$ of c_α is calculated by

$$u_c(c_\alpha) = \sqrt{u_c^2(c_\alpha)} \tag{A7}$$

A3. Calculating the Critical Level

A3.1 General Principles

An analysis of a radiological sample will produce a gross signal response that is related to the quantity of analyte present in the sample. However, random measurement uncertainties will cause this signal to vary somewhat if the measurement is repeated on the same sample. A nonzero signal may be (and usually is) produced even when no analyte is present. For this reason, a laboratory analyzes a blank (or an instrument background) to determine the signal observed when no analyte is present in the blank sample and subtracts this blank signal from the gross signal to obtain a net signal for the sample being analyzed. In fact, because the signal varies when the blank measurement is repeated, there is a blank signal distribution whose parameters must be estimated. In a similar manner, the net signal response will have a distribution with an average value (assumed to be zero or μ_0) and a standard deviation (σ_0).

To determine how large an instrument signal must be to provide high confidence for the presence of the analyte in a sample, one calculates a threshold value for the net signal called the critical level L_C (Currie, 1968). The critical level also is denoted by S_C (MARLAP, 2004, chapter 20). If the observed net signal for a sample being analyzed exceeds the critical level, the radiological constituent is considered detected; otherwise, it is not detected. Because the measurement process is statistical in nature, it is possible for the net signal related to a blank sample to exceed the critical level, leading one to conclude incorrectly that the sample contains a positive amount of the analyte. Such an error is sometimes called a “false positive,” although the term Type I error terminology is favored by MARLAP (2004, chapter 20). The probability of a Type I error is often denoted by α. Before calculating the critical level, one must choose a level for α. The most commonly used level is 0.05, or 5 percent. If $\alpha = 0.05$, then one expects the net instrument signal to exceed the critical level in only about 5 percent of cases (1 in 20) when analyte-free samples or blank samples are analyzed. These concepts are shown graphically in figure A1.

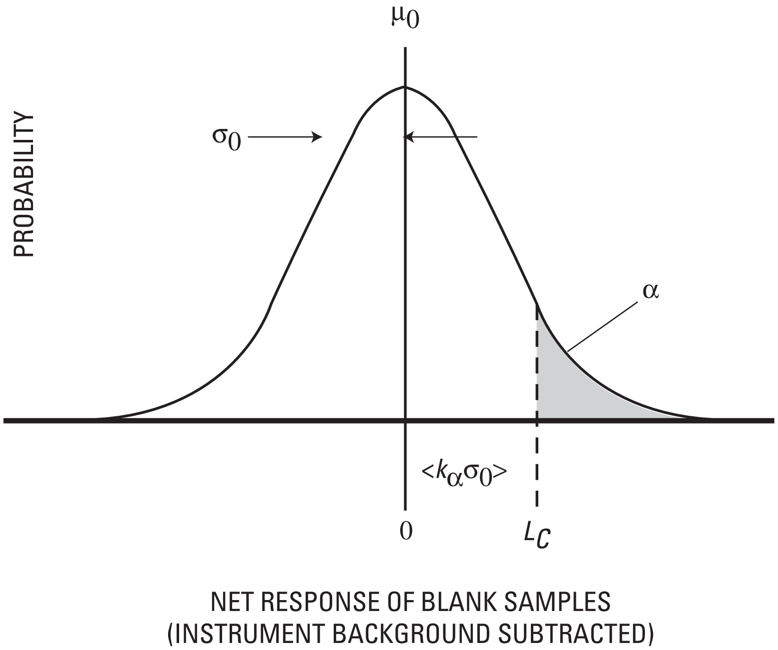

Figure A1. The curve representing the critical level concept by using a theoretical distribution of the net instrument signal (concentration) obtained when analyzing an analyte-free sample. The chosen Type I error probability σ determines the location of the critical level of the net signal L_C. For an $\alpha = 0.05$ or 5 percent, the critical level corresponds to the 95th quantile value of the normal distribution. The probability α is depicted as the area under the curve to the right of the dashed line. Note that decreasing the value of α requires increasing the critical value (shifting the dashed line to the right) and increasing the value of α decreasing the critical level (shifting the dashed line to the left). The α quantile of the standard normal distribution, using a default level of 1.645, is k_α; the mean of the net signal responses of blank sample distribution is μ_0; and the standard deviation of the net signal response of blank sample distribution is σ_0.

