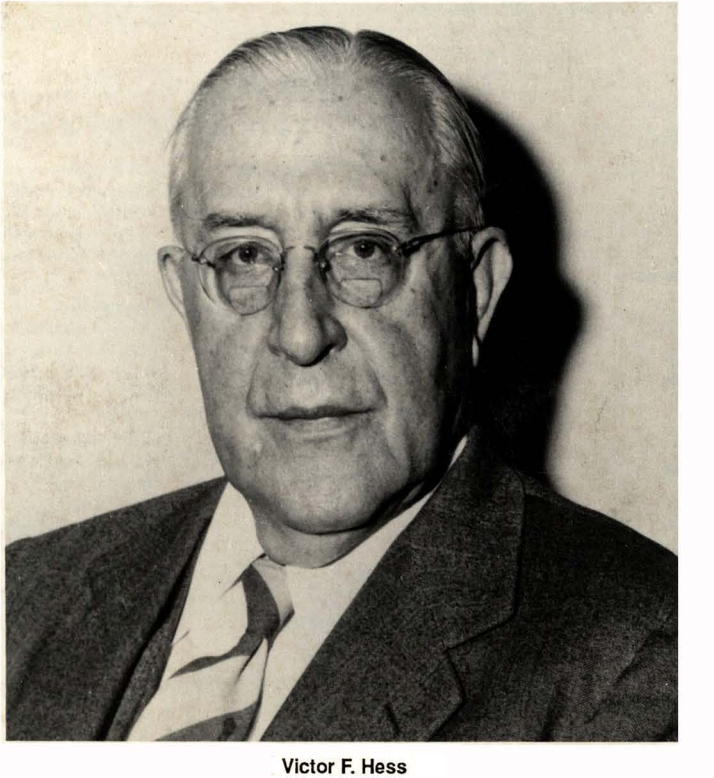
VOLUME 19, NUMBER 5, 1993 ISSN 0160-4120

SPECIAL ISSUE

HISTORY OF RADIUM, URANIUM, THORIUM, AND RELATED NUCLIDES IN INDUSTRY AND MEDICINE

ENVIRONMENT INTERNATIONAL

A Journal of Science, Technology, Health, Monitoring and Policy



Victor F. Hess

Pergamon Press
New York Oxford Seoul Tokyo

ENVIRONMENT INTERNATIONAL

A Journal of Science, Technology, Health, Monitoring and Policy

Editor-in-Chief A. Alan Moghissi
Temple University

Managing Editor Barbara Moghissi
Regulatory Science Press

Production Editor: *Lorraine Petrella, Pergamon Press, Tarrytown, New York.*

Editorial Office: *Environment International,* P. O. Box 7166, Alexandria, Virginia 22307, USA.

Publishing, Subscription and Advertising Offices: Pergamon Press Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, USA INTERNET "PPI@PERGAMON.COM" and Pergamon Press Ltd., Headington Hill Hall, Oxford, OX3 0BW, England.

Published Bimonthly. *Annual Institutional Subscription Rate* (1993): £270.00 (US$432.00). Sterling prices are definitive. US dollar prices are quoted for convenience only, and are subject to exchange rate fluctuation. Prices include postage and insurance and are subject to change without notice.

HISTORY OF RADIUM, URANIUM, THORIUM, AND RELATED NUCLIDES IN INDUSTRY AND MEDICINE

A SPECIAL ISSUE OF
ENVIRONMENT INTERNATIONAL

Pergamon Press
New York • Oxford • Seoul • Tokyo

Pergamon Press Offices:

U.S.A.: Pergamon Press, 660 White Plains Road, Tarrytown, NY 10591-5153, USA

U.K.: Pergamon Press, Headington Hill Hall, Oxford, OX3 0BW, England

KOREA: Pergamon Press, K. P. O. Box 315, Seoul 110-603, Korea

JAPAN: Pergamon Press, Tsunashima Building Annex, 3-20-12 Yushima, Bunkyo-ku, Tokyo, 113, Japan

Environment International, Vol. 19, p. 419, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

EDITORIAL

HISTORY OF RADIUM, URANIUM, THORIUM, AND RELATED NUCLIDES IN INDUSTRY AND MEDICINE

No scientific field has fascinated the scientific community and the general public as much as radioactivity and nuclear reactions. To this date, pioneers of this field are both revered and feared. Recognizing the fascination for the history of radiation and radioactivity, the University of Salzburg organized an international workshop on the history and current uses of radium, uranium, thorium, and related nuclides in industry and medicine. The workshop was held in Badgastein, Austria on October 1-3, 1991. The Radium Dial Painters' Project of Argonne National Laboratory was a valuable resource to the organizers of the workshop.

This special issue is devoted to the history of radium, uranium, thorium, and related nuclides in industry and medicine. The editors and authors have drawn upon historical materials and insights gathered in the course of their primary work on the physiological fate of ingested or inhaled radionuclides, the remediation of sites involved in the early uses of radioactive substances or application of radionuclides in research, medicine, and industry.

After the discovery of radioactivity by Henri Becquerel and the discovery of radium by Marie Curie, radioactivity and radionuclides acquired magical fascination. The medical profession embraced the application of radionuclides for treatment of many important diseases. Radium was considered a natural resource with a limited supply, but of exceptional significance.

The application of radium was not limited to the field of medicine. Once radium (and to a lesser extent thorium and uranium) became available in larger quantities, their applications spread to industry. Probably the largest quantity of radium was used in the luminizing industry. Suddenly, watches and clocks could be read in the dark and pilots could see their instruments at night. Unfortunately, occasionally radium was also carelessly used or abused in medicine and industry. Probably, the true carelessness in the usage of radium occurred in certain factories during this early application of radium. Victor Hess contributed significantly to the protection of workers from the deleterious effects of exposure to radiation and thus an editorial is devoted to this Austrian physicist.

Why devote an entire issue of an environmental science to the history of radium and related subjects? There are several reasons for such a decision. First, radium was the one substance that saved a large number of lives at a time when there was no other method for treating cancer. Second, radium is clearly a pollutant with the potential for causing significant harm as demonstrated by the occurance of bone cancer in former radium dial painters. Third, it is naturally occurring and its concentration in environmental media (water, soil, and food) often exceeds the levels considered safe by regulatory agencies. Finally, while at one time it was an exceptionally valuable natural resource, today it is totally replaceable by artificially produced radionuclides, such as cobalt 60 and cesium 137.

Much can be learned from the history of radioactivity. Natural resources should be used prudently and cautiously. However, there is no reason for not using them. One should remember that the loss of 20 g of radium during World War I was considered one of the most important losses of that war because it was an irreplaceable natural resource.

The preparation of this topical issue was both difficult and fascinating. The documentation of historical events was often poor. The editors had to balance the requirements for accuracy and their desire to tell the story of this interesting subject. During this process, several papers had to be excluded.

Despite this shortcoming, the editors believe the topical issue provides a unique reference on a poorly-documented subject. We are hopeful that lessons can be learned and future mistakes can be avoided.

Fritz Steinhäusler
Edward R. Landa
John Rundo
James H. Stebbings
A. Alan Moghissi

Environment International, Vol. 19, pp. 421-423, 1993
Printed in the U.S.A. All rights reserved.

0160-4120/93 $6.00 +.00

EDITORIAL

VICTOR F. HESS

The symposium upon which this issue of *Environment International* is based was dedicated to the work of the Austrian physicist, Victor F. Hess. Hess is the discoverer of cosmic rays and an outstanding contributor to the scientific field which is now called health physics.

The discovery of cosmic radiation occurred in 1911 by Victor Franz Hess. Hess used an ionization chamber in his experiments, which was already in use for the detection of radioactivity. Since radiation is absorbed by matter, e.g. air, it was obvious that with increasing distance from the earth with its natural radioactivity, radiation intensity should decrease.

For the world exhibition of 1889 in Paris, Eiffel built a steel tower of 300 m in height. When an ionization chamber was taken to the top, a decrease in the radiation intensity with altitude was found, but it was less than had been expected taking into account the absorption observed in air. The question arose whether the ionization chamber recorded a second kind of radiation which, in contrast to radioactivity, came from the sky. At that time, airplanes were unsuited for high altitudes, but balloon ascents were well developed. Victor Hess, accompanied by two copilots, entered a balloon basket and measured the radiation intensity at various altitudes up to 5 350 m, breathing oxygen at higher altitudes. The intensity at high altitudes turned out to be not lower, but twice as high as that at sea level. This was a strong indication for the existence of a cosmic radiation.

Who was this daring young man, Victor Hess, in his flying basket? According to his hand-written autobiography, he was born on June 24, 1883 at Schloss Waldstein near Graz in Austria. He was the son of a forest official, Vincens Hess. Victor was educated at the Graz High School and then studied physics and astronomy at the University of Graz. Because of his excellent grades at the university, he received a *promotio sub auspiciis imperatoris* Ph.D. on June 16, 1906, which included a precious ring donated by the Emperor of Austria. The topic of his thesis was "About the refractive index of mixtures of two liquids taking into account the volume change which occurs upon mixing "(Hess 1905).

Hess then switched to Franz Exner at the University of Vienna and in 1910 he became lecturer (*Privatdozent*) after submitting a paper "Absolute determination of the content of radon in the atmosphere". In the same year, 1910, the Vienna Academy of Sciences opened its Institute for Radium Research with Professor Stefan Meyer as the head, and Hess joined the institute as First Assistant. He then worked on the absorption of gamma radiation in air, on the definition of radium standards, and on the heat production of radium after it had been separated from its decay products. Previously, he had made experiments on how to separate uranium X ($^{234}Th/^{234}Pa$) from uranium. Beginning in the winter term of 1908/09, he taught experimental physics at the Vienna Veterinary University. It was in this period that his balloon ascents and his discovery of cosmic radiation occurred. The discovery was first met with skepticism. It was not until 15 y later that all doubts about the existence of the new radiation were removed. Ten more years elapsed until he was honored with a Nobel Prize in 1936 for his discovery.

During World War I, Hess became head of the x-ray division of a reserve military hospital. While there, he worked on a precise determination of the number of alpha particles emitted by radium per unit time with an Englishman, Lawson, who had been trapped in Vienna by the outbreak of the war. Hess was the first to apply Geiger's counting chamber to detect gamma rays. In 1919, his efforts were honored with the Lieben prize of the Viennese Academy of Sciences.

In 1920 at the age of 37, Hess became Associate Professor of Physics at the University of Graz, which meant tenure and a chance to start a family. On September 6, 1920, he married the 52-year old Jewish widow with a son and a daughter, Maria Breisky née Wärmer. Hess had lived with the Breiskys and sampled the excellent products of Mrs. Breisky's culinary art, and Hess loved to dine.

In 1920, the U.S. Radium Corporation in Orange, N.Y. offered him a position of chief physicist, and the means to organize a research laboratory. Soon,

the U.S. Bureau of Mines appointed him a Consulting Physicist. His publications now dealt with medical applications of radium and with an improved method for recognizing radium in ore. He wrote a paper on the avoidance of burns of the skin in the radium treatment of deeper-seated cancers (Hess 1921). It had already been recognized that certain kinds of cancer were by factors of 4 to 7 times more sensitive to radioactive treatment than normal cells. Hess made an estimation of a suitable distance between the radioactive source and the human body. When Professor Stefan Meyer in Vienna asked him for proposals for a radioactive mineral springs law, he replied: "The American Medical Association in principle rejects the use of radioactive water for purposes of drinking, bathing, and inhaling because there is no evidence of a curing effect, and the cumulative effect of radioactive final products in the body at a long-term application is not at all considered harmless. Emanation cures are forbidden."

In 1925, after returning to Graz, Hess became full Professor. Since there were no radium samples available at Graz, Hess continued his former observations of electricity in air (Hess 1926; Hess and Benndorf 1928). In 1929 and 1930, Hess served as Dean of the Philosophical Faculty.

In the summer of 1931, Victor Hess was permitted to establish an observatory for cosmic rays on the Hafelekar, a mountaintop in the Alps with an altitude of 2300 m, easily accessible by a funicular. At the same time, he was offered a professorship at the nearby University of Innsbruck. For financial support of his research projects, Hess managed to interest the U.S. Rockefeller Foundation, which in 1933 granted him $5600 and one year later another $1000. In addition, Hess and the biologist Jakob Eugster investigated the biological effects of cosmic rays in animals, bacteria, and seeds. The results were published in a book (Eugster and Hess 1940).

