Waste Operations Evaluations Project
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ABSTRACT

The DOE Western Environmental Technology Office (WETO) is supporting Oak Ridge National Laboratory (ORNL) by conducting an evaluation of the Liquid and Gaseous Waste Operations (LGWO) at ORNL for future waste generation.

In August 2003, UT-Battelle issued *The ORNL Liquid and Gaseous Waste Treatment System Strategic Plan* that provides their prioritized roadmap for the development of cost-effective and upgraded liquid and gaseous waste collection and treatment systems as a part of the revitalization effort to modernize ORNL. Waste management activities at ORNL for process waste, liquid low level waste, and gaseous waste are currently managed by Bechtel Jacobs Company LLC (BJC), the DOE Office of Environmental Management (EM) management and integrating contractor. DOE-EM’s mission is currently planned to end at the ORNL site when the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) Record of Decision requirements for Bethel and Melton Valleys have been implemented, which is expected to be in the 2015 timeframe. DOE-EM proposes to transfer responsibility for newly generated waste management to the U.S. Department of Energy Office of Science (DOE-SC) by 2006.

After the release of the *LGWTS Strategic Plan*, DOE-EM requested that MSE Technology Applications, Inc. (MSE) conduct an evaluation of the LGWO as an independent party to assist DOE-EM in their decision-making process to transfer waste management responsibilities to DOE-SC.

During fiscal year 2004, MSE designed a draft interactive model to create a material balance around the LGWO Facilities. The model consisted of a series of electronic spreadsheets showing the inputs, outputs, and interactions of all of the LGWO systems. MSE expanded on the model during fiscal year 2005 and used the model to evaluate ORNL’s future waste treatment needs. In addition to the interactive model, MSE determined the future waste generators, estimated volume of waste streams for fiscal year 2015, and conducted a literature search and evaluated alternative technologies for the current waste treatment processes.

This approach to consider the ever-changing waste stream contents and volumes was effective in supporting the decision-making process.

INTRODUCTION

This paper summarizes the work conducted by MSE in support of an evaluation of the LGWO at ORNL. MSE supported the DOE-EM in this effort by evaluating the current LGWO for future use, estimating the generation of liquid and gaseous waste in fiscal year 2015 (FY15), and searching for alternative technologies to the current treatment processes for treating the liquid and gaseous wastes.

MSE's evaluation focused on future systems for treating and disposing process waste, liquid low-level waste, and gaseous waste. This paper is based on a review of documentation describing existing and proposed waste streams generated at the ORNL, existing management and treatment processes, regulatory requirements, new and innovative treatment processes and best management practices. Specific recommendations are based on best available data and anticipated waste stream volume and contaminant concentrations. Changes to these parameters could significantly alter the preferred management and treatment strategy.

Background

ORNL is one of the nation's largest and most diverse energy research and development (R&D) institutions in the DOE laboratory complex. The DOE-SC is the ORNL landlord and is responsible for facility operations. UT-Battelle, LLC (UT-B) is the DOE-SC management and operating (M&O) contractor for the ORNL [1].

The DOE-EM is currently responsible for waste management activities at ORNL for process waste, liquid low level waste, and gaseous waste and remediation of the Bethel and Melton Valleys in accordance with the terms and conditions of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) Record of Decision (ROD). Bechtel Jacobs Company LLC (BJC) is the DOE-EM waste management contractor. DOE-EM's mission is currently planned to end at the ORNL site when the CERCLA ROD requirements for Bethel and Melton Valleys have been implemented, which is expected to be in the 2015 timeframe. DOE-EM proposes to transfer responsibility for newly generated waste management to DOE-SC by 2006.

The ORNL *Liquid & Gaseous Waste Treatment System (LGWTS) Strategic Plan* was developed by UT-B in August 2003 to provide an approach for cost effectively upgrading waste collection and treatment systems in support of the revitalization effort to modernize ORNL into one of DOE's premier 21st Century Laboratories [1]. The *2005 ORNL LGWTS Strategic Plan Update* summarizes modifications to the original strategy document that have resulted from new information, including results of the DOE-SC funded Operational Improvements Project and Health & Safety Improvements Program initiatives, operational changes at existing DOE-EM treatment facilities, DOE-EM plans for use and transition of existing treatment facilities to DOE-SC, and evolving ORNL hot cell consolidation and nuclear programmatic plans. The *LGWTS Strategic Plan* demonstrates DOE-SC's commitment to DOE and national pollution prevention and waste minimization goals by providing a plan for minimizing liquid and gaseous waste production, providing paths for disposal, de-centralizing treatment facilities, and dramatically reducing the dependence on buried tanks and piping. The plan provides specific recommendations and a road map for funding and scheduling.

After the release of the *LGWTS Strategic Plan*, DOE-EM requested that MSE conduct an evaluation of the LGWO as an independent party to assist DOE-EM in their decision-making process to transfer waste management responsibilities to DOE-SC.

In support of the evaluation, MSE developed an interactive model that contains a series of linked electronic spreadsheets to define the influents and effluents for all the LGWO systems. In order to develop a model to be used to evaluate future changes, we started with a baseline or "known" numbers. The average flow rates used to develop the baseline model were CY04 values from the Excel file *Waste Generation at LGWO CY04.xls* provided by the LGWO subcontractor Duratek [2].

The model contains six worksheets: a "map" of the model, Building 3544 Process Waste Treatment Complex facility, Building 3608 PWTC facility, LLLW facility, Gaseous Waste Treatment System (GWTS), and a flowchart showing the interconnections of the LGWO facilities. Each facility worksheet defines the waste stream generators (influent streams) to the facilities and the waste streams produced (effluent streams). Some of the effluent streams are influent streams to other LGWO facilities for treatment. These waste streams are linked between the worksheets and define the interactions between treatment facilities. This model can be used to examine the material balance for each LGWO system (Buildings 3544 and 3608, the LLLW system, the off gas system, and the cell ventilation system) as well as the interconnections between the LGWO facilities.