A3.2 Calculating the Sample-Specific Critical Level (ssL_C)

The sample-specific critical level (ssL_C) is calculated by the contract laboratory using all the sample-specific parameter values. A sample-specific critical level is calculated using the net instrument background counting distribution and sample-specific factors, and by assuming that the mean value of the net instrument background distribution is zero. The ssL_c is commonly calculated using equations A8a and A8b (Currie, 1968).

$$ssL_C = \frac{1.645 \times \sigma_0}{CF \times t_s \times V \times B \times \varepsilon \times Y \times DF \times I} \tag{A8a}$$

and

$$\sigma_0 = \sqrt{N_B \frac{t_S}{t_B}\left(1 + \frac{t_S}{t_B}\right)} \tag{A8b}$$

where:

σ_0 is the standard deviation of the net instrument background (counts);
N_B is the background counts in background counting interval
CF is the unit conversion factor, typically 2.22 disintegrations per minute per picocurie;
t_S is the counting time of sample (minute);
t_B is the counting time of background (minute);
V is the sample size (mass or volume);
B is the branching fraction of the particle emission being counted;
ε is the fractional detector efficiency for the particular emission of the radionuclide;
Y is the chemical yield of the analysis;
DF is the decay factor; and
I is the correction factor for the ingrowth of progeny used to calculate the activity of a parent.

Equation A8a is not suitable when the number of detector background counts is small or zero over the background counting time. This may be the case for analyses that use low background detection systems, such as alpha or gamma ray spectrometers. For such applications, the equations discussed in chapter 20, attachment 20A of MARLAP (2004) need to be reviewed and applied.

The specific equation that is used by the contract laboratories to calculate the ssL_C for various radioanalytical methods will be specified by the NWQL in the PWS.

A3.3 Practical Approach for Verifying the Reported Sample-Specific Critical Level

From a practical point of view, when the sample and background counting times are nearly equal, multiplying the CSU associated with a QC blank's result by 2 approximates the critical level of a method (not an individual result). However, this approximation is only good when the parameters used to calculate the concentration for the blank are nearly equal to the average values used for the method.

An approximation of 2 times the CSU of the sample or blank result (or sometimes slightly greater depending on the background of the instrument used for the measurement) can be used for verifying the reported ssL_C. This assumes near equal sample and background counting times. For example, if a radiological result is 0.121 ± 0.091 pCi/L, the ssL_C can be estimated to be 0.18 or about 0.20 pCi/L. This ssL_C approximation is useful to verify that the laboratory has not made a substantial error in calculating the sample-specific critical level. The approximation of 2 times the CSU may provide reasonable estimates for most applications but should not be used to categorically indicate that a mistake has been made without evaluating the relation between the reported result, CSU, ssL_C, and ssMDC.

A4. Calculating the Minimum Detectable Concentration

A4.1 General Principles

The *a priori* minimum detectable concentration (*a priori* MDC) is a hypothetical predictive concept that is used to estimate the detection capability of a measurement process (method) under defined circumstances. The concept is *a priori* or before the fact (before a sample measurement) and is not to be used to evaluate individual measurement results. After a measurement has been made, the result and its Combined Standard Uncertainty (CSU) are the important quantities that are used to compare analytical results to historical values for the sampling site.

Once the critical level L_C (instrument response) has been defined for a method based on a distribution of blank samples and an α probability of Type I error (false detection) rate, an *a priori* MDC may be established by specifying the acceptable Type II error rate β (false nondetection) and the standard deviation of the probability distribution σ_D of the net signal response when the true value S_D is equal to the MDC value (Currie, 1968). The concept assumes no systematic errors such as method bias. The *a priori* MDC is defined so that the probability distribution of the possible measurement responses when S_D equals the MDC crosses the critical level L_C at the 1-β fraction of the distribution.

Graphically, the theoretical *a priori* MDC distribution and its relation to the critical level are shown in figure A2 (based on figure 20.1 in MARLAP, 2004). In this figure, S_D corresponds to the *a priori* MDC value. Under the *a priori* MDC concept, the shaded area under the S_D distribution corresponds to the β or the lower 5 percent of the distribution. Analytical results with values in the shaded area would not be considered different from background because their values are below the critical level. The nonshaded area of the S_D distribution corresponds to the 1-β or the upper 95 percent of the distribution. Analytical results with values in this region would be considered positive because their values are greater than the critical level. Note that all possible analytical result values between L_C and S_D (45 percent of the S_D distribution) would be considered different from background and thus positive values.

