

A. Introduction

The Portsmouth Gaseous Diffusion Plant was built in 1952, and began operations at the end of 1954, to increase the government's capacity to produce highly enriched uranium for naval reactors and the burgeoning nuclear power industry. Two gaseous diffusion plants (GDP), at Paducah (PGDP) and Oak Ridge (OGDP), were already operating at the time. Now, only one is operating, the Paducah plant. In 2001 the United States Enrichment Corporation (USEC), the present operator, placed the Portsmouth plant on cold standby. Meanwhile, the Department of Energy (DOE) is presently engaged in a major remediation effort to correct contamination caused by past operations. The site is located in the southern part of Ohio (see Fig. 1) and is quite large, covering an area of about 3,700 acres.

The purpose of this report is to review the present and past environmental record of the Portsmouth facility, focusing on groundwater and the facilities on the east side of the site. To prepare this report, we reviewed a large number of federal and contractor reports and letters, some recently released under the Freedom of Information Act (see Bibliography), Ohio EPA reports and letters, toured the Portsmouth site, performed some independent calculations and discussed some issues with former workers. This work was sponsored by two public interest groups, PRESS and the Uranium Enrichment Project; who were extremely helpful in assisting us in this report. The report has a different and larger perspective than reports by federal and state agencies and their contractors, nor are we bound by political and financial considerations. But our review is necessarily limited by the data collected. We have not independently sampled soil, groundwater, sediment and air. This report also suffers from the fact that not all radionuclides and volatile organics, and all pathways were monitored by the DOE and its predecessors in the earlier years.

While this report reviews the environmental impact of the entire Portsmouth GDP (PORTS), we focus on groundwater and those facilities that are associated with releases of technetium-99 (Tc-99), transuranics, and the hazardous chemical trichloroethylene (TCE), such as the oxide conversion facility X-705E and the 701B Holding Pond. As will be discussed later, for remediation purposes and as a description of groundwater regimes, the site is divided into four Quadrants. X-705E and X-701B are located in Quadrant II on the eastern side of the site. Another contributor to a groundwater plume of contaminants is the X-749 landfill in Quadrant I, located on the southern side of the site (see Fig. 2). As we discuss in this report, the DOE has a major remediation effort underway to interdict, pump and treat groundwater plumes. We do not discuss fluoride contamination to the air and water, even though this might be an important environmental impact.

Because of the use of depleted uranium in Kosovo and Iraq, there has recently been heightened interest in some of the impurities brought to the GDP by returned or recycled uranium. Recycled uranium (RU) contains U-236, technetium-99 (Tc-99) and transuranic elements (TRU), such as neptunium (Np) and plutonium (Pu). Some of these impurities may be drawn out of the GDP with depleted uranium, and then be shaped into armaments. This matter is also discussed in this report, but is not our major focus.

The basic function of a gaseous diffusion plant (GDP) is to enrich uranium in the fissionable isotope U-235, but as a byproduct, toxic and radioactive materials are released into the air and water. The GDP process involves passing uranium in gaseous form, uranium hexafluoride (UF₆), under pressure, through over 4,000 sieves or stages that selectively pass the lighter isotope U-235 and restrain the heavier isotope U-238. Thousands of motors must be cooled and maintained, necessitating lubricants and chemicals such as degreasers (TCE). The cooling water contained the toxic metal, hexavalent chromium. As a result of introducing recycled uranium into the GDP, miles of pipes and tons of other equipment were coated with radionuclides, such as technetium-99. When the pipes and equipment were dismantled and brought to Building 705 for decontamination, Tc-99 escaped into the X-705 building.

Also, in the past DOE operated an incinerator near the X-705 building. For most, but not all, effluent sources, filters, scrubbers and resins were used to capture effluents before they reached the environment, but this material would then be buried in landfills that inevitably leaked and now occupy a sizable percentage of the plant area. For an operation this size, it was inevitable that toxic chemicals and radionuclides would be released to the environment, in the air and the water. However, since the site is so large, about 6 square miles, it may be years before some of the toxic or radioactive materials actually reach the plant boundary through groundwater, though the groundwater from the site has contaminated surface water off-site.

As we discuss in this report, the Department of Energy at Portsmouth is making progress in attacking contaminated facilities and soil that exist on the site. Progress on groundwater contamination, and the ultimate status and use of the site, however, remain areas of concern, and now a more pressing concern with the recent plan to convert 340 acres of the site to industrial use. Active maintenance will be required for the indefinite future.

It is also important to point out that DOE and local residents differ on the impact of the Portsmouth plant on health. Local residents have been and still are concerned about releases from the plant and the effect on their health. Residents feel that the local cancer rate is excessive. The Department views present soil and water concentrations as indicative of the minor impact the plant has had on the environment. The response by the DOE has not allayed local concerns. In response, or non-response, the Department points out how it is cleaning up the site and the "fact" that radioactive and toxic materials have not moved off-site. But present soil and water radionuclide concentrations would not explain an increased cancer rate. Depending on the location within the human body, hard cancers have a latency period of 10 to 20 years. That is, an elevated cancer rate, if one exists, is likely due to releases in the '60's and '70's, when the concentrations of radioactive and toxic materials in the air and water were likely much greater. In many cases, the Department did not sample for these concentrations at the time and we may never know how much uranium or technetium was released. Current surface soil samples, if undisturbed, sometimes provide a

clue on past releases. As we discuss later, it is likely that radioactive materials were released off-site from the Portsmouth plant.

In addition, the amount of contaminated soil in general has been grossly underestimated. At different times, Tc-99 and TRU have been detected in and around various facilities in Quadrant II, and have seeped via ground and surface water to on-site drainage ditches and eventually to Little Beaver Creek, Big Beaver Creek, Big Run Creek, and the Scioto River, presenting health risks for an even larger community than was expected.

In Sections B and C we discuss the historical background and site setting of the PORTS facility. In Section D we discuss in detail the gaseous diffusion process and facilities within the four quadrants of the Portsmouth site and how Tc-99, transuranics and TCE arise at Portsmouth. In Section E we discuss groundwater contamination in the four quadrants of the Portsmouth site, focusing on Quadrant II. Section F has a brief discussion of off-site contamination, the health implications of earlier radioactive and toxic chemical releases from PORTS, and DOE's cleanup strategy. Section G contains our conclusions and recommendations.

B. Historical Background

Peter Kiewit and Sons constructed the Portsmouth Gaseous Diffusion Plant in Piketon, OH beginning in 1952, and operations began in the end of 1954. PORTS' original mission was to enrich uranium, in the form of UF₆ (feed material), to assays up to 97% U-235. The highly enriched uranium was used in military applications, particularly in nuclear weapons production and naval nuclear propulsion programs. Enriched uranium with assays of 3% to 5% U-235 was removed as a side product for use in light water reactors [1, 2]. In more recent years, PORTS only produced low enriched uranium for use as commercial reactor fuel. At present, the plant is on cold standby. The Bush administration is trying to decide what to do about USEC, including PORTS. The intent of the administration is unknown, although it appears to be leaning towards keeping PORTS on standby. At the same time, the Ohio congressional delegation and governor are lobbying DOE to keep the plant open, at least for R&D purposes.

Most of the UF₆ feed that PORTS processed came from the Paducah Gaseous Diffusion Plant, Oak Ridge Gaseous Diffusion Plant, PORTS' own feed manufacturing plant, and commercial customers. From 1958 to 1962, PORTS produced its UF₆ feed material from uranium tetrafluoride (UF₄, green salt) in the X-344 Feed Manufacturing Plant. Another source of UF₆ feed for PORTS was recycled uranium converted to UF₆ in the X-705E Oxide Conversion Facility from 1957 to 1978 [3].

1 USDOE, *Independent Investigation Vol. 1*, sec. 1.2, 2.2.

2 M Resnikoff, *Cost/Benefits of U/Pu Recycle*, R-804948, Oct 11, 1982.

3 USDOE, *Independent Investigation Vol. 1*, sec. 1.2, 2.2.

Until 2001, PORTS and the Paducah Gaseous Diffusion Plant were the two operating uranium enrichment facilities in the United States that concentrated or enriched uranium in the isotope U-235 [4]. Separation and enrichment occur as a result of pumping and compressing uranium hexafluoride (UF₆) through sieves at closely controlled pressure and temperature through several thousands of stages known as cascades.

Natural uranium contains approximately 0.7% U-235. The Paducah facility enriched the U-235 concentration in uranium to 2-2.5%, and then shipped it to the Portsmouth Gaseous Diffusion Plant (PORTS) for further enrichment. PORTS increases the U-235 content to 3% to 5%, used as raw material for uranium pellets used in fuel elements of nuclear reactors. In the past, PORTS also enriched uranium to a U-235 concentration as high as 97%, which was used as fuel for nuclear vessels and atomic weapons, but production of this highly enriched uranium (HEU) was discontinued beginning in 1991 [5, 6]. While the Portsmouth plant is on cold standby, DOE intends to have the Paducah GDP enrich U to 3 % and more for use in commercial reactors.

Plant construction at PORTS began in late 1952, shortly after the Atomic Energy Commission (AEC) chose the site. Enrichment operations began in 1954 while construction was still underway. Construction was not completed until early 1956 [7]. The original contractor, Goodyear Atomic Corporation (GAT), operated PORTS under contract with the AEC (and its successor agencies, Energy Research and Development Administration (ERDA) and the US Department of Energy (DOE)) until 1986. Lockheed Martin Energy Systems (LMES), formerly Martin Marietta Energy Systems (MMES) took over from GAT and operated the facility from 1986 to 1998. In April 1998, Bechtel Jacobs Corporation (BJC) was named as DOE's management and integrating contractor for PORTS [8].

In the late 1970's, PORTS was selected as the site for a new uranium enrichment facility that would employ the gas centrifuge technology. Construction of the Gaseous Centrifuge Enrichment Plant (GCEP), located southwest of the existing gaseous diffusion plant, began in 1979 but was halted in 1985 [9].

On July 1, 1993, control of PORTS passed from DOE to the United States Enrichment Corporation (USEC), a wholly owned government corporation created by Congress in an attempt to transform the DOE's uranium enrichment enterprise into a profitable business. Under this arrangement, DOE leases to USEC the gaseous diffusion facilities, while DOE

4 There was another uranium enrichment plant in Oak Ridge, TN, called the Oak Ridge Gaseous Diffusion Plant. It was put on standby in 1985 and finally closed in 1987.

5 Theta Technologies, Inc. (Theta Tech), *Recycled Uranium Mass Balance Project, Portsmouth, Ohio Site Report*, BJC/PORTS-139, Apr 14, 2000.

6 Memo from M Moore to A Mazzocchi, *Workplace Health Problems*, May 30, 1979.

7 Theta Tech.

8 US Department of Energy (USDOE), *Independent Investigation of the Portsmouth Gaseous Diffusion Plant, Volume 1*, Office of Oversight, May 2000, sec. 1.2, 2.1.

9 Theta Tech.

retains responsibility for environmental restoration and management (treatment, storage, and disposal) of wastes resulting from enrichment operations prior to July 1, 1993. USEC, which also operates the Paducah facility, was later privatized on July 28, 1998 [10].

USEC announced it would cease enrichment operations at PORTS in July 2001; enrichment actually ceased two months earlier in May. The plant is presently on cold standby; DOE intends to conduct research on new enrichment technologies at the site. As for DOE's environmental management program at PORTS, long-term surveillance, maintenance, and institutional controls will continue indefinitely. Future use of the site has been the subject of discussion among stakeholders. Maintenance of industrial land use within Perimeter Road and mixed industrial/commercial use for areas outside of Perimeter Road but still within the DOE site boundary is preferred. Potential recreational use of land outside Perimeter Road is being considered. Development of the PORTS site for future residential land use has not been recommended [11].

C. Site Setting

Geographical Setting

The Portsmouth Gaseous Diffusion Plant is located in Pike County, in the south central region of Ohio. It is approximately 80 miles south of Columbus, 2 miles east of the Scioto River. The industrialized portion of PORTS, consisting of more than 100 plant buildings, occupies approximately 1,000 acres of a 3,700-acre DOE-owned site near the Village of Piketon [12]. Almost 120 acres of the site now consist of landfills.

Pike County has a population of approximately 23,000 residents. Within the county, there are several communities that lie within a few miles of the plant, such as Piketon, Wakefield, and Jasper. Piketon is the residential center nearest to PORTS, and is located about 5 miles north of the plant on US Route 23. It has a population of about 1,700. Waverly, Pike County's largest community, lies 10 miles north of the plant and is populated by about 5,100 residents.

