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Abstract

The enrichment of uranium, often for nuclear weapons programs and commercial nuclear reactors, produces higher concentrations of radioactive uranium 235 (^{235}U) than what naturally occurs, which can pose a human health hazard. The most abundant naturally occurring uranium isotope is ^{238}U , which is still radioactive, however a higher concentration of ^{235}U skews the observed isotopic uranium distribution. The Department of Energy Portsmouth Gaseous Diffusion Plant, located near Piketon, OH, enriched uranium from 1954 to 2001 and ^{235}U pollution has recently been detected in air and sediment samples in the surrounding community. The extent of the ^{235}U contamination was tested using sediment core samples from ponds within the vicinity of the plant. Cores were processed in the laboratory by depth intervals to capture the history of atmospheric deposition of ^{235}U . Analyzing the samples with an ICPMS instrument allowed for ratios between enriched ^{235}U and naturally occurring ^{238}U to be calculated, thus unearthing the level of contamination. These results will provide the citizens of Piketon, OH with scientific evidence to facilitate their understanding of the uranium contamination within their community.

Keywords

Uranium, Enriched uranium, Mass spectrometry, Sediments, Health

Publication Statement

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Records of Enriched Uranium Atmospheric Deposition in Pond Sediments in Piketon, OH

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Abstract

The enrichment of uranium, often for nuclear weapons programs and commercial nuclear reactors, produces higher concentrations of radioactive uranium 235 (^{235}U) than what naturally occurs, which can pose a human health hazard. The most abundant naturally occurring uranium isotope is ^{238}U , which is still radioactive; however, a higher concentration of ^{235}U skews the observed isotopic uranium distribution. The Department of Energy Portsmouth Gaseous Diffusion Plant, located near Piketon, OH, enriched uranium from 1954 to 2001 and ^{235}U pollution has recently been detected in air and sediment samples in the surrounding community. The extent of the ^{235}U contamination was tested using sediment core samples from ponds within the vicinity of the plant. Cores were processed in the laboratory by depth intervals to capture the history of atmospheric deposition of ^{235}U . Analyzing the samples with mass spectrometry allowed for ratios between enriched ^{235}U and naturally occurring ^{238}U to be calculated, thus unearthing the level of contamination. Clear evidence of enriched uranium in the Piketon, OH community was found in all sediment core and surface sediment grab samples, which each exhibited some degree of elevated $^{235}\text{U}/^{238}\text{U}$. These results will provide the citizens of Piketon, OH with scientific evidence to facilitate their understanding of the uranium contamination within their community.

1 INTRODUCTION

Naturally occurring uranium (U) comes in two major isotopes, 99.3% uranium 238 (^{238}U) and 0.7% uranium 235 (^{235}U), and two minor isotopes, 0.0055% uranium 234 (^{234}U) and <10-8 % uranium 236 (^{236}U ¹). When uranium is enriched, this naturally occurring ratio shifts so that there is a greater abundance of ^{235}U . While the uranium isotopes ^{238}U and ^{235}U are chemically identical, they have different masses. The difference in mass allows the uranium isotopes to be separated, thus making it possible to increase the percentage of ^{235}U in a given sample¹.

Just south of the rural town of Piketon, OH in Pike County is the former Department of Energy Portsmouth Gaseous Diffusion Plant (coordinates: 39.012982, -83.000872). This Department of Energy (DOE) facility enriched uranium from 1954 to 2001. Initially this uranium was for use in the United States' nuclear weapons program during the Cold War. Following the Cold War, the DOE leased the facility to the private sector and uranium, which was no longer weapons-grade, was enriched for commercial nuclear reactors. This uranium enrichment aimed to create large quantities of ^{235}U , the desired uranium isotope for nuclear applications. The enrichment process relies on conver-

sion of U compounds to uranium hexafluoride (UF_6 ; g), which is separated isotopically based upon differences in the rate of effusion through a porous barrier. An environmental cleanup program was implemented at the site in 1989 by the DOE's Office of Environmental Management, and decontamination and decommissioning of the plant started in 2011².

In general, greater amounts of uranium coincide with an increase in human health hazards. The presence of enriched uranium in an environment particularly escalates health hazards because it is often associated with an elevation in total uranium concentration. Additionally, as uranium is enriched, the proportions of the shorter-lived, more radioactive isotopes ^{234}U and ^{235}U are enhanced versus nature; therefore, enriched uranium is more radioactive than naturally occurring uranium on a mass basis. Studies have found that uranium poses human health risks beyond those caused by radiation exposure; uranium exposure can be genotoxic and mutagenic by forming strand breaks in DNA³. Uranium exposure also has the potential to be carcinogenic, and a 2011 study found that incidences of colorectal, breast, kidney, and total cancer may be greater in areas that suffer from elevated levels of U in groundwater and experience more frequent groundwater usage⁴.

There is currently radioactive pollution, particularly

in the form of enriched ^{235}U , in the Piketon community surrounding the Portsmouth Gaseous Diffusion site. Enriched uranium has been found in several sediment and air samples in the Piketon area within the past year. In April 2019, sediments, interior dusts, and surface waters in the community were confirmed to be contaminated with ^{235}U and non-fallout plutonium and neptunium ^{237}U . Shortly after in June 2019, it was found that the local Zahn's Corner Middle School was contaminated with ^{235}U ⁶. Determining the history of enriched uranium from this gaseous diffusion facility will grant the citizens of Piketon, OH scientific evidence that will facilitate their understanding of the uranium contamination issue in their community. The extent of the enriched uranium contamination is not well understood at this time; the DOE is in the process of conducting a "third party" study of the environment within a 7-mile radius of the facility. Naturally, the Piketon community is immensely concerned about the potential health implications of this probable uranium pollution and would like answers in order to take action. The mission of this research project is to determine if there is history of atmospheric deposition of enriched uranium in the areas surrounding the Portsmouth Gaseous Diffusion Plant, and if so, what the extent of that pollution is.

