Radium-226 in Creek Foam / Water

from the Portsmouth Gaseous Diffusion Plant

September 2005

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The RadioActivist Campaign

TRAC

P.R.E.S.S.

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Front Cover Photo: Foam / Water Sample: Coffee Filter After Immersion in Creek

Summary

Citizen activists collected foam and water filtrate flowing from the Portsmouth Gaseous Diffusion Plant, in November 2003. They identified beta radioactivity in the sample at least 100 times normal background levels by simple analysis with a Geiger counter and intervening aluminum foils. Authorities followed up, taking radiation measurements and collecting samples for analysis. A year later, they reported gross beta radioactivity in foam samples between 287 and 1,010 picocuries/liter (pCi/L) of liquid, which is more than 100 times normal beta levels in typical ambient water, confirming the citizens' discovery. The official identification of the beta emitter was: *not* technetium–99.

Several candidate sources of this elevated beta radioactivity have been suggested: naturally occurring uranium, uranium separated from its decay products, enriched uranium, depleted uranium, and naturally occurring potassium-40. The RadioActivist Campaign (TRAC) has reviewed the available information in order to identify the main source of radioactivity in the foam / water samples in order to provide a basis for future sampling and radiological analyses.

TRAC identifies the main source of radioactivity in the foam / water to be radium-226. This identification is based on the citizen activists' Geiger counter measurements in November 2003 and on a later gamma spectrometer measurement. There are several possible explanations for the presence of radium-226, much above normal background activity, in water flowing out of the Portsmouth Gaseous Diffusion Plant.

This identification of radium-226 as the main source of radioactivity in the creek foam / water increases the level of concern for public health tenfold. The enforceable Maximum Contaminant Level of radium-226 in drinking water is 5 pCi/L. This limit is probably exceeded in creek water flowing from the plant.

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Chronological Introduction

In November 2003, Sergey Pashenko of the non-profit organization, Siberian Scientists for Global Responsibility (SSGR), demonstrated and taught simple methods that had been developed in the 1960s by the USSR Ministry for Medium Machine-Building that allow citizen-activists to acquire their own information about radiological conditions in their environment. The demonstration employed an "Inspector" Geiger counter, produced by International Medcom, Sebastopol, California, Fig. 1.

Fig. 1. Front (readout) and back (window) views of Inspector [International Medcom].

Pashenko and Vina Colley of the Portsmouth/Piketon Residents for Environmental Safety and Security (PRESS) measured radioactivity around the Portsmouth Gaseous Diffusion Plant (GDP) in Ohio. The work was part of an exchange program facilitated by ISAR: Resources for Environmental Activists. The use of the Inspector at Portsmouth GDP is described in "A Citizen's Guide to Monitor Radioactivity around the Energy Department's Nuclear Facilities [ISAR, Washington DC, www.isar.org (May 2005)]."

Several hundred meters from the GDP building, Colley noted an accumulation of white foam flowing from beneath the plant fenceline on Big Run creek. The flow of Big Run, at the sample location, was estimated visually at 10 liters per second. Big Run flows into the Scioto River which flows into the Ohio River.

Pashenko chose to sample foam, instead of water, because it would have taken him two days to evaporate a sufficient water sample ["DexV. Description of expeditions in the vicinity of the Portsmouth Gaseous Diffusion Plant, November 2003," DEBvar21-Eng.doc, ISAR, www.isar.org (April 2005)]. Pashenko skimmed about 1.5 liters of foam / water from Big Run into a plastic container. The foam / water was evaporated to a dry residue and placed into a jig for counting under the window of the Geiger-Muller detector in the Inspector. (See disk in right photo in Fig. 1.) The number of counts per 10 minutes was recorded.

Pashenko removed the Inspector from the jig, placed one Russian aluminum foil over the sample, and replaced the Inspector. The number of counts per 10 minutes was recorded. This counting procedure was repeated, with several more aluminum foils added between the sample and the Inspector; see Fig. 2.

Fig. 2. Placing a number of aluminum foils over the dried sample, before counting [modified from SSGR].

After Pashenko completed counting with 8 intervening foils, he began to prepare for a lecture, for which he then interrupted the counting. The next day, Pashenko resumed counting. The counting results are the blue squares in Fig. 3.

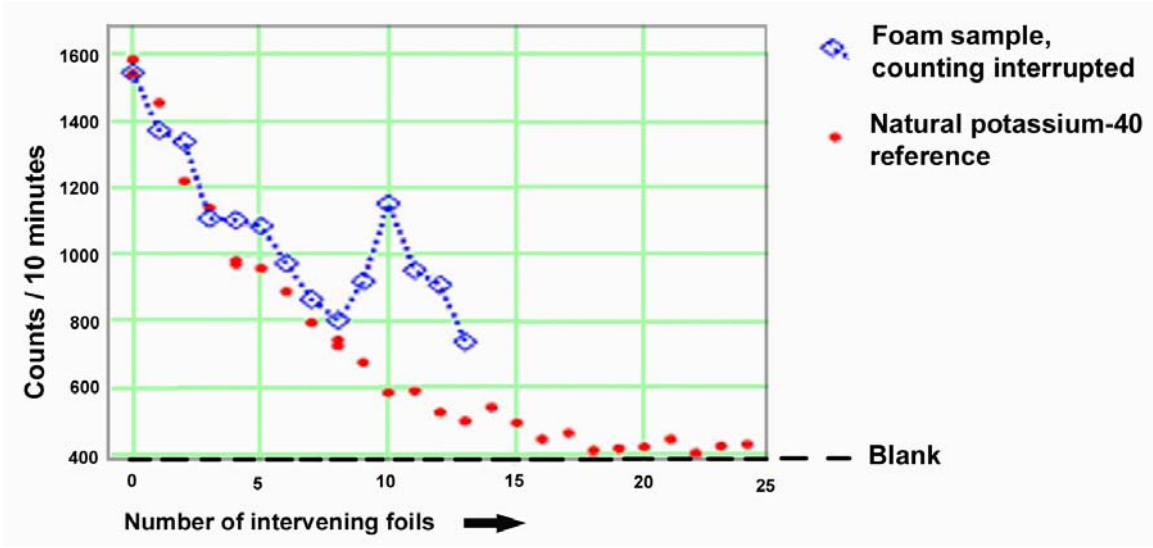

Fig. 3. Foam sample counts (blue) with intervening foils, in comparison to counts (red) with a calibrated reference material containing primarily natural potassium-40 [modified from SSGR]. "Blank" is the number of counts with no sample in the jig.

The spike at 10 foils in Fig. 3 is explained as follows: With the sample left under aluminum foils in the jig, radon gas produced by the continuing decay of radium-226 was partly blocked from dispersing out of the enclosure. Radioactive products of radon decay began to accumulate within the jig, adding to the measured radioactivity. Pashenko described this result as follows:

> This result was the first indication for me in [the] expedition around Portsmouth that we have emanation (release) of radon (daughter products) [if] the sample is closed [a] long time [e-mail, 26 March 2005, brackets added]."

Prompt products of radon decay include radioactive lead-214 and bismuth-214. Both are beta particle emitters.

On November 18, 2003, Pashenko held an urgent press conference in which he announced beta radiation in the foam sample "at least 100 times higher than normal background levels."

After the press conference, Pashenko and Colley proceeded to a second location, below a drainage pipe, where foam was also accumulating and water was discharging into the Scioto River and hence into the Ohio River. A water filter was fabricated from an American coffee filter and immersed in the flow for almost 24 hours, Fig. 4.

Fig. 4. Coffee filter discolored after filtering creek water for 24-hours [SSGR]. This discolored filter is the original sample for the "Coffee-filtered water sample," in Fig. 5.

Pashenko burned the coffee filter and subjected the ashes to analysis with the Inspector, in the jig, adding aluminum foils between 10-minute counts. The results appear in Fig. 5.