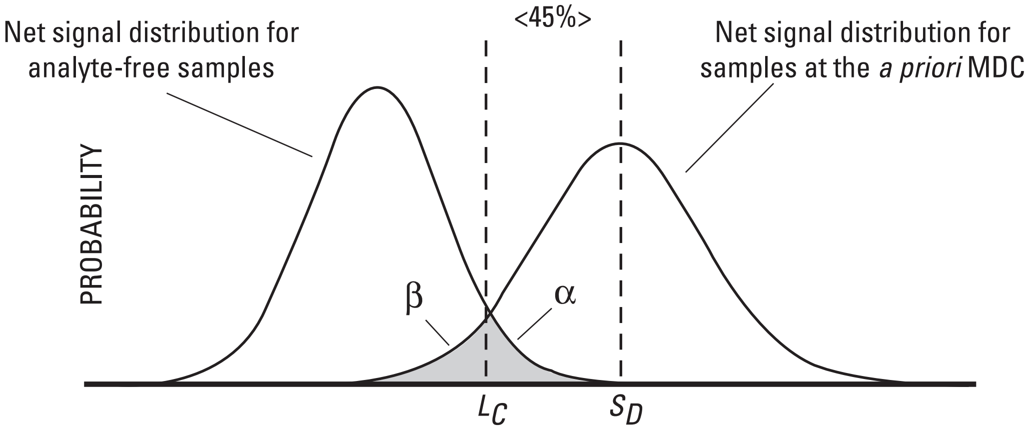

Figure A2. Graphical representation of the *a priori* Minimum Detectable Concentration concept (taken from figure 20.1; in MARLAP, 2004). The false nondetection Type I error (α, 5-percent probability), the false detection Type II error (β, 5-percent probability), the critical level (L_c), and the *a priori* minimum detectable concentration (S_D) are shown in the figure.

If each sample in a set of 100 samples were spiked with a radionuclide at the *a priori* MDC concentration and analyzed, the mean concentration of the 100 analyses would be the *a priori* MDC value (S_D) and the relative one standard deviation of the distribution of analytical results around the *a priori* MDC would be about 30 percent. Therefore, the spread in the individual results from the set of 100 samples would range from slightly less (about 5 percent) than the critical level to about twice the *a priori* MDC value. However, 95 out of the 100 results would exceed the critical level. By specifying the *a priori* MDC in the PWS, a false nondetection β probability of 5 percent could be assumed for a distribution of results (from multiple samples or analyses of the same sample) when the true concentration is at the hypothetical MDC. For USGS applications, typically only one sample from a particular site may be provided and only one analysis of the sample performed. A result from a single analysis that is less than the hypothetical *a priori* MDC may be from the MDC distribution, but it also could be from distributions whose true concentration is less than or greater than the MDC. When a single analysis of an individual sample is made, the true concentration in the sample can be estimated with approximately 95-percent confidence to be within the range described by the reported value ± 2 CSU.

Mathematically, the *a priori* MDC S_D can be calculated using many equations, each having a different assumption and approach. MARLAP (2004, chapter 20) discusses these different approaches. The most simplified approach assumes a Gaussian distribution for L_C and S_D instrument responses. Currie (1968) used a version of the following simplified equation for instrument response, not concentration:

$$S_D = L_C + k_\beta \times \sigma_D \qquad \text{(A9)}$$

where:

k_β is the $1 - \beta$ quantile of the standard normal distribution, default value of 1.645;
L_C is the critical level response, defined earlier; and

σ_D is the standard deviation of the S_D distribution (net response).

When $\alpha=\beta$, $k_\alpha=k_\beta$, and $\sigma_D \sim \sigma_0$ and where σ_0 is the standard deviation of the net background response distribution, equation A9 reduces to the most simplified form:

$$S_D = 2.71 + 3.29 \times \sigma_0 \tag{A10}$$

When the net response background distribution is more like a Poisson distribution (when the background or sample counts for the analysis are less than 70) rather than like the assumed Gaussian distribution, the use of this equation may give an MDC wherein the observed probability of false detection may be higher than the assumed α value.

An approach has been suggested by Rucker (2001) for determining a method *a priori* MDC-based net background standard deviation S_{B0}, in picocuries per liter, from a population of blank sample results (instrument background subtracted) as a substitute for use in equations A8 and A11. In addition, the k_α and k_β for equations A10 and A11 are replaced with the Student's *t* factor for the appropriate number of degrees of freedom. The MDC equation proposed by Rucker is

$$MDC = t^2 + 2 \times t \times S_{B0} \tag{A11}$$

where:

S_{B0} is the standard deviation of the distribution net blank results in picocuries per liter; and
t is the Student's *t* factor for the number of blank samples used to determine S_B and for the default α and β probabilities of 0.05.

As noted by Rucker, this approach accounts for all of the uncertainty in the measurements (due to the variability in the parameter values used to calculate the result), not just the counting uncertainty. As such, this approach is useful in estimating the *a priori* MDC for a method, not a sample-specific MDC, and can be applied to defining the sensitivity requirements for contract laboratory work.

