Protecting Fresh Water Resources during a Large-scale Low-Level Radioactive Waste Clean-up Project with Best Available Technology

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The Port Hope Area Initiative involves the development of two new long-term waste management facilities at separate Ontario sites, currently containing low-level radioactive waste materials. Water treatment for contaminant removal has been an integral operating component at these sites since the late 1970s. This paper describes a staged development strategy for ensuring that Best Available Technology options will be considered during the specification of the water treatment requirements for these two projects. The objective behind this strategy is to enable the projects to make a positive contribution to the preservation of a healthy, prosperous and sustainable Great Lakes Basin ecosystem.

1. Project background

1.1 Project scope

The Port Hope Area Initiative (PHAI) is focused on developing two new long-term waste management facilities in the form of above-ground engineered mounds for low-level radioactive wastes (LLRW) that are mostly by-products of uranium and radium ore processing activities of the former crown corporation Eldorado Nuclear Limited (Eldorado) and its private sector predecessors in Port Hope dating from 1932 through 1988. A contextual overview of the PHAI is provided in a companion paper "Making Strides on the Port Hope Area Initiative: Canada's Largest Low-Level Radioactive Waste Clean-up Project" [1]. The PHAI is being led by the Port Hope Area Initiative Management Office (PHAI MO), which consists of a partnership of Atomic Energy of Canada Limited (AECL), Natural Resources Canada and Public Works and Government Services Canada. AECL has the lead role for developing the water treatment technology.

AECL received a Waste Nuclear Substance License for the Port Hope Project, which includes the on-going operation of the Welcome Waste Management Facility (WMF), in 2009 October. This licensing action triggered a process to transfer the WMF and the ownership of the lands on which it is situated from Cameco to the Government of Canada. The transfer, and AECL's tenure as licensee on behalf of the federal government, is currently expected to take effect in early April 2010. This is at least two years prior to the start of the construction associated with the new longterm waste management facility (LTWMF). A similar license issue (to AECL) and land transfer (to the Government of Canada) regarding the Port Granby WMF in Clarington is expected to take place in 2011.

1.2 History of existing waste sites

1.2.1 Welcome Waste Management Facility

From 1948 to 1954, solid wastes from Eldorado's operations were deposited at the Welcome WMF, just outside Port Hope and near the village of Welcome. Eldorado's waste management practices of that era were typical of the industry. Due to the potential commercial value of some of the waste products, which contained residual processing chemicals as well as the naturally occurring radioactive elements and various metal elements (e. g., cobalt, copper, nickel and silver) separated from the original pitchblende ore concentrate, they were simply end-dumped on the ground in a designated area of the site and left exposed to the elements. Arsenic was also a significant contaminant associated with these wastes due to its high levels in ore concentrates derived from mines in the Northwest Territories.

Erosion of the waste at the Welcome WMF resulted in off-site transport and environmental concerns within the local surface water drainage system. In response, Eldorado constructed water collection and retention ponds and, in 1956, installed a pumping system and a buried three kilometre-long pipeline to transfer the collected water directly to Lake Ontario. In 1978, Eldorado installed a water treatment plant to reduce the loading of arsenic and radium-226 to the lake. The treatment process was based on ferric chloride (FeCl₃) injection, as a solution, to precipitate these elements as insoluble solids, which were separated from the flow by gravity settling in a series of holding ponds. Clarified water continues to be pumped to Lake Ontario via the pipeline. Accumulated precipitates are periodically removed from the treatment ponds and relocated to the original waste burial area.

Treatment system performance data obtained from Cameco for the 12-year period 1997 through 2008 are summarized in Table 1 below. It shows that very good removal efficiency exists for arsenic but less so for radium-226 and uranium.

