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MODIFICATION OF THE NATURAL RADIONUCLIDE DISTRIBUTION BY SOME HUMAN ACTIVITIES IN CANADA

by

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ABSTRACT

Examples are presented of three types of human activity that have resulted in elevated radiation levels. Investigations carried out by a Federal-Provincial Task Force are described. The distributions of grab sample measurements of radon and radon daughter concentrations are compared for the Bancroft area, Cobourg, Deloro, Elliot Lake and Port Hope in Ontario, and Uranium city in Saskatchewan; it is concluded that the major point of difference between the communities that were investigated and the reference community of Cobourg is the departure from a symmetrical log-normal distribution at the higher concentrations.

INTRODUCTION

In Canada, responsibility for the control and supervision of the development, application and use of atomic energy is assigned to the Atomic Energy Control Board (AECB). The AECB receives its authority from the Atomic Energy Control Act which was promulgated in 1946 and empowers the AECB to write regulations and to administer a comprehensive system of licences for nuclear facilities and prescribed substances. Further information on the AECB is given in its annual reports.

During 1975, a number of investigations were initiated by the AECB that were concerned with radioactive contamination that may have resulted from past operations and disposal practices. By early 1976, the increasing number and magnitude of the investigations led to a Federal Cabinet decision to form a Federal-Provincial Task Force under the leadership of the AECB to expedite the investigation across Canada and to co-ordinate the use of Federal and Provincial resources. The investigation has followed a similar pattern in each area; following a preliminary radiation survey by the appropriate member agencies of the Federal-Provincial Task Force, the normal approach has been to hire a consultant to act as Programme Manager in each area. The Programme Manager carries out a more comprehensive radiation survey and determines the appropriate remedial measures for those homes where the radiation levels exceed the criteria (AECB, 1977) that have been adopted by the Task Force, and, in consultation with the AECB and the home-owner, carries out the remedial measures. Upon completion of the remedial work, the AECB and other regulatory bodies make protracted measurements to confirm that the average radiation levels are below the criteria.

This paper describes the preliminary radiation surveys conducted in several areas across Canada and discusses the results of these surveys.
PRELIMINARY RADIATION SURVEYS

Purpose

The purpose of the preliminary radiation survey in each selected area was quite specific; it was to provide a fast answer to the question "Does a problem exist?" If the answer was affirmative, the survey provided information on the magnitude of the radiation levels and the approximate number of homes and buildings involved.

Organization

The radiation surveys have been conducted at various times over a two-year period in several different communities; as a consequence, the approach to team organization and type of survey has changed. Initially, it was thought that a gamma radiation survey of the streets would provide a rapid guide to the "worst" areas, but it was soon found that the correlation was poor between gamma exposure rates outdoors and the radon or daughter concentrations inside houses in the vicinity. This type of survey was quickly replaced by an "on demand" programme which responded to those home-owners who requested a survey. It was soon evident from the random nature of the "discoveries" that a complete survey of the whole community would be necessary to provide assurance that significant cases of contamination had not been overlooked, and, incidentally, to give a balanced view of the extent of the problem.

The decision to survey the whole community in each case has allowed a more efficient organization to evolve. Emphasis has been placed on measurement of the radon daughter concentration in the basement and in a representative living area in each house; radon concentrations and gamma exposure rates have been measured only in those homes where a significant daughter concentration was found. The surveys have been carried out by teams of two people; one filling out the information sheet while the other collects the samples. After visiting four or five homes, the team returns the samples to a central point for counting, and then continues visiting more homes. An information office has been set up in each community to provide a working base for the survey teams and a communication point for local residents. The staff manning the information office schedule the homes to be sampled each day and have usually followed a street-by-street plan. Home-owners have been warned ahead of time either by announcements in the local newspaper or by telephone. It is usually necessary to repeat coverage of an area two or three times and to make visits during the evening in order to find the majority of residents "at home". This approach has resulted in a relatively small percentage of homes being "missed" in each community. These "missed" homes are checked subsequently by the Programme Manager.

Experienced survey teams have been provided by Health and Welfare Canada, the Department of Labour in Saskatchewan, and in Ontario, the Ministry of Labour which replaces the Ministry of Health in this role by a recent Provincial re-organization. This experienced core has been
considerably augmented by teams trained by the AECB which have been made up of members of the Nuclear Accident Support Teams from the Department of National Defence and University students hired for the summer.