At the age of 50, Hess was at the culminating point of his career. Unfortunately, his health began to decline. In 1933, he had an operation; and two years later an amputation of one of his thumbs became necessary. It may have been caused by radium burning during his Vienna work two decades earlier. In 1934, Hess, who passionately loved to smoke cigars, had to undergo an operation of his larynx which was under suspicion of cancer. Even so, after one year of nonsmoking, he started it again. His voice, which he needed for teaching, was heavily impaired.

In 1938, Hess and his wife were allowed to emigrate to the U.S., but not until the Swedish crowns that had come with the Nobel Prize had been confiscated. Hess was offered a professorship at Fordham University in New York City. His teaching obligation was five hours per week at the Graduate School where he taught for 18 years. In June 1948, he spent a month as a guest professor at Innsbruck University. In 1955, the President of Austria offered him a "außerordentlicher Versorgungsgenuß", a retiring pension for the years he had served at Austrian Universities. On the occasion of his 75th birthday, he again visited his home country. In 1959, he was honored with the Austrian *Ehrenzeichen* for Science and Art. He became an honorary doctor of the University of Innsbruck and of the Veterinary University of Vienna, and a Corresponding Member of the Austrian Academy of Sciences. In addition, he was honored in the U.S. with honorary doctorates from Fordham University and the Jesuit Loyola University of Chicago, and was named a Fellow of the American Physical Society. He had previously been elected to the Pontifical Academy of Science at Rome. In 1932, he received the Ernst Abbé Memorial Prize of the Carl Zeiss Foundation at Jena, Germany.

Of the 128 works (including books) which Hess published, 27 date from 1939 to 1950. These include studies of cosmic radiation, its seasonal and temperature variation, and effects on it by magnetic disturbances of the earth; the use of radioactive tracer methods; the radioactivity of minerals; the influence of the atomic bomb tests on the ionization of the atmosphere; and the ionization balance of the atmosphere. One important paper is about the radium poisoning of persons employed in the radium industry, where phosphorescent materials were impregnated with radium and the mixture was applied to dials and hands of watches. Together with McNiff, Hess developed two methods of diagnosis of radium poisoning where the radon content of breath samples was measured (Hess and McNiff 1947). From these results, the radium content of the body was calculated.

In 1956, at the age of 73 and after 18 years at Fordham, he retired. On December 17, 1964, at the age of 81, Hess died of pneumonia at his home in Mount Vernon, N.Y., after a long suffering from Parkinson's disease. He had been an American citizen for two decades and had become known in the U.S. for his work on the health of American workers in the radium industry.

Karlheinz Seeger
Ludwig Boltzmann Institut für Festkörperphysik
und Institut für Festkörperphysik
Universität Wien
Vienna, Austria

Acknowledgment — Let me thank Dr. Kerber and Mrs. Mag. Smola from the Central Library of the Physics Department of the University of Vienna for their cooperation in the literature search. Also it is my pleasure to acknowledge Dr. James Stebbings and Dr. J. Rundo for sending me copies of some important papers from the Fordham Archives about Victor Hess.

REFERENCES

Hess, V.F. Über das Brechungsvermögen von Mischungen zweier Flüssigkeiten unter Berücksichtigung der beim Mischen eintretenden Volumänderung, Diss. Univ. Graz; 1905. Sitzungsber. Akad. Wiss. Wien 114: 1231-1251; 1905.

Hess, V.F. On a simple method to avoid burns of the skin in the radium treatment of deeper-seated cancers. New York: U.S. Radium Corporation; Sept. 1, 1921.

Hess, V.F. Die elektrische Leitfähigkeit der Atmosphäre und ihre Ursachen. Braunschweig: Friedrich Vieweg & Sohn Verlag; 1926. English translation: London: Constable & Co.; 1928.

Hess, V.F.; Benndorf, H. Luftelektriziträt, Müller-Pouillets Lehrbuch der Physik, Vol. 5, 11. Edition. Braunschweig: Friedrich Vieweg & Sohn Verlag; 1928.

Eugster, J.; Hess, V.F. Die Weltraumstrahlung und ihre Biologischen Wirkungen. Zürich: Orell-Füssli Verlag; 1940. Fordham University Press; 1949.

Hess, V.F.; McNiff, W.T. Quantitative determination of the radium content of the human body and of the radon control of radium poisoning in persons employed in the radium industry. Am. J. Roentgenol. Radium Ther. 57: 91-102; 1947.

Environment International, Vol. 19, pp. 425-438, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

HISTORY OF THE DETERMINATION OF RADIUM IN MAN SINCE 1915

J. Rundo*
Argonne National Laboratory, Argonne, IL 60439-4833, U.S.A.

EI 9203-132 M (Received 4March 1992; accepted 9 May 1993)

In 1915, volunteers were injected with, or ingested, solutions of a radium salt, and the excreta were analysed for radium, but the first direct demonstrations of the presence of radium *in vivo* years after exposure were reported in 1925 by Martland et al.; they were also able to demonstrate the presence of radon isotopes in the exhaled breath. In 1929, Schlundt et al. reported radium levels in six living persons based on a calibration that was derived from measurements on a seventh who subsequently died and whose partial skeleton was analysed. In 1933, Ives et al. reported results for 40 subjects who showed gamma-ray results of 1 μg radium (i.e., retained radon) or more. There was an improvement in sensitivity by an order of magnitude in 1937 when Evans described the use of a Geiger-Müller detector and the metre-arc geometry; the method was still incapable of measuring radium at the "tolerance dose" of 0.1 μg. Ten years later, Hess and McNiff published details of an ionization chamber method that did permit such levels to be determined. In the early 1950s, three groups independently described the use of long high-pressure ionization chambers that were sensitive enough to detect the radiation from the naturally radioactive potassium content of the human body. In 1955, the introduction of sodium iodide as gamma-ray detector, with its high detection efficiency and spectrometric capabilities, and of large organic scintillators, revolutionized the assay of radium (and all other gamma-ray emitters) *in vivo*. The lower limit of detection of radium in man showed a steady decrease that was roughly exponential from 1929 to the mid-1950s with a half-period of a little more than 3 y.

INTRODUCTION

The history of the determination of radium in man is inextricably linked to the history of radium poisoning, primarily in the luminous dial workers (who are commonly but inaccurately referred to as the "dial painters"), but, in the very early literature, there are a few papers concerning the metabolism of radium in laboratory animals and, more importantly, in man. We might call this the "pre-history" of the determination of radium in man.

In 1915, Seil, Viol, and Gordon (1915) reported on the excretion of radium by a "normal" man aged 23, following intravenous injection of 100 μg radium (probably as the chloride) on two occasions 54 d apart. All excreta were collected and analysed for radium. It was quickly established that the urinary excretion of radium was far less than the faecal, and urine collection was stopped after 3 or 4 d. In the first experiment, faecal collection and analysis were complete for 9 d; subtraction of the cumulative excretion

*Present address: P.O. Box 675, Westmont, IL 60559, USA.

of radium from the amount injected gave the retention. In the second experiment, faecal excretion was only continuous for 7 d, but three additional samples were collected to 20 d post-injection. The results from the two experiments were in reasonable agreement with one another, indicating a retention at 9 d of 50-60%. Two points may be noted. Firstly, this value for the retention is substantially higher than both the prediction of 17% from the power function equation of Norris et al. (1955) and that of 13.3% from the ICRP model of alkaline earth metabolism in man (Marshall et al. 1973). Secondly, on two subsequent occasions, a man ingested 50 μg of radium, and Seil et al. reported that 25-35% of the ingested dose was retained 4-5 d later. This is somewhat higher than the value of 20% for the uptake of ingested radium reported more than half a century later by Maletskos et al. (1969).

Seil et al. determined the concentration of radon in the breath after the first ingestion, reporting the result both in nCi L^{-1} of air, and in Maché units (1 Maché unit was a unit of concentration of radon, corresponding to about 13.3 Bq L^{-1}). Their results could have been used to relate exhalation of radon to body content of radium, but were not. They made no attempts to measure gamma rays from radium in the body.

THE NINETEEN TWENTIES

A publication by Hoffman (1925) on radium necrosis provoked H. S. Martland into what he obviously regarded as premature publication of his continuing work on radium poisoning. In this paper we find the first reference to the detection of gamma radiation from radium in the living human, with the aid of an electrometer

". . . of the Wulf type . . . placed about 18 inches from the chest of the patient and 9 inches above the level of the bed. Readings were then made and showed . . . normal leak before test, 10 subdivisions per hour; total leak during test, 14 subdivisions per hour. The difference of four subdivisions per hour was caused by penetrative radiations coming from the . . . patient." (Martland et al. 1925).

No calibration was made or perhaps even possible; we believe now that this patient contained a substantial proportion of ^{228}Ra with the ^{226}Ra. This was certainly the case with the second patient discussed by Martland et al.; the leak of the electrometer when filled with the subject's dried exhaled breath was more than twice as great in the first 10 min as in subsequent 10-min periods, as shown in Table 1. Again, no calibration measurements were made because all these measurements were made merely to demonstrate the presence of radium *in vivo*.

Table 1. Martland's (1925) results of electrometer leak when filled with dried breath from a radium patient,demonstrating the presence of ^{220}Rn (thoron) as well as ^{222}Rn.

Period	Leak (subdivisions)
1st 10 min	33.5
2nd 10 min	16.2
3rd 10 min	15.7
4th 10 min	16.6
5th 10 min	17.7
Natural leak in 50 min (Background)	8.5

The first publication that described quantitative determinations of radium *in vivo* was by Schlundt et al. (1929), four years after Martland's first paper. These workers used a "Wulf-Hess quartz fiber electroscope" placed on a table "near to the middle of the back of the subject" who was seated in a chair. (Hess's modifications to the instrument used by Wulf were described in an article by Hess (1940) on his discovery of cosmic radiation, which earned him the 1936 Nobel Prize in Physics:

"I used a modification of Th. Wulf's apparatus with walls of zinc, thick enough to withstand the excess pressure of one atmosphere and a temperature compensation for the fiber electrometer."

The modified instrument would certainly qualify to be called a Wulf-Hess electroscope but it is not clear if the one used by Schlundt et al. (1929) was modified in exactly the same way.)

The procedure used by Schlundt et al. was to determine the time for the electroscope fibers to drift over the same 10 divisions for the following measurements:

Natural drift, scale readings 60-50
Drift, subject under test in chair, 60-50
Drift, subject under test in chair, 60-50
Natural drift, scale readings 60-50

If the first and last of these (what we would today call the background measurements) were not "practically identical," this was "a sure way of detecting an erratic behavior of the electroscope. . ." Some experimental results obtained by Schlundt et al. (1929)

are set out in Table 2. Except for the first, all the values for the radium content are estimated from a calibration factor of 8.57×10^{-5} scale divisions s^{-1} μg^{-1} radium. This value was obtained from the observed rate of drift

"on subject A a few months before death. After death the radium content of the bony skeleton and body parts was determined by means of an extensive series of radium determinations by the well-known emanation method, which when summed up gave the total radium present in the body of Subject A as 126 micrograms ..."

From the values in the second column of Table 2, it is clear that the background was extremely variable, being in the range 0.00330-01128 scale divisions s^{-1}, yet the net increase for each of the subjects B to G was less than this variability. It may be noted that the difference between the two extremes of background was the same as the response to 93 μg of radium, but presumably such large variations did not occur during the course of one day. Schlundt et al. (1929) commented that the "natural drift will vary with weather conditions" but they

"reached the conclusion that .... in the hands of an experienced operator working under carefully controlled conditions, 2 or 3 micrograms will give a positive test."

The average of 2.5 μg may be taken as their limit of detection.

Complicating factors that these workers recognized but only partially allowed for were the presence with the ^{226}Ra of substantial amounts of the much shorter-lived ^{228}Ra (more commonly known in those days as mesothorium, or MsTh1), and the emanation and exhalation of isotopes of radon in the two decay chains, ^{222}Rn and ^{220}Rn (thoron). Thus, they related the gamma-ray measurements on Subject B to total radium content on the basis of the analyses for radium in the body of Subject A, but then added the emanating part, deduced as 2.5 μg, to get a total of 92.5 μg. No allowance was made for the fact that the value of 90 μg resulted from ionization from a combination of the two radium isotopes. In this way, they deduced a value for the emanating fraction of ^{226}Ra that was much too small. Then, they assumed that the emanating power of ^{224}Ra (the parent of thoron, ^{220}Rn) was the same as that of ^{226}Ra. We now know that, when produced in the bone, only about 1% or less of ^{220}Rn is exhaled in the breath, because of its short half-life, compared with 60-70% for ^{222}Rn, the daughter of ^{226}Ra. Thus, Schlundt et al. (1929) would have seriously underestimated the body content of ^{224}Ra and consequently of ^{228}Ra, while overestimating the content of ^{226}Ra. The value of 2 or 3 μg that "give a positive test" must be construed as referring to ^{226}Ra free of ^{228}Ra.

