

Toxicity Limitation on Radioactive Liquid Waste Discharge at OPG Nuclear Stations

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Abstract

The Municipal and Industrial Strategy for Abatement (MISA) regulation, which came into effect in 1995 in Ontario (Ontario Regulation 215/95 under the Environmental Protection Act), imposed additional limitations on liquid discharges from power generating stations. The MISA regulation has divided discharges into non-event and event streams, which have to be monitored for the prescribed parameters and for toxicity. Radioactive Waste Management Systems fall into the category of non-event streams. Standard toxicity testing involves monitoring lethality of *Daphnia Magna* and Rainbow Trout in the effluent. The new legislation has imposed a need to address several issues: acute toxicity, complying with the specific limits prescribed by the regulation and, in the long run chronic toxicity.

In the first phase, the correlation between various chemical parameters and acute toxicity was established and several investigations were initiated. The effects of microbial activities in the waste streams were the most difficult to address because many synergies between chemical toxicants and microbial toxins were not known. Due to a limited time schedule to achieve MISA compliance, a wide approach was taken. Approaches included best management practices in the power plants, waste stream segregation and a choice of treatment technology that could simultaneously address a number of potential toxins. Furthermore, the variability of the waste streams in present and in future had to be taken into account. This approach had a drawback in potentially generating significant costs of consumables and volumes of secondary solid radioactive waste. A strategy to monitor treatment technology performance and to optimize processes and costs in the long run, was therefore devised. Control of waste generation and centralizing laundry facilities from Pickering and Darlington to the Bruce site have enabled achievement of MISA compliance with simple processes and modifications at Pickering and Darlington stations (laundry is a key source of organic toxins). In parallel, more elaborate technologies were applied at Bruce A site for treatment of additional laundry wastes.

Originally, Active Liquid Waste Treatment (ALW) systems were designed to remove radioactivity from the liquid waste streams. Achieving radioactivity Derived Emission Limits (DEL's) and Station Target Limits was never a problem in Ontario Power Generation stations and the ALW treatment systems were rarely required for this purpose. Modifications of these existing systems and installation of the new treatment systems, were a part of the strategy developed at OPG.

At Pickering a completely new treatment system was designed and installed. The process consists of an ultraviolet biocide unit, cartridge filtration, Granulated Activated Carbon (GAC), cation resin and neutralizing filter (CaCO_3) bed. This combination of processes

enabled the use of adsorption for reduction of organics and cation IX resin for removal of dissolved cations. Filter optimization will have to be considered for long term operation.

The Darlington ALW system was modified so that a redundant IX column was used for GAC (later switched for Macronet resin). For long term operation, filtration, cation resin and a CaCO_3 bed are being considered.

The Bruce A ALW system is a complex, multi-stage treatment system consisting of pretreatment, a multi-stage reverse osmosis system (ROS), and an evaporator solidification system (ESS). The concentrates from the ROS are fed to the ESS and evaporated via the use of a thin film evaporative process. The evaporator bottoms are solidified in bitumen and the solidified residue placed in a container for storage on-site as a low-level radioactive waste. In the final polishing stage of the process, the permeate from the ROS is treated via a calcium carbonate bed for pH adjustment and this water discharged to the lake.

The overall effort to achieve MISA compliance at OPG Nuclear sites was extremely challenging, in particular because of the very tight schedules. Lessons have been learned in the areas of procurement and design. Internal management processes have been established to ensure continued MISA compliance.

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Introduction to MISA Compliance Requirement

In 1990 the Ontario Ministry of Environment issued the Development Document for the Effluent Monitoring Regulation for the Electric Power Generation Sector under the MISA strategy (MISA = Municipal/Industrial Strategy for Abatement; Reference 1). The MISA strategy intent was to stop water pollution at its source. At that time, it became clear that toxic discharges from power generating stations would have to be eliminated. Bruce A NGS had an early start in search of processes and technology to eliminate pollutants from Active Liquid Waste effluent. The Bruce A active liquid waste treatment system project started in 1993 with no definite idea of what would have to be eliminated from the waste streams to achieve MISA compliance. The actual legislative act was issued in April 1995 as Ontario regulation 215/95 under the Environmental Protection Act and with the title: "Effluent Monitoring and Effluent Limits - Electric Power Generation Sector".