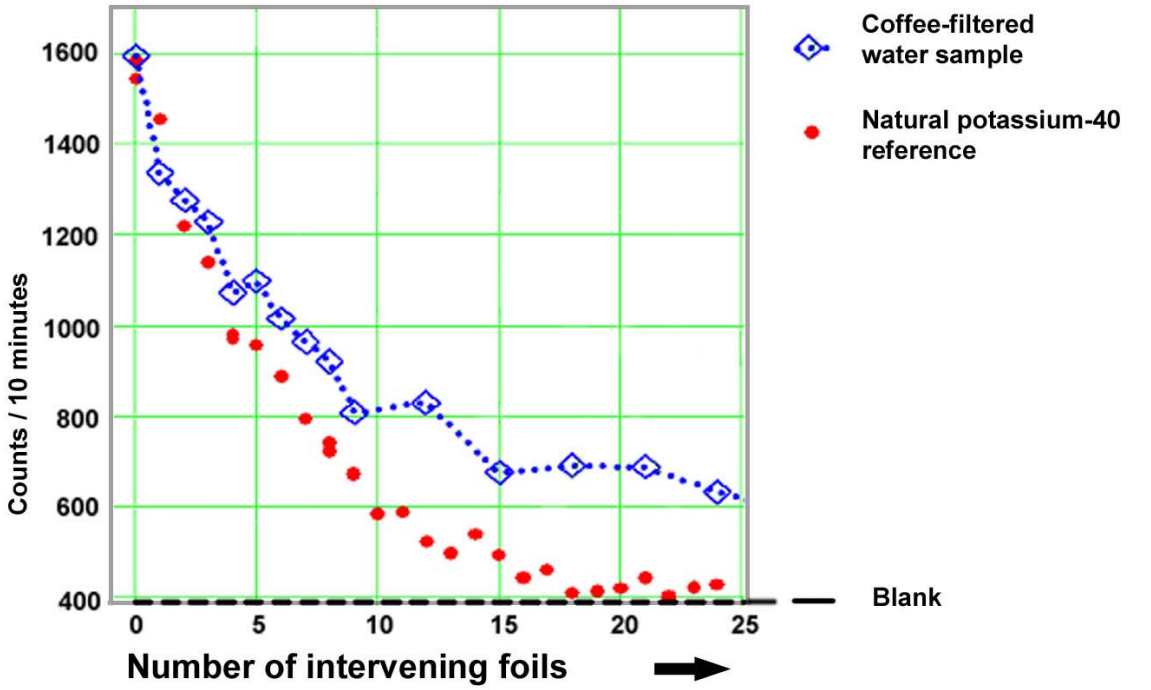

Fig. 5. Coffee-filtered water sample counts (blue) with intervening foils, in comparison to counts (red) with a calibrated reference material containing primarily natural potassium-40. "Blank" is the number of counts with no sample in the jig [modified from SSGR].

Pashenko attributed the relatively high count rates, acquired with large numbers of aluminum foils intervening, to beta radioactivity with energy exceeding 2,000 keV [S.E. Pashenko, "DexV. Description of expeditions in the vicinity of the Portsmouth Gaseous Diffusion Plant," November 2003, DEBvar21-Eng.doc, ISAR, www.isar.org (April 2005)].

Based on his years of experience with uranium in the environment in Novosibirsk, Siberia, Pashenko provided a generic identification as "uranium [S.E. Pashenko, 'English_Content bucleta var 11,' ISAR, www.isar.org (March 2005) Fig 4-2]."

There are two beta emitters with energies exceeding 2,000 keV in the uranium (U) decay chains:

U-238 decay: protactinium-234 (Pa-234): 2,200 keV
U-238 decay: bismuth-214 (Bi-214): 3,270 keV

The latter has already been mentioned in regard to the results of Fig. 3.

Meanwhile, the GDP operator, the United States Enrichment Corporation (USEC) responded to Pashenko's urgent press conference by sending environmental and health physics professionals to measure radioactivity and to collect samples on November 19, 2003 [Attachment 2]. At a public meeting on December 2, 2003, DOE assured the public that the results did not indicate the presence of radionuclides above normal background levels. USEC provided the analytical results to the Ohio Environmental Protection Agency on December 10, 2003.

After completing his study at Portsmouth, Pashenko took the Coffee-filtered water sample, Fig. 5, back to Novosibirsk and re-analyzed it with a FieldSPEC, NaI scintillation spectrometer, manufactured by Target Systemelectronic GmbH., Solingen, Germany. This gamma spectrometer was calibrated and stabilized with a cesium-137 calibrator. Pashenko counted the Coffee-filtered water sample and a Blank for 23 hours. Then he subtracted the Blank energy spectrum from the Coffee-filtered water sample spectrum. The *difference spectrum* is Fig. 6.

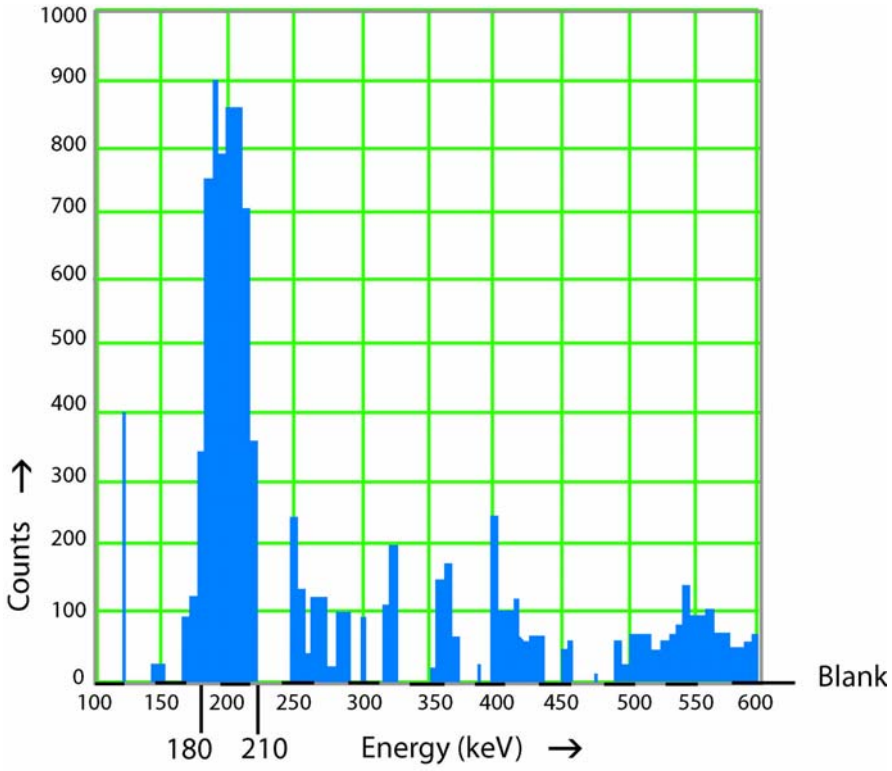

Fig. 6. Coffee-filtered water sample energy spectrum (100 - 600 keV), with Blank energy spectrum subtracted [modified from S.E. Pashenko, A.E. Osochenko, et al, "The Use of Filters for Sampling Radioactive Aerosols and Hydrosols from the Atmosphere and Water for Monitoring Studies by Independent Environmentalists Near Nuclear Sites," Lecture 1 filters_English.doc, ISAR, www.isar.org (March 2005) final figure].

Pashenko noted that the gamma energy of the sample (with Blank subtracted) was primarily in the energy range of 180 to 210 keV (highlighted in red in Pashenko's original). He concluded this "may well correspond to U-235 content in water in the pipe." Uranium-235 (U-235) has three main gamma energy emissions:

163 keV, with 4.7% intensity
186 keV, with 53.5% intensity
205 keV, with 4.7% intensity

The issues raised by Pashenko's foam / water sampling and DOE/USEC's follow-up sampling remained unsettled over the next year. Then DOE published some of USEC's foam analysis results on February 24, 2005; see Attachment 2. The gross beta results for two foam samples collected by USEC were: 287 and 1,010 picocuries/liter (pCi/L). (DOE expressed those results per milliliter.) DOE did not detect technetium-99 in the samples.

The results of the foam / water analyses that had been reported by the time TRAC became actively involved in this matter, in March 2005, are summarized in the following section.

Summary of Prior Results

Prior results refers to relevant data available before March 2005 and specifically to the foam / water measurements described in the last section. Those prior results are summarized as bulleted *features*, as follows:

SSGR foam analysis, interrupted:

- foam contains radon emanation
- with decay products having many times normal background beta activity
- not potassium-40.

SSGR Coffee-filtered water analysis:

- "uranium"
- with beta activity >2,000 keV.

SSGR gamma spectrum of Coffee-filtered water sample:

- gamma energy primarily in range of 180 - 210 keV

DOE/USEC gross beta analysis of two foam samples:

- relatively low survey (gamma) activity
- with gross beta: 287 and 1,010 pCi/L
- but beta emitter is not technetium-99.

In the remainder of this report, other information is included as *prior results*, as follows: the USEC data in Attachment 1, the DOE report of those data in Attachment 2, and generic and monitoring information cited in Attachment 3 of this report.

Identity of Main Radioactivity in the Foam / Water

This section is the crux of the report. The purpose is to identify the radionuclide(s) contributing most beta radioactivity to the foam / water samples collected by Pashenko and USEC in 2003. The general method of this section is to begin with a reasonable set of candidate radionuclides and to compare their radiological features with the features listed in the Summary of Prior Results, in the last section. The plan is to eliminate unlikely candidates on the basis of few *matches* between features of their radiology and features of the prior results.

