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Tritiated Water Disposition Strategies

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September 2020

SRNL-STI-2020-00332, Revision 0

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Printed in the United States of America

Prepared for
U.S. Department of Energy

SRNL-STI-2020-00332
Revision 0

Keywords: *tritium, water*

Retention: *Permanent*

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Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

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ACKNOWLEDGEMENTS

The authors gratefully acknowledge the funding and support from NNSA NA-231 Mo-99 Program Office.

EXECUTIVE SUMMARY

Tritium, a radioactive isotope of hydrogen, is typically confined within one or more confinement barriers. Tritium readily reacts with oxygen to form tritiated water or can exchange with protium or deuterium atoms in water to create tritiated water which is around 10,000 times more toxic than elemental tritium. Primary tritium confinement systems can create small volumes of highly tritiated water. Secondary confinement systems, like inert gloveboxes, will produce larger volumes of lower concentration tritiated water. Much larger volumes of tritiated water of very low tritium content are produced by tritium permeation/diffusion. Tritium from inside primary and secondary confinement barriers will diffuse from these systems into cooling water systems. Closed loop, recirculating cooling water systems will slowly accumulate tritium and create concentrations which exceed drinking water (or permitted discharge) concentrations. The cooling water systems designed as series of interconnected cooling loops that can slowly transfer tritium from one cooling loop to another cooling loop through heat exchangers. These large volumes of tritiated water may be above federal discharge limits and also lack convenient disposition pathways.

This document will discuss tritiated water management strategies from the perspective of large volume, low tritium concentrations that exceed drinking water or permitted discharge limits. Applicable US regulatory requirements for a Molybdenum-99 (Mo-99) production facility concerning tritiated water are also cited. Tritiated water disposition options, based on typical waste minimization strategies will guide dispositions options. A Case Study example is presented to illustrate the pros and cons of several disposition options. The report does not recommend any particular disposition pathway but discusses strategies to be explored by a producer to guide selection of processes which best fits the needs of a particular waste stream.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF ABBREVIATIONS

ALARA	As Low As Reasonably Achievable
AMSB	Ambient temperature molecular sieve bed
Bq	Becquerel
CANDU	Canada Deuterium Uranium
cc	Cubic centimeter
CFR	Code of Federal Regulations
Ci	Curies
CPCL	Cooling Pool Coolant Loop
DAC	Derived Air Concentration
DAC-HT	Derived Air Concentration for elemental tritium, 2×10^{-1} μCi/cc
DAC-HTO	Derived Air Concentration for tritium oxide, 2×10^{-5} μCi/cc
DF	Decontamination factor, initial concentration divided by concentration
FCL	Facility Coolant Loop
FSAR	Final Safety Analysis Report
HTO	Tritiated Water
L	Liter
LCO	Limiting Conditions of Operability
MSR	Methane Steam Reforming
NPDES	National Pollution Discharge Elimination System
NRC	Nuclear Regulatory Commission
NUREG	U.S Nuclear Regulatory Commission Technical Report Designation
ODCM	Offsite Dose Calculation Manual
PCL	Process Coolant Loop
PMR	Palladium Membrane Reactor
PRF	Permeation Reduction Factor
RCA	Radiological Control Area
RCACL	RCA Coolant Loop
REMP	Radiological Environmental Monitoring Program
RETS	Radiological Effluent Technical Specifications Program
TRF	Tritium Removal Facility
TRM	Technical Requirements Manual
TS	Technical Specifications
UFSAR	Updated Final Safety Analysis Report
WAC	Waste Acceptance Criteria
WGS	Water Gas Shift

WHO	World Health Organization

Additional Subscripts

i	Initial value
f	Final value
e	Enriched stream
d	Depleted stream

1.0 Introduction

In applications where tritium is handled, tritium confinement is performed using different barriers to minimize releases to the environment. For gaseous (elemental) and liquid (oxide) tritium, process piping and components provide the primary confinement function. Secondary tritium confinement is typically provided by inert (i.e. non-flammable gases such as nitrogen, argon, or helium) gloveboxes which are connected to a tritium stripper system. Tritium confinement will produce various amounts and concentrations of tritiated water/tritium oxide (e.g. HTO). Small volumes at high tritium concentrations can be produced in process system components. Secondary confinement systems (e.g. glovebox) tritium recovery/stripper systems many times rely on the creation and absorption of tritiated water to mitigate environmental emissions.

Tritium contaminated water is also created by tritium permeation/diffusion. Tritium from inside the process piping will diffuse through the primary confinement material and contaminate the glovebox atmosphere. Non-contact water cooling coils on heated process vessel flange connections water will become contaminated with tritium. Even water cooling lines penetrating a glovebox boundary or water chilled glovebox heat exchangers will slowly become contaminated with tritium from the glovebox atmosphere. Closed loop, recirculating cooling water systems will slowly accumulate tritium which can eventually produce large volumes of low activity water. Tritium concentrations in these systems can build-up and exceed drinking water (or permitted discharge) concentrations creating large volumes of water without convenient disposition pathways.

Cooling water systems, especially those used for radiological processes such as fission power plants, typically consist as a series of interconnected cooling loops to create a series of systems without a direct pathway for process contamination to be released outside the facility. This document will discuss tritiated water management strategies from this perspective based on a series of systems interconnected by heat exchangers. Applicable US regulatory requirements for a Molybdenum-99 (Mo-99) production facility concerning tritiated water will also be cited. Tritiated water disposition options, based on typical waste minimization strategies will guide dispositions options.

2.0 Regulatory Requirements

Facilities using tritium will eventually have tritium discharges whether they are solid, liquid, and/or airborne emissions – it is just the magnitude of the emissions that are in question. This report will focus mostly on tritiated water generated from indirect contact of tritium with non-contaminated water. Tritium activity will accumulate in several closed water-cooling systems pertaining to tritium processing and Mo-99 production through permeation through components. Several storage, processing, release and/or disposal strategies for tritiated water need to be developed. It is assumed the Mo-99 facility is licensed through the US Nuclear Regulatory Commission (NRC).

