# Environmental Impact of the CANDU Fuel Cycle

**Charlene Fawcett, PEng**
Department of Chemistry and Chemical Engineering
Royal Military College of Canada/Collège militaire royal du Canada
Box 17000, Station Forces
Kingston, Ontario CANADA K7K 7B4
Email: fawcett--@rmc.ca
**Supervisors:** Dr. H.W. Bonin & Dr. K.A.M. Creber

## Abstract

The last half century has seen the birth and advancement in CANDU nuclear reactor technology. Concommitant with that technology has been the planned and unscheduled release of radioactive particles into the environment. This study looks at the impact of the CANDU fuel cycle on the Canadian environment over the past 50 years. Included in the analysis is a quantification of radionuclides released during mining, milling, refining, fuel fabrication, in-core power reactor operation, and spent fuel storage. Best available technologies to remediate existing radioactively contaminated sites and decommission power reactors are identified. Finally, an economic analysis is performed to estimate costs to remediate and isolate nuclear wastes from the general population.

This paper discusses findings to date on the graduate thesis topic outlined above.

## Nuclear Industry

The Canadian nuclear energy industry consists of power reactors situated in Ontario, Québec, and New Brunswick. Total capacity from these reactors is 15 048 MW, with Ontario Power Generation (OPG) reporting a capacity factor of 53.5% (7328 MW of 13 693 MW) ,Gentilly-2 operating at 88.0 % of 675 MW and Point Lepreau generating 71.4% of 680 MW in the year 2000 (1). Ontario Power Generation's total installed generating capacity from nuclear, fossil fuel and hydroelectric is 30 900 MW and the Ontario market consumed an average of approximately 14 900 MW in 1999 (2) with peak power demand at approximately 23 000 MW.

## Uranium Mining and Milling

Canada is the largest producer of uranium in the world, supplying approximately 25 % (8214 tonnes) of the world's supply in 1999 (1). In the mining of uranium, the rock matrix is first crushed and then dissolved in acid or base, in order to extract the uranium (3). The typical concentrations of uranium in early mining of uranium ranged from 2 to 6 pounds of $U_3O_8$ per tonne, or 0.1 to .3 % uranium. Because of this small percentage of uranium in the bulk rock mass, it was important to use a low-cost reagent to dissolve the matrix, and typically sulphuric acid was the acid of choice. The simplified chemical reaction is as follows:

$$UO_2 + H_2SO_4 + \tfrac{1}{2}\,O2 \rightarrow UO_2SO4 + H_2O$$

Pitchblende sulphuric acid oxidant uranyl sulphate water

The oxidants most often used include manganese dioxide and sodium chlorate. Dissolution in an alkaline solution involves using sodium carbonate or sodium bicarbonate, in the reaction:

$UO_2 + \frac{1}{2} O_2 + NaCO_3 \rightarrow UO_2(CO_3)_3Na_4 + H_2O$

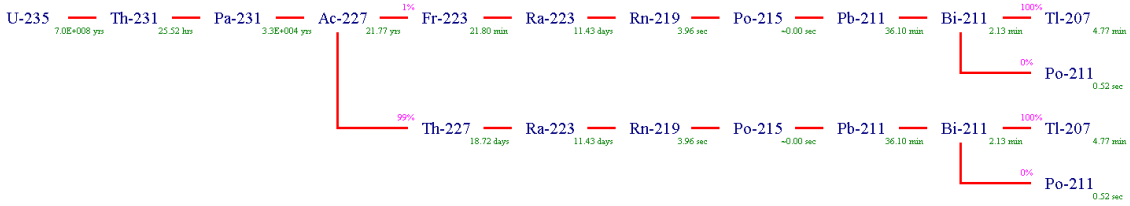
Pitchblend oxidant sodium carbonate sodium uranyl tricarbonate water

The oxidants used are potassium permanganate and copper salts in ammonium.

The recapture of uranium out of solution is achieved using either ion exchange resins or solvent extraction. The ion exchange process involves binding of aqueous uranium to an anion resin, rinsing with water to remove the impurities from the resins, ion exchange of the uranium from the bound resin, and rinsing of the free uranium from the resin. This procedure purifies uranium from an initial concentration of .1 to 1 g/l by a factor of 10 to 20 times.