The plan of this section is then to scrutinize more closely the candidate radionuclide(s) that have features that best correspond to the prior results. This is intended to help the reader appraise what confidence to place on the identification of the main beta radioactivity in the foam / water samples.

Following that general identification of candidate(s), other alternative radionuclides that were not included in the beginning set of reasonable candidates will be examined. This will help extend the appraisal of confidence in the identified candidate(s).

Finally, a few radiological implications of the identified radionuclide(s) will be mentioned to help concerned parties decide on any follow-up measures that might be appropriate.

The author has simplified the radiological considerations as much as seemed feasible. However, comparison of alternative radionuclides as the main source of beta activity in the foam / water samples is inherently technical. This identification seems important enough to warrant attention to these technical matters.

The members of the natural uranium decay chains comprise the beginning set of radionuclides to be examined in this section. This set accords with the Pashenko's expert opinion that the SSGR Coffee-filtered water analysis showed "uranium."

For this presentation, the uranium decay chains are simplified by splitting the uranium–238 (U–238) decay series into two parts: U–238 to U–234, here called the "U–238 decay chain." Then this chain is taken up anew with U–234 and continued through lead–210 (Pb–210), here called the "U–234 decay chain."

According to this simplification, there are three natural "uranium" decay chains. These three radioactive decay chains are summarized in Fig. 7, on the next page. Notice that the U-238 decay chain of Fig. 7(a) leads into the U-234 decay chain of Fig. 7(b). Notes for Fig. 7 appear on the page after the figure.

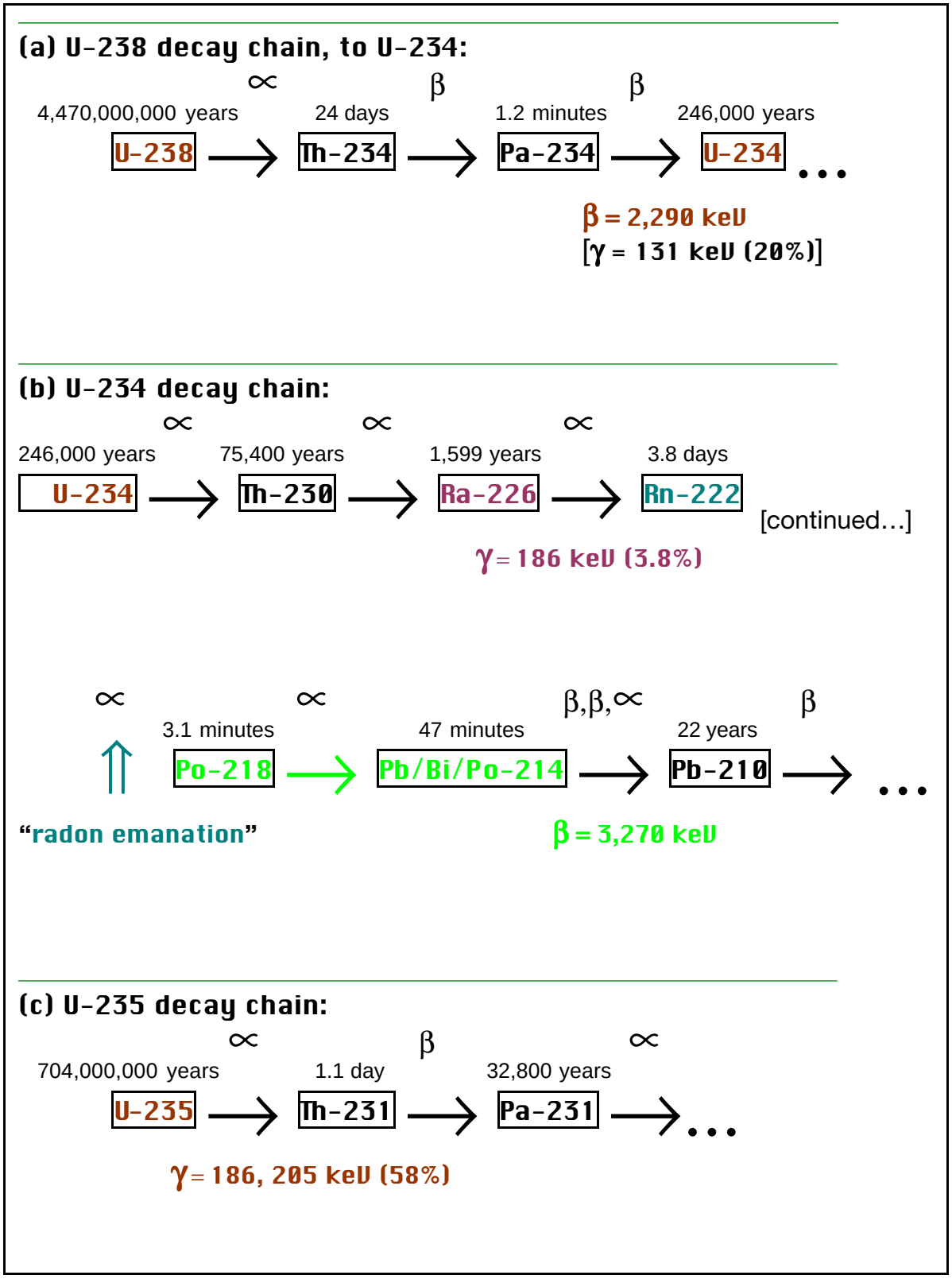

Fig. 7. Alternative "uranium" explanations

Notes for Fig. 7 (on the previous page).

- The elements are symbolized, in Fig. 7, as follows:

Bi	bismuth	Pa	protactinium	Pb	lead	Po	polonium
Ra	radium	Rn	radon	Th	thorium	U	uranium

- "α" or "β" over an arrow indicates alpha or beta radiation.
- Radioactive decay half-life is listed above each isotope in Fig. 7.
- "γ" under an arrow indicates the associated gamma energy in keV. Relevant beta energies are listed below the isotopes, in keV.
- The products of radioactive decay that match prior results are listed below each decay in Fig. 7.
- One gamma (γ) that is not a match to the prior results bulleted in the last section is also shown [in brackets] below Pa–234, for special consideration later in this section.

The plan of this section begins with comparison of the features of the three natural uranium decay chains, summarized in Fig. 7, against the bulleted items in the Summary of Prior Results. Non-uranium possibilities are generally eliminated as likely candidates by comparison to the SSGR Coffee-filtered water analysis result: "uranium." Natural potassium–40 is further eliminated as a candidate by the SSGR foam analysis, interrupted result: "not potassium–40." Likewise, technetium–99 is further eliminated by the DOE/USEC gross beta analysis of two foam samples result: "beta emitter is not technetium–99."

The idea now is to compare how well the radiological features of each of the three natural uranium decay chains *matches* the prior results. For present purposes features and bulleted prior results are considered to *match* if they accord, and the number of these matches is counted. The number of such matches is listed, and the particular matches are identified, as follows:

- **U-238 decay chain** has two features that match the prior results: "uranium" and beta radiation exceeding 2,000 keV.
- **U-234 decay chain** has 5 features that match all prior results: "uranium", radon emanation, gamma energy in the range 180-210 keV, relatively low gamma intensity (3.8% at 186 keV) coupled with intense beta activity (200%), and beta energy exceeding 2,000 keV.
- **U-235 decay chain** has two features that match the prior results: "uranium" and gamma energy in the range 180-210 keV. (The energy of the single beta decay is only 305 keV.)

TRAC eliminates U–238, U–235, and their decay chains as candidates for the main source of the radioactivity measured in the foam / water samples by Pashenko and USEC in 2003, on the basis of only two matches, each, in comparison to 5 matches for the U-234 decay chain.

The elimination of uranium–238 as a candidate for the main source of radioactivity in the foam / water samples is specially important for the plan of this section. Many *scenarios* for (-or causes of-) the main source of radioactivity inherently include U–238.

- **Naturally occurring uranium** with its decay chains more or less in secular equilibrium.
- **Separated uranium** with its decay products more or less eliminated.
- **Enriched uranium**.
- **Depleted uranium**.

Each of these four scenarios requires substantial U–238 content. Hence, the elimination of U–238 as a candidate for the main source of radioactivity eliminates each of these four scenarios as a likely explanation. (The absence of the 20% intensity gamma radiation of Pa–234 at 131 keV in Fig. 6 also weighs against abundance of U–238 in the samples. Note that the energy in that region is lower and too narrow for a true peak.)

The focus of the remainder of this section turns both to candidate sources of the observed radioactivity *within the U–234 decay chain* and to other alternative candidate sources that are unrelated to uranium and that have not yet been evaluated.