	Units	Treatment	Treatment	Removal	Licensed
		Inflow	Discharge	%	Release
					Limit
Volume	m ³ /year	115,185	110,807	NA	NA
Arsenic	mg/L	0.545	0.014	97.5	0.5
Uranium	mg/L	0.333	0.166	50.1	NA
Radium-226	Bq/L	0.096	0.057	40.6	0.37
NA = Not Applicable					

Table 1. Summary of Welcome WMF Water Treatment System Performance

1.2.2 Port Granby Waste Management Facility

From 1955 until 1988, wastes from Eldorado's operation were emplaced at a site it had purchased near the hamlet of Port Granby in the southeast corner of the Municipality of Clarington on the north shore of Lake Ontario. In fact, this site was selected so that leachate and runoff could flow directly into the lake (i.e., no need for a pipeline). By 1954, radium recovery was being phased out of Eldorado's production operations and a number of new uranium refining and conversion processing operations were being introduced. These new processes, which included the production of uranium trioxide, uranium dioxide, uranium hexafluoride and uranium metal, resulted in a greater variety of waste types and characteristics being present at Port Granby when compared to the materials at the Welcome WMF. Consequently the characteristics of leachate from the Port Granby site was also more diverse than that at the Welcome WMF.

In 1977, water collection and treatment facilities were installed to limit the release of contaminants to the environment. Initially, the treatment system was based on barium chloride $(BaCl_2)$ and aluminium sulphate $(Al_2(SO_4)_3)$ addition for co-precipitation of radium-226 and ferric chloride (FeCl₃) addition for arsenic removal. By the early 1980s, it was observed that FeCl₃ alone was satisfactory for both arsenic and radium-226. In the mid 1980s, however, it became evident that the treatment process was losing efficiency in terms of arsenic removal. Eldorado's investigations into the cause of the decreasing performance concluded that it was due to a shift in the chemical form of the arsenic in the collected waters. At that time, it was determined that one of the refinery waste types (calcium fluoride residue) had started to contribute a second source, and form of arsenic in the leachate collected for treatment. The speciation was subsequently determined to be arsenic hexafluoride (AsF₆), which was also found to not be amenable to removal by common precipitation techniques.

Treatment system performance data obtained from Cameco for the 12-year period from 1997 through 2008 are summarized in Table 2 below. It shows that moderately good removal efficiency exists for Ra-226 but not so for arsenic. Because uranium is not specifically targeted by the Port Granby treatment process, it is not monitored in the inflow or discharge. However, uranium concentrations are monitored in the final discharge from the Port Granby site, which includes the Treatment Discharge, and which for this same period, averaged 1.9 mg/L.

	Units	Treatment	Treatment	Removal	Licensed
		Inflow	Discharge	%	Release
					Limit
Volume	m ³ /year	40,235	NA	NA	NA
Arsenic	mg/L	1.664	1.283	22.9	NA
Uranium	mg/L	NA	NA	NA	NA
Radium-226	Bq/L	0.672	0.253	62.4	0.37
NA = Not Applicable					

Table 2. Summary of Port Granby WMF Water Treatment System Performance

1.3 PHAI environmental assessment of current conditions

AECL started environmental Baseline Characterization Studies for the Port Hope and Port Granby Projects in 2002. The Port Hope Environmental Assessment Screening Report (2006 December) [2] concluded that there would be no significant adverse effect associated with the discharge of effluent from the water treatment process, which was assumed to be based on a technology comparable to that currently employed at the Welcome WMF. Likewise, the Port Granby Environmental Assessment Screening Report (2009 August) [3] also concluded that

there would be no significant adverse effect associated with the discharge of effluent from the water treatment process, which was also assumed to be based on a technology comparable to that currently employed at the Port Granby WMF. Nevertheless, it was deemed prudent to reconsider the overall effects from the context of the ALARA¹ principle and from the perspective of minimizing pollution. An examination of options for enhancing treatment effectiveness was therefore carried out as part of specifying details of the requirements for the Port Hope Project. In 2008, this resulted in the formulation of a strategy for addressing water treatment requirements applicable to the PHAI objectives.

