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Characterizing Radionuclides in the B Plant HEPA Filters

P. E. Roege
B&W Hanford Company

D. W. Wootan
Fluor Daniel Northwest, Inc.

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H. Author/Requestor

Paul E. Roege
(Print and Sign)

Responsible Manager

Robert E. Heineman, Jr.
(Print and Sign)

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P. E. Roege
B&W Hanford Company
P.O. Box 1200, S4-66
Richland, Washington 99352
(509) 372-0443 / fax (509) 372-1836
email: paul_e_roege@rl.gov

D. W. Wootan
Fluor Daniel Northwest, Inc.
2920 George Washington Way, H0-31
Richland, Washington 99352
(509) 376-4635 / fax (509) 376-1293
email: david_w_wootan@rl.gov

ABSTRACT

B Plant was built during World War II to separate plutonium for nuclear weapons from reactor fuel. Later, the plant was re-equipped and used to separate radioactive fission products from the Hanford Site's nuclear processing waste tanks. The facility is now being deactivated: eliminating, stabilizing, and documenting existing hazards to allow safe surveillance and maintenance pending a final disposition which is yet to be determined.

The processing areas of the plant, including process cells and exhaust air system, are heavily contaminated with radioactive cesium and strontium from the tank waste separation process. However, detailed characterization is difficult because many of these areas are inaccessible because of physical barriers and high radiological dose rates. The five existing canyon high efficiency particulate air (HEPA) filters were thought to contain a significant fraction of the inventory, but estimates were highly uncertain. This paper describes the process used to inspect and characterize the radionuclide content in one of these filters.

The investigation required a collaborative effort among field and technical personnel. Sophisticated computer modeling and detector technologies were employed in conjunction with sound radiological control and field work practices. The outcome of the effort was a considerable reduction in the filter inventory estimate, accompanied by a greatly improved level of confidence in the data. The information derived from this project will provide a sound basis for future decisions regarding filter disposition.

BACKGROUND

B Plant was built in 1943-44 to separate plutonium for nuclear weapons from reactor fuel as part of the Corps of Engineers' Manhattan Project. During the

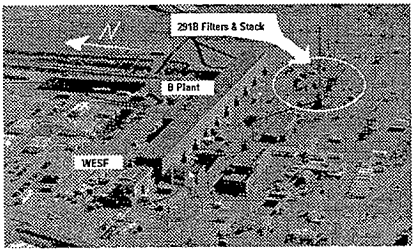

Figure 1 - B Plant and Filters

1960s, the B Plant canyon building was re-equipped and used to separate high-heat components (^{137}Cs and ^{90}Sr) from the Hanford Site's nuclear processing waste tanks. Although the separation mission ended in 1983, the facility remained in a standby condition until, in October, 1995, the U. S. Department of Energy ordered the facility shut down. B Plant is now in the process of deactivation, which involves eliminating, stabilizing and documenting existing hazards to allow safe surveillance and maintenance of the facility for an extended period pending final disposition.

The process areas of the plant, including process cells, canyon and exhaust air system, are heavily contaminated with radioactive cesium and strontium from the tank waste separation process. However, detailed characterization is difficult because many of these areas are inaccessible due to physical barriers and high radiological dose rates. The five existing canyon HEPA filters were believed to contain a significant fraction of the radiological inventory in the plant, but estimates were highly uncertain.

This paper describes the process used to inspect and characterize the radionuclide content in one of these filters. The facility built a diverse team which was equipped with both the theoretical and the field capabilities necessary to do the job. The team executed the task using sophisticated computational tools and sound field work practices. The characterization effort proved a success, not only in terms of its data output, but as a demonstration of how successfully to integrate scientific and field endeavors.

B Plant Filters

The first canyon exhaust HEPA filters were built in the 1960s, replacing the existing sand filter and increasing the exhaust system's capacity to support the facility's cesium/strontium separation mission. Due to the nature of the process, the filters were exposured to nitric acid vapors and a sometimes rapid buildup of radiation laden aerosols and particulate matter. In anticipation of this severe service and high radiation fields, the filters were built in underground chambers. New filters could be added alongside, leaving the old filters retired in place. The filters received letter designations, with A and B Filters the first and F Filter the last to be built.