Ontario Regulation 215/95

The Ontario Regulation 215/95 lays out the definitions and conditions of effluent monitoring and effluent limits for all specified power generation stations listed in Schedule 1 of the regulation (Reference 2). The purpose of the legislation is to monitor and control the quality of effluent discharged from the plants listed in Schedule 1 of the regulation. It is important to say that this regulation does not apply with respect to the discharge of effluent to a municipal sanitary sewer.

The discharges from the power generating stations have been divided into event process streams, non-event process streams and building effluents. The non-event process effluent streams seem to be related more to non-radioactive systems and non-nuclear plants.

The event process effluents are defined in the regulation under the list of examples of various waste streams. Radioactive Liquid Waste Management System (RLWMTS) is typically under the event process effluent streams.

Regulation 215/95 furthermore defines the way of establishing sampling points and sampling frequency for the event and non-event process effluent streams.

Specific limits on effluent parameters are given in the Schedule 2 of the Regulation 215/95. The list of these limits on effluent parameters for active liquid waste discharges from the Ontario Power Generation nuclear stations is given in Table 1. Although the limiting parameters are listed specifically for each station, they are the same for all Radioactive Liquid Waste Management System (RLWMSTE) discharges. In general, there have not been many problems to comply with the limits specified in Table 1.

The sampling points establishment and analysis methods are described in the "Protocol for the Sampling and Analysis of Industrial/Municipal Wastewater" issued by Ministry of Environment and Energy Publication (Reference 3).

The requirement that had the largest impact on radioactive liquid waste management systems discharges was the requirement that all the effluents be non-toxic at all times. The non-toxic discharge was defined as lethality of less than 50 % to both *Daphnia magna* and rainbow trout.

The protocol for the acute toxicity testing requires a lengthy exposure of *Daphnia magna* (48 hours) and rainbow trout (96 hours) to the effluent sample. Toxicity testing was therefore not a practical means of controlling effluents in the operating station environment given that there was not enough spare capacity in RLWMS tanks to hold wastewater until toxicity testing is performed.

The first problem that was encountered was to find the causes of toxicity in RLWMTS effluents and to try to translate them to the measurable chemical parameters that would provide an indication of toxicity in reasonably shorter time than toxicity testing. Knowing the chemical parameters that cause toxicity would also enable us to select the most adequate treatment and apply it selectively, depending on the characteristics of each waste batch or stream. It was also important to treat waste batches selectively in order to minimize unnecessary generation of the secondary solid radioactive waste (i.e. filter cartridges, IX resin etc.).

Several studies were performed by OPG in conjunction with Ontario Power Technologies (OPT) to gain the necessary information about causes of toxicity in RLMTS effluents and about treatability of waste streams.

Causes of Toxicity in RLWMTS Effluents and Derived Toxicity Limits

A first toxicity study was conducted in 1995. A later and more extensive study on the causes of toxicity was issued in September 1998 (Reference 4). This later study was utilized more because it was based on more recent and more detailed sampling and analysis results. The results of the 1998 study have pointed out that there are several suspected causes of toxicity in RLMTS streams specific to each OPG nuclear station.

Table 1; Chemical Parameters in Radioactive Liquid Waste Management System Effluents for all Nuclear Stations Limited by Regulation 215/95

Parameter	Monitoring Frequency	Daily Concentration Limit (mg/L)	Monthly Average Concentration limit (mg/L)
Total Phosphorus	Weekly	-	1.0
Total Suspended Solids (TSS)	Daily	73.0	21.0
Zinc	Weekly	1.0	0.5
Iron	Weekly	9.0	3.0
Oil and Grease	Weekly	36.0	13.0

Darlington. From the 60 samples analyzed from Darlington RLWMTS, 38% were non toxic (neither *Daphnia magna* nor rainbow trout. The rest, 62 % of samples were toxic either to *Daphnia Magna* or rainbow trout or to both. The study has shown that there was a clear relationship between the concentration of the total organic carbon (TOC) and toxicity. At TOC concentrations below 10 mg/L, effluent was consistently non-toxic while at concentrations above 20 mg/L effluents were consistently toxic. Adjustment to the lake hardness did not effectively reduce toxicity of Darlington RLWMTS effluents.

