Conditional Clearance of Very Low-Dose Spent Solvents: Regulatory Issues from a Practitioner’s Perspective

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ABSTRACT

Spent solvents generated from Boiler Chemical Cleanings (BCC) at nuclear reactor sites may contain radionuclide activities in amounts that exceed IAEA exemption levels. A pathways analysis can be undertaken to estimate the dose consequences associated with site-specific release and disposal options for such waste streams. Historical practise in Canada has been that if the dose consequence is less than *de minimis* for all potential receptors, then it should be possible to release and dispose of such wastes as non-radioactive materials.

A pathways analysis for spent solvents and associated rinse water from BCCs at the Pickering Nuclear Generating Station (PNGS) was carried out to illustrate this concept. Several transportation and disposal options were considered. The results indicate that the doses are less than *de minimis* for all the options considered in this study. Conditional clearance levels can also be derived from the same calculations. These levels correspond to the maximum activities which would result in a *de minimis* dose. Site-specific conditional clearance levels can be used to illustrate the conservative nature of the IAEA exemption activities. At the moment, Canada is moving towards establishing exemption levels for disposal of radioactive wastes.

INTRODUCTION

Spent solvents are generated as a result of Boiler Chemical Cleanings (BCC) at CANDU reactor sites. These solutions contain small amount of radioactivity from a number of different sources including:

- Cut tubes – short sections of boiler tubes are infrequently removed from the boilers for a detailed characterization. These tubes are typically only plugged at the tubesheet allowing the primary side deposits to be exposed to BCC solvents.
- Tube leaks – primary to secondary side leaks also occur infrequently as a result of tube degradation. Radioactivity from the leaking fluid can consequently be deposited in the sludge on the secondary side of the tubes.
- Diffusion of tritium – during normal operation of the reactor units, tritium slowly diffuses from the heavy water in the primary heat-transfer system to the light-water coolant on the secondary side. Some of this tritium is retained in the secondary side deposits.

The Pickering Nuclear Generating Station (PNGS) would like the flexibility to have several options for handling the spent solvent waste and associated rinse water from BCC. To this end, a radiological pathways analysis was undertaken to determine dose consequences associated with each option. Sample results from this study are included in this paper.

The pathways analysis is used in this study to calculate dose to hypothetical receptors including individuals such as truck drivers, incinerator workers, residue (ash) handlers, residents who live near the landfill, inadvertent intruders into the landfill after closure and residents who live near the outfall. This dose is compared to a *de minimis* dose. A *de minimis* dose or dose rate represents a level of risk, which is generally accepted as being of no significance. Shipments of spent solvents and rinse water with corresponding doses below *de minimis* can be sent to conventional (i.e., non-radioactive) landfills for incineration and disposal as the radioactive dose associated with them is much less than natural background. A similar approach was previously undertaken by Leung [1], Benovich [2], Garisto and Strain [3] and Garisto and Belanger [4].

This particular paper focuses on the methodology used to estimate the dose for each option of managing the BCC solutions. It illustrates the methodology with example calculations assuming one cleaning per year and discusses implications to the planned BCC at PNGS. Other aspects of the ongoing study such as derivation of conditional clearance concentrations and/or activities are beyond the scope of this paper. These details are provided in Garisto and Eslami [5].

WASTE STREAMS

The waste streams considered in this study include:

- Spent Solvent (SS) – generated during the chemical cleaning process;
- Rinse Water (RW) - generated during the chemical cleaning process;
- Concentrate – generated from processing Rinse Water via Ultra Filtration / Reverse Osmosis (UF/RO) facility;
- Sludge- generated from the UF/RO permeate at a Sewage Processing Plant (SPP); and
- Liquid Effluent – generated from the UF/RO permeate at the SPP.

- Illustrative radionuclide concentrations of the waste streams are presented in Table I and II for the purposes of illustrative dose estimates presented below. These tables include two sets of concentrations based on sample radiological analyses representing dilution of the estimated radionuclides in minimum and maximum volumes (V_{min}, V_{max}.) of various waste streams (see Table III).

