TRITIUM EMISSION REDUCTION INITIATIVES AT CANDU REACTORS

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Abstract

CANDU[1] reactors produce significantly more tritium than any other type of reactor. Therefore, control of tritium releases to the environment is particularly important for these reactors. Ontario Power Generation Nuclear (OPGN) has implemented detritiation technology to reduce tritium in their CANDU reactors. This has resulted in reduced environmental tritium emissions, and dose to workers and the public from reactor operations. However, application of this technology results in the release of both elemental tritium and tritiated water vapor during operation. This paper reviews the Darlington Tritium Removal Facility (DTRF) operation and its contribution to tritium emissions reduction at OPGN generating stations.

1. Introduction

CANDU reactors use heavy water in their moderator and primary heat transport (PHT) systems. Tritium is produced in heavy water reactors through neutron capture by deuterium atoms in the heavy water. The annual production of tritium in a CANDU reactor is typically 1.4-2.0 kCi[2]/MW(e) and most of this tritium (~97%) is produced in the moderator [1]. This is because ~90 percent of the moderator water in the calandria is exposed to a large thermal neutron flux, whereas ~5% of the total PHT system water is subject to high flux. If the tritium produced is not removed through detritiation or by swapping with low-activity heavy water, the tritium activities in both the moderator and PHT systems will increase with time until they reach equilibrium values. At equilibrium, the PHT and moderator systems for OPGN reactors are expected to reach tritium activities of 2-3 Ci/kg and 60-100 Ci/kg, respectively. During normal operations and maintenance activities, tritiated heavy water can escape from the heavy water systems. While most of the heavy water is recovered and recycled, a small fraction is lost to the environment contributing to both airborne and waterborne tritium emissions. Tritium releases to the environment are strictly controlled and are maintained as low as reasonably achievable (ALARA). OPGN station emissions generally represent only a small fraction of the regulatory limits (DRL[3]).

Tritium releases from CANDU stations are a complex function of operational activities, maintenance activities, operating and maintenance practices, age of systems and components, tritium activity of heavy water systems etc. The PHT system has been shown to contribute

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[1] CANada Deuterium Uranium, Registered Trademark
[2] 1 Ci = 37 GBq
[3] Authorized releases to the environment, in Canada, are governed by Derived Release Limits (DRLs). DRLs represent an estimate of a release of a radionuclide that could result in a dose of 1 mSv to an exposed member of the public. The DRL values are specific for each facility and take into account various regional factors such as meteorological and geographical conditions and the population density.

the most to heavy water losses from CANDU reactors while the moderator has shown to contribute the most to tritium emissions due to its high tritium activity [2], [3], [4].

CANDU reactors produce significantly more tritium than any other type of reactor. Consequently, the control of tritium releases to the environment is of significant importance to ensure that public dose is ALARA, and to maintain public confidence in CANDU technology.

OPGN operates a number of nuclear generating stations (NGSs) based on CANDU, pressurized heavy water, reactors. These stations include Darlington NGS (DNGS), Pickering NGS (PNGS (A) and (B))[4]. Bruce NGS (BNGS (A) and (B))[5] was also operated by OPGN, until it was leased to Bruce Power in 2001. DNGS, PNGS and BNGS have four 905 MWe, eight 540 MWe and eight 840 MWe reactors respectively. OPGN opted to remove tritium from their heavy water systems to reduce occupational exposure and environmental emissions. OPGN constructed and commissioned the DTRF at DNGS in 1989. In addition to providing tritium-removal services to OPG reactors, DTRF also provides tritium removal services to other CANDU reactors in Canada. Although DTRF reduces tritium in OPG reactors to reduce emissions, it has also become an important source of tritiated water (DTO) vapor and elemental tritium (DT) emissions at DNGS. This paper reviews the DTRF contribution to tritium emissions reduction at OPG nuclear generating stations in Ontario. Proposals to improve DRTF operations to further reduce tritium emissions are also discussed.

2. Effect of moderator tritium activity on environmental tritium emissions

Since 1989, the DTRF has been operating to reduce and control the tritium activities in the moderator and the PHT systems of OPGN reactors. Significant reductions in tritium activities in OPGN reactors have been achieved since 1989. In the last decade (2000-2010)[6], the average moderator tritium activities in DNGS and PNGS (B) reactors were in the range 6.7-10.4 Ci/kg, and 12.5-18.9 Ci/kg respectively. In the same period, the average PHT activities were in the range 0.6-0.8 Ci/kg and 1.0-1.2 Ci/kg respectively. During this period, the moderator tritium activities for all four reactors at DNGS were in a very narrow range of 5.6-10.7 Ci/kg while for PNGS(B) four reactors they were in the range 10.4-21.0 Ci/kg.