Once the model was complete, we started evaluating changes to the system to predict future use of the LGWO system. The cumulative and/or synergistic impacts associated with different options or decisions were evaluated based on material balances for the individual operations and the overall LGWO system as changes were made. The sensitivity of the system to the timing and magnitude of these changes was also evaluated. This type of information provided the technical basis for streamlining the LGWO operations as waste treatment needs change over time.

Present Waste Treatment System

The Liquid and Gaseous Waste Operations (LGWO) is a centralized system designed to treat liquid and gaseous waste streams emanating from research and operating facilities located in Bethel and Melton Valleys. The LGWO consists of the Process Wastewater Treatment Complex (PWTC), Liquid Low Level Waste (LLLW) Treatment System, and the Gaseous Waste Treatment System (GWTS).

The LGWO was originally constructed in the 1950's and remains in service today. Significant upgrades, as recent as the 1990's, have been implemented as necessary to prevent pollution and comply with more stringent environmental protection requirements. However, the system has become outdated, oversized, and portions of the system are rapidly deteriorating. Operation of this system and other facilities in the Bethel and Melton Valleys has led to widespread environmental contamination. Fig. 1 depicts the liquid and gaseous waste operations, the interactions between the individual treatment facilities, and the current usage of the LGWO system based on CY04 average flowrates.

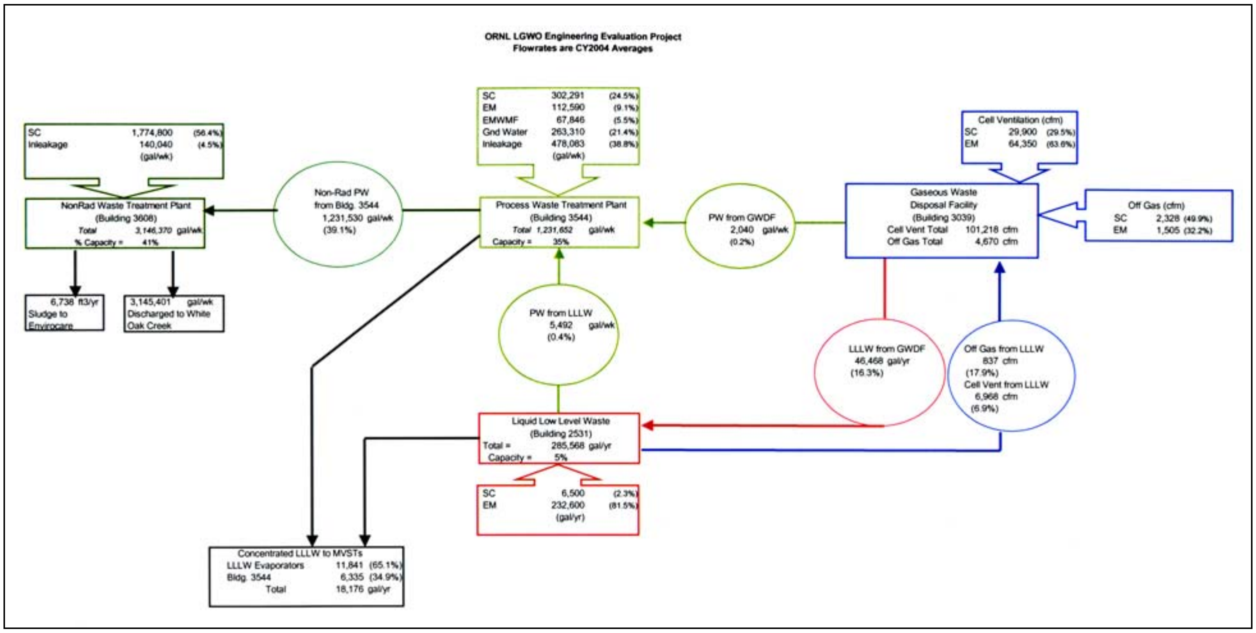

Fig. 1. Model depicting current (CY04) usage of LGWO.

Future Waste Treatment Needs

DOE-SC-funded operations are dependent upon the availability of a wide variety of facility infrastructure including experimental laboratories, hot cells, nuclear reactors and their associated waste collection and treatment systems to accomplish its mission of scientific research and technology development. One of these systems is the LGWO.

The *LGWTS Strategic Plan* is a management strategy for ORNL to consolidate new radioactive wastewater treatment systems in Melton Valley and eliminate the use of the aging centralized gaseous waste, process waste, and LLLW systems, which are primarily located in Bethel Valley on the ORNL Central Campus. Facilities to process newly generated R&D waste recommended in the strategic plan, are scoped to accommodate existing and new waste streams such as those expected to be generated by the Spallation Neutron Source (SNS) and proposed Nuclear Initiative programs. Nuclear and radiological R&D facilities in Bethel Valley will have local gaseous waste handling systems and local LLLW collection systems. This will allow upgrades to be implemented on a building-by-building basis as hot cell consolidation activities and the ORNL Ten Year Site Plan evolve.

The majority of the LGWO facilities are located in Bethel Valley on the Central Campus, which is the oldest part of the ORNL. Many of the buildings on the Central Campus have been shutdown, or are scheduled for shutdown, deactivation and destruction in accordance with the CERCLA ROD. DOE-EM remains on schedule to remove legacy waste and remediate the Bethel and Melton Valley areas as required by the ROD by 2015. These activities will dramatically reduced the volume of effluents and gaseous waste currently treated by the LGWO.

Fig. 2. is a flowchart from the interactive model showing the estimated future flowrates (FY15) for the current LGWO facilities.