Schlundt et al. drew attention to the importance of the subject showering, of changing into inactive clothing, and of removing all jewellery, before observations of gamma radiation from the human body were made. Flinn (1929) stressed the importance of measuring the natural drift of the electroscope with an unexposed person "placed in the same position and distance from the electroscope as the suspected person would be placed." He went further and recommended the use of corrugated cardboard between the person and the electroscope as an insulator to eliminate the effect of the body's heat on the instru-

Table 2. Some results reported by Schlundt, Barker, and Flinn (1929) showing the range of activities observed.

Subject	Nat. Drift (Av.) Div. per Sec.	Observed Drift Subject	Drift Increase	Approx. Radium Content, μg
A	0.00632	0.01711	0.01080	*126*
B	0.01128	0.01902	0.00740	90
C	0.00543	0.00576	0.00033	4
D	0.00713	0.00813	0.00100	12
E	0.00582	0.00768	0.00186	22
F	0.00813	0.01063	0.00250	29
G	0.00330	0.00405	0.00075	9

ment. Although colleagues suggested that there might have been shielding by the body of local gamma radiation or of cosmic rays, Flinn was not convinced. Examination of his data suggests that the latter effect was in fact present.

THE NEXT DECADE

The 1930s saw two very important papers. On the occasion of a conference on radium poisoning held in Washington, D.C., on 20 December 1928, the Surgeon General of the United States appointed a committee to act in an advisory capacity as to the character of work to be undertaken and the nature and efficiency of tests to be used in the study of radium poisoning. The first paper of the 1930s (Ives et al. 1933) was one of four from the U.S. Public Health Service, with the general title of "Health Aspects of Radium Dial Painting," that resulted from the recommendations of this committee.

Like Schlundt and his colleagues, Ives et al. used a "Wulf bifilar (Hess type) electroscope" for making the gamma-ray determinations. The instrument was

"placed as close as possible to the centre of gravity of the seated body . . . with the feet and legs under a specially constructed table, and with the body touching the edge of the table."

To minimize the possibility of surface contamination giving a falsely high reading, "the workers were required, before their examination, to bathe, shampoo their hair, and wear clothes that had not been worn in the factory." Watches and jewellery were not permitted to be worn. Ives et al. (1933) also recognized the possibility that inhaled radon might be retained and affect both the gamma-ray and radon tests; accordingly they did not allow "the girls to enter the factory during the day of examination" for gamma rays.

As an example of the results, to indicate the time taken for a test, and as an illustration of the sensitivity of their method, Ives et al. quoted the following data obtained on one subject:

Natural drift	0.000770 div. per sec (43-min observation)
Subject reading	0.000852 " " " (40-min observation)
Natural drift	0.000750 " " " (44-min observation)
Average natural drift	0.000760 " " "
Net rate from subject	0.000092 " " "

This subject was considered to contain 0.61 μg radium. On another subject, the two natural drift (i.e., background) measurements were 7.01×10^{-4} and 6.99×10^{-4} div. s^{-1}, and the net rate from the subject was 1.97×10^{-4} div. s^{-1}, corresponding to 1.30 μg radium. It can be seen that with a calibration factor of 1.51×10^{-4} div. s^{-1} for 1 μg of radium, this system was apparently almost twice as sensitive as that of Schlundt et al. (1929), where the calibration factor was 8.57×10^{-5} div. s^{-1} for 1 μg of radium (but see comments below after discussion of Evans's metre-arc method). As already noted, the data of Schlundt et al. in Table 2 show considerable variability in the natural drift of the electroscope; Ives et al. gave no quantitative data on this, but they noted: "The procedure of taking readings depended on the variability of the natural drift of the electroscope."

The calibration method used by these workers is of some interest. They injected 11 radon seeds of known strength into the body of a cadaver in such a way that the activity in each body part was proportional to the mass of the skeleton in that part. The locations of the seeds were "in all cases at the surfaces of the bones." Observations of the rate of drift of the electroscope fibres was then made with the cadaver arranged in the chair in the same way as the living subjects under test. Ten observations were made on three different days, yielding the average calibration value for the rate of drift of the electroscope of 1.51 (range 1.46-1.57) $\times 10^{-4}$ div. s^{-1} for 1 μg of radium. From a series of observations on 31 control subjects with no exposure to radium, Ives et al. determined the standard deviation for their measurements as 0.42 μg. This may be taken as their limit of detection.

Ives et al. also tested the exhaled breath for the presence of both radon (i.e., ^{222}Rn) and thoron (i.e., ^{220}Rn). By recording the drift of the Lind aluminium foil electroscope every 54.5 s, they were able to demonstrate unequivocally the presence of thoron in 18 of 40 subjects tested; for 10 of these, they plotted the net rate of drift as a function of time and the agreement between the experimental observations and the theoretical decay curves was very good.

The second of the two papers of the 1930s presented major improvements in technique, sensitivity, and precision. Evans (1937) replaced the electroscope that had been used by previous workers, by a Geiger-Müller tube with a direct reading counting-rate meter. This detector was more than an order of magnitude more sensitive than its predecessors. Furthermore, Evans devised a method of calibration that circumvented the need for cadavers, phantoms, or post-

mortem analyses of bones. In essence, the method consisted of measurements of the counting-rate from the subject lying on a light wooden examining table (to minimize scattering of the gamma rays) with the body bent in an arc of 1-m radius of curvature, facing alternately towards and away from the detector; the gamma-ray detector was at the centre of curvature of the arc. The counting-rates were combined with measurements of standardized sources of radium also placed on the table, 1 m from the detector, with and without the subject in place, to determine the attenuation of radium gamma rays by the body. The body's content of non-emanating radium, R, was determined from the following equation:

$$R = A\,(S_3S_4N_1N_2/N_3\,N_4)^{0.5},$$

where

$$A = \left(1 - \frac{d}{u} + \frac{x\,(d-x)}{u^2}\right)^{0.5n}$$

$$\approx 1 - \frac{nd}{2u}$$

In this expression, N_1 and N_2 are the counting rates observed from the subject; N_3 is the additional counting rate from a radium source of strength S_3 μg placed behind the subject (in practice, N_3 may be determined by placing three or four sources of equal strengths behind the subject's head, shoulders, back and thighs); N_4 is the counting rate from a bare radium source of strength S_4 μg at distance u (i.e., 1 m) from the detector to the back of the subject; x is the effective depth of a point source in the body that gives the same response as the distributed source in the body; d is the mean of the thicknesses of the subject at the shoulders, hips, and thighs; and n is the exponent in what is commonly called the inverse square law, but which may be as low as 1.51-1.7 unless precautions are taken to eliminate scattered radiation. In the equation for A, there is no term involving attenuation and the correction term involving x is negligible.

Evans's method gave the first reliable estimates of radium content but it suffered from the same drawback as all its predecessors, viz., no means to distinguish between ^{226}Ra and ^{228}Ra, a point of some importance in the follow-up of former dial painters in plants where ^{228}Ra had been much used for some period of time (it was to be almost 20 y before this disadvantage could be overcome). Nevertheless, Evans' contribution was outstanding; he reduced the limit of detection for the assay of radium *in vivo* to about 0.2 μg of radium, expressed as the corresponding activity (7.4 kBq) of the daughter-product, ^{214}Bi (RaC). Evans (1943) commented that this "corresponded to about 0.4 micrograms of total radium burden within the body. This value is four times the present accepted tolerance." However, it should be pointed out that at that time much smaller quantities of the body's content of emanating radium could be quantitated by determination of the exhalation rate of radon. However, the determination of the gamma-ray activity is always to be preferred for the assay of radium *in vivo*.

It is to be noted that Evans (1937) published some data that contradict the assertion above that the method of Ives et al. (1933) was almost twice as sensitive as that of Schlundt et al. (1929). In fact, Evans showed that the geometry of Schlundt et al. (electroscope behind the spine) was about 1.7 times as sensitive as that of Ives et al. (electroscope in front of the lower sternum).

THE CONTRIBUTIONS OF VICTOR HESS IN THE 1940s

In 1938, V.F. Hess, who was awarded the 1936 Nobel Prize in Physics, left Austria and became professor of physics at Fordham University in New York City. He had had a thumb amputated in 1934 as a result of radiation damage and had become concerned about and interested in radiation protection. Hess and McNiff (1947) described a technique that permitted the quantitation of radium *in vivo* by the gamma-ray method at a substantially lower level than was previously possible. As detector they used a cylindrical ionization chamber 40.6 cm high by 20.3 cm in diameter (giving it a volume of 13 L), with a 2.5-mm thick brass wall and filled with dry nitrogen at atmospheric pressure. A Lindemann electrometer was connected to the inner electrode of the ionization chamber but instead of observing the drift of the needle per unit time, Hess and McNiff used the electrometer as a null instrument. They connected the guard ring with a potentiometer circuit to add a compensating potential to it, and adjusted the potentiometer to keep the electrometer needle at zero. The compensating voltage was read with a precision voltmeter at intervals of a few minutes. Division of this voltage by the time elapsed and multiplication by a predetermined calibration factor gave I, the ionization produced in the chamber, in ion pairs $cm^{-3}\ s^{-1}$. Indoors, the average value of the background ionization of 7-9 I was made up of ~ 5 I from local gamma rays, ~ 2 I from cosmic rays, and ~ 1 I from alpha-

particles arising from contamination in the inner wall of the ionization chamber. This background ionization was rather stable, the changes observed for each 15-min period of observation usually being less than ± 0.1 I.

Hess and McNiff's choice of geometrical arrangement of detector and subject, and their justification of it, seem a little odd. They commented,

"Since most of the radium is lodged in the thorax of persons with incipient radium poisoning, it was thought best always to have the patient sitting upright at a standard distance (35 cm) from the vertical axis of the ionization vessel, first with his back and then with his chest toward the chamber."

The statement about radium being concentrated in the thorax is curious indeed. Ten years earlier, Evans (1937) had written, "studies of radium poisoning demand accurate quantitative knowledge of the total amount of radium present in the *patient's skeleton.*" (Emphasis added.) Despite this, Hess and McNiff calibrated their measurement geometry with a water-filled tank in the shape of the human thorax, with a small source of radium placed in turn in each of 57 positions within the tank. The average of 2.90 I for 1 μg radium was used as the calibration factor for measurements *in vivo*. It is important to note that although Hess and McNiff studied the absorption of the radium gamma rays in the water of their phantom, they made no allowance for the lower absorption that would have resulted from the lower density of the lungs in the case of a living subject if the radium were indeed confined to the thorax.

Instead of following Evans' recommendation (1937) of making background measurements with a noncontaminated person sitting in the chair to allow for the absorption of local gamma-radiation, Hess and McNiff determined the average absorption effect for persons of various weights, and obtained a value of 0.37 I. Table 3 shows their example of the application of this correction and indicates the kind of precision they obtained. They used the calibration factor given above to deduce a body content of 0.11 μg; this was of course only the nonemanating part, and could be more accurately described as 4 kBq ^{214}Bi.

These workers went one step further, in that they built what amounted to the first "shadow shield." By erecting a 10.5-cm thick wall of iron bars, 110 cm high and 82 cm wide behind the subject's chair, they not only eliminated the local gamma-ray absorption effect of the body, but also reduced the background ionization by about 2 I.

Like Evans (1937) and Schlundt et al. (1929), Hess and McNiff emphasized the need for a subject to bathe and wash the hair, and to wear special clothing. They also forbade subjects from entering a radium plant in the 12 h before the tests. They summarized their conclusion as follows:

". . . we may say that our gamma-ray method *allows the detection of the tolerance dose of radium in the body (0.1 microgram) with sufficient accuracy within one hour, and of even smaller quantities if several tests are made in succession.*"

Hess and McNiff (1947) also described in detail their technique for routine measurements of radon in breath samples. They used two 4-L ionization cham-

Table 3. Example of interpretation of observed ionization from radium contaminated subject, with corrections for background and subject absorption effect (Hess and McNiff 1947).

Background observed (ground floor)	7.27 ± 0.1 I
Background corrected for the case of an inactive patient sitting in front of the apparatus (0.37 I)	6.90 ± 0.1 I
Patient with radium poisoning in front of apparatus -- observed	7.22 ± 0.1 I
Actual gamma-ray ionization due to radium in the body	0.32 ± 0.14 I

bers in a differential arrangement, the two having equal voltages but of opposite signs applied to their central electrodes. A Lindemann electrometer was used to measure the differential ionization current. Since the main objective in determining radon in the breath was whether or not the subject under test contained more or less than the "tolerance" limit, which was considered to correspond to 10^{-12} curie/L (i.e., 37 Bq m^{-3}), extremely high sensitivity was not needed. Hess and McNiff showed that 15-min observations were adequate once the radon (in the chamber with the positive voltage) had had enough time for its short-lived daughter-products to be collected on the central electrode, and to grow into equilibrium (3 h). Using this technique, Hess reported thousands of concentrations of radon in breath samples from people employed in the radium industry (chemists, dial workers, etc.) during the years 1945 to about 1965 (Argonne National Laboratory).