The solutions of uranium are then precipitated out using sodium hydroxide, ammonium hydroxide, calcium hydroxide or magnesium hydroxide. The product is a mix of uranate and hydroxide, which is separated by filtration. The final concentration is approximately 70 % uranium, with alkali metals or alkaline earth bases and oxygen representing the balance. This is commonly referred to as “Yellowcake”.

The uranium mine and mill tailings contain high levels of uranium decay products that are effectively liberated from the rock matrix upon chemical processing for uranium extraction. Of the approximately 10 tonnes of uranium produced annually in Canada, another 1 to 10 million tonnes of mine tailings is generated. The accumulated volume of tailings is in the order of hundreds of millions of tonnes. Natural uranium consists of approximately 99.3% U-235, 0.7 % U-238 and <.1% U-234. U-235 undergoes decay in the following series (4):




Of these decay products, radium, radon and its radon daughters have received extensive review for the roles that they play in lung and bone cancers. In the mining of uranium ores, the percentage of cancer deaths due to radiation exposures has been estimated at

upwards to 50% of all miners within 10 to 20 years of exposure (7). With improved ventilation standards, one would expect to see a longer latency period, yet with the high grade ores currently being mined in Canada, one could expect continued excess lung cancers due to occupational exposures. In Canada, uranium is primarily mined in Saskatchewan. Uranium ore is also found in British Columbia, Nova Scotia and Labrador, but public opposition has prevented mining from taking place. (8)

It is a sad commentary on rights of the worker when one learns of the plight of the Dene Indian miners of Great Bear Lake. From the 1940s until 1960, the Dene Indians mined uranium at Port Radium, under federally owned Eldorado Mining and Refining: they have been plagued with lung, colon, stomach and other cancers ever since. Even with today's worldwide knowledge of uranium miner cancer risks, the Dene community had still not received compensation as late as 1998.(7) When this author inquired with CNSC in 2000 on the status of their compensation claims, there was no reported knowledge of the issue.