Table IIIThe analysis is required due to some uncertainties in the number of chemical steps, which will be performed during the cleaning, and which will impact the total volume of waste generated.

The total radionuclide activities in these waste streams ecxeed exemption activities (e.g., IAEA [6]) for many of the radionuclides. Therefore, the release of these wastes from PNGS requires a pathways analysis to demonstrate that the associated dose to all potential receptors is below *de minimis* dose.

Table I. Radionuclide Concentrations in Maximum Volume of Waste Streams (µCi/Cm^3)

Nuclide	SS	RW	Concentrate	Permeate	Liquid Effluent	Sludge
H-3	1.80E-03 [a]	1.80E-03 [a]	1.80E-03 [a]	1.80E-03 [a]	2.34E-04	1.65E-05
Mn-54	5.20E-09	2.17E-11	2.17E-10	4.82E-13	3.14E-14	1.10E-12
Fe-55	9.64E-06	4.02E-08	4.02E-07	8.92E-10	5.82E-11	2.04E-09
Co-60	4.10E-06	1.71E-08	1.71E-07	3.79E-10	2.47E-11	8.67E-10
Zn-65	2.23E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Sr-90	1.84E-06	1.89E-07	1.89E-06	4.19E-09	2.73E-10	9.58E-09
Nb-94	4.31E-08	1.80E-10	1.80E-09	3.99E-12	2.60E-13	9.12E-12
Ru-106	1.32E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Sb-125	1.86E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Cs-134	4.34E-09	5.88E-11	5.88E-10	1.31E-12	8.52E-14	2.99E-12
Cs-137	1.49E-07	2.02E-09	2.02E-08	4.49E-11	2.93E-12	1.03E-10
Ce-144	1.05E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Eu-152	9.57E-08	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Eu-154	9.16E-08	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Pu-238	1.13E-07	1.16E-08	1.16E-07	2.58E-10	1.68E-11	5.90E-10
Pu-239	4.42E-07	4.54E-08	4.54E-07	1.01E-09	6.58E-11	2.30E-09
Am-241	3.60E-07	3.69E-08	3.69E-07	8.20E-10	5.35E-11	1.87E-09
Cm-242	2.56E-10	2.62E-11	2.62E-10	5.82E-13	3.80E-14	1.33E-12
Cm-244	3.83E-08	3.93E-09	3.93E-08	8.72E-11	5.69E-12	1.99E-10

Table II. Radionuclide Concentrations in Minimum Volume of Waste Streams (µCi/Cm^3)

Nuclide	**SS**	**RW**	**Concentrate**	**Permeate**	**Liquid Effluent**	**Sludge**
H-3	1.80E-03 [a]	1.80E-03 [a]	1.80E-03 [a]	1.80E-03 [a]	1.05E-04	7.39E-06
Mn-54	7.28E-09	4.83E-11	4.83E-10	1.07E-12	3.14E-14	1.10E-12
Fe-55	1.35E-05	8.95E-08	8.95E-07	1.99E-09	5.82E-11	2.04E-09
Co-60	5.73E-06	3.80E-08	3.80E-07	8.45E-10	2.47E-11	8.67E-10
Zn-65	3.12E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Sr-90	2.57E-06	4.20E-07	4.20E-06	9.33E-09	2.73E-10	9.58E-09
Nb-94	6.03E-08	4.00E-10	4.00E-09	8.89E-12	2.60E-13	9.12E-12
Ru-106	1.84E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Sb-125	2.60E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Cs-134	6.07E-09	1.31E-10	1.31E-09	2.91E-12	8.52E-14	2.99E-12
Cs-137	2.09E-07	4.51E-09	4.51E-08	1.00E-10	2.93E-12	1.03E-10
Ce-144	1.47E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Eu-152	1.34E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Eu-154	1.28E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Pu-238	1.59E-07	2.59E-08	2.59E-07	5.75E-10	1.68E-11	5.90E-10
Pu-239	6.19E-07	1.01E-07	1.01E-06	2.25E-09	6.58E-11	2.30E-09
Am-241	5.04E-07	8.22E-08	8.22E-07	1.83E-09	5.35E-11	1.87E-09
Cm-242	3.58E-10	5.84E-11	5.84E-10	1.30E-12	3.80E-14	1.33E-12
Cm-244	5.36E-08	8.75E-09	8.75E-08	1.94E-10	5.69E-12	1.99E-10