2 METHODS

Pond sediment core samples and surface sediment grab samples were collected in the Piketon area within the vicinity of the DOE facility on October 5, 2019 by Dr. Michael Ketterer, Mrs. Elizabeth Lamerson, and Mr. Josh Lamerson. Samples were collected from three different farm ponds known by locals to have not been dredged or disturbed since the 1950s when the DOE facility was built. These ponds are the Bobo Road pond (coordinates: 39.010000, -82.972778), the Bailey Chapel pond (coordinates: 38.993056, -82.995278), and the Dutch Run pond (coordinates: 39.010556, -82.983889; Figure 1).

The first samples were collected from the Bobo Road pond, which was on the Lamerson's property. The pond was approximately 10-12 ft. deep at its deepest sections, had an outflow and moving water, and had been drained to remove built-up surface material approximately 8 years ago (this did not affect the sediment in the pond). Two sediment core samples (named Lam0098 and Lam0099) were each collected by plunging a 19mm internal diameter copper pipe straight down into the pond and underlying sediment to maximum depth. Pipes were pulled out, capped with copper end caps, sealed with electrical tape, and labeled with sample name and collection date. A surface sediment grab sample was also collected from the Bobo Road pond (named Lam0100) by scooping up sediment with a gallon sized bucket and pouring it into a labeled Ziploc bag.

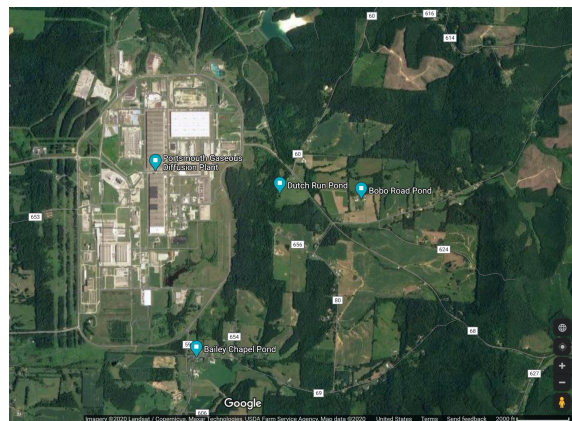


Figure 1. Three pond sampling locations (the Bobo Road pond, the Bailey Chapel pond, and the Dutch Run pond) for collecting sediment core samples to analyze for enriched uranium were located within 2 miles of the Portsmouth Gaseous Diffusion Plant, the location of uranium enrichment activities for several decades⁷.

The Bailey Chapel pond was the second sampling location. Samples were collected closer to the edge of this pond due to lower accessibility caused by cattails, weeds, and fencing. This lower accessibility resulted in a surface sediment grab sample using the gallon bucket method not being collected at this location. However, the sampling team was successful in extracting two sediment core samples (named Lam0101 and Lam0102) using the copper piping method.

The final sampling location was the Dutch Run pond. Barriers such as plants and fencing did not cause sample collection issues at this pond like they had at the previous location. However, this pond was more stagnant than the others and had some algae on its surface, though any changes in water chemistry caused by these algae would likely have no impact on any uranium present in sediments. Two sediment core samples (named Lam0103 and Lam0105) were collected via the copper piping method and one surface sediment grab sample (named Lam0104) was collected via the gallon bucket method.

Two months after the sample collection was completed and samples were certain to be well-contained with no leaks, Mrs. Elizabeth Lamerson shipped all 8 collected samples as well as one empty copper pipe as a blank sample from Piketon, OH to the University of Denver (DU) in Denver, CO (Figure 2). The samples were mailed out on December 27, 2019 and were received by Dr. Michael Ketterer at DU on January 3, 2020. After the samples were unpacked, lab processing and analysis of samples were performed by the student researcher.

In the DU Department of Chemistry and Biochemistry Majestic Lab, the copper pipes containing the sediment cores were each measured and marked with a Sharpie marker at 3cm depth intervals and cut at these

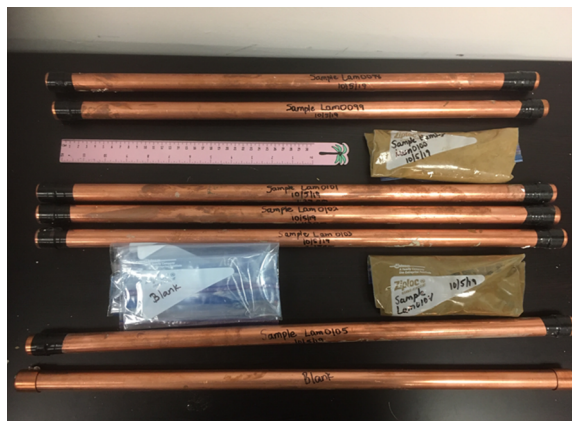


Figure 2. Piketon, OH pond sediment samples from October 2019 collection were mailed to the University of Denver in the wintertime. These samples included 6 sediment cores in copper pipes, 2 surface sediment grab samples in Ziploc bags, and 1 blank in a copper pipe to be analyzed for evidence of enriched uranium contamination.

intervals using a handheld pipe cutter. Each 3cm sample was transferred to a glass vial and labeled according to the core it came from and its depth range. The two surface sediment grab samples were transferred from their plastic bags to glass beakers and labeled according to sample number. Core and surface sediment grab samples were then heated at 85°C in a small Sargent-Welch S-63995 oven for several days to evaporate all water content.