Pickering A and B. From the 104 samples of RLWMTS effluents tested from Pickering, 55% passed both acute toxicity tests. The rest, 45% of samples have failed either the *Daphnia magna* or the rainbow trout test or both tests. Detailed analysis of results by tanks of origin has indicated that TOC and Cu were significant contributors to toxicity. In most cases, TOC or Cu alone were likely causes of toxicity (23 % and 22% of toxic

samples respectively). It was also concluded that Li, NH₃ and low hardness were occasionally contributors to toxicity.

Bruce A. Only three analyzed samples were reported in this study. Only one of the three was toxic and the main suspect in this case was hydrazine. The reason for the limited number of samples was that Bruce A is to some extent, in their early start in MISA compliance project. Since the legislation was not yet issued at the time of their start, they have chosen an advanced combination of processes to treat RWMTS as well as a non-selective treatment approach and Bruce A had confidence that this approach would render their effluent non-toxic.

Bruce B. From the 17 analyzed samples from Bruce B, 13 (76%) have failed either the *Daphnia magna* or the rainbow trout tests or both. Based on study results and some other related studies, it was concluded that hydrazine was the cause of toxicity in most of the RLWMTS samples at this site.

Later on, supplementary treatability studies were performed on samples from non laundry tanks from Darlington and Pickering stations (Reference 5 and 6). The results of these studies have confirmed the prediction that most toxic samples could be treated successfully with granulated active carbon (GAC) or IX resin (very often just cation resin). It also showed that in approximately 4 % of all samples none of the treatment worked in the first pass through both media in series. For such cases, mixing with other non-toxic waste batches was foreseen.

As mentioned while Bruce A and B have chosen the approach to treat all waste streams, Darlington and Pickering have adopted a selective treatment approach.

For Pickering and Darlington it was therefore necessary to translate toxicity into chemical parameters so that the most adequate treatment could be selected. The considered treatment was based mainly on filtration, GAC and IX resin. The additional reason for a selective treatment approach was that the large volume of discharged effluents in a day, in particular at Pickering (up to 500 m³ per day), would result in large volumes of secondary solid radioactive waste and high cost of consumables (e.g. filter cartridges, GAC, IX resin).

To enable a selective treatment approach, Derived Toxicity Limits (DTL) were developed for Pickering and Darlington stations. Derived Toxicity Limits are a combination of literature data and specific data obtained by OPT studies (Reference 7). Derived Toxicity limits are shown in Table 2.

DTL and TU represent a concentration or a value of a chemical parameter above which this parameter alone would cause more than 50 % lethality by either toxicity testing method. Synergies of several chemical parameters in causing toxicity were not originally

included in TU's or DTL's. DTL for OPG power plants are not considered to be a fixed value, but rather the best limit obtained from the available information. Learning about synergies between specific toxicants has already and may in future change the values of DTLs.

A new method of toxicity testing with *Daphnia magna* called "Daphnia IQ" was introduced later in the MISA compliance project. This method of checking the toxicity of the waste stream sample can be done within one hour. The results of this method were compared to the test results performed according to the MISA protocol and they are often on the conservative side i.e. more samples failed IQ tests than the regular toxicity tests by the MISA protocol. It was estimated that Daphnia IQ testing is showing failure in 10% more cases than regular *Daphnia magna* testing (Reference 8). The benefit of the "Daphnia IQ" tests far exceeds the drawback of potential unnecessary treatment of small volumes of liquid waste.