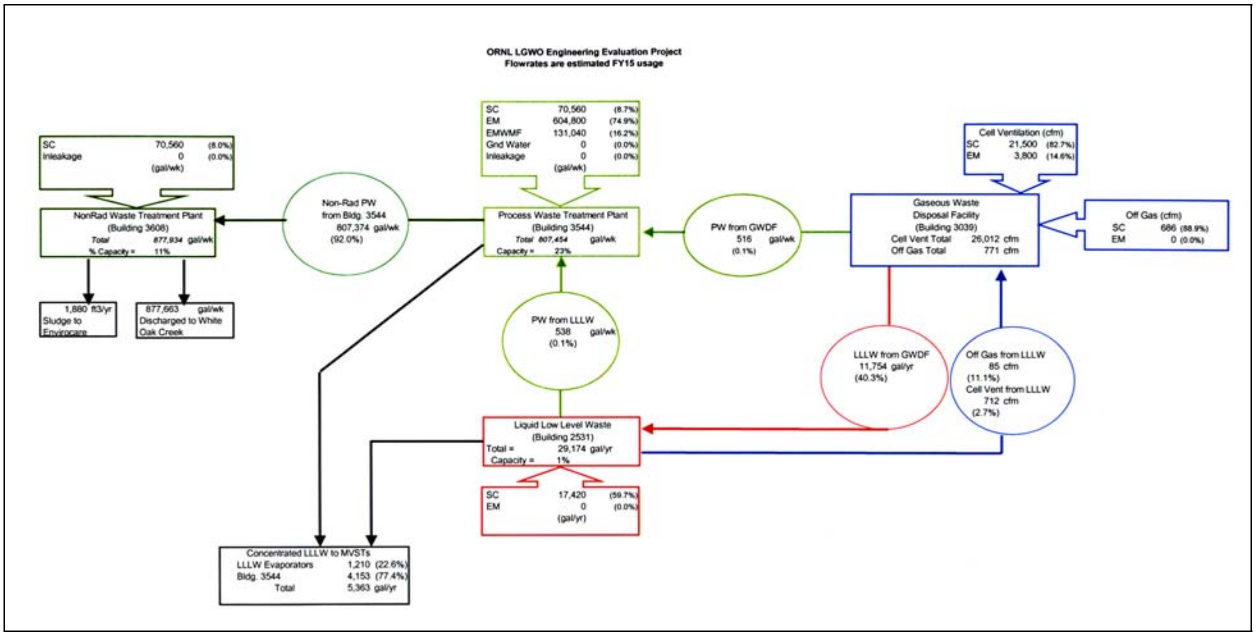

Fig. 2. Model depicting future (FY15) usage of LGWO.

PROCESS WASTE TREATMENT FACILITIES

Current Process Waste Treatment Facilities

The ORNL PWTC is designed to treat process wastewater generated at ORNL facilities in Bethel and Melton Valleys and condensate from the LLLW Evaporator Facility. Process waste is collected from a variety of sources including laboratory sink, floor, and hood drains; contaminated groundwater; rainwater from contaminated or potentially contaminated facilities, etc. Process waste is wastewater that is contaminated with radioactivity at concentrations below the defined minimum concentrations required for acceptance for treatment by the LLLW System. These minimum concentrations are defined in the *Waste Acceptance Criteria (WAC) for Systems Operated by the Liquid and Gaseous Waste Operations Project at Oak Ridge National Laboratory for the LLLW System, and PWTC Buildings 3544 and 3608* [3].

The PWTC consists of two facilities: the radiological wastewater treatment facility (Building 3544) and the non-radiological wastewater treatment facility (Building 3608). Building 3544 and part of Building 3608 provide the radiological contaminant treatment process for the PWTC. The radioactivity removal process consists of three basic operations: precipitation, filtration, and ion exchange. A precipitation operation was installed in late 1996 in Building 3608 to provide an additional throughput capacity at Building 3544. Nonradiological wastewater entering Building 3608 receives physical and chemical treatment to remove organics, particulates, and heavy metals. Treated effluent from Building 3608 nonradiological treatment facility is discharged to White Oak Creek through NPDES Outfall X12 [4].

The nominal treatment capacity of the PWTC is 760 gallons per minute (gpm). In CY04, the PWTC treated approximately 312 gpm (41% of capacity).

Building 3544 is designed to treat large volumes of slightly radioactive wastewater generated from different areas of ORNL to permitted discharge limits (i.e., WAC) before discharging the wastewater to Building 3608. It is optimized for Sr-90 removal but is also effective for Cs-137. Building 3544 also provides a backup capability to reduce the water hardness prior to radiological treatment [5].

The influent to Building 3544 is routinely softened at Building 3608 prior to transfer to Building 3544 for additional treatment. The composition of waste streams entering Building 3544 must meet criteria in *WM-LWS-WAC, Waste Acceptance Criteria for Systems Operated by the Liquid and Gaseous Waste Operations Project at Oak Ridge National Laboratory* [3]. The process consists of three basic operations: precipitation, which is typically bypassed since influent is softened at Building 3608, filtration, and ion exchange. Normally the precipitation is performed at Building 3608, but Building 3544 provides backup operation [5].

Typical contaminant concentrations in Bldg. 3544 influent are provided in Table I. The effluent from Building 3544 typically contains 1 Becquerel per liter (Bq/L) Sr-90 (99.9% removal) and 59 Bq/L Cs-137 (46% removal). Metals concentrations in the effluent are not measured, but the precipitation and ion exchange processes should be very efficient at removing heavy metals.

Table I. Typical Contaminant Concentrations In Building 3544 Influent

Contaminant	Concentration	Units
Strontium-90	750	Bq/L
Cesium-137	110	Bq/L
Cobalt-60	25	Bq/L
Europium-153	30	Bq/L
Zirconium-95	50	Bq/L
Silver	0.006	mg/L*
Arsenic	0.095	mg/L*
Cadmium	<0.005	mg/L*
Chromium	0.008	mg/L*
Copper	0.037	mg/L*
Mercury	0.0006	mg/L*
Lead	<0.2	mg/L*
Zinc	0.27	mg/L*
Total Organic Carbon	1.9	mg/L*
*mg/L = milligrams per Liter		

Building 3608 is designed to pretreat (i.e., soften) radiological process water, which is then sent to Building 3544 and to treat non-radiological process wastewater generated at ORNL to levels of pollutants specified in the National Pollutant Discharge Elimination System (NPDES) permit. Non-radiological process wastewater is treated for removal of organics, particulates, and heavy metals and to adjust the pH value of the wastewater prior to its discharge to White Oak Creek [5].

