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Supplementary Information

Written submission from Frank R. Greening

In the Matter of

Ontario Power Generation Inc., Pickering Nuclear Generating Station

Request for a ten-year renewal of its Nuclear Power Reactor Operating Licence for the Pickering Nuclear Generating Station

Commission Public Hearing – Part 2

June 2018

Renseignements supplémentaires

Mémoire de Frank R. Greening

À l'égard de

Ontario Power Generation Inc., centrale nucléaire de Pickering

Demande de renouvellement, pour une période de dix ans, de son permis d'exploitation d'un réacteur nucléaire de puissance à la centrale nucléaire de Pickering

Audience publique de la Commission – Partie 2

Juin 2018

Canada

From:	Frank Greening
Sent:	Tuesday, June 05, 2018 9:22 AM
To:	Interventions (CNSC/CCSN); Levert, Louise (CNSC/CCSN)
Cc:	gerry.frappier@canad.ca; Elder, Peter (CNSC/CCSN); Mendoza, Melissa (CNSC/CCSN); McDougall, Glen (CNSC/CCSN); Jin, John (CNSC/CCSN); Lojk, Robert (CNSC/CCSN); Rinker, Michael (CNSC/CCSN); Torrie, Brian (CNSC/CCSN); Sauvé, Kiza (CNSC/CCSN)
Subject:	Additional Supplementary Submission for 2018-H-03
Attachments:	PINT2018B.docx

To: The Senior Tribunal Officer
Secretariat of the Canadian Nuclear Safety Commission (CNSC)
280 Slater Street, P.O. Box 1046, Station B Ottawa, ON K1P 5S9

Tel: 1-800-668-5284 or 1-613-996-9063 Fax: 613-995-5086
Email: Interventions@cnsc-ccsn.gc.ca

Re: Supplemental material for my submission to the CNSC Public Hearing to consider Ontario Power Generation's (OPG's)
application to renew its Nuclear Power Operating Licence for the Pickering Nuclear Generating Stations, A and B, for a period of 10 years, Hearing Number Ref. 2018-H-03

To whom it may concern:

In my original submission, dated May 4th 2018, for the upcoming Pickering Hearings, I included the following data request:

1. All available fitness-for-service inspection data, (as required by CSA N285.4 and N285.8), for pressure tubes at Pickering A and Pickering B over the past 5 years (2013 – 2017). This should, as a minimum, include:

(i) Hydrogen/Deuterium data derived from concentration measurements of scrapes taken at different axial locations and after different Effective Full Power Hours (EFPH) or Hot Hours (HH) of Unit Operation.

(ii) All available pressure tube oxide thickness data measured on removed pressure tubes from Pickering A and Pickering B.

(iii) Hydrogen pickup data at the rolled joints of Pickering pressure tubes.

I requested this data in light of the fact that OPG's Licence renewal request is to operate Pickering B to December 2024, which requires exposing the fuel channels in Units 5 – 8 to in excess of 274,000 Effective Full Power Hours. This represents uncharted territory for any CANDU reactor worldwide, and is accompanied by great uncertainty and risk in the fitness-for-service of over 1500 pressure tubes, as required by CSA N285.4 and CSA N285.8.

I therefore now wish to provide supplementary material to support my claim that CSA N.285.4 and CSA N285.8 may not be satisfied in the run-up to 2024. Thus, please find as an attachment to this email, a WORD file (PINT2018B) that contains an analysis of deuterium pickup data for typical CANDU plant up to 2005, and explains the need for more recent data as I have requested.

Please include this email and its attachment as supplementary material to my original submission.

Thank you,

Sincerely,

Dr. F. R. Greening

CANDU Pressure Tube Corrosion and Hydrogen Isotope Pickup:

A design basis accident potentially resulting in major fuel damage and the release of fission products is a pressure tube rupture. In 1975 cracks were observed just inboard of the rolled joints in some Zr-2.5%Nb pressure tubes in Pickering NGS and it was subsequently recognized that deuterium picked up at a rolled joint will diffuse inboard into the body of the pressure tube. Once the terminal solute solubility (TSS) is exceeded, hydride precipitation occurs in high stress regions followed by delayed hydride cracking. For Pickering Units known to have sufficiently large/sharp flaws, thermal cycle restrictions must be imposed.