[a] assumed at 95% of the *Packaging and Transportation of Nuclear Substances Regulation*'s H-3 limit, which is 70 Bq/g (1.89E-03 µCi/cm^3 for a density of 1 g/cm^3). Therefore, concentration does not depend on volume.

OPTIONS FOR MANAGING THE BCC SOLUTIONS

- Several options were considered for handling BCC solutions generated from an assumed two boiler chemical cleanings per year (see

Table III):

- Direct shipment of Spent Solvent and Rinse Water to an incineration facility for processing and subsequent landfill disposal.
- Shipment of Spent Solvent to an incineration facility for processing and subsequent landfill disposal. Shipment of Rinse Water for treatment by Ultra Filtration / Reverse Osmosis/ (UF/RO). The concentrate from the UF/RO will be shipped to the incineration facility whereas the permeate will be processed in a Sewage Treatment Plant (SPP). The liquid waste from the SPP will be released to an outfall and the sludge will be shipped to another landfill.
- Shipment of Spent Solvent and Rinse Water to a storage facility. Shipment from this facility to an incineration facility for processing and subsequent landfill disposal.
- Shipment of Spent Solvent to a storage facility and then to an incineration facility for processing and subsequent landfill disposal; Shipment of Rinse Water to treatment by Ultra Filtration / Reverse Osmosis (UF/RO). The concentrate from the UF/RO will be shipped to the landfill whereas the permeate will be processed in a Sewage Processing Plant (SPP). The liquid waste from this process will be released to an outfall and the sludge will be shipped to another landfill.

Table III. Definition of Hypothetical Options for Dose Calculation

Option #	Case	Transport Route	Type of Vehicle /Assumed # of Drivers	Distance (km)	Material Transported	Total Volume (m^3/yr)		# of Shipments	
						Max	Min	V_{max}	V_{min}
S1	C1	PN to Landfill 1	Tanker/5	323	Spent Solvent	1400	1000	56	40
	C2				Rinse Water	2540	1140	102	46
S2	C1	PN to Landfill 1	Tanker/5	323	Spent Solvent	1400	1000	56	40
	C2	PN to RO/UF	Tanker/5	290	Rinse Water	2540	1140	102	46
	C3	RO/UF to Landfill 1	Tanker/1	205	Concentrate	254	114	10	5
	C4	SPP to Landfill 2	Roll-off-on truck/1	2	Sludge	500	500	22	22
S3	C1	PN to RO/UF	Tanker/5	290	Spent Solvent	1400	1000	56	40
	C1	RO/UF to Landfill 1	Tanker/1	205	Spent Solvent	2540	1000	102	40
	C2	PN to RO/UF	Tanker/5	290	Rinse Water	1400	1140	56	46
	C2	RO/UF to Landfill 1	Tanker/1	205	Rinse Water	2540	1140	102	46
S4	C1	PN to RO/UF	Tanker/5	290	Spent Solvent	1400	1000	56	40
	C1	RO/UF to Landfill 1	Tanker/1	205	Spent Solvent	2540	1000	102	40
	C2	PN to RO/UF	Tanker/5	290	Rinse Water	1400	1140	56	46
	C3	RO/UF to Landfill 1	Tanker/1	205	Concentrate	254	114	10	5
	C4	SPP to Landfill 2	Roll-off-on truck/1	2	Sludge	500	500	22	22

DE MINIMIS DOSE CONCEPT

A *de minimis* dose or dose rate represents a level of risk, which is generally accepted as being of no significance. This level is derived from a small fraction of the annual dose due to natural background radiation, and thus represents an insignificant risk (AECB [7]). In Ontario, the individual dose from all natural sources is approximately 300 mrem (3 mSv) per year. The Atomic Energy Control Board (AECB) Regulatory Document, R-85, states that the AECB will use a *de minimis* dose criterion of 5 mrem (50 μSv) in a year to an individual provided that the potential for exposure to large populations is small (AECB[7]). Below this dose criterion, regulatory controls by the CNSC are not required.