1.4 Regulatory direction

Based on 1) regulatory agency review comments on the two PHAI Environmental Assessment Study Reports and 2) Canadian Nuclear Safety Commission (CNSC) staff review of preliminary design description documentation for the respective projects, it became evident that apparent changes in regulatory direction would likely have some bearing on the specification and development of water treatment requirements. The following principal aspects were thought likely to impact this development effort:

- The CNSC, in its mandate to ensure licensees are in compliance with the General Nuclear Safety and Control Regulations to take all reasonable precautions to control the release of nuclear substances or hazardous substances into the environment, is looking to [licensees] to consider the application of Ecological Risk Assessment to aid in the definition of regulatory expectations and requirements [4].
- CNSC staff expect licensees to consider treatment requirements and environmental protection objectives in the context of using Best Available Technology (BAT)² that is economically achievable [5].

2. Approach to developing water treatment requirements

2.1 Overall approach

The first critical challenge faced was predicting future water flow rates and contaminant concentrations. The Welcome WMF has been in a relatively static condition since the mid 1980s and the Port Granby WMF since 1988. All wastes are under fill cover and no waste excavation or disruption has taken place. However, during the remediation of the existing WMFs and during the Construction and Development Phase of the projects, the existing on-site conditions will change dramatically due to:

¹ Proponents are generally encouraged to apply the ALARA principle (As Low As Reasonable Achievable, social and economic factors considered) where human health or environmental safety concerns exist to ensure that impacts are not managed to just meet regulatory objectives, but to do better if it is reasonably achievable.

² Various forms of this term are in use, e.g., BDAT (Best Demonstrated Available Technology) or BATEA (Best Available Technology Economically Achievable). These are assumed to have essentially the same sense and meaning. Consequently, only BAT is used in this paper.

- Waste materials being openly exposed to the weather for varying periods of time;
- Greater direct contact between waste and storm water flows resulting in potentially higher concentrations of Contaminants of Concern (COCs); and
- The potential for more water to treat due to waste excavation area dewatering requirements and the large open area of the new LTWMF that will be impervious to water infiltration.

In addition, the new LTWMF in Port Hope will be receiving wastes from a number of sites within the urban part of the municipality that might introduce a different suite of contaminants than currently found at the Welcome WMF (e.g., Coal Gasification Plant residue, Chemetron lagoon residue). Consequently, these may contribute unique water quality characteristics that will combine into an overall treatment inflow.

To address these challenges a progressive development strategy was adopted in order to determine the optimum water treatment requirements for the Port Hope and Port Granby projects. Separate, but coordinated specialist consulting teams were engaged to start the water treatment requirements definition process for each project.

The approach to the effort for both projects was structured to consist of the following four stages, two of which have been completed and two are planned for 2010:

- i. Assessment of Treatment Requirements and Applicable Technologies (2008)
- ii. Bench Scale Testing Program (2009)
- iii. Pilot Scale Testing Program (2010)
- iv. Detailed Design (2010)

The scope, status and major findings to date (where applicable) are discussed in the following sections.

2.2 Assessment of treatment requirements and applicable technologies

2.2.1 Treatment requirements (2008)

The assessment of treatment requirements consisted of two basic components:

- i. Develop estimates for potential total inflow quantities.
- ii. Estimate potential future inflow quality.

This assessment was made possible by consulting the existing and extensive database on:

- i. Waste type characteristics;
- ii. Associated site groundwater quality data;
- iii. The results of previous studies on waste leaching potential;

- iv. The results of hydrological studies carried out for the respective Environmental Assessments; and
- v. Preliminary engineering design information regarding the sites and facilities.

2.2.1.1 Future flows and water quality – Port Hope

The existing Welcome WMF treatment inflow currently consists of flows of surface water runoff and groundwater discharge to surface that are collected in the on-site drainage/collection network. Since the new LTWMF will be on the same site as the existing Welcome WMF, it is expected that the existing water treatment system will be utilized for the Port Hope project. As a result, during the Construction and Development phase, four additional contributing water sources will be present in the inflow:

- i. On-site waste excavation area dewatering flows;
- ii. LTWMF base sub-drain water;
- iii. LTWMF leachate; and
- iv. Water from equipment washing and decontamination operations.