Instruments and Procedures

A variety of instruments and techniques has been used in the different surveys to measure the concentration of radon in picocuries per litre of air, the concentration of radon daughters in Working Levels (WL), and the gamma radiation exposure rates in milli- or micro-roentgens per hour.

Radon. Measurements made by the Ontario Ministry of Labour are based on the work of Cowper and Simpson (1953) with modifications introduced by Tai-Pow (1978). Briefly, a sample of room air is collected by opening an evacuated 2 litre glass bottle; the sample bottle is then sealed and returned to the laboratory where the sampled air is transferred to a counting flask that has a thin aluminum collection foil in contact with a Lucite window coated with zinc sulphide. The air sample is left for 4 hours for the radon daughters to come to equilibrium, and the alpha activity is then counted for 2 to 16 hours. The system is calibrated with a standard radium solution obtained from the U.S. National Bureau of Standards.

Measurements made by Health and Welfare Canada followed the Lucas chamber method (Lucas, 1957) using commercially available cells and a portable counting unit. The cells were calibrated using standard radium solutions obtained from the U.S. National Bureau of Standards.

Radon daughters. Measurements made by Health and Welfare Canada depended upon the modified Kusnetz method (Kusnetz, 1956). Air samples were taken using a portable air pump and a 25 mm diameter filter; samples were taken at a flow rate of 10 litres per minute for 5 minutes. The same commercially available portable counting units that are used for radon measurements were modified to hold the filter papers in contact with a disc coated with zinc sulphide.

Measurements made by the AECB followed the same general procedure as that just described except that in one community (Elliot Lake) a different procedure was adopted that depended on counting the beta radiation from the decay of the RaB and RaC daughters (Horwood, 1978). Briefly, an air sample is drawn through a Whatman 41 coarse filter at a flow rate of 850 litres per minute for 5 minutes. The filter is then folded and placed between two pancake geiger detectors and the beta activity is counted for 5 minutes. The system is calibrated by comparison with the modified Kusnetz method which includes a correction for the age of the filter.

Gamma radiation. Measurements of the gamma ray exposure rate were intended to give an indication of the presence of excessive concentra-
tions of gamma-emitting sources like radium and thorium. Commercially-available survey meters employing geiger detectors were used to make measurements at 1 metre above the ground and in contact with the ground. All meters were calibrated using a radium source.

Data analysis. The distribution of radon daughter concentrations was analyzed in each community except one (Port Hope) where the radon concentration was analyzed in order to determine the best fit for the data, the mean, and the upper limit as defined by the mean plus three times the standard deviation; the cumulative frequency of the concentration distribution was also examined. These analyses were in addition to the simple distributions made according to arbitrary concentration intervals which have been reported previously (Aitken, 1977; Taniguchi, 1977).

No special statistical analysis was made of the gamma exposure rates.

AREAS SURVEYED

The areas selected for investigation were those where radioactive contamination was already suspected, and, in the first instance, the selection was based on a knowledge of the past operations in each area.

The first area selected for investigation was the community in which a radium-uranium refinery had operated for several decades; investigation of this community led to investigation of other communities in which non-nuclear metallurgical operations had made use of the refinery residues, and, finally, following the "trail" in reverse, it was speculated that there could be a "contamination" problem in the uranium mining communities. Each of these potential problem areas is considered below with a brief review of the human activities that may have affected the human exposures to radiation.

Operation of a radium-uranium refinery in Port Hope.

Port Hope is a community located on the north shore of Lake Ontario about 100 km east of Toronto in the Province of Ontario; the population today is about 9,000.