RAPID DEVELOPMENTS IN THE 1950s

A major improvement in the sensitivity for the determination of radium *in vivo* was reported in 1951, only four years after the appearance of Hess and McNiff's paper. Sievert (1951) described equipment which, for the first time, permitted the detection of the gamma rays from the naturally radioactive potassium content of the human body. He assembled 10 ionization chambers, 0.22 m in diameter by 1.25 m long and filled with nitrogen at 2.45 MPa, in a ring 0.5 m in diameter in which the subject to be tested lay on a curved aluminium plate. The equipment was placed below ground and shielded somewhat incompletely by concrete and by tanks filled with water from the River Thames in England, because the local water supply was considered to have too high a radium content (Sievert 1952). The inadequacy of the shielding against local gamma radiation was demonstrated by a reduction in the ionization when a water-filled dummy was placed in the measuring location; the magnitude of the reduction depended on the weight of the dummy.

The anode current of the electrometer tube was balanced with a battery and potentiometer, and when the ionization changed, the out-of-balance current was recorded on photographic paper. Figure 1, taken from the paper by Sievert (1951) was described as a typical record, showing from left to right: background; a 4-h calibration observation with 0.1 μg radium; another background; the absorption effect for a person with a radium burden (quantity unspecified); a third background observation; and finally a background with a 50-kg water-filled dummy. It can be seen that the reduction in the ionization due to absorption of local gamma-radiation by the latter amounted to a little over half of the increased ionization due to the radium standard. This made determination of potassium contents quite difficult and time-consuming. It can also be seen that there was a slow but steady decrease in the background over the period of these observations. This was almost certainly due to a change in the intensity of the cosmic radiation, presumably associated with a change in the barometric pressure.

Sievert noted that "under favourable conditions, the standard error can be reduced to ± 0.002 μg Ra eqv. with 6-8 observations for persons with no radioac-

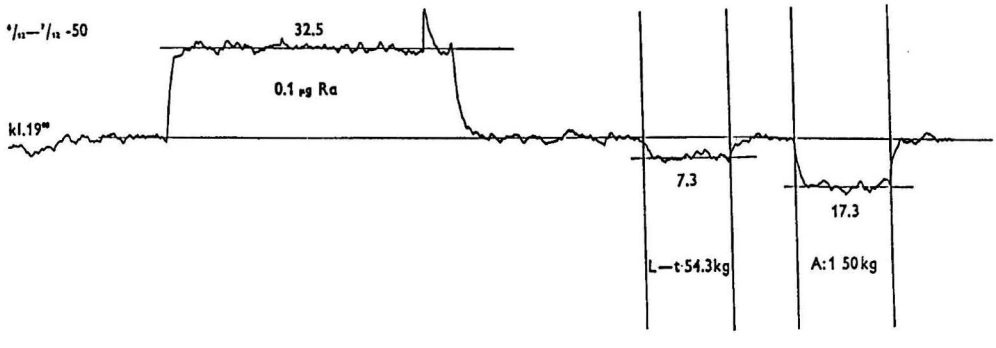


Fig. 1. A record of ionization current, showing from left to right: background; a 4-h calibration observation with 0.1 g radium; another background; the absorption effect for a person with a radium burden (quantity unspecified); a third background observation; a background with a 50-kg water-filled dummy; and finally a short background (Sievert 1951). The effect of a cosmic-ray shower is seen near the end of the radium calibration observation.

tive contamination." The limit of detection for a single observation may be taken as 0.015 μg radium, expressed as ^{214}Bi, i.e., 550 Bq.

In a later development, Sievert (1956) installed two similar equipments in a laboratory built in the side of a mountain in the outskirts of Stockholm, so that there was at least 50 m of granite for shielding of cosmic radiation. To reduce environmental gamma radiation, the walls of the laboratory were lined with water-filled tanks, each 4 × 2 × 1 m, yielding a room 4 × 8 × 2.5 m, shielded in all directions by at least 1 m of water. The ionization chambers were increased in size to 2 m long by 0.4 m in diameter, and each apparatus had 12 chambers. The natural radiation in the laboratory was about 1% of that in an ordinary Swedish house, being about 0.07 I (ion pairs cm^{-3} s^{-1}). This may be compared with Hess's best value of about 6 I, almost 90 times higher, and it is a dramatic illustration of the enormous advantages to be gained by very large amounts of shielding. Cosmic radiation still contributed significantly, as evidenced by the observation of sudden increases in the ionization due to showers. The absorption of environmental gamma radiation by the presence of the human body was essentially zero, and even radiation from school children could be readily determined. By attributing to ^{40}K all the radiation observed, Sievert calculated the potassium content of many persons over a range of ages and a range of body weights. His results were in essential agreement with other contemporary published data.

The emphasis of measurement changed with Sievert's two publications. Initially he had been attempting to determine the natural radium content of the human body, to resolve a major discrepancy (a factor of about 100) between the results of two sets of analyses of cremation ash for radium (Krebs 1942; Hursh and Gates 1950). He was unsuccessful in this endeavour, but his preliminary results were the incentive to build and equip the underground laboratory. With the availability of equipment that could quantitate the radiation from ^{40}K, the subject now broadened to include assay of potassium (of interest to physiologists and dietitians), as well as numerous other radionuclides of importance for radiological protection. We shall review subsequent developments with an attempt to keep the emphasis on radium.

Two years after Sievert's first publication (1951), two papers appeared in the issue of *Nature* for September 19, 1953, on the subject of body radioactivity measurement not necessarily related to radium poisoning or metabolism. The first of these (appearing on pages 519-521) (Burch and Spiers 1953) was actually received by the journal a month later than the second (on pages 521-523) (Reines et al. 1953). Burch and Spiers used four high- pressure ionization chambers, 0.29 m in diameter and 1.98 m long, filled with commercial nitrogen at 1.37 MPa but instead of countering the electrometer voltage with a variable potential, they used another set of seven ionization ("backing") chambers in much the same way as Hess and McNiff did for their radon determinations (Fig. 2). The differential ionization current was determined with the aid of a vibrating reed electrometer. By adjusting (doubling) the pressure of the nitrogen in the backing chambers, they were able to reduce substantially the effects on the differential ionization current of changes in the barometric pressure. There was still a quite considerable absorption of local gamma radiation, probably a consequence of imperfect shielding. It was determined by ensuring that background measurements were always made with a water-filled phantom of appropriate size. Table 4 shows results for three subjects, a man with no known exposure to radioactivity, one exposed to ^{60}Co, and a former mesothorium (^{228}Ra) worker. It is seen that the magnitude of the background absorption correction was almost as large as the emissive effect (first subject). The activity in excess of the expected potassium content was considered to be 555 Bq ^{60}Co and 4.44 kBq ^{228}Ra, for the two contaminated subjects, respectively. The equipment response was calibrated for potassium from the response to a small known quantity of ^{42}K 4-8 h after oral administration to the subject, and comparison with the responses to known amounts of ^{42}K and potassium chloride in a phantom. For other radionuclides, corrections were calculated from the attenuation coefficients for different quantum energies. A value of 2 ng radium with all daughter-products in equilibrium, was quoted for the probable error for a 2-h observation of body radioactivity, i.e., a limit of detection of about 330 Bq ^{214}Bi.

One other development of the same type may be mentioned here, although it is out of the strictly chronological sequence that has been followed so far. In many respects, the equipment used by Rundo (1955) was similar to that of Burch and Spiers (1953). Four high-pressure ionization chambers filled with argon at 20 atmospheres were used for detecting the radiation emitted by the subject; they were 160 mm in diameter by 0.18 m long, with collector electrodes of 6.4-mm diameter rods. Four similar chambers were connected differentially with them, and separated from them by 0.6 m of water shielding, to reduce the background current. The differential ionization cur-

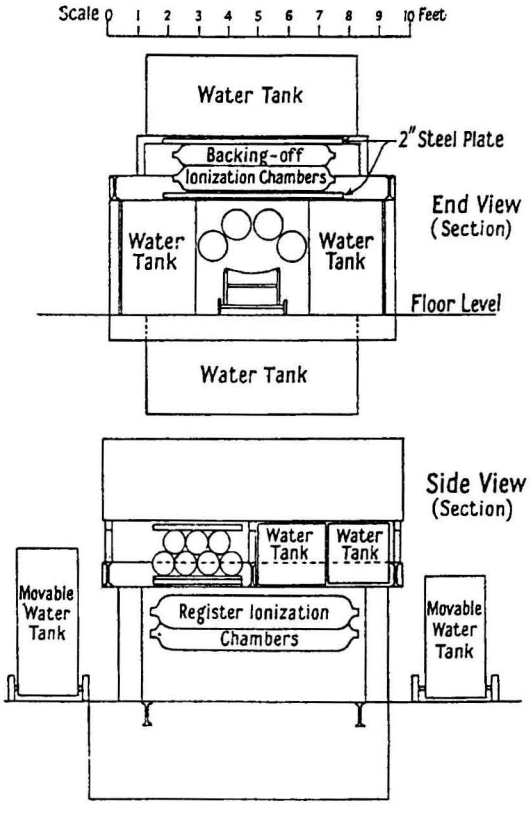


Fig. 2. Diagram of the first apparatus that used high-pressure ionization chambers for registering the response from a human, and also for reducing ("backing-off") the background (Burch and Spiers 1953).

Table 4. Examples of radioactivity measurements in three individuals with different exposure histories, showing the ability to detect the naturally radioactive potassium (Burch and Spiers 1953). Response and absorption units are 10^{-15} amp.

Exposure	Weight, kg	Response above Background	Background Absorption Correction	Effective Response	Potassium Equivalent, g
None	68.0	1.2 ± 1.2	+ 6.7	7.9 ± 1.2	150 ± 23
Cobalt-60	55.7	17.7 ± 1.4	+ 6.3	24.0 ± 1.4	413 ± 24
Radium-228	70.8	79.6 ± 1.2	+ 6.8	86.4 ± 1.2	1665 ± 23

rent from the chambers developed a potential across a stable resistor (selectable in the range 10^8 to 10^{12} ohm), and this was measured with a vibrating reed electrometer. The ionization chambers were mounted in a steel tank, 3.78 × 2.53 × 3.72 m high, filled with water so that there was a minimum of 0.6 m shielding for each set of chambers in any direction. The shielding above the "backing" chambers (arranged in pairs above one another) was 0.9 m, while above the two measuring chambers immediately below them there was 1.5 m of water. The two other measuring chambers had 2.1 m of water immediately above them. Because of this, background absorption by the body was not detectable, but because of the different shielding of the two sets of chambers there was a marked effect of changes in the barometric pressure on the differential ionization current, e.g., $(1^{-6}) \times 10^{-14}$ A for pressures in the range 96.4-103.7 kPa.

This apparatus was designed for the study of *Thorotrast* poisoning so high sensitivity was not a primary requirement. Nevertheless, it proved to have a limit of detection a little lower than that reported by Burch and Spiers (1953), viz. a standard error for a 2-h observation of ± 30 g potassium, or ± 2 ng of ^{226}Ra in equilibrium with its daughter-products, corresponding to a limit of detection of about 220 Bq ^{214}Bi.

The second paper in the issue of *Nature* for 19 September 1953 described a radical departure from the use of ionization chambers that had held sway in the present context for so long. Reines et al. (1953) described the first use of large-volume liquid scintillation detectors for the determination of radioactivity in the human body. Unlike the apparatus described in the preceding reports, the equipment they used was not designed specifically for the determination of body radioactivity, but the application suggested itself in the course of development of a large detector for neutrinos (Cowan et al. 1953). Nevertheless, the experiments reported must be regarded as a landmark.

The equipment used by Reines et al. consisted of an annular liquid scintillator, 0.76 m high, with an outer diameter of 0.76 m and a thickness of 0.13 m (volume ~0.2 m^3), and viewed by 45 photomultipliers (Fig. 3). The whole was shielded by 127 mm of lead except for a 0.5-m aperture at the top. By adopting what can best be described as an upright "foetal position," a human body was entirely within the detector. This arrangement was the first approach to counting radioactivity in people in almost 4π geometry.