Consideration of the U–234 decay chain begins with attention to U–234 *itself*, as a candidate for the main source of elevated radioactivity, in the U–234 decay chain. Inasmuch as both U–238 and U–235 have been eliminated as candidates, this consideration turns to the question: Under what circumstances might substantial U–234 occur in environmental samples without much either U–238 or U–235 accompaniment? If U–234 is the main source of elevated radioactivity in foam / water samples, each of the four scenarios, displayed above, would also contain substantial U–238 and/or U–235. Naturally occurring and separated uranium would exhibit as much U–238 activity as U–234 activity. Enriched uranium would exhibit elevated U–235 along with elevated U–234. Depleted uranium has diminished U–234 as well as diminished U–235 in comparison to U–238 and so does not provide a scenario for U–234 dominating environmental samples.

The second member of the U–234 decay chain, thorium–230, matches all the prior results. Naturally occurring Th–230 is not reported separately from its parent, U–234, in the environment [M. Eisenbud and T. Gesell, “Environmental Radioactivity From Natural, Industrial, and Military Sources,” (4th ed) Academic Press, San Diego, California

(1997) p. 563]. The elimination of U–234, itself, as a main source of the radioactivity in the foam / water thus eliminates *natural* Th–234 as a likely candidate also. The Handbook of Chemistry and Physics and the Encyclopedia of the Chemical Elements list no commercial, industrial, or military uses for Th-230. Thus, the GDP is probably not using substantial Th–230. On this basis, Th-230 is eliminated as a likely candidate source of the elevated radioactivity in the foam / water.

The third member of the U–234 decay chain, Ra–226, matches all five prior results, exactly as Th–230 does. The distinction of Ra–226 is its history of special commercial, industrial, and military uses (discussed in the next section of this report). Therefore, Ra-226 passes screening as a candidate of the main source of elevated beta radioactivity in the foam / water samples collected in November 2003. The Ra–226 candidate is sketched in Fig. 8.

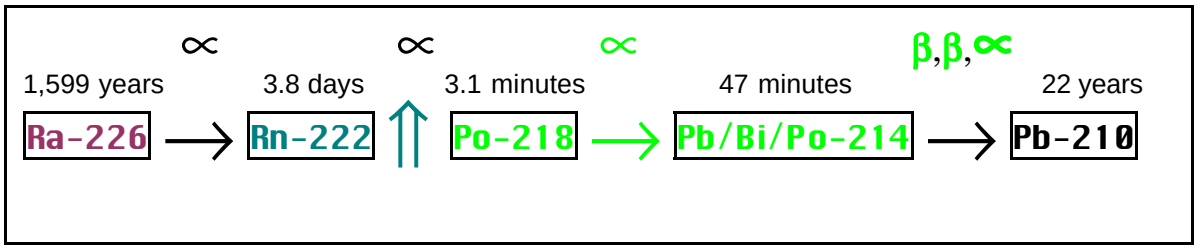

Fig. 8. Summary decay chain of radium-226.

Continuing down the U–234 decay chain, in Fig. 7(b), Rn–222 is itself the "radon emanation" that Pashenko identified (accumulating from decay of parent Ra–226) in Fig. 3. The short, 3.8-day, half-life of Rn–222 eliminates it as a candidate source of the radioactivity in the foam / water. If Rn–222 were the source, it would have decayed out of the sample after a few days. Furthermore, Rn–222, as a source, does not account for the 180-210 keV gamma emission in Fig. 6.

Each radionuclide even farther down the U–234 decay chain shares the mis-matches with the Summary of Prior Results that Rn–222 has. Furthermore, each radionuclide farther down the decay chain lacks the match of "radon emanation."

This completes the elimination of all members of the uranium decay chains, except for Ra–226, as the main source of excessive beta radioactivity in the foam / water samples.

TRAC has eliminated non-uranium radionuclides as candidate sources of the measured radioactivity, as follows:

(1) Any strictly non-"uranium" source would not match Pashenko's expert opinion that the source was "uranium".

(2) TRAC has searched radioactive decay libraries for major gamma emissions in the range of 180-210 keV, shown in Fig. 6. Only two candidates were identified: U-235 itself and Ra-226 in the U-234 decay chain.

(3) TRAC has searched the Chart of the Nuclides for other radionuclide candidate sources / scenarios, including decay chains, that have scenarios more or less consistent with the prior results [E.M. Baum, H.D. Know, et al (rev.), "Nuclides and Isotopes," (16th ed), Lockheed Martin Distribution Services, nuclides.chart@lmco.com (2002)]. This search turned up actinium–227 (Ac-227) part way down the U–235 decay chain and Th–229 in the artificial U–233 decay chain. These decay scenarios involved radioactive in-growth of short-lived radium isotopes as the "radon emanations" that Pashenko reported. TRAC eliminated these scenarios as inconsistent with the prior results.

Based on the elimination of alternatives, the main source of radioactivity in the foam / water samples is identified as radium–226.

Relevant considerations regarding this identification of Ra–226 as the source of elevated radioactivity follow:

The hazard associated with Ra–226 in waters is indicated by the enforceable Maximum Contaminant Level (MCL) allowed by the Environmental Protection Agency in primary drinking water. The MCL for Ra–226 is **5 pCi/L**. **T**he health-based target Maximum Contaminant Level Goal (MCLG) for Ra–226 in drinking water is **zero pCi/L** ["National Primary Drinking Water Regulations; Final Rule," 40 CFR Parts 9, 141, and 142, Federal Register (Vol. 65, No. 236) (December 7, 2000) p. 76722].

This enforceable limit of Ra–226 $\leq$ 5 pCi/L in drinking water compares to the screening level of **50 pCi/L** for gross beta radioactivity in ambient waters.

The public health concern for a given amount of radioactivity in drinking water varies as the reciprocal of regulatory limits: 50 pCi/L for gross beta $\div$ 5 pCi/L for Ra–226. That is to say, the hazard attending Ra–226 in drinking water is about ten times the hazard attending gross beta radioactivity.

The identification of Ra-226 as the main contaminant source in the creek foam / water samples collected in 2003 raises the level of public concern for health by a factor of ten.

Environmental monitoring around Portsmouth is insensitive to this concern for Ra–226. Neither Ra–226 nor gross beta are measured or reported in surface waters around the GDP in DOE's annual 2003 environmental data report for Portsmouth. The state of Ohio EPA last measured gross beta activities in surface waters collected from 6 locations in Little Beaver Creek in 1997, which is a different drainage, but they did not measure Ra–226 anywhere.

Ohio EPA project coordinator, M. Galanti, who oversees cleanup of the Portsmouth GDP site for the state, had accompanied USEC during the sampling on November 19, 2003 [M. Lafferty, "State Hopes to Unravel Radiation Readings near Piketon," Columbus Dispatch (July 30, 2005) P. B4]. Ohio EPA received both the USEC results in the Memorandum dated December 1, 2003 [Attachment 1] and additional DOE/USEC information dated February 24, 2005 [Attachment 2].

In April, 2005, Ohio EPA drafted plans to collect surface water samples from 17 locations around the GDP, between August 4 and September 4, 2005, as part of its regular monitoring program, ["USEC Piketon Facility Draft DSW Workplan," Ohio EPA, http://www.epa.state.oh.us/dsw/documents/lbeac97.pdf (April 29, 2005, revised July 1, 2005)]. Ohio EPA will analyze those samples for gross beta radioactivity.

On August 4, 2005, TRAC requested that Ohio EPA add Ra–226 to the list of analytes in its sampling plan for USEC Piketon. TRAC also suggested that foam samples be collected and analyzed from three locations, as new references. Ohio EPA declined, because the sampling plan could not be modified because it had

> **been approved and is not subject to revision. We can not sample for foam since there are no approved EPA methods to do so. We could not verify the data. We need to take over a liter of water to sample for radionuclides. It would be impossible to determine the amount of foam needed to meet the standards for water samples** [M. Galanti, e-mail, Ohio EPA (August 4, 2005)].

—This statement suggests that Ra–226 flowing out of the plant is not yet on th*e regulatory radar screen*.

As a matter of procedure, it would be quite easy to collect and weigh a sample of foam. That foam could then be reduced to liquid by adding a measure of anti-surfactant and distilled water to increase the total volume to one liter. That liquid could be analyzed for gross beta according to the EPA method for water. The analytical result could then be back calculated for the foam on the basis of its original weight.

Explanations of Ra-226 in Foam / Water from the GDP

This section explores alternative explanations for the Ra-226 in creek foam / water flowing out of the Portsmouth GDP, far above normal background levels.

The last section concluded by identifying Ra–226 as the main contaminant in creek foam / water samples collected in November 2003. Although Ra–226 is a member of the U–234 decay chain, U–234 and other uranium isotopes were eliminated as candidates for the main source of radioactivity in foam / water.