The nonradiological influent to Building 3608 for treatment comes from several sources and typically contains occasional surges of heavy metals [parts per million levels or less] and a fairly steady flow of a wide variety of organics at the parts per billion level. Sources of feed to the plant include drainage from laboratories, Building 3544 effluent, once-through cooling water, and aqueous streams from radiochemical processing plants and reactor operations. The composition of waste streams entering Building 3608 must meet criteria in WM-LWS-WAC, *Waste Acceptance Criteria for Systems Operated by the Liquid and Gaseous Waste Operations Project at Oak Ridge National Laboratory* [3], unless the waste stream is to be treated only by the clarifier (F-1006) and pumped back to

Building 3544. The WAC limits radioactivity levels in wastewater treated only in Building 3608 to less than the Derived Concentration Guide levels specified in DOE Order 5400.5 [5].

Table II shows the average metal concentrations in the influent to Bldg. 3608, the effluent concentrations at the NPDES discharge point, metal removal efficiencies, and the maximum daily limits for metals in the NPDES permit.

Table II. Building 3608 Influent and Effluent Metals Concentrations and Daily NPDES Limits

Contaminant	Concentration (mg/L)			
	Nonmetals Tank Influent	Effluent	Removal (%)	NPDES Limit
Arsenic	<0.05	0.0015	<97	0.14
Cadmium	<0.003	0.0001 6	<95	0.034
Chromium	<0.01	0.0017	<89	0.44
Copper	0.029	0.0057	80	0.11
Lead	0.090	0.0016	98	0.69
Mercury	0.001	<0.000 2	>80	0.0003
Nickle	<0.05	0.0014	<97	3.98
Selenium	<0.05	0.0025	<95	0.01
Silver	0.010	0.0001 7	98	0.008
Zinc	0.330	0.046	86	0.95

The wastewater from the nonmetals tank is treated by filtration, air stripping and granular activated carbon (GAC) adsorption. The Building 3608 metals tank wastewater is precipitated at a pH of 10.5 and clarified before it joins the nonmetals tank water. The flow rate of the metals tank wastewater is very low, averaging 8 gpm, with most of the wastewater coming from backwashing the filters and GAC columns [1].

Future Projected Process Waste

Four future process waste streams are anticipated to be above the UT-B process wastewater discharge criteria, including the:

1. High Flux Isotope Reactor (HFIR);
2. Radiochemical Engineering Development Center (REDC);
3. Spallation Neutron Source (SNS); and
4. ORNL Radiological Laundry [1].

In addition, the Melton Valley Groundwater, EMWMF Stormwater, and SWSA Leachate are expected to need treatment for radionuclides and/or heavy metals.

Projected future process waste generation flow rates from these sources and the UT-B proposed actions are provided in Table III.

Table III. Process Waste Future Projections

Source	Estimated 2003 Flow (gpm)	Estimated Future Flow (gpm)	UT-B Proposed Action
Radiological Process Wastewater			
Melton Valley Groundwater	100	50	Remediate to reduce flow
Cooling Water	33	0	Divert to storm drans
LLLW Evaporator	2	0	Remediate
HFIR	3	3	Treat in new facility
SNS	0	3	Treat in new facility
Radiological Laundry	1	1	Eliminate stream
EMWMF Stormwater	13	13	Treat in new facility
SWSA Leachate	0	60	Treat in new facility
Incidental Wastewater	2	0	Divert to sanitary
Total 3544 Wastewater	*54*	*80*	
Non-radiological Process Wastewater			
Manhole 190			
Cooling Water	164	0	Divert to storm drains
Incidental wastewater	2	0	Divert to sanitary
REDC	7	7	Treat in new facility
ESD	4	0	Divert to sanitary
Total 3608 Wastewater	*177*	*11*	

The projected future flowrate for the process waste treatment facilities is 87.5 gpm (11% of capacity of current facilities).

The process wastewater from the High Flux Isotope Reactor (HFIR) had an average flow rate of 2.2 gpm in early FY03, and contained:

- 420,000 Bq/L of tritium (H-3),
- 2,295 Bq/L chromium-51 (Cr-51),
- 15.5 Bq/L americium-241 (Am-241),
- low concentrations of several other metallic radionuclides, and
- nonradioactive Cr.

All of the radionuclides are below the Derived Concentration Guide (DCG) values in DOE Order 5400.5 except H-3 and Am-241[1].

The future estimated flow rate for the HFIR process wastewater is projected to be 3.0 gpm. Future contaminants are assumed to be similar to the contaminants listed above.

The process wastewater from the Radiochemical Engineering Development Center (REDC) had an average flow rate of 5.2 gpm in early FY03 and did not normally contain any measurable radionuclides or other contaminants [1].

Current activities at the REDC involve storage of uranium-233 (U-233), which generates a minimal amount of waste. The DOE Office of Nuclear Energy, Science and Technology (NE) is seeking a private sector contractor to process ORNL’s inventory of U-233 to extract thorium-299 (Th-299) to produce actinium-255 (Ac-255) and bismuth-213 (Bi-213) for medical applications. This will involve chemical processing activities comparable to those currently performed at the REDC, and has the potential to generate significant amounts of LLLW from FY06 through FY14. Future process waste generation will not be available until the contract has been awarded; therefore, MSE assumed the REDC process waste would be the same in the future.

The Spallation Neutron Source facility is expected to begin operation in 2007. The process wastewater from the SNS is expected to have an average flow rate of 2.6 gpm and contain a range of metallic radionuclides as shown in Table IV [1].

The ORNL Radiological Laundry generates 1 gpm of wastewater contaminated with Sr-90, Cs-137, detergents, and particulates [1]. It is assumed from the available information that the Radiological Laundry, generation will be the same in the future.

