CHARACTERIZATION OF THE RADIOCHEMICAL ACTIVITY IN CANDU® STEAM GENERATORS

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ABSTRACT

The scope of mid-life refurbishment activities at some CANDU® plants includes replacement of the existing steam generators[1]. Shipment of the discarded steam generators for interim storage, intact disposal or for recycling via metal melting requires an assessment of the inventory of radionuclides within the components and also dose rates.

Kinectrics was contracted by Bruce Power to develop radionuclide inventory and dose rate data for steam generators from its four-unit Station A located in Kincardine, Ontario, Canada. Units 1 & 2 are currently being refurbished while Units 3 & 4 are planned for refurbishment in 2016. The scope of the refurbishment activities for all units includes replacement of the existing steam generators. Those from Units 1 & 2 have already been placed into interim storage at the Western Waste Management Facility (WWMF) which is also located on site but operated by Ontario Power Generation.

This paper presents the detailed methodology employed to develop radionuclide inventory and dose rate data for radionuclides within (characterize the steam generators that are) both in-service and out-of-service (in-storage) steam generators. The data were developed as follows: a) archived tube sections from various steam generators were characterized and scaling factors for Difficult-to-Measure radionuclides were derived from the measured data, b) in-situ gamma spectrometry (using germanium and cadmium zinc telluride detectors) and dose rate surveys were performed at various steam generators and c) a detailed assessment of the tritium inventory in various primary and secondary side components was performed.

1. INTRODUCTION AND BACKGROUND

Worldwide a number of CANDU® utilities are planning or undertaking reactor refurbishment activities for the purpose of extending plant life. Typically, refurbishment entails the replacement of fuel channel components consisting of pressure tubes, calandria tubes and end fittings. At specific plants, the scope of the refurbishment activities may also include replacement of carbon steel feeders/headers and also steam generators.

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[1] CANDU® is a registered trade mark of Atomic Energy of Canada Limited.

While the replaced fuel channel components are significantly activated and must be stored as radioactive waste, components such as steam generator shells can be readily melted down for metal recycling. The tube bundle, which contains most of the activity, is typically separated from the shell, segmented into small sections and disposed of as radioactive waste. To limit dose uptake, the tube bundle may be mechanically cleaned prior to dismemberment. Services for disposing and/or processing of intact steam generators are offered by vendors internationally.

Assessment of detailed radionuclide inventories and dose rates is a pre-requisite for interim storage, intact disposal or recycling of discarded steam generators. Radionuclides of particular interest include the long-lived gamma emitters such as Co 60, Cs-137, Nb-94 and Eu isotopes, the alpha emitters Pu-238, Pu-239, Pu-240, Am-241and Cm-244, the beta emitters H-3, C-14, Ni-63, Sr-90, Pu-241, Tc-99and I-129, and the X-ray emitter Fe-55. These radionuclides are principally confined to the oxide layer present on tube internals as well as other primary side surfaces, namely, bowl, tube sheet and divider plate, which during operation are exposed to the contaminated heavy water coolant (the secondary side of the tubes are exposed to light water).

Kinectrics was contracted by Bruce Power to develop radionuclide inventory and dose rate data for the steam generators associated with its four unit Station A located in Kincardine, Ontario. Units 1 & 2 are currently being refurbished[2] while Units 3 & 4 are planned for refurbishment in 2016. The scope of the refurbishment activities at all the four units includes replacement of the existing steam generators. Those from Units 1 & 2 have already been placed in interim storage at Ontario Power Generation's Western Waste Management Facility (WWMF) which is also located on the same site but operated by Ontario Power Generation. Data for both stored and in-service steam generators were developed as follows:

- Archived tube sections, previously removed from various steam generators, were radiochemically characterized. Scaling factors relative to Easy-to-Measure gamma emitters were derived from the data for the Difficult-to-Measure (DTM) alpha and beta activity data and applied to determine the radionuclide inventory of the entire steam generator.

- In-situ gamma spectrometry and dose rate surveys were performed on various steam generators. Gamma spectrometry was performed on the steam generators in interim storage as well as those still in service. Measurements were performed using both a hyperpure germanium detector as well as a miniature cadmium zinc telluride (CZT) detector; the latter was inserted into a tube through the tube sheet during an outage.

- A detailed assessment of tritium in both the primary and secondary side components was performed to obtain the overall tritium inventory. This was based on a combination of approaches, including estimations based on permeability of tritium through metals and measurement of tritium activity in the tube oxide.