Calibrations were made in two ways. In the first, about 45 kg of water was placed in a "shaped plastic bag" and counting rates were determined before and after the addition of 450 g of potassium chloride. The net counting rate from the potassium was 125 s^{-1}, so with an expected potassium content of about 150 g, a rate of 78 counts s^{-1} would be expected from a 70-kg man. The data in Table 5 indicate that potassium was indeed the only significant gamma-ray emitter in seven of the nine subjects in the table. In the

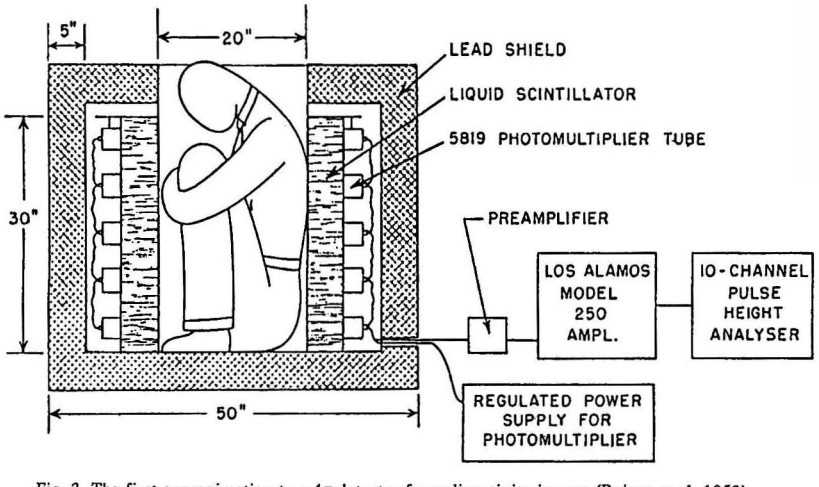


Fig. 3. The first approximation to a 4π detector for radioactivity in man (Reines et al. 1953).

Table 5. "Summary of results for experiments with human beings" as reported by Reines et al. (1953)

Subject	Sex	Weight, kg	Counts s^{-1} above Background
W.L.	M	59.1	57
D.F.	F	47.7	30
W.B.	M	65.9	801
P.M.	M	72.7	784
L.B.	M	63.6	42
E.A.	M	61.4	75
P.H.	M	63.6	80
D.H.	F	56.8	57
G.L.	F	56.8	63
Background			837

case of subject W. B., most of the additional counting rate was found to be attributable to external contamination in his clothing.

A simple calibration was also made for radium; subject W. L. (Table 5) was counted while holding a small radium source against his stomach so that it was entirely shielded by his body. The efficiency was about 162 counts s^{-1} for 1 kBq radium with daughter-products in equilibrium, and the fluctuations in the background, which were greater than statistical variations, corresponded to about 37 Bq ^{214}Bi. In the absence of potassium, the limit of detection would therefore be about 100 Bq, but because of uncertainties in the predicted potassium content, the limit was greater, and probably about the same as reported by Burch and Spiers (1953), i.e., 330 Bq ^{214}Bi.

In a later development at Los Alamos (Anderson et al. 1956), a so-called "human counter" was built specifically for the purpose of determining radioactivity in people, although not necessarily radium. Like its pioneering predecessor, it used a large liquid scintillator but the rather poor spectrometric properties precluded the determination of very low levels of radium because of the response from the body's potassium content. It is therefore not possible to quote a limit of detection for radium, but the average potassium content of an adult male could be determined with a statistical accuracy of no more than about ± 20 g (corresponding to about ± 14% of the body content) with two observations of 100 s each. The authors noted that a lead door was needed to eliminate the background reduction caused by absorption of soft local gamma-radiation in the body of the subject under test, although the magnitude of this was not reported.

The enormous potential for identification of gamma-ray emitters *in vivo* offered by the spectrometric capabilities of the new detectors was of course recognized by all these workers, but the large liquid scintillators had only limited energy resolution. On the other hand, the increasing availability, in the early to mid-1950s, of large crystals of thallium-activated sodium iodide with its superior energy resolution, was exploited to the full by the workers in the field of body radioactivity measurement. Because of their work at Argonne National Laboratory on radium poisoning, Marinelli et al. (1955a) were the pioneers in this application, although their stated objective was a method capable of measuring the natural levels of radium (10^{-10} g, or if 30% of the radon is retained, about 1 Bq of ^{214}Bi).

In their first report, these workers described the use of an NaI(Tl) crystal, 57 mm long by 38 mm in diameter with minimal shielding (no more than 13 mm of lead on the floor and 6 mm on the walls), and a single-channel analyzer. They used the metre-arc method of Evans (1937) for the subject-detector geometry, but because the efficiency of the NaI(Tl) detector increases with decreasing photon energy (the opposite of the Geiger-Müller counter's behaviour), the attenuation curves showed marked deviations from exponential, resembling depth dose curves. To allow for this, they had to use a fictitious attenuation coefficient and an empirical scattering correction factor that was dependent on the average depth of the source within the body (assumed) or in the calibration phantom (known). To increase the counting efficiency, Marinelli et al. adopted Evans' suggestion of placing the person in a chair, with the detector 100 mm from the twelfth thoracic vertebral body. With this simple arrangement, they were able to report an overall average standard error of $\pm 9 \times 10^{-10}$ µg of radium (33 Bq of ^{214}Bi) for a 2-h observation Marinelli et al. (1955b). However, it should be noted that a realistic limit of detection cannot be derived from this because it is based on the assumption that all the observed response was from radium (more accurately, radon) daughter-products, whereas of course there is an unavoidable and irreducible contribution from ^{40}K at photon energies up to a little above 1.46 MeV. Furthermore, it made

no allowance for effects of absorption by the body of local gamma or soft cosmic rays.

These problems were all overcome by the use of substantial shields (200 mm of steel, 100 mm of lead, or the equivalent) that were large enough to contain both the subject under test and an operator, as well as the detector(s) and supporting mounts. Two papers describing slightly different approaches to this were presented at the Second United Nations Conference on the Peaceful Uses of Atomic Energy. In the first of these (Rundo 1958), the emphasis was on health physics aspects rather than research. The equipment used four NaI(Tl) scintillation detectors, 110 mm in diameter by 51 mm thick, and a multichannel pulse height analyzer for gamma-ray spectrometry; the detectors were disposed about the supine subject, two above and two below. Despite the substantial shielding (100 mm of lead), there was still an effect on the background of placing a water-filled phantom on the stretcher, but spectrometry showed that there was a decrease at energies above 0.25 MeV and an increase below that energy. At the time, these effects were attributed entirely to scattering and absorption of radiation from radioactivity in the detectors themselves, but in retrospect it seems likely that much, if not all, of the effect was due to absorption and scattering of gamma rays produced in the shield by cosmic rays. The standard error (statistical only) for a 50-min measurement of potassium in a man and a long observation of background, was ± 7 g, i.e., ± 5% for an average content of 140 g. For radium, a limit of detection was quoted of 8 ng (defined as three times the standard deviation of the background for photon energies greater than 1.46 MeV — the energy of the gamma rays emitted by ^{40}K) of ^{226}Ra with 30% retention of the radon and its daughters, i.e., 90 Bq of ^{214}Bi.

The second paper at the U.N. Conference (Miller 1958) described the use of a single NaI(Tl) scintillation detector, 200 mm in diameter by 100 mm thick; this detector contained about 60% more material than did the four detectors of the preceding paper (Rundo 1958), so one would expect somewhat better performance. Indeed the assertion was made that a body content of 10^{-9} g of radium could be measured quickly, although there was no further discussion of this. The single detector was suspended above the subject, who sat in a chair that was tilted back so that movement of any part of the body was opposed by gravity. The statistical standard error for the measurement of potassium was quoted as less than 5% with a 10-min observation, quite similar to that obtained in a 50-min observation with a smaller detector system (Rundo 1958). No mention was made of an effect on the background of the presence of a phantom in the measuring position; the absence of such an effect might have been a consequence of the use of steel (200 mm thickness) instead of lead as a shielding material.

Techniques such as these were widely copied over the next few years, although not primarily for the investigation of body contents of radium, with one notable exception (Wenger and Miller 1962). Shields were usually of steel or lead, although chalk and water were also used (International Atomic Energy Agency 1964, 1970). The geometrical arrangements were commonly metre-arc or similar or copies of the Argonne National Laboratory tilting chair for single detector systems, or with the subject supine on a stretcher for multiple detector equipment. Calibration techniques of various kinds were used — arc geometry (Evans 1937; Marinelli et al. 1955a; Lillegraven and Rundo 1965), phantom and gamma-ray spectrum matching, water-filled phantoms with either "point" or distributed sources (Rundo 1958), or realistic phantoms of differing degrees of complexity (Rundo 1957; Alderson Research Laboratories 1962). In any one case, the arrangement chosen was dictated by the specific requirements of the user.

Our history of the determination of radium in man is nearly complete, but there are three other developments worth mentioning. The first of these (Miller and Steingraber 1958, 1959) was an attempt to take advantage of the coincidences between some of the gamma rays emitted by ^{214}Bi. Thus, the 0.609-MeV gamma ray is emitted in coincidence with the 0.77-Mev and also with the 1.12-MeV photons, and if a source of ^{226}Ra is placed between two NaI(Tl) detectors, a sum coincidence spectrum shows peaks at 1.38 (= 0.609 + 0.77) MeV and 1.73 (= 0.609 + 1.12) MeV with an extremely low background contribution. Unfortunately, the technique is of limited application in the determination of radium *in vivo* because the detector pair can only "view" a very small part of the body.

Secondly, all the determinations of radium in man by the direct observation of gamma-ray emissions that have been discussed so far suffer from the same shortcoming, viz., the need to determine or assume the fraction of radon retained in the body, because this is what is quantitated. Miller and Steingraber (1958, 1959) reported attempts to use the 186-keV gamma ray emitted by ^{226}Ra itself in about 6% of its disintegrations to make direct determinations of the radium content without assumptions about, or measurement of the radon retention. Unfortunately, the

energy of this gamma ray is virtually the same as that of the backscattered radiation from the much more intense 0.609-MeV photons; however, if a collimated detector is placed near a region of the body (e.g., knee, elbow, ankle) that contains a large mass of bone with minimum amounts of scattering tissue, it seemed to be possible to quantitate the radium/retained radon ratio directly. Despite a promising start, this development seemed to lead no further. The present-day availability of quite large solid-state detectors with their greatly superior energy resolution, should make this an entirely practicable method although the limit of detection may be relatively high because of the small fraction of gamma rays that can be intercepted by the detector.

The third development worth mentioning involved the use of the low-energy photons (0.25, 0.29, and 0.35 MeV) from ^{214}Pb (radium B). It was shown (Bengtsson 1968) that this resulted in a lower limit of detection than did the use of the gamma rays with energy greater than 1.46 Mev (^{40}K), but there was a limitation in the determination of the contributions from ^{40}K and the not inconsiderable amounts of ^{137}Cs that were present *in vivo* at that time. As a result, the limit of detection (3 standard deviations) for the low energy region was 37 Bq compared to 107 Bq for the high-energy region, based only on counting statistics; the former was increased to 59 Bq because of uncertainty in the prediction of the "subject background" counting rates. The improvement was thus not a dramatic one.

Lastly, although it has nothing to do with radium in man, we may demonstrate the capability of the very large organic scintillators by mentioning that the efficiency of equipment that used three large plastic scintillators (508 mm long × 254 mm wide × 165 mm thick) was such that the standard error for a 15-min observation of body potassium was just over ± 1% (Burch et al. 1962). This is substantially better than the performance of the first Los Alamos "human counter," where nonstatistical background fluctuations limited the precision with which potassium could be determined to about ± 14% (Anderson et al. 1956). However, subsequent developments

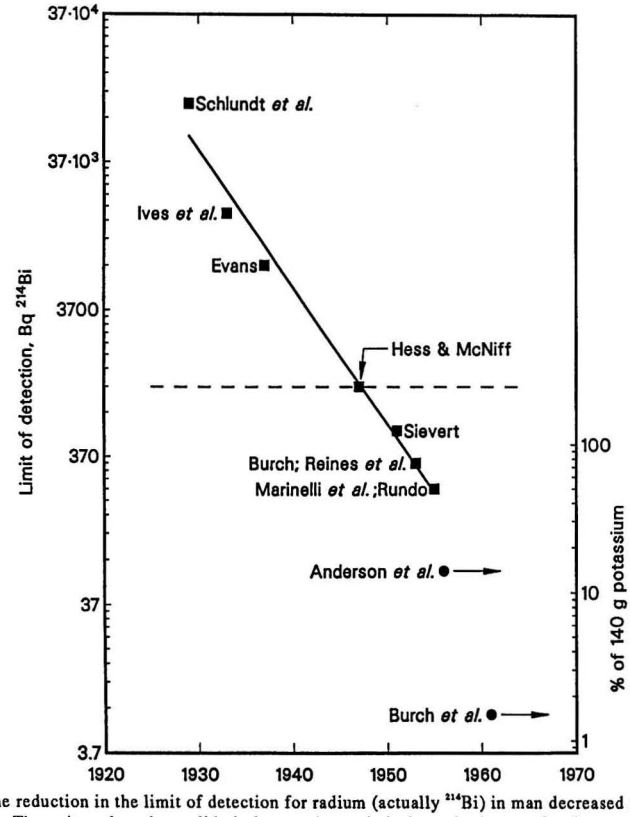


Fig. 4. Showing how the reduction in the limit of detection for radium (actually ^{214}Bi) in man decreased roughly exponentially from 1929 to the mid-1950s. The points plotted as solid circles are the statistical standard errors for the measurement of potassium and their ordinate values are to be read on the right-hand scale.

with the large liquid scintillators resulted in a second "human counter" which permitted the determination of the body's potassium content with a statistical precision of ± 2% (Van Dilla and Anderson 1962).