Several residences are adjacent to the southern half of the eastern boundary of the plant and along Wakefield Mound Road (old US 23), directly west of the plant. Two nursing homes, with a combined capacity of 60 persons, are located along Wakefield Mound Road.

10 SI Schwartz, *US Nuclear Weapons Research, Development, Testing, and Production, and Naval Nuclear Propulsion Facilities*, Oct 12, 1999.

11 *Portsmouth Gaseous Diffusion Plant Quadrant I CAS/CMS Final Report*, Mar 29, 2000, p. ES-1.

12 Theta 00, p. 13.

Below the Cuyahoga shale is the Sunbury formation, composed of black, carbonaceous, fissile shale. It is approximately 20 ft thick on the eastern portion of the PORTS site, but is absent on the western side. Where present and more than 4 ft thick, the Sunbury shale forms a confining layer for the Minford silt layer and the Berea sandstone.

The Berea sandstone is continuous across the PORTS facility, with a thickness of about 30 ft. It underlies the Sunbury shale on the eastern portion of the site, and the Minford and Gallia layers on the western portion of the site. Its upper 20 ft is composed of light-gray, hard, thickly bedded, fine-grained sandstone, while the lower 10 ft has interlayered shale laminations and very similar to the underlying Bedford formation.

The Bedford formation consists of thinly bedded shale with laminations of hard, grayish, fine-grained sandstone and siltstone. Approximately 100 ft thick, the Bedford is continuous across the PORTS site.

The water-transmitting layers at PORTS are the Gallia sand and the Berea sandstone. The Gallia aquifer is considered the primary groundwater migration pathway, with a hydraulic conductivity ranging from 0.11 to 150 ft/day (mean: 3.4 ft/day). The Berea aquifer has a hydraulic conductivity ranging from 0.0045 to 15 ft/day, with a mean value of 0.16 ft/day. Groundwater in the Gallia sand and Berea sandstone formations that underlie PORTS are not reportedly used as domestic, municipal, or industrial water supplies.

PORTS receives its water from the X-608, X-605G, and X-6609 well fields, which tap the Scioto River Valley buried aquifer. Groundwater contamination at the PORTS site reportedly has not affected this aquifer, and is limited to the on-site groundwater aquifer. In the documents that we reviewed, we found no information that implied that the Scioto River Valley Aquifer has been contaminated. However, nearby residents that do not receive their water from the municipal water supply have drinking water wells that draw water from the aquifer on which the site is located.

Groundwater Flow by Quadrant

For purposes of classifying areas of the site that require remediation, the DOE compartmented the site into four quadrants, QI – QIV. See Fig. 2. These quadrants are also related to the direction of ground and surface water flow. QI, generally the southern section of the Portsmouth site, contains the Peter Kiewit and low-level waste landfills, X-749. Surface water drainage in the landfills area is controlled by Big Run Creek, to the east and south of the landfill, and by several smaller drainage ditches that discharge to Big Run Creek. Big Run Creek flows in a westerly direction into the Scioto River. Ground water flow in QI is primarily to the south, towards Big Run Creek.

QII includes the 701B Holding Pond and the East Drainage Ditch. Surface drainage is to the drainage ditch that empties into Little Beaver Creek. Ground water flow in QII is to the

east and Little Beaver Creek. Little Beaver Creek runs north and west and empties into Big Beaver Creek that runs south into the Scioto River. See Fig. 2.

QIII includes the two main process buildings X-326 and X-330. Both surface and ground water flow west towards the West Drainage Ditch and several holding ponds. QIV is the northern discharge region. Surface and ground water of this Quadrant discharge to Little Beaver Creek in the north.

Background Radiation

The area surrounding Portsmouth Gaseous Diffusion Plant is mainly agricultural. Other businesses in the vicinity include lumberyards, asphalt-gravel companies, and the State Highway Department office and garage. None of these facilities is expected to have any significant environmental impact. Also, no major nuclear facilities are located in the area aside from PORTS [21].

Uranium in soil attributable to background is 1.4 ± 0.79 pCi/g U-238 for Ohio. Background concentrations of uranium in air for January to June 1985 were $2.3 \pm 0.8 \times 10^{-19}$ microcuries per milliliter ($\mu\text{Ci/mL}$) U-235 and $33.3 \pm 4.2 \times 10^{-18}$ $\mu\text{Ci/mL}$ U-238, based on USEPA measurements at a Columbus, Ohio monitoring station. 1985 measurements at the Scioto River upstream from the PORTS' discharge point yielded background activities of $<0.14 \times 10^{-8}$ $\mu\text{Ci/mL}$ (detection limit) to 1.35×10^{-8} $\mu\text{Ci/mL}$ with an average of $<0.91 \pm 0.16 \times 10^{-8}$ $\mu\text{Ci/mL}$ for gross alpha (combined α radiation) and $<4.50 \times 10^{-8}$ (det. limit) to 5.86×10^{-8} $\mu\text{Ci/mL}$ with an average of $<4.54 \pm 0.6 \times 10^{-8}$ $\mu\text{Ci/mL}$ for gross beta. Uranium in water ranged from <1 $\mu\text{g/L}$ (detection limit) to 23 $\mu\text{g/L}$ with an average of $<2.2 \pm 1$ $\mu\text{g/L}$. [22]

Tc-99 and neptunium-237 are not naturally found in the environment and their background concentrations are therefore below detectable limits. But as we discuss later, both radionuclides are found in the Portsmouth environment.

D. Process and Facilities

In the following, the gaseous diffusion process and most important facilities at PORTS are explained, together with the possible related problems within each quadrant. Much of the information is based on a report by Mary Davis [23]. As an overview, uranium is enriched in the isotope U-235 by passing uranium hexafluoride, a gas, through a series of compressors and sieves. To do this, uranium must first be converted into hexafluoride form. The cascades must be cooled and maintained. Contaminants and uranium must be filtered out

21 USDOE, *Environmental Survey Preliminary Report*, p. 2-1.

22 USDOE, *Environmental Survey Preliminary Report*, pp. 3-1, 3-17, 3-31.

23 Davis MB., *A Guide to Key Facilities and Sites at the Portsmouth Gaseous Diffusion Plant*, October 2000.

before air is released. Wastes materials, such as sludges and filters, are generated and disposed of on the site. From this large complex, which processed over 100,000 tons of uranium, including uranium recycled from reactors containing Tc, Np and Pu, considerable amounts of radioactive contaminants and TCE, a degreaser, have entered the air and groundwater.

Quadrant III

The Cascades: X-330, -333, -326 (1954-present)

The enrichment of U-235 occurs as UF₆ passes through thousands of stages. These stages are a series of compressors, heat exchangers, control valves and motors, converters, and supporting pipes arranged in stages, cells, and units that progressively enrich the UF₆ feed. The barriers in the converters favor isotopes of lower molecular weight (U-235 as opposed to U-238) in the UF₆ to pass through. This stream becomes slightly enriched in U-235 at each stage from the feed point to the top of the cascade. Conversely, there is another stream that becomes depleted in U-235 from the feed point to the bottom of the cascade. Both the enriched product and the depleted tails are fed into cylinders and allowed to cool until solid. The product is then sent to other DOE sites and fuel fabricators for further processing, while the tails are re-fed to the cascade or stored on site [24]. Some tails are converted to metal.

The Cascades are composed of 4,080 stages of process equipment distributed over 3 buildings: X-333 (Q IV), X-330, and X-326 (both Q III). X-333 and X-330 were built in 1955. X-333 was used for the initial phase of uranium enrichment. X-330 was used for the intermediate enrichment of uranium. X-326 was built in 1956 and used for the high-enrichment phase. In 1991, production of highly enriched uranium ceased; the X-326 building is used only for product withdrawal and side feeding. From early 1997 to mid-1998, the X-326's product withdrawal equipment was also used for downblending highly enriched uranium [25].

The PORTS cascade is capable of enriching uranium to 97% U-235. The side purge cascade at PORTS consists of 2 high-speed cells, 1 intermediate-speed cell, and 2 low-speed cells. They are responsible for removing the lightweight molecular gases or "lites", such as air, F₂, and HF from the cascade, and separating them from the UF₆ gas stream. After lites separation in the side purge, trace quantities of U and Tc in the lites stream are reduced further by flowing through chemical traps before they (lites) are discharged into the vent stack and out to the atmosphere [26].

24 USDOE, *Independent Investigation Vol. 1*, sec. 2.2.

25 Theta Tech, p. 15.

26 RL Faulkner, *Trip Report on PORTS Purge Cascade High Emissions*, GDP-0460, OR0040885, Aug 30, 1993.

Both natural uranium and recycled uranium have been introduced to the PORTS cascade. Natural uranium contains the U isotopes U-234, U-235 (0.7% by wt) and U-238 (99.2% by wt), but no transuranics or fission products. First converted to UF₆, and then introduced into the cascade stages, the lighter isotopes U-234 and U-235 move more easily through a succession of sieves, and their concentration increases towards the top of the cascade. Commercial nuclear reactors require an enrichment of 3% to 5% U-235. U-238 is removed as depleted uranium with U-235 assay 0.3%.

During its time of operation, a total of 320,817 MTU were fed to the PORTS cascades between 1955 and 1999, 1,086 of which were recycled uranium from nuclear reactors [27]. 796 MT of this recycled uranium were received from Paducah, whereas the remaining 290 MTU came from other sources, such as Oak Ridge, Fernald, the Division of International Affairs, USAEC, Babcock & Wilcox, NUMEC, Allied Chemical Corporation, K-25, Y-12, France, Germany and the United Kingdom. We are uncertain about the fate of the hundreds of thousands MT of recycled uranium that were processed at Paducah. To our knowledge, the majority of the uranium that was processed at Paducah was subsequently shipped to PORTS for further enrichment. In this context, we are surprised about the comparably very low number of 1,124 MT recycled uranium that were reportedly received at PORTS (38 MTU were either stored on site or shipped away). The recycled uranium arrived at PORTS in the chemical forms of UF₆, UF₄, UNH, U₃O₈, UO₃, UO₂ and UNH [28].

The top purge cascade at PORTS consists of 3 intermediate-speed cells and 12 low-speed cells. The top purge cascade separates the intermediate molecular weight gases (such as R-114 or freon, ClF₃, etc.) from the UF₆ gas stream. Trace quantities of radionuclides, such as Tc-99, are also reduced further by flowing through chemical traps before the intermediate gases are discharged into the vent stack for release to the atmosphere.

According to Makhijani [29], the chemical traps used in the top purge cascade consisted of activated alumina and had an efficiency of 90 % for uranium. For comparison, the traps used in the Oak Ridge plant used a potassium hydroxide scrubber with 99 % efficiency. The top and side purges were the largest source of monitored uranium releases.

Intentionally introduced into the top purge is a dry air bleed to facilitate the upward flow of the intermediate molecular weight gases out the top purge and prevent overloading the intermediate speed cells from very high concentrations of R-114, a refrigerant [30].

In X-326, after cells were washed and serviced, a wet air evacuation system was used to transfer wet air from cells to the atmosphere, before UF₆ was reintroduced to the cells.

27 Theta Tech 2000, p. 86.

28 Theta Tech 2000, p. 83.

29 Makhijani A, "Preliminary Estimates of Emissions of Radioactive Materials and Fluorides to the Air from the Portsmouth Gaseous Diffusion Plant, 1954-1984," October 14, 1992, p. 3.

30 Faulkner.

According to a 1985 vent committee investigation, the potential for loss of highly enriched uranium from this system was “very high” (Makhijani *et al*).

Possible releases to the air include uranium, technetium, transuranics, hydrogen fluoride, fluorine, chlorine, sulfur dioxide, SO₂F₂, Freon 114, ClF₃ and CF₄. Chemical traps were installed to capture these gases, but they did not completely succeed in doing so. Pipes and other parts of the cascades were coated with Tc. When these cascade parts were replaced and decontaminated at the X-705 Decontamination building (see below), liquid wastes containing technetium, transuranics and VOC resulted and contaminated drainage areas on the east side of the site. Solid wastes emerged from the cascades in form of spent Alumina, NaF and MgF₂ traps, and absorbent material soaked with PCB's. The solid wastes were deposited in several landfills, depending on their contents. Technetium-contaminated material was predominantly disposed of in the X-749 landfill.