[2] Unit 1 and 2 were shutdown in October 1997 and October 1995, respectively.

- Measured tube gamma activity data corresponding to different operating periods were utilized to estimate the future gamma activities of Units 3 & 4 steam generators which are still in service. The estimated gamma activities were used to calculate dose rates in contact with fully drained, bare (insulation stripped) steam generators at shutdown.

This paper presents an account of the various methodologies employed to determine the radiochemical inventory and contact dose rates for all Station A steam generators.

2. CHARACTERIZATION OF ARCHIVED TUBE SECTIONS

Typically, oxide deposits and associated radiochemical activity within CANDU® steam generators vary significantly along the tube length with deposits on the cold leg side being generally more copious [1]. Although a number of hot leg sections were readily available, availability of archived cold leg side tube sections was somewhat limited. The available tube sections were part of longer sections which had been removed from a region between the tube sheet and the bottom support plate.

Tube activity is associated principally with the oxide layer, with much lower levels being present in the base metal. The internals of tube sections selected for characterization were exposed to fresh concentrated acid in two steps to dissolve the oxide and associated activity: most of the oxide was dissolved in the first step while the remaining activity was dissolved in the second step. The weight loss in the first step provided a measure of the tube oxide loading (the contribution of base metal dissolution to the weight loss was judged to be relatively small). The acids from both steps were consolidated to determine the activity loading on the tube surfaces.

Solutions containing the dissolved activity were analyzed for various alpha, beta and gamma emitting radionuclides. A separate procedure was, however, required to measure the C-14 and tritium present: this is because C-14 is not retained in acidic solutions (it being evolved in the form of CO_2 gas) and the dissolved tritium would exclude that present in the base metal. These radionuclides were, therefore, measured by exposing selected tube sections to approximately 1000°C in a tube furnace; oxidized copper shavings were used to catalyze the oxidation of any C-14 present in non-CO_2 form and any tritium present in elemental form. The evolved C-14 and tritium were subsequently trapped for analysis. Measured radiochemical data were suitably decay-corrected.

Scaling factor data for the DTM radionuclides, namely, the plutonium isotopes, Am-241, Cm-244, Sr-90, Ni-63, Fe-55 and C-14, were developed from the measured data [2]. Mean values were estimated by combining the measured data for both the hot and cold leg sections. The scaling factors were subsequently utilized to estimate DTM radionuclide inventories for the entire steam generators. Levels of I-129 and Tc-99, which were too low to be measured reliably, were estimated from theoretically derived scaling factor values relative to Cs-137 and Co-60, respectively [3, 4].

3. CHARACTERIZATION OF GAMMA ACTIVITY IN STEAM GENERATORS

As stated earlier, in-situ gamma spectrometry and dose rate surveys were performed to characterize the gamma activity in various steam generators [5]. Those from Units 1 & 2 were surveyed inside the storage building at WWMF while those from the operating Units 3 & 4 were surveyed during an outage. Survey methodologies for both the in-storage as well as the in-service steam generators are described in this section.

3.1. In-storage steam generators

Figure 1 is a view of in-storage steam generators. The orientation of every second steam generator was reversed to attain greater storage efficiency. Because of their close proximity to each other, access to the steam generators was generally limited and only the extremity ones with unencumbered access were surveyed.