A4.2 Calculating the Sample-Specific Minimum Detectable Concentration (ssMDC)

Some laboratories calculate the sample-specific Minimum Detectable Concentration (ssMDC) based on the instrument background and applicable sample-specific parameters according to the following general equation:

$$ssMDC = \frac{3.29 \times \sigma_0 + 2.71}{CF \times t_s \times B \times Y \times \varepsilon \times V \times DF \times I} \tag{A12a}$$

and

$$\sigma_0 = \sqrt{N_B \frac{t_S}{t_B}\left(1 + \frac{t_S}{t_B}\right)} \tag{A12b}$$

where:

σ_0 is the standard deviation of the net instrument background (counts);
N_B is the background counts in background counting interval;
CF is the unit conversion factor, typically 2.22 disintegrations per picocurie;
t_S is the counting time of sample (minute);
t_B is the counting time of background (minute);
V is the sample size (mass or volume);
B is the branching fraction of the particle emission being counted;
ε is the fractional detector efficiency for the particular emission of the radionuclide;
Y is the chemical yield of the analysis;
DF is the decay factor; and
I is the correction factor for the ingrowth of progeny used to calculate the activity of a parent.

Equations A12a and A12b are not used when the number of detector background counts is small or zero over the background counting time. This may be the case for analyses that use low background detection systems, such as alpha- or gamma-ray spectrometers. For such applications, the equations discussed by MARLAP (2004, chapter 20, attachment 20A) need to be reviewed and applied.

In the original concept, the average values of the applicable parameters were used to calculate the *a priori* MDC for a method. However, more recently, sample-specific values of the parameters are used to calculate the ssMDC to show that the laboratory has met the required *a priori* MDC specified in a contract. The ssMDC is not used to determine if results are different from an instrument background or a blank sample.

The specific equation that is used by the contract laboratories to calculate the ssMDC for various radioanalytical methods will be specified in the PWS by the NWQL.

A4.3 Practical Approach for Verifying the Reported Sample-Specific Minimum Detectable Concentration

There are at least two approaches or rules of thumb that can be used to estimate whether a reported ssMDC has been calculated properly. The first approach compares the reported ssMDC to the reported ssL_C. The ssMDC should be approximately 2 times the reported ssL_C (or slightly greater depending on the background of the instrument used for the measurement). The second approach is to compare the reported ssMDC of a sample to a multiple of the reported CSU. This approach is only applicable when dealing with acceptable blank and negative results or with acceptable positive results whose absolute value is less than approximately 3 times the reported ssMDC value. For such cases, the reported ssMDC should be approximately 3 to 4 times the CSU of the sample result. Different multipliers may be applied for certain methods of analysis and instrument backgrounds. Analytical concentrations at or near the ssMDC should have a relative CSU of approximately 30 percent for most methods, with the exception of methods for the low-level determination of alpha-emitting nuclides. The relative CSU decreases with successively higher concentrations above the MDC. These rules of thumb are based on the assumed relation of the relative CSU at the MDC for paired observations.

A4.4 The Effect of Sample Size and Counting Time on the Reported Sample-Specific Minimum Detectable Concentration (ssMDC)

The magnitude of the ssMDC is inversely proportional to the sample volume analyzed and inversely proportional to the square root of the counting time (see equations A12a and A12b in section A4.2). Figure A3 shows the effect of reducing the size of a 1-liter sample on the magnitude of the ssMDC. When the sample volume analyzed is reduced by a factor of 2 (1 L to 0.5 L), the magnitude of ssMDC is increased by a factor of 2 (1 pCi/L to 2 pCi/L). Figure A4 shows the effect of reducing the amount of time a sample is analyzed by a radiation detector (instrument counting time) on the magnitude of the ssMDC. When the counting time is reduced by a factor of 2 from 200 minutes to 100 minutes, the magnitude of the MDC is increased by a factor of $\sqrt{2}$ from 1 pCi/L to 1.41 pCi/L (see equations A12a and A12b).

A4.5 The Relation between the Combined Standard Uncertainty and the Calculated Activity in a Sample for Two Radioanalytical Measurement Techniques

Several graphical illustrations of the relation between the reported activity in a sample and the Combined Standard Uncertainty (CSU) of the result have been developed. These relations are shown in figures A5a and A5b for gross beta activity analysis of water and in figures A6a and A6b for isotopic uranium analysis of water by alpha spectrometry techniques. Typical sources (detector efficiency and chemical yield) and their standard uncertainties were used (for illustrative purposes only) in the calculation of the CSU and the development of the graphs.