Since the issuance of R-85, the Advisory Committee on Radiological Protection (ACRP), an advisory body to the CNSC, have recommended a *de minimis* individual dose rate of 1 mrem (10 μSv) per year (ACRP [8]). Their recommendation was based largely on that of the International Atomic Energy Agency (IAEA) regarding the principles to be used when exempting radiation sources and practices from regulatory control (IAEA [9]). The recommended annual individual dose takes into account the potential exposure from multiple sources.

The CNSC (in draft) uses a dose of 10 μSv/y for the disposal requirements of nuclear substances (CNSC [10]). It is interesting to note that the CNSC still considers a dose of 50 μSv/y to a member of the public to be a sufficiently low dose that below which an As Low As Reasonably Achievable (ALARA) assessment is not necessary (CNSC [11]).

For conservative purposes, this paper uses an individual dose rate of 1 mrem (10 μSv) per year as a *de minimis* level.

METHODOLOGY

This section describes the calculation of dose to hypothetical receptors from the transportation and disposal of BCC chemicals. The dose calculations were carried out using several models as follows:

- Receptors potentially impacted by the landfill disposal were assessed by IMPACTS-BRC (O’Neal and Lee[12]);
- The driver(s) transporting the wastes were assessed by MicroShield (Framatome [13]); and
- Receptors potentially impacted by release of liquid effluent from the SPP to the outfall were assessed analytically, using available Derived Release Limits (DRL) for the facility.

(i) IMPACTS-BRC and MicroShield

The IMPACTS-BRC, Version 2.1 computer model (O’Neal and Lee [12]) was used to estimate the dose to several hypothetical receptors. This code is a generic, radiological assessment code that was developed for use by the U.S. NRC to assist in the classification of waste streams as Below Regulatory Concern (BRC). The IMPACTS-BRC model estimates annual radiological doses to maximally exposed, hypothetical receptors as a result of transportation, treatment and disposal of wastes.

Dose coefficients for effective dose from inhalation and ingestion of radionuclides used in the IMPACTS-BRC model are based on tissue weighting factors recommended in ICRP 26 [14]. The effective dose coefficients were updated in this study to reflect ICRP72 [15] recommendations.

The IMPACTS-BRC model was used to estimate doses associated with disposal of BCC chemicals. However, the IMPACTS-BRC model does not provide much flexibility in terms of defining potential receptors or specifying the exposure scenarios. In particular, the transportation model in IMPACTS-BRC has pre-defined transportation parameters. For example, the truck capacity is fixed at 5 tonnes and the drivers are exposed to hazardous wastes for a fixed period of time regardless of the distance from the site to the landfill. Therefore, the driver dose was calculated using MicroShield v 6 (Framatome [13]).

(ii) Receptors

Several hypothetical receptors were defined in order to perform the pathways analysis. The selected receptors were expected to represent maximally exposed individuals (see Table IV).

(iii) Release of SPP Effluent to the Outfall

For the SPP discharges, the waterborne DRLs for the facility (Benovich[16]) can serve as a conservative surrogate for estimating a hypothetical resident dose from the release of SPP effluent to the outfall. DRLs represent the activity which will lead to an exposure dose of 1 mSv/y of each radionuclide to a receptor. The doses for the SPP discharge were estimated using proportionality with the DRLs.

The exposure groups (relatively homogeneous groups of members of the public, who represent the most highly exposed to the releases of radionuclides from a facility) used in the DRL calculation included a dairy farm, lakeshore homes, inland residences, a trailer park and an industrial site near the facility.