After initial sample drying, sediment samples were ground one by one using a mortar and pestle and placed back into their corresponding vial or beaker. Approximately 0.5g of each sample was then weighed and placed into a smaller labeled glass vial, producing 66 samples. Also included in the set of small glass vials with individual samples were 10 random duplicates of the samples, 5 known control samples and duplicates for each, and 5 blanks. This produced a total of 91 samples, which were dry ashed in a furnace that could maintain steady high temperatures for extended periods of time at 600°C for three hours. Following the dry ashing, 3mL of 2M nitric acid (HNO₃) was added to each sample to leach them. The 91 sample bottles with the nitric acid were left overnight in the Sargent-Welch S-63995 oven at 80°C and were pulled out and shaken approximately every few hours when personnel were available to come into the lab.

Due to the onset of the COVID-19 pandemic and restrictions of student access to labs, the following steps were performed by Dr. Michael Ketterer instead of the student researcher. Using a 7mL Globe Scientific column pipet with a cotton plug, the samples were filtered and 2.0-2.2mL of each sample solution was recovered. Next, 50mg of UTEVA resin was added to each sample; it was pipetted in the form of a concentrated aqueous slurry. In this step, the UTEVA resin polymeric beads were

used in solid-phase extraction in which the uranium was purified (separated from the nitric acid) by binding to polymeric beads⁸. The UTEVA resin-sample mix was transferred to a 7mL Globe Scientific column pipette with a cotton plug for filtration, and the pass-through solution was discarded. Each column was then rinsed twice with 1mL of 2M HNO₃ to remove un-retained components of the solution, while U remained bound to the resin column, and the rinse solution was discarded. The U was eluted with a sequence of the following: 0.5mL of H₂O, 0.5mL of 0.05 M aqueous ammonium oxalate, and 0.5mL of H₂O. The eluted samples were collected in the same 10mL tube which was used to collect the filtered sample leach solution.

At Northern Arizona University, a Thermo X2 quadrupole inductively coupled plasma mass spectrometer (ICPMS) instrument was used by Dr. Ketterer to determine the relative abundances of ²³⁴U, ²³⁵U, ²³⁶U, and ²³⁸U in each sample. The instrument was equipped with an APEX HF high efficiency desolvating sample introduction system⁹. The ICPMS instrument converts the uranium present in the sample extract into gas phase ions (i.e., U⁺ (g)) and then sorts the ions in a quadrupole mass analyzer according to their mass¹⁰. This instrument records counts of the levels of each uranium isotope in a sample because different isotopes have different masses. To run the UTEVA-prepared samples through the ICPMS, the samples' ammonium oxalate fractions from the UTEVA columns were diluted to yield approximately 1 million cps (counts per second) at m/z 238 with 1% HNO₃, so that relatively consistent signal levels were recorded for all samples. This obviates several possible sources of systematic error such as "detector dead time."

The ICPMS analyzed each sample with either 3 or 5 runs with 46 second acquisitions, 10 seconds per mass, and a 10 ms dwell and 1000 sweeps. A control sample of naturally occurring U, derived from IRMM-056 (a uranium standard solution that can be used as a control for naturally occurring uranium) was diluted to give 10⁶ cps at ²³⁸U and run first as a control; the IRMM-056 solution was analyzed again in the middle and at the end of the analytical session.

The raw ICPMS data were transferred from Dr. Ketterer to the student researcher for analysis. Data were put into Microsoft Excel to run calculations that produced uranium isotope ratio values for every sample (²³⁴U/²³⁸U, ²³⁵U/²³⁸U, and ²³⁶U/²³⁸U). First, calculations were performed on the 4 blank samples that were run through the ICPMS. The average concentration of each uranium isotope (²³⁴U, ²³⁵U, ²³⁶U, and ²³⁸U) was calculated from these 4 blanks to be subtracted from the isotope signals of the rest of the data from the ICPMS to improve accuracy. For each isotope value for each run in the ICPMS (either 3 or 5 runs per sample), the average blank value for the corresponding isotope was

subtracted. Next, isotopic ratios were calculated for each run by dividing the ^{234}U , ^{235}U , and ^{236}U values, respectively, by the ^{238}U value that were made from blank subtraction. ^{238}U was used as the denominator in the ratios because it is naturally the most abundant uranium isotope and will therefore have the most counts per second in each ICPMS run. The average of each $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$, and $^{236}\text{U}/^{238}\text{U}$ ratio was then calculated for each sample, and one standard deviation for each ratio was calculated as well. For statistical analysis of these data, single factor ANOVA tests were performed in Microsoft Excel for each sediment core sample.

3 RESULTS

Naturally occurring ratios for $^{234}\text{U}/^{238}\text{U}$, $^{235}\text{U}/^{238}\text{U}$, and $^{236}\text{U}/^{238}\text{U}$ in nature, respectively, are $5.5\text{E-}5$, 0.00725 , and 0 , and ICPMS results were compared to these thresholds. The natural value for $^{235}\text{U}/^{238}\text{U}$ is the primary ratio of focus in this study as enriched ^{235}U is radioactive and was likely emitted during the operation of the Portsmouth Gaseous Diffusion facility. This natural $^{235}\text{U}/^{238}\text{U}$ ratio of 0.00725 is relatively stable in nature; however, anthropogenic activities such as uranium enrichment for nuclear weapons can create large variations in this ratio¹¹. Calculated $^{235}\text{U}/^{238}\text{U}$ ratios that are significantly greater than this natural ratio indicate higher levels of enriched ^{235}U from anthropogenic sources. Across all sediment core and surface sediment grab samples, above-natural ratios of $^{235}\text{U}/^{238}\text{U}$ were observed at different intensities, and these intensities varied with depth for the core samples. Depth variations correspond to variations in time due to the deepest layers of sediment being the oldest and the surface levels of sediment having been recently deposited. Out of 66 sediment samples (representing the sectioned sediment cores or surface sediment grab samples), only two samples did not have $^{235}\text{U}/^{238}\text{U}$ ratios that exceeded the naturally occurring value (the 22.5cm depth sample from Lam0099 and the 52.5cm depth sample from Lam0101). When one standard deviation below the calculated $^{235}\text{U}/^{238}\text{U}$ ratio was computed, again only the same two out of the 66 samples had $^{235}\text{U}/^{238}\text{U}$ ratios that were less than the naturally occurring value.