Figure 1 shows the effect of moderator tritium activity on airborne DTO emissions for PNGS (B). As Figure 1 shows, a strong correlation exists between the moderator activity and the airborne DTO emissions for PNGS(B). Correlations also exist between the moderator activity and the total DTO emissions for PNGS (B). Similar correlations were also found with other OPGN reactors.

Figure 2 shows the average annual airborne tritium emissions from all OPG stations (DNGS, PNGS and BNGS) as a function of the average moderator tritium activity. The data include the BNGS data up to 2001. Figure 2 does not include data for PNGS (A) Units 1-4 and

[4] PNGS Units 1-4 were laid up in 1997. PNGS Units 1 and 4 came on line in 2005 and 2003 respectively. PNGS Units 2 and 3 are scheduled for decommissioning.
[5] OPG operated BNGS Units 1-8 until 2001. BNGS Units 1-4 were laid up from 1998 to 2000. BNGS was leased to Bruce Power in 2001.
[6] Since not all reactors were in operation at PNGS (B) during 2000-2010 and BNGS was leased to Bruce Power in 2001, the data from these two sites during this period are not considered in the analysis.

BNGS (A) Units 1-4 after 1997. As the data show, a strong correlation between moderator activity and airborne DTO emissions exist for all OPG reactors. Similar correlations also exist for the average total DTO emissions.