With these multiple sources, it is estimated that during the Construction and Development phase of the Project, flow volumes may be up to twice that currently being treated at the Welcome WMF (i.e., $250,000 \text{ m}^3/\text{a} \text{ vs.} 124,000 \text{ m}^3/\text{a}$) [6]. It was concluded that the best way to deal with the increased flow volume is to increase the capacity of the discharge pumping to Lake Ontario. Late in 2008 AECL was in dialogue with Cameco regarding waste site operations when Cameco advised that they were planning to replace the existing 75 mm diameter pipeline with a new 100 mm diameter line during 2009. At AECL's request, Cameco designed the replacement to consist of a double 100 mm diameter pipeline installation. This work was completed and commissioned in the fall of 2009.

A conservative approach was employed to evaluate potential future inflow concentrations, in which the projected maximum concentrations derived from the available data for the each of the five individual sources noted above (existing inflow plus four additional water sources) were considered to be typical for that source. Note that the LTWMF leachate source above includes potential contributions from the waste remediation sites in the urban area of the Municipality of Port Hope. A weighted mass balance approach, based on source waste volume, was used to estimate the resulting combined influent water quality that could be expected in the existing water collection ponds during on-site construction and operations. The resulting projected maximum influent concentrations for the primary Port Hope COCs are shown in Table 3 [6].

As a result of this assessment, one small (non-radioactive) waste source was identified as contributing substantially to the list of potential contaminants due to the presence of Polyaromatic Hydrocarbons (PAHs). Since this waste has no LLRW, an alternate to disposal in the Port Hope LTWMF is being pursued, as opposed to pursuing treatment technologies which may have to deal with PAHs.

2.2.1.2 Future flows and water quality – Port Granby

Because the new Port Granby LTWMF will be located several hundred metres away from the existing Port Granby WMF, it is expected that there will be separate water collection and treatment facilities at each location. The existing water treatment system will be used to treat existing flows in addition to water collected as a result of runoff and ground water infiltration into waste excavation areas and from small miscellaneous sources such as equipment decontamination facility discharges. A new water treatment system will be constructed adjacent to the new Port Granby LTWMF to treat water and leachate collected during the time the facility is receiving waste materials prior to the final cover installation.

Based on site-specific hydrological assessment, it was estimated that the peak flow rate at the existing water treatment system would be about 1.5 times the recent historic maximum (monthly) flow rate, increasing from about 9,000 m³ / month to about 13,000 m³ / month. Peak monthly flow rates for the new water treatment facility were estimated to be about 12,000 m³ [7].

The determination of potential future inflow concentrations for the Port Granby project was more straight-forward than for the Port Hope project. Because the existing Port Granby WMF is the only waste source, projection could be inferred from groundwater monitoring results obtained during the Port Granby Environmental Assessment. In addition, the results of laboratory leach tests done in 1994 on Port Granby wastes were reviewed and considered for projecting potential future water quality, especially at the new LTWMF. The resulting projected maximum influent concentrations for primary Port Granby COCs are also shown in Table 3 [7].

	Arsenic mg/L	Uranium mg/L	Radium-226 Bq/L
Port Hope – New LTWMF	26.6	11.6	10.6
Port Granby – Existing WMF	10	9	22
Port Granby – New LTWMF	10	20	75

Table 3. Projected Maximum Concentrations for Primary COCs in Treatment Inflows

2.2.2 Technology assessment (2008)

The core of this assessment, for both projects, consisted of the following steps:

i. Conduct comprehensive technology applicability assessment. In order to identify processes that have potentially high treatment efficiencies for the COCs in the treatment inflow, a survey of applicable technologies was carried out. The survey examined a wide array of technologies under the basic processes of sorption, biological, membrane, chemical, and other physical processes. The technology survey was deliberately broad to consider a wide range of options.

- ii. Conduct review of precedents from previous studies. The review included information from existing wastewater treatment plants at conceptually similar sites such as uranium mines and mills, tailings management facilities and radioactive waste management sites, mostly in Canada or the United States.
- iii. Screen potentially applicable technologies. A two-stage screening process was employed to determine the broad applicability of the technology for the Port Hope and Port Granby projects through consideration of the relative ease of implementation and overall treatment efficiency. The technologies determined to be most applicable were then evaluated by means of an advantages / disadvantages matrix to identify a preferred system for each project. Based on the comprehensiveness of the screening process, the preferred system is considered to be an example of BAT that is economically achievable in the context of the PHAI projects.