Pressure tube flaws such as scratches and dents in as-installed tubes, or debris and fuel bearing pad fretting marks, are important fuel channel aging issues. In order to prevent tube failures developing from these flaws, CSA N285.4 & N285.8 standards were issued in 2005 as the applicable codes for flaw fitness-for-service assessment of CANDU pressure tubes. When tensile stresses are applied to a crack tip the crack can grow when the hydrogen (or deuterium) concentration exceeds the so-called terminal solid solubility limit (TSS) for the precipitation of hydrides. For flaws with peak notch tip stresses greater than the threshold stress for delayed hydride cracking the flaw is restricted to a limited number of thermal (heat-up and cool-down) cycles.

Remarkably, even after more than 40 years of intensive research into CANDU pressure tube corrosion and hydrogen isotope pickup, the mechanisms by which H/D/T enter the wall of a zirconium alloy pressure tube are still poorly understood. Thus, consider the *Ontario Energy Board* hearing held on 17th Aug, 2010 (EB-2010-0008) and in particular the *PWU Interrogatory #014*. Here we find the following:

> *Until recently, Pickering B was not expected to exceed EOL limits during the pressure tube nominal operating life of 210k EFPH. This expectation was related to the lower operating temperatures in Pickering B. However, the hydrogen and deuterium profiles through the inlet and outlet rolled joint regions of surveillance tube P6 M14 have challenged this belief (report issued December 2008). It appears that P6 M14 has much higher deuterium uptake in the compressive regions of the pressure tube and H_{eq} exceeds the solubility limit at both inlet and outlet rolled joint burnish marks.*

From a purely chemical perspective, zirconium corrosion is a simple aqueous oxidation reaction:

$$Zr + 2D_2O = ZrO_2 + 2D_2$$

However, in a CANDU PHTS this reaction is complicated by the fact that some of the released deuterium enters the zirconium base metal where it forms an insoluble hydride/deuteride phase:

$$Zr + xD = ZrD_x$$

The amount of deuterium that is actually picked up by the base metal is indeterminate but theoretical limits may be calculated as follows:

For 100 % D Pickup, 1 mole of Zr picks up 2 moles of D_2 gas

Consider 1 gram of Zr = 1/91.2 moles of Zr =0.01086 moles

For 100 % pickup we have 2 × 0.01086 moles of D_2 = 4× 0.01086 moles of D atoms = 0.08688 grams.