Tc-99: A Special Concern

Recycled uranium contains an additional uranium isotope U-236, fission products and transuranics, such as plutonium and neptunium. In reacting UO₃ or UF₄ with fluorine gas, UF₆ is created, vaporized and collected in cold traps. For the most part, the reaction process at the conversion stage separates uranium from these other impurities, which generally remain in the ash at the conversion plant. The fission product technetium is the exception; like uranium, it forms a hexavalent compound and vaporizes at about the same temperatures. Introduced to the cascade stages, it coats metal surfaces and moves rapidly to the high end. GDP operators remove Tc-99 using MgF₂ traps at side traps. This captured Tc-99 must then be stored or put into landfills at Portsmouth. The Department of Energy has made the following quantitative assessment of the movement of Tc-99 within PORTS [31]. Approximately 15% of Tc-99 remained in the feed plant and cylinder heels. Of the remaining amount introduced to the Cascades, approximately 2/3 was retained in the piping or the MgF₂ filters. The remainder exited the facility as enriched product, though a small percent was released to the air through the side purges. The DOE reports do not estimate how much of the captured or retained Tc-99 was subsequently released to the environment in the X-705 facility when the Cascades piping was opened for CIP/CUP improvements in the 1970's, or how much Tc-99 was buried in landfills, but 57% of admitted Tc represents a large inventory.

Unlike Tc-99, 75% of Np was removed in the feed plant and cylinders, and only 25% was introduced to the cascades. Very little, less than 1 %, remained in the final product. For plutonium, almost all remained in the ash at the conversion plant. The DOE estimates 99.85% to 99.97% remained in the ash that was subsequently buried in the landfill [32].

31 USDOE, *Recycled Uranium Project Report*, Ohio Field Office, May 15, 2000

32 USDOE, *ORGDP Recycled U Report*, Fig. 5.1-2

decontamination process, Tc-99 would have been released to the air and the east drainage ditch, as occurred during the 1970's, but monitoring for Tc-99 did not begin until 1975, so records are not available. A major change-out of cascade equipment occurred beginning in the mid '70's, accounting for much higher environmental concentrations.

From FY 1968 to FY 1969, 568 MT of recycled uranium were received from Paducah. 168 MT were fed to the cascade in FY 1970 (Oct-Nov 1969) while the rest (400 MT RU) was fed in January 1974. The 400 MT of recycled uranium was converted to UF₆ in the X-705 facility [38]. During this conversion process, Pu was released to the air or was present in the ash. Little Pu would actually have been introduced to the GDP stages.

A major equipment change-out was begun in FY 1973 and was not completed until FY 1983. Process equipment and piping were removed, decontaminated in the X-705 facility, and changed as part of the cascade improvement program. The Cascade Improvement Program/Cascade Upgrade Program (CIP/CUP) was also associated with higher gross alpha air concentrations, as seen in Fig. 4. The process of opening and decontaminating piping coated with Np, Pu and Tc-99 led to major contamination in the X-701B holding pond. An estimated 12.8 g Np and 1.2 mg Pu were removed from the cascades. We do know that the beta/gamma readings in monitoring wells near the east drainage basin X-701 greatly increased during this period. For example, monitoring location 8 had the following readings: yr 1973, 2,550 pCi/L; yr 1974, 17,478 pCi/L; yr 1975, 150,000 pCi/L; yr 1976, 3,400 pCi/L. Sampling locations 7 and 11 had similar increases during CIP/CUP. If it is assumed that these increases are entirely due to Tc-99, in yr 1975, the levels are 40 times over the USEPA drinking water standard of 4 mrem/year [39].

PORTS used MgF₂ side-stream traps to reduce technetium in the upper cascades. These traps allowed the selective removal of volatile technetium from the UF₆ gas stream [40]. However, only one set of traps was being used. When they became saturated, they were taken to the X-705 building for cleaning. Meanwhile, the cascade was operating with no traps. A 1980 memo from Engineering requested that another set of MgF₂ side-stream traps be placed at X-326 [41]. The more highly enriched uranium and Tc-99 would be found in X-326. One year later, no additional traps had been installed. As far as we are aware, even up to 1993, the additional traps were not installed [42]. A 1981 memo emphasized the need for additional technetium traps at the X-326 side purge cascades, as technetium-associated problems and costs had been mounting. For one, technetium caused plugging of cascade equipment. This resulted in additional costs associated with cell maintenance activities, such as equipment removal, cleaning and recharging the MgF₂ traps, separation and disposal of spent ion-exchange resins used in Tc-99 removal, and servicing by Health Physics and Laboratory Analysis groups. Also, vent emissions exceeded recommended airborne effluent

38 USDOE, PORTS Recycled U Report, App. II

39 Based on a water intake of 2 L per day, 4 mrem/year are reached with a concentration of 3749 pCi/L

40 DW Simmons, , p. 18.

41 Memo from JA Weber to RD Bush, *Technetium trapping of "side stream"*, GAT-841-80-75, July 3, 1980.

42 GDP-0460, August 30, 1993.

radionuclide concentrations. According to EIS data (ERDA-1555), Tc venting from the cascades was up to 5.826 Ci/yr, which was unacceptable to GAT and the DOE. EPA calculations showed that a release of only 1 Ci Tc-99/year would lead to unacceptable air concentrations at the fence post [43]. Additional equipment had to be provided to monitor these vent losses [44]. A DOE draft environmental survey [45] stated that uncontrolled releases of Tc-99 were known to take place, because only partial trapping with MgF₂ traps takes place when the Tc-99 bubble flows through the cascade. This was confirmed in a Los Alamos National Laboratory (LANL) report [46], according to which breakthrough of Tc-99 occurs when the flow rate of radioactive gases through chemical traps exceeded the recommended rate. The report also stated that such large quantities of radioactivity paralyzed the ionization chambers of the space recorders that were used to measure losses of Tc-99 and TRU, and that the downtime of the space recorders was high. In addition, MgF₂ pellets of high quality for Tc-99 traps were rare at PORTS due to manufacturing problems [47]. The original MgF₂ traps that were used to prevent the Tc-99 from being vented to the atmosphere had a decreased ability to absorb Tc-99 at very low levels and allowed a constant bleed-through of about 1 ppm [48].

During the HEU shutdown program, stack emissions from the cascades increased from 0.04 to 0.23 mrem/year between 1986 and 1991, and to 0.87 in the first half of 1993, mainly due to Tc-99 recovered from the X-326 cells [49]. Again it was noted that most of the emissions occurred from discrete, single events (“burps”). In the first half of 1993 alone, at least 10 “burps” were measured, with the largest one emitting 100 g of Tc-99 on June 23 [50]. Since PORTS monitored the stacks only by taking grab samples every two hours until the installation of a continuous monitoring system in September 1993, it is probable that several “burps” remained undetected.

According to Makhijani, the total amount of Tc-99 released to the atmosphere and surface water by 1983 was estimated between 57.89 and 405.87 Ci [51], as opposed to the 212.85 Ci reported by PORTS [52].

43 Till, J, et al, *Assessment of 99Tc Releases to the Atmosphere – A Plea for Applied Research*, ORNL-TM-6260, June 1978.

44 Memo from Dept. 841 to RR Miller, *Technetium trapping of side stream, X-326*, GAT-841-81-216, July 13, 1981.

45 USDOE, *Draft Environmental Survey 1986*, p. 3-9

46 Picard et al, *Study of Waste Streams at the Portsmouth Gaseous Diffusion Plant (Q4/84-219)*, Los Alamos National Laboratory, April 1984, p. 19.

47 Faulkner, 1993, p. 11

48 Simmons, 1996, p. 18

49 R. Faulkner, Internal Correspondence, Martin Marietta Energy Systems, August 30, 1993

50 Faulkner, 1993, p. 5

51 Makijani 1992, p. 27.

52 BJC/PORTS-139, p.50.

According to DOE, the UF₄ that was fed to the reaction towers was all “fresh” or “virgin” uranium. None of the UF₄ was recycled reactor material. However, the X-344 feed manufacturing plant was contaminated with Tc-99 and TRU, which could only have come from recycled reactor material. DOE offered two explanations. First, there was a time, specifically from August 1958 to October 1961, when purge gas from the cascades was processed in X-344 through the cleanup towers to recover excess F₂. Filter plugging, presumably due to Tc-99 contamination, was frequently observed in the cleanup reactor system, following initiation of fluorine recovery operations using the purge gas. Second, waste liquids from X-344 that were sent to X-705 for uranium recovery and then returned to X-344 as uranium oxide were contaminated with TRU by the X-705 uranium recovery system [58]. Prior to 1970, a large amount of Tc-99 was introduced to the PORTS facility, mostly in the form of UF₆, and therefore not introduced directly into the X-344 facility; but other sources, such as UNH from ORGDP, had to be converted to a gaseous form at X-344.

The grinder that was used to grind the ash prior to its blending with fresh UF₄ had a high rate of mechanical failure and leaks. When operated or repaired, it produced considerable dust and resulted in extensive airborne contamination.

When the plant closed in 1962, all material inventories were removed from the system, containerized, and moved to storage for final disposition. The remaining ash from X-344, about 23.3 MTU, was removed, pulverized, containerized, and sent to Paducah for uranium recovery and processing [59, 60]. This ash would contain Pu and Np.

After closure of X-344, recycled uranium would enter PORTS as UF₆ from Paducah feed or directly from other sources.

Transuranics and fission products in depleted uranium

UF₆ feed that was produced from recycled uranium (RU) contains TRU and fission products. The feed is shipped and fed in cylinders of various sizes. Small cylinders with a diameter of 5, 8 and 12 inches were used at PORTS to transport and store UF₆ produced at the X-705 conversion plant [61]. This feed was usually highly enriched. Larger (10 and 14 tons) cylinders were used for UF₆ that was converted at Paducah and shipped to PORTS.

The cascade tails or depleted uranium (DU) contains most of the mass of the uranium, but with a lower concentration of U-235, which is enriched in the cascade product. Because TRU has a great affinity to metal surfaces, most TRU do not appear in the product or the tails. Pu itself does not readily form a +6 valence state and therefore does not exist as a

58 USDOE, *Independent Investigation Vol. 1*, sec. 2.2, 3.2.1.

59 Theta Tech, pp. 17, 19.

60 USDOE, *Independent Investigation Vol. 1*, sec. 2.2, 3.2.1.

61 BJC, Recycled Uranium Mass Balance Project, BJC/PORTS 139, April 2000, p. 47.

hexafluoride to any great extent. However, small amounts of Np, Am and Pu have been found in the cascade tails.

No Tc-99 is found in the cascade tails, because besides plating out in the pipes, Tc-99 is lighter than uranium and migrates upward in the cascade, away from the tails.

When the cylinders are fed, not all of the TRU and Tc-99 enters the diffusion cascade. Estimates of the fraction that remains in the feed cylinders range [62][63] from 60 to 75 % for Pu, 75 to 90 % of Np, and 10 – 15 % for Tc-99. When the same cylinders are re-filled with RU without cleaning, an accumulation of TRU and FP occurs in the cylinder heels. Many cylinders were known to be re-used without cleaning, including a number of 10-ton cylinders that were filled four consecutive times with RU feed, without being cleaned between fills [64].

Some cylinders with Tc-99 and TRU-heels were filled with depleted uranium from the cascade tails, without the benefit of washing. The exact number of such DU cylinders that contain TRU and Tc-99 is unknown, but estimated to be a few hundred [65].

Hence, depleted uranium may contain TRU in two main forms: small particulates uniformly dispersed throughout the DUF6 contents (drawn from the cascade tails), and consolidated heels from the original feed.

Finally, the DU is shipped out of PORTS and either stored at other sites, or used for the manufacture of armor-piercing ammunition. As proof that Pu, Np and Tc remain with DU, the Army has sampled DU metal used in tanks. Tests show a high concentration of Tc-99 and low concentrations and approximately equal concentrations of Np-237 and Pu. [66]

X-616 Liquid Effluent Control Facility (Chromium Sludge Lagoons, 1976-85)

Also located in QIII, the X-616 facility was used to treat blowdown from the recirculating coolant water (RCW) system. Chromium was added to coolant water to inhibit corrosion. The X-616 facility precipitated chromium from the water, generating chromium hydroxide

62 BJC 2000, p. 46

63 US DOE, Strategy for Characterizing Transuranics and Technetium Contamination in Depleted UF6 Cylinders, Report by Hightower JR et al, ORNL/TM-2000/242, Oct 2000, p. 3.