2.1 Applicable US Federal Regulations

Some of the US Code of Federal Regulation (CFR) relevant to the licensing and operations of a Mo-99 facility are listed below.

- 10 CFR Part 50 Domestic Licensing of Production and Utilization Facilities
  This includes construction permit and operating licenses. Also contains information on radioactive release limits to the public/environment in Appendix I.

- 10 CFR 70 Domestic Licensing of Special Nuclear Material

Possession and use of Special Nuclear Material - low enriched uranium if applicable to the Mo-99 production process.

- 10 CFR 30 Rules of General Applicability to Domestic Licensing of Byproduct Material
  Production of Byproduct Materials - Molybdenum-99
- 10 CFR 51 Environmental Protection Regulations for Domestic Licensing and Related Regulatory Functions
  Environmental Impacts
- 10 CFR 20 Standards for Protection Against Radiation
  Radiation Protection/Radioactive Releases/Radioactive Shipments
- 10 CFR 19 Notices, Instructions, and Reports to Workers: Inspections and Investigations

2.2 Applicable Implementation Methodologies

The previous section listed some of the regulations/requirements that must be met for the Mo-99 producer. Methodologies to meet these requirements are listed below.

- Regulation Guide 8.2 Administrative Practices in Radiation Surveys and Monitoring
- Regulation Guide 4.1 Programs for Monitoring Radioactivity in the Environs of Nuclear Power Plants
- NUREG-1301 Offsite Dose Calculation Manual Guidance: Standard Radiological Effluent Controls for Pressurized Water Reactors.

2.3 Required Licensing Documents

The pertinent requirements of the above regulations and the producer's methodologies for implementing and adhering to these requirements will be detailed in several "License" documents:

1) Final Safety Analysis Report (FSAR)
    a. Following major changes (site configuration or major regulation changes) can be renamed Updated Final Safety Analysis Report (UFSAR)
2) Technical Specifications (TS)
    a. Contains systems surveillance requirements, control parameters, and Limiting Conditions of Operability (LCOs)
3) Technical Requirements Manual (TRM)
    a. Contains additional systems surveillance requirements, control parameters, and Limiting Conditions of Operability (LCOs)
4) Offsite Dose Calculation Manual (ODCM)
    a. Is a license document that details how the Mo-99 producer will meet the requirements of 10 CFR 50, 10 CFR 20, and Reg. Guide 4.1. The document will contain surveillance frequencies for effluent and environmental sampling and analysis, Radiation Monitor acceptance criteria, and Radiation Monitor setpoint, release rate, and dose calculation methodologies and the implementation of Radiological Environmental Monitoring Program (REMP) and Radiological Effluent Technical Specifications Program (RETS)

All these documents will contain information on releasing radioactive material to the environment and the public.

3.0 Radiological Cooling Water Loops

Figure 3-1 illustrates the way interconnected cooling loops may be designed to minimize environmental releases with potential contamination levels of the water, decreasing from left to right in the figure. The highest concentration water is likely formed from cooling components which would be the primary confinement boundary for tritium and will be designated as the Process Coolant Loop (PCL). Another source of tritiated water may be a cooling pool of water, which may be used as radiation shielding but has the potential to become contaminated from tritium permeation from components in the pool or from atmospheric exposure to gaseous tritium/oxide in the surroundings. This loop will be designated as the Cooling Pool Coolant Loop (CPCL). The PCL and the CPCL would likely be closed looped, recirculating systems.

The PCL and the CPCL would reject heat to another cooling loop via heat exchangers. Indirect contact between the water in the PCL and CPCL and the next cooling loop via heat exchangers will reduce, but not eliminate, contamination of this intermediate coolant loop. This loop may be considered part of the Radiological Control Area (RCA), due to its potential for contamination and will be referred to as the RCA Coolant Loop (RCACL). The RCACL acts as an intermediate barrier between water in close contact with tritiated components and another facility coolant loop which rejects heat to the environment: designated as the Facility Coolant Loop (FCL). The FCL is assumed to be a "clean" (non-radiological) system which does not release radionuclides to the environment.