D Filter was operated from 1979 until 1996, when E Filter was activated. In order to extend its service life, D filter was designed with two stages of 85% efficient prefilters in series, followed by three HEPA filters. All but the two final banks were designed with movable sections, allowing the each stage to be bypassed, once the filter airflow resistance built up to its operating limit. All three movable filter banks had been bypassed over the years, allowing significant dust loading on all but the final filter.

Throughout the plant's processing history, the staff maintained records of radioactive material inventories, including estimates of the exhaust filter loading. However, the estimates were based primarily upon processing losses and a limited number of radiation readings taken inside the filter housings. Because of the difficulty in obtaining measurements and the limited calculation tools,

the resulting filter inventory estimates contained significant uncertainties. D Filter was particularly uncertain, given 12 years of operation since its last radiation reading.

In 1992, the facility's engineering staff recognized the potential for radiation damage to some of the materials of construction in the filters, due to the large inventory they held. Each of the first three filters (A, B and C) was estimated to contain on the order of 10^4 curies of ^{137}Cs, ^{90}Sr and their radioactive decay progeny. D Filter had a much larger, although highly uncertain estimate of up to 1.5×10^6 curies. This latter estimate was based upon radiation measurements taken when processing ended in 1984, with a conservative assumption that radionuclide deposition had continued at a linear rate.

D Filter thus contributed a large share of the risk calculated in the facility's safety analysis, particularly in the event of earthquake. Because of this predicted risk, the facility sought ways to reduce the vulnerability of the filters following facility deactivation. In order to confirm the technical bases for the safety analysis and for follow-on projects to stabilize or mitigate the filters, the facility elected to develop a more reliable inventory estimate for D Filter.

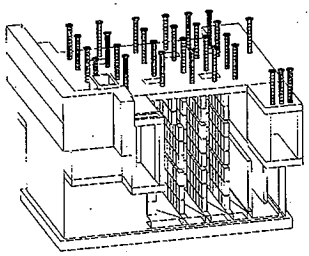

Figure 2 - 3-D Model of D Filter

Constraints

Clearly, the conditions inside the filter housing would be inhospitable. The radioactive dust collected over 17 years of operation would present a significant contamination hazard. Radiation doses inside the chamber were predicted to range into the hundreds of rad/h. The insertion and removal of the detection equipment would require well-planned technique to protect the workers and prevent contamination of the equipment, which would be re-used for each set of readings.

The geometry of the filter would require a complex calculational model. As stated above, D Filter consists of an underground chamber with a series of five filter banks. Each bank is loaded with 240 filters measuring 60cm x 60cm x 30cm deep. All but the final bank contained significant quantities of contamination, as did the walls, ceiling and floor of the filter housing. An accurate characterization would require a large number of readings, a complex three-dimensional model and an accounting for shielding due to the materials present.

Finally, D Filter was not designed to allow manned access or filter changes. Access to the filters could be gained through a number of 25 cm diameter inspection ports, which were plugged with 150 cm long sand-filled pipes to provide radiation shielding for workers during normal operation. When removed, these plugs would present the combined hazards of hoisting and rigging, contamination and dose.

Uncertainties

The primary unknown was how much deposited material had been loaded onto each filter, recognizing that HEPA filters sometimes double in weight due to deposited material. This material must be included in the calculation model.

The estimated inventory inside D Filter was highly uncertain, both in terms of total inventory and with respect to the relative split between cesium (gamma) and strontium (beta) isotopes. Radiation detectors selected must be sensitive at the upper and lower ranges of expected field strength; immediate feedback was required to adjust exposure times for cumulative devices (thermoluminescent detectors). In order to avoid challenges in collection, handling and disposal of a physical sample, the project also required equipment and calculations which could determine the isotopic split from dose readings alone.

The actual conditions could not be determined until the filters' inspection ports were opened. These uncertainties would require a conservative but flexible approach to both the data collection and radiological work practices. The team addressed these issues through conservative planning, careful selection of equipment, mockups, early feedback and the extensive use of hold points.

APPROACH

Due to the complex geometry, limited physical access and severe radiological conditions, the project to characterize D Filter demanded a blend of state-of-the-art computational tools, technology and field work practices. In response to the challenge, the facility built a multi-disciplinary project team consisting of scientists, technicians, engineers, crafts and operations personnel. This team worked together to integrate the use of a three-dimensional computer model, radiation detection and video equipment and sound radiological field practices.