A refinery has been operating in Port Hope since 1932. Initially, radium was recovered from ores mined in Port Radium, Northwest Territories; the location of the refinery was chosen on economic grounds since it was less costly to ship each ton of pitchblende concentrates than to ship the seven tons of chemicals required for treatment. During the 1940's and early 50's the refinery operation was converted to the recovery of uranium although some radium refining continued until 1953 when it ceased altogether. Since the mid-1950's, the refinery has obtained feed material in the form of uranium concentrates from mines in northern Ontario and northern Saskatchewan as well as from various mines in the United States. During the whole of this period, the refinery has produced residues which have been more or less
radioactive according to the treatment of the concentrates prior to shipment from the mine; in the early years, the concentrates received little or no treatment. The first residues were placed on the refinery property; later residues were deposited in areas close to the refinery. In following years, the residues were reprocessed and re-deposited and some were sold for the recovery of other important metals. There were several periods during which there was an active building programme at the refinery when old buildings were demolished to make way for new construction. In consequence, contamination of the town of Port Hope could have resulted from any of the following causes:

a) spillage of residue during loading for shipment by road or rail.
b) contamination as a result of temporary storage of residues in various locations in the town while awaiting reprocessing.
c) distribution of contaminated fill, building rubble, and reclaimed building materials that originated from the refinery property.
d) the existence of past residue disposal areas within the present built-up area of the town.

Further details of the historical background of the Port Hope situation are given by Knight (1976).

The preliminary survey began in Port Hope early in 1976 and has extended over a period of two years to complete measurements in each of the approximately 3,000 homes, schools, and business premises.

Since the Radiation Protection Laboratory of the Ontario Ministry of Health was located in nearby Toronto and possessed an excellent capability for measuring radon concentrations, it was decided to perform this analysis on the air samples taken in Port Hope.

Non-nuclear metallurgical operations.

Another type of operation that is more difficult to recognize is that of a non-nuclear operation, usually metallurgical, where the use of radioactive material is quite incidental to the end-product and, hence, may not be recognized by the operator or by the regulatory agency. The particular concern in such situations is with the waste disposal practices employed by the processor which do not take account of the radioactive aspect of their waste. Two examples are given below to illustrate this particular problem.

An abandoned cobalt smelter. Deloro is a small village of about 70 houses situated in Hastings County in Ontario about 65 km east of Peterborough. A custom smelter specializing in handling arsenical ores operated in Deloro from 1907 to 1961. The smelter also specialized in the extraction of cobalt using feed materials from northern Ontario, Morocco, Rhodesia, and, of particular significance in this discussion, from the radium-uranium refinery in Port Hope. The smelter was abandoned in 1961 leaving a major problem of handling large volumes of arsenic-contaminated waste. In 1975, following preliminary investiga-
tions in Port Hope, the AECB became aware of radioactive contamination of the waste at Deloro resulting from the sale of residues to the smelting company for the extraction of cobalt. Measurements of the gamma exposure rates on the abandoned smelter property made in the Fall of 1975 showed a widespread, low-level radioactive contamination of slag and other waste that had been deposited on the property. Radon measurements were made in those houses that were closest to the abandoned property, and one family whose home was only a metre from the property boundary was evacuated by the Provincial Ministry of Health when it was found that the father had lung cancer. A complete survey of the community was made in 1977.

Ferro-columbium alloy production. Gloucester Township adjoins the Federal capital Ottawa. In 1976, three locations were found to be radioactively contaminated in the Township; there were two abandoned and one operating site where ferro-columbium alloys were/are produced from pyrochlore concentrates which contained thorium and radium. The feed materials originated from Brazil and eastern Canada. After processing, the thorium remains with the slag which had been buried on the two abandoned sites, and is currently under company control at the operating site. A complicating factor from a jurisdictional point of view has been the additional presence of chemically toxic contaminants. As abandoned sites, the first two did not present a serious problem, since access to them could have been restricted, but plans for building development at both sites led to both areas being cleaned up in order to avoid possible elevated radon levels inside any homes built there.

Elevated radiation levels in uranium mining communities.

Acting on the experience of Grand Junction, Colorado (Aspinall, 1971), and Port Hope, Ontario, it was suspected that a radioactive contamination problem could exist in uranium mining communities resulting from the use of waste rock from the mines being used on drive-ways or as backfill, or from the siting of houses on or close to natural occurrences.

Consequently, preliminary radiation surveys have been conducted in all the major uranium mining communities in Canada. In each case, the survey has stressed the measurement of radon daughter concentrations in Working Levels as being most meaningful to the mining population, but measurements of radon concentrations and gamma exposure rates have been made in those locations where the radon daughter concentrations were found to be significant.