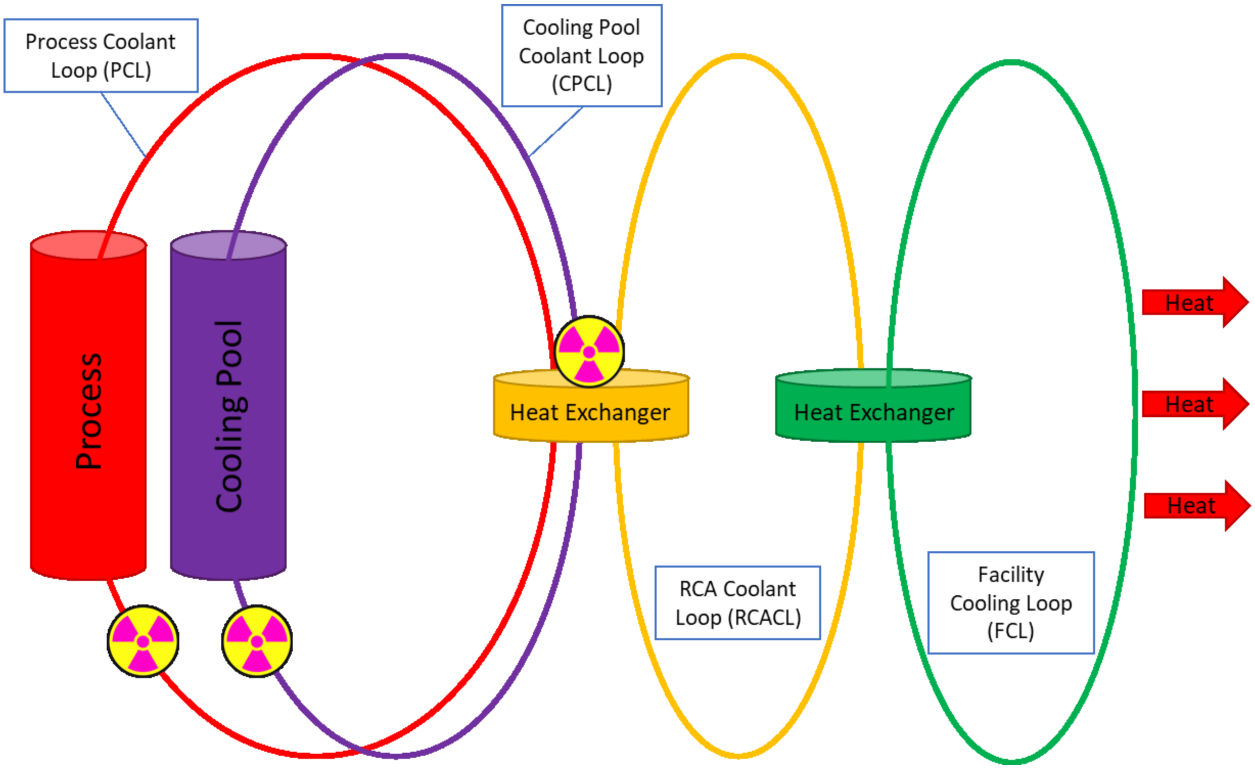

Figure 3-1: Typical Radiological Cooling Loops

4.0 Tritiated Water Perspective

Figure 4-1 shows tritiated water activity, in mCi per Liter (L) versus volume. The US drinking water limit is 20 pCi per cc (2×10^{-5} mCi/L) and is shown along with the World Health Organization (WHO) drinking water limit of 10,000 Bq/L (270 pCi/L) and the Canadian drinking water limit of 7,000 Bq/L (189 pCi/L). Water at or below the drinking water limit can be discharged but facility monitoring is required as part of obtaining an NRC license to operate. For comparison, tritium concentrations in heavy water CANDU reactors can cycle up to 50 Ci/L (5×10^{4} mCi/L) before the water is processed by a heavy water tritium removal facility (TRF). Decontamination factors (DFs) for TRFs run on the order of 50 to 100, so a lower level activity of 0.5 Ci/L is also shown in the figure. The volume of water for a single reactor is on the order of 10^{5} kg [Ref. 1] and is indicated in the figure by the double ended arrow between 0.5 and 50 Ci/L.

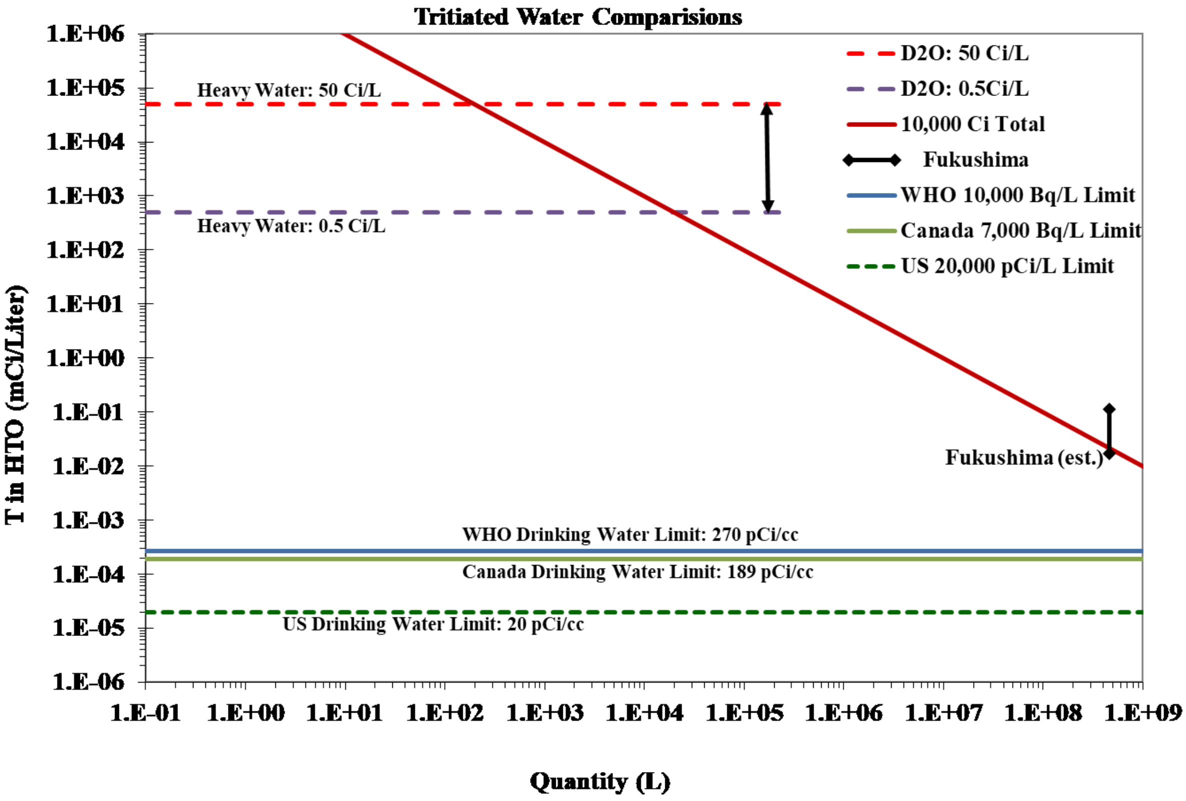

Figure 4-1: Tritiated Water: Concentration versus Volume Perspective

Quantities and concentrations of tritium contaminate water for Fukushima vary with one-time concentration values ranging between 6.3×10^{5} to 4.2×10^{6} Bq/L (1.7×10^{-2} to 1.14×10^{-1} mCi/L) in 460,000 m^3 (4.6×10^{8} L) of water and are also included in Figure 4-1 for comparison. The negative sloping line in the figure is for a constant quantity of tritium, chosen as 10,000 Ci (nominally 1 g of tritium) with the concentration calculated by dividing the total tritium content by the volume of water. This report will focus on tritiated water below the extremes of CANDU heavy water tritium concentrations and Fukushima volumes of water needing dispositioning.