Table IV. Estimated Maximum Radionuclide Concentrations in Process Wastewater from the SNS and DCG Values

Isotope	Concentration (Bq/L)	DCG Value (Bq/L)
H-3	14,100	74,000
Be-7	17,100	37,000
Al-26	78	370
Cr-51	17,500	37,000
Fe-55	1200	7400
Mn-54	42	1850
Co-56	84	370
Co-57	61	3700
Co-58	141	1850
Co-60	6	185
Ni-63	66	11,100
Cu-64	110,900	11,100

Recommended Process Waste Treatment Facilities

MSE focused its evaluation on treatment technologies that treat process wastewater by producing clean water and concentrating the radionuclides in a filter or on a media that can be contained and disposed. Treatment options for each process wastewater will be discussed in the following sections. Treatment options consist of individual treatment units and/or combinations of various different treatment units. Most treatment units fall under the following categories:

1. Coagulation/filtration;
2. Ion exchange;
3. Media adsorption (i.e., zeolite); and
4. Mixing/dilution.

All alternative treatment technologies discussed below to treat the process wastewater at the ORNL were identified through literature searches. The alternative treatment systems were identified because they showed promise for treating process waters contaminated with heavy metals and radionuclides.

REDC Process Wastewater

Future projections indicate the REDC process waste stream will not contain significant levels of contaminants of concern. If these projections are accurate, MSE recommends considering direct discharge of REDC process wastewater to the sanitary sewer system.

HFIR Process Wastewater

The Am-241 and H-3 contaminant concentrations in the current and projected HFIR process wastewater exceed the DCG values in DOE Order 5400.5. MSE recommends conducting an as low as reasonably achievable (ALARA) evaluation as required by DOE Order 5400.5 and, if necessary, identifying a treatment process to remove H-3. Through the literature search, MSE identified standard technologies for separating tritium from wastewater(s).

MSE's evaluation of available tritium removal technologies indicates that treatment of the HFIR process wastewater for tritium may not be economically feasible (see Table V) [6].

Table V. Comparison Of Estimated Costs And Energy Consumption For Tritium Separation Processes

Process	Feed Flow Rate (Liter/min)	Capital Equipment Costs ($ in thousands)	Operating Costs ($ in thousands/yr)	Safety Concerns
Water Distillation	~100	10,000	--	Low
Hydrogen Distillation	8	20	12	High
Electrolysis	8	150	34	High
Combined electrolysis & catalytic exchange	95	340	200	Moderate
Girdler-sulfide	95	6.1	2.5	High

MSE also identified alternative commercial technologies to process tritiated water. These technologies are listed below.

1. Palladium Membrane Reactor (PMR), a catalytic reactor in combination with palladium-silver membranes [7].
2. Savannah River Site – magnesium based process called Z-bed Recovery; Circulation loop consisting of a metal-bellows pump train, a zeolite bed furnace, and a heated magnesium reactor, filled with 5 to 10 kg of reagent-grade magnesium turnings [7].
3. Cryogenic Distillation, Kinectrics Incorporated;
4. Palladium-Coated Kieselguhr
5. Hydrophobic Catalyst for Tritium Separation from nuclear effluents [8].
6. Molecular Separations, Inc.'s proprietary tritium loading bed [9].

In addition to the process to treat tritium, there will also have to be a support system associated with handling, sensors, and monitoring at the treatment facility.

If the removal of tritium is not an issue when considering the treatment of the HFIR, SNS, and REDC combination process wastewaters and only heavy metals concentrations are the concern, MSE recommends that a coagulation/filtration system may be a more appropriate treatment process to remove the heavy metals. To save money, an ion exchange process could be available on standby. If the radionuclide concentrations reach a pre-determined level, the process wastewater could then be diverted to the ion exchange treatment system.

SNS Process Wastewater

The radionuclide of concern in the SNS process wastewater is Cu-64 at 110,900 Bq/L compared to the DCG Value of 11,100 Bq/L (See Table IV). The *LGWTS Strategic Plan* states that the SNS should have lower concentrations of radionuclides than what is shown in Table IV; however, the 2004 SNS Waste Management Plan revision for future radionuclide concentrations was not available. The MSE-recommended treatment options for the SNS process wastewater are filtration combined with ion exchange or another effective adsorption media.

Radiological Laundry

The Radiological Laundry process wastewater is contaminated with Cs and Sr. The water also contains particulates and detergents, which make the process water inappropriate to mix with the HFIR, SNS, and REDC waters. Since the Radiological Laundry wastewater has Cs and Sr, MSE recommends treating the water at the collection site using a process consisting of coagulation/filtration, ion exchange and/or media adsorption.

MSE also recommends considering transferring the Radiological Laundry process wastewater following pre-filtration to either the Melton Valley Groundwater, EMWMF Stormwater, and/or Melton SWSA Leachate new treatment facilities. This option may be the most cost-effective approach to treat the Radiological Laundry wastewater if this wastewater did not negatively impact the new proposed groundwater treatment systems.

Melton Valley Groundwater, EMWMF Stormwater, and Melton Valley SWSA Leachate

Current treatment processes for the decontamination of groundwater usually involves the removal of the radionuclides by ion exchange on organic resins or inorganic zeolites. One of the drawbacks in using these technologies is that they are not adequately selective for the removal of radionuclides in the presence of large concentrations of alkaline and alkaline-earth metals typically found in groundwater. Consequently, the sorbents are quickly exhausted and generate large amounts of solid secondary waste.

Despite the problems noted above, ion exchange is the compelling choice for wastewater treatment, offering:

- Simplicity of operation in modular units or equipment;
- High waste concentration factors;
- Well-understood principles of operation; and
- An acceptable solid waste form [10].

Other Process Wastewater Treatment Technologies

Other treatment technologies that may be considered to treat radionuclide contaminated process wastewaters are listed below.

1. 3M Selective Separation Cartridge with Pre-Filtration – Applicable to most situations where ion exchange columns are utilized. The separation cartridges have been used in treating groundwater remediation systems, purge-water from groundwater sampling activities, decontaminate and decommission activities, reactor fuel storage and disassembly basins, and surface water treatment. Pre-filtration will remove particulates and the selective separation cartridges containing ion exchange resins will remove the radionuclides and heavy metals Cs, Tc, Sr, Co, Pb, and Cu. Cartridges to remove the radionuclides U and Pu are currently under development. [11]
2. Electrosorption and multiplayer adsorption/ion exchange column – has potential for removing Cs and Sr from wastewaters; however, limited information treating actual radioactive liquid waste was found.
3. AMED 1.2 Unit – to be developed under the Arctic Military Environmental Cooperation [12].
4. ECO-3 Mobile Liquid Radioactive Waste Treatment Unit – manufactured by Radon, Moscow (MOSRADON) [12].