The limits of detection for the various methods described in this paper are plotted in Fig. 4. It can be seen that the straight line is a reasonable fit to the data, indicating a reduction in the limit of detection from 1929 to the mid-1950s with a half-time of a little more than 3 y. Perhaps this is a reasonable rate of improvement for a technology over that particular time period.

Acknowledgment — The work was supported by the U.S. Department of Energy, Office of Health and Environmental Research, under Contract W-31-109-ENG-38.

REFERENCES

Alderson Research Laboratories, Inc. Alderson Organ-Scanning Phantom. Technical Bulletin No. 41 (1962).

Anderson, E.C.; Schuch; R.L.; Perrings, J.D.; Langham, W.H. The Los Alamos Human Counter. Nucleonics 14: 26-29; 1956.

Argonne National Laboratory, Records of Radium Worker Studies, unpublished.

Bengtsson, L.G. Human radium assay at nanocurie levels using external counting of low energy photons. Acta Radiol. 7 :148-158; 1968.

Burch, P.R.J.; Spiers, F.W. Measurement of the γ-radiation from the human body. Nature 172: 519-521; 1953.

Burch, P.R.J.; Hughes, D.; Iinuma, T.A.; Overton, T.R.; Appleby, D.B. The construction and performance of a three-unit plastic scintillator whole-body counter. In: Proc. symposium on whole-body counting. Vienna, International Atomic Energy Agency; 1962: 59-69. Available from: IAEA, Vienna.

Cowan, C.L.; Reines, F.; Harrison, F.B.; Anderson, E.C.; Hayes, F.N. Large liquid scintillation detectors. Phys. Rev. 90: 493-494; 1953.

Evans, R.D. Radium poisoning II. The quantitative determination of the radium content and radium elimination rate of living persons. Amer. J. Roentgenol. Rad. Ther. 37: 368-378; 1937.

Evans, R.D. Protection of radium dial workers and radiologists from injury by radium. J. Indust. Hyg. Toxicol. 25: 253-269; 1943.

Flinn, F.B. Some precautions to be taken when making tests for radioactivity in the living body. Amer. J. Roentgenol. Rad. Ther. 22: 554-556; 1929.

Ives, J.E.; Knowles, F.L.; Britten, R.H. Health aspects of radium dial painting. III. Measurements of radioactivity in workers. J. Indust. Hyg. 15: 433-446; 1933.

Hess, V.F.; McNiff, W.T. Quantitative determination of the radium content of the human body and of the radon content of breath samples for the prevention and control of radium poisoning in persons employed in the radium industry. Amer. J. Roentgenol. Rad. Ther. 57: 91-102; 1947.

Hess, V.F. The Discovery of cosmic radiation. "Thought" (Fordham University Quarterly) 15: 225-236; 1940.

Hoffman, F.L. Radium (Mesothorium) necrosis. J. Amer. Med. Assoc. 85: 961-965; 1925.

Hursh, J.B.; Gates, A.A. Body radium content of individuals with no known occupational exposure. Nucleonics 7: 46-59; 1950.International Atomic Energy Agency. Directory of whole-body radioactivity monitors. (Vienna, International Atomic Energy Agency) First edition 1964; Second edition 1970.

Krebs, A.T. Untersuchungen zum Problem der Radiumvergiftung; der Gesamtradiumgehalt des menschlichen Organismus. Strahlentherapie 72: 164-169; 1942.

Lillegraven, A.L.; Rundo, J. Systematic arc calibration method for body radioactivity measurements. Acta Radiol. 3: 369-383; 1965.

Maletskos, C.J.; Keane, A.T.; Telles, N.C.; Evans, R.D. Retention and absorption of ^{224}Ra and ^{234}Th and some dosimetric considerations of ^{224}Ra in human beings. In: Mays, C.W.; Jee, W.S.S.; Lloyd, R.D.; Stover, B.J.; Dougherty, G.H.; Taylor, G.N. eds. Delayed effects of bone-seeking radionuclides. Salt Lake City, UT: University of Utah Press; 1969: 29-49.

Marinelli, L.D.; Miller, C.E.; Gustafson, P.F.; Rowland, R.E. The quantitative determination of gamma-ray emitting elements in living persons. Amer. J. Roentgenol. Rad. Ther. Nucl. Med. 73:661-671; 1955a.

Marinelli, L.D.; Miller, C.E.; Rowland, R.E.; Rose, J.E. The measurement *in vivo* of Ra gamma-ray activities lower than K^{40} levels existing in the human body. Argonne National Laboratory Report ANL-5378; 1955: 27-29; Radiology 64: 116; 1955b.

Marshall, J.H.; Lloyd, E.L.; Rundo, J.; Liniecki, J.; Marotti, G.; Mays, C.W.; Sissons, H.A.; Snyder, W.S. Alkaline earth metabolism in adult man (ICRP Publication 20). Health Phys. 24: 125-221; 1973.

Martland, H.S.; Conlon, P.; Knef, J.P. Some unrecognized dangers in the use and handling of radioactive substances. J. Amer. Med. Assoc. 85: 1769-1776; 1925.

Miller, C.E. Low-intensity spectrometry of the gamma radiation emitted by human beings. In: Proc. 2nd int. conf. peaceful uses of atomic energy; 23: 113-122; 1958. Available from: United Nations, Geneva.

Miller, C.E.; Steingraber, O.J. In: Argonne National Laboratory Report ANL-5919; 1958: 67-70; Rad. Res. 11:455; 1959.

Norris, W.P.; Speckman, T.W.; Gustafson, P.F. Studies of the metabolism of radium in man. Amer. J. Roentgenol. Rad. Ther. Nucl. Med. 73: 785-802; 1955.

Reines, F.; Schuch, R.L.; Cowan, C.L.; Harrison, F.B.; Anderson, E.C.; Hayes, F.N. Determination of total body radioactivity using liquid scintillation detectors. Nature 172: 521-523; 1953.

Rundo, J. An apparatus for the measurement of γ-radiation from the human body. J. Sci. Inst. 32: 379-384; 1955.

Rundo, J. Body radioactivity measurement as an aid in assessing contamination by radionuclides. In: Proc. 2nd int. conf. peaceful uses of atomic energy. 23: 101-112; 1958; Prog. Nucl. Energy, I, Series XII, 1958: 286-306.

Rundo, J. The determination of the distribution of internally deposited thorium by means of studies with a realistic phantom. Acta Radiol. 47: 65-78; 1957.

Schlundt, H.; Barker, H.H.; Flinn, F.B. The detection and estimation of radium and mesothorium in living persons. I. Amer. J. Roentgenol. Rad. Ther. 21: 345-354; 1929.

Seil, H.A.; Viol, C.H.; Gordon, M.A. The elimination of soluble radium salts taken intravenously and per os. New York Med. J. 101: 896-898; 1915.

Sievert, R.M. Measurements of γ-radiation from the human body. Arkiv för Fysik 3: 317-346; 1951.

Sievert, R.M. Personal communication; 1952.

Sievert, R.M. Untersuchungen über die Gammastrahlung des menschlichen Körpers. Strahlentherapie 99: 185-195; 1956.

Van Dilla, M.A.; Anderson, E.C. Human counters using liquid scintillators. In: Proc. symposium on whole-body counting. Vienna, International Atomic Energy Agency; 1962: 41-58. Available from: IAEA, Vienna.

Wenger, P.; Miller, C.E. Recherches sur l'accumulation et la toxicité du radium et du radiostrontium dans le corps humain. II. Helv. Chim. Acta 46: 467-479; 1962.

Environment International, Vol. 19, pp. 439-447, 1993
Printed in the U.S.A. All rights reserved.

0160-4120/93 $6.00 +.00

RADON LEVELS IN A PENNSYLVANIA DIAL PAINTING FACILITY FROM MEASUREMENTS BY VICTOR HESS

James H. Stebbings

Epidemiology and Biostatistics Program, School of Public Health, The University of Illinois at Chicago, Chicago, IL 60680, U.S.A.

EI 9203-136 M (Received 4 March 1992; accepted 29 May 1993)

The U.S. Radium Corporation plant in Bloomsburg, PA, was one of two major radium dial painting facilities in the United States during the post-World War II era. Victor Hess, of the Physics Department of Fordham University, was responsible for radon breath measurements from workers at this plant. His room air sample results, over 300 measurements dating from 1949 through 1966, form a unique data base for reconstructing radium dial worker radon exposures in that era. Monday morning radon levels averaged about 150 Bq m^{-3}, well below the tolerance level of 370 Bq m^{-3}. That average is representative of radium dial painting areas of the plant. Most areas, particularly offices and technical support areas, had radon levels comparable to local residential averages. Levels in the radium laboratory areas throughout most of the study period were above the tolerance limit; the highest location averaged 603 Bq m^{-3}. Radon levels in laboratory areas were significantly reduced in the last six years of the study, but radon levels in other parts of the plant remained relatively constant. These values, because of time of collection, may modestly underestimate radon concentrations during work hours in much of the plant and significantly overestimate it in laboratory areas.

INTRODUCTION

Radon in radium workplaces has for decades been believed to present a carcinogenic hazard to the respiratory tract. The earliest published measurements (Bloomfield and Knowles 1933) were made in that context. An early study of the pre-1930 cohort of U.S. radium dial workers (Polednak et al. 1978) did not find excess lung cancer, but a later study, updating the original and incorporating additional cohorts (Stebbings et al. 1984) found an excess of lung cancer consistent with that predicted on the basis of our limited knowledge of radon and external gamma radiation levels in the workplaces.

The radium dial workers are the only known female occupational cohorts exposed to high levels of radon in the workplace. The atmosphere of the generally clean radium workplaces is more reasonably compared to domestic atmospheres than to that of hard rock mines. As the dial workers were most commonly employed early in life, in most cases for a short interval before marriage or child-rearing, these cohorts are a good analogy for females living for a limited period in a high radon house early in adult life.

Correspondence and reprint requests to: Midwest Epidemiology Associates, 1122 Windsor Dr., Shorewood, IL 60436 USA.

Stebbings et al. (1984) review in detail the available radiation exposure data for radium dial workers. A few radon, but no radon daughter, measurements have been published for early radium workplaces. A mean level of about 1850 Bq m^{-3} was reported by Bloomfield and Knowles (1933) based on nine readings in five plants around 1930. Their observed values were curiously bimodal, three values being around 370 Bq m^{-3} and four in the 2400-3000 Bq m^{-3} range. Conditions improved during the 1930s and in the early 1940s: Evans (1943) reported on 47 analyses of room air in ten plants and found most measurements below the tolerance limit of 370 Bq m^{-3}, only three being above 740 Bq m^{-3}. Duggan and Godfrey (1967) reported modal radon levels near 370 Bq m^{-3} for contemporary British luminizing facilities, with a range of observations from 370 to 3000 Bq m^{-3}. Radium dust values, when measured contemporaneously with the radon values, have been much lower. Despite the paucity of radon measurements in the scientific literature, we believe that radon levels in industrial facilities were, in fact, relatively well known to the industrial hygienists in the industry and to state agencies since the mid-1930s.

A safe working concentration of 370 Bq m^{-3} had been arrived at by Evans and Goodman (1940) and was widely accepted (NBS 1941) in the United States. Evans (1981) reviewed the history of the inception of standards for radon.