The technical approach was based upon earlier work (Lan, et. al., 1991) and depended upon several key assumptions, including:

- All of the radiation originates from ^{137}Cs or ^{90}Sr or their daughter products. This is likely a good assumption since they are the primary radionuclides present in B Plant.

- The geometry and material configurations in D Filter are known. The construction details of the filters themselves were available from drawings; video inspection at the time of the radiation measurements was used to verify the overall cell and filter configuration.

- The cesium and strontium were distributed uniformly across each bank of filters. All of the radiation was assumed to be emitted from the filters, with no contribution from the floors or walls. It is not known what the air flow patterns were in the filter cell; thus, it is possible that material preferentially deposited in the lower regions or settled on the floor and walls.

Equipment

Radiation detection and video equipment were selected to provide adequate data and mutual verification. One of the more notable elements of the characterization effort was development of the B Plant D-Filter Detector System (BDDS). This system provided the means to deploy detection devices into the filter, while minimizing exposure to radiation or contamination. The team developed and improved this system through an extensive series of discussions and exercises using a mock-up of a D Filter inspection riser. Video images taken inside the filter were also used to provide early feedback regarding the actual conditions.

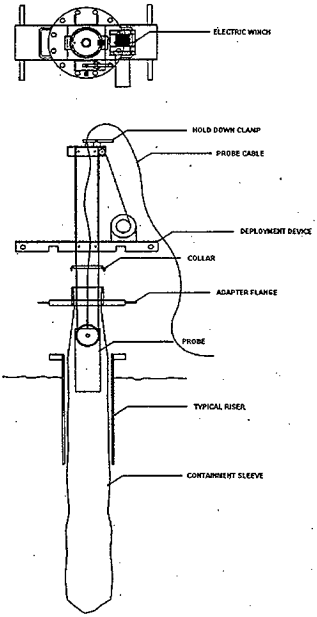

Figure 3 - B Plant D Filter Detector System

The B Plant D-Filter Detector System (BDDS), shown in Figure 3, includes the following components:

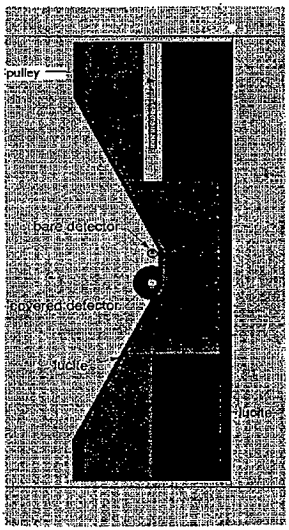

Figure 4 - Detector Probe Assembly

- The detector probe assembly with separate detectors for beta-gamma and gamma dose rates and a lead collimator. Shown in Figure 4, the assembly consists of a fixed lead collimator, a Lucite and aluminum container, a pulley, and two detector elements.
- A deployment device to lower and raise the probe assembly.
- An adapter flange with containment sleeve to provide contamination control for the equipment.
- Thermoluminescent dosimeter (TLD) strings mounted in nylon holders and attached to measuring tapes, which are deployed along with the probe.

Detector chambers

The detector assembly contains two thin-wall beta/gamma ionization chambers. The chamber wall is steel, and the fill gas is nitrogen. Each detector chamber is mounted horizontally along the axial centerline of the detector assembly. The top chamber is bare, while the bottom chamber is covered by Lucite to shield out all beta radiation. Electrical connections from both chambers pass through the Lucite covering to standard cable connectors located on the top aluminum plate.

Thermoluminescent Detectors

Thermoluminescent detectors (TLDs), similar to those commonly used for personnel dosimetry, were used to provide independent dose rate measurements. TLDs passively collect energy deposited by beta or gamma radiation; a TLD reader is then used to determine the cumulative incident radiation dose, based upon the release the stored energy in the form of light. Four strings of TLDs in holders were used. The TLD strings were made up of nine TLD holders arranged in a specific orientation. Each TLD holder contained nine standard lithium fluoride TLDs. The TLDs were mounted in holders, which were attached to a nylon measuring tape at 61 cm intervals, corresponding to the same axial elevations as the detector probe measurements.