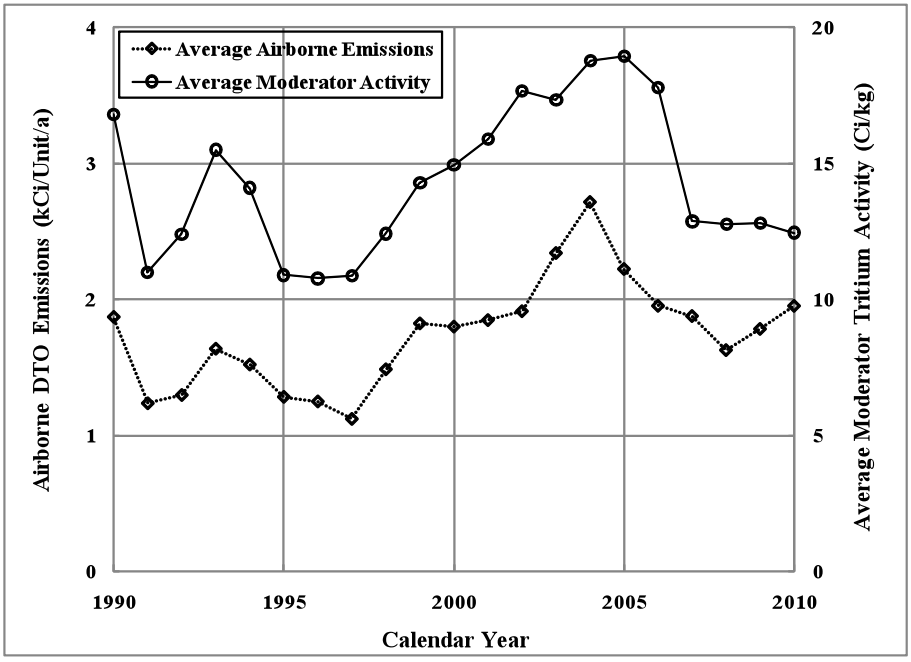

Figure 1 Effect of moderator tritium activity on PNGS (B) DTO emissions

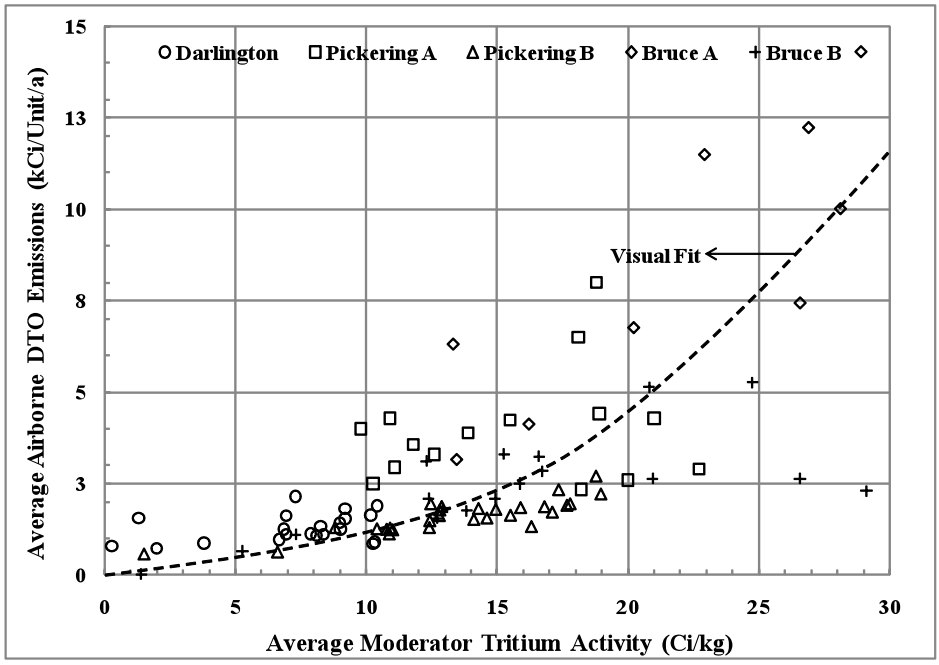

Figure 2 Total average DTO emissions versus moderator tritium activity

During 2000-2010, the average airborne DTO emissions for DNGS and PNGS (B) were 1.23±0.33 kCi/Unit/a and 2.00±0.31 kCi/Unit/a respectively. During the same period, the average waterborne DTO emissions were 1.24±0.70 kCi/Unit/a and 1.26±0.23 kCi/Unit/a respectively. The DNGS reactors had the lowest moderator and PHT tritium activities and the lowest airborne DTO emissions (0.86-1.89 kCi/Unit/a) among the OPGN reactors, in spite of the fact that DNGS HTO emissions also include DTO emissions from the DTRF.

The tritium emission data reported above show that the moderator is a dominant contributor to tritium emissions. The data also show that DTRF has contributed significantly in reducing and maintaining tritium activities in heavy water systems at OPGN stations and tritium emissions low.

3. A detritiation strategy to reduce OPGN tritium emissions

Although the DTRF heavy water processing capacity is used to reduce the tritium activities in heavy water systems, allocation of processing capacity between OPGN stations is not optimized for reducing emissions [5]. Based on an analysis of historical emission data from OPGN stations and heavy water processed from these stations at DTRF, it was concluded that further reductions in tritium emissions may be achieved at OPGN stations, by allocating DTRF capacity to each station based on its historical contribution to DTO emissions [5]. Since moderator activity strongly correlates with the airborne DTO emissions at OPGN stations, it was suggested that DTRF capacity be allocated based on the airborne DTO emission contributions (i.e., allocate more DTRF capacity to the station contributing more to airborne DTO emissions and vice versa).

The airborne DTO emission contributions from DNGS and PNGS (A&B) as a percentage of the total airborne emissions from the two sites are shown in Figure 3 during 2000-2010. As Figure 3 shows, the percentage DNGS contribution to the total airborne DTO emissions from the two sites is significantly smaller compared to the PNGS contribution. In the same period, the DNGS contribution decreased slowly over time while the PNGS contribution increased slowly. During 2000-2010, the average percentage contributions from the DNGS and PNGS to the total airborne DTO emissions from the two sites were ~24% and ~76%, respectively. According to the detritiation strategy discussed, further reductions in the overall tritium emissions at OPGN stations may be achieved by allocating the available DTRF detritiation capacity to OPGN stations based on the percentage contributions to the total airborne DTO emissions given above.

4. Tritium emissions from DTRF

In the last decade (2000-2010), the DTRF, average airborne DTO and DT emissions were 0.14±0.08% and 0.03±0.05% of the DNGS site DRLs [6], respectively. In 2004, the airborne DTO emissions and in 2004 and 2005 the DT emissions from DTRF were significantly higher than the normal. The average airborne DTO and DT emissions without these data are 0.12±0.05% and 0.01±0.01% of the DNGS site DRLs, respectively. During this period, the DTRF contribution to the airborne DTO emissions from DNGS has been significant, ~36.9±9.5%. However, the DTRF contribution decreased significantly from 47.6% in 2008 to 24.1% in 2010. Although the DNGS airborne DTO emissions during the same period were well below 1% of the DRL, the significant DTRF contribution to DNGS emissions prompted

a study aimed at identifying DTRF emission sources and solutions for further emissions reduction.