64 BJC 2000, p. 46.

65 US DOE Oct. 2000, p. 3.

66 Ramachandra, KB, USATA, Radiation Research Office, to Lapajenko-Maguire, Commander, US Tank and Automotive Command, Dept of the Army, memo, January 19, 2000.

The uranium recovery process at X-705 salvaged the uranium contained in (1) decontamination solutions from equipment removed from the cascades and other areas, (2) trap media, (3) oxide conversion ashes and filtrates, (4) UF₄ conversion ashes and filtrates, (5) incinerator ashes, (6) UF₆ cylinder cleaning, (7) field decontamination solutions, and (8) laboratory sources [70]. Decontamination solutions would be digested with nitric acid, concentrated, extracted and calcinated to produce uranium oxide. The uranium oxide would then be converted to UF₆ and fed to the cascades.

A major upgrade of the cascades beginning in the mid 1970's led to the decontamination of cascade equipment and the subsequent release of Tc. The resulting depleted acid (raffinate) was discharged into the X-701B Holding Pond (see below) and led to greatly increased concentrations of Tc and a major Tc groundwater plume (Q II), as will be discussed below.

At X-705, transuranics remained with uranium for the most part, but technetium entered the raffinate stream and was subsequently released to the environment. The raffinate also contained uranium daughter products and TCE. The equipment decontamination process at X-705 depended on the type and size of equipment. Large equipment would be disassembled and vacuumed first to remove uranium compounds and other particulates. Exhaust from the vacuum would be filtered through a HEPA filter. Large parts would be placed on a dolly and passed through a series of spray booths (tunnel). The parts would be washed with recirculating nitric acid, ammonium carbonate, and rinse water. These solutions would later be processed at the uranium recovery portion of X-705. In 1984, discharge to X-701B was permanently suspended with the startup of the heavy metals precipitation process, Tc-99 ion exchange process (both in X-705), and biodentrification process (X-700). The treated solutions were discharged to the sanitary sewer and further treated in the X-6619 Sewage Treatment Plant before discharge to the Scioto River. When PORTS began treating the raffinate with ion exchange resins, technetium was disposed of together with the spent resin at the X-749 landfill.

Prior to the closure of the X-701B Holding Pond in late 1988, water from the final rinse was discharged to X-701B at a rate of 50,000 gallons per month, the total uranium concentration reportedly being 1 to 2 ppm. Meanwhile, small parts would be decontaminated by hand with water, nitric acid, ammonium carbonate, and isopropyl alcohol. The final step in decontamination was accomplished by spraying the parts with steam guns. The effluent was also discharged to X-701B [71].

Cleaning of UF₆ cylinders was done using a boric acid wash solution, sometimes mixed with nitric or citric acid. The cleaning solution was then discharged to the RCW blowdown line, which was further treated at the X-616 chromate treatment facility before being discharged to the Scioto River. Beginning November 1988, solutions from this process are

70 Theta Technologies, Apr 2000, p. 28.

71 Geraghty & Miller, Inc., *Quadrant II RFI Final Report*, DOE/OR/11-1232/V1&D3, POEF-ER-4583/V1&D3, Vol. 1, Sept 30, 1996, p. 83.

passed through a microfiltration system installed in X-705 prior to discharge to the RCW blowdown line.

Also housed within the X-705 building were a laundry service and a process laboratory. The laundry facility was used to wash protective clothing worn by workers. Discharges from the laundry were sent to a sump that drained to the X-615 Sewage Treatment Facility via the sanitary sewer. This would then be released to the Scioto River. Uranium accumulation at X-615 was estimated to be 10 kg/year. Present discharges, amounting to about 15,000 gallons per month, are now directed to X-6619 through the sanitary sewer line. The X-705 Process Laboratory provides routine chemical analyses for the X-705, X-700, and X-342 facilities. Most laboratory drains discharge to the X-6619 through the sanitary sewer line. Dilute chemical solutions from the laboratory were discharged to X-701B, as long as it was operating [72].

It is important to note that X-705 did not have a stack sampling program during the 1970's when the CIP/CUP program was underway. For X-705, the main emissions seemed to be through the raffinate output, rather than through vent stacks.

There are indications that about 19 kg, or 325 Ci, of Tc-99 may have been released prior to 1978 [73]. Measurements of Tc-99 during 1982 indicate that most losses were of a sudden nature, because the venting happened mainly when the cells were disconnected from the cascade and cleaned out prior to sending it to X-705 for further decontamination.

Present in all soil samples collected in the X-705 vicinity was total uranium at concentrations ranging from 3.3 mg/kg to 70.6 mg/kg. Gross alpha was detected in some soil samples at activities ranging from 6 pCi/g to 169 pCi/g. Gross beta activities in three samples were 20, 28, and 169 pCi/g. Technetium was in more than half of the samples at levels ranging from 0.2 pCi/g to 85 pCi/g [74]. These data are above background and indicate the presence of plant effluents.

Transuranics have accumulated in stream sediments to the east of the site. Pu and Np could only have arisen from the X-705 facility. PORTS did not sample consistently for TRU contaminants until 1999. [75]

X-705E: Oxide Conversion Facility (1957-78)

The purpose of the Oxide Conversion Facility's was to produce UF₆ feed from uranium oxides recovered from decontamination solutions in X-705 and incinerator ash [76].

72 Geraghty & Miller, *Quadrant II RFI*, Sect. 4, p. 84.

73 Makhijani, p. 16

74 Geraghty & Miller, *Quadrant II RFI*, Sect. 4, pp. 88-90.

75 DOE, "Independent Investigation of the Portsmouth Gaseous Diffusion Plant, Vol. 2," May 2000, p. 22.

76 The X-705E facility is located in the northeast section of the X-705 Building.

solution – either PORTS modifies and contains the old system or designs and builds a new contained system [103].

After the test run, an engineering study of the X-705 Oxide Conversion Facility was ordered to look into the problem of high airborne contamination within the operator's work area. A preliminary investigation found low-level contamination within the facility. Heated process gas housings were heavily contaminated, which became a source of airborne contamination during maintenance operations or when struck in an accidental manner. Many routine production and maintenance operations involved use of equipment that was not housed within the glove box, the primary containment area. The overhead utility piping and ventilation ducts were arranged in an inaccessible manner in an area with 16' high ceilings, which made it difficult to thoroughly decontaminate the area after a high airborne contamination exposure, leaving the top surfaces contaminated. A sudden room air disturbance could generate localized contamination. Also, a majority of the process valves were unbuffered plug valves, which made it difficult, even impossible, to detect low-level leaks when they failed. These valves had a high, unpredictable rate of failure, resulting in valve failures becoming common within the facility. Lastly, seals were not used to isolate pneumatic instruments from process piping. This enabled process gas to back up through instrument lines, resulting in plugged instrument lines. Repairs of these plugged lines became a source of airborne contamination [104].

The study team preferred that the existing facility be abandoned and a new one built, although the team noted that it was also possible to strip and modify the existing facility. Extensive modifications would have to be made since the existing facility was not originally designed to process material contaminated with transuranics. It was estimated that 8%w to 20%w of all oxide feed ended up as unreacted tower ash, causing material handling problems and increasing the airborne contamination risk. The team also recommended that a thorough study of the behavior of uranium daughter products and transuranics within the system be done. It was estimated that approximately 75%w of all Pu and 25%w of all Np entering the system was deposited in the purge gas filter and tower ash while approximately 95%w of remaining Pu and Np deposited in the MgF₂ traps. If the MgF₂ traps were overloaded with chromium, transuranics would be displaced and proceed to contaminate the remainder of system [105]. As stated, in late 1978 X-705E operations were shut down.

X-705A: Waste Incinerator (1950s-1986)

Uranium-contaminated wastes generated from plant operations were burnt here. Because the original two incinerators had routinely produced heavy black smoke, they were

103 Collier and Smith.

104 PE Cross, TE Noel, and SJ Wethington, *Preliminary Study Oxide Conversion, X-705*, Dec 12, 1978.

105 Cross, Noel, and Wethington.

smokeless but due to improper use of the incinerator (examples are incineration of plastics and non-combustible items, lack of maintenance, introduction of oils and solvents into the incinerator to enhance combustion), there were several incidents of severe smoking and smoke incursions into nearby buildings [112].

Ash resulting from the incineration was shoveled and vacuumed from the combustion chambers, collected in cylinders, and sampled. If the laboratory analysis showed a uranium concentration that was recoverable, the ash was sent to the X-705 uranium recovery process for recycling. Otherwise, the ashes (in cylinders) were placed in wooden boxes and disposed of in the X-749 landfill. Measurable radioactive contamination was present on most surfaces inside the building. Inhalation of airborne radioactive materials while handling the incinerator ash presented the greatest hazard to the workers. Although required, respirators were not normally worn [113, 114].

The radiator was ordered shut down in 1986 after it was discovered that, between August 1984 and April 1986, 370 gallons of flammable liquids (waste oil and tributyl phosphate) were improperly incinerated [115], that is, contrary to licensed activities. ORO wanted to develop specific procedures concerning the receipt of acceptable wastes to correct the problem but the State revoked the facility's permit. Testing of the oils, solvents, and incinerator ash indicated it to be RCRA hazardous waste because of the presence of cadmium and barium [116]. Some ash samples reportedly exceeded 100 ppm of barium and 1 ppm cadmium while some of the waste oil batches failed the EP toxicity test for mercury. The incinerator was never used again and was closed under RCRA authority in the 1990s [117]. As seen in Fig. 4, gross alpha concentrations increased in 1984, presumably due to the burning of these hazardous chemicals.

With the closure of the incinerator, 1700 containers of burnable waste materials are currently stored on site. Also, PORTS continues to store the residual incinerator ash. Sampling of this incinerator ash revealed that it contained enriched uranium and trace quantities of plutonium, neptunium, and hazardous metals [118].

The soils surrounding the X-705A incinerator building and the X-705B storage pad were contaminated with uranium, technetium and transuranics at above-background levels. The soil concentration of Pu-239 ranged from near background up to 5.37 pCi/g; the soil concentration of Tc-99 ranged from near background up to 4,700 pCi/g [119]. The elevated soil concentrations indicate elevated air concentrations that Portsmouth workers would

112 CDM, pp. B-22 to B-25.

113 USDOE, *Independent Investigation Vol. 1*, sec. 3.2.7, 4.1.

114 Geraghty & Miller, *Quadrant II RFI*, p. 97.

115 CDM, pp. B-22 to B-25.

116 USDOE, *Independent Investigation Vol. 1*, sec. 3.2.7, 4.1.

117 CDM, pp. B-22 to B-25.

118 CDM, pp. B-22 to B-25.

119 Department of Energy, "Quadrant II CAS/CMS Final Report, Portsmouth Gaseous Diffusion Plant," August 15, 2000.

inhale. Working backward, one could estimate the air concentrations and likely radiation dose to Portsmouth workers due to operation of the incinerator. There is some uncertainty though about the presence of uranium-contaminated soils under the incinerator building, storage pad, and aboveground storage tanks. Contamination is attributed to past operation of the incinerator, specifically on the airborne transport of contaminants, spills or leaks of handled waste materials, and spills and runoff from the storage lot [120].

During Phase I of the Quadrant II RCRA Facility Investigation (RFI), radiological and toxic chemical contaminants were detected in soils around the incinerator building and storage lot. Found were polycyclic aromatic hydrocarbons (PAH), Tc-99, and uranium. PAH were detected at 3 locations up to 430 ppb. The radiological contamination was highest near the incinerator and within the top 2 feet of soil. Gross alpha ranged from 22 pCi/g to 6,516 pCi/g, while gross beta contamination ranged from 21 pCi/g to 1,621 pCi/g. Technetium was found at levels ranging from 6.3 pCi/g to 333 pCi/g. Total uranium concentrations ranged from 5 ppm to 2,180 ppm. At that time, isotopic composition of the uranium present in soil had not been determined, but was assumed to be similar to naturally occurring uranium in setting up Preliminary Remediation Goals (PRGs) [121, 122, 123]. Transuranics were not factored into the risk analysis since they were not detected in the original RFI data. But post RFI analysis does show areas near X-705A and X-701B with transuranics, including plutonium in Little Beaver Creek. [124]

Because of the high levels of radiological contamination in X-705A/B, additional soil samples were collected during Phase II of the Quadrant II RFI and analyzed for isotopic uranium, technetium, and transuranics. Gross alpha and gross beta activities varied from 11 pCi/g to 351 pCi/g and from 46 pCi/g to 258 pCi/g, respectively. Total uranium ranged from 2.4 ppm to 84 ppm. U-234 was detected in one sample at 270 pCi/g while U-235 was detected in two samples at activities of 7.06 pCi/g and 14.20 pCi/g. Technetium varied from 4.2 pCi/g to 189 pCi/g. No transuranics were detected.