LIQUID LOW LEVEL WASTE TREATMENT FACILITIES

Current LLLW Treatment Process

LLLW are those that are contaminated with radioactivity at concentrations above minimum levels as defined in the ORNL WAC document [3]. There are upper limits on total activity, two curies per gallon Sr-90 equivalent, and upper limits for total specific activity, 100 nanocuries per gram for transuranic isotopes and U-233. Also, solutions of U-233, U-235, Pu-239, or Pu-241, and other fissile nuclides must be denatured or isotopically diluted to certain ratios prior to disposal in the LLLW system. In addition to radioactivity, the wastes may contain nonradiological contaminants.

The LLLW Evaporator Facility receives wastewater from a variety of sources, including waste solutions from reactors, radioactive fuel and target processing facilities, decontamination operations, hot cells, the Gaseous Waste Treatment Facility wet scrubber, and analytical laboratories. Wastewater is transferred by means of vehicle or underground piping to the LLLW Evaporator Facility and accumulated in collection tanks in the LLLW Collection, Transfer, and Storage Facility. The LLLW treatment system is comprised of two 600-gallon-per-hour, steam-heated evaporators, numerous underground storage tanks, thousands of feet of underground piping and all the ancillary pumping, monitoring, and control systems required to service all LLLW generators in Bethel and Melton Valleys. The evaporation process operates at an average volume reduction ratio of 30:1[4]. Overheads from the evaporators are condensed and transferred to the PWTC for removal of radiochemicals from the evaporation process. Concentrate from the evaporation process is currently stored in underground storage tanks in Melton Valley.

The LLLW system currently processes approximately 225,000 gallons per year (gpy) and may be required to process up to approximately 500,000 gpy in the near future. Current LLLW volumes are shown in Table VI.

Table VI. Liquid Low Level Waste Generation Rates

Source	Present Waste Generation Rate (gpy)	Projected Waste Generation Rate 2015 (gpy)	UT-B/ DOE-EM Proposed Action
2026	3741	0	Consolidate at Building 7920 &4501
3025E	0	0	Consolidate at Building
3047	1	0	Consolidate at Building
3074	720	4,420	Move to Melton Valley and truck to new treatment facility
3525	0	5	Truck to new treatment facility
4500N	2	2	Bottle for new treatment facility
4500S	2	2	Bottle for new treatment facility
4501	8	250	Bottle for new treatment facility
7920-30	11,000	11,000	Hard pipe to new treatment facility
SNS	0	3,000	Truck to new treatment facility
3019	191	0	Treat processing waste in new temporary treatment facility and truck TRU waste to new treatment facility
3092	42,637	0	Shutdown prior to TPF shutdown
3517	37,095	0	Remediate
3544	5,746	0	Upgrade or replace to eliminate use of regenerable organic ion exchange resins
7900	7,500	0	Truck to new treatment facility
2008	4	5	Bottle for new treatment facility
Hot Storage Garden	200	0	Remediate
Solid Waste Storage Areas	650	0	Remediate
Tank WC-10	750	0	Remediate
Tanks T-1, T-2, and HFIR	74,000	0	Remediate
New Hydrofracture Facility	6,020	0	Remediate
Well Performance Assessment	2,250	0	Remediate or divert to new groundwater treatment facility

The current method of treatment is sufficient for the near term. The LLLW collection piping and treatment facility has been upgraded, and can be expected to support the Bethel and Melton Valley remediation as scheduled, and possibly the start-up of the SNS, with routine maintenance. However, as DOE-EM and DOE-SC continue to shutdown and decontaminate existing facilities, it will become increasingly obvious that the LLLW treatment system is no longer required.

Future Projected LLLW

After completion of remediation activities by DOE-EM and hot cell consolidation efforts, future LLLW quantities will be reduced to those generated by DOE-SC research and development programs. This volume is expected to decrease to approximately 20,000 gpy by 2015, as shown in Table VII.

Table VII. Projected Future LLLW Streams Contaminants and Concentrations

Isotope	SNS	Isotope	REDC	Manipulator Repair Shop
	Concentration (Bq/L)		Concentration (Bq/L)	
H-3	3.33E+10	Pu-238	6.70E+06	
Be-7	4.85E+11	Pu-240	3.40E+06	
C-14	1.08E+09	Pu-241	4.70E+07	
Na-22	5.22E+06	Pu-242	5.00E+04	
Na-24	6.22E+04	Am-241	1.50E+05	4.90E+04
Si-32	3.00E+04	Cm-244	1.80E+07	
P-33	3.48E+06	Cf-252	2.20E+05	
S-35	2.71E+07	Ce-144	1.80E+05	
Ar-37	1.26E+08	Co-60	1.00E+05	
Ar-39	7.55E+04	Cs-134	7.60E+06	1.20E+04
K-42	2.25E+05	Cs-137	1.60E+09	1.40E+06
K-43	6.62E+04	Eu-154	5.20E+04	
Ca-45	4.55E+06	Eu-155	4.50E+04	4.01E+04
Sc-43	1.10E+04	Nb-95	2.20E+05	
Sc-44	3.25E+04	Ru-106	2.40E+07	
Sc-46	7.88E+07	Sb-125	5.80E+05	5.40E+04
Sc-47	4.77E+06	Sr-90	5.10E+08	
Ti-44	1.68E+05	Zr-95	1.10E+05	
Ti-45	1.88E+03	Co-60		9.00E+03
V-48	2.90E+08			
V-49	4.66E+08			
Cr-48	5.00E+05			
Cr-51	7.44E+10			
Mn-52	2.03E+08			
Mn-54	2.09E+09			
Mn-56	2.21E+04			
Fe-52	4.29E+04			
Fe-55	2.31E+10			
Fe-59	1.85E+09			
Co-55	4.70E+05			
Co-56	1.00E+08			
Co-57	5.48E+07			
Co-58	8.25E+07			
Co-60	1.23E+07			
Ni-56	6.40E+05			
Ni-57	1.36E+05			
Ni-63	4.40E+08			

Each of the three major future LLLW generating facilities (SNS, REDC, and Manipulator Repair Shop) will generate a diverse low-level liquid waste stream as shown in Table VII. The projected future contaminants and concentrations shown in these tables are based on existing operations; these streams are not expected to change significantly.