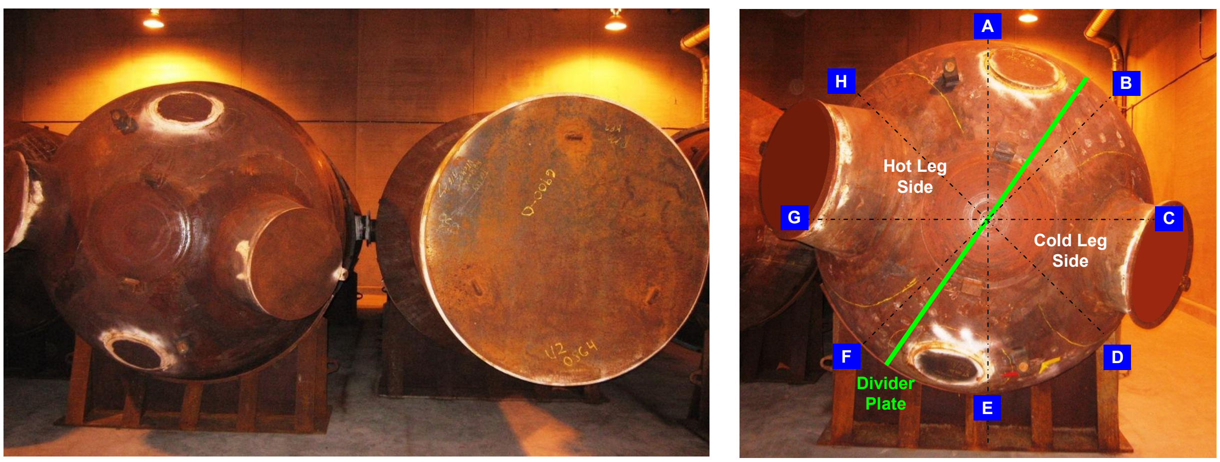

Figure 1. End view of an in-storage steam generator

Dose rate surveys were performed at several distances from the tube sheet. However, at each location, they were recorded only at Positions A-D because dose rates at Position E had a significant scatter contribution from the concrete floor while those at Positions F-H had a significant shine effect from the adjacent steam generators. The measured dose rate at Position A was considered to be representative of the hot leg while those at Positions B-D were averaged to obtain the representative value for the cold leg.

Contact dose rates in general were low (2 - 8 mrem/h) because of the previous shutdown history of Units 1 & 2. Figure 2 shows the cold leg to hot leg contact dose rate ratio measured for one steam generator as a function of distance from the tube sheet. It indicates the cold and hot leg dose rates to be generally similar, except near the tube sheet where the cold leg dose rates were up to a factor of 2 greater than the hot leg dose rates.

Gamma spectra were recorded using a germanium detector at a distance of approximately 1 m from the surface of the steam generators and at 2 m and 5 m from the tube sheet (along the length of the steam generator). Measurements, which could only be recorded on the cold leg side, indicated that Co-60 was the only radionuclide present. The data were interpreted using a shielding model for the steam generator as shown in Figure 3. The model considered the tube bundle to be a homogeneous source cylinder surrounded by layers of water, metal and insulation. Zone 1 represents the tube bundle with primary water within the tubes and secondary side water outside the tubes; shielding properties of the bundle, namely attenuation coefficient and buildup factor were calculated accordingly.

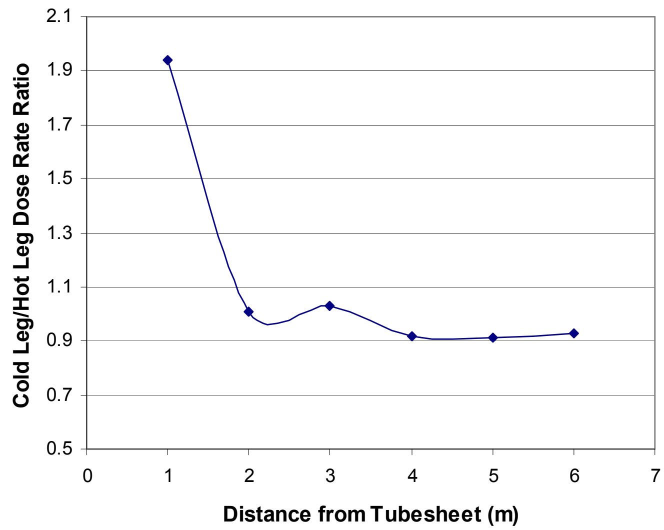

Figure 2. Variation in cold leg to hot leg dose rate ratio with distance from the tube sheet