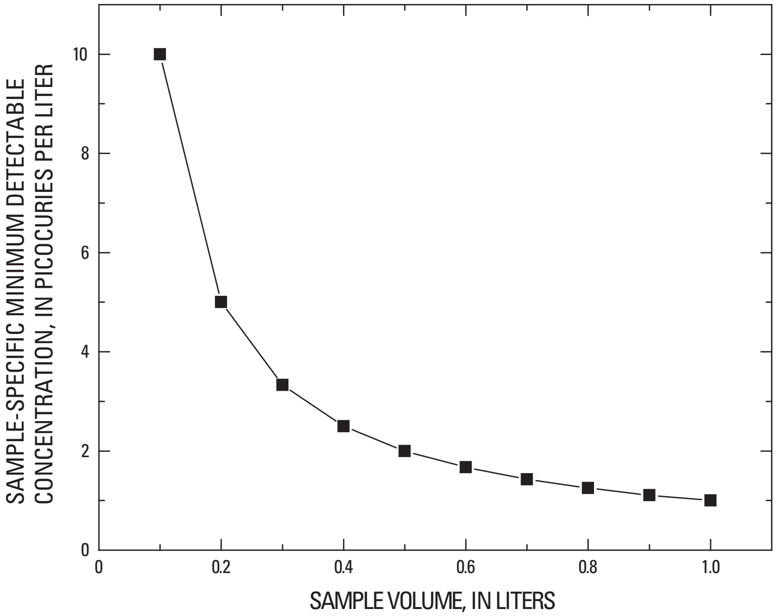

Figure A3. The sample-specific Minimum Detectable Concentration (ssMDC) as a function of analysis sample volume.

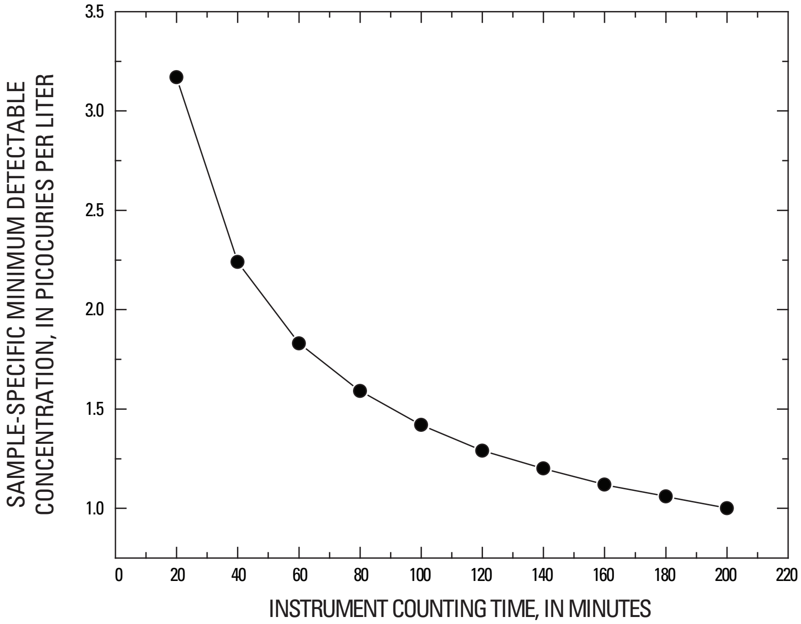

Figure A4. The sample-specific Minimum Detectable Concentration (ssMDC) as a function of analysis counting time.

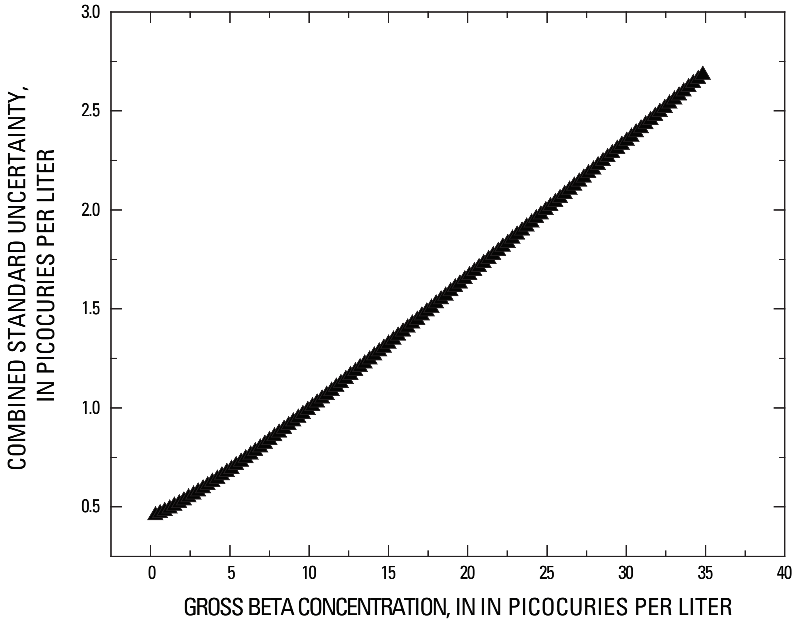

Figure A5a. Typical Combined Standard Uncertainty (CSU) as a function of gross beta concentration.