Table 2.: Currently Applied DTLs at Darlington and Pickering

Parameter	Limit or Range
pH	6 - 9.5
Ammonia	0.8 mg/L
Hydrazine	0.1 mg/L
Copper	0.05* mg/L
Iron	3.0 mg/L
Lithium	0.5 mg/L
Zinc	0.5** mg/L
Total residual Cl (TRC)	0.5 mg/L
Total Organic Carbon (TOC)	15 mg/L (7.5 ppm for DNGD)
Oil in Water	13.0 mg/L
Conductivity	10 to 150 mS/m
Total Phosphorus	1 mg/L
Total Suspended Solids (TSS)	21*** mg/L

*There are further restrictions to this limit related to hardness and Zn concentration

** There are further restrictions to this limit related to hardness and Cu concentration

*** This is the monthly average daily limit for and individual daily limit is 73 ppm

Wide Approach to Elimination of Acute Toxicity from OPG RLWMTS Effluents

The initial notion was that toxicity of radioactive liquid waste effluent could be eliminated just by selective or full waste stream treatment. This notion had to be abandoned early on, based on the information that treatment did not produce non-toxic streams in all cases.

A much wider approach was taken to eliminate toxicity. This approach included:

1. Best management practice, which is mainly minimizing the use of toxic chemicals in the plant (selection of non-toxic detergents and other chemicals).
2. Cleanup of active liquid waste tanks and sumps.
3. Minimizing the volume of generated liquid waste at the source.
4. Waste segregation. It was shown that in many stations, laundry waste was a significant contributor to toxicity. It was necessary, as a minimum, to separate the laundry waste from other “cleaner” waste streams. Historically, waste batches were mixed and in many cases, caused cross contamination with toxins.

As Bruce A had an early start in building technology to treat radioactive liquid waste for MISA compliance, they have chosen a more advanced technology consisting of filtration, waste stream pretreatment and reverse osmosis. Furthermore, additional washing machine capacity was installed at Bruce A. At the same time, there were limited treatment capabilities planned for Darlington and Pickering liquid waste treatment due to tight schedules. A logical decision was then made to transfer all laundry from Darlington and Pickering to Bruce A. This move has immediately decreased the burden of toxicity on both the Pickering and Darlington stations.

Modifications and New Treatment Systems for Radioactive Liquid Waste Effluents

The original RLWMTS systems were designed to reduce radioactivity to maintain effluents within Derived Emission Limits (DELs) and Station Target Limits (STLs) as well as to help reduce activity in the case of accidents. Historically, there were not many problems maintaining DELs and STLs even without the use of these systems. The systems typically consisted of filters and IX resins and were, in most cases, not capable of eliminating toxicity.

Bruce A started working on its new Radioactive Liquid Waste Treatment System in 1993. The Active Liquid Waste Treatment System (ALWTS) at B A is a complex, multi-stage treatment system (Reference 9). At the “heart” of the Bruce A ALWTS is a multi-stage reverse osmosis system (ROS) consisting of two separate RO stages (RO-1 and RO-2), followed by chemical precipitation / decantation and ultrafiltration (UF). As can be seen from the flowsheet in [Figure 1](#), the distinct yet highly interactive unit operations in the ROS are as follows.

- Pre-treated and filtered water is concentrated in the first stage of the ROS (i.e. RO-1). Processing of the water in RO-1 is facilitated by the addition of acid and antiscalant to prevent scale on the RO membrane surface.

- The concentrate from RO-1 is fed to the second stage (RO-2) for further processing and concentration. Acid is added and the pH carefully controlled to prevent scale formation. The permeate from RO-2 is recycled back to RO-1.
- Sodium hydroxide is added to the concentrate from RO-2 to adjust the pH and precipitate sparingly soluble compounds such as calcium carbonate, silica and metallic hydroxides. The precipitation occurs in the reaction/decant tank.
- The supernatant from the decant tank is clarified via the use of tubular ultrafiltration (UF) membranes, which remove suspended solids down to a pore size of 0.05 microns. The "softened" UF permeate is recycled back to RO-2 to enhance overall system recovery.

The ROS has been specifically designed to meet the very stringent performance criteria, which were originally specified by OPG.

A comprehensive study was undertaken to quantify the performance of the ROS system, since it is a crucial component towards the Station's goal of achieving MISA (environmental) compliance.

Pre-treatment to the ROS is currently provided by a "temporary" system consisting of polymer addition followed by disposable filtration (DFS). The disposable filters currently in use are the same filters that were put into service when the station was commissioned (at that time, the filters were used in conjunction with / as a pre-treatment to ion exchange (IX)). The DFS system is outdated and inefficient. Operating costs for the current pre-treatment scheme using DFS are extremely high, as the disposable filters are being replaced very frequently, at an enormous cost.

