# The Quantitative Analysis of Uranium Isotopes in the Population of Port Hope, Ontario Canada

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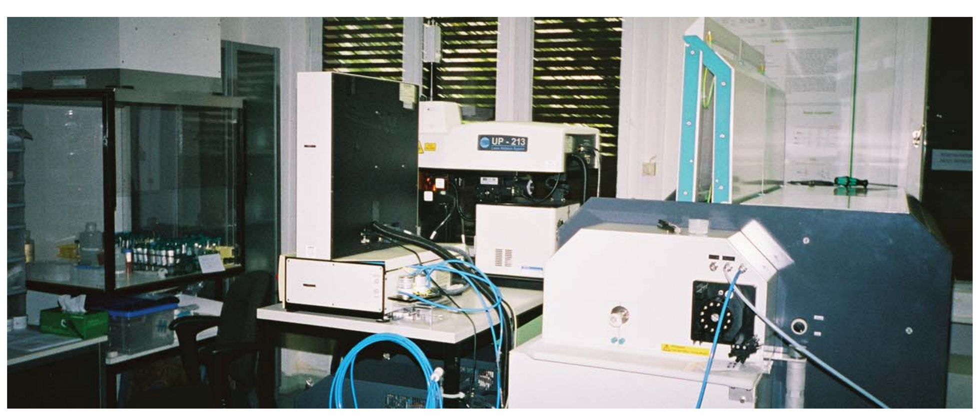
## Introduction

The contamination of the civilian population living in the vicinity of nuclear fuel processing plants has been a subject of numerous studies and controversy regarding the adverse effects of internal contamination with uranium isotopes released into the environment. Among many sites in North America, particular interest has been given to facilities such as Fernald, Ohio and Paducah, Kentucky although they were never followed up by objective research studies of the quantities and ratios of uranium isotopes. Likewise the oldest uranium processing facility in the world, located in Port Hope, Ontario Canada, although being studied by epidemiological research, the objective analytical study of uranium isotopes in the Port Hope population has never been conducted. The purpose of our study was the quantitative analysis of the internal contamination with four uranium isotopes in the population living near the uranium conversion facility in Port Hope, Ontario Canada.

Port Hope harbor back dropped by the Cameco facility, $UF_6$ drums are visible behind the fence

## Materials and Methods

The urine samples of subjects presenting with multi-system, non-specific symptoms of immune system alterations, musculo-skeletal, central nervous system, and neoplastic disease were obtained from residents of Port Hope and analyzed in reference to the control samples from residents of other parts of Ontario. The samples were analyzed at the Institute for Mineralogy, J.W. Goethe University, Frankfurt, Germany in a specialized radiochemistry laboratory by mass spectrometry. The analytical methodology included pre-concentration of urine by co-precipitation, oxidation of organic matter, uranium purification by ion-exchange chromatography, and ICP-MS double-focusing Thermo Finnigan Neptune multi-collector.



The laboratory at the Institute for Mineralogy, J.W. Goethe University

Table 1: $^{238}U/^{235}U$ Isotopic Ratio, Total Uranium, and $^{236}U$ Concentration