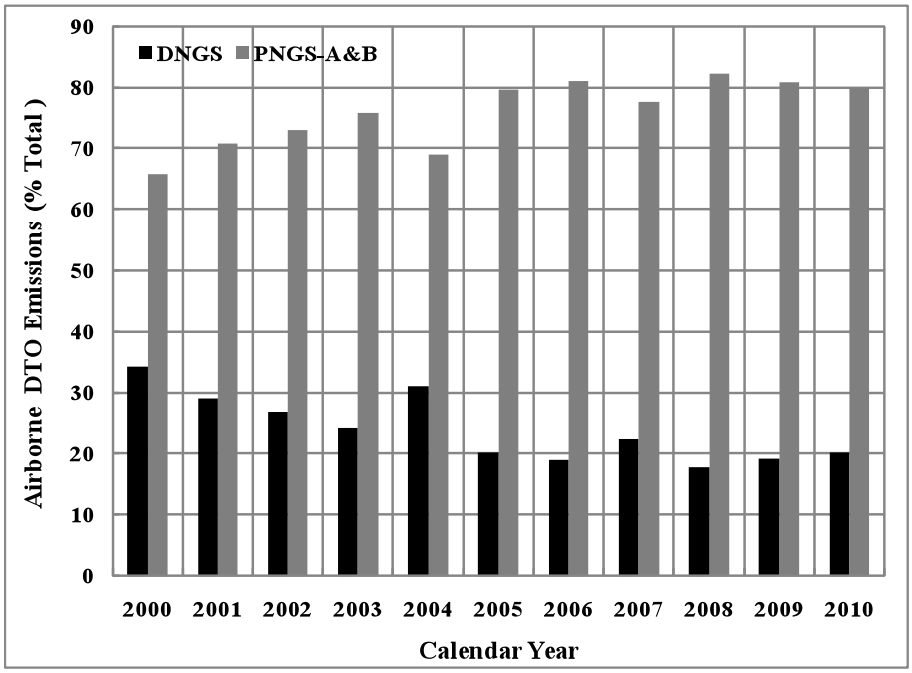

Figure 3 Station airborne DTO emissions contribution to the total OPGN sites

4.1 DTRF – A brief description

DTRF is located in the Heavy Water Management Building (HWMB) at DNGS. The HWMB houses facilities to cleanup, upgrade and store heavy water for all four reactor units at DNGS. The facility provides a means of transferring moderator water between reactor units, the DTRF and the heavy water upgrader (HWU). The HWMB also handles shipments of tritiated heavy water in drums or Type B transportation packages, as part of tritium removal services provided to other customers.

The DTRF houses the heavy water detritiation process and associated systems [7]. A schematic diagram of the DTRF process is shown in Figure 4.

The heavy water detritiation process essentially consists of two stages. In the first stage, tritium in the heavy water is transferred to a deuterium (D_2) carrier gas in the form of DT gas, while detritiating the heavy water to the desired level using the vapor phase catalytic exchange (VPCE) process. In the second stage, pure elemental tritium (T_2) is separated from the D_2 carrier gas using cryogenic distillation (CD) and the detritiated D_2 gas is recycled back to the first stage. A schematic of the TRF process, showing the key systems, is shown in Figure 4 [7]. A detailed description of the DTRF process is beyond the scope of this paper.