The remaining major component in the ALWTS is the evaporator solidification system (ESS). The concentrates from the ROS are fed to the ESS and evaporated via the use of a thin film evaporative process. The distillate from the ESS is processed via a polishing system prior to being fed to the permeate verification tank. The distillate polishing consists of UV oxidation with hydrogen peroxide to eliminate volatile organics from the distillate. The evaporator bottoms are solidified in bitumen and the solidified residue placed in a container for storage on-site as a low-level radioactive waste.

Here is a summary of the results of a comprehensive evaluation, which was conducted on the ROS system between September, and mid-December 1999. Key findings are as follows:

- First Stage RO membranes (RO-1) are delivering the design flux of approximately 17 US gallons per sq. ft. per day (GFD) after approximately one year of operation, albeit for frequent membrane cleaning averaging 1 – 2 cleaning applications per week (versus a typical frequency of once per month).

- The ROS system is delivering very high permeate recoveries ranging from 97.5% to 98.5%, compared to the maximum design value of 99.1%.
- The quality of treated RO-1 membrane permeate meets or exceeds expected performance from the ROS, having a typical conductivity in the range 15 – 20 $\mu\text{S}/\text{cm}$.
- The treated membrane permeate meets MISA parameters including Total Suspended Solids (TSS), total phosphorus, zinc, iron and Oil and Grease. The pH of the treated RO permeate is typically in the range 5.5 – 6.5, compared to the MISA range of 6 – 9.5. Treated permeate pH is adjusted via the use of a Neutralizing Filter (calcium carbonate bed). The RO permeate is sometimes toxic to fish. After treatment with the Neutralizing Filter, the RO permeate is non-toxic. The pH of the final discharge after neutralization is typically in the range 7.5 – 8 and the hardness is in the range 50 – 80 mg/L as CaCO_3 .
- The ROS efficiently reduces Mixed Gamma radioactivity levels. Mixed Gamma levels in the Discharge Tanks range from 0.002 $\mu\text{Ci}/\text{kg}$ to 0.03 $\mu\text{Ci}/\text{kg}$ which are approximately two orders of magnitude (i.e. 99%) lower than their concentration in the ALW Collection Tanks (typically 0.25 $\mu\text{Ci}/\text{kg}$ – 1.0 $\mu\text{Ci}/\text{kg}$).
- The concentration of heavy metals in the RO permeate including iron, copper, nickel, zinc, barium, manganese, chromium, mercury, lead, arsenic, antimony, selenium, cobalt, vanadium, beryllium, cadmium, molybdenum, silver and thallium were all below detection limits, with typical rejection efficiencies > 99%.
- The overall concentration of organic compounds as Total Organic Carbon (TOC), including Total Oil and Grease (TOG), Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD) in the RO permeate were all at about 1.0 mg/L or less, confirming the RO membranes' very high separation efficiency of 90 - 95% for organic molecules such as laundry surfactant, lubricating and hydraulic oils, water treatment (flocculating) polymers and polymeric scale inhibitor.
- The current ROS pretreatment is inadequate, resulting in very high concentrations of colloidal suspended solids, Silt Density Index (SDI), turbidity and iron fed to the ROS system. This poor water quality has caused severe fouling and scale formation on the second stage RO (RO-2) and ultrafiltration (UF) membranes as well as requiring frequent and very costly replacement of the DFS filters.

After operating for almost a year using the "temporary" DFS filters, two new pre-treatment processes have been selected and installed and are currently awaiting to be commissioned (see [Figure 1](#)). Gray Water Processing Unit -GWPU by Aquatronics, which utilizes dissolved air flotation (DAF) ozonization, and filtration will be used to treat the laundry waste. A proprietary clay-based chemical addition process, supplied by

CETCO, which brings about flocculation, coarse filtration, excess polymer removal and fine filtration to produce water with low SDI will also be implemented. Additional monitoring of the Bruce A ROS system is planned after start-up of these pre-treatment systems. These new pre-treatment systems are expected to greatly reduce the frequency of DFS filter change-outs.