The sediment samples from the Bobo Road pond (cores Lam0098 and Lam0099 and surface sediment grab sample Lam0100) all exhibited elevated levels of enriched ^{235}U (Figures 3 and 4). The shortest sediment core, Lam0098, did not show as high of ^{235}U contamination as Lam0099, which was approximately 9cm longer. The peak $^{235}\text{U}/^{238}\text{U}$ value for Lam0098 was 0.00739 , which occurred both at average depths of 1.5cm and 13.5cm . The peak $^{235}\text{U}/^{238}\text{U}$ value for Lam0099 occurred at an average depth of 4.5cm and was 0.00853 .

Sediment samples collected from the Bailey Chapel

Sediment Core Sample	Minimum $^{235}\text{U}/^{238}\text{U}$	Maximum $^{235}\text{U}/^{238}\text{U}$
Lam0098	0.00730	0.00739
Lam0099	0.00725	0.00853
Lam0101	0.00723	0.00761
Lam0102	0.00735	0.00763
Lam0103	0.00736	0.00934
Lam0105	0.00739	0.00941

Table 1 Maximum and minimum $^{235}\text{U}/^{238}\text{U}$ values for all 6 sediment core samples collected from three different ponds in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant. Each value is for a different average core depth; during lab processing, each sediment core was cut into 3cm depth intervals where each depth interval was analyzed to produce uranium isotope concentration data.

pond (core samples Lam0101 and Lam0102) showed elevated levels of $^{235}\text{U}/^{238}\text{U}$ (Figure 3). These isotope ratios fluctuated over depth and therefore time significantly more than the samples from the other locations. The peak $^{235}\text{U}/^{238}\text{U}$ value for Lam0101 was 0.00761 at 31.5cm , and the peak value for Lam0102 was 0.00763 at 28.5cm .

The sediment samples from the final location, Dutch Run pond, (core samples Lam0103 and Lam0105 and surface sediment grab sample Lam0104) exhibited the highest levels of enriched uranium (Figures 3 and 4). The peak $^{235}\text{U}/^{238}\text{U}$ value was 0.00934 at an average depth of 16.5cm for Lam0103 and was 0.00941 at an average depth of 13.5cm for Lam0105.

To statistically analyze the results from the sediment core samples, ANOVA single factor tests were performed in Microsoft Excel. A test was executed for each of the 6 core samples with an alpha value of 0.05 using the calculated $^{235}\text{U}/^{238}\text{U}$ values (one value per 3cm depth interval) and the naturally occurring $^{235}\text{U}/^{238}\text{U}$ value. Only one of the 6 samples returned a p -value greater than 0.05 (sediment core Lam0099; Table 2).

Isotopic ratios of all samples were also graphically compared to one another. The $^{235}\text{U}/^{238}\text{U}$ values were plotted against both the $^{236}\text{U}/^{238}\text{U}$ and the $^{234}\text{U}/^{238}\text{U}$ values. Both graphs showed a positive correlation where as ^{235}U increased, so did ^{234}U and ^{236}U . However, the correlation was much stronger for $^{235}\text{U}/^{238}\text{U}$ vs. $^{236}\text{U}/^{238}\text{U}$ than for $^{235}\text{U}/^{238}\text{U}$ vs. $^{234}\text{U}/^{238}\text{U}$ (Figures 5 and 6).

Uranium Deposition

Sediment Core	df Between	df Within	F	F Critical Value	p-Value
Lam0098	1	8	26.789	5.3177	0.0085
Lam0099	1	14	4.1572	4.6001	0.06080
Lam0101	1	34	85.222	4.1300	868E-11
Lam0102	1	30	113.98	4.1709	9.76E-12
Lam0103	1	18	18.086	4.4139	0.00048
Lam0105	1	12	12.765	4.7472	0.00383

Table 2 ANOVA single factor test results for all 6 sediment core samples extracted from three ponds in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant, which historically enriched uranium. An ANOVA test was run for each sediment core using the $^{235}\text{U}/^{238}\text{U}$ values (one value per 3cm depth interval) and the naturally occurring $^{235}\text{U}/^{238}\text{U}$ value.