There are three long-established, major uranium mining communities in Canada; Uranium City in Saskatchewan, and Elliot Lake and the Bancroft area in Ontario. Recently-opened uranium mines in Saskatchewan do not have local communities.
Uranium City, Saskatchewan. Uranium City is located in northern Saskatchewan near the north shore of Lake Athabaska. The town was first laid out in 1953 and currently has a population of about 2,200 living in about 500 homes. Radiation surveys of the community were undertaken by Health and Welfare Canada and the Provincial Department of Labour from February to June 1976 and was completed in the Fall of that year (Taniguchi, 1977).

Elliot Lake, Ontario. Elliot Lake is located about 20 km north of the northern shore of Lake Huron and about 135 km west of Sudbury. The town was first laid out in 1955 and has a present-day population of about 9,000 which is rapidly increasing as a result of an expansion in the mining activity; there are about 2,000 homes and other buildings. Radiation surveys of the community were undertaken by AECB-trained teams made up of students and Canadian Forces personnel, with assistance from Health and Welfare Canada staff.

Bancroft area, Ontario. The village of Bancroft is situated in central Ontario and is surrounded by many smaller communities that have some association with uranium mining in the area. Currently, only one of the four mines in the area is operating. Central Ontario is a vacation area and many of the homes pre-date the establishment of the uranium mines in 1953; there are about 1200 homes in the uranium mining area. Radiation surveys of the area were undertaken in the Summer of 1977 by teams of students working under the guidance of experienced staff from AECB, Health and Welfare Canada, and the Provincial Ministry of Labour.

RESULTS

The distribution of the maximum measured radon concentrations found in each of 2,961 homes in Port Hope is shown in Figure 1; it is noted that this maximum concentration was usually found in the basement. For comparison, Figures 2a and 2b show the distribution in Cobourg, Ontario. Cobourg is located on the north shore of Lake Ontario about 10 km east of Port Hope, and, having no suspected contamination problem, was used as a convenient reference for evaluating the measurements obtained in Port Hope. Measurements were made by the Ontario Ministry of Health in the Spring and Fall of 1976 (Aitken et al, 1977). The distribution of the maximum measured radon daughter concentrations found in the homes of Uranium City, Elliot Lake, and the Bancroft area are shown in Figures 3, 4 and 5, respectively. Attempts were made to fit the data to the normal and Poisson distributions without success; finally, it was found that the lognormal distribution when fitted to the logarithms to base 10 of the data gave an acceptable value for the chi square statistic in each case. The significant statistics calculated were the mean and standard deviation of the log-normal distribution and the upper 99% confidence level; for comparison with other studies the percentage of observations greater than 0.02 WL was also calculated since this is the primary criterion for remedial work.
The antilog of the log-normal statistics for the air samples are listed in Table 1 for each of the communities where these measurements were made; included for comparison are the same statistics for Cobourg. It is noted that the values for the radon daughter concentrations in Table 1 for Port Hope and Cobourg were calculated from the radon figures using an equilibrium factor of 30% to convert to Working Levels. The statistics for Deloro are also listed, but, with only 68 homes sampled, a graph showing the distribution was not thought to be worthwhile.

The cumulative frequency plots for Port Hope, Cobourg, Uranium City, Elliot Lake and the Bancroft area are shown in Figures 6, 7, 8, 9 and 10 respectively.

A summary of the soil sample analysis from the locations in Gloucester Township is given in Table 2; the gamma exposure rates measured in contact with the ground at the site of each sample ranged from 0.2 to 5.0 mR/h, but the correlation between these field measurements and the laboratory analyses of the principal radionuclides was poor. Generally, the estimated errors for the laboratory analyses are better than a few percent, but the presence of low energy radiation plus the energy dependence characteristics of the field survey instrument introduce relatively large errors so that a poor correlation is not surprising. The laboratory analyses were performed by the Radiation Protection Laboratory of the Ontario Ministry of Labour.

DISCUSSION OF RESULTS

Before drawing any conclusions, it is necessary to establish the value of the data to be discussed. With respect to the radon and radon daughter measurements, the surveys were very broad in that virtually every house in the community was sampled, however, the sample times were of very short duration (a matter of minutes) while the whole survey in a single community extended over a relatively long time, and, in some cases, over several seasons. It is well known that the radon and daughter concentrations vary considerably according to ventilation and meteorological conditions. Hence, the one-time measurement recorded for a single house has little meaning, but the total distribution provides a useful qualitative picture for each community and comparisons will be made on a community basis.