Bruce B has build a new system based on the toxicity studies (Reference 4) that hydrazine is the main cause of toxicity in their waste streams. They have a NaOCl addition for hydrazine oxidation with GAC downstream to remove residual chlorine. With these simple modifications, they have managed to maintain their RLWMTS effluents non-toxic.

Darlington has chosen a two-phase approach to the MISA compliance project.

In the short-term phase, MISA compliance was achieved with the modification of the existing ALW treatment system. The modification consists of using one of two IX vessels for GAC and adding the piping between two IX vessels to enable the use of the GAC and IX resin in series. There is existing filtration in the system which uses cartridges with 40 μ pore size. Initial GAC loading problems have necessitated the search for a media with similar processing characteristics, but different physical characteristics. A mixture of non-polar and weakly basic Macronet resin was selected. Handling of the Macronet was the same as that of IX resin and the adsorption properties were very similar to those of GAC. The initial study of treatability with Macronet at OPT has indicated an effectiveness in TOC and Cu reduction. Furthermore, the TOC values have decreased since removal of the laundry from Darlington site and it was therefore expected that Macronet would be used less frequently and for lower concentrations of TOC.

The long-term solution will be confirmed by monitoring the performance of the short-term modification. For the long tem solution, a combination of filter, Macronet cation resin and neutralizing filter is recommended (Reference 10). The operation of the modified ALW system has confirmed the laboratory results that Macronet can successfully reduce TOC in the waste stream.

Macronet also successfully reduces copper in the waste stream. This was found to be important because GAC treatment does not affect hardness in the waste stream. The disadvantages of treating copper and zinc with IX resin is that it also removes hardness and makes the traces of copper and zinc more toxic.

Pickerin station has designed and built a new system for RLWMTS. The system consists of a UV unit to prevent biofouling in the system, cartridge filtration, granulated active carbon, cation bed IX exchanger and neutralizing bed.

The block diagram of the new treatment system is shown in [Figure 2](#).

The system is designed to treat liquid waste selectively. Waste streams that contain high TOC will be treated with GAC. If metals and other cations are indicated to be the cause of toxicity, cation IX resin will be used. As the design of the system has started when there was no decision on removal of laundry from the Pickering site, the UV unit was originally designed for organic destruction with hydrogen peroxide. After the decision about laundry elimination from the site, UV was applied as a biocide (or biostat) before filtration. The need for this was discovered in early attempts to use the existing old treatment system when frequent biofouling of filters was experienced. The piping was designed so that UV oxidation with hydrogen peroxide could be used in the future if necessary.

In the original design of the new system, one vessel was foreseen for anion resin if it was shown to be necessary in the future. Since of all ionic compounds, the cations are indicated as causes of toxicity, the last IX column was modified into a neutralizing filter with a CaCO_3 bed. The benefit of this neutralizing bed is multifaceted as was found later during the MISA project. The CaCO_3 bed neutralizes acidic effluent after cation resin and also acts as polishing filter for the effluent. Furthermore, the CaCO_3 bed adds some hardness to the waste stream, which very often helps to reduce the effects of traces of untreated toxins. Examples of this are with copper and zinc, which are more toxic without the presence of hardness. Therefore, treatment with IX resin very often does not eliminate toxicity of copper and zinc. It was found out that passing the waste stream through a neutralizing filter after treatment with IX resin renders it non-toxic.