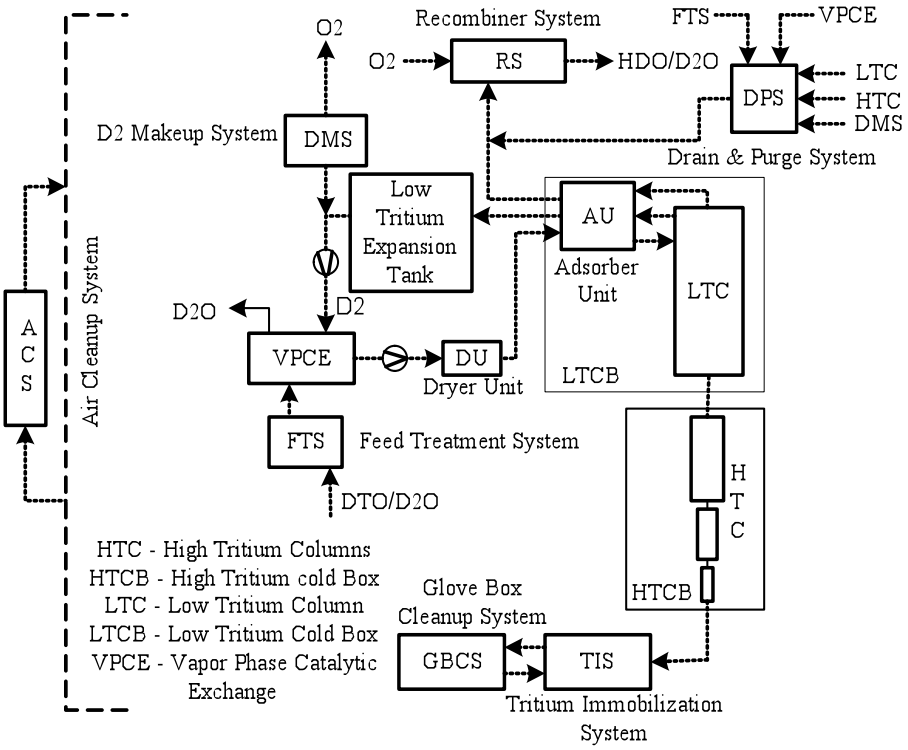

Figure 4 DTRF Simplified Flow Diagram

4.1.1 DTRF emission sources and pathways

The daily tritium emission data from the main stack and detailed event logs from 2005 to 2009 were reviewed in an attempt to identify emission sources. The emissions were found to be largely due to acute events. A study led by DTRF staff, has identified several administrative measures that, when implemented, resulted in significant reductions in DTRF DTO emissions. These measures included improvements to operating procedures, prompt investigation of daily emissions >5 Ci/day and continuous focus on reducing human error aimed at reducing acute emission events. The recent reduction in the DTRF contribution to DNGS emissions (from 48% in 2008 to 24% in 2010) has been attributed to these measures.

Sources contributing to tritium emissions, that were identified during this review, are: Flame Recombiner System (RS) operations, Feed Treatment System (FTS) venting, Tritiated-Heavy-Water-Transportation-Package (THWTP) unloading, Heavy-Water-Storage-Tanks (HWSTs) filling and emptying operations, and Tritium Immobilization System (TIS) operational activities. Figure 4 shows the schematic of the DTRF contaminated ventilation system showing the emission pathways from these systems.

RS operation: The RS is a flame recombiner which is used to safely burn hydrogen isotope gas mixtures during various DTRF operating and maintenance activities. The hot combustion gases are cooled on heat exchanger coils in the burner vessel and a chilled-water-cooled condenser. The condensate is collected in a tank. The unconverted elemental tritium (DT) and non-condensed DTO along with non-condensable gases (O_2, N_2, He) are vented to the process vent (PV) that exhausts to the TRF stack (Figure 5). The amount of DTO in the non-

condensable exhaust gases is dependent on the dew point, and the flow rate of the non-condensable gases. The chilled water temperature and the condenser efficiency limit the minimum humidity and DTO emissions that can be achieved at the condenser outlet.

The head space of the condensate collection tank is vented to the HWMB stack through a chilled-water cooled heat exchanger (Figure 5), HX1, and can also contribute to DTO emissions through HWMB depending on HX1 efficiency.

The gas phase in the condenser is isolated from the condensate tank with a heavy-water-loop seal. Operation of the RS without a loop seal can lead to residual DT and DTO bypassing the condenser. Depending on the efficiency of HX1, loop seal failure can lead to increased DTO emissions.

FTS cover gas purges: The Drain and Purge System (DPS) provides a mechanism to allow the transfer of process gas or purge gas between systems for maintenance. Several DTRF systems utilize DPS, including FTS (Figure 1). There is seat leakage of some of the DPS drain valves to FTS. Consequently, DT has been found in the FTS cover gas system. FTS cover gas is purged to DPS when the hydrogen isotopes concentration reaches 4%. This purge gas is eventually burned in the RS. DTRF has experienced higher than normal DTO levels in the exhaust when FTS cover gas is purged as a result of tritium ingress from passing DPS valves. Emissions through this pathway can be reduced by replacing the passing drain valves.