Upon review of cleanup alternatives, OEPA and USEPA recommended remediation of the X-705A/B site by excavation, washing, and disposal of the contaminated soil. The plan included decontamination and demolition of the X-705A incinerator and building prior to soil excavation [125]. The incinerator and the building were demolished and removed in 1996 [126]. A total of 214 boxes or 1,260,009 pounds of excavated soil were sent to Envirocare in

120 Science Applications International Corp. (SAIC), *X-705A/B Draft Cleanup Alternatives Study / Corrective Measures Study Report for the Portsmouth Gaseous Diffusion Plant, Piketon, Ohio*, DOE/OR/12-1239&D1, POEF-ER-4573&D1, Mar 1994, pp. 3-4, 9, 90.

121 Geraghty & Miller, *Quadrant II RFI*, p. 100-101.

122 SAIC, pp. 3, 9-10.

123 *The Ohio EPA's and US EPA's Preferred Plan for the X-705A/B Solid Waste Management Unit, US DOE-PORTS Site*, Mar 1995, p. 9.

124 *Independent Investigation*, vol 2, p. 12.

125 *Preferred Plan for X-705A/B*, pp. 28-29.

126 USDOE, *Independent Investigation Vol. 1*, sec. 3.2.7, 4.1.

FY 1999 [127]. We are uncertain if all the contaminated soil has been removed, or if some contamination remains at the X-705A-site. Recent soil and groundwater samples north of the plant area, on land scheduled for release for industrial use show trace levels of neptunium and plutonium in the soil indicating the effluents from the incinerator and conversion facility have spread over a much larger area than previously supposed.

X-701B: Holding Pond (1960s-1988)

This unlined pond was used for the neutralization and settling of metal-bearing waste water, solvent-contaminated solutions, and acidic waste water, with most of the wastes coming from the X-700 Chemical Cleaning Facility and the X-705 Decontamination Building [128]. An estimated 1,415 g of Tc-99 was sent to the holding pond along with the decontamination raffinate [129]. Effluent from the holding pond flows through the East Drainage Ditch into Little Beaver Creek. Starting in 1971, slaked lime was added to neutralize the low pH and promote precipitation; once or twice a year, the pond was dredged and the sludge pumped to containment ponds. It can be assumed prior to 1971, much higher concentrations of uranium and Tc were released to Little Beaver Creek, though measurements are not available. In 1984 the discharge of raffinate to X-701B was permanently suspended due to the startup of treatment processes in X-705 and X-700.

From 1954 to November 1988, the unlined X-701B holding pond was used for the neutralization and settling of liquid process effluents received from the X-700 Chemical Cleaning Facility and the X-705 Decontamination Building via the X-701C Neutralization Pit and Process Waste Lines. The effluents were primarily wastewaters and cleaning solutions contaminated with acids, organic solvents, metals, and radionuclides. Common wastes discharged into the pond were nitric acid, chromic acid, TCE and TCA. The latter two solvents were usually contaminated with Tc-99 and uranium [130]. Uranium reportedly comprised 92% (76.5 kg) of the total radioactivity released to the holding pond in 1969 and 90% (117 kg) of the total in 1970 [131].

In 1974, PORTS began adding slaked lime to the X-701B influent at the X-701E Neutralization Facility to neutralize the low pH and induce the precipitation of radionuclides. This caused large amounts of sludge to accumulate in the holding pond, necessitating annual dredging of the sludge (until 1985, when the pond was last dredged). The sludge removed from the pond was dewatered and stored in two basins known as the X-701B East and West Retention Basins. The East Retention Basin was built in 1973 and received sludge from

127 "More than 4.6M Pounds of Waste Shipped in FY 1999", *Portsmouth Environmental Bulletin*, Jan 2000, p. 3.

128 USDOE, *Independent Investigation Vol. 1*, 2000

129 Theta Technologies, *Recycled Uranium Mass Balance Project*, Portsmouth, Ohio, Site Report, prepared for Bechtel Jacobs Company, BJC/PORTS, April 2000.

130 Geraghty & Miller, *Quadrant II RFI*, pp. 65, 67.

131 USDOE, *Independent Investigation Vol. 1*.

1974 to 1980, when it became full. The West Retention Basin was then built in 1980 and used until 1988 [132]. Like the holding pond, these retention basins were unlined and did not have a leachate collection system [133]. A groundwater plume of TCE and radioactive materials originates near the X-705 facility and empties into X-701B and Little Beaver Creek. This is discussed in Section E below.

X-626/630/633: Recirculating Cooling Water (RCW) System (1954-present)

The separation of uranium isotopes via gaseous diffusion, which involves repeated compression of the UF₆ gas in the cascades, generates a tremendous amount of heat. PORTS' RCW system collects this heat and dissipates it to the atmosphere at cooling tower complexes.

Water pumps and cooling towers circulate water that is used to remove the heat of compression from the process gas in the cascades, along with the waste heat in a few auxiliary processes, and dissipate this heat to the environment. The cooling water was to some extent in contact with materials and substances used in the cascade. To limit the concentration of solids that will occur in the recirculating water, a quantity of water called blowdown is removed from the system continuously and discharged to the X-616 Liquid Effluent Control Facility. The solids in the water included chromium, zinc, copper, iron, sulfate, silica and pentachlorophenate (a fungicide applied to the wood of the cooling towers). Gas emissions included chloride, freon 114 and freon 113.

The Quadrant II RCW system in X-633 consists of RCW lines surrounding the X-330 and X-333 process building, the X-633 recirculating water pump house, and four cooling towers with associated basins [134].

Analyses of groundwater samples collected at X-633 during the Quadrant II RFI indicated chemical and radiological contamination.

X-700: Maintenance Building (1954-?)

The building was used for maintenance and cleaning of non-radioactive or low-level radioactively contaminated equipment from the cascades and other parts of the plant. Heated TCE was used for degreasing until 1987 when it was replaced with 1,1,1-trichloroethane (TCA).

132 Geraghty & Miller, *Quadrant II RFI*, pp. 65, 67.

133 LMES, *1995 Environmental Report*, p. 6-25.

134 Geraghty & Miller, *Quadrant II RFI*.

Quadrant I

X-231A/B: Oil Biodegradation Plots (1971-1983) (Quadrant I)

At PORTS, waste oils were treated according to a process developed in Oak Ridge. However, the fertilizing, tilling and disking that the process required were not adequately implemented. Plot A received about 24,500 gallons of waste oil contaminated with solvents and radionuclides, 124,300 pounds of oil-soaked fuller's earth, 60 gallons of TCE, and 1,000 gallons of chlorinated solvents. Plot B received uranium-contaminated waste oil, PCBs, and solvents, but no records were kept of the quantities applied [135].

a. X-231A Southeast Oil Degradation Area

Covering approximately 54,000 ft², the X-231A area was used for the disposal of wastes contaminated with volatile organic compounds (VOC) during the 1970s. [136]

TCE is the primary groundwater contaminant in the X-231A area. The highest TCE concentration detected is 120 ppb. Within the TCE plume, other VOC contaminants found were TCA, DCE, and DCA [137].

X-231A was treated with agricultural lime and fertilizer, which were then disced into the soil. A soil berm was added along with a reinforced geosynthetic membrane cover. In 1996-1997, treatability studies were conducted at the site [138], as discussed below.

b. X-231B Oil Biodegradation Plots

The X-231B unit was used for the disposal of wastes contaminated with volatile organic compounds (VOC), uranium, and technetium. It covers approximately 37,000 ft² and received hazardous wastes from 1976 to 1983 [139]. The waste oils and degreasing solvents received were applied on 2 plots of land, which were periodically fertilized and disced for aeration and the promotion of natural biological degradation of waste oil [140].

RFI data indicate that TCE and technetium levels in the soil exceed leaching levels established by OEPA, while uranium is present at above background concentrations. TCE was found at various depths in the oil plots while uranium and technetium were confined to

135 USDOE, *Independent Investigation Vol. 1*, 2000.

136 *Quadrant I CAS/CMS*, p. ES-3.

137 LMES, *1995 Environmental Report*, p. 6-41.

138 *Quadrant I CAS/CMS*, p. 1-34.

139 *Quadrant I CAS/CMS*, p. ES-3.

140 LMES, *1995 Environmental Report*, pp. 6-27, 6-29.

depths less than 6 feet. Groundwater investigations at the X-231B unit indicate that the VOC (primarily TCE) plume extends about 1,000 ft to the south toward the X-230K Holding Pond. The northern boundary of the plume is near the south end of the X-710 building [141].

In 1994, RCRA closure of the X-231B unit resulted in source removal actions, which partly removed some of the VOC contamination. TCE, however, remained at concentrations exceeding its soil leaching levels [142].

In July 1994, IRM at X-231B was completed. It consisted of shallow soil mixing to a depth of ~22 ft below ground surface combined with TEVE. Off-gases were treated through a series of activated carbon and HEPA filters. DOE estimated that about 70% of the VOC were removed. After treatment, the site was covered with a layer of clean soil and graded to promote precipitation runoff. Underground pipes from the three groundwater extraction wells near X-231B were installed to deliver groundwater to the X-622 Groundwater Treatment Facility [143, 144].

X-749A: Classified Materials Burial Ground (1953-1988)

The X-749A landfill is a 5.9-acre area east of the X-231B Oil Biodegradation Plot. Operated from 1953 through 1988, the X-749A facility served as the disposal site for classified materials. RCRA closure of the X-749A as a solid waste landfill began in June 1993 and was completed in April 1994. Remediation consisted of installing a drainage system to collect surface water runoff and covering the landfill with a multimedia cap [145, 146, 147].

The X-749A vicinity is included in the Quadrant I 5-unit investigative area. PORTS claims that RFI data indicate X-749A is not a contamination source for the 5-unit investigative area groundwater plume, although groundwater west of X-749A is contaminated. Apparently, no inorganics, metals, or radionuclides are leaching from the X-749A unit. However, according to Lehman & Associates, the X-749A monitoring plan was not adequate to detect the contamination gradient in the Berea and to fully define the southern extent of contamination [148]. The southernmost wells in the monitoring network exhibited uranium concentrations that indicate that the contaminant plume from the X-749 RCRA has migrated beyond the existing monitoring network.

141 LMES, *1995 Environmental Report*, pp. 6-28 to 6-30.

142 *Quadrant I CAS/CMS*, p. ES-3.

143 The X-622 facility uses an activated carbon filtration system to treat the contaminated groundwater.

144 *Quadrant I CAS/CMS*, p. 1-34.

145 LMES, *1995 Environmental Report*, p. 6-21.

146 *Quadrant I CAS/CMS*, p. 1-35.

147 US Department of Energy (USDOE), *Portsmouth Environmental Restoration Activities*.

148 L. Lehman & Associates, *Opinions Regarding Assessments at PORTS*, yearxxx?

X-749: Contaminated Materials Disposal Facility (1955-1990)

Low-level radioactive wastes were buried in this landfill beginning in the late '50's and continuing until 1992. This landfill received all kinds of contaminated solid wastes, including spent trap material containing Tc-99, ion exchange resin, incinerator ashes, replaced equipment etc. Unsealed chemical traps contained water-soluble Tc-99 [149]. Because of the inappropriate burial of RCRA waste, Ohio EPA directed that the landfill be closed, and it was shut down in 1993 [150]. The contaminants leaking into the groundwater from X-749 included uranium, TRU, Tc-99, metal hydroxides, TCE, and other VOC, all located in an extensive groundwater plume that follows the potentiometric surface. See Figs. 5 and 6.

The X-749 landfill was used for the disposal of hazardous and low-level radioactive waste materials and contaminated equipment. The northern portion received wastes contaminated with solvents, oil, and wastewater treatment sludge from 1955 through 1989. It was closed in September 1993 in accordance with RCRA Subtitle C requirements. The southern portion received "nonhazardous" low-level radioactive wastes from 1986 through 1988 and was closed in March 1993 under RCRA Subtitle D. A major groundwater plume of TCE extends from X-749 to the south towards Big Run Creek, as discussed in Section E below.