Therefore, D concentration = 0.08688 grams of deuterium per gram of zirconium

But this is for the complete oxidation of 1 gram of zirconium

In fact, an oxide layer forms on the base metal that is x microns, (μm), thick

Pressure tube wall thickness = 0.406 cm

Density of Zr = 6.49 g/cm^3; Density of ZrO_2 = 5.83 g/cm^3

So, we have 0.406 cm^3 of zirconium with 1 ×10^{-4} cm^3 of oxide

Hence, we have 0.406 × 6.49 g of Zr with 1 ×10^{-4} × 5.83 g of ZrO_2

Or 2.635 g of Zr with 5.83 ×10^{-4} g of ZrO_2

Or 1 g of Zr with 2.21×10^{-4} g of ZrO_2 = (2.21×10^{-4})/ 123.22 moles

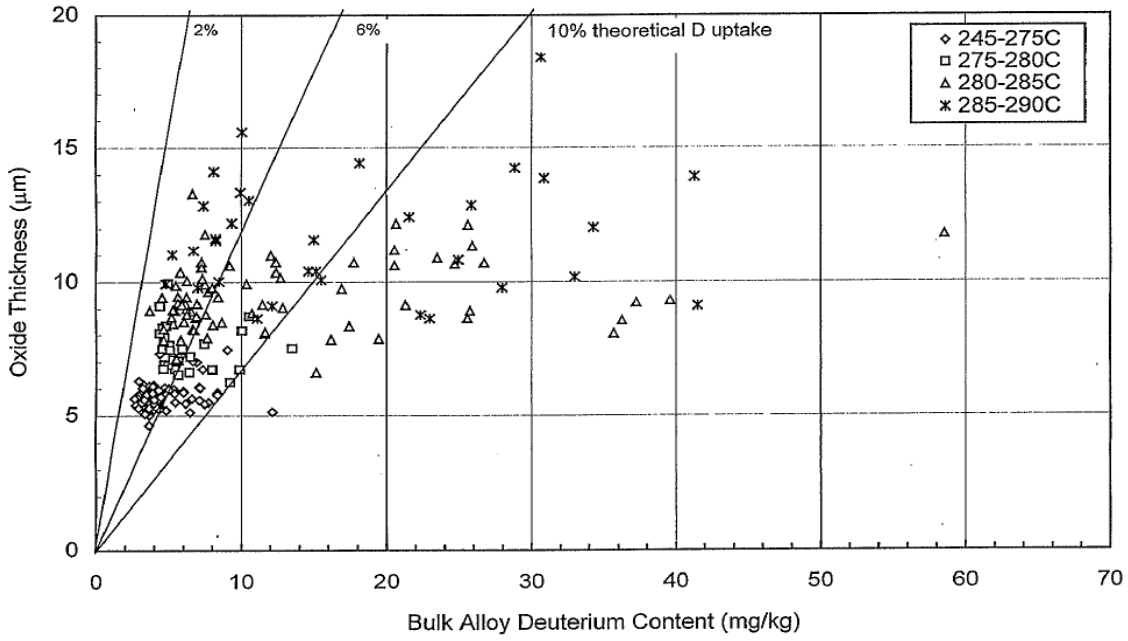
= 1.79 ×10^{-6} moles of oxide

At 100 % pickup this would be associated with 4 × 1.79 ×10^{-6} moles of deuterium atoms = 14.3 × 10^{-6} g of D

Hence, the D concentration for 100 % pickup by formation of 1 μm of oxide is 14.3 ppm

OPG has compiled a vast amount of data on deuterium pickup and oxide thicknesses for pressure tubes exposed to varying effective full power years (EFPY) in its Pickering, Bruce and Darlington reactors. An example of such data is presented below which is a plot of oxide thickness (in μm) vs. deuterium pickup (in mg/kg) at different temperatures. (A Pickering outlet operates at about 290 °C).

Pressure Tube Deuterium Pickup as a Function of Oxide Thickness



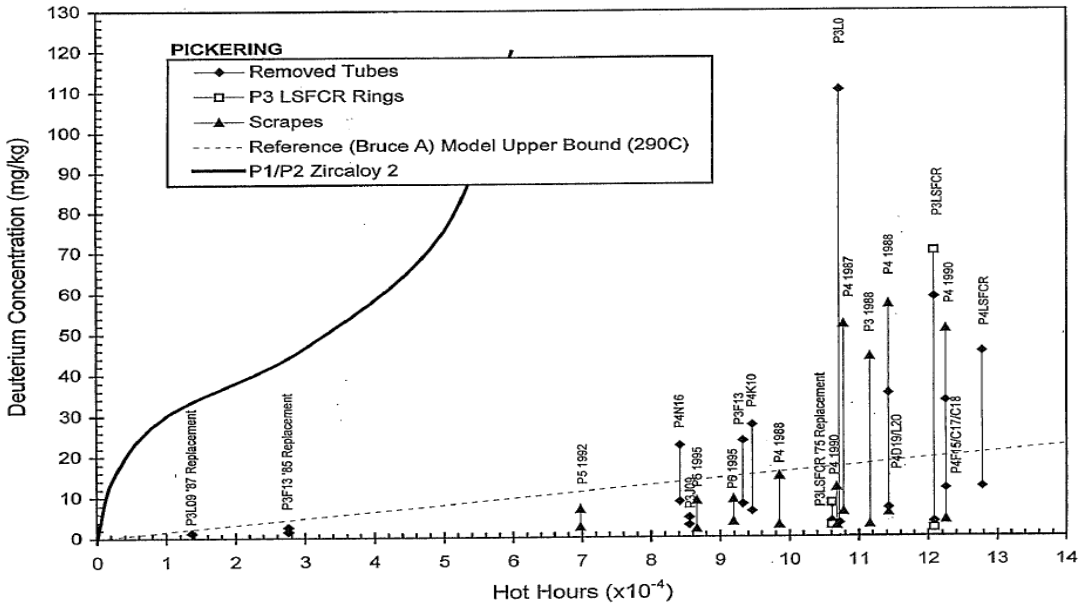
From this plot it is evident that there is great variability in the percent theoretical uptake of deuterium by CANDU pressure tubes. This variability was acknowledged in a recent review of the corrosion and deuterium pickup by CANDU pressure tubes and the reasons for this variability are discussed in the extract below - See CNL Nuclear Review Vol 5 (1), published June 2016:

> *Along the main body of a pressure tube the deuterium concentration increases and peaks near the outlet end. Approximately 2% - 10% of the deuterium generated by the corrosion process is absorbed. In general, the deuterium concentration in the main body of the pressure tube increases with time. At the 1.5 m axial location the increase is approximately linear with time, whereas at the 4 m and 5 m axial locations the uptake rate is increasing with time. However, monitoring programs have indicated that there is reactor-to-reactor and tube-to-tube variability in deuterium ingress.*
>
> *Reactor-to-reactor variability is likely a result of variations in reactor operating conditions such as: temperature, neutron flux, thermal hydraulics, and water chemistry. Tube-to-tube variability likely results from differences in the manufacturing process such as alloy chemistry, quenching practices, extrusion variables, microstructure, texture, coldwork, surface finish, and stress-relieving treatments. In addition to the monitoring program, research programs are conducted to understand the factors that influence in reactor corrosion and deuterium ingress. In many cases there are synergistic relationships between the variables making it difficult to determine the relative importance of any one variable in terms of its*

influence on pressure-tube corrosion and deuterium ingress behavior from reactor data alone.

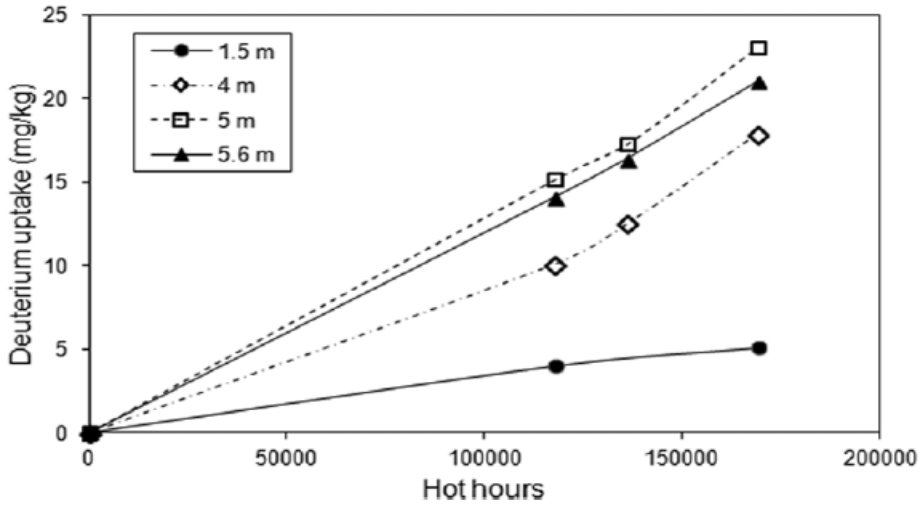
Although not explicitly stated, this quote amounts to an admission that deuterium pickup by CANDU pressure tubes is subject to so many causative factors *that it is essentially impossible to reliably predict future trends in the deuterium content of any particular pressure tube*. This is especially evident when deuterium pickup data are plotted as a function of the in-reactor exposure of the pressure tube, as shown below, where the large variability in deuterium pickup is clearly evident.

Deuterium Pickup by Pickering Pressure Tubes as a Function of Hot Hours of Exposure



The CNL Report noted above provides a recent version of this plot for so-called “representative uptake behavior”, (that ignores the scatter in the data):

CNL Deuterium Uptake vs. Hot Hours of In-Reactor Exposure



A comparison of CNL's plot with OPG's data for Pickering reveals that many pressure tubes show deuterium pickups that are well above the values depicted in the CNL plot. Thus, for example, the CNL "representative behavior' for deuterium pickup near the outlet of a pressure tube is about 15 mg/kg after an in-reactor exposure of 120,000 hot hours, which is equal to 1.25 mg/kg/10^4 Hot Hours. By comparison, OPG's data indicate that many Pickering pressure tubes show deuterium pickups in excess of 40 mg/kg after in-reactor exposures of 120,000 hot hours, which is equal to 3.3 mg/kg/10^4 Hot Hours.