Deployment device

The deployment device consists of an adapter flange, probe support stand, and an electric winch to lower and raise the probe. The adapter flange fits on top of the filter cell riser flange and also supports the probe containment sleeve. The containment sleeve is a plastic sleeve, closed and weighted at the bottom, that fits down the riser and provides a protected environment for the probe and TLD deployment. The support stand contains the winch and supports and orients the probe and TLD strings.

Orientation of the probe with respect to the filter cell is provided by clearly marked arrows that indicate the direction the probe is pointed. The probe cable and TLD strings are marked with stopping points for each axial measurement location. The TLD strings were deployed by attaching each string to the top of the probe, and lowering the probe assembly rapidly to the lowest probe measurement location. Because the TLD strings were attached to the top of the probe assembly, no TLD measurements were possible at the lowest probe measurement location.

Calibration

In order to interpret the measured radiation fields in terms of radionuclide inventories, it is necessary to establish a correlation between the measured and calculated detector response to known sources of both beta and gamma radiation. Calibration of the detector provides the means to convert the detector output current in picoAmps to a quantity that can be calculated directly, such as dose rate in rad/h.

Rather than working in absolute dose rates, the approach taken was to measure the detector response for a range of known source/detector configurations, and then calculate the detector response with a detailed computer model. Comparison of the measured and calculated responses provides a means for comparing the response of the detector in the actual filter cell measurements to calculated responses in the filter cell model. A limited calibration using available beta and gamma sources was conducted since it was not feasible to mockup the entire D Filter geometry and potential radionuclide inventory.

Multiple calibration factors were generated because the response of the chambers changed during the measurements and recalibration was necessary. The covered chamber was calibrated for gamma dose. The bare chamber was calibrated for both gamma and beta dose.

CALCULATION METHODOLOGY

D Filter Model

The D Filter cell was modeled in detail to provide a realistic evaluation of the radiation environment and source contributions to the detector response. Unfortunately, the team did not have the capability to import existing drawings from a computer-aided design (CAD) format for use in the Monte Carlo N-Particle (MCNP) analysis (Breismeister 1993). However, a recently developed visual editor was helpful in creating and checking MCNP input (Schwarz 1997). Initially, the model was based upon the available information in the plant drawings and discussions with plant personnel. Later, the model was updated using results from a video inspection done in conjunction with the dose readings.

During the planning process, the team's scientists performed a preliminary MCNP analysis of D Filter using the initial model and expected conditions. This analysis allowed the team to prioritize the radiation readings by riser number. Later, this information proved valuable, when two of the less important inspection points were found to be obstructed inside the filter housing. The team was able to bypass those readings, with little impact on the quality of the result.

Calculational model

The MCNP code was used to calculate the detector and TLD responses for both the calibration measurements and the D Filter measurements. An extensive computer modeling effort was required because of the difficulty in providing a calibration environment that was representative of the actual filter cell environment. Once the model had been shown to accurately compute the calibration configurations, the same techniques were used to account for the differences between the calibration configuration and the actual measurements. The models were used to provide the response functions to determine the radioisotope inventory that would produce the measured dose rates.

Version 4A of MCNP has been certified for use on Hanford site computers (Carter, 1996). However, problems were encountered with Version 4A using the combination of sources in lattices for the D Filter configuration. Discussions with the MCNP code custodian and Los Alamos National Laboratory indicated that the problem was likely related to known problems with Version 4A.

A Version 4B executable was obtained from Los Alamos National Laboratory (LANL) that did not have the same problems. No changes were made to the code to run at Hanford. To verify operation of this version of the code, the 25 LANL test problems used to verify the code were run and compared. In addition, selected D Filter cases that would run using Version 4A were compared with Version 4B.

Gamma Calculations

The penetrating nature of gamma rays from cesium results in sources throughout the filter cell contributing to each measurement location. The fact that the collimator allows some penetration also required that the backside contributions had to be calculated. Scatter off the walls, floor and ceiling also had to be included.

The approach taken was to calculate the dose rates in air for all of the measurement locations with a 1 Ci source uniformly distributed in a single filter bank. This was repeated for all five filter banks. Separate calculations with the same filter cell model but with the detector probe at selected locations were made to generate front and back side correction factors for each of the chambers. Similarly, TLD packages were included in the filter cell model to characterize the TLD response. Dose rates in the TLD package consisting of the TLD holder and the stack of TLDs were calculated and compared with the dose rate in air.