5.0 Tritiated Water Management Strategies

The use of tritium will always have some emissions whether it is atmospheric, liquid, and/or solid/contaminated material discharges. Hydrogen atoms are present in almost all materials and migrate through materials. The low energy beta emissions of tritium decay supplies sufficient energy to catalyze reactions needing vastly different conditions to be observed with protium and deuterium. This is one of the reason why discussions regarding the use of tritium use the term "confinement" instead of "containment", since tritium atoms can be confined (i.e. their movement out of barriers slowed down) but not realistically contained (i.e. their movement out of barriers prevented), which is unobtainable and a matter of how well the material is confined.

Figure 5-1 is a plot of tritium concentration versus quantity of water and will guide the discussion in this section. As before, lines of constant tritium content are shown as negative sloped lines in the figure with values of 1, 10, 100, and 1,000 Ci shown. Additional horizontal lines are shown for vapor phase tritium concentrations for the derived air concentration (DAC) of tritium oxide (DAC-HTO) of 2×10^{-5} μCi per cc and the DAC of elemental tritium (DAC-HT) of 2×10^{-1} μCi per cc, converted to vapor phase water saturated at 22 °C and 760 torr to liquid water concentrations. The DAC-HT converts to approximately 1.03×10^{4} mCi/L (liquid) and DAC-HTO converts to 1.03×10^{1} mCi/L (liquid).

Typical waste minimization strategies employ the following hierarchy from most-to-least preferred options: 1) Prevent, 2) Minimize/Reduce, 3) Reuse, 4) Recycle, 5) Treat, and 6) Dispose. Tritiated water disposition strategies can utilize this hierarchy but some treatment technologies do not scale-up economically with volume for even relatively low concentrations of tritium. Technology choices for disposition of tritiated water will depend on the activity and volume of water with different options depending on the specific conditions to be managed. For this application Reuse will be defined as using tritiated water in the same or similar process without any chemical and/or physical treatment. Recycle will be defined as using tritiated water is the same or similar process following chemical and/or physical treatment.

5.1 Avoidance/Prevention

Creation of tritiated water can be prevented by use of another fluid as the heat transfer media or use of alternate cooling technologies (e.g. thermoelectric devices). Substitution of another liquid as the coolant for water may avoid creation of tritiated water but may create a different, but even harder to manage tritiated liquid stream. Use of air or another gas (e.g. nitrogen, helium) instead of water for the coolant will require larger cooling systems due to the lower heat capacity of gas relative to liquids – even when compressed to reasonable pressures and used at high flow rates. A gas phase cooling system will also require some tritium management strategies or detritiation technologies, but system designs may be more straight-forward than those of a water detritiation system.

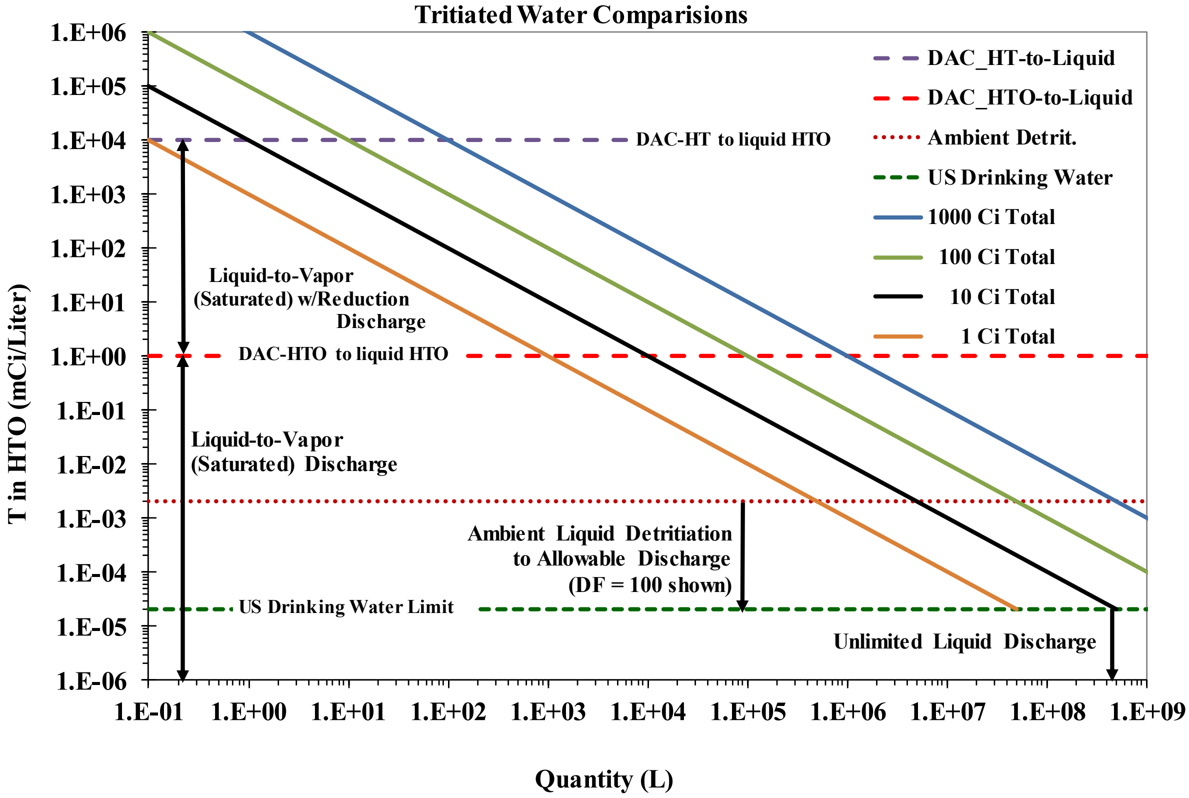

Figure 5-1: Tritiated Water Concentrations from DAC Values

5.2 Mitigation/Minimization

If the goal is to prevent creation of tritiated water exceeding the drinking water (or permitted discharge) concentration limit one can either reduce the amount of tritium entering the water or increase the volume of water which contains the same amount of tritium. Permeation barriers can be used to reduce the flux of tritium into the water but does not prevent it from entering the water. Permeation reduction factors (PRFs) can be on the order of 100's to 1000's but will still create tritiated water of lower activity than without use of a barrier in the same amount of time.