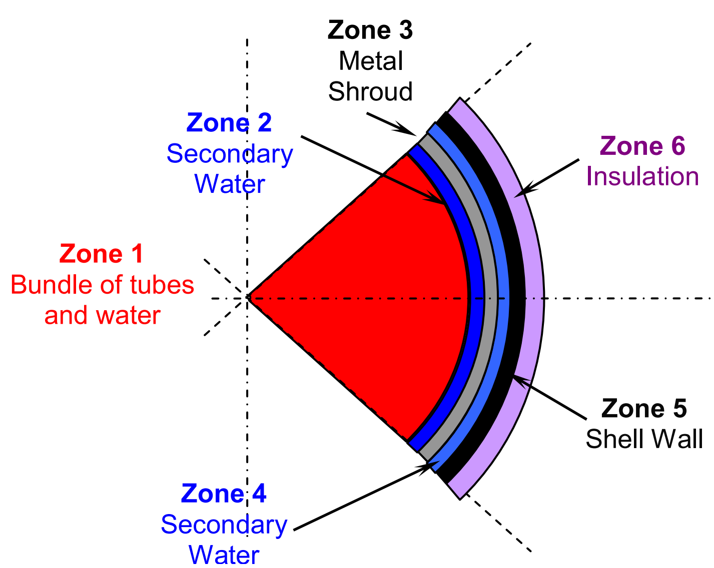

Figure 3. Shielding model for steam generator

Results based on the model interpretation were generally consistent with those obtained from the analysis of tube sections discussed earlier. The activities obtained for the cold leg were combined with the dose rate profile along the length of the steam generators to estimate the Co-60 activity as a function of tube length. The Co-60 inventory was subsequently estimated based on the total number of tubes in each steam generator.

3.2. In-Service Steam Generators

Selected in-service steam generators were gamma scanned (during an outage) at several elevations above the tube sheet using the germanium detector. Because the distance between neighboring steam generators is sufficiently large, both cold as well as hot leg side measurements were performed at each elevation. Complementary dose rate measurements were also recorded at each elevation at up to 8 circumferential locations around the periphery of the steam generators.

In addition to measurements using the germanium detector, a miniature CZT detector was also utilized to scan one steam generator. It was inserted through the tube sheet into a central tube within the tube bundle, thus permitting gamma spectra to be recorded as a function of distance along the tube length [1]. The CZT technique was chosen because of the relative ease with which radionuclide activity can be profiled along the tube length in a steam generator. On the other hand, the bulkier germanium detector system can only be used at a limited number of access levels to the steam generators.

Figure 4 shows the simple equipment used to insert the CZT probe through the man-way into a selected steam generator tube. Spectra were successfully recorded at several hot and cold leg locations along the length of the selected tube.

(a) (b) (c)

Figure 4. CZT detector system showing (a) pole mounted delivery system inserted through man-way of mock-up (b) couplant used to engage with the tube sheet and (c) probe inserted through couplant into tube sheet

Data obtained using the germanium detector indicated the presence of Co-60 and several short-lived radionuclides. Depending on the radionuclide, cold leg activities (data were interpreted using the shielding model) were up to a factor of 3 higher than hot leg activities. In contrast with the spectra recorded using the germanium detector, those obtained using the CZT detector showed the presence of only Co-60 and Zr-95/Nb-95, with the latter being somewhat poorly defined. Data recorded using the CZT detector were interpreted considering the detector to be surrounded by a uniformly distributed source cylinder.

Figure 5 shows the Co-60 activity data obtained using both the germanium and CZT detectors. Clearly, both techniques yielded consistent results with the Co-60 activity decreasing systematically from the cold leg to the hot leg. The decrease over a tube length of 1260 cm was a factor of approximately 2.7.

Compared with the activities determined for the removed tube sections, the in-situ activity results were significantly greater. This is because of the additional activity deposition which would have occurred within the operating steam generators since the tube sections were removed. This is discussed further in Section 4.

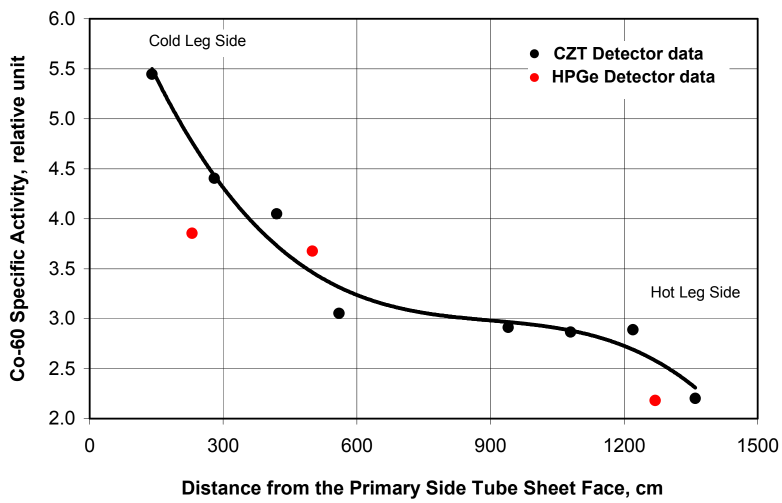

Figure 5. Variation in measured Co-60 activity with tube length for an in-service steam generator