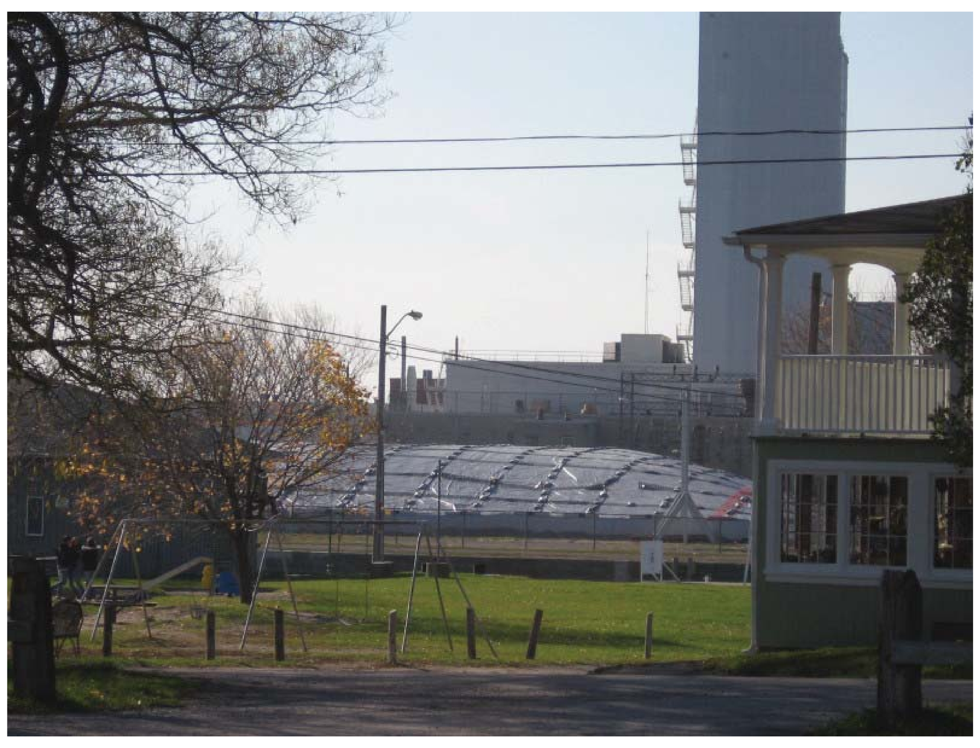
| Subject | $^{238}U/^{235}U$ | 2 SD | U ng/L | $^{236}U$ fg/L |
|---|---|---|---|---|
| 1 | 137.97 | 0.31 | 8.5 | < 1 |
| 2 | 137.99 | 0.57 | 24.8 | 1.7 |
| 3 | 147.11 | 1.42 | 7.0 | 31 |
| 4 | 138.75 | 1.12 | 5.1 | < 1 |
| 5 | 139.26 | 1.52 | 2.7 | < 1 |
| 6 | 137.71 | 0.67 | 9.4 | 517 |
| 7 | 138.22 | 0.83 | 8.8 | < 1 |
| 8 | 138.49 | 1.79 | 3.0 | < 1 |
| 9 | 137.34 | 0.78 | 3.7 | < 1 |
| Control 1 | 138.74 | 0.41 | 5.6 | < 1 |
| Control 2 | 138.15 | 1.54 | 2.1 | < 1 |

Table 2: $^{234}U/^{238}U$ and $^{236}U/^{238}U$ Isotopic Ratios

| Subject | $^{234}U/^{238}U$ | 2 SD | $^{236}U/^{238}U$ | 2 SD |
|---|---|---|---|---|
| 1 | $6.71 \times 10^{-5}$ | $8.88 \times 10^{-6}$ | | |
| 2 | $5.65 \times 10^{-5}$ | $1.11 \times 10^{-6}$ | $6.53 \times 10^{-8}$ | $8.6 \times 10^{-9}$ |
| 3 | $5.17 \times 10^{-5}$ | $5.03 \times 10^{-6}$ | $4.38 \times 10^{-6}$ | $4.3 \times 10^{-7}$ |
| 4 | $6.78 \times 10^{-5}$ | $9.43 \times 10^{-6}$ | $7.48 \times 10^{-8}$ | $4.3 \times 10^{-8}$ |
| 5 | $6.81 \times 10^{-5}$ | $5.06 \times 10^{-6}$ | | |
| 6 | $5.97 \times 10^{-5}$ | $4.69 \times 10^{-6}$ | $5.53 \times 10^{-5}$ | $3.9 \times 10^{-6}$ |
| 7 | $6.01 \times 10^{-5}$ | $4.50 \times 10^{-6}$ | | |
| 8 | $5.56 \times 10^{-5}$ | $7.09 \times 10^{-6}$ | | |
| 9 | $7.07 \times 10^{-5}$ | $3.16 \times 10^{-6}$ | | |
| Control 1 | $4.80 \times 10^{-5}$ | $9.82 \times 10^{-7}$ | | |
| Control 2 | $4.62 \times 10^{-5}$ | $5.50 \times 10^{-6}$ | | |