Now the approach is to estimate the fraction of gross beta radioactivity in the foam / water samples that ***is*** attributable to naturally present radionuclides in the uranium and thorium decay chains and potassium–40. The residual radioactivity is then attributed to Ra–226.

Many explanations for the occurrence of radium in the environment involve its position in the natural U–234 radioactive decay chain, with decay products more or less in secular equilibrium with U–234 and U–238. Those explanations are ruled out, for the present case, by the evidence presented in the last section. That evidence weighed against uranium isotopes themselves being the main source of the radioactivity reported in foam / water samples from Portsmouth. Yet, there are natural and artificial processes that leach soluble radium decay products from naturally rather insoluble uranium and so result in more radium than uranium in groundwater or surface water, even though the actual source material in the ground is uranium.

Fortunately, USEC provides some analytical results for uranium in their foam / water samples, in addition to gross beta results [Attachment 1]. This allows a conservative estimation of the "excess beta" radioactivity that is not attributable to natural uranium in the foam / water samples. The gross beta and uranium results for the DOE/USEC samples of creek foam / water appear in Table 1 on the next page.

Units of measure in the original data have been converted to pCi/L for consistent comparisons in this table. The symbol "β" is used to emphasize a particular numerical value of "beta radioactivity." Wherever the connotation is less numerical in the text, "beta" is written instead of the symbol.

Table 1. Radioactivities in USEC foam / water samples of November 2003.

Sample	Gross β [pCi/L]	U [pCi/L]	Gross β / U [no units]
Foam:	287.	11.182	25.7
Foam:	1,010.	38.073	26.5
Water:	<50.*	0.375	<140.
Water:	<50.*	0.789	<140.

* Listed as 0.0 pCi/mL in the original [Attachment 1]. On August 8, 2005, TRAC requested from USEC the full-precision, gross beta results for the water samples. No reply was received from DOE/USEC by the publication date of this report. According to the usual rule of rounding: Gross β ≥ 0.050 pCi/mL rounds up to 0.1 pCi/mL; and Gross β ≤ 0.049 pCi/mL rounds down to 0.0 pCi/mL. Therefore, a listing of Gross β = 0.0 pCi/mL implies that Gross β < 0.05 pCi/mL, which is Gross β <50 pCi/L. Note: The screening limit for gross beta radioactivity in ambient water is 50 pCi/L.

Conversion factors from original data, for this tabulation:

Volume:	1000. mL/L
Liquid density:	1. kg/L
Natural Uranium radioactivity:	0.686 pCi/µg

Gross beta radioactivity is listed as "Gross β" in the first numerical column of Table 1. Uranium radioactivity is listed as "U" in the second numerical column. The dimensionless ratios, "Gross β / U" in the right-hand column are the values in the first numerical column divided by the values in the second numerical column.

The U values measured by USEC in foam / water samples in 2003, Table 1, are now used to calculate the amount of naturally present beta activity, as background in these foam / water samples, in accord with the radiological context that is developed in Attachment 3.

Background beta radioactivity attributable to the uranium and thorium decay chains is estimated at 6.6 times the activity of uranium in a foam / water sample. (See Attachment 3, Table A2 and below it: Net β / U = 6.6.) Background K–40 activity is estimated at 3.9 pCi/L. (See end of Attachment 3.) Thus, naturally present "Background Gross β" radioactivity in the foam / water samples of Table 1 is estimated as 6.6 times the uranium activity in Table 1, in addition to 3.9 pCi/L for natural background potassium radioactivity.

The uranium activities in Table 1 that USEC measured are repeated in the first numerical column of Table 2, on the next page. Those "U" values are multiplied by 6.6 in the second numerical column. 3.9 pCi/L of potassium ("K") is added to each of those values to yield "Background Gross β" in the fourth numerical column of Table 2. "Gross β" in the USEC samples is copied from Table 1 to the fifth numerical column of Table 2.

Table 2. Excess β calculated in USEC foam / water samples [pCi/L liquid]

Sample	U	6.6 U	K	Background Gross β	Gross β	**Excess β**
Foam:	11.182	73.8	3.9	77.7	287.	**209.**
Foam:	38.073	251.3	3.9	255.2	1,010.	**755.**
Water:	0.375	2.5	3.9	6.6	<50.	**<43.4**
Water:	0.789	5.2	3.9	9.1	<50.	**<40.9**

Notes: "<" means "less than."
Background Gross β = 6.6 U + K.
Excess β = Gross β – Background Gross β

"Excess β" here refers to the beta radioactivity in a sample that is not attributable to naturally present "Background Gross β." This beta activity in excess of background is calculated as the value of "Gross β" in fifth numerical column of Table 2 minus the value of "Background Gross β" in the fourth numerical column. Excess β is the beta radioactivity in a sample that exceeds what is contextually accounted in Attachment 3 as naturally present, from the uranium and thorium decay chains and potassium.

"Excess β" radioactivity in Table 2 is attributable to the plant.

Gross beta radioactivity in Table 2 is 73% "Excess β" in the first foam sample and 75% "Excess β" in the second foam sample. Gross β in the two water samples is less than 87% "Excess β."

About three-quarters of the beta radioactivity in the foam samples is unaccountable as naturally present *background*. According to the conclusion in the last section, this above-background beta radioactivity results from the beta decays in the Ra–226 decay chain in Fig. 8.

Before exploring alternative processes that might account for the excess beta radioactivity from the plant, an initial estimate of the *actual* excess beta activity (listed merely as "<50 pCi/L") in the water samples of Table 1 is required. This initial estimate is obtained by extrapolation. This extrapolation serves in lieu precise values of gross beta in the DOE/USEC water samples. (DOE/USEC has not yet responded to TRAC's request on August 8, 2005, for information.) The USEC data in Attachment 1 only allow the interpretation that gross beta in the two water samples was <50 pCi/L, as in Table 1 and repeated in Table 2.

For this extrapolation, assume that excess beta activity in the water samples is proportional to excess beta in the foam samples, and that both those excesses are proportional to their uranium contents. (The latter assumption is of constant ratios of

uranium and thorium; see discussion at Table A1 in Attachment 3.) This can be written as an equation, with each term as an activity with units of pCi/L, and then values can be plugged in from Table 2, as follows:

(excess beta in water) = (excess beta in foam) x (uranium in water) ÷ (uranium in foam)

(excess beta in water) = (209) x (0.375) ÷ (11.182) = 7.0 pCi/L
(excess beta in water) = (755) x (0.789) ÷ (38.073) = 15.6 pCi/L

The abbreviated radium-226 decay chain in Fig. 8 shows that radioactive decay of one Ra–226 nucleus is followed within a few days by two beta (β) emissions. That is, two above-background beta emissions result from one Ra–226 decay (neglecting loss of Rn–222). Thus, the above-background excess Ra–226 radioactivity in the USEC water samples is about half the excess beta radioactivity. Table 3 gives the just-displayed extrapolations, divided by two:

Table 3. Extrapolated excess Ra-226 in USEC water samples [pCi/L]

extrapolated excess Ra–226 in water:	7.0 / 2	=	3.5
extrapolated excess Ra–226 in water:	15.6 / 2	=	7.8

These extrapolations of excess Ra–226 in the USEC water sample data compare to the U.S. EPA enforceable limit for drinking water of 5 pCi/L, as follows: The first is less than the limit, the second exceeds the limit by half. Note that the U.S. EPA enforceable limit applies to total Ra–226, which includes both excess and naturally present Ra–226.

These extrapolations identify Ra–226 as a radionuclide of public concern for health in surface waters that might feed drinking waters, around the plant. The actual magnitude of that concern can be determined after Ra–226 has been measured specifically in creek water samples.

The estimate, above, that about three-fourths of beta radioactivity in the foam samples is above background differs markedly from Pashenko's on-the-spot conclusion in November 2003, that the beta activity in the foam / water samples was more than 100 times background. In either case, there is cause for serious public concern, but the difference in these estimates invites more scrutiny here.

The difference between these estimates is in Ohio EPA's measurement of mean gross beta radioactivity in Little Beaver Creek water in 1997 at 16 pCi/L [Table A2 in

Attachment 3]. That value is relatively high in comparison to gross beta radioactivity of about 2 pCi/L in Clinch River water at the DOE Oak Ridge, Tennessee site and about 1 pCi/L in Columbia River water at the DOE Hanford, Washington site. The conservative explanation is that there is a high background beta activity in surface waters in the Portsmouth area, attributable to naturally high uranium and potassium content. That conservative explanation was an assumption in Attachment 3 and hence in Table 2.