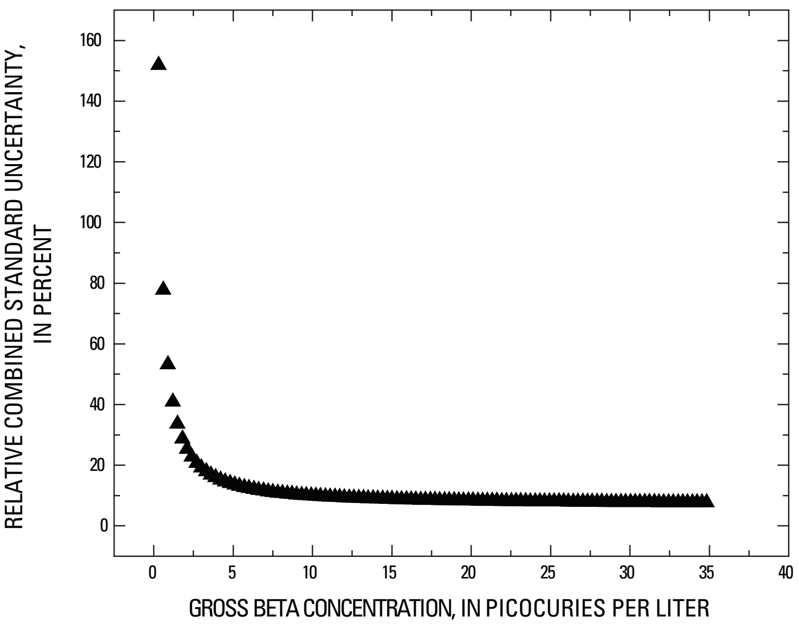

Figure A5b. Typical relative Combined Standard Uncertainty (CSU) as a function of gross beta concentration.

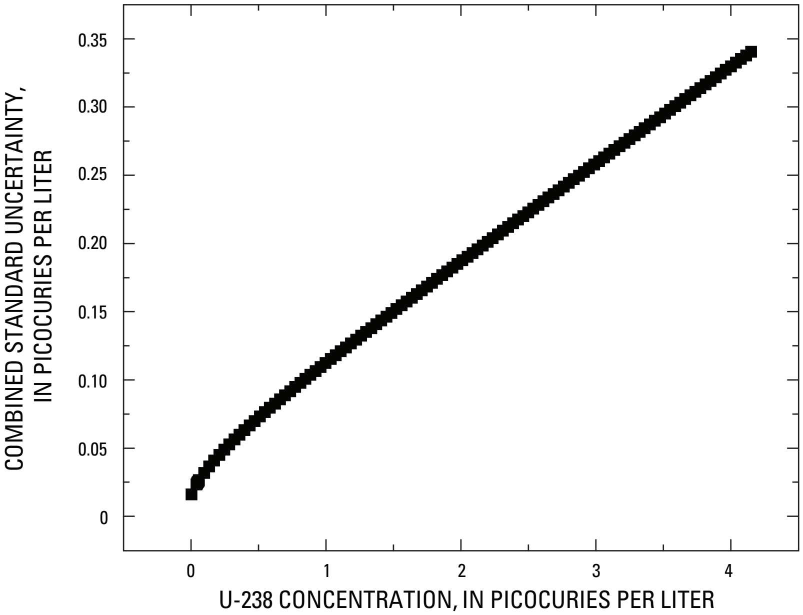

Figure A6a. Typical Combined Standard Uncertainty (CSU) as a function of Uranium-238 concentration.