Whatever the flux of tritium into water, exceeding threshold values can be avoided by monitoring the tritium activity in the water and then replacing the water with fresh or reduced activity water before the threshold value is exceeded. For closed, recirculation cooling systems, replacement of the water before it exceeds drinking water limits will allow discharge of the water and the system can be refilled with fresh water. This is a variant of a single-pass cooling system where batches of water are used and then periodically discharged or purged from the system.

5.3 Reuse and Recycle

If tritiated water is created, methods in which the water can be reused should be considered. In this context, reuse is considered without detritiation processing, which is covered in another section of this document, or any other chemical and/or physical treatment. If water in one process exceeds a set-point concentration, it could potentially be reused in another contaminated system as a source of make-up water. The reuse of water would be from lower activity systems or processes to higher activity systems or processes. Although practical for typical industrial processes, this may be more problematic for radiological facilities. The volume of very low activity tritiated water typically exceeds the volume of higher activity streams by orders of magnitude, so complete utilization of the very low activity water may not be practical. The use of corrosion inhibitors or other chemical additives in some cooling water systems may also require pre-treatment of the water before reuse in another part of the process, further complicating the ability of the water to be reused in the process.

5.4 Treatment/Disposition

Treatment and disposition options will be presented in this section. Although not all options presented are practical, they are included in the discussion for completeness of the options considered.

5.4.1 *Decay Storage*

Storage of tritiated water is not a practical option but is included for completeness. Storage of tritiated water until it decays to allowable discharge levels is not practical due to the low required tritium concentrations. It takes approximately 41 years for tritium to decay an order of magnitude in concentration. For the Fukushima water shown in Figure 4-1, some of the water would need to be stored for around 123 years before it would reach the WHO drinking water concentration and 164 years before it would reach the US drinking water limit.

5.4.2 *Solid Phase Options*

Encapsulation, where the water is confined inside a waste package, is not considered a practical option, but is included for completeness. An example would be filling a stainless steel vessel with water and then welding closed the vessel. This is a near-zero volume increase strategy but many disposal facilities do not accept containers with liquids so this may not be practical.

Freezing of the water does little to aid in a disposition strategy. Freezing reduces the mobility/movement of the water. Ice walls have been created to reduce mobility of contaminated water already in the ground (e.g. Fukushima) but is not practical as a strategy for disposition of the water.

Commercial solidification products are available for converting liquid waste streams into solid waste streams. The amount of solidification agent is typically two to three parts per one part water, so the total volume increase of the waste is three to four times the initial volume.

Creation of cement or grout waste forms are also options. These waste forms may also need an engineered secondary confinement barrier due to the mobility of the tritium. This, like use of commercial solidification products, would need to be compared against the waste acceptance criteria (WAC) for the disposal facility.

5.4.3 Liquid Phase Options

If the water is below the drinking water (or permitted discharge) concentration limit, the water can be discharged without treatment. Monitoring of the discharge stream for tritium activity (and other hazardous and radiological) will likely be part of the facility operating license and permit.

If the water is at or above the drinking water/permitted discharge concentration limit, the water could be combined with other effluent discharge streams for disposition. The ability to combine the tritiated water with other discharge streams will depend on the concentration and volume of the tritiated water and the volume (flow rate) of the other effluent stream(s). Although not labeled explicitly as dilution, this would be the net effect. The volume of dilution water is easily calculated by performing a tritium mass balance of the two streams.

There are several liquid phase options for transfer of tritium from one volume of water to another water or solid stream. There is ample literature on proposed processes for treatment of the Fukushima water so these processes will only be discussed at a high level.

One promising SRNL technology being developed [Ref. 2] passes low activity tritiated water through a column packed with specialized material to isotopically exchange tritium from the water molecule to the column material, resulting in “clean” detritiated water as the effluent. The process does not change the volume of the water but creates a secondary waste solid stream which must be processed or managed. These technologies are under development and the magnitude of decontamination of the water stream is still being determined. Figure 5-1 shows tritiated water concentrations 100 times greater than the drinking water limit. It is believed this technology can achieve or exceed DFs of 100.

There are other detritiation technologies/processes which produce a lower volume, higher concentration tritium stream such as liquid phase catalytic exchange, vapor phase catalytic exchange, combined electrolysis catalytic exchange, etc. that are used to detritiate water. The draw-back of these processes besides the relatively large capital and operating costs is the fact they are not typically used to treat large volumes of low activity water, but rather smaller volumes of higher activity water. For example, some CANDU reactors remove the heavy water from the reactor when it reaches a threshold value, refill the reactor with reduced tritium content heavy water, ship the high activity water to a TRF for processing, and then return the heavy water for reuse in a reactor. The TRF process design does not create a water discharge stream anywhere near drinking water concentrations. The TRF process has DFs on the order of 100 for water approximately eight orders of magnitude greater higher in tritium concentration than the US drinking water limit as illustrated in Figure 4-1.