Figure 1 Bruce 1-4 Active Liquid Waste Treatment System (ALWTS) Flowsheet

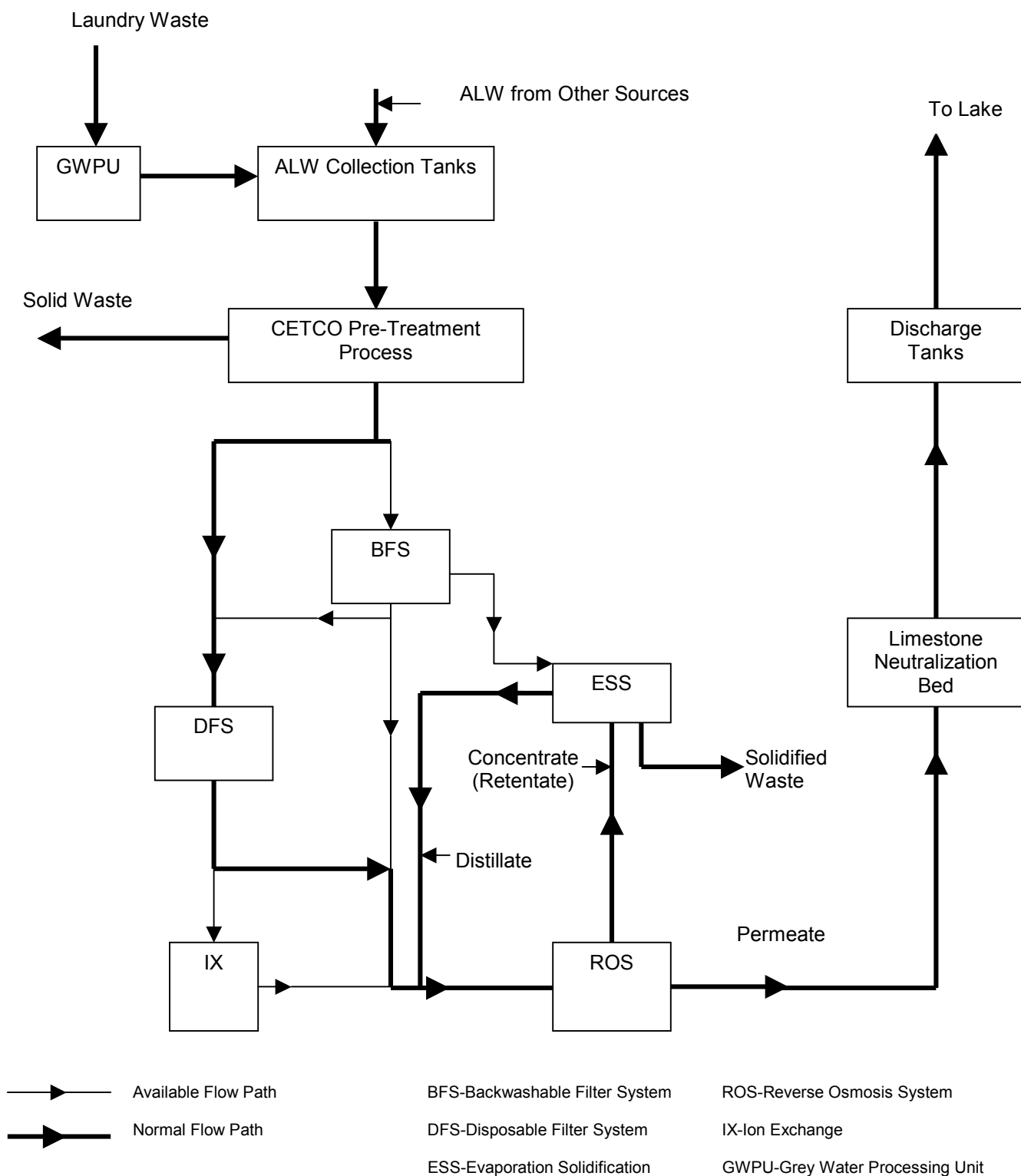
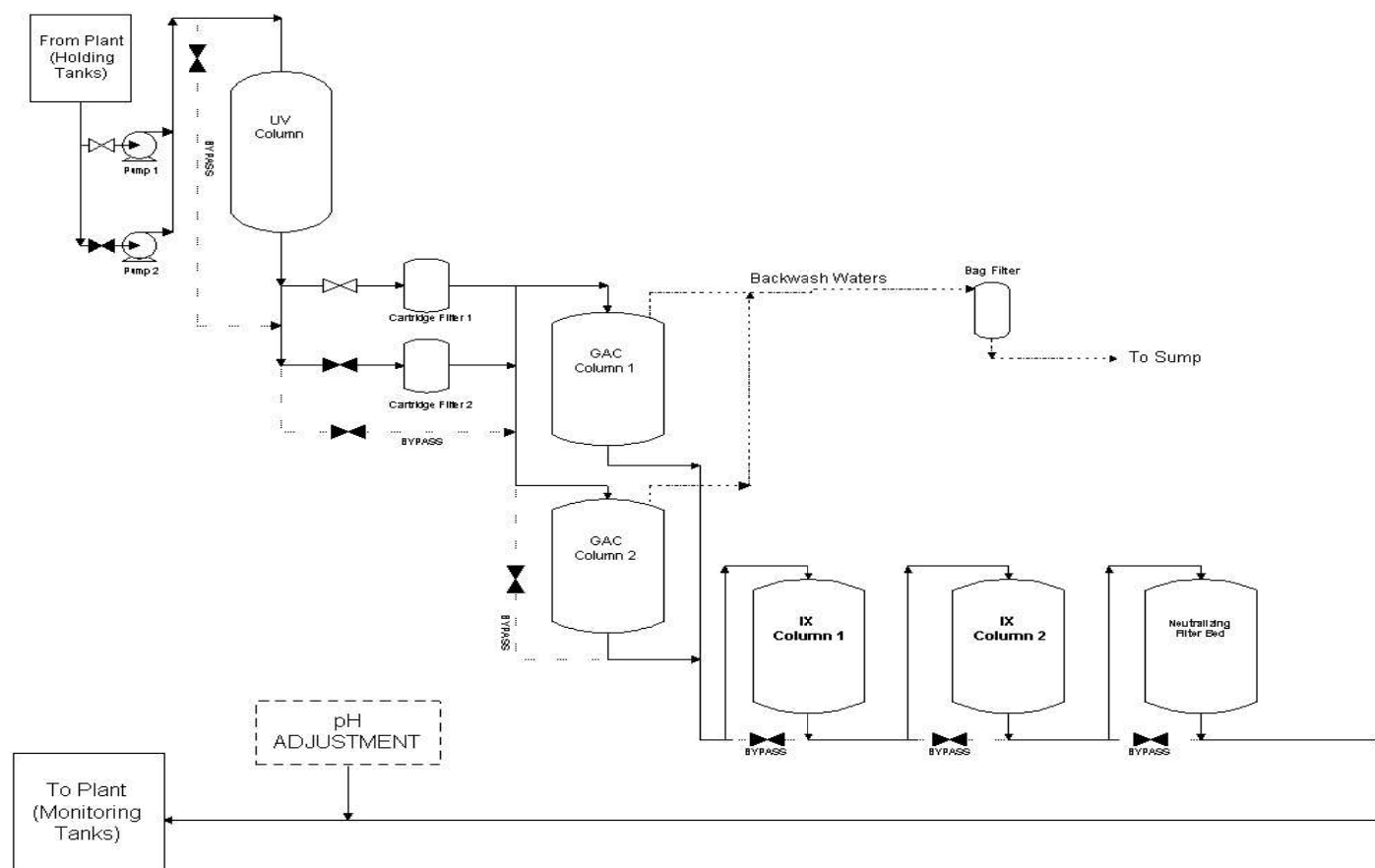


Figure 2; New Pickering Radioactive Liquid Waste Treatment System



Conclusion and Future Activities Planned

The overall effort to achieve MISA compliance at OPG Nuclear sites was extremely challenging, in particular because of the very tight time frame. Lessons have been learned in the areas of procurement and design. Internal management processes have been established to ensure continued MISA compliance.

The installed systems are capable of maintaining MISA compliance with the attentive operations staff. For the plants that have a selective treatment approach, close monitoring of system performance is necessary. Besides maintaining MISA compliance, close monitoring can also help minimize secondary solid radioactive waste generation. Neutralizing filtration with CaCO_3 provides an additional shield and edge against MISA non-compliance. Combined with cation resin CaCO_3 bed can also reduce resin consumption of IX resin to one third of when mixed bed IX resin is used.

There are indications that filtration at Pickering and Darlington could be optimized with respect to pore size and filtration technology. This would protect GAC/ Macronet and IX resin from being used as filters.

A multifaceted approach to MISA compliance, which includes reduction of toxicity at the source, was necessary because treatment technology alone could not guarantee MISA compliance. Best management practices like tank cleanup, control of chemicals used in the plant etc., will be continued and expanded if necessary.

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