Table IV. Hypothetical Receptors for Pathways Analysis

Exposure Mode	Hypothetical Receptor	Description
Transport	Truck Driver	Driver of truck taking spent solvent, rinse water concentrate or sludge exposed to external gamma from radionuclides in the shipment
Landfill	Intruder (Construction)	An individual who intrudes into the facility after closure and constructs a house
	Intruder (Agricultural)	An individual who intrudes into the facility after closure, living in a house, consuming food grown on-site and drinking from an on-site well
	Resident (Intruder-initiated release to air)	A nearby resident who is exposed to air-borne dispersion of radionuclides as a result of intruder activity
	Resident (Natural erosion release to air)	A nearby resident who is exposed to air-borne dispersion of radionuclides as a result of natural erosion
	Resident (Intruder-initiated release to water)	A nearby resident who is exposed to surface water containing site run-off as a result of intruder activity
	Resident (Natural erosion-initiated release to water)	A nearby resident who is exposed to surface water containing site run-off as a result of natural erosion
	Resident (Intruder-initiated release to air)	A nearby resident who is exposed to air-borne radionuclides as a result of intruder activity
	Resident (Natural erosion-initiated release to air)	A nearby resident who is exposed to air-borne radionuclides as a result of natural erosion
	Resident (Leachate treatment)	A nearby resident who is exposed to surface water containing treated leachate from the landfill during operation
	Resident (Leachate overflow)	A nearby resident who is exposed to surface water containing leachate that overflows from the landfill after closure
	Resident (Evaporation)	A nearby resident who is exposed to air-borne radionuclides from evaporation
	Intruder (Well)	An individual who intrudes into the facility after closure drinking from an on-site well
	Resident (Well)	A nearby resident who drinks from a well containing leachate-contaminated groundwater
	Resident (Surface water)	A nearby resident who drinks surface water containing leachate-contaminated groundwater
	Worker (Residue Handler)	A worker at the landfill/ incinerator handling ash/ residue

RESULTS

The dose calculations are discussed in detail by Garisto and Eslami. The results of dose calculations are summarized in Table V and Table VI. Table V presents the driver dose calculations using MicroShield while Table VI presents the dose to the most exposed receptors for each waste management option (as defined in

Table III). Different receptors receive the highest dose in different waste management options. The driver is not the limiting receptor in any of the cases analyzed in this study.

The results show that for the illustrative example waste streams and waste management options, all doses are less than *de minimis*. It is interesting to note that minor differences were observed between the V_{max} and V_{min} cases, i.e., the main factor affecting dose is the total activity per cleaning shipped offsite.

Table V. Individual Driver Doses for Four Shipment Options (Calculated by MicroShield for both Minimum and Maximum Volumes of Waste Streams)

Option #	Case	Transport Route	Material Transported	Dose to Driver µSv/y	
				1 Driver per Route	5 Drivers per Route
S1	C1	PN to Landfill 1	Spent Solvent	-	1.2
	C2		Rinse Water	-	0.009
S2	C1	PN to Landfill 1	Spent Solvent	-	1.2
	C2	PN to RO/UF	Rinse Water	-	0.008
	C3	RO/UF to Landfill 1	Concentrate	0.03	-
	C4	SPP to Landfill 2	Sludge	7.6E-05	-
S3	C1	PN to RO/UF	Spent Solvent	-	1.1
		RO/UF to Landfill 1	Spent Solvent	4.0	-
	C2	PN to RO/UF	Rinse Water	-	0.008
		RO/UF to Landfill 1	Rinse Water	0.03	-
S4	C1	PN to RO/UF	Spent Solvent	-	1.1
		RO/UF to Landfill 1	Spent Solvent	4.0	-
	C2	PN to RO/UF	Rinse Water	-	0.008
	C3	RO/UF to Landfill 1	Concentrate	0.03	-
	C4	SPP to Landfill 2	Sludge	7.6E-05	-