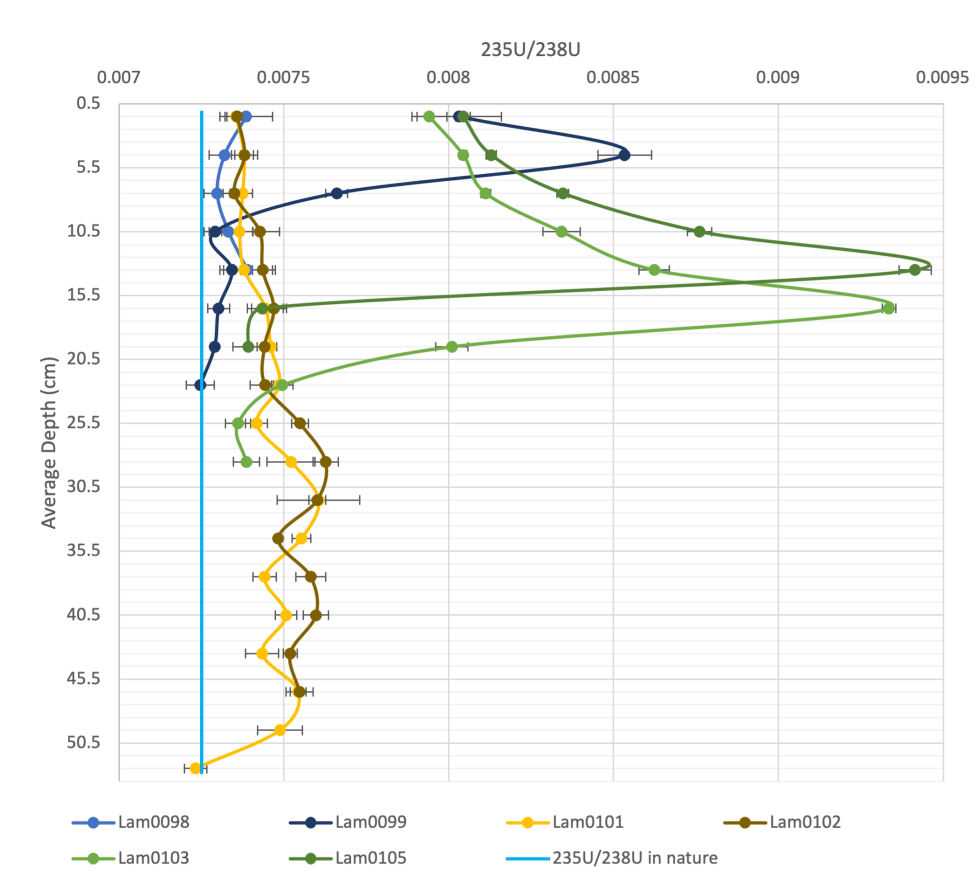


Figure 3. Uranium $^{235}\text{U}/^{238}\text{U}$ ratios in 6 sediment core samples collected from 3 different ponds versus the naturally occurring uranium $^{235}\text{U}/^{238}\text{U}$ ratio. Error bars are one standard deviation of the $^{235}\text{U}/^{238}\text{U}$ values. Sediment cores Lam0098 and Lam0099 were extracted from the Bobo Road pond, cores Lam0101 and Lam0102 were extracted from the Bailey Chapel pond, and cores Lam0103 and Lam0105 were extracted from the Dutch Run pond. All ponds are located in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant, which historically enriched uranium, and analyzed in a lab by depth intervals using an ICPMS instrument to produce uranium isotope concentration data.

4 DISCUSSION

Careful analysis of pond sediments provided clear evidence of enriched uranium in the Piketon, OH commu-

nity. All 6 sediment cores and 2 surface sediment grab samples exhibited some degree of elevated $^{235}\text{U}/^{238}\text{U}$, with the overall lowest value being 0.00723 at an average depth of 52.5cm for Lam0101 and the overall highest

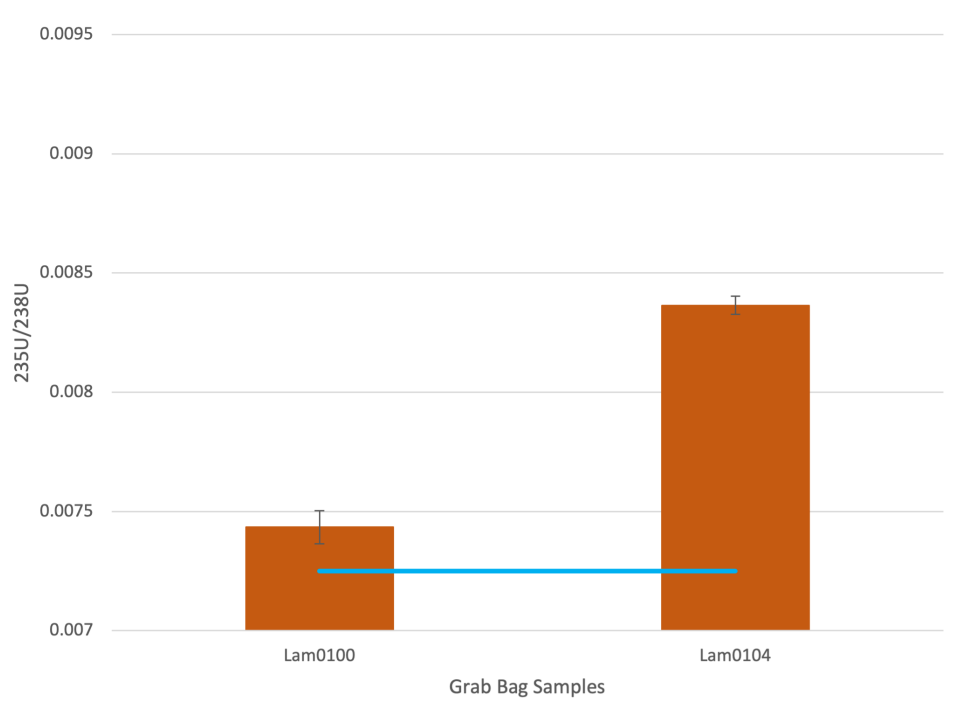


Figure 4. Uranium $^{235}\text{U}/^{238}\text{U}$ ratios in the surface sediment grab samples (Lam0100 from Bobo Road pond and Lam0104 from Dutch Run pond) versus the naturally occurring uranium $^{235}\text{U}/^{238}\text{U}$ ratio (seen as the blue line). Error bars are one standard deviation of the $^{235}\text{U}/^{238}\text{U}$ values. The surface sediment grab samples were collected via gallon bucket from two ponds in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant, which historically enriched uranium, and analyzed in a lab using an ICPMS instrument to produce uranium isotope concentration data.

value being 0.00941 at an average depth of 13.5cm for Lam0105 (Table 1, Figure 3).