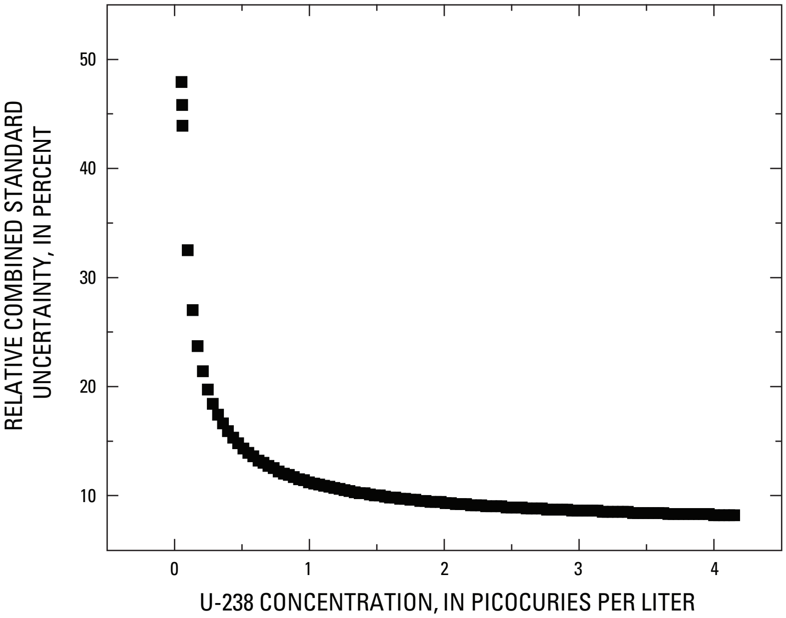

Figure A6b. Typical relative Combined Standard Uncertainty (CSU) as a function of Uranium-238 concentration.

Manuscript approved for publication June 13, 2008

Prepared by the U.S. Geological Survey Publishing Service Center, Denver, Colorado

USGS Publishing Staff
  Edited by Mary A. Kidd
  Report approved by Keith J. Lucey
  Cover designed by Suzanne C. Roberts and Sharon Powers
  Report prepared by Sharon Powers

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  World Wide Web: *http://nwql.cr.usgs.gov*

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McCurdy and others—**Interpreting and Reporting Radiological Water-Quality Data**—USGS TM 5–B6

ISBN 978-141132223-3
9 781411 322233

Printed on recycled paper

Exhibit E

SIERRA CLUB
ATLANTIC CHAPTER

744 Broadway ● Albany, NY 12207

March 6, 2019

VIA EMAIL

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Re: Suggestions for Additional Radioactivity Testing at Hakes Landfill

Dear Supervisor Horton and Members of the Campbell Town Board:

Following up on our letter of February 22, 2019, we would like to offer some suggestions for additional testing and evaluation of radioactivity in the Hakes landfill.

We enclose a short memorandum from Dr. Raymond Vaughan describing some relatively simple radon testing that could be conducted of the Hakes landfill gas (LFG) and some information about the protocols that need to be following in continuing to test the Hakes landfill leachate for Lead-214 and Bismuth-214.

Respectfully,

Kate Bartholomew

Kate Bartholomew, Chair
Sierra Club Atlantic Chapter

Karen G Ash

Karen Ash, Chair
Concerned Citizens of Allegany County

Gary McCaslin

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Enclosure

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From:	Raymond C. Vaughan, Ph.D., P.G.
To:	Sierra Club Atlantic Chapter
Subject:	Recommended testing of Hakes landfill gas (LFG) and leachate
Date:	March 5, 2019

This memo outlines the testing needed to reach a better understanding of **a**) the intermittently high levels of Lead-214, Bismuth-214, and Radon-222 in leachate from Hakes C&D landfill, **b**) the radium in the landfill from which these radionuclides were generated, and **c**) the associated health risk. Depending on the results, followup testing may be needed for more detailed characterization of radium in the landfill and the associated health risk

Radon is a radioactive gas that's a good "tracer" for radium, in the same sense that a visible plume of smoke can be traced back to the fire that generated the smoke. This is not a perfect analogy because radon is a chemically inert gas that is not destroyed by fire; it simply passes through a flame without reacting. However, given radon's usefulness as a tracer, the testing outlined here can provide a better understanding of the radon concentrations in both landfill gas (LFG) and leachate, including a determination of whether these levels are routinely high, routinely low, or highly variable. Then, if the radon levels are found to be routinely high or highly variable, additional testing would likely be needed to trace the radon gas back to its radium source(s) within the landfill. Focusing on radon in the tests outlined here has the advantage of being minimally invasive. Additional testing, if needed, would likely require excavation and/or new boreholes to identify and quantify the radium source(s).

Leachate and LFG are the two main landfill fluids into which radon may mix. Both are accessible for sample collection and testing. Leachate is generally sampled for individual cells or pairs of cells, while samples of LFG can be taken from the collection system that combines the LFG from various cells. According to the Draft Supplemental Environmental Impact Statement (DSEIS) for the Hakes C&D Landfill Expansion Project, page 36, the LFG collection system connects "the ten vertical landfill gas wells, the four horizontal collectors, and seven leachate cleanouts, to the blower and flare system. The blower creates a vacuum and provides for the collection and treatment of more gas than would be treated through a passive collection system. Within the flare, the landfill gasses are mixed with air and combusted." As noted above, radon does not burn; it passes through the flare into the surrounding air.

Four types of LFG and leachate testing are recommended:

1. Arrange to have a qualified testing company perform monthly testing of radon concentration (activity) in Hakes LFG, reported in pCi/L, for at least a year. The testing company should have

a record of experience in sampling and testing for radon in natural gas pipelines,[1] and the monthly testing should meet the scheduling and other conditions expressed below as 1.a, 1.b, 1.c., and 1.d.