Table VI. Highest Exposure Doses and the Respective Receptors

Option /Case	Receptor	Main Contributor to Dose	Highest Total Dose (μSv/yr)	
			V_{max}	V_{min}
Option S1, Case C1	Residue Handler/ Maintenance Worker	Co-60	1.83E+00	1.82E+00
Option S1, Case C2	Leachate Treatment	H-3	4.00E-01	1.80E-01
Option S1, Overall (Cases C1 & C2)	Residue Handler/ Maintenance Worker	Co-60	1.85E+00	1.84E+00
Option S2, Case C1	Residue Handler/ Maintenance Worker	Co-60	1.83E+00	1.82E+00
Option S2, Case C2	Transport	Co-60	7.62E-03	7.62E-03
Option S2, Case C3	Erosion Air	Pu-239	1.76E-01	1.76E-01
Option S2, Case C4	Intruder [a]	H-3, Pu-239 [c]	1.13E+00	6.30E-01
Option S2, Case C5	3-mon Nursing Infant	H-3	<7.51E-02	<7.51E-02
Option S2, Overall (Cases C1 to C4)	Residue Handler/ Maintenance Worker	Co-60	1.85E+00	1.84E+00
Option S3, Case C1	Transport	Co-60	4.00E+00	4.00E+00
Option S3, Case C2	Leachate Treatment [b]	H-3	4.00E-01	1.80E-01
Option S3, Overall (Cases C1 & C2)	Transport	Co-60	4.03E+00	4.03E+00
Option S4, Case C1	Transport	Co-60	4.00E+00	4.00E+00
Option S4, Case C2	Transport	Co-60	7.62E-03	7.62E-03
Option S4, Case C3	Erosion Air	Pu-239	1.76E-01	1.76E-01
Option S4, Case C4	Intruder	H-3, Pu-239	1.13E+00	6.30E-01
Option S4, Case C5	3-mon Nursing Infant	H-3	<7.51E-02	<7.51E-02
Option S4, Overall (Cases C1 to C4)	Transport	Co-60	4.03E+00	4.03E+00

a) Intruder- Agriculture for the case of Vmax and Intruder-Air for the case of V_{min}
b) Leachate Treatment is only relevant in facilities which plan post-closure overflow treatment.
c) H-3 for the case of V_{max} and Pu-239 for the case of V_{min}

CONCLUSION

A pathway analysis was carried out to estimate radioactive dose impacts for the transportation and disposal of BCC solutions from the Pickering Nuclear site. The methodology was based on the application of three models: IMPACTS BRC, MICROSHIELD and a DRL model. Regarding methodological aspects of this study, the analysis has shown that (i) Dose Conversion Factors (DCFs) in IMPACTS BRC can be readily updated to reflect current values, and (ii) the use of MICROSHIELD allows for site-specific driver exposure calculations whereas IMPACTS BRC uses default transportation parameters which may not reflect site-specific situations.

Several sample calculations are presented in this paper. These calculations show that for the particular waste streams considered in the calculations, the dose to all hypothetical receptors is less than *de minimis.* These results imply that *for the options considered in this study*, these particular waste streams can be handled as non-radioactive materials.

The most exposed receptors are different for different waste management options, as shown in Table VI. The most exposed receptor overall is the residue handler at the landfill/ incinerator facility. The main radionuclide contributing to his/her dose is Co-60. The dose to this receptor depends on the total activity shipped (i.e., concentration x volume).

A similar dependence is also observed for other receptors, where an increase in volume is cancelled out by a corresponding decrease in concentration (see Table VI). In the case of post-closure receptors (e.g., from a future overflow treatment or inadvertent intrusion), the overall dose is also proportional to activity. However, in these cases, the main contributor to dose is H-3. The H-3 concentration is assumed to be the same regardless of dilution (see Table I and Table II).

The calculations presented in this paper will be used to derive maximum allowable concentrations and activities that can be shipped from the site and disposed of as non-radioactive materials (Garisto and Eslami).

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