Beta Calculations

MCNP provides a full treatment of electron and photon transport. Unlike gamma rays or neutrons, beta particles (electrons) are electrically charged and thus interact with every atom they encounter. They are capable of producing large amounts of ionization as they penetrate a substance, losing energy with each interaction.

The D Filter model was used to calculate the beta dose rates from unit $^{90}Sr/Y$ and $^{137}Cs/Ba$ sources distributed uniformly in each of the filter banks. The beta particles are limited in range, so each riser location would generally only receive contributions from the adjacent filter banks. The highest energy beta particles would not penetrate from one side of a filter to the next.

Dose rates in air were calculated for all of the measurement locations. For selected locations, the detector probe was included in the model and the response in each of the detector chambers was calculated in order to determine a correction factor to account for the perturbing effect of the

probe. The response of the bare chamber to beta radiation in the D Filter environment was found to be 68% less than the dose rate in air. This includes the effect of the chamber wall, the plastic sleeve, and the collimator effects.

Variance reduction techniques such as varying the cell importances were used to increase the population of beta particles in the vicinity of the chambers. Energy cutoffs were used to increase the calculational efficiency by eliminating time spent on tracking particles that could not contribute to the detector response.

Sensitivities

Calculations were made to judge the sensitivity of the calculated responses to assumptions on filter densities. While the gamma calculations are fairly insensitive to assumptions on material densities, the beta calculations depend greatly on the accuracy of the geometric and material descriptions. These comparisons indicated that the calculated beta dose response factors, and thus the strontium inventory predictions, could easily be off by 50% just due to uncertainty in the filter densities. The filter screens were shown to have a major impact on the beta dose reaching the detectors.

Another effect that must be included in the analysis is the contribution of ^{137}Cs beta decay to the beta dose rate. For comparable amounts of ^{137}Cs and ^{90}Sr in the D Filter environment, the ^{137}Cs beta dose is a factor of 10 to 30 smaller than the beta dose from ^{90}Sr. Thus, large ^{90}Sr inventories would dominate the beta contribution. The beta energy from ^{137}Cs decay is smaller, so the beta particles penetrate less. For the D Filter analysis, the contribution of beta decay from ^{137}Cs was included.

DATA COLLECTION

The task was planned and executed by an established team of trained facility workers, using "enhanced work planning" techniques. This process involves all team members in the planning, and integrates all types of safety, health and environmental considerations with the work scope.

Models, drawings and mock-ups played important roles in the project. Because of the high level of attention given the filters, the facility previously had prepared a three-dimensional CAD drawing of the filters. The team used the model to familiarize the team, and as a visual aid for planning. In addition, the team used a full-scale mock up of one inspection riser to validate and refine its procedures, as well as the detector deployment system.

Through its planning and rehearsal process, the team identified a variety of innovative techniques to minimize contamination and radiation hazards. For example:

- The team specified and procured a prefabricated containment structure, which housed the

entire top of D Filter. This housing accommodated the entire job, without the need to rebuild the containment.

- Two workers used long-handled rollers to fix any contamination on the plug as it was hoisted from the filter inspection riser. The plug was then slid away from the hole and enclosed in a split section of PVC pipe for shielding.

- A prefabricated, disposable plastic sleeve was deployed into the open inspection riser to protect the detection equipment from contamination. After use, the sleeve was left inside the filter, greatly reducing the risk of contamination.

Detector Probe Measurements

The D Filter characterization measurements were conducted between November 19 and December 20, 1996. Video inspection of the interior of the filter cell was performed before the detector probe or TLDs were deployed. Dose rate measurements were made in nine inspection risers. Two risers originally planned for measurements were canceled after video inspection indicated a high potential for interference of the filter bank with the detector probe. Instead, two alternate risers were selected, based upon the measurements that had already been made.

During the inspection, the detector assembly was lowered into the filter cell between filter banks through the 25 cm access risers. Vertical scans were made with the probe facing both north and south in each riser, with the detector rotated 180 degrees to look at the filter banks on each side of the riser. The probe was stopped at each of the marked measurement elevations long enough for the detector output reading to stabilize before collecting at least five readings of the current output of the two chambers at each location.