A closure plan for the X-749 landfill was approved in June 1989. A multimedia cap was installed over the northern and southern portions of the facility. Construction began in February 1991. The northern portion was completed in June 1992 and the southern portion in December 1992. A slurry wall was installed along the northern and western boundaries of the landfill to restrict the movement of groundwater into the landfill area. A groundwater collection system was installed along the southwestern boundary and the northern portion of the eastern boundary of the X-749 to capture contaminated groundwater flow from the landfill. The captured groundwater is pumped to and treated at the X-622 Groundwater Treatment Facility [151]. This active maintenance must likely continue into the indefinite future.

Peter Kiewit (PK) Landfill (1954-68)

Opened in 1952, the Peter Kiewit (PK) Landfill was used as a salvage yard, burn pit, and trash disposal area by the construction contractor, Peter Kiewit and Sons, during the construction of PORTS. After plant construction, the site was used as a sanitary landfill until 1968, when the landfill was backfilled with soil, graded, and seeded. PORTS reported there

149 *Independent Investigation*, vol. 1, p. 23.

150 *Ibid.*

151 *Quadrant I CAS/CMS*, pp. 1-35, 8-6.

Groundwater in the X-120 area is contaminated with TCE. In 1989, TCE concentrations of 100 and 800 ppb were detected in 2 groundwater wells located at the X-120 area. At first, because of its proximity to the X-749 facility, it was assumed that the TCE contamination originated from the X-749 unit. However, further investigation during the Quadrant I RFI revealed that X-120 is a source of TCE contamination, independent of the X-749 unit. TCE concentrations in 4 wells ranged from 18 ppb to 1,200 ppb.

The X-120 plume is a long and narrow stretch of TCE-tainted groundwater, extending about 1,400 ft to the southwest from south of Hewes St. The southeastern edge of the X-120 plume nearly converges with the X-749 plume [158]. In 1996, a horizontal well was installed along the axis of the plume, with the extracted groundwater being sent to the X-625 Groundwater Treatment Facility 159.

Soil samples taken from the X-120 area during RFI indicate that contaminants are no longer present above OEPA's leaching levels, hence, X-120 no longer appears to be a source of contamination. After review of the RFI Final Report, it is not clear to us how the contaminants were removed from the soil.

Quadrant IV

X-611A Lime Sludge Lagoon

X-611A is a series of three lagoons, surface area 11.2 acres, in Q IV that accepted lime sludge from 1954 to 1960 generated by the water treatment process. Coolant water was treated with acid at the X-611 facility, and then mixed with high pH sludge in the lagoon, prior to release to Little Beaver Creek. The contaminants are total chromium and zinc from the RCW. The lagoons were covered in 1996. As expected, total chromium was detected in sediment samples at concentrations ranging from 3.5 to 5,230 ppm. Zinc was detected at concentrations ranging from 12 ppm to 382 ppm. Total uranium and gross alpha concentrations were above background [160]. The lagoon was capped in 1996. 161

X-734 Old Sanitary Landfill; X-734A Construction Spoils Landfills; X-734B Old Construction Spoils Landfill

158 LMES, *1995 Environmental Report*, pp. 6-39, 6-41.

159 DOE, *1999 Environmental Report*, p. 6-7

160 *Quadrant IV, RFI Draft Report*, Section 4, p. 64.

161 Davis report, p. 15.

These 3 landfills with a surface area 13.6 acres, contained non-radioactive trash and garbage, organic solvents, and solid waste, including construction spoils, trees and wood. Apparently empty paint cans and 55-gallon drums containing hazardous materials were also disposed in X-734B [162]. Elevated levels of gross alpha and technetium were detected in surface and ground water. TCE was detected in subsurface soil samples and in groundwater in the unconsolidated aquifer. Seeps located on the northeast side of the landfill have elevated concentrations of TCE and technetium. [163]

X-735A Sanitary Landfill

This 7.9-acre landfill accepted solid wastes, including sludge from coal storage runoff, grit screening from the sewage treatment plant and fluorine sludge. This solid material was placed in trenches [164]. No releases of VOC or PCB's to groundwater were detected. Closure of the southern part of this landfill was completed in 1998. [165]

E. Groundwater Contamination

Quadrant II

X-705 Decontamination and Uranium Recovery Facility

The X-705 facility was responsible for decontamination of all plant equipment and recovery of U from solution. X-705 also recovered U from ashes, cylinders and laboratory solutions. A major decontamination effort began in the mid '70's when the GDP stages were upgraded. As a result numerous GDP stages were removed and decontaminated at X-705. This led to major releases of U and Tc, and less concentrated releases of Pu and Np to the air and water. The releases of alpha emitters to the air were recorded at station 12 (see Fig. 4) and peaked in 1973 and 1976. Releases of beta-gamma emitters, primarily Tc, to X-701B holding pond are shown in Fig. 7. As seen, the beta-gamma releases peaked in 1976 and 1984, consistent with air emissions. Degreasers were used in X-705, leading to major TCE contamination.

The oxide conversion facility X-705E converted U oxides and UNH to UF₆ from X-705 decontamination solutions and incinerator ash. Between FY 1968 and FY 1978, various

162 *Quadrant IV, RFI Draft Report*, Section 4, p. 88.

163 *Quadrant IV, RFI Draft Report*, Section 4, p. 96.

164 *Quadrant IV, RFI Draft Report*, Section 4, p. 99.

165 Davis report, p. 16.

quantities and assays of recycled uranium were received as UF₆ or converted to UF₆ at the X-705E Oxide Conversion facility.

The acid waste stream (raffinate) from these operations, containing Tc, Pu and TCE, was initially discharged to an onsite ditch that flowed to Little Beaver Creek. Later, the raffinate was directed to an on-site settling pond, X-701B, where the raffinate was neutralized. The resultant sludge was dredged from X-701B once or twice a year and pumped to containment ponds.

As a result of past operations, the drainage ditch, holding pond and containment ponds have become contaminated. Wastewater collected at X-705 during Phase I of the Quadrant II RFI indicated elevated VOC and radiological levels. Two (of two) samples had TCE concentrations of 830 µg/L and 8,700 µg/L. Gross alpha was detected at activities of 44,855 pCi/L and 300,305 pCi/L while total uranium was measured at 20.8 mg/L and 66 mg/L. Gross beta was detected at activities of 874,584 pCi/L and 937,993 pCi/L while technetium levels were measured to be 469,858 pCi/L and 565,836 pCi/L. These Tc-99 concentrations were 7.8 and 9.4 times the maximum permissible concentrations for off-site waters.

The X-705 building was also found to be a source of groundwater contamination. Groundwater samples collected at X-705 were analyzed as part of a "Quadrant II Investigative Area", which includes X-705, X-700, X-700CT, X-700T, X-700C, and X-720/X7-20NP. The X-700 building was used for chemical cleaning, using heated fluorine gas and vapor degreasers. This building released TCE to the air and through floor drains. X-720 was another maintenance building also used for cleaning and conditioning parts. RFI data for this investigative unit delineated the extent and nature of the groundwater contamination in the Gallia aquifer consisting of VOC, chromium and technetium (Figs 8-10). Of the several VOC detected, the primary contaminant is TCE at concentrations ranging from 0.88 µg/L to 23,000 µg/L (or ppb). Radiological parameters detected in the groundwater of the Quadrant II Investigate Area were gross alpha (15 pCi/L to 477 pCi/L), gross beta (47 pCi/L to 6,039 pCi/L), and technetium (31 pCi/L to 7,650 pCi/L). In the vicinity of X-705, the highest TCE concentrations were 1,100 µg/L and 1,300 µg/L (in contrast, the EPA's National Primary Drinking Water standard is 5 ppb, while the highest technetium concentrations were 5,117 pCi/L and 7,650 pCi/L). It was noted that the X-705 samples with the highest TCE levels also had the highest technetium concentrations.

According to PORTS modeling, the lateral extent of the groundwater plume is determined by the pumping of the X-705/X-700 basement sump pumps, while the vertical extent of the plume is reportedly restricted by the Sunbury shale underlying the area, which retards the movement of contaminants to the Berea aquifer [166]. According to DOE, the use of the sump pumps means that the groundwater does not flow to the east, as it would naturally in this area, but towards the pumps, where it is collected and then treated at the X-

166 Geraghty & Miller, *Quadrant II RFI*, pp. 201-211.

gross alpha (80 - 510 pCi/L) and gross beta (57 – 910 pCi/L). The groundwater plume of the beta-emitting technetium also reaches Little Beaver Creek (Fig. 12). Tc-99 was detected at activities ranging from 91 pCi/L to 2,800 pCi/L while uranium, an alpha-emitter, was not detected [173].

PORTS claims that the TCE plume is limited to the Gallia aquifer. However, small amounts of TCE have already been detected in the deeper Berea sandstone. Contamination has also been detected in the East Drainage Ditch and the Little Beaver Creek. PORTS reported in 1995 that no contamination has been detected beyond or east of Little Beaver Creek [174]. However, in 1990 USEPA stated that existing contamination in Little Beaver Creek had to be considered “off-site”, and that it was a threat to human health and the environment [175]. During Phase I of the RFI in 1996, VOC and radionuclides were detected in surface water samples from the creek. TCE was found at concentrations ranging from 2 µg/L to 110 µg/L. Gross alpha activity ranged from 12 pCi/L to 29 pCi/L while gross beta ranged from 4 pCi/L to 61 pCi/L. Tc-99 was detected at levels ranging from 54 pCi/L to 68 pCi/L [176]. Surrounding sediment samples contained DCE (1,900 µg/kg) and TCE (12 µg/kg, 23 µg/kg).

In 1991, as an Interim Remedial Measure, a T-shaped interceptor trench was installed to prevent contaminated water from discharging into Little Beaver Creek and the East Drainage Ditch. The trench consists of 2 branches: one running north-south parallel to Little Beaver Creek and the other running east-west along the south side of the East Drainage Ditch east of X-230J7. The trench is lined with a geotextile, backfilled with gravel surrounding a perforated collection pipe. Two pumps remove groundwater from the trench and discharge it to the X-624 groundwater treatment facility. Whereas this effectively prevents contaminated groundwater from seeping through the trench, the X-624 groundwater treatment facility only removes a part of the TCE, and no radionuclides, before discharging the water to the Scioto River. Also, if the sump pumps in the trenches cease operation out of any reason, contaminants could accumulate in the trench and eventually seep to the other side. In 1994, the second phase of closure of the X-701B unit was initiated with the construction of a groundwater pump-and-treat system and in situ treatment of the soils in the bottom of the holding pond with thermally enhanced vapor extraction (TEVE). Use of the TEVE technology was not successful and subsequently terminated. The limestone riprap and gravel that was placed on the bottom of the Holding Pond to support the soil treatment equipment was left in place [177]. We could not confirm independently whether enough soil was removed from the bottom of the holding pond.

In FY 1999, PORTS shipped some of the X-701B-related wastes to Envirocare in Utah for disposal. The shipment included 1,899,535 lb (384 boxes) of sludge from the retention

173 Geraghty & Miller, *Quadrant II RFI*, pp. 66, 69.

174 LMES, *1995 Environmental Report*, pp. 6-27, 6-44.

175 Muno W, Memo, ???

176 Geraghty & Miller, *Quadrant II RFI*, p. 152.

177 Geraghty & Miller, *Quadrant II RFI*, p. 66.

basins that were originally stored onsite and 919,245 lb (560 drums) of soil from the interceptor trench. In FY 1998, 173 drums of interceptor trench soil were shipped to Envirocare [178].

X-633 Recirculating Cooling Water (RCW) System

The separation of uranium isotopes via gaseous diffusion, which involves repeated compression of the UF₆ gas in the cascades, generates a tremendous amount of heat. PORTS' RCW system collects this heat and dissipates it to the atmosphere at cooling tower complexes. The Quadrant II RCW system in X-633 consists of RCW lines surrounding the X-330 and X-333 process building, the X-633 recirculating water pump house, and four cooling towers with associated basins [179].

Analyses of groundwater samples collected at X-633 during the Quadrant II RFI indicated chemical and radiological contamination. Chromium was detected at groundwater concentrations ranging from 16 µg/L to 780 µg/L. These are approximately 0.2 to 8 times the EPA drinking water standard of 100 µg/L (0.1 mg/L) [180]. Zinc was detected at concentrations ranging from 76 µg/L to 37,000 µg/L, which are 0.02 to 7.4 times the drinking water standard for zinc of 5,000 µg/L (5 mg/L) [181, 182].