That conservative explanation attributes a factor of up to 20 to unusually high background radioactivity in water at Portsmouth. Which is to suggest, that beta radioactivity in surface water around Portsmouth contains an abnormally large amount of naturally present beta radioactivity. That explanation thus accounts for a factor of up to 20 of Pashenko's conclusion that beta radioactivity is elevated "at least 100 times ***normal*** background levels [emphasis added]." Yet, even **such naturally high background beta activity is overwhelmed by radium-226 that is not accountable as high background**.

That easy reconciliation between Pashenko's on-the-spot conclusion and the estimates in radiological context in Table 2 has an important consequence. That consequence derives from the conservativeness of the assumption that the beta radioactivities measured in surface waters near the plant in 1997 were naturally occurring and not attributable to the plant itself. That consequence implies that excess beta radioactivity in foam / water around the plant, measured by Pashenko and USEC in 2003, began after 1997:

Excess radioactivity in foam / water from the plant is recent.

This implication heightens public concerns, because Ohio EPA's routine monitoring program is unresponsive to new kinds of radioactivity, such as Ra–226, that have not been included in their historic cleanup investigations for Portsmouth: "We are looking for contaminants most likely to be associated with the production processes at the plant **based on our ongoing cleanup investigation** [emphasis added, M. Galanti, e–mail, Ohio EPA (August 4, 2005)]."

(The alternative of assuming less conservatively that some of the beta radioactivities measured in surface waters near the plant in 1997 were of plant origin, rather than naturally occurring, would lead to higher estimates of "Excess β" in Table 2 and higher extrapolations of excess Ra–226 in Table 3. That is to say, by using *conservative* assumptions, the pollution is probably worse than described, in one way or another.)

Consideration now turns to the probable cause of the (extrapolated) Ra–226 in foam / water running out of the plant.

Historically, radium-226 had several uses, including luminous paints and the contents of "needles" implanted surgically into a human organ to be treated by irradiation. In most commercial applications, radium has been supplanted by less hazardous materials.

Thus, most explanations for elevated Ra-226 activities involve archaic accumulations. One such possible explanation of the Ra-226 in creek foam / water flowing out of the Portsmouth GDP is storage of radium left over from the Manhattan Project era. The GDP might be a storage or disposal site, like the Niagara Falls Storage Site (NFSS) in Niagara County, New York, which holds half the world's stock of Ra–226 in residues from uranium extraction from high-grade ores [A. Witryol, e-mail (October 2004); and Commission on Geosciences, Environment and Resources (CGER), "Safety of the High-Level Uranium Ore Residues at the Niagara Falls Storage Site, Lewiston, New York," http://books.nap.edu/ (1995)]. However, unreported residues at Portsmouth, from the old Manhattan Project era, does not provide a pleasing explanation of the sudden emergence of overwhelming Ra–226 from the GDP, seemingly only within the last few years. Such a sudden emergence after many years of non-detection would foreshadow staggering contamination within another few years.

Other explanations involve the presence of old Ra–226 materials that might have been shipped to the plant for processing, storage, or disposal within the past decade or so. The record shows that shipments within the U.S. nuclear complex were not too thoroughly documented through the 1950s [J.H. Robillard, "KAPL Radioactive Waste Information Related to the Lake Ontario Ordnance Works," REC&SD:DAD05-17, letter to S. Gavitt, Schenectady Naval Reactors Office, Department of Energy, Schenectady, NY (August 10, 2005) 28 pp. with 8 attachments]. Maybe the documentation has been lost for a tenth of the old shipments. Materials having little interest, such as "Misc. scrap" in wooden boxes was poorly documented. For example a Bill of Materials, dated June 26, 1958 for such a shipment in Box Car ATMX 209, lists only the number (30) of wooden boxes, the estimated weight of the load (22,500 pounds), and the external radiation outside the box car (10 mr/hour at 12 feet from the sides and 5 feet from the ends). In cases of more interest, more detail was provided. For example, the first item on a Bill of Materials, dated February 14, 1958 for Box Car SAL 10063 lists four pallets of capsules containing uranium residue with a maximum dose rate of 5 mr/hour. The maximum weight of each of these pallets is listed as 1000 pounds. The contents are listed as "Cesium and strontium gaps capsulated in 65 gal. c/s [carbon steel] drums."

Record-keeping routines for shipments between nuclear sites has improved over the years as rules and regulations have tightened. From the examples mentioned in the last paragraph, it seems highly unlikely that massive amounts of Ra-226 laden materials might *ever* have been shipped from off-site to the plant without hundreds or thousands of Bills of Material still existing at either the point of origin or at the plant.

Attention now turns to the prospect of some recent or current, on-site source of Ra-226 contamination that might not yet have been published. Several commercial / industrial / military sources and applications of Ra–226 have been reported [M. Eisenbud and T. Gesell, “Environmental Radioactivity From Natural, Industrial, and Military Sources,” (4th ed) Academic Press, San Diego, California (1997)]. One such process involves mixing radium decay products with beryllium to provide intense sources of neutrons [D.R. Lide (ed.), “CRC Handbook of Chemistry and Physics,” 84th ed., CRC Press, Boca Raton, Florida (2003) p. 4-25]. This possibility is explored here in a little more detail.

According to this latter possibility, the noble gas, Rn–222, is easily extracted from a bed of finely divided radium. The products of the decay of the extracted radon gas are Groups 14, 15, and 16 elements. These can be extracted from the inert radon gas. The short-lived products of radon decay release two alpha particles and two beta particles. The half-life of this short Po-218, Pb/Bi/Po-214 decay chain is only 50 minutes. This short half-life allows an extra-ordinarily high density of alpha and beta particles.

Alpha and beta particles, impacting light atoms like beryllium (Be) and tritium (T), yield neutrons. This production of neutrons is diagrammed in Fig. 9, on the next page.

Figure 9 diagrams an intense, compact neutron source.

In 2004, TRAC sampled Ra-226 in water coming from DOE’s Y-12 facility at Oak Ridge. TRAC attributed that Ra-226 to new nuclear weapons material production at Y-12 [N. Buske, "Radioactive Pathways" (Discussion) and "Data Report of a Radiological Study...", both at www.radioactivist.org/ORRreports.html, TRAC (2005)]. DOE has denied new weapons production at Y-12.

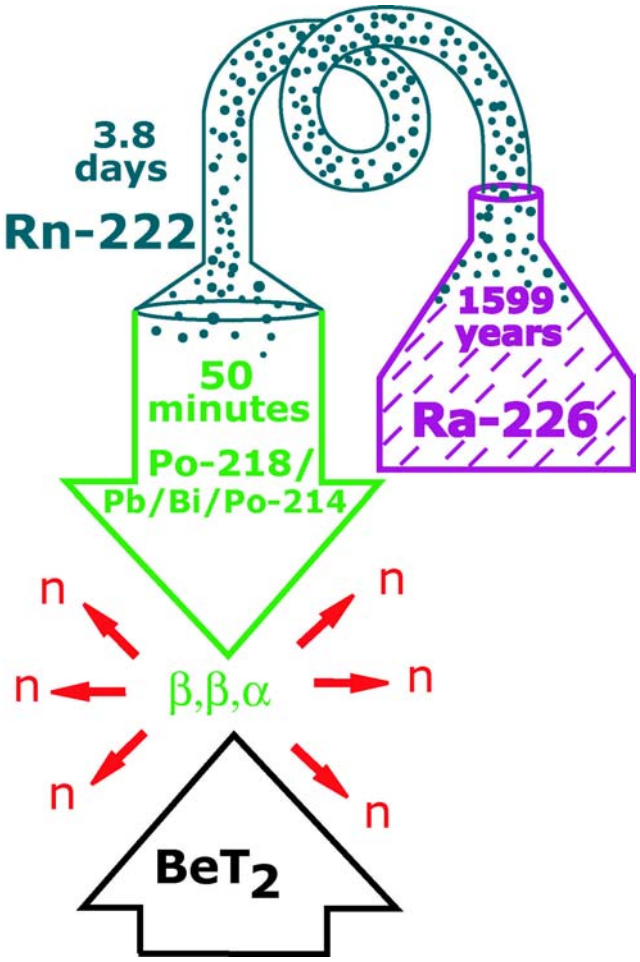

Fig. 9. Simple diagram of a compact neutron (n) source, powered by Ra-226. The half-life of the radionuclides in each unit of the neutron-production process is included above the radionuclides in that unit.

Production of new materials for next-generation, "micro" nuclear weapons is one possible explanation for Ra-226 running out of the Portsmouth GDP. The release of Ra–226 to creeks running from the GDP might then be attributed to maintenance scrubbing of radioactive impurities growing into the radium source stock of the intense, compact neutron sources.