Recommended Technologies for LLLW Treatment

MSE recommends individual source treatment systems for the REDC and SNS because of the diversity in the characteristics and quantities of contaminants in the projected future sources of liquid low level waste. DOE-SC should consider planning and implementing a "cradle to grave" treatment process for each individual waste stream as part of the laboratories of the 21^{st} century vision. Final treatment at the generator source promotes waste minimization and utilization of innovative waste treatment processes. Listed below are three technologies MSE evaluated for the treatment of LLLW.

- Shielded Hot Air Drum Evaporator (SHADE)
- Centrifugal Membrane Separation
- Selective Separation Cartridges

SHADE System

The Shielded Hot Air Drum Evaporator (SHADE) system combines evaporation, shielding, and contaminant storage in a single relatively inexpensive and disposable unit. SHADE systems are currently installed and operating at the Argonne National Laboratory West (ANLW) in the new LLLW treatment facility. The SHADE system offers numerous advantages over the existing LLLW treatment system. These advantages include:

- Right sized for the quantity of projected future LLLW;
- Low capital cost;
- Ease of operation;
- Containment of contaminants; and
- A pathway for solids disposal.

The SHADE system also has some disadvantages, including:

- The inability to effectively treat H-3;
- The cost of operation in terms of energy required to complete the evaporation process;
- The ratio of the size and weight of the material to be disposed versus the actual quantity of material removed from the waste stream;
- The individual SHADE units are not DOT approved and require extra size and bulk in the form of an over pack for shipping purposes; and
- The atmospheric release of the vapor phase effluent will require treatment and monitoring.

MSE has discussed operations of the SHADE system with operations personnel at ANLW and reviewed process throughput and level of contaminant concentration records from their operations. These discussions and records indicate the importance of pre-treatment, especially for the REDC LLLW in order to significantly reduce the level of contaminant concentration in the waste stream prior to entry in the SHADE system.

This SHADE system requires an ancillary system capable of providing up to 500 cubic feet per minute (cfm) of ambient air pre-heated to 250^{o}F and delivered to the SHADE system.

Other Treatment Processes

A number of new and innovative LLLW treatment systems and treatment technologies are emerging from the private sector as a result of government sponsored research and development. These systems are currently in use and being installed at other DOE facilities for treating waste streams similar to the ORNL pre-treated streams. These systems provide the same advantages offered by the SHADE system and deserve consideration and comparison.

Centrifugal Membrane Separation

Centrifugal filtration processes supplied by SpinTek Filtration Co. are currently in use and being installed for LLLW treatment at the Los Alamos National Lab and at the Savannah River Site. Savannah River is also working on a handling system for the SpinTek concentrate that should be available to other DOE sites when it is perfected.

The SpinTek Filtration unit features a ceramic membrane disk spinning at high speeds within a pressure vessel to filter various fluids without heavy build up on the filtration media. The unit concentrates aqueous solutions containing both dissolved and suspended solids and produces a concentrate up to 20-50% total solids. The unit feed is typically 1-10% solids. The SpinTek unit uses variable sized semi-permeable flat sheet membranes depending on the application. The unit will pass material from 300 molecular weight to 3 microns depending on the membrane used, and concentrate the material above this size.

The SpinTek field unit consists of one membrane containing one square foot of active membrane area. The permeate flow will vary from 25 to 500 gallons per day depending on the membrane used and the concentration and characteristics of the feed.

The field unit is small and easy to install, requiring only connection to the supply tank, 120 volt ac single phase 15 amp electrical service, and ½" concentrate and permeate discharge lines. The unit is 304 stainless steel construction and accepts fluid pH ranges from 0 to 14.

Advantages of SpinTek

- SpinTek requires far less space
- SpinTek does not discharge to the atmosphere
- SpinTek does not require a hot air delivery system.
- SpinTek does not require vapor phase filtration and monitoring.
- SpinTek concentrate can be captured and disposed in a container not grossly oversized for the quantity of concentrate contained.

Disadvantages of SpinTek

- SpinTek may require precipitation/coagulation prior to treatment.
- Spintek requires the treatment facility to develop and install a concentrate handling system.
- Spintek requires the treatment facility to provide an aqueous effluent path of disposal.

Selective Separation Cartridges

The 3M Company has developed a Selective Separation Cartridge designed to remove specific radionuclides from aqueous solutions. The cartridge is based on an innovative membrane technology made up of sorbent particles loaded or enmeshed onto a membrane. The membrane is fabricated into a spiral wound cartridge filter. Several classes of materials have been successfully incorporated into the membranes including ion-exchange materials, inorganic absorbents, zeolites, and sophisticated macrocyclic recognition compounds. Membranes have been developed for removing Cs, Sr, Co, and Tc. Membranes for removing U and Pu are being developed.

The individual cartridges can be installed as multiple elements in a single housing or as single elements in a single housing. They can be configured as parallel systems or series systems containing different cartridges. They have tremendous loading ability and would require shielding when used in multiple cartridge containers or as single cartridges removing Cs. This choice, i.e., shielding versus disposal, would be up to the user.

Advantages of Selective Separation Cartridge

- Compact light-weight design
- Ease of operation
- System design flexibility
- Faster flow rate
- Very low operating cost
- Ease of disposal
- Effluent treated to discharge standards

Disadvantages of Selective Separation Cartridge

- May require pre-filtration to prevent loading.