Gross alpha was detected at activities ranging from 46 pCi/L to 274 pCi/L, while gross beta activity ranged from 55 pCi/L to 301 pCi/L. The maximum contaminant level (MCL) of gross alpha in drinking water allowed by the EPA is 15 pCi/L, while the MCL for gross beta is the activity of a beta-emitter that would result in a dose of 4 mrem/yr [183]. However, uranium and Tc-99, alpha and beta emitters respectively, were not detected in groundwater. This led DOE to suggest that the elevated levels of gross alpha and gross beta may be due to the presence of natural radium and radon in the Sunbury shale bedrock [184]. However, DOE did not elaborate about the probability of this explanation, nor are we aware of high levels of natural radionuclides below the PORTS site.

Quadrant I: Groundwater Investigative Area

178 *Portsmouth Environmental Bulletin*, p. 3.

179 Geraghty & Miller, *Quadrant II RFI*.

180 US Environmental Protection Agency, *National Primary Drinking Water Standards*, EPA 810-F-94-001, Dec 1999.

181 US Environmental Protection Agency, *Current Drinking Water Standards*, Table 2, July 24, 2000, accessed 5 Jan 2001, <<http://www.epa.gov/safewater/mcl.html>>.

182 Geraghty & Miller, *Quadrant II RFI*, pp. 34, 37, 181.

183 *National Primary Drinking Water Regulations; Radionuclides; Final Rule*, 65 FR 76708-76753, Dec 7, 2000.

184 Geraghty & Miller, *Quadrant II RFI*, p. 40.

The 5-unit Quadrant I Investigative Area contains a groundwater contaminant plume of primarily TCE, but it also includes Tc-99. It encompasses an area extending south from the X-710 Technical Services Building to the X-230K South Holding Pond and east from the southwest corner of the X-326 Process Building to the X-749A Classified Materials Burial Ground [185]. The area includes the X-231A Southeast Oil degradation area and the X-231B Biodegradation plots.

X-749 Landfill

The primary contaminant of concern in groundwater in the X-749 area is TCE. It is believed that TCE was released when the landfill was still in use. The primary release pathway to the groundwater is through the soils beneath the unit [186]. Some of the other VOC detected and contained within the TCE plume are TCA, DCE, DCA, chloroform, and freon-113. The Gallia sand contamination plume at the X-749 facility extends about 1,500 ft to the south-southwest, about 1,000 ft to the south, and about 200 ft to the southeast of the X-749. Field investigations in 1992 and 1993 revealed the TCE plume is much closer to the DOE boundary than expected – just 55 ft of the southern boundary [187]. In the spring of 1994, as an Intermediate Remedial Measure (IRM), the OEPA and USEPA approved the installation of a 1,077 ft-long subsurface barrier wall near the southern DOE property line to prevent the X-749/X-120 TCE-contaminated groundwater plume from moving outside the PORTS boundary. Construction began in April 1994 and completed in September 1994 [188].

The wells in trenches had Tc-99 concentrations of up to 12,607 pCi/L in 1999, whereas Berea wells reported concentrations of up to 440 pCi/L. The highest concentration of TCE measured in 1999 was 3,700 µg/L at X749-06G [189].

The multimedia cap, barrier walls, and groundwater collection systems mentioned above reportedly removed the X-749 landfill as a source of ongoing contamination, [190] but it remains an issue how long the slurry wall and groundwater collection systems will remain effective. The Independent Oversight panel pointed out that PORTS did not monitor south of the slurry wall and therefore could not confirm its effectiveness in restraining the groundwater plume. [191]

185 *Quadrant I CAS/CMS*, p. ES-6.

186 *Quadrant I CAS/CMS*, p. 8-5.

187 LMES, *1995 Environmental Report*, p. 6-35.

188 *Quadrant I CAS/CMS*, pp. 1-35, 8-6.

189, EQ Midwest, *Portsmouth Annual Environmental Data for 1999, Piketon, Ohio* (DOE/OR/11-3053&D1), Jan 2001, pp. 4-3 to 4-10

190 *Quadrant I CAS/CMS*, p. ES-9.

191 DOE, *Independent Investigation*, Vol. 2, p. 12.

Peter Kiewit (PK) Landfill (1954-68)

The underground plume extending from the Peter Kiewit landfill and the X-749 landfill is shown in Fig. 6. Near the PK landfill, the TCE concentrations are over 100 ppb and moving in a southern direction. Nearer the X-749 landfill, the TCE concentrations are over 1000 ppb. The concentrations appear to be declining over time, likely due to the RCRA caps placed over the landfills and the interceptor trenches bounding the south and east side of the landfills.

Quadrant III

X-616 Liquid Effluent Control Facility (Chromium Sludge Lagoons, 1976-82)

Groundwater investigations revealed that concentrations of total chromium were elevated and exceeded regulatory limits. TCE was also detected “at low levels (less than 30 ppb)”; the maximum contaminant limit allowable in drinking water for TCE is 5 ppb (5 µg/L). In this area of PORTS, the Gallia sand directly overlies the Berea sandstone and thins in all directions away from the lagoons. There is no Sunbury shale to retard the movement of groundwater from the Gallia to the Berea aquifer. It is no surprise then that chromium and VOC (TCE and TCA) contamination have been detected in X-616 wells screened in the Gallia and Berea aquifers. Models constructed by PORTS predict that groundwater from both the Gallia and Berea discharges to the West Drainage Ditch, with an estimated travel time of more than 30 years for both [192].

F. Off-site Contamination and Remediation

Contamination

PORTS repeatedly claimed that there were no off-site releases, or that such releases were always under the allowed limit, or that the measured concentrations off-site were always below existing thresholds. This contradicts findings of several studies, including releases reported by PORTS itself.

PORTS published estimates about the releases to the surface water to be 221 Ci between 1975 and 1999 [193]. IN 1975 alone, the reported releases were 77.5 Ci, which is equivalent to a gross beta concentration of 6,830 pCi/L in Little Beaver Creek [194]. This is almost twice the concentration of 3,749 pCi/L of Tc-99, which causes a yearly dose of 4 mrem/year,

192 LMES, *1995 Environmental Report*, pp. 6-30 to 6-31, 6-49.

193 Theta Tech 2000, p. 5

194 Annual flow from <http://waterdata.usgs.gov/nwis-w/OH>, downloaded on April 11, 2001

Water treatment facilities

Five groundwater treatment facilities are operating on the site at present. In 1999, these facilities combined treated 24.7 million gallons of groundwater and reportedly removed about 100 gallons of TCE. The water treatment facilities do not remove radionuclides from the groundwater. After treatment, the water is discharged in so-called outfalls to the surrounding rivers [202]. In 1999, one sample of the discharge water from each treatment system was taken and tested for TRU. The results were always negative. However, Tc-99 was never measured. Because of the continuous extraction of groundwater, the natural groundwater flow is changed in the regions where groundwater is pumped out. Instead of flowing towards the natural discharge direction that originally defined the four quadrants, the plume is supposed to remain in place.

X-622 Groundwater Treatment Facility

This facility processes groundwater from the Quadrant I Groundwater Investigative Area and from the X-749/X-120/PK groundwater collection systems, by using activated water and green sand filtration. In 1999, the unit processed about 7.6 million gallons of groundwater, but removed only 1 gallon of TCE or 0.13 gallon TCE per million gallons groundwater. Compared with the contamination and the sizes of the two groundwater plumes, this seems very little.

X-622T Groundwater Treatment Facility

Contaminated groundwater from the basements of the X-700 and X-705 buildings are treated here, both of which are part of the Quadrant II Groundwater Investigative Area. The treatment process uses activated carbon and extracted 2 gallons of TCE from 10.7 million gallons groundwater in 1999, or 0.19 gallons TCE per million gallons water. However, measurements from 2000 showed concentrations of up to 7,000 µg/L in the X-705-BSW basement sump, or 7 ppm [203]. This value is worrying in two ways: First, it is 1,400 times over the MCL (Maximum Contaminant Level) as issued by US EPA; second, the treatment does not seem to work very well, because of the 7 ppm TCE, only 0.19 seem to be removed before discharge.

X-623 Groundwater Treatment Facility

This facility treats groundwater from the X-701B Holding Pond plume, using an air stripper with off-gas activated carbon filtration, and aqueous-phase activated carbon

202 EQ Midwest, Environmental Report 1999 for PORTS, p. 6-24 to 6-28.

203 USDOE, *Independent Investigation Volume 2*, p. 23.

filtration. In 1999, 55 gallons of TCE were extracted from 3.3 million gallons of groundwater, or 16.67 gallons TCE per million gallons water.

X-624 Groundwater Treatment Facility

This facility uses the same treatment methods as X-623 and treats water from the Interceptor Trench associated with the X-701B plume. The 1999 ratio (gallons TCE/million gallons water) was 11.38. Measurements from the Interceptor Trench sumps in 2000 showed concentrations of 24,000 µg/L or 24 ppm TCE. This concentration is 4,800 times over the MCL, and of the 24 ppm, less than half seem to be removed in the treatment process before discharge.

X-625 Groundwater Treatment Facility

This facility treats groundwater from a horizontal well associated with the X-749X-120 groundwater plume, as does X-622. The treatment technique in 1999 was mainly passive media, such as iron filings. Before discharge, the water passes an additional activated carbon filter. In 1999, 0.015 gallons of TCE were removed from 142,000 gallons groundwater, or 0.11 per million. Samples from the horizontal well in 2000 had concentrations of 190 µg/L or 0.19 ppm TCE, which is more than the 0.11 removed by the treatment facility.

The two treatment facilities that receive groundwater from the X-701B Holding pond plume clearly remove the greatest amount of TCE per extracted water. This seems to be due to the much higher concentration of TCE in the groundwater from the X-701B plume, rather than due to a more efficient treatment system, as the calculations for X-623 and X-624 show.

Technology Applications

Like the groundwater treatment systems, the following applications are applied to remove TCE and other VOC from the groundwater. Radionuclides cannot be extracted along with these contaminants, because their chemical and physical characteristics are completely different than those of VOC.

Steam stripping and hydrous pyrolysis/oxidation

Steam Stripping was applied to remove VOC from the groundwater by injecting steam underground through multiple wells, thus heating the area to above the contaminants' boiling point. The vaporized contaminants were then to be removed by vacuum extraction wells. Another process, called hydrous pyrolysis/oxidation would destroy the

contaminants that were not removed by the extraction wells. The system operated between January 29 and June 12, and removed 68 gallons of TCE -or 80 % of the total amount estimated by PORTS- from the X-701B Holding Pond plume, and the process was described as being a viable alternative [204]. However, during the subsequent monitoring, TCE levels in the plume were not found to be decreased, reaching concentrations of up to 670,000 µg/L at well X701-08G, or 134,000 times the drinking water standard of 5 µg/L [205]. Further, the process was expensive, costing about \$6 million to remove 825 pounds of TCE. [206]

X-701B *in situ* chemical oxidation

This was another process to remediate VOC in groundwater. Chemical oxidants were injected into the ground in order to transform TCE into nontoxic chemical compounds. A test in 1999 indicated that the reaction worked, but the system was shut down because the oxidant sodium permanganate was moving from below the ground, where it was injected, to the surface [207].

X-749/X-120 vacuum enhanced recovery systems:

This application uses a vacuum to increase the amount of water that can be pumped from a groundwater recovery well, and is useful in groundwater areas that are mainly made up of clay, which has a low permeability and makes it difficult to extract water. In addition, the movement of air due to the vacuum helps chemicals in the groundwater to volatilize, i.e. to move from the soil or water into the air [208]. Results from this pilot project have been incorporated for consideration as alternatives in the QI and II Corrective Action Study/Corrective Measure Study (CAS/CMS) reports.

Phytoremediation system

This system was used at the X-740 Oil Handling Facility to extract VOC from the ground [209]. Planting of 765 1-year-old poplar trees was completed in March 1999. Beginning in 2001, the trees are expected to have roots that are mature enough for this process. Organic compounds are expected to be captured in the trees' root system and transported to the leaves, where ultraviolet light degrades them as they are transpired

204 EQ Midwest, *Portsmouth Annual Environmental Report for 1999*, DOE/OR/11-3052&D2, Jan 2001, p. 3-6.

205 EQ Midwest, *Portsmouth Annual Environmental Data 1999*, DOE/OR/11-3053&D1, Jan 2001, p. 4-20 to 4-24.

206 DOE Portsmouth Environmental Bulletin, January 2000, p. 2.

207 EQ Midwest, *Environmental Report for 1999*, 2001, p. 3-4 f.

208 *Ibid*, p. 3-5.