In comparing the log-normal distributions shown in Figures 1 to 5, inclusive, it is noted that only the "normal" community of Cobourg has a symmetrical distribution and that all of the others exhibit a marked asymmetry by having a significant number of homes with higher concentrations.

Comparison of the cumulative frequency plots shown in Figures 6 to 10, inclusive, exhibit the same departure from a symmetrical distribution for the investigated communities as was noted in Figures 1 to 5.

Using Bartlett's Method (Bartlett, 1937) for testing the homogeneity of the variances of the seven log-normal distributions listed in Table 1, it is concluded that at the 95% confidence level:
1) there is no significant difference between the Spring and Fall
distributions for Cobourg

2) there are three mutually exclusive populations i.e. Cobourg, Port
Hope, and the remaining communities which correspond respectively
to a normal, uncontaminated community in Southern Ontario, a
community in the same area contaminated by human activities, and
a group of communities sited in or near uranium mining areas.

It will be interesting to compare these results with those of the
survey conducted by Health and Welfare Canada during the Summer of 1977
which are to be published shortly (Letourneau et al, 1978).

The analyses listed in Table 2 have no particular significance
other than to demonstrate the concentrations found in areas where un­
controlled disposals have taken place.

Finally, it is noted that two types of mechanism have come into
play in the communities under discussion which have modified the dis­
tribution of the natural radionuclides concerned. In the cases of Port
Hope, Deloro, and Gloucester Township, radioactive material was brought
from other places for processing purposes and subsequently resulted in
contamination of the local communities with possible unanticipated ex­
posure of the local populations. In the cases of the uranium mining
communities, the mechanism is slightly different in that the local pop­
ulation has been deliberately introduced to the area where the concen­
trations of the natural radionuclides are higher than elsewhere in
order to mine them.

FUTURE WORK

Programme Managers have been appointed for Port Hope, Uranium
City, Elliot Lake, and the Bancroft area. In each case they have
carried out a much more detailed investigation and further interesting
data can be expected in the future.

A particular shortcoming of the preliminary investigation has
already been noted, i.e. the inability of the grab sampling technique
to allow for variations in the air concentration. In order to deal
with this problem, G. Cowper at the Chalk River Nuclear Laboratories of
Atomic Energy of Canada Limited has developed an instrument for inte­
grating the exposure to radon in air; it is planned to use many of
these instruments in making compliance inspections at those sites where
remedial work has been completed.

ACKNOWLEDGEMENTS

The gathering and analysis of the large body of data that has been
discussed here has involved a large number of people from the member
agencies of the Federal-Provincial Task Force on Radioactivity and
their help and assistance is gratefully acknowledged.
REFERENCES


Fig. 1  Log-normal distribution of radon concentrations inside homes in Port Hope.
Fig. 2A Log-normal distribution of radon concentrations inside homes in Cobourg; Spring, 1976.

Fig. 2B Log-normal distribution of radon concentrations inside homes in Cobourg; Fall, 1976.
Fig. 3 Log-normal distribution of radon daughters concentrations inside homes in Uranium City.
Fig. 4  Log-normal distribution of radon daughters concentrations inside homes in Elliot Lake.
Fig. 5  Log-normal distribution of radon daughters concentrations inside homes in the Bancroft area.
Fig. 6 Cumulative frequency plot of the logarithm of radon concentrations inside homes in Port Hope.
Fig. 7 Cumulative frequency plots of the logarithm of radon concentrations inside homes in Cobourg during the Spring and Fall, 1976.
Fig. 8 Cumulative frequency plot of the logarithm of radon daughters concentrations inside homes in Uranium City.
Fig. 9 Cumulative frequency plot of the logarithm of radon daughters concentrations inside homes in Elliot Lake.
Fig. 10 Cumulative frequency plot of the logarithm of radon daughters concentrations inside homes in the Bancroft area.
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* Radon daughter concentrations for Port Hope and Cobourg are calculated from the radon concentrations using an equilibrium factor of 0.3.
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