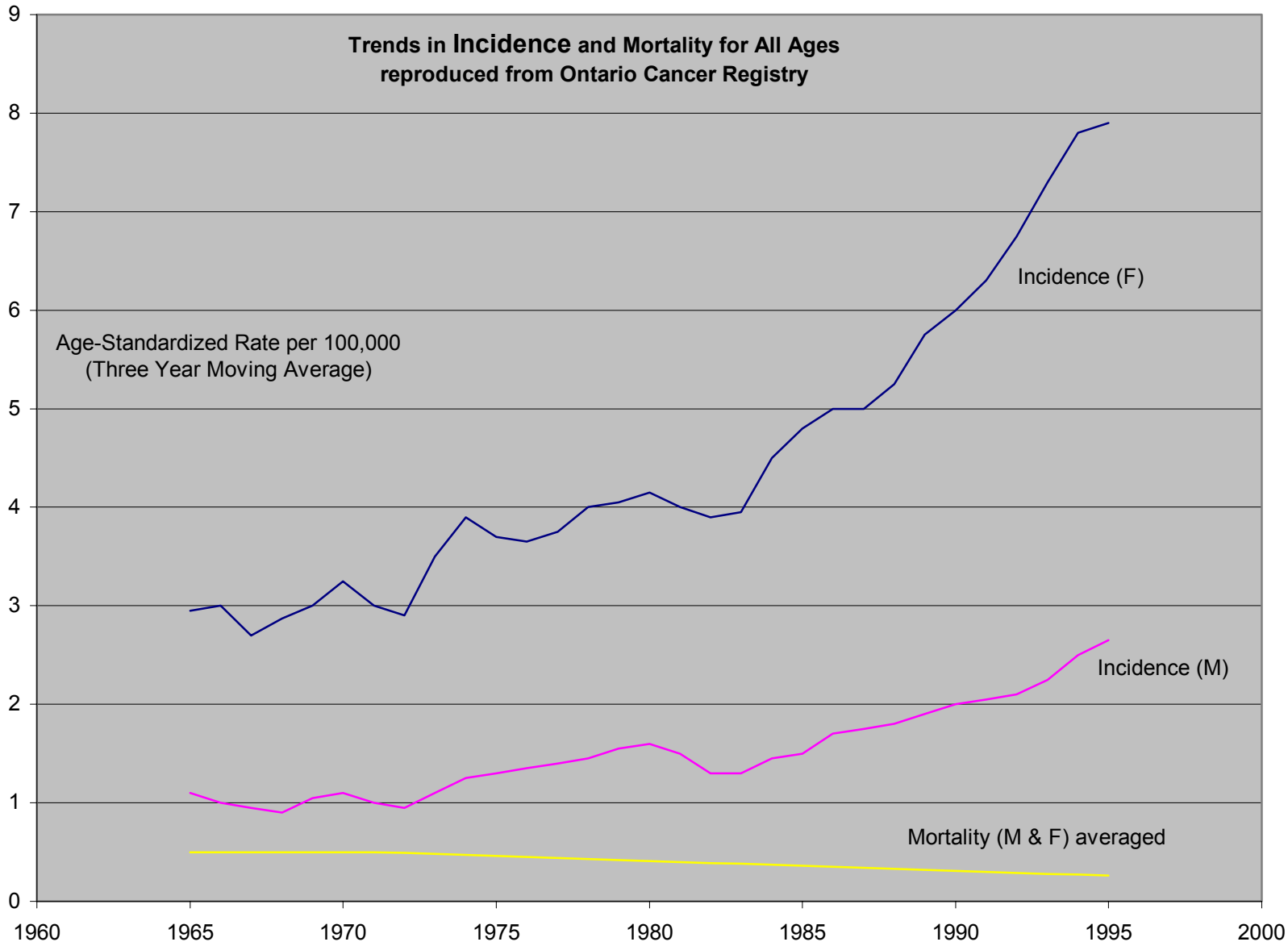
Uranium refineries are situated in Blind River and Port Hope Ontario, while fuel fabrication plant licenses are held at GE Canada in Toronto and Peterborough, Cameco Corp. in Blind River and Port Hope and Zircatec in Port Hope. One would expect to see occupational exposure challenges in these industries similar to that of mining of uranium. Engineering controls would determine the extent of exposure.

## CANDU Reactor Facilities

Nuclear reactors continuously emit radioisotopes into the environment. During power plant operations, fuel clad failures and pinhole leaks result in rapid escape of fission products into the containment system. Fission-products from compromised cladding as well as neutron activation products from the reactor are released into fuel storage bays. These radioisotopes easily work their way into the ventilation system, and on occasion into the groundwater, as has been seen at Pickering Nuclear Generating Station. Some fission products are captured in resin filters while others, such as tritium (H-3) and radioisotopes of iodine are more difficult to capture.

Of all possible cancer outcomes from exposure to nuclear reactor fission products, it is thyroid cancer that is the most clearly linked to radiation exposure. This fact is born out in Chernobyl cancer studies as well as follow-up of atomic bomb survivors and is cited in recent Cancer Care Ontario studies. Given the relative rarity of thyroid cancer incidence (3 in 1 million new cancers per year) any increases must be scrutinized with radiation exposure in mind.

Recent studies of thyroid cancer incidence show a troubling trend in Ontario (8):




Investigation further the possible causes of such increases in thyroid cancer, the following possible routes were considered:

1) Nuclear Weapons Test fallout;
2) Nuclear Reactor emissions; and
3) Other unknown mechanisms.

**Nuclear Reactor Emissions**

OPG monitors their station emissions as part of an ongoing requirement of relicensing. Because nuclear reactors are regulated by the federal government agency, Canadian Nuclear Safety Commission, it is the author's view that the provincial government does not play a dominant role in regulatory oversight. The reports that have written on levels of fission products escaping into the environment are therefore publications produced by OPG directly.