OPG's most recent Pickering Fuel Channel Fitness-For-Service Report states:

> *Scrape sampling and material surveillance examinations provide measurements of hydrogen content in body of tube, as well as rolled joint regions of the pressure tubes. CSA N285.4 has established acceptance criteria for maximum hydrogen concentration values as well as maximum allowable rate of change in hydrogen concentration. Measurements have shown that hydrogen content is projected to remain within acceptance limits.*

The current CSA N285.4 and N285.8 limits on H_{Eq} are as follows:

> Maximum allowable hydrogen equivalent concentration at the outlet of a pressure tube is 100 mg/kg

12.3.5.2 Acceptance criteria

The determination of H_{eq} shall be considered acceptable when

(a) the predicted concentration value at the end of the next periodic measurement interval is below the level at which hydrides are present at sustained operating conditions; and

(b) the measured/determined rates of change in H_{eq} are less than those defined in the following Table:

Maximum channel outlet temperature	Maximum allowable rate of change in H_{eq} concentration per 10 000 hot operating hours
< 315 °C	3 ppm H_{eq}
< 305 °C	2 ppm H_{eq}
< 295 °C	1 ppm H_{eq}

The channel outlet temperature of Pickering Units is ~ 292 °C, then as noted above, the limit on deuterium uptake per 10^4 hot hours is 1 ppm, or 1 mg/kg H_{eq}. With this limit in mind it is useful to consider data for one of the earliest evaluations of Pickering B deuterium uptakes, namely the 1995 scrape campaign for Pickering Unit 6 – see OH Report A-FC-97-113-P, issued in July 1998:

Measured Deuterium Uptakes for Pickering Unit 6 After 91,890 Hot Hours

Channel ID	Axial Position (m)	[D] (mg/kg)	D Uptake Rate (mg/kg/10^4 HH)
P6C15	1.5	3.7	0.40
P6C15	4.0	4.2	0.40
P6C15	5.0	4.3	0.47
P6C15	5.8	7.1	0.77
P6F14	1.5	9.7	1.06
P6F14	4.0	6.8	0.74
P6F14	5.0	9.8	1.07
P6F14	5.8	13.8	1.50
P6M13	1.5	5.5	0.60
P6M13	4.0	8.0	0.87
P6M13	5.0	9.2	1.00
P6M13	5.8	11.1	1.21
P6M22	1.5	3.7	0.40
P6M22	4.0	5.9	0.64
P6M22	5.0	4.7	0.51

P6M22	5.8	9.0	0.98
P6T16	1.5	0.45	0.45
P6T16	4.0	0.84	0.84
P6T16	5.0	11.3	1.23
P6T16	5.8	15.9	1.73

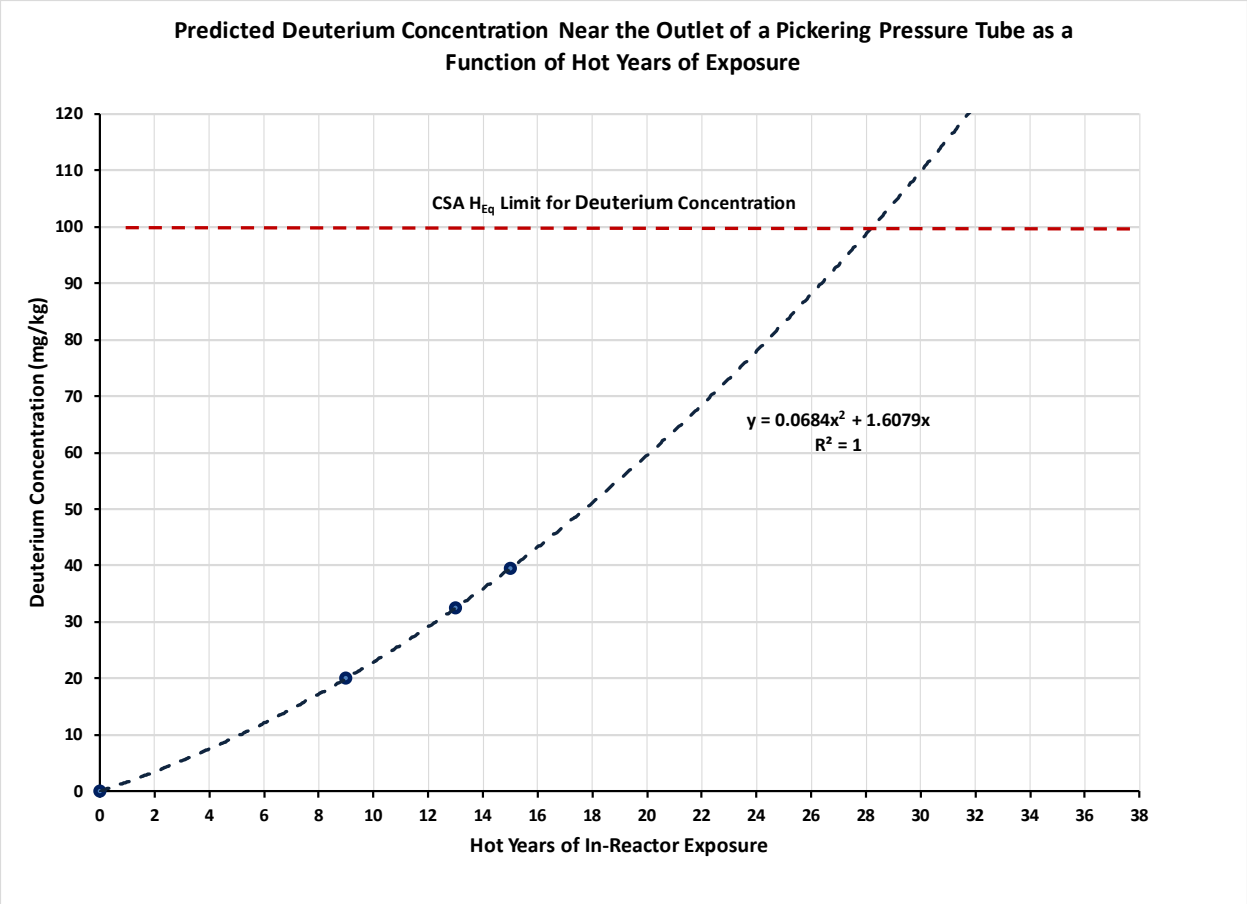
This Table of data shows:

(i) 6 out of 20 pressure tubes exceed the 1 mg/kg/10^4 Hot Hours limit.
(ii) The D uptakes near pressure tube outlets, (the 5.0 and 5.8-meter axial locations), vary by more than a factor of three.

Unfortunately, no oxide thickness data are available for oxide scrape samples. Nevertheless, techniques such as carbon-13 oxide dating have shown that accelerated growth of inside surface pressure tube oxides is to be expected for oxides thicker than about 10 μm, with concomitant accelerated deuterium pickup. For the current relicensing of Pickering B, OPG has applied to the CNSC to extend reactor operations to December 31st, 2024, implying exposures of pressure tubes in Units 5, 6, 7 and 8 to over 274,000 hot hours or about 31 hot years. Unfortunately, very little data on deuterium uptakes by Pickering pressure tubes beyond 150,000 hot hours, is publicly available.

Nevertheless, it is possible to extrapolate from published deuterium pickup data based on measurements up to 15 years of in-reactor exposure. As previously noted in this report, such an extrapolation is fraught with many uncertainties, but serves as a predictive tool that may be compared to subsequent data derived from real measurements as they become available

A best fit of such data, extrapolated to 32 hot years, is shown below. The plotted data clearly demonstrate that Pickering B is predicted to reach the CSA-imposed limit of 100 mg/kg H_{Eq} *well before the required 31 hot years*. It would be very helpful and informative for OPG to publish ***measured*** deuterium pickup data for Pickering B to confirm or deny this conclusion.



That such data are mandated, and are readily available is demonstrated by the Pickering License Condition Handbook which states (*Emphasis added*):

> **Predicted Maximum Heq Concentration**
>
> The predicted Heq concentration at the inlet and outlet burnish marks at the end of the evaluation period should be determined through a station or unit-specific model. The initial Hydrogen concentration should be from off-cut measurements and be channel-specific, the unit-specific bounding value, or the station specific bounding value. Operating conditions such as temperature and fast flux, where applicable to the model or its components, should be channel-specific, the unit-specific bounding combination, or the station-specific bounding combination. If any inputs are sampled from a distribution, the inputs as well as their percentiles should be justified. For a parametric or probabilistic approach, the input for and choice of the upper-bound percentile for the Heq prediction at the end of the evaluation period should be justified. In accordance with Clauses 12.3.4.6 and 12.4.4.6 of CSA N285.4, *OPG should report all of the parametric data and inputs used in the determination and prediction of the Heq concentration values*.

To this end, I request that OPG/CNSC provide electronic copies of the following reports:

1. OPG's revised compliance plan N-REP-31100-10061 R002 (N-CORR-00531-17932, e-Doc 4895642) for the use of CSA N285.8-15 "In-Service Evaluation of Zirconium Alloy Pressure Tubes".

2. OPG's procedural updates and technical justifications for pressure tube material testing submitted in e-Doc 3848127, N-CORR-00531-05488.

3. All reports containing data for deuterium pickup and oxide thickness measurements for pressure tubes removed from Pickering B in the period 2000 to 2017.