## Results

Our results show 4 of 9 samples containing uranium of non-natural origin. Subject 3 was highly positive for depleted uranium with a $^{238}U/^{235}U$ ratio of 147.11 ± 1.42 and a relatively normal abundance of total uranium. This sample contained a concentration of $^{236}U$ with a $^{236}U/^{238}U$ ratio of $4.38 \times 10^{-6} \pm 4.3 \times 10^{-7}$ indicating its reactor origin. Three other subjects (2, 4, and 6) contained detectable amounts of $^{236}U$. Subject 6 had a paradoxically high $^{236}U/^{238}U$ ratio of $5.53 \times 10^{-5} \pm 3.9 \times 10^{-6}$. Subject 2 also had a higher than normal concentration of total uranium at 24.8 ng/L. The $^{236}U$ in these samples indicates its origin as contamination with non-natural uranium. The remaining five subjects were negative for both depleted uranium and uranium-236. Control subjects had no detectable $^{236}U$ and a normal concentration of total uranium in their urine. Control subject 2 had a natural $^{238}U/^{235}U$ ratio. However control subject 1 had $^{238}U/^{235}U$ ratio that was slightly depleted. It was learned after testing that this person had visited Port Hope at some time prior to giving their sample.



A street, private home, and children's playground in the immediate vicinity of the Cameco facility; a tarp covers uranium tailings

## Discussion

The inadvertent exposure and toxicology of uranium isotopes in both military personnel and civilians employed in the nuclear industry or living in the vicinity of uranium processing plants has been well documented. Both parenteral and oral administration of uranium isotopes has been studied in animal studies and humans. Of particular interest are inhalational pathway toxicity studies which confirmed significant renal and pulmonary damage with eleven uranium compounds including oxides, fluorides, tetrachlorides, and nitrates in six different animal species as well as humans. Most recent studies of the Gulf War veterans have estimated a significant carcinogenic risk of inhaled depleted uranium. Uranium containing dust has been identified as the most important source of radiation exposure in uranium mining and processing. Toxicity of uranium in the ground waters (Saskatchewan, Canada), higher risk of lung cancer in uranium miners (New Mexico, Arizona), overall cancer risk in workers involved in the uranium processing industry (Ohio, Colorado), and numerous published studies from around the world all point to the realistic probability of adverse health effects of uranium isotopes in the human population living in the vicinity of nuclear processing plants.

## Summary

The contamination with depleted uranium has been verified and well documented in the studies on the military personnel in the conflicts in Iraq and Eastern Afghanistan, as well as, in the civilian population. The history of uranium contamination in Port Hope is well documented. Our results provide the first objective analytical study of long-term contamination and possible association with adverse health effects in the current population of Port Hope. These preliminary results warrant additional multidisciplinary studies.