The most pronounced contamination out of all of the samples was seen in the Dutch Run pond, which was the pond closest to the Portsmouth Gaseous Diffusion Plant. The two core samples from the location exhibited the highest $^{235}\text{U}/^{238}\text{U}$ ratios with 6 of 10 (Lam0103) and 5 of 7 (Lam0105) depth interval samples exceeding a ratio of 0.008 (Figure 3). The surface sediment grab sample from this location also exceeded a ratio of 0.008 with a value of 0.00836 (Lam0104; Figure 4). Out of the 5 other samples (3 from Bobo Road pond and 2 from Bailey Chapel pond) only 1 other sample, from Bobo Road pond, had any $^{235}\text{U}/^{238}\text{U}$ values exceed 0.008 (2 of 8 samples in Lam0099). These findings are statistically significant as the ANOVA test produced $F(1,18) = 18.086$, $p = 0.00048$ for Lam0103 and $F(1,12) = 12.765$, $p = 0.00383$ for Lam0105 (Table 2). This pond is also located directly east of the plant, so it is probable that the westerly winds blew the contamination from the plant in the direction of the pond. These findings indicate that the Dutch Run pond was subjected to the most enriched uranium atmospheric deposition out of the sampling locations, which makes sense due to the proximity of the plant to the pond and natural wind direction making the area more susceptible to deposition accumulation.

The Bobo Road pond displayed the second highest level of contamination, which makes sense due to it also being directly east of the plant and therefore in the direct path of westerlies blowing from the plant. The first sediment core collected from this pond (Lam0098) was also the first core collected out of all the samples, and it was the shortest (Figure 3). Less accurate sampling due to it being the first one may have an influence on this sample; this could explain the discrepancy with the second sample (Lam0099) taken from this pond, which was significantly longer and contained more enriched uranium (Figure 3). No other sets of cores taken from the same pond displayed the same discrepancy as the two taken from the Bobo Road pond, so this is likely a result of inaccuracies from attempting to perfect the sampling technique. However, the ANOVA test found that the uranium concentrations in the second of the two cores, Lam0099, were not statistically significant with $F(1,14) = 4.1572$, $p = 0.06080$ (Table 2). This was the only one out of all 6 sediment core samples where the p -value exceeded 0.05. ANOVA results for the shorter of the Bobo Road pond cores, Lam0098, were $F(1,8) = 26.789$, $p = 0.00085$, which displays a significantly different p value (Table 2). The non-statistically significant p -value for Lam0099 may be due to this being the only core where within-group variance was greater than between-group variance. The high $^{235}\text{U}/^{238}\text{U}$ variance by depth

Uranium Deposition

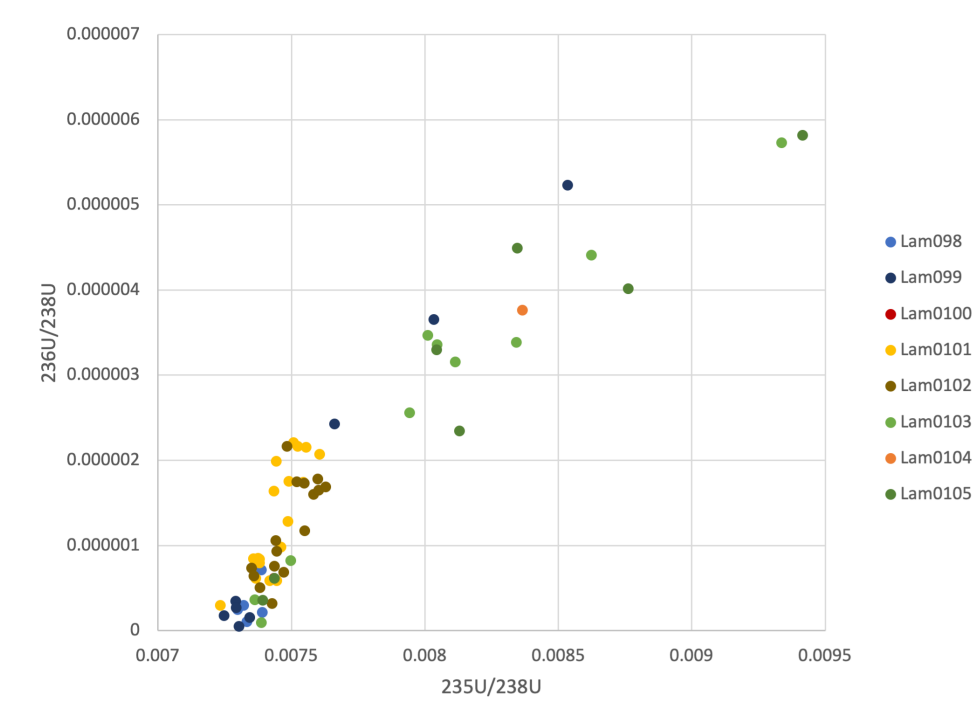


Figure 5. Uranium $^{235}\text{U}/^{238}\text{U}$ ratios versus uranium $^{236}\text{U}/^{238}\text{U}$ ratios for all sediment core and surface sediment grab samples, calculated from ICPMS results; a positive correlation is evident. The sediment core and surface sediment grab samples were collected from the three ponds in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant, which historically enriched uranium, and analyzed in a lab using an ICPMS instrument to produce uranium isotope concentration data. For the sediment core data, each data point on the graph is a different 3cm depth interval.

in the core could have resulted from a sudden and drastic increase in uranium deposition at the pond after a long period of low deposition, which may be the jump in data seen graphed from a depth of 10.5cm to 7.5cm (Figure 3).