FTS cover gas exhausts to the PV, through a chilled-water cooled cold trap, via a pressure regulating valve (PRV) set to open at 110 kPa(g). The PRV is activated during daily filling of the DTRF FTS feed tank. Both DT and DTO are released to the PV through this pathway.

THWTP unloading: Heavy water in THWTPs is unloaded to a storage tank with air pressure. During this process, air saturated with DTO is released to the HWMB stack via HX1. The extent of the emissions depends on the HX1 efficiency and the volume of vented air.

HWSTs filling and emptying process: Emissions from the HWSTs result from venting of the cover gas to the stack during heavy water filling operations. The HWSTs in the HWMB are maintained under air cover gas. The air pressure in the HWSTs is maintained at 13.8 kPa(g). The cover gas in these tanks is vented to the stack through a chilled-water cooled heat exchanger HX2 to the HWMB stack before filling and then re-pressurized to 13.8 kPa(g) after filling. The cover gas purges can result in DTO emissions through the HWMB stack.

Emissions from TIS: Emissions from the TIS were found to result from several sources. During normal operations, for every batch (~45 kCi) of pure elemental tritium (T_2) removed from the CD, a sample is injected in to a mass spectrometer for tritium purity analysis. The sample gas is exhausted through the mass spectrometer pumping system directly to the TRF stack.

The immobilized tritium storage container (ITC) is located inside a separate airlock (argon atmosphere) attached to the TIS glove box. During ITC changes, the argon gas in the airlock is purged to the TRF stack and can potentially contribute to emissions if tritium is present in the airlock.

Elemental tritium is removed from the CD system to the TIS through a transfer line protected by a rupture disc-relief valve combination. In the event of a rupture disc failure tritium will be released to the relief valve discharge line and into the TRF stack. An overpressure event can result in significant T_2 and DTO (from oxidation of the elemental tritium on the moist stainless steel piping surfaces) emissions.

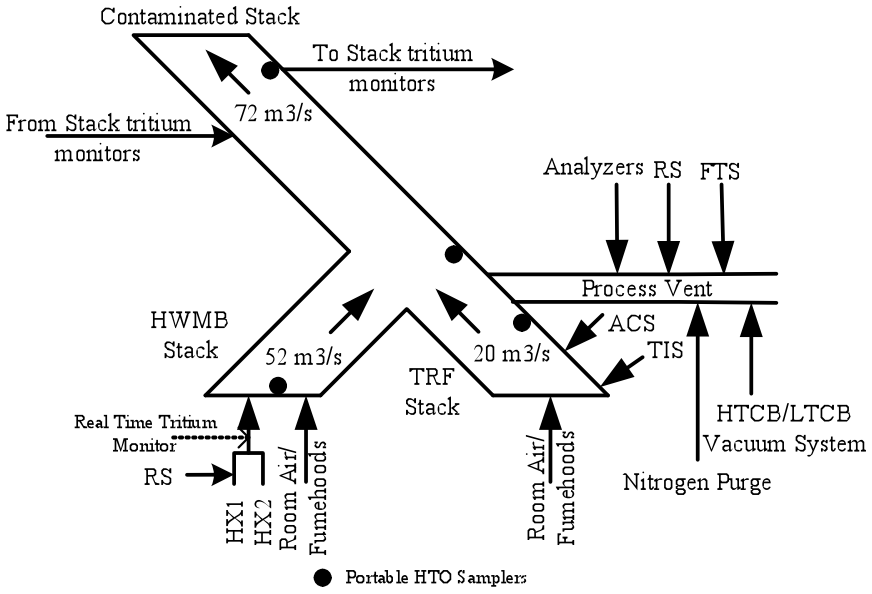

Figure 5 TRF contaminated ventilation system schematic

4.1.2 Contribution from HWMB, TRF stacks and PV to emissions

All emissions are routed to the main stack via the HWMB and TRF stacks. To estimate the contributions from HWMB, TRF and PV to the main stack emissions, DTO emissions were monitored for a long period at several locations in the DTRF ventilation system (see Figure 5). The measured DTO-emission data indicate that majority of the DTO emissions from the main stack come from the HWMB stack (~74%). The rest (~26%) come from the TRF stack. The TRF stack emissions come mainly from the PV (22%). The remainder comes from the combined exhausts from room air, fume hoods in TRF and TIS. Consequently, DTO-emission-reduction options that target emissions from the HWMB stack should contribute the most to DTO emission reduction.