4. ASSESSMENT OF ACTIVITY INCREASE WITH OPERATING TIME

It was important to estimate the final shutdown radionuclide activities of in-service steam generators based on their present values. In this respect, Co-60 is of particular interest since it is the dominant contributor to the overall activity and dose rate. Considering that the increase in tube activity with time from additional deposition is offset by a decrease in activity from radioactive decay, the total Co-60 activity at shutdown can be expressed as

$$A(t_2) = A_{initial}(t_1) \cdot \exp(-\lambda \cdot (t_2 - t_1)) + A_{deposited}(t_2) \tag{1}$$

where, the first term represents the contribution of the present or initial activity $A_{initial}(t_1)$ decay-corrected to the final shutdown date and the second term represents the decay-corrected contribution of the additional activity which will deposit between now and final shutdown. $A_{deposited}$ is determined using the following equation:

$$dN = R \cdot dt - \lambda \cdot N \cdot dt \tag{2}$$

where, *R* denotes the deposition rate (atoms/s) and *N* the atoms of Co-60 deposited. Integration yields, after re-arrangement,

$$A_{deposited}(t_2) = R \cdot (1 - \exp(-\lambda \cdot (t_2 - t_1))) \tag{3}$$

Application of Equations (1) - (3) using appropriate hot leg tube activity data indicated that the average value of *R* for the hot leg side of in-service steam generators to be 6E+04 atoms/s/cm. The corresponding value for the cold leg could not be estimated because of unavailability of suitable cold leg tube activity data. Therefore, a conservative value of *R* for the cold leg, which was a factor of 3higher than that for the hot leg, was assumed, consistent with the greater magnitude of cold leg deposits. The *R* values were then used to obtain the Co-60 activity at final shutdown.

5. PROJECTIONS OF MEASURED DOSE RATES TO SHUTDOWN DATES

Limits imposed by transportation and disposal requirements necessitated an estimation of the maximum dose rates that would exist for the in-service steam generators at final shutdown. The dose rate values of interest are those in contact with the bare surfaces (after insulation has been removed) of fully drained steam generators. On the other hand, measured dose rates were obtained in contact with the surface of the insulation, with the steam generators being drained on the primary side but full on the secondary side. The following considerations were involved in estimating the expected maximum dose rates:

- Based on Figure 5, activity and hence dose rate is expected to increase as the tube sheet is approached on the cold leg side of the steam generator. The resulting increase, however, would be offset by an increased shielding effect in the region close to the tube sheet. Based on this, the maximum dose rate was considered to occur at a distance of approximately 0.5 m above the secondary side tube sheet face or equivalently at 0.9 m above the primary side tube sheet face.

- Based on the measured value @ 1.4 m above the primary tube sheet face on March 25, 2008, the corresponding value @ 0.9 m above the primary face was first estimated. This was then extrapolated to the final shutdown date based on Equations (1) and (3) using an appropriate value for the Co-60 cold leg deposition rate.

- Based on the model shown in Figure 3, MicroShield was finally used to estimate the expected dose rate at shutdown considering the steam generators to be fully drained and bare (non-insulated).

The maximum contact dose rate at shutdown was thus estimated to be approximately 74 mrem/h, which is significantly greater than the 2008 measured value but much lower than the transportation limit of 200 mrem/h. The increase in dose rate from the 2008 measured value was because of (a) the projected growth in activity between 2008 and final shutdown (b) differences in the applicable geometry (presence/absence of insulation) and (c) differences in the dose point.

6. ESTIMATION OF OVERALL TRITIUM INVENTORIES

Besides being present on internal tube surfaces, tritium is also associated with various other primary and secondary side surfaces of the steam generators. A systematic assessment of the various contributions was performed using typical tritium levels in the Primary Heat Transport System (about 3.7E+10 Bq/kg) and in the secondary water (about 3.7E+04 Bq/kg). Several specific contributions to the primary and secondary side tritium inventories were assessed:

6.1. Tritium associated with primary side surfaces

<u>Tritium associated with tube oxide and tube base metal:</u> Based on data for steam generator tube sections, a) the average tritium concentration in removed cold leg tube sections was a factor of 6 higher than in the hot leg tube sections, and b) most of the tritium was bound within the tube oxide layer. The maximum amount of tritium in the base metal was estimated from equilibrium solubility considerations to be about 64 Bq/cm.