Attachment 1: USEC Foam / Water Data

This attachment is also "Attachment 1" to a Memorandum from T. Taulbee [X-710, MS-2209, PORTS (2052), "Off-Site Monitoring for Increased Levels of Radioactivity," USEC File - POEF-X38300-03-280 (December 1, 2003) 4 pp.]. This attachment, below, was provided to TRAC by Ohio EPA on August 9, 2005:

Attachment I

Summary of Laboratory Sample Results:

				Gamma Spec results in pCi/g (solid) pCi/l (liquid)						
Date/Type	Alpha/Beta/Uranium			Tc-99/U-234/U-235/U-236/U-238					K-40	Bi-214
11/19/03 Water	0.0	0.0	0.521		na	na	na	na	na	na
11/19/03 Water	0.0	0.0	1.15		na	na	na	na	na	na
11/19/03 Foam	0.0	1.01	0.0555		na	na	na	na	na	na
11/19/03 Sediment	2.14	0.0	1.1489	0.0	0.3935	0.0	0.0	0.3861	9.9	0.0
11/19/03 Sediment	2.65	3.72	1.5462	0.0	0.5519	0.0	0.0	0.5196	11.1	0.0
11/19/03 Sediment	3.04	1.22	1.1871	0.0	0.5178	0.0394	0.0	0.3928	23.4	3.2
11/19/03 Rock	na	na	na	na	na	na	na	na	na	2.3
11/20/03 Foam	0.0	0.287	0.0163	na	na	na	na	na	na	na

na = not analyzed

Alpha Water Sample results are in pCi/ml
Beta Water Sample results are in pCi/ml
U water samples results are in ug/l
Foam U results are in mg/kg
Alpha sediment results are in pCi/g
Beta sediment results are in pCi/g
Tc-99 sediment results are in pCi/g

Due to the size and make-up of the sample, gamma spectroscopy was not performed on liquid samples. There was no technetium found in any soil samples. The results for uranium, uranium-234 and uranium-238 in secular equilibrium, and the almost total lack of U-235 in any sample indicates all samples contained only naturally occurring uranium, potassium (K-40) and bismuth (Bi-214). Potassium levels found (9.9 pCi/g, 11.1 pCi/g, and 23.4 pCi/g) closely match those levels expected for sandstone and shale (noted above).

Attachment 2: Letter by W.E. Murphie

—This attachment is a letter by W.E. Murphie, "Additional information regarding radiation levels associated with off-site creek foam," PPPO-01-227-05, Portsmouth/Paducah Project Office, Department of Energy, Lexington, KY (February 24, 2005), 3 pp. It is stamped, "ATTACHMENT ?4900500?".

(Text of this attachment is on the next three pages.)

ATTACHMENT

(049005008)

Department of Energy

Portsmouth/Paducah Project Office
1017 Majestic Drive, Suite 200
Lexington, Kentucky 40513
(859) 219-4000

8 2005

FEB 24 2005

Ms. Vina Colley
Portsmouth/Piketon Residents for Environmental
Safety and Security (PRESS)
3706 McDermott Pond Creek Road
McDermott, OH 45652

PPPO-01-277-05

Dear Ms. Colley

ADDITIONAL INFORMATION REGARDING RADIATION LEVELS ASSOCIATED WITH OFF-SITE CREEK FOAM

The purpose of this letter is to provide additional information regarding an issue you raised during the December 2, 2004, public meeting updating the community on Department of Energy (DOE) programs at the Portsmouth Gaseous Diffusion Plant (PORTS). The issue is related to the presence of foam in creeks and associated radiation levels near PORTS.

DOE understands that the issue regarding the creek foam was first raised at a November 18, 2003 press conference held by Portsmouth/Piketon Residents for Environmental Safety and Security (PRESS). At that time, Sergei Paschenko stated that foam and water samples he had taken from creeks near the plant showed field readings of beta radiation "at least 100 times higher than normal background levels." The site responded immediately by forming a team to evaluate the situation.

On November 19, 2003, the United States Enrichment Corporation (USEC) dispatched environmental and health physics professionals to the locations of concern referenced at the press conference to take radiation readings with state-of-the-art sampling and radiological monitoring instrumentation. Samples were collected from foam, surface water, and soil west of the plant at a drainage ditch near Wakefield Mound Road/Sargents Lane and at a location just west of Big Run Road, south of the plant property. Representatives from the Ohio Environmental Protection Agency's Southeast District Office observed the sampling activities.

Background radiation readings were taken with radiation detection field instruments capable of detecting below one-millionth of a Roentgen (microR). These instruments, known as Ludlum 12S mircoR meters, indicated normal background radiation levels of 10 microR per hour in both sample locations. As a reference, average background levels for the area surrounding the plant are historically 8-12 microR per hour. Radiation readings were also taken with a Geiger-Mueller equipped radiation detection meter.

All readings were less than 100 counts per minute (100 counts per minute is considered the minimum count rate detectable with this type of instrument). The results from this instrument provide additional confirmation that no unusual radiation was present at any sample location.

In addition to background radiation readings, environmental media samples were taken from both locations and analyzed at a laboratory. Results from the analysis of one of the two foam samples indicated gross beta radiation of 1.01pCi/ml (one-trillionth of a Curie of activity per milliliter of liquid). The second foam sample was 0.3pCi/ml. These results were at or near the limits of detection for the laboratory equipment used to conduct the analyses. These results did not indicate the presence of radionuclides above normal background levels. No Technetium-99 (^{99}Tc) was detected in the analyzed samples, and the historical surface water monitoring data has not indicated elevated ^{99}Tc levels. Additionally, soil and water samples analyzed from these locations did not indicate the presence of radionuclides above normal background levels. The analytical results were provided by USEC to the Ohio Environmental Protection Agency on December 10, 2003.

The appearance of foam on streams and creeks may be due to a number of causes. Streams that originate from woody areas often have a brown tint in the water caused by the presence of tannin, which is a substance that gives wood its color. Tannin is released during the decomposition of wood along with other materials that often cause foaming when they are introduced in water. Foam is common in dark-colored streams, especially in the late fall and winter when plant materials are decomposing in the water. During the November 19, 2003 sampling event, furniture and other man-made debris were observed in the off-site portions of the ditches/streams. These objects can also contribute to the formation of foam.

A comprehensive environmental monitoring program has been in place at the PORTS for many years to ensure the protection of the public and environment in accordance with all state and federal regulations. Water discharges are monitored periodically. Historical monitoring data shows that water discharges have not exceeded the applicable Nuclear Regulatory Commission discharge limits for radionuclides or corresponding DOE discharge requirements (DOE Order 5400.5). DOE provides monitoring data annually to the public in the Portsmouth Annual Environmental Report, which is available to stakeholders in the Environmental Information Center or by calling (740) 289-3317. We sent a copy of the most recent Annual Environmental Report to you under separate cover.

Please note that Lucy Henry with the Institute for Soviet and American Relations (ISAR), the organization that sponsored Mr. Pashenko's visit, was contacted in an effort to obtain the sampling data collected by Mr. Pashenko. Ms. Henry indicated that a report is not expected to be published until the spring of 2005. Accordingly, DOE has not had an opportunity to review the data from Mr. Pashenko's sampling activities.

If you have any additional questions or need further information, please contact Ms. Laura Schachter at (859) 219-4010.

Sincerely,

William E. Murphie
Manager
Portsmouth/Paducah Project Office

cc:
R. Miskelley, DOE/PPPO
K. Dewey, OH-EPA
J. Meersman. BJC/PORTS
P. Musser, USEC/Piketon

Sandy Cheldin
Jack Williams
Rosemary Richmond
Patrick Willsie

Attachment 3: Radiological Context of Foam / Water

——This attachment provides a somewhat technical, radiological context for the section, "Explanations of Ra-226 in Foam / Water from the GDP." An excellent reference for this development of context is E.M. Baum, H.D. Knox, and T.R. Miller's "Nuclides and Isotopes" (16th edition), Lockheed Martin Distribution Services, nuclides.chart@lmco.com (2002).

Pashenko was probably the first to sample and measure radioactivity in foam from creek water around the plant, in November 2003. USEC followed-up with foam sample results in Attachment 1, that G. Mitchell of the Ohio EPA characterized as "high but that it's impossible to know what they really indicate without more information [M. Lafferty, 'State Hopes to Unravel Radiation Readings near Piketon,' Columbus Dispatch (July 30, 2005) P. B4]]."

There is some relevant information. Radioactivity in the environment around the Portsmouth GDP has been monitored by DOE/USEC, Ohio state EPA, and others, over many years. However, almost all previous results do not relate directly to the Pashenko and USEC measurements of foam and water in 2003. The radiological context of the Pashenko and USEC measurements can be estimated from prior results by converting data to the same units of measure, by ordinary radiological calculations, and by extrapolations.