For both the Port Hope and the Port Granby projects, it was determined that in order to achieve the desired discharge water quality objectives, the existing water treatment processes would require augmentation of the coagulation-precipitation-flocculation systems already in operation. Table 4 identifies the resulting potentially applicable technologies and conceptual treatment process configurations for the Port Hope and Port Granby projects.

	Port Hope		Port Granby
I.	General pre-treatment (e.g., filtration or pH adjustment) followed by a two- stage process utilizing coagulation / precipitation (with ferric chloride) and Reverse Osmosis (RO).	I.	General pre-treatment (e.g., filtration or pH adjustment) followed by coagulation / precipitation using ferric chloride followed by IX.
II.	General pre-treatment followed by coagulation / precipitation using ferric chloride followed by Ion Exchange (IX).	II.	General pre-treatment followed by coagulation / precipitation using ferric chloride followed by RO.
		III.	General pre-treatment followed by coagulation / precipitation using ferric chloride followed by evaporation.

 Table 4. Preferred Feasible Water Treatment Technologies

Evaporation was not considered a feasible post-treatment solution for the Port Hope project due to the much higher volume of water subject to treatment.

For the Port Hope preferred technology approach (I), two alternatives were proposed:

• Alternative IA – is based on the ferric chloride coagulation / precipitation being the lead step, which would be followed by RO for final polishing.

• Alternative IB – is based on RO being the lead step followed by the ferric chloride coagulation / precipitation process for treating the reject (concentrate) stream from the RO stage.

2.3 Bench scale programs

Bench scale testing programs to confirm the feasibility of the preferred technologies were developed and carried out during the fall of 2009. The same sets of consulting teams were employed as for the requirements and technology assessments. The bench scale testing programs include the following primary elements:

- i. Background data collection
- ii. Testing program design
- iii. Water Sample Collection
- iv. Preliminary bench top screening tests (for concept feasibility confirmation)
- v. Comprehensive bench top testing (for preliminary system design)

2.3.1 Approach to experimental design

A second major challenge in the overall water treatment requirements determination was the acquisition of representative water samples. As noted above, there is a high likelihood that the levels of COCs in the inflows will be different (higher) during the PHAI Construction and Development stage than what is currently collected. The approach used to address this concern – there is no present source of future water – consisted of preparing various composite samples by collecting water from a number of different individual sources, including groundwater monitoring wells located within the waste burial areas. The individual source sites were selected on the basis of associated waste types in the area, water quantity yield and expected relatively high contaminant concentrations.

A series of jar tests for coagulation / precipitation experiments were designed and conducted to determine factors such as optimal ferric chloride dose rates, flocculent requirements, mixing time, settling time and the like. All jar tests were carried out at the investigating consultant's laboratories.

For the Port Hope project, a custom-built RO test unit (see Figure 1) was installed at the Welcome WMF to conduct the comprehensive testing program. This unit was supplied by a vendor specializing in RO equipment for waste water treatment applications. It features a unique disk-shaped membrane configuration, which is claimed to be superior to more conventional tabular construction.

During the preliminary bench top screening tests for the Port Granby project, it was determined that due to the high levels and composition of total dissolved solids (calcium, magnesium, sodium, potassium, sulphate, ammonia, nitrate and nitrite) in the composite samples, the efficiency of an IX process would not likely be satisfactory to achieve good water quality results. Consequently, the decision was made to focus the comprehensive bench scale program on the

second preferred approach – RO. In the case of the Port Granby project, containers of the composite samples were shipped to the investigating consultant's laboratory to carry out the tests. The comprehensive RO testing on Port Granby composite water samples was carried out with an available conventional lab-scale unit.



Figure 1. Custom-made Bench Scale RO Unit at Existing Welcome WMF Water Treatment Plant

2.3.2 Summary of Preliminary Results

Final reporting on the bench scale testing program has not yet been completed. However, some of the key findings are listed below.