5.4.4 Liquid-to-Vapor Phase Options

There are several processes which involve changing the water from liquid to vapor phases. One simple process is to take a dehumidified gas (e.g. air) stream and pass it through a bubbler containing the tritiated water. The liquid water evaporates to vapor which is carried by the gas stream and can be discharged from the facility through a ventilation system. In Figure 5-1, one horizontal line is for saturated water vapor at 22 °C for the air concentration at the DAC-HTO. Discharges at ambient temperatures above 22 °C will ensure the water will not condense with additional flow in the ventilation flow, likely further reducing the likelihood of water condensation. Caution must be exercised if the water is heated or boiled to create water vapor due to concerns of condensing the water on surfaces cooler than the saturation temperature of the heated water vapor.

Electrolysis is another option for processing the water. Electrolysis electrolytically splits water into an HT and oxygen streams. Depending on the type of electrolyzer used, a drier system will need to be installed on one or both effluent streams so any tritiated water entrained in the stream can be recovered and electrolyzed. Electrolysis allows processing of water at concentrations higher than 1 mCi/L, but less than $1x10^4$ mCi/L, if discharges of an HT stream less than the DAC-HT is required. Dilution of the HT discharge stream for deflagration prevention purposes likely allows for higher activity water processing. Caution should be exercised when considering the use of electrolyzers since some technologies can concentrate the water tritium concentration up to 10X the feed water concentration. Another consideration for use of electrolyzers is the effect of higher tritium concentrations on many of the polymeric materials used in commercially available electrolyzers.

In additional to electrolytic reactions, there are other reaction mechanisms for treating tritiated water and releasing the tritium in the form of HT. Many times, heated (“hot”) metals are used to “crack” (remove the oxygen) from the water releasing the HT. Example metals include Fe, Cu, U, and Mg which form metal oxides and release HT. The metal/metal oxide bed is a secondary waste stream that will need disposition.

The effectiveness/efficiency of metal conversion to metal oxide varies greatly with bed design and process conditions so expectations of 100% utilization of the metal is likely to be unrealistic.

Gas phase reactions which transfer the oxygen atom from the water to another gas phase species includes the Water-Gas-Shift (WGS) reduction reaction:

$$CO + HTO \rightleftharpoons CO_2 + HT \tag{1}$$

The reaction will reach equilibrium based on processing conditions (temperature, pressure, feed composition). Removal of HT through a Pd-Ag tube ("Palladium Membrane Reactor – PMR") during the reaction can increase the cracking efficiency of the water since HT removal will shift the reactor towards the product side of the reaction. Another common reaction for removing the oxygen from the water to another gas phase species is the Methane-Steam-Reforming (MSR) reaction:

$$CH_4 + HTO \rightleftharpoons CO + HT + 2H_2 \tag{2}$$

which can be further reacted following Equation 1 with an additional equivalent of HTO to further produce molecular hydrogen isotopologues and reduce CO which may poison some catalytic systems. Similar comments can be made about reaction equilibrium, greater efficiency via use of a PMR, etc.

A final gas phase reaction to be considered would be an isotopic exchange reaction of the tritiated water with a material where the protium (H) of the material exchanges with the tritium in the water (e.g. Ref. 2):

$$X\text{-}H + HTO \rightleftharpoons X\text{-}T + H_2O \tag{3}$$

The efficiencies of some of these reactions are not very high and activity reduction of the water vapor may be considered more along the lines of "contamination" of the vessel rather than an efficient isotopic exchange process. This process produces a secondary waste stream which also must undergo disposal or treatment.

6.0 Tritiated Water Case Study

A Case Study example is presented to illustrate some of the options previously described in the document. Case Study example conditions were chosen as 1000 L of tritiated water at a concentration of 2 mCi/L. The total tritium content of the water is 2 Ci. Figure 6-1 is a plot of water concentration versus quantity with some of the same reference plots as shown in previous figures: the 1 Ci Total line included for refence. Case Study conditions are represented as a single circle in the figure with the different disposition strategies represented as movement from the case study initial conditions.

6.1 Dilution

The first water disposition option presented will be the dilution of the water to US drinking water limits. A tritium mass balance is

$$C_iV_i + C_cV_c = C_fV_f \tag{4}$$

where C_i and V_i are the initial concentration and volume of the tritiated water, respectively; C_c and V_c are the concentration and volume of the clean (dilutant) water, respectively; and C_f and V_f are the concentration and volume of the final, blended water stream. Rearranging and solving for the volume of clean water needed gives

$$V_c = V_i \frac{\left(\frac{C_i}{C_f}-1\right)}{\left(1-\frac{C_c}{C_f}\right)} \tag{5}$$