TLDs

TLD strings were exposed for 30 minutes each. It took 90 seconds to lower or raise each string, so the total transition time was kept to about 10% of the exposure time. After exposure, the TLD strings were surveyed for contamination and then sent to the laboratory for reading. Control TLD packages identical to those on the exposed strings were also maintained and read.

Characterization Results

Dose conversion factors developed from the analysis of the calibration measurements were applied to the measured detector output. Linear regression analysis was used to perform a least squares fit of the calculated and measured detector responses for all of the measurement locations. This analysis provided estimates of the ^{137}Cs and ^{90}Sr inventories in each bank of filters, as well as the total filter cell inventory.

This assessment indicates that D Filter contains 56,000 Ci of ^{137}Cs with an uncertainty of 25%, and 7,000 Ci of ^{90}Sr with an uncertainty of 100%. Based upon these figures, the facility has revised its bounding estimate from 550,000 Ci to 70,000 Ci of ^{137}Cs, nearly an eight-fold reduction, and from 50,000 Ci to 14,000 Ci of ^{90}Sr, or a reduction factor of 3.6.

CONCLUSIONS

The project met its original goal to provide reasonable estimates of the inventories of ^{137}Cs and ^{90}Sr in the B Plant D Filter cells. In addition, it validated methodologies used in both theoretical and field endeavors; in fact, it provided a model for integration between the two worlds.

The analysis demonstrated a viable means for determining inventories and distribution of a mixture of beta and gamma emitting waste radioisotopes. The process of combining specific targeted measurements with extensive analytic analysis to unfold the contributing source can be applied to many different situations, and provides a means for determining upper bounds as well as best estimates.

The planning, preparation and execution of the filter inspection demonstrated the value of enhanced work planning. Taking advantage of the diversity of its members, the team worked together to understand the work scope, identify hazards, incorporate safety controls into the work process, and use rehearsals and feedback to improve the work process. As a result, the job proceeded with minimal delay, without injury or contamination; radiation exposures were well below historical expectations.

The inventories developed in this study are much lower than previous worst case estimates. Utilization of these lower inventories in future planning could significantly impact decisions based on risk, ALARA, waste generation, and eventual disposition alternatives.

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Louise Arnold - NPO
Dono Balero - NPO
Ted Bienerth - RCT, FDH
Gary Clark - engineer, BWHC
Victor Cruz - NPO, FDH
Mike Dawson - pipefitter, FDH
Marie Desoto - SOE , FDH
Paul Fiskum - pipefitter, FDH
Paul Gallager - radiological engineer, BWHC
Ed Gonsalves - cognizant engineer, BWHC
Lawrence Groce - NPO, FDH
Dave Harrold - RCT, FDH
John Hasson - haz waste coordinator, BWHC
Steven Hathaway - radcon support, BWHC
Elaine Hawkins - NPO, FDH
Bonnie Judy - RCT, FDH
Butch Judycki - RCT, FDH
Keith Keever - team coordinator, BWHC

Sarah Larrinaga - clerk, BWHC
Nancy Lenhart - NPO, FDH
Bernie Lueck - project activity mgr, BWHC
Mike Messinger - NPO, FDH
Dave Meyer - NPO, FDH
Jim Millbauer - pipefitter, FDH
Mark Murphy - detector dsn & calibr, FDNW
Rod Nelson - RCT, FDH
Tom Pedersen - video technician, FDNW
Glenn Ritter - deploymt system dsn, FDNW
Randy Robinson - SOE, FDH
Paul Roege - engineering mgr, BWHC
Kent Smith - project activity mgr, BWHC
Richard Smith - team coordinator, BWHC
Ron Sparks - video inspection, NHC
Jessie Weathers - RCT, FDH
Ron Weissenfels - engineer, BWHC
Ray Williams - radcon support lead, FDH
Steve Williams - NPO, FDH
Rebecca Wilson - safety engineer, BWHC
David Wootan - data collect. & anal, FDNW

Job titles:
NPO = nuclear process operator
RCT = radiological control technician
SOE = stationary operating engineer

Companies:
BWHC = B&W Hanford Company
FDH = Fluor Daniel Hanford Company
FDNW = Fluor Daniel Northwest, Inc.

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FIGURES

1. B Plant and Filters

2. 3-D Model of D Filter

3. B Plant D Filter Detector System

4. Detector Probe Assembly