<u>Tritium associated with bowl, divider plate and tube sheet surfaces:</u> During operation, tritium is expected to also permeate into the primary side, oxide covered surfaces of the carbon steel bowl, divider plate and the Inconel-600 clad carbon steel tube sheet. The oxide contribution was obtained by scaling up the measured activity data (Bq/cm^2) for tritium in a carbon steel feeder sample. Inventory estimates for tritium in the base metal of each component were obtained theoretically from solubility considerations.

<u>Tritium associated with wet primary side tube surfaces:</u> A wet (non-dried) steam generator will contain an additional inventory of tritium associated with the film of primary heat transport water remaining on the tube surfaces. Tests indicated that upon prolonged standing, a wet tube retains approximately 3.0 10^{-3} g of moisture per cm length. The additional contribution of this wet layer to the tritium inventory was estimated using the activity concentration of tritium in the primary heat transport water.

<u>Tritium associated with ingress of primary water into intact plugged tubes:</u> Some of the tubes in each steam generator may have been plugged because of wall thinning without

necessarily having through-wall defects. Such tubes could fill up and trap primary water if the plugs are defective. The additional contribution to the tritium inventory from this was estimated from the tube hold-up (about 1.35 L per plugged tube) and the activity of tritium in the primary heat transport water.

6.2. Tritium associated with secondary side surfaces

Permeation of tritium across the tube surfaces and the tube sheet typically results in a steady state tritium concentration of about 3.7E+04 Bq/kg in the secondary water. The inventory of tritium on the secondary side was estimated using this concentration and the associated amount of secondary side water.

- Tritium associated with external shell surfaces: Based on the low secondary side tritium concentration, the amount of tritium which permeates into the shell wall is expected to be negligible. Hence, the inventory of tritium in the steam generator shell is expected to be insignificant.

- Tritium associated with water on the tube sheet: This was estimated based on tritium concentration in the secondary water and the quantity of secondary side water which remains after draining; the latter was estimated to be approximately 76 kg.

- Tritium associated with water within plugged defected tubes: Plugged tubes with through-wall defects may contain varying quantities of secondary side water depending on the location of the defect. For a conservative estimate, a) 50% of the plugged tubes were considered to have cracks just above the tube sheet and thus would contain negligible quantities of water, b) 25% of the tubes were considered to have a crack in the straight leg - one straight length of each such tube was assumed to be water filled, and c) the remaining 25% of the tubes were considered to have a crack in the U-bend - the entire length of each such tube was assumed to be water filled. The total hold-up of secondary water in the plugged tubes was thus estimated to be 490 L.

- Tritium associated with water in the sludge pile: Water present within the pores of the tube sheet sludge pile will likely be retained when the secondary side is drained. Based on the known geometry of the sludge pile within in-service steam generators, the pore volume associated with the sludge pile was estimated to be 65 L.

6.3. Tritium inventory - summary of overall findings

As expected, the primary side tritium inventory was estimated to be orders of magnitude higher than the tritium inventory on the drained secondary side. Most of this is associated with the primary side oxide layer on the tube surfaces with relatively small levels present in the base metal. Drying the steam generator would reduce the primary side tritium inventory by a factor of about 25.

7. CONCLUSIONS

This paper presents a comprehensive approach for the development of radionuclide inventory and dose rate data for steam generators planned to be discarded and disposed/stored. Radiochemical data were developed both from analysis of archived tube sections and using in-situ gamma spectrometry techniques. The in-situ gamma spectrometry data yielded activity data as a function of tube length and hence provided a more accurate assessment of the overall inventory of gamma emitters than is obtained solely based on the analysis of archived tube sections (these represent activities in the tube sheet region only). Because of significant variation in activity along the length of the tubes, it is preferable to derive scaling factors for DTM radionuclides from the analytical data for the archived tube sections and then apply them along with the total estimated inventory of Co-60 to determine the overall inventory of DTM radionuclides in the steam generators.

Availability of tube activity data (from tube sections or from in-situ measurements on the steam generators) corresponding to varying periods of operation permitted an estimation of the tube activity at a future shutdown date. This activity was then used to estimate future dose rates for in-service steam generators.

Detailed assessment of the tritium inventory indicated that most of the tritium is associated with the primary side oxide surfaces (tube, tubesheet, bowl and divider plate) with relatively small levels present in the base metal and in the residual secondary side water of the drained steam generator.

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