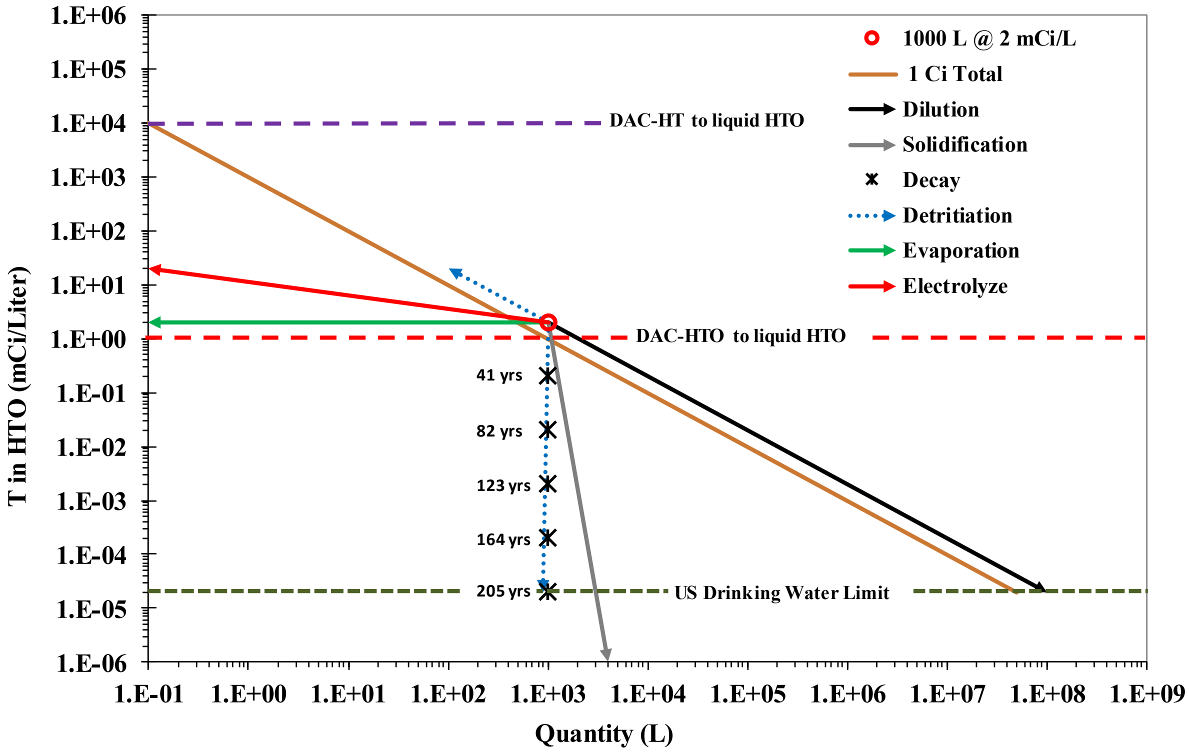

Figure 6-1: Tritiated Water Case Study

The reason for writing the equations in this form is to illustrate that the concentration of water used for diluting the tritiated water stream does not have to be "clean" (i.e. free of tritium) water, but could be two to three orders of magnitude lower than the final concentration of the stream – especially if the ratio of C_i divided by C_f is large, to get similar results. For this example, C_i divided by C_f is 1×10^5, so C_c being two orders of magnitude lower than C_f does little to alter the total volume of "clean" water needed for dilution. The total volume of the stream for discharge is

$$V_f = V_i \frac{\left(\frac{C_i}{C_f}-\frac{C_c}{C_f}\right)}{\left(1-\frac{C_c}{C_f}\right)} \approx V_i \left(\frac{C_i}{C_f}\right) \; for \; \frac{C_c}{C_f} \ll 1 \tag{6}$$

For use of "clean" water (C_c equals zero), the final water volume is 1×10^8 L. The dilution of this water is shown in Figure 6-1 as a negatively sloped line of constant tritium content starting at the Case Study conditions, ending at 1×10^8 L and at the concentration of the US Drinking Water Limit.

6.2 Solidification

The next water disposition option presented is for solidification of the water with a commercial solidification product. For 3 parts solidification product added to 1 part tritiated water, the final volume of the waste (neglecting any significant volume change/swelling of the final product) will be four times the initial volume of the tritiated water: 4,000 L or about twenty 55-gallon drums of waste. The illustrative line in Figure 6-1 is from the initial Case Study conditions to 4000 L at the lowest concentration, shown in the figure for graphical representation rather than terminating the line at a concentration of 25% of the starting concentration.

6.3 Decay Storage

An impractical water disposition option is decay storage of the tritiated water until it reaches the current US Drinking Water limit concentration. As mentioned earlier, it takes approximately 41 years for tritium to decay to 1/10th of its initial value. Figure 6-1 shows no volume change of the liquid as the tritium decays with the years of storage required to reach each decade of concentration reduction, indicated with plot symbols: 205 years total decay before the water reaches US Drinking Water concentrations.

6.4 Tritium Separation

If the water stream could somehow be separated into two liquid streams with one depleted in tritium and the other enriched in tritium, the volume of the enriched water for disposition/disposal would be smaller if the depleted stream could be discharged without further treatment. A mass balance on tritium produces

$$C_i V_i = C_d V_d + C_e V_e \tag{7}$$

where C_d and V_d are the concentration and volume of the tritium depleted stream, respectively; and C_e and V_e are the concentration and volume of the tritium enriched stream. V_d will be some fraction (f) of the V_i so the concentration of the enriched stream (C_e) can be written as

$$C_e = C_i \frac{\left(1 - \frac{f C_d}{C_i}\right)}{(1-f)} \approx \frac{C_i}{(1-f)} \ for \ \frac{C_d}{C_i} \ll 1, \ f = \frac{V_d}{V_i} \tag{8}$$

The f value can range from 0 (no enrichment) to near (but less than) 1. For the depleted water being at the US Drinking Water Limit concentration, the ratio of C_d to C_i is 1×10^{-5}. Arbitrarily selecting f as 0.90 (V_d being 90% of V_i and, V_e being 10% of V_i), the enriched volume is 100 L with a concentration of almost 20 (19.99982) mCi/L and V_d being 900 L.

Figure 6-1 shows the change of this split stream as two dashed lines emanating from the case study starting point. The first line for the "Detritiation" depleted stream terminates at 900 L and the US Drinking Water Limit concentration. The second line terminates at 100 L and approximately 20 mCi/L. Larger f values would have the termination point for the depleted stream line move from 900 L towards 1000 L. The termination point for the enriched stream line would extend further up and to the left in the figure as the enriched volume decreases and concentration increases. The enriched stream line is almost parallel to the 1 Ci Total line since most of the tritium resides in the enriched stream.

6.5 Evaporation/Bubbling

Another water disposition option illustrated in Figure 6-1 would be bubbling/saturation of a dry carrier gas as an emission from the facility. As the carrier gas is humidified by the water, the volume of water decreases. Any difference in vapor pressure of HTO relative to H_2O are ignored for this example. The line in the figure goes horizontally from the starting Case Study point to the left of the figure until the water has been fully evaporated.

6.6 Electrolysis

The water disposition option illustrated in Figure 6-1 would be the use of an electrolyzer to split the water into HT and oxygen. Recognizing that some electrolysis technologies produce an enrichment of heavier hydrogen isotopes in the water phase, the line is drawn from the Case Study conditions to a concentration 10 times the starting concentration and at the smallest volume shown in the figure. The electrolyze line in the figure is for illustrative purposes only and the actual concentration-versus-volume trace as the water is converted to HT and oxygen will be different than shown.