A review of recent environmental assessment data from OPG (9) reveals some concerns with regards to monitoring, including:

1) No monitoring of I-129. Only short-lived radioisotopes I-131 to I-135 are being monitored, despite the fact that I-129 is the more significant radioisotope of concern.
2) Use of passive air samplers in obtaining H-3 samples. Reporting of H-3 in air data without any discussion of actual values in air versus percent efficiency of sampling device.
3) Reporting of negative values for air noble gas emissions in 1999 at the Pickering Nuclear Boundary.
4) The sum of the fractions rule in occupational health and safety regulations involves measuring quantities of all fission products released and adding up the sums of the allowable limits, to determine if total dose is greater than the maximum allowable limit of 1. Until such time as all fission products are monitored, the total dose to the workers and public cannot be ascertained.
5) Reliance on theoretical CANDU release mechanisms to determine monitoring program. In particular, I-129 because the theoretical release limits are below regulatory concern.

## Tritium in Groundwater at Pickering NGS

The design of a CANDU reactor is such that tritium is produced at an increased rate, as compared to pressurized water reactors and boiling water reactors. The tritium is separated out of the cooling water, but inevitably, much escapes into the environment. One particular area of potential environmental impact is the release of both tritium and other fission products into the groundwater at Pickering Nuclear Generating Station.(10)

The natural background level of tritium in groundwater is under 1 Bq/l. The Ontario drinking water standard has been set at 7000 Bq/l: well above natural background, but below the theoretical level of concern. Due to faults in the drain system at Pickering, tritium and other fission products have escaped into the groundwater around the reactor. Levels as high as 2.7E8 Bq/l have been measured in the shallow groundwater in the area. OPG has set its own generic screening criterion of 3E6 Bq/l, in the absence of other regulatory limits.

Tritium toxicology study is limited to two published studies as of 1991(11). These studies demonstrated radiation-induced mammary tumours in female rats, and a third study was underway on radiation-induced myeloid leukemia in mice. The report recommended “Further studies of the RBE of tritium beta rays compared to X-rays at low dose-rate for stochastic endpoints such as induction of cancers”.

## Conclusions

Interim findings lead to unexplained increases in thyroid cancers in Ontario. The good news is that thyroid cancer is dropping, despite increased incidence. With regards to

environmental monitoring, it is clear that there is limited toxicology data to support the ambitious levels of tritium that OPG wishes to release into the environment. Until such time as there is evidence to support their screening criterion, it is the author's belief that the Ontario drinking water standard should be applied equally to groundwater. Dilute and disperse has never been an effective means of protecting the environment. Its widespread application within the nuclear industry is resulting in increased background levels that will not be easily remediated.

**References**


(1) Canadian Nuclear Association. Nuclear Canada Yearbook 2001. Ottawa, Ontario. (2001), 42.-43.
(2) Ontario Power Generation Inc. 1999 Annual Report. Analysis of Operating Results. (97.9 TWh over 9 months projected to 12 months and divided by 8760 hrs (1 year)), 24.
(3) Rozon, D., *"Gestion du Combustible Nucléaire- Document IGN-194"*, Ecole Polytechnique de Montréal, Québec. 1976, Pp. 25-30.
(4) Hacker, Charles. Radiation Decay Freeware Program Version 3.5, October 2000.
(5) Atomic Energy of Canada Limited., AECL-10718, "The Disposal of Canada's Nuclear Fuel Waste: Engineered Barriers Alternatives" (1994). pp. 12-14.
(6) Framand. K-H, and D'Aragon, P., "Uranium Resources in Canada and in the World" in Proceedings of 23rd CAN/CNS Annual Student Conference, March 1998., Kingston.
(7) Nikiforuk, A. "Echos of the Atomic Age" in Calgary Herald, March 14, 1998 edition.
(8) Ontario Cancer Registry, Cancer Care Ontario, Thyroid (ICD9 193). Cancer Incidence and Mortality in Ontario, 1964-1996. August 1999.
(9) Ontario Power Generation, Annual Summary and Assessment of Environmental Radiological Data for 1999. Report Number N-REP-03419-10000
(10) CH2M Gore and Storrie and Ontario Power Generation, "Pickering Nuclear Division Tritium in Groundwater Study", Report NA44-REP-07010-10001, September 2000.
(11) Myers, D.K., and Johnson, J.R., Advisory Committee on Radiological Protection ACRP-10, "Toxicity and Dosimetry of Tritium: A Review. January 1991. 21.