Victor Hess at Fordham University

Victor Hess came as a refugee to Fordham University in 1938. His activities seem to have been restricted to Fordham University during World War II, although there is evidence that he unsuccessfully attempted to contribute along with others of the refugee physics community to the American war effort. During World War II, firms dealing with radioactive materials were of direct interest to the Manhattan Project. This included the wholesalers and distributors of radium and related radionuclides in New York City. It is likely that he had retained personal contacts in this industry from his stint as Chief Physicist of the U.S. Radium Corporation in Orange, NJ, from 1921 to 1923. Immediately after World War II, Victor Hess was found to be active in the measurement of radium body burdens in employees of the firms in New York importing and selling (or leasing) radium for medical use and for industrial applications.

In 1947, he published his methods for determination of radium in workers (Hess and McNiff 1947). This health physics effort gradually expanded and continued into the mid-1960s. There is evidence in his correspondence that the desire to develop financial savings for retirement was an important element in the work. On the other hand, in the letters which invariably accompanied the breath radon results back to management, it is clear from Hess's tone when reporting levels above the tolerance limit in expired air (37 Bq m^{-3}), especially when they had been repeatedly above tolerance, that these were matters of serious personal concern to him. In general, management seems to have responded effectively, either by modification of work practices or the workplace, or by temporary reassignment of personnel.

From a start with a few small importers and distributors in New York, Hess's radon breath measurement activities expanded widely, even to such places as Little Rock, AK and Amarillo, TX. Many of the workplaces he studied have not been among the dial worker cohorts studied at Argonne National Laboratory. His largest and longest standing effort, however, was the new U.S. Radium dial painting facility at Bloomsburg, PA, which began operations about 1947. At this facility, he carried out thousands of breath measurements over the years 1949 to 1966, and concurrently some hundreds of room air samples from the workplace.

In the 1960s, radium was phased out in favor of tritium both in Bloomsburg and in Ottawa, IL. At Bloomsburg, there was no more radium on site by July, 1969. Victor Hess's efforts ended early in 1966.

During the two decades following World War II, approximately 1946 to 1965, no other individual seems to have contributed as much to the health physics protection of workers in the luminizing industry as Victor Hess, along with his coworkers at Fordham, William McNiff, Alfons Weber, and Bernard Dunn.

METHODS

U.S. Radium Corporation in the late 1970s voluntarily furnished Argonne National Laboratory's Center for Human Radiobiology with complete personnel records, which included individuals' health physics measurements from Victor Hess. The numerous brief lists of radon measurements (which included the room air levels along with names and expired air radon measurements for individuals measured the same week) had been filed multiple times, once each in each named individual's medical record folder. It was not economical to access them. From the files of Victor Hess in the archives of Fordham University the worksheets and transmitted lists of radon measurements in both expired and room air were obtained

by the Center for Human Radiobiology for its archives; these were used in this study.

Air samples were collected at various plant locations by U.S. Radium's health physicists, and shipped to Fordham University where they were analyzed during the next few days, with account taken, of course, of radioactive decay since time of collection. The double chamber differential method of Hess and McNiff (1947) was used. It was sensitive to radon levels of 7.4 Bq m^{-3} and greater, and precise in repeat measurements of individuals to within ±20%.

The individual variables available for each observation are the report date, building, room, date of measurement, day of week of measurement, and time of day of measurement. As room terminologies were not always consistent, and changes in process locations within the plant over the years seem to have occurred, data were recoded to consistent room identifiers. In 1984, Richard E. Toohey of ANL interviewed several former employees of the Bloomsburg plant (J. Sorber, C. Berlin, J. Powlus, L. Harmon, D. Kingsbury, and possibly J. Watts and K. Sneidman) who helped locate many radon measurements on blueprints of the plant. Even with this effort, ambiguities remain in the definition of rooms and spaces. No detailed study of the Bloomsburg cohort involving careful industrial hygiene and health physics assessment of individual job titles and assignments, with concurrent description of operations by department or room over time, has been carried out.

For the Bloomsburg plant, 320 measurements of room or control air collected between 1949 and 1966 were found. Thirteen observations remaining ambiguous as to placement are excluded from analysis. Four inactive measurements (i.e., < 3.7 Bq m^{-3}) were recoded (extrapolating the frequency distribution) as 2.59 Bq m^{-3}. Three measurements were corrected as to time or date by reference to the original calculation sheets. The final data set included 307 observations, including outdoor and control measurements. Data coding, editing, and preliminary analyses were carried out at Argonne National Laboratory, and resulted in a preliminary report (Hartmann 1986).

RESULTS

The image of the classic American watch/clock dial painting facility, where timepiece dials are received from the manufacturer, painted with radium paint, and shipped to the end-user does not fit this plant. From review of personnel records, and from an undated sales brochure in the Argonne National Laboratory radium archives (U.S. Radium Corp. undated), it is clear that a large and profitable part of the luminizing operation included nameplates, instrument dials, various scientific specialty items, some classified military items along with military items like deck markers, and, of course, watch and clock dials. Secondary products, to quote the brochure, included: luminescent materials, cathode-ray tube powder, x-ray screens, radium locators, luminous reticles, ionotron static eliminators, radioactive foils and radiation standards, neutron sources, and various processed isotopes. Americium and polonium were in use over portions of the period of operation. The manufacture of custom dials, panels, and scales using a "Lackon Process," licensed from the Sun Dial Corporation, was also a major part of the operation.

The hand painting department of the plant is pictured in Fig. 1; this photograph had been reproduced in the sales brochure cited above.

These operations were carried out in an installation composed of one large main building, a smaller one-story etching building, and several minor structures. The main building had a two-story front, was single-story behind, and had some basement areas. There also appears to have been a small third floor area at the center of the front of the building. The geometric mean radon level for the two largest buildings was 150 Bq m^{-3} (n = 246).

The number of rooms and work areas was large relative to the number of radon measurements. This report focuses primarily upon mean radon levels by location. First, however, it is necessary to review a potential confounding variable, the times of day of the measurements.

Sampling-time effects

While 89% of the 246 measurements in the two larger buildings were obtained early in the mornings on Mondays, the remainder were subjected to a preliminary analysis to insure their comparability with the Monday morning data.

The atypical measurements were concentrated in the early years and in the main building: nearly all were both carried out in the afternoons and on days other than Mondays. To each such measurement was matched a Monday a.m. control measurement: the control value was the first previously unused prior measurement in the same room, if that existed, otherwise, the first previously unselected following measurement. Only 24 of 26 measurements could be matched. Outside of laboratory areas, the geometric mean of the 9 non-Monday p.m. measurements (196 Bq m^{-3}) was very similar to that of the Monday a.m. measurements (216 Bq m^{-3}). A very different pattern appeared in the laboratory areas, how-

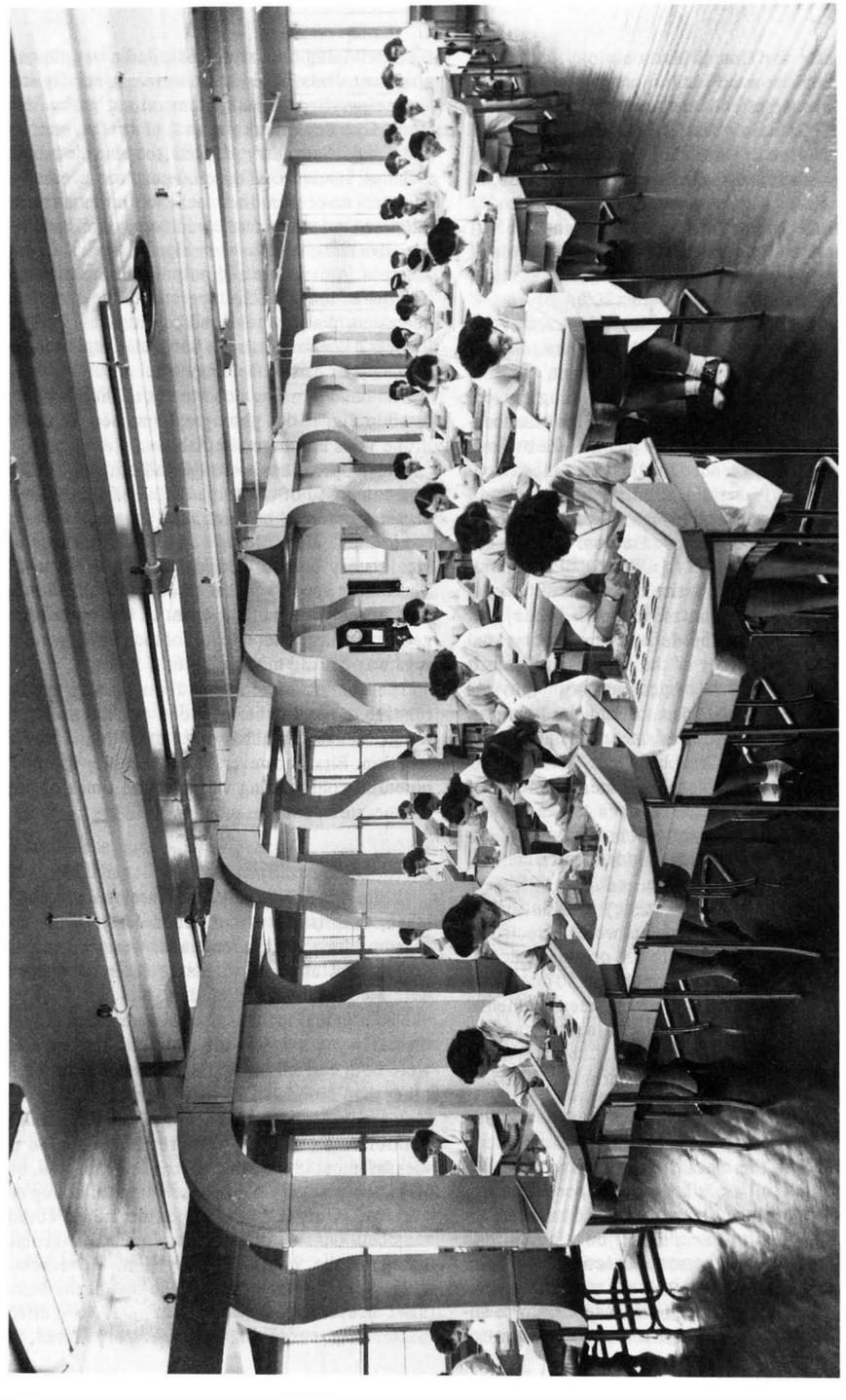


Fig. 1. The "Hand Painting Department" at the Bloomsburg plant (U.S. Radium Corporation photograph).

ever, where the 8 non-Monday p.m. measurements (175 Bq m^{-3}) were much lower than the Monday a.m. measurements (863 Bq m^{-3}). The sparse data from the other pairs were consistent with these findings. In analyses following, a.m. measurements are separated from p.m. measurements only for the laboratory areas.

Radon levels by area and room

Workers, predominantly women, who hand-painted dials worked in the "by hand" area on the second floor of the main building. This second-story front had no adjoining second story areas to the rear: it was, thus, naturally both well-ventilated and lighted. It is believed that the area shown in Fig. 1 is part of this area. Table 1 gives mean radon levels for the various rooms identified with this area: levels were highest in the painting room, but except for the office, all areas were similar.

The dial process, office, and technical support areas were on the first floor: the dial process areas were central; the office areas were west of center, to the front; and the technical support areas were east of center, at the front. The radon levels in the dial process areas and main office (Table 2), which approach those in the "by hand" area, may reflect lessened ventilation in the building center and/or air movement from the adjacent laboratory areas. The extreme variability in the dial process areas in particular may reflect variable ventilation patterns. The technical support areas were naturally better ventilated and more distant from sources, except, of course, the darkroom (which, in the context of this industry, usually refers to a darkened room for inspection of luminized objects).

Radon levels in laboratory areas are given in Table 3. Even the general laboratory areas have radon levels slightly above those in the hand painting areas, while the radium laboratory areas have much higher levels, in the 400 to 600 Bq m^{-3} range. The area where the phosphor was compounded showed the highest levels. These areas are also quite variable, more so than the hand-painting areas, but less so than the dial process areas, which adjoined the general laboratories.

Table 4 shows radon levels in the one-story etching building. Although generally low, they approximate those in the hand-painting department in a few rooms. The high levels in the maintenance rooms are notable; maintenance rooms seem not to have been measured elsewhere. In general, variability was low.

Radon levels in three peripheral structures, the radium vault, the waste disposal build, and the laboratory evaporation building were high: 3051 (n = 9), 399 (n = 3), and 564 (n = 6) Bq m^{-3} respectively.