Uranium 235 concentrations in the Bailey Chapel pond fluctuated the most across depth intervals of the two core samples, Lam0101 and Lam0102 (Figure 3). This pond was close in proximity to the Portsmouth Gaseous Diffusion Plant and was located to the southeast of it. The westerlies may also play a role in the deposition that reached this pond, as not being in the direct path of the westerly winds may have reduced instances of constant pollution but fluctuations in wind patterns may have brought more sporadic pollution. This also may attribute to the overall lower $^{235}\text{U}/^{238}\text{U}$ concentrations seen in the Bailey Chapel pond samples compared to the other samples; the lowest overall value seen out of all of the samples came from the first of the two Bailey Chapel samples (Lam0101). These lower, fluctuating data however are statistically significant; ANOVA results for Lam0101 were $F(1,34) = 85.222$, $p = 8.68\text{E-}11$ and for Lam0102 were $F(1,30) = 113.98$, $p = 9.76\text{E-}12$ (Table 2).

While the numeric depth of the sediment cores is of little meaning due to the cores not having been chronologically dated, it is evident that the uranium concen-

trations do change with depth and therefore there is a definite timeline of enriched uranium deposition from the Portsmouth Gaseous Diffusion facility. The Dutch Run pond samples exhibit this the most clearly because the graphs of the two core samples are strikingly similar in their shapes (Figure 3). Peak $^{235}\text{U}/^{238}\text{U}$ values for these samples are very close (0.00934 for Lam0103 and 0.00941 for Lam0105) and occurred at similar average depths (16.5cm for Lam0103 and 13.5cm for Lam0105; Table 1). The peak values graphically indicate sharp increases from the $^{235}\text{U}/^{238}\text{U}$ values of the sample directly below the peak samples in core depth, which may indicate a drastic increase in uranium enrichment operations during the Cold War; Lam0103 values rose from 0.00749 to 0.00801 to 0.00941 within 6cm and Lam0105 values rose from 0.00743 to 0.00941 within 3cm (Figure 3). The faster increase seen in the latter of these samples may be a result of a slower sedimentation rate in the specific section of the pond from which it was extracted.

Beyond the $^{235}\text{U}/^{238}\text{U}$ concentrations seen in the samples, the relationship between ^{235}U concentrations and ^{234}U and ^{236}U concentrations points to anthropogenic influences on the presence of uranium. On plots comparing $^{235}\text{U}/^{238}\text{U}$ to $^{234}\text{U}/^{238}\text{U}$ or $^{236}\text{U}/^{238}\text{U}$, the cluster of samples closer to the vertex that have overall low uranium concentrations are close to naturally occurring levels, and as the samples move further away from the

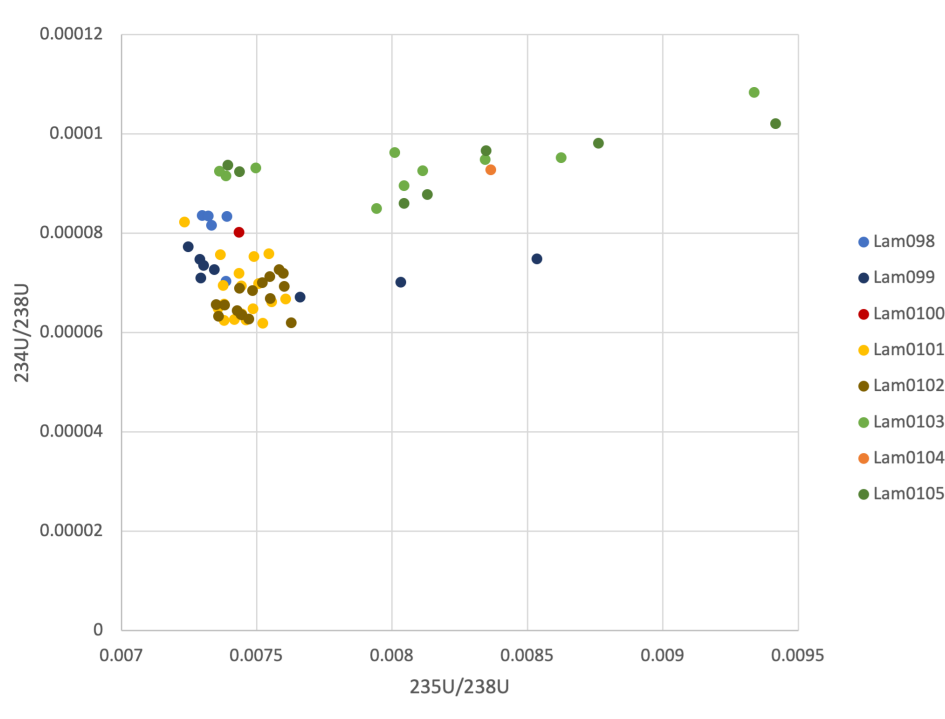


Figure 6. Uranium $^{235}\text{U}/^{238}\text{U}$ ratios versus uranium $^{234}\text{U}/^{238}\text{U}$ ratios for all sediment core and surface sediment grab samples, calculated from ICPMS results; a positive correlation is evident. The sediment core and surface sediment grab samples were collected from the three ponds in Piketon, OH within the vicinity of the Portsmouth Gaseous Diffusion Plant, which historically enriched uranium, and analyzed in a lab using an ICPMS instrument to produce uranium isotope concentration data. For the sediment core data, each data point on the graph is a different 3cm depth interval.