This attachment employs algebraic methods to make prior radiological data consistent with the new data of November 2003 and so to allow comparisons. In the text of this attachment and in the main report, terms like "U–238" describe an isotope of uranium having atomic number 238. In the displays of this attachment, "U" means "uranium" in an algebraic sense, that is either as a *variable* or as an *unknown* activity of uranium. Similarly, displayed "β" means a variable or unknown quantity of beta radioactivity. This nomenclature applies to other terms, as displayed.

A starting point for this development of radiological context is the conversion of prior measurements of elemental uranium concentration [in units of micrograms per liter = μg/L] in environmental samples into radioactivity [in units of pCi/L]. At the same time, the beta activity of the measured uranium can be calculated, assuming that the uranium decay chains are in *secular equilibrium*. This means that the absolute radioactivity of each and every radionuclide in a decay chain is equal, except for the stable end product of decay, which is not radioactive.

For the purpose of this attachment, the natural uranium decay chains are treated as in Fig. 7. The U–238 decay is thus considered distinctly from the U–234 decay chain, although U–234 is a member of the U–238 decay chain.

More than 95% of radioactivity in natural uranium is due to U–238 and U–234 decay, and less than 5% is due to U–235 decay. The U–238 decay chain emits two betas down to U–234. The U–234 decay chain then emits four betas down to stable Pb–206 at the end of the decay chain. Assuming secular equilibrium, the activities of U–238 and U–234 are equal. Therefore, the equilibrium ratio of beta emissions to natural uranium radioactivity is [(2+4)/2 =]

$$\beta/U = 3 \quad \lfloor_{equilibrium}.$$

Likewise, the natural thorium (Th) decay chain emits two betas from Th–232 down to Th–228. The Th–228 decay chain then emits two betas down to stable Pb–208. Assuming secular equilibrium, the activities of Th–232 and Th–228 are equal. Therefore, the equilibrium ratio of beta emissions to natural thorium radioactivity is [(2+2)/2 =]

$$\beta/Th = 2 \quad \lfloor_{equilibrium}..$$

In addition to the uranium and thorium decay chains, the main source of beta radioactivity in terrestrial materials is potassium-40 (K–40). One beta emission accompanies each K–40 decay:

$$\beta/K = 1.$$

In common, natural rocks in which the uranium and thorium decay chains are nearly in secular equilibrium, the *gross beta* radioactivity is nearly the sum of beta emissions from the decay chains of uranium, thorium, and potassium:

$$\text{Gross } \beta = 3U + 2Th + K \qquad \text{Eq.1}$$

where “Eq.1” references the first equation to which reference will be made later.

Uranium and thorium are in the relatively insoluble Actinide Group of elements; whereas, potassium belongs to the relatively soluble Group 1 elements. In consideration of these different solubilities, the actinide primary sources of beta radioactivity are separated from potassium, by defining a *net beta* radioactivity, assuming secular equilibrium of the decay chains:

$$\text{Net } \beta = \text{Gross } \beta - K \qquad \text{Eq.2}$$

$$\text{Net } \beta = 3U + 2Th \qquad \text{Eq.3}$$

$$\text{Net } \beta / U \text{ (in common rocks)} = 3 + 2Th/U \qquad \text{Eq.4}$$

Thorium and uranium radioactivities, and their ratios, are listed for common rocks in Table A1, on the next page.

Table A1. Radioactivities of uranium and thorium in common rocks [pCi/g]*

Kind of Rock:	Limestone	Sandstone	Shale
Thorium:	0.14	0.65	1.09
Uranium:	0.43	0.40	0.40
Th/U [no units]:	0.33	1.62	2.72

* 'g' = gram. [Memorandum from T. Taulbee, X-710, MS-2209, PORTS (2052), "Off-Site Monitoring for Increased Levels of Radioactivity," USEC File - POEF-X38300-03-280 (December 1, 2003) p. 2; original reference: UNSCEAR (1958) p. 52.]

For the purpose of this discussion, "common rocks" means natural rocks having radioactive contents within the range listed in Table A1.

From the bottom line in Table A1, the range of Th/U radioactivities in common rocks ranges from about 0.33 in limestone to about 2.72 in shale, with sandstone intermediate:

$$0.33 \text{ (in limestone)} \leq \text{Th/U (in common rocks)} \leq 2.72 \text{ (in shale)}$$

This range of Th/U ratios substitutes into Eq.4, yielding a range of ratios of Net beta radioactivity to uranium radioactivity in common rocks:

$$3.7 \text{ (in limestone)} < \text{Net } \beta \text{ / U (in common rocks)} < 8.6 \text{ (in shale)}$$

An *Average* Net β/U in common rocks, as the mean of the three Th/U values in the bottom line of Table A1 is

$$\textit{Average} \text{ Net } \beta \text{ / U (in common rocks)} = 6.1.$$

Net β / U in common rocks compares to Ohio EPA's measurements in 1997 of Net β / U in surface waters in Little Beaver Creek, near the Portsmouth GDP. Ohio EPA collected four water samples at each of 6 locations and analyzed them for gross beta, uranium, and potassium. The four surface water samples collected from location RM 1.00 on Little Beaver Creek had one report of "less than lab detection" for gross beta, but there were no "less than lab detection" reports for uranium or potassium in water from that location.

Ohio EPA results for each of the other 5 locations had at least three reports of "less than lab detection" for gross beta and/or uranium. Non-quantitative reports of "less than lab detection" bias the mean values of the quantitative results upward. Therefore, the ratios of mean Gross β ÷ mean U for the other 5 locations are less reliable and are not used here.

The uranium (U) and potassium (K) results for location RM 1.00 are translated from chemical units into radioactivities in Table A2.

Table A2. Beta and uranium in Little Beaver Creek water (RM 1.00)]

Gross β *	U	Gross β / U	K	Net β / U
16. pCi/L	1.85 pCi/L	8.6	3.8 pCi/L	**6.6**

* Each tabulated value is the mean of four water sample analyses during 1997, with the exception of Gross β, for which one of the four samples was reported as 'less than lab detection.' ["Biological and Water Quality Study of Little Beaver Creek and Big Beaver Creek –1997, Portsmouth Gaseous Diffusion Plant, Pike County, Ohio," DSW/MAS 1998-5-1, Ohio state Environmental Protection Agency, Ecological Assessment, Division of Surface Water, Columbus, Ohio (June 4, 1998) Table 5.] The tabulated value for Gross β is the mean of three sample reports.

Conversion factors from chemical to radiological data:

Natural Uranium radioactivity:	0.686 pCi/µg
Potassium-40 (K-40) radioactivity in natural potassium:	1.224 pCi/mg

Net β / U = 6.6, in Table A2, for surface water around Portsmouth, is a little higher than the *Average* Net β / U = 6.1 in common kinds of rocks observed in the Pike County, Ohio, as previously mentioned.

For this discussion, assume that the Net β / U value in Table A2 represents natural background for the creek foam / water samples collected by Pashenko and by USEC in November 2003:

Net β / U (in background surface water or foam) = 6.6

—Whatever uranium value is reported in a sample, 6.6 times that value of Net β / U is treated as natural or background Net β / U.

This assumption (that background Net β / U = 6.6) implies that all beta radioactivity that was measured at location RM 1.00 in Little Beaver Creek in 1997 was naturally present. If there happens to be any beta emitting technetium-99 or other artificial beta emitter in the surface water samples from location RM 1.00 in Table A2, that artificial beta activity is treated as natural. This assumption is somewhat conservative in the present context.

Yet this does not seem an overly conservative assumption. Net β / U = 6.6 is only slightly higher than an average value (6.1) in common rocks. Furthermore, preferential solution of naturally present Ra–226 into creek water in comparison to relatively

insoluble actinide precursors U–234 and Th–230 might easily account for beta emissions elevated above secular equilibrium. Finally, official monitoring results for beta emitting technetium–99 and protactinium-233 (as parent radionuclide neptunium–237) do not evidence the presence of *artificial* beta-emitting radionuclides in surface waters around the GDP ["U.S. Department of Energy Portsmouth Annual Environmental Data for 2003, Piketon, Ohio," DOE/OR/11-3153&D1, EQ Midwest, Inc., Cincinnati, Ohio (November 2004) Table 4.21]. Therefore, there is no reason to suppose that significant beta activity originating from the GDP might be concealed in Net β / U = 6.6.

Finally, the range of beta-emitting potassium-40 (K) measured by Ohio EPA in surface waters around Portsmouth was narrow, between 3.47 and 3.88 pCi/L; that is:

$$3.47 \text{ pCi/L} \leq K \leq 3.88 \text{ pCi/L}.$$

In the same sense that Net β / U=6.6 is slightly conservative, a slightly conservative estimate of K–40 that is naturally present in creek foam / water is:

K (in surface water or foam) = 3.9 pCi/L

This completes the particular radiological context in which Pashenko and USEC collected foam / water samples from creeks around the Portsmouth GDP in November 2003.