209 *Ibid*, p. 3.2 f.

along with the water vapor. The system has yet to be monitored and evaluated to examine its effectiveness.

Low-level waste cell

A feasibility study is under way that could result in the construction of a low-level waste cell 210. Such a cell is designed to keep surface water from infiltrating into the material stored inside the cell, and if water seeps in nevertheless, then it is drained to a catch basin. There, contamination levels can be examined. Presumably, this cell should be built above the water table.

Preliminary Remediation Goals

Preliminary Remediation Goals (PRG's) developed for future residential property use of the site were 46 pCi/g for U-234, 0.17 pCi/g for U-235, 1.1 pCi/g for U-238, and 590 pCi/g for Tc-99 [211]. For total uranium, a PRG of 4.7 ppm was assigned, assuming the isotopes were present in naturally occurring proportions. Comparing these PRG's with the initial RFI data, the only chemical of concern that PORTS identified at the X-705A/B site was uranium [212]. In our view, the selected PRG's are not protective of public health for future residents. That is, cleanup to the above PRG's guarantee that the Portsmouth site must be safeguarded essentially forever. We ran the DOE program RESRAD (v. 6.0) to estimate radioactive doses to a future resident of the Portsmouth site, using default assumptions for resident farmers and assuming a soil contamination level for Tc-99 of 590 pCi/g, the PRG for Tc-99. RESRAD calculates the annual dose rate from various pathways as a function of model inputs, including levels of contamination, climate, expected radionuclide intakes of future residents etc. The potential radiation dose due to Tc-99 alone is 734 mrem/y, far exceeding the decommissioning guideline of 25 mrem/y. The dose is primarily due to ingestion of well water, but 120 mrem/y is due to plant and milk ingestion. Including uranium and transuranics and the inhalation pathway would further increase the dose. Thus, the cleanup PRG's are much too high to protect the health of a future on-site resident. The PRG are designed to comply only with the limits of the industrial use of the site. This makes sense for the foreseeable future. However, because institutional control will be lost at some point, nobody knows what the future brings in 100, 500 or 10,000 years, it would be prudent to assume that at some point, people will be living on the site. Risk calculations at most DOE sites actually assume that institutional control will be lost, and calculate a future dose to a person living as a residential farmer on the examined site. This hypothetical person is called "maximally exposed resident". It is not clear to us why DOE did not choose this approach for the risk calculation at PORTS.

210 Yoakum R, Chillicothe Gazette, January 26, 2002.

211 *Preferred Plan for X-705A/B*, p. 29.

212 SAIC, pp. 14-15.

The RESRAD analysis of groundwater contamination due to high Tc-99 soil concentrations agrees with an analysis conducted by Lehman & Associates, according to which there is a high probability for downward movement of the X-701B groundwater contaminant plume. This plume would move below and to the east of Little Beaver Creek into the Berea [213], from where it could reach residential drinking water wells.

In May 2000, the DOE Oversight Team criticized PORTS' assumption of natural uranium isotopic ratios in its risk analysis, which led to an inadequate evaluation of remediation plans. The Oversight Team noted that PORTS seemed to have overlooked the fact that (1) it is a uranium enrichment plant and (2) an actual isotopic uranium result indicated 13% enrichment. Based on the RFI data and a natural uranium distribution assumption, it was determined that 100 yd³ of soil needed to be removed from the X-705 A/B area to meet cleanup goals. However, shortly after excavation began, enriched uranium was detected at levels between 6% and 12%. Clearly, the amount of soil that needed to be excavated had been underestimated, and the extent and cost of remediation underrated. In the documents that we reviewed, no updated calculation about the amount of soil that needs to be excavated is given.

Another issue raised by the Oversight Team is the non-detection of transuranics in the X-705A and B samples during the Quadrant II RFI. Post-RFI analyses of some samples apparently indicate the presence of transuranics. Since PORTS did not detect transuranics, they were not considered in the risk analysis [214].

The focus of recent DOE reports has been on the decontamination and decommissioning of parts of the Portsmouth site. Soil and groundwater sampling and analysis, and risk assessments are being conducted in the present with an eye towards the future. But past air concentrations due to the incinerator and water concentrations due to the Holding Pond were much higher than they are today. Because of the latency period for hard cancers, ingestion and inhalation of radionuclides and toxic chemicals in the '60's and '70's could give rise to elevated cancer rates today. Residents near the Portsmouth facility have complained of elevated cancer rates that many of the DOE studies, focused on today's contamination, do not address.

In its budget document for FY 2003, the Bush administration proposed an 800-million dollar "reserve" fund to implement fundamental program changes for DOE's Environmental Management Program, with the expectation that the proposed reforms will improve cleanup efficiency by completing construction projects within baselines, reducing the cost of waste treatment and disposal, and integrating cleanup strategies across different sites [215]. As a response, DOE proposes to speed up remediation and cut costs by incomplete remediation of

213 Lehman L, *Opinions Regarding Assessments at Portsmouth Gaseous Diffusion Facility*, p. 1.

214 US Department of Energy, *Independent Investigation of the Portsmouth Gaseous Diffusion Plant, Volume 2*, Office of Oversight, May 2000, pp. 10, 12.

215 IEER Press release, February 4th, 2002.

its sites. The Institute of Energy and Environmental Research (IEER) argues that the administration is correct in being worried about the remediation efficiency of the DOE, but that it is not the Environmental Management program itself that has to be changed, but its implementation. Among the DOE propositions to cut costs are to mix of highly radioactive waste with concrete and bury it in trenches at Hanford, which would turn the site into a high-level waste dump. At other sites, dumps have been capped with concrete (including PORTS) and wastes cemented to cut costs, which creates worse long-term problems because the wastes can't be retrieved anymore.

G. Conclusions and Recommendations

In this report we reviewed past operation of the PORTS facility up to the present day remediation efforts. This report identifies the sources of groundwater contamination and highlights as well as possible with the available data past operations. Compared to other regional offices of the Department, its Oak Ridge Office has not been citizen friendly. Not till they were threatened with a lawsuit, did they relinquish documents almost a year after our FOIA request even though the documents are referenced in their publications.

The basic problem in earlier years and the reason for the lack of environmental data is that the Department was learning as they were going. Radionuclides, such as Tc-99, and hazardous chemicals, such as TCE, were initially put out into the environment with no containment. Over time, monitoring and containment was added, but "after the horses were out of the barn." Now it is a problem of costly catch-up and remediation.

What is obvious in retrospect is that large quantities of radioactive materials and toxic chemicals have entered the environment through the air and ground and surface water discharges. But monitoring of these discharges has been wanting. Monitoring for Tc-99 began in 1975. In earlier years, before incorporating MgF₂ filters and ion exchange resins, Tc was directly released to the environment. Most of the Tc was introduced to the Portsmouth plant from UF₆ received from the Paducah GDP. Similarly TCE degreasers, used throughout the DOE complex, were released to surface or ground water, or buried in landfills on the site.

Health effects due to radioactive and toxic chemical releases in the '50's and '60's would be showing up today in the form of cancer and other diseases to former workers and the general public. While the Department is right to carry out and take credit for the remediation effort taking place today, it should also recognize the legitimate grievances of former workers and the general public. The Department calculates today's low fence post dose to the public due to today's releases of radioactive materials. From estimates of the past amount of uranium and Tc-99 entering the Portsmouth facility and the amount of TCE used, and the containment technology available at the time, the Department could fairly easily estimate past fence post and on-site doses and calculate the risk to the public and former workers. We recommend that the Department and/or an independent group-do this

As for the future use of the site and the remediation standards used at Portsmouth, we strongly suggest that any risk assessment be based on the maximally exposed resident farmer scenario normally used at DOE site. It is simply irresponsible to assume that over the next thousands of years (i.e. before present contaminants are broken down or have decayed), DOE will retain control of the site. In contrary, it has to be expected that at some point in the future, people will live on the current site as subsistence farmers, growing most of their own food on-site and using local groundwater.

List of Acronyms and Abbreviations

AEC	Atomic Energy Commission
BJC	Bechtel Jacobs Corporation
CAS/CMS	Corrective action study/corrective measure study
Ci	Curie (measure for radiation)
CIP/CUP	Cascade Improvement Program/Cascade Upgrade Program
DCE	Dichloroethene
EIS	Environmental Impact Statement
ERDA	Energy Research and Development Administration
F	Fluorine
FY	Fiscal Year
GAT	Goodyear Atomic Corporation
GCEP	Gaseous centrifuge enrichment plant
GDP	Gaseous diffusion plant
H	Hydrogen
HEPA	High-efficiency particulate air
HEU	Highly enriched uranium
Gross alpha	Total alpha contamination from all sources
IRM	Intermedial remedial measure
LMES	Lockheed Martin Energy Systems
MCL	Maximum contaminant level
Mg	Magnesium
MMES	Martin Marietta Energy Systems
MTU	Metric tons of uranium
N	Nitrogen
Na	Sodium
Np	Neptunium
O	Oxygen
OEPA	Ohio State Environmental Protection Agency
OGDP	Oak Ridge Gaseous Diffusion Plant
ORO	Oak Ridge Operations of USDOE
PAH	Polycyclic aromatic hydrocarbons
PGDP	Paducah Gaseous Diffusion Plant
PORTS	Portsmouth gaseous diffusion plant
ppm	parts per million
PRG	Preliminary remediation goals
Pu	Plutonium
RCRA	Recourse Conservation and Recovery Act
RCW	Recirculating cooling water
RFI	RCRA Facility Investigation
RU	Recycled uranium
SODI	Southern Ohio Diversification Initiative
Tc-99	Technetium 99
TCA	1,1,1-Trichloroethane
TCE	Trichloroethylene
TEVE	Thermally enhanced vapor extraction

TRU	Transuranic elements
U	Uranium
UF4	Uranium tetrafluoride
UF6	Uranium hexafluoride
USDOE	U.S. Department of Energy
USEC	United States Enrichment Corporation
USEPA	U.S. Environmental Protection Agency
VOC	Volatile organic compounds
wt	wet tons

References

1. Blackledge, CP Jr. *Analysis of Samples Collected during Experimental Oxide Conversion Process* (GAT-923-77-321). Nov 3, 1977.
2. Blackledge, CP Jr. Memo to JP Vournazos, *Oxide Conversion Facility* (GAT-923-78-197). Sept 13, 1978.
3. Blackledge, CP Jr. *Transuranic Elements Committee* (GAT-923-77-13). Jan 21, 1977.
4. Brown, RW. *Contamination from Transuranic Elements* (GAT-922-76-184). Dec 30, 1976.
5. CDM Federal Programs Corp. *Air Pathway RCRA Facility Investigation Report for the Portsmouth Gaseous Diffusion Plant, Piketon, Ohio, Vol. 2* (DOE/OR/11-1350/V2&D1, POEF-ER-4614-V2&D1). Feb 1995.
6. Clary, AH. *Transuranic Test Run*. Sept 12, 1977.
7. Collier, R and D Smith. *Trip to Portsmouth: Transuranic Study – July 1978 Test Run and Employee Complaint regarding X-330 Cold Recovery Operations*. Sept 25, 1978.
8. Coyle D, Finaldi L, Greenfield E, Hamilton M, Hedemann E, McDonnell W, Resnikoff M, Scarlot J, Tichenor J. *Deadly Defense-Military Radioactive Landfills. A Citizen Guide by Radioactive Waste Campaign, New York*. 1988
9. Crawford, JG. *Fluorination of All Enrichments of Uranium Oxides* (GAT-P-43). June 24, 1968.
10. Cross, PE, TE Noel, and SJ Wethington. *Preliminary Study Oxide Conversion, X-705*. Dec 12, 1978.
11. Davis M. *A Guide to Key Facilities and Sites at the Portsmouth Gaseous Diffusion Plant*, October 2000.
12. Dept 841. Memo to RR Miller, *Technetium trapping of side stream, X-326*, (GAT-841-81-216). July 13, 1981.
13. Emler, VS. *Experimental Processing of Transuranic-Contaminated Uranium Oxide* (GAT-920-77-86). Aug 16, 1977.
14. Emler, VS. Memo to JG Crawford, *Transuranics in Solution Recovery Materials* (GAT-920-76-76). Dec 28, 1976.
15. EQ Midwest. *Portsmouth Annual Environmental Report for 1999, Piketon, Ohio* (SOE/OR/11-3052&D2). Jan 2001.
16. EQ Midwest. *Portsmouth Annual Environmental Data for 1999, Piketon, Ohio* (DOE/OR/11-3053&D1). Jan 2001.