vertex, there is an increasing anthropogenic influence on the U concentration. Because the plots have points across this range, it is likely that the uranium observed in the samples is a mixture of naturally occurring and enriched uranium from the Portsmouth plant. When the $^{235}\text{U}/^{238}\text{U}$ values were graphed against the $^{234}\text{U}/^{238}\text{U}$ values for all samples, a positive relationship was found in that as $^{235}\text{U}/^{238}\text{U}$ values increased, so did $^{234}\text{U}/^{238}\text{U}$ values (Figure 6). However, an even stronger positive correlation was observed for the plot of $^{235}\text{U}/^{238}\text{U}$ versus $^{236}\text{U}/^{238}\text{U}$ (Figure 5). The ^{236}U isotope is mainly of synthetic origin, as it is produced by neutron irradiation of ^{235}U ; uranium recovered from plutonium production reactors, referred to as “recycled uranium,” is known to contain ^{236}U and this material was processed at the Portsmouth Gaseous Diffusion Plant⁵. The obvious strong correlation between elevated $^{235}\text{U}/^{238}\text{U}$ and the detection of nonzero $^{236}\text{U}/^{238}\text{U}$ ratios, with the highest values being from the Dutch Run pond, further suggest that this contamination deviates from nature and is a direct result of the activities at the Portsmouth Gaseous Diffusion Plant (Figure 5).

Several limitations are included in this study that could be alleviated with further research efforts. First and foremost, the sediment samples that were collected are arguably not of the highest quality and more accurate, extensive sediment core extraction methods

popular in the soil coring field could have been employed. The ponds that were sampled also may have experienced confounding influences unknown to the researchers; local knowledge that the ponds had not been dredged since the construction of the Portsmouth Gaseous Diffusion Plant may not be entirely accurate. Despite this, the samples that were obtained produced results that tell a compelling story and clearly exhibit trends of enriched uranium pollution. Another method that would improve upon this study is establishing a chronology. Previous studies have established sediment core chronologies by measuring the 1952 appearance and 1963 peak activity of plutonium in individual sediment layers¹². Due to plutonium being a component of open atmosphere nuclear testing during the Cold War and subsequent tropospheric fallout, low concentrations of plutonium are found non-uniformly in small quantities around the world from deposition¹³. With more time and resources, establishing a chronology via plutonium dating could greatly strengthen the findings of this study.

5 CONCLUSION

Detection of pervasive enriched uranium in sediment samples from the Piketon, OH community has shined a light on the dark truth of the pollution in the area. It

is evident that enriched uranium from the Portsmouth Gaseous Diffusion Plant has been deposited in the air, soils, and water of the surrounding community and has persisted long since the closure of the facility. The findings of this study will provide the citizens of Piketon scientific evidence that will facilitate their understanding of the uranium contamination issue in their community. Piketon locals have immense concerns about the health impacts of this uranium pollution (as seen on their CARD, or Citizens Advocating for Responsible Disposal, in Pike Facebook page), and therefore solid scientific data detailing the extent of the enriched uranium contamination will hopefully provide further insight into how pressing the health hazards are and help those in charge implement a thorough and timely plan of action for cleaning up the pollution.

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7 EDITOR'S NOTES

This work was adapted from a senior thesis and has been condensed for publication. Contact DUURJ staff for the full publication.

REFERENCES

- [1] World Nuclear Association. Uranium Enrichment (2019).
- [2] Energy.gov. Portsmouth Site.
- [3] Stearns, D. *et al.* Uranyl acetate induces hprt mutations and uranium–DNA adducts in Chinese hamster ovary EM9 cells. *Mutagenesis* **20**, 417–423 (2005).
- [4] Wanger, S. *et al.* Groundwater uranium and cancer incidence in South Carolina. *Cancer Causes & Control* **22**, 41–50 (2011).
- [5] Ketterer, M. & Szechenyi, S. Investigation of anthropogenic uranium, neptunium, and plutonium in environmental samples near Piketon, Ohio (2019).
- [6] Ketterer, M. & Szechenyi, S. Uranium isotopes in samples from Zahn's Corner Middle School (2019).
- [7] Bobo Road Pond, Bailey Chapel Pond, Dutch Run Pond. Google Maps. URL maps.google.com.
- [8] Horwitz, E. *et al.* Separation and Preconcentration of Uranium from Acidic Media by Extraction Chromatography. *Analytica Chimica Acta* **266**, 25–37 (1992).
- [9] Elemental Scientific. apex HF Desolvating Nebulizer. URL <http://www.icpms.com/products/apexHF.php>.
- [10] Agilent Technologies. ICP-MS: A Primer (2005).
- [11] Stirling, C., Anderson, M., Potter, E. & Halliday, A. Low-temperature isotopic fractionation of uranium. *Earth and Planetary Science Letters* **264**, 208–225 (2007).
- [12] Ketterer, M., Hafer, K., Jones, V. & Appleby, P. Rapid dating of recent sediments in Loch Ness: inductively coupled plasma mass spectrometric measurements of global fallout plutonium. *Science of the Total Environment* **322**, 221–229 (2004).
- [13] Simon, S., Bouville, A. & Beck, H. The geographic distribution of radionuclide deposition across the continental US from atmospheric nuclear testing. *Journal of Environmental Radioactivity* **74**, 91–105 (2004).