## *History of the Nuclear Industry in Port Hope*

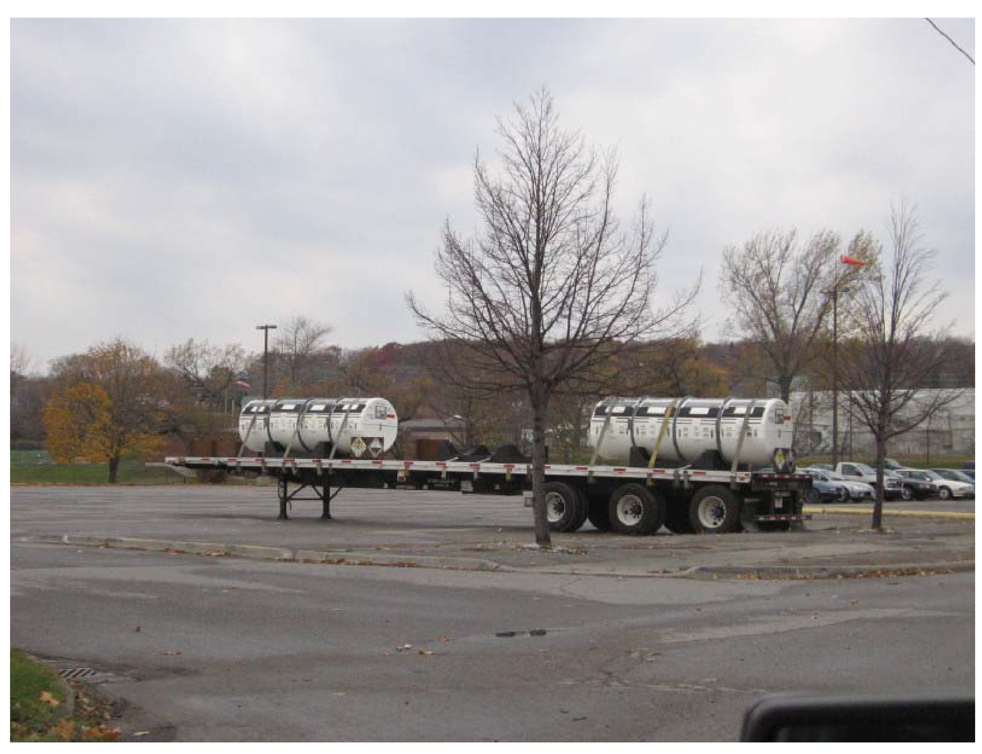
*Canada began mining uranium ores in the early 20th century for their radium content. In 1930, uranium ores were discovered in the Great Bear Lake deposit in the North West Territories and were developed by Eldorado Gold Mines for radium and uranium extraction. The refinery in Port Hope, Ontario was the first facility of its kind built and the only one in North America in the early 1940s that was equipped to refine uranium. Uranium concentrates (yellowcake) were shipped to the refinery where uranium was refined into uranium oxides ($UO_2$ and $UO_3$) as well as uranium hexafluoride ($UF_6$).*

*From 1941 to 1945, the entire production of refined uranium was supplied to the United States for use in the Manhattan project. The Port Hope facility had hundreds of tons of uranium concentrate on site from years of radium extraction but to meet demand Eldorado reopened the mine at Great Bear Lake which had shut down two years earlier. The facility also refined uranium from ores purchased by the US from Union Minière, a Belgian company that developed a deposit in the African Congo.*

*Canada's uranium mining and processing industry continued to sell uranium for nuclear weapons until 1959 when United States stopped purchasing uranium from Canada. Production slowed but continued under the Canadian government's uranium stockpiling program until the mid 1980s. Eldorado Nuclear built a new uranium refinery at Blind River, Ontario (early 1980s). The Blind River facility refined uranium concentrate into $UO_3$ which was shipped to Port Hope. In Port Hope, $UO_3$ was converted into $UO_2$ and $UF_6$. The $UO_2$ was then sold as fuel for CANDU reactors. The $UF_6$ was exported to enrichment facilities. The Port Hope facility also produced depleted uranium metals until 1992 and processed enriched uranium from 1966 to 1987. Port Hope also blended enriched and depleted uranium powders to specific isotopic concentrations.*

*In 1988, Cameco Corporation was formed by the privatization of Canada's uranium industry and the merger of two government owned corporations Eldorado Nuclear and Saskatchewan Mining Development Corporation. Cameco is the only Canadian company and one of only four companies currently providing uranium refining and conversion services to the western world; the other three being Honeywell in the United States, British Nuclear Fuels Limited in the United Kingdom, and Comurhex in France. Cameco is the world's largest uranium producer with four operating mines in Canada and the United States and two new mines being developed, one in Canada and the other in Central Asia. It has about 40% of the capacity in the western world to produce $UF_6$. It is also the only producer of ceramic uranium oxides for fuel in Canadian-built CANDU reactors.*

*Port Hope is also home to a facility established in 1965 to develop fuel to support Canada's nuclear energy program. This facility produces fuel pellets from refined $UO_2$ and assembles fuel bundles for CANDU reactors. In 2006, the plant was acquired by Cameco from Zircatec Precision Industries.*



Two $UF_6$ drums left on a trailer in a publicly accessible parking lot

Street in Port Hope with $UF_6$ drums just on the other side of the fence