4.1.3 Emission reduction options

Tritium (DT and DTO) emissions from the DTRF can be significantly reduced by treating the combined exhaust from HX1 and HX2 and the PV.

Emission reductions up to ~74% in DTO may be achieved by treating the exhaust gases from HX1 and HX2 to the HWMB stack. Except for room air and fume hood exhausts, all discharges from process and tank vents to the HWMB stack are through HX1 and HX2. Operating experience has indicated low HX1 and HX2 efficiencies. Emissions may be reduced by replacing HX1 and HX2 with high-efficiency heat exchangers. However, much higher reductions in DTO emissions may be achieved using dryers to treat the combined exhaust from HX1 and HX2. Reductions in DTO emissions from HX1 and HX2 in excess of 99% may be achieved using dryers.

Emissions from the TRF stack come mainly from PV and TIS. The key contributors to emissions from the PV are RS operations and FTS. Both DT and DTO contribute to emissions from the PV. The emissions from the two key contributors to the PV (RS and FTS) can be reduced using high-efficiency vent condensers. However, the minimum dew point of the vent gas and the emission levels that can be achieved using a condenser are limited by the chilled-water temperature and the condenser efficiency. To further reduce emissions from the PV, the other option is to treat the PV exhaust gas using molecular sieve dryers before the TRF stack. Since the PV exhaust can contain both DT and DTO, a catalytic recombiner may need to be incorporated into the design, upstream of the dryers, to convert DT to DTO.

The TIS exhaust can contain both DT and DTO. The TIS exhaust is vented directly to the TRF stack without pretreatment to reduce tritium emissions. Currently efforts are underway to treat the TIS exhaust using the ACS.

5. Conclusions

Since its commissioning in 1989, the DTRF has been operating to reduce and control the tritium activities in the moderator and the PHT systems of OPGN reactors. As a consequence, OPG reactors were able maintain low tritium emissions since 1989. Although the DTRF has reduced the tritium activities in the moderator and PHT systems in OPGN reactors, the allocation of DTRF detritiation capacity among OPGN stations is not considered optimized for reducing tritium emissions. A detritiation strategy based on optimal allocation of DTRF detriation capacity among OPGN stations may be used to further reduce the total tritium emissions from OPGN stations.

Since its beginning, airborne DTO emissions from the DTRF have constituted a significant fraction of the total DNGS airborne DTO emissions. According to the DTRF daily emissions data, under normal operating conditions, emissions from the DTRF mostly come from routine, operations–related, activities. Some of these emissions have been identified with human error and procedural weaknesses and these have been already addressed through administrative measures. Other emissions are related to operational activities that include RS operations, FTS venting, and TIS operations. The key emission pathways from these sources are the PV, TIS vents to the TRF stack. There is also a significant source of DTO release from the HWMB stack via HX1 and HX2, during THWTP unloading and HWSTs filling. High-efficiency vent condensers and/or dryers can be used to further reduce emissions from the DTRF and HWMB systems. The proposed design changes to the DTRF and HWMB system vents can be used to reduce overall tritium emissions from DNGS. However, the proposed changes require detailed design and review to determine cost-effectiveness and to ensure that

the changes would not result in operational challenges. This cost benefit review needs to also consider the need to invest in the plant to increase equipment reliability, in order to further enhance tritium removal from the reactors.

6. Acronyms

ACS	Atmosphere Cleanup System
DMS	D_2 Makeup System
DPS	Drain and Purge System
DRL	Derived Release Limit
DTO	Tritiated Water
DTRF	Darlington Tritium Removal Facility
DU	Dryer Unit
FTS	Feed Treatment System
GBCS	Glove Box Cleanup System
HT	Elemental tritium
HTC	High Tritium Column
HTCB	High Tritium Cold Box
HTDC	High Tritium Distillation Column
HWMB	Heavy Water Management Building
LTC	Low Tritium Column
LTCB	Low Tritium Cold Box
LTDC	Low Tritium Distillation Column
OPGN	Ontario Power Generation Nuclear
PHTS	Primary Heat Transport System
PV	Process Vent
RS	Recombiner System
SCR	Station Condition Report
TIS	Tritium Immobilization System
VPCE	Vapour Phase Catalytic Exchange

7. References

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