REDC Pre-Treatment

The REDC LLLW stream will require pre-treatment for the removal of high gamma and TRU elements. Pre-treatment will increase the throughput volume of a treatment system resulting in lower operation and disposal costs. The final design and success/failure of the new LLLW treatment facility correlates directly to the final design and success/failure of the REDC pre-treatment. For these reasons, MSE recommends constructing the new LLLW treatment facility directly adjacent to the REDC and considering it to be the REDC LLLW treatment facility. The facility would be managed and staffed by REDC operations personnel and designed primarily to treat REDC LLLW. The facility should also be designed with provisions to accept and treat similar waste streams such as the LLLW from the Manipulator Repair Shop and other small quantities from laboratories. MSE recommends treating the SNS LLLW separately.

SNS LLLW Treatment

SNS LLLW is not an acceptable waste stream for any of the proposed or alternative LLLW treatment processes previously discussed because of the concentration of the H-3 isotope. None of the processes are capable of removing the H-3 from the waste streams; therefore, the H-3 would pass through and be released to the environment. In addition, because of the concentration of H-3 in the projected SNS LLLW stream, remote handling and shielding during the treatment process could be required. Other contaminants will probably have to be removed from the SNS LLLW stream prior to H-3 removal (depending on the H-3 removal technology selected) to prevent fouling of the H-3 removal media. Removal of other contaminants could be accomplished by a centrifugal separation ultrafiltration, ion exchange, or zeolite depending on the particle size and other characteristics of the final waste stream. This type of process would allow for final treatment at the SNS.

The H-3 isotope in the SNS LLLW, at the concentration shown in Table VIII, is of high value to the DOE for strategic stockpiling when removed from the waste stream and concentrated. However, an economically feasible technology for detritiating and capturing the H-3 from the SNS waste stream is not presently available. One recognized method for recovering tritium from wastewater is cryogenic distillation and containment of the H-3 in metal hydride filled containers. This type of process, sized specifically for the SNS flow, would cost in the neighborhood of $3 to $5 Million for the main process components [13]. This estimate does not include facility, storage, ancillary systems or pre-treatment to remove other contaminants.

As an alternative to treating the SNS LLW stream to recover H-3, MSE recommends that the SNS LLLW stream be solidified at the SNS facility and land disposed. This technology appears to be the most cost effective solution for treating the H-3 waste stream.

Current research involving emerging new generation polymer sorbents has demonstrated cost-effective methods for using the sorbents to solidify aqueous waste streams and provides evidence of the ability of new generation sorbents to easily mix and completely sorb and solidify aqueous waste streams. In fact, mixing is not required for some of the polymer sorbents; the new generation polymers are simply added to the waste stream. Sorption is complete with no residual pockets of free liquid remaining in the waste form. This research also provides evidence of the sorbents ability to pass various liquid release tests that indicate the ability to pass shallow land disposal waste acceptance criteria and DOT transportation criteria.

These new polymer sorbents have been shown to be effective when mixed 1 part of polymer to 20 parts of water on a weight basis. However, regulations applicable to shallow land disposal mandate using twice the volume of sorbent required to completely sorb the liquid waste. For this application, assume a ratio of 1 part sorbent to 5 – 10 parts of liquid, by weight, as a conservative estimate for cost purposes. At this ratio we can expect a volumetric increase of approximately 50%. New generation polymers exhibit a specific gravity typically in the range of 0.5 to 0.7.

A process for treating SNS LLLW would involve adding the polymer to the LLLW at the correct waste loading ratio in a one-cubic-meter, high-density polyethylene (HDPE) shipping container in a shielded cell. The container would have to be sealed in the cell and the wall thickness would have to be sufficient to provide contact handling when removed from the cell for disposal. This waste form would most likely be classified as a Class B waste and suitable for disposal at a class B landfill.

GASEOUS WASTE TREATMENT FACILITY

The Gaseous Waste Treatment System (GWTS) at ORNL is a centralized system designed to accept and treat approximately 160,000 cfm of cell ventilation air and off-gas from research facilities in Bethel Valley. The system was installed in the 1950's and has not been significantly upgraded since. The system includes the centrally located off-gas stack and numerous above and belowground ducts. Approximately 1900 feet of underground ducting serving the 3500 and 4500 areas has serious leaks and periodically floods, releasing contaminants back into the surrounding soil and groundwater. Effluent from the wet scrubber, located at the central off gas stack area, discharges to the LLLW treatment facility. The system users currently utilize approximately half of the system capacity.

Bethel Valley Remediation and Hot Cell consolidation efforts are projected to reduce the future volume of gaseous waste generated from the current level of approximately 80,000 cfm to approximately 27,000 cfm by 2015 or 15% of the existing system capacity.

MSE recommends abandoning the existing GWTS and installing new stacks at Buildings 4501 and 3525. Discontinuing use and remediating the 3500 and 4500 area underground ducts should be considered a high priority. A wet scrubber may be required at the new Building 4501 stack; this requirement should be investigated and perhaps an alternative method identified to avoid generating an additional liquid waste stream requiring treatment prior to disposal. If the wet scrubber is mandatory, a path of disposal for the scrubber liquid should be determined.

CONCLUSIONS

Table VIII summarizes MSE's recommendations for modernizing the treatment of liquid and gaseous wastes at ORNL.

Table VIII. Summary of Process Wastewater Recommendations

Waste Stream	Recommendation
Process Wastewater	
SNS, HFIR, and REDC combined stream	Ion exchange
REDC	Discharge to sanitary sewer
HFIR	Coagulation/filtration
SNS	Ion exchange
Radiological Laundry	Coagulation/filtration w/ ion exchange and/or media adsorption
Groundwater	Ion Exchange
EMWMF	Ion Exchange
SWSA Leachate	Ion Exchange
LLLW	
SNS	Solidification via sorbents
REDC	Pre-treatment process and SHADE system, Centrifugal Membrane Separation, 3M Selective Separation Cartridges
Manipulator Repair Shop	Treat at REDC LLLW Treatment Facility
Gaseous Waste	
3500 Area	Build new stack
4500 Area	Build new stack

MSE recommends that DOE-SC develop policy and infrastructure designed to promote deployment of new and innovative technologies in the waste treatment, disposal and recycling arena. Waste treatment at the point of generation promotes accountability and professionalism at the generator facility, reduces cost, avoids confusion and creates the flexibility necessary to explore and implement innovative technologies, processes and procedures for streamlining the treatment process.

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