2.3.2.1 Port Hope

- 1. The RO process appears capable of achieving very high rejection rates (typically > 99%) for all contaminants in all the composite samples tested. When combined with the ferric chloride coagulation / precipitation process, an overall (BAT) system will be capable of achieving enhanced water treatment objectives.
- 2. The testing has shown that a flexible configuration, switchable between Alternatives I and II may be the best overall approach. Alternative II may be more suitable at lower contaminant levels in the inflow, i.e., base flow conditions whereas Alternatives I may be better during periods when higher concentrations are expected, i.e., when certain waste types are being excavated or placed into the new LTWMF.

2.3.2.2 Port Granby

- 1. The RO process appears capable of achieving very high rejection rates (typically > 99%) for all contaminants in all the composite samples tested, including the arsenic hexafluoride currently not removed with the existing process. When combined with the ferric chloride coagulation / precipitation process, an overall (BAT) system will be capable of achieving enhanced water treatment objectives.
- 2. Pre-treatment for ammonia and/or nitrate/ nitrite removal may be required.
- 3. Evaporation may be feasible for final brine (concentrate) treatment.

2.4 Pilot scale program and preliminary design

A pilot scale testing program needs to be designed so that it can validate the bench scale testing program outputs as well as identify those process issues that would likely become apparent only during real-world operations, thereby improving the robustness of the final design. Pilot scale testing is expected to be carried out later in 2010. Following the pilot scale testing program, design requirements will be prepared to enable the preparation of detailed design documents, drawing and cost estimates.

Secondly, following completion of the bench scale testing program, a preliminary design can be prepared, which could include, but not be restricted to, tentative equipment specifications, operation process parameters, facility layout recommendations and the like. The preliminary design will enable AECL to inform stakeholders of the more specific details with respect to water treatment system requirements and to pursue the planning of the pilot scale testing program. The preliminary design is expected to be completed early in 2010.

3. Issues and challenges

Residual uncertainties remain regarding future water volumes and contaminant loads. These cannot be resolved until waste excavation begins and leachate from the new LTWMFs starts to accumulate. However, the bench scale testing program results do suggest sufficient robustness to accommodate a range of inflow characteristics.

The best method to achieve an appropriate balance between BAT and regulatory expectations still needs to be evaluated. At the time of writing, preliminary system design information, including potential implementation costs are not yet available. Final decisions with respect to treatment system installations will need to consider cost versus improved performance.

4. Summary & conclusions

AECL's approach to defining water treatment requirements for the Port Hope and Port Granby projects appears to be leading to the application of new Best Available Technology installations. Based on the high contaminant rejection rates observed in the bench scale tests for the primary COCs, it is concluded that the proposed technology approach would be capable of minimizing pollution and thereby contributing to the preservation of a healthy, prosperous and sustainable Great Lakes Basin ecosystem. In addition, these bench scale test results are consistent with the general experience noted at other waste water treatment facilities at the sites considered during the assessment described in Section 2.2.2 as being conceptually similar to the Port Hope / Port Granby facilities. For example, Metzler, et al, (2001) [8] and SENES (2006) [9] showed that the kinds of technologies currently proposed for the PHAI projects are already being utilized in generally comparable settings and producing good quality effluent. As a result, it is expected that the technologies evaluated during the PHAI bench scale testing program will be capable of producing effluent that will be protective of the local fresh water resources potentially impacted by the two projects.

For the Port Hope Area Initiative's two distinct, but comparable water treatment projects, a common approach of sequential investigation initially yielded two distinct theoretical solutions. A common, sequential approach to feasibility testing confirmed one of the theoretical solutions but ruled out the other. As a result of the coordinated approach between the two separate projects, the results of the one become the catalyst for a favourable solution to the other. Lessons learned by each project were shared to yield stronger results in both individual projects.

Further testing and design, and likely refinement along the way, will be required to complete the water treatment requirements definition for the PHAI. It is expected that specific effluent quality predictions will be possible once the next stage of the water treatment requirements determination process – Pilot Scale Testing – is completed, by late in 2010. At such time, the actual degree of fresh water resource protection to be provided by the water treatment technology can be quantified.

5. References

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