Hess took a number of control measurements in an unspecified manner as well as some immediately outside certain plant buildings. He also took some in the physician's house, near the center of Bloomsburg, where breath radon measurements were taken from workers on Monday mornings. Both the plant location and the physician's residence were on topographically low areas associated with low house radon levels (Janssen et al. 1991). These values are shown in Table 5, along with measurements taken decades later from the residences of individuals who had (or still did) work at the Bloomsburg plant (Janssen et al. 1991).

Table 1. Radon levels (Bq m^{-3}) in "by hand" dial painting areas of main building.

	N	Geom. mean	Geom. s.d.
"By hand" area, 2nd fl.	49	134	2.61
Application room	23	167	2.24
Office	6	47	1.55
Oven room	12	151	3.58
Spray room	4	120	1.84
Inspection darkroom	4	146	2.36
Rest and lunch areas, 3rd fl.	6	28	4.02

Table 2. Radon levels (Bq m^{-3}) in dial process and office areas of the first floor, main building.

	N	Geom. mean	Geom. s.d.
Dial process areas	13	164	7.36
Dial process and laboratory	8	113	8.20
Dial plant	4	373	7.96
Lackon eng. office	1	116	--
Administrative office areas	12	69	2.84
Main office	5	137	2.36
General sales office	4	80	1.38
R&D office	3	18	1.99
Technical support	19	97	5.16
Health physics office	3	45	2.03
Library	4	25	1.83
Main dark room	12	185	5.55

Temporal trends

The preliminary analysis (Hartmann 1986) suggested a significant downward trend in overall plant radon levels from the beginning to the end of the study period. Clearly in the last few years of the study, there were both a decline in radium usage and an increased interest in monitoring. Visual examination of residuals from an overall regression on time suggested only a reduction in the frequency of high outliers during the first 14 y, but no reduction of median values until about 1963.

In this analysis, where the individual work room or area is taken as the prime determinant of radon levels, only two rooms (the "by hand" application room and the radium laboratory) have enough measurements to permit a temporal analysis. The work areas are not sufficiently homogeneous to encourage temporal analyses by area. The eighteen years (1949-1966) of the study had been divided equally into three sexennial periods; general scarcity of measurements in some early years and the early predominance of afternoon measurements permitted comparison of only the last two sexennia for these two rooms. Results for the "by hand" application room differed little over time: geometric means were 185 Bq m^{-3} (n = 9) in the middle period and 164 Bq m^{-3} (n = 9) in the last period. Results for the radium laboratory were strikingly different: the geometric mean for the middle period was 616 Bq m^{-3} (n = 11), and but 210 Bq m^{-3} for the last period. Clearly attention had been paid to reducing radon levels in the laboratory areas in the last few years of radium operations.

The preliminary analysis also investigated seasonal effects, and found marginally significantly higher levels in the spring and fall (periods of relative atmospheric stability in that climate), lower levels in the winter, and lowest in the summer. Those results would be expected on a priori grounds, and are within the range -29% (summer) to +35% (autumn) relative to annual averages.

DISCUSSION

The Bloomsburg site and the major buildings maintained average radon levels well below the 370 Bq m^{-3} tolerance limit in effect (U.S. Public Health Service 1954) for radium workplaces. The U.S. Public Health Service recommended permissible level of radon daughters in mines (Holaday et al. 1957) however implied a permissible radon level approximately 20 times the tolerance limit in radium workplaces, as atmospheres in luminizing facilities were normally quite clean for the practical reason that luminized articles were sticky and were set out to dry.

Table 3. Radon levels (Bq m^{-3}) in laboratory areas of the main building, by time of day.

	Time	N	Geom. mean	Geom. s.d.
General laboratory area	a.m.	9	175	3.80
General laboratory	a.m.	8	164	4.11
Adhesives laboratory	a.m.	1	296	--
	p.m.	3	96	1.79
Laboratory annex rooms	a.m.	18	57	1.90
Annex and krypton rooms	a.m.	13	49	1.89
Laboratory office	a.m.	4	92	1.73
Isotope laboratory	a.m.	1	58	--
Ra/Po laboratory areas	a.m.	55	457	3.69
	p.m.	10	197	2.16
Radium laboratory	a.m.	23	442	3.59
	p.m.	7	233	2.10
Polonium area	a.m.	3	238	2.12
	p.m.	1	52	--
Compounding area	a.m.	11	603	3.85
	p.m.	1	195	--
Inspection darkroom	a.m.	5	577	2.00
Weighing room	a.m.	13	407	5.15
	p.m.	1	232	--
Ra lab basement weigh room	a.m.	11	225	2.57

Geometric mean exposure levels in individual work rooms, including those where hand or machine application of radium to dials took place, appear to have remained below the tolerance limit. A few radiation laboratory areas had Monday morning levels routinely above the tolerance limit, but these had been controlled in the last few years of radium usage, and seem, from the few afternoon radon measurements in earlier years, to have been below the tolerance level for some of the workday. Many office and administrative areas of the plant had radon levels no higher than those recorded many years later in a survey of residences of ex-employees of the plant (Table 5).

Room values should not be extrapolated to worker exposure levels without careful examination of time records and departmental assignments, and personal interviews. Work was at times sporadic; official department assignments on employee records do not necessarily reflect real assignments, especially in slack or excessively busy periods, and some workers were subject to sporadic layoff. The higher routine exposures are likely to have been suffered by the more highly technically skilled individuals working in laboratories or doing custom work; this fits the observed pattern of health effects in the industry in New York in the World War II era (Silson et al. 1955) and the Ottawa, IL facility in the late 1930s (unpublished data). Dial workers themselves, after approximately 1928, have been less at risk of health effects than technical personnel, and the external gamma exposures are believed to have played a major role in observed health outcomes in the technical staff (Silson et al. 1955; Stebbings et al. 1984).

Other serious issues remain concerning the extrapolation from these observed mean values to ac-

Table 4. Radon levels (Bq m^{-3}) in etching building watch dial (WD) areas.

	N	Geom. mean	Geom. s.d.
Etching building WD areas	35	92	2.41
Embossing room	4	41	1.56
Inspection area	4	61	1.55
Maintenance rooms	7	151	1.57
Oven room	5	219	1.39
Ra screen room	6	65	2.16
Shave, blank, packing rooms	2	50	1.23
Shipping room	6	89	4.14
Spray room	1	147	--

tual exposures of workers. For a few years, data are missing or sparse (1951, 1953, 1957-58), and season of measurement and calendar year are confounded to some degree. The major issue, however, derives from the fact that nearly all measurements were taken on Monday mornings at the beginning of the workday (when, quite properly, breath radon samples were taken, at the company physicians's residence). If the radon source term is primarily radium contamination and long-term storage, as is likely the case in the laboratory areas, these results will yield conservative estimates. If radium in actual use during the week is the primary source term, radon levels would be expected to be higher later in the day in those production areas. Any such effect would be exaggerated if there was a planned weekly production cycle such that a week's production was packed and/or shipped prior to the weekend, but during the week was exposed to air in the workrooms. Central areas in the main building may also have had higher radon levels later in the day from movement of radon out of the laboratory areas, and the observed extreme variability in these areas supports that concern.

The morning measurements were collected predominantly in the 0630-0900 h period; afternoon measurements were frequently taken about 1530 h,

Table 5. Radon levels (Bq m^{-3}) in control and comparison areas.

Measurements	N	Dates (19xx)	Geom. mean
From Victor Hess			
Outside plant buildings	11	49-66	30
Unspecified control	17	50-64	15
Physician's residence	11	54-55	35
From Janssen et al. (1991)			
Ex-employee residences			
Basements	189	83-85	149
First floors	198	83-85	66
Second floor	113	83-85	52

presumably at the end of the workday. In April, 1984, ANL staff carried out a 24-h survey of radon levels in two locations in the main building. In the main office, radon peaked at 1000 h and at 1700 h. In the library, levels peaked at 1200 h and reached a minimum at 1600 h. During these measurements, radium was not on site except as contamination, and the numbers of employees and usage of the building were much reduced from the period 1949-1966. Nevertheless, these observations support concern that Monday morning measurements may have underestimated radon levels in central areas of the building with unexceptionable Monday morning averages.

It does not appear that the plant represented any major improvement over the conditions described by Evans (1943) for luminizing facilities early in World War II. At that point, careful scientific attention was being paid to potential hazards in the radium industry (Evans 1943; Evans 1981; Evans and Goodman 1940) which, producing dials for aircraft and navy ships, was an important defense industry. In postwar years, although still active in producing for the military, the luminizing industry was at best in stasis. Given the equivalently much looser standards promulgated for the uranium mining industry at the time, it is quite reasonable that there was no push to reduce radon exposures to as low a level as possible, but, rather, only to keep average levels distinctly below the tolerance limit.

Acknowlegment — Gratitude is owed first to the U.S. Radium Corporation which furnished Argonne National Laboratory with their personnel and health physics records; their corporate successor at Bloomsburg, Safety Light Corporation, has been equally cooperative. The Rev. Edward Dunn, S.J., archivist of Fordham University, New York, granted willing access to the files of Victor Hess. Dr. John Rundo of Argonne National Laboratory furnished data necessary for this writing up of the study. Work at Argonne National Laboratory was supported by the U.S. Department of Energy under Contract W-31-109-ENG-38.

REFERENCES

Bloomfield, J. J.; Knowles F. L. Health aspects of radium dial painting. II. Occupational environment. J. Indust. Hygiene, 15: 368-382; 1933.

Duggan, M. J.; Godfrey B. E. Some factors contributing to the internal radiation hazard in the radium luminizing industry. Health Phys. 13: 613-623; 1967.

Evans, R.D. Protection of radium workers. J. Indust. Hygiene Toxicol. 25: 253-269; 1943.

Evans, R.D. Inception of standards for internal emitters, radon and radium. Health Phys. 41: 437-448; 1981.

Evans, R.D.; Goodman, C. Determination of the thoron content of air and its bearing on lung cancer hazards in industry. J. Indust. Hygiene Toxicol. 22: 89-99; 1940.

Hartmann, H.M. Analysis of 1949-1966 radon levels in the Bloomsburg, Pennsylvania U. S. Radium Corp. plant. Plan B Master's Degree Program paper. Division of Environmental Health, University of Minnesota, Minneapolis; 1986.

Hess, V.F.; McNiff, W.T. Quantitative determination of the radium content of the human body and of the radon content of breath samples for the prevention and control of radium poisoning in persons employed in the radium industry. Amer. J. Roentgenol. Radium Ther. 57: 91-102; 1947.

Holaday, D.A.; Rushing, D.E.; Coleman, R.D.; Woolrich, P.F.; Kusnetz, H.L.; Bale, W.F. Control of radon and daughters in uranium mines and calculations on biological effects. U.S. Public Health Service Publication No. 494. Washington, D.C.: U.S. Government Printing Office; 1957.

Janssen, I.; Stebbings, J.H.; Essling, M.A.; Dimino, K.M.; Rodgers D.L. Prediction of ^{222}Rn from topography in Pennsylvania. Health Phys. 61: 775-783; 1991.

NBS (National Bureau of Standards). Safe handling of radioactive luminous compounds. Handbook H27. Washington, D.C.: National Bureau of Standards; 1941.

Polednak, A.P.; Stehney, A.F.; Rowland, R.E. Mortality among women first employed before 1930 in the U.S. radium dial painting industry. Amer. J. Epidemiol. 111: 175-182; 1978.

Silson, J.E.; Kleinfeld, M.; Harris, S.J. External radiation exposure in the radium-dial painting industry. Arch. Indust. Health 12: 503-508; 1955.

Stebbings, J.H.; Lucas, H.F.; Stehney, A.F. Mortality from cancers of major sites in female radium dial workers. Amer. J. Indust. Med. 5: 435-459; 1984.

U. S. Public Health Service. Radiological Health Handbook. Washington, D.C.: U.S. Government Printing Office; 1954.

U.S. Radium Corporation. Untitled sales brochure. New York: United States Radium Corporation; pp. 8; undated.

Environment International, Vol. 19, pp. 449-454, 1993
Printed in the U.S.A. All rights reserved.

0160-4120/93 $6.00 +.00

RADIOLOGICAL ASPECTS OF FORMER MINING ACTIVITIES IN THE SAXON ERZGEBIRGE, GERMANY

Gert Keller
Institut für Biophysik, Universitätskliniken, D-6650 Homburg/Saar, Germany

EI 9203-126 M (Received 4 March 1992; accepted 9May 1993)

In the Middle Ages, rich silver occurrences were exploited intensively in the Saxon Erzgebirge. The unprofitable ores, the socalled "pechblende", were dumped as waste materials in the environment. The high uranium and radium contents of these rocks prompted later investigations by Henri Bequerel and