> a. Based on descriptions of the Hakes LFG system (LFG wells, collectors, blower, flare) in the DSEIS p. 36 and Appendix H, the monthly testing of radon in LFG should *use the same sampling port* (at flare inlet) that has been routinely used to test LFG for other analytes such as methane, CO_2, and H_2S.
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> b. Ensure and document that the Hakes LFG system has been in its normal mode of operation for at least 3 days prior to each monthly test of radon in LFG.
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> c. For each monthly test of radon in LFG that is close to a semiannual leachate sample collection date for Method 901.1 gamma spectroscopy/spectrometry leachate testing, *coordinate the two tests such that the test of radon in LFG is 5 ± 2 days prior to the semiannual leachate sample collection date for Method 901.1 testing*.
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> d. For each monthly test of radon in LFG that is close to a quarterly test of LFG for other analytes, *coordinate the two tests such that both tests are done at approximately the same time, preferably on the same day*.

2. Continue semiannual EPA Method 901.1 gamma spectroscopy/spectrometry leachate testing for Lead-214 and Bismuth-214 in both unfiltered and filtered ("dissolved") samples, with a few additional conditions or precautions as expressed below as 2.a, 2.b, and 3. This type of testing has been done routinely on Hakes leachate and should be continued.

> a. Adopt a uniform sample holding period (between sample collection and testing) of 23 ± 2 days.[2]
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> b. Ensure that the Method 901.1 test procedure is set up and conducted so that quantitative results ranging from non-detect to at least 7000 pCi/L can be reported within a quantified uncertainty.[3]

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[1] As an example, see https://www.sgsgroup.us.com/en/news/2018/05/radon-testing-of-natural-gas-and-light-hydrocarbons. No endorsement is intended.

[2] A recent sample holding period (Hakes leachate samples collected 6/26/18 and tested by ALS Fort Collins) was 25 days for some samples and 26 days for others. A holding period of 25 days, if adopted as a standardized hold time for all semiannual Hakes 901.1 samples, would be acceptable by the above criterion. A holding period of 26 days is excessive.

[3] Method 901.1 test results, as reported in the past by Pace (Pittsburgh/Greensburg) for Hakes leachate samples, have generally met this condition. However, several of the recent Method 901.1 test results reported by ALS Fort Collins for Hakes leachate samples collected on 6/26/18 are flagged with an "NQ" qualifier, indicating spike sample recovery outside control limits which may cause the test result to be interpreted as qualitative rather than a quantitative value. Quantitative results, uncertainties, and MDCs are needed.

3. Perform Lead-210 testing of semiannual leachate samples in conjunction with the Method 901.1 testing of such samples, in the same general manner as the Lead-210 testing that was done on Hakes leachate samples collected 2/27/18. Such testing of each sample will serve as a check on the Lead-214 and Bismuth-214 results and the associated radon levels.

4. For all of the leachate samples collected semiannually for Method 901.1 testing, *set up and follow a protocol to minimize loss of radon from the sample containers during the standardized holding period of 23 ± 2 days, and set up and follow a protocol to measure and document the approximate loss of radon from the sample containers*. These steps are needed to resolve the question of likely radon loss indicated by certain past samples, especially where results reported from filtered samples substantially exceed those reported for otherwise identical unfiltered samples.

> a. For filtered samples, current procedure apparently requires the containers to be opened twice during the sample holding period – once to check pH and/or add HNO_3, and again for filtration – as compared to only once for unfiltered samples. *Each time a sample is opened, four types of data should be recorded*: 1) the date and time that the sample is opened, 2) whether the sample container appears to be tightly capped/sealed when it is opened, 3) either the estimated head space in milliliters, or the estimated distance in cm from lip of sample container to top of liquid, and 4) the length of time (in minutes or seconds) that the sample remains in an unsealed condition.
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> b. Various protocols could be set up as a check on radon loss from capped sample containers during the holding period. As an example (not prescriptive but illustrative of a method), each leachate sample may be collected and its container put immediately into a relatively gas-tight plastic pouch along with a readily available radon test kit, with each sample and its accompanying radon test kit remaining its own sealed pouch throughout the holding period except when opened for filtration, pH checking, or addition of HNO_3. Results from the radon test kits would provide approximate measures of radon loss from each capped sample container during the holding period.
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> c. As another example (not prescriptive but illustrative of an alternative method), duplicates of all leachate samples may be collected and sent to different analytical labs. For a given sample, any substantial difference between the results reported by the two labs for Lead-214, Bismuth-214, and/or Lead-210 would be indicative of radon loss from the capped sample container that showed the lower result.