7.0 Implementation Strategies

7.1 Liquid Discharges

All liquids that leave the producer site as intended releases are required to be accounted for, even if at non detectable or at drinking water levels. Intended releases are accounted for through a permitting process that involves sample collection and analysis for activity and specialized software used to calculate radiation monitor setpoints and ensures release limits of 10 CFR 20 and 10 CFR 50 are not exceeded. During the release, flow rate, duration, and tank levels (as applicable) are recorded and then used to determined how much radioactive material (if any) was released to the environment.

In addition, for liquid radioactive releases, there are additional sampling and analysis requirements of the liquid radioactive effluent stream. The producer will also need to obtain a National Pollution Discharge Elimination System (NPDES) permit through the state and comply with additional laboratory certification and sampling requirements of the NPDES program. Furthermore, the producer may choose to utilize water treatment methods in cooling water systems to protect against corrosion, fouling, and other damaging conditions. If the producer utilizes water treatment methods, such as chemical additives, in cooling water systems that they plan to release off site, additional permitting, treatment, sample, and analysis may be required by local, state, and federal laws prior to release. Additional system sampling and analysis will also be required to monitor water treatment methods.

7.1.1 Low Level Water Outfall Discharge with Freshwater Replenishment

For liquid releases to an outfall, the producer will need to identify a suitable release outfall, a source of dilution flow water, and add a liquid effluent process radiation monitor with flow isolating capabilities to isolate flow if the monitor setpoint is exceeded during the release. A site location near a river, a commonly used practice for siting industrial, chemical, and nuclear power facilities, allows these types of discharges which may not be an option for facilities sited miles away from suitable outfall.

7.1.2 Low Level Water Public Discharge with Freshwater Replenishment

Any low level radioactive active waste release to public waters/storm sewers will have to meet the requirements of 10 CFR 20, Radiation Protection/Radioactive Releases/Radioactive Shipments, 10 CFR 50, Domestic Licensing of Production and Utilization Facilities. Additionally, 10 CFR 20 requires adherence to 40 CFR 190, Environmental Radiation Protection Standards for Nuclear Power Operations for liquid radioactive releases.

A potential method of release could be utilizing a specified release drain to municipal water treatment facility with deep well water for dilution flow following utilization of an ion-exchange system for the removal of activation products to ensure the releases are As Low As Reasonably Achievable (ALARA) and a separate storage tank for the liquid effluent to be stored prior to release. The city/county having jurisdiction and the NRC will have to approve this methodology of utilizing a drain for intended, monitored releases.

For intended releases, this scenario is not recommended for the producer due to additional equipment, treatment methods, sample, and analysis requirements needed to meet the standards contained in 10 CFR 20, 10 CFR 50, and 40 CFR 190. This method, if chosen, can be further analyzed for unplanned releases or accident scenarios to determine the level of activity released to the environment and the public.

7.2 Water Treatment

7.2.1 Water Recycle

A separate storage tank(s) could be utilized to periodically receive waters from the closed-cooling loop and cooling pool. These waters could then be passed through an ion-exchanger system to remove potential activation products and potential incorporation of a detritiation system for tritium removal. Once processed, the water could be stored in a tank and used for make-up water to the cooling loops.

7.2.2 Water Reduction

Producer facilities will likely have an Aqueous Radioactive Liquid Waste Storage system (RWLS) that provides receipt, mixing, and buffer storage for aqueous radioactive wastes generated by Mo-99 processing operations within the Radiation Controlled Area. This RWLS could possibly be utilized and/or expanded to also periodically receive contaminated water from the cooling water loops to reduce tritium concentrations in the loops after refilling with water. The contaminated water could then be sent to a radioactive waste evaporation and immobilization system where the waste is concentrated by evaporation and then immobilized for disposal. This is like the previously described flowing stream humidification (evaporation) process for disposition of the water but takes advantage of an existing radiological system. Modifications may include increasing tank volumes and creating tie-in points for addition of the low level tritiated water. Residual tritium in the concentrated waste is likely to be a lower hazard constituent of the waste than the other radionuclides.

8.0 Disposal Pathways

Ref. 3 “Pathways for Disposal of Commercially-Generated Tritiated Waste”, discusses available disposal options for tritiated liquid and solid wastes. Disposition of gaseous waste streams are assumed to be processed through a stripper system which may create a solid or liquid waste stream or can be directly vented as a discharge from the producer’s facility as discussed in this document. For tritiated liquid and solid waste, there are several disposal facilities across the continental United States that receive low level radioactive wastes (LLW) from commercial producers. In additional to the disposal facilities, there are also commercial processing and recycling facilities that have the ability to process and treat radioactive waste streams, unable to be directly accepted by the disposal facilities, into forms acceptable by the disposal facility. The producer will need to work directly with a disposal and/or processing facility on their specific disposal needs.

9.0 Conclusion

In applications where tritium is handled, the interconnected water systems utilizing or exposed to tritium will accumulate tritium and potentially create large volumes of tritiated water without convenient disposition pathways. This document has highlighted several liquid tritium waste disposition options that a Mo-99 procedure could utilize. It is the intent of this report not to recommend a disposition strategy or procedure, but to discuss the methodologies of each strategy so that a procedure could utilize the disposition strategy that best fits their individual needs.

10.0 References

(1) S. K. Sood et al., “A Compact, Low Cost, Tritium Removal Plant for CANDU 6 Reactors,” Proc. Annual Conf. of Canadian Nuclear Society, Toronto, Canada, June 8–11, 1997, p. 11, Canadian Nuclear Association (**1997**)
(2) Xiao, X., *Decontamination of Tritiated Water*, WO 2015/072981 A1, (**2015**).
(3) Halverson, N., *Pathways for Disposal of Commercially-Generated Tritiated Waste*, SRNL-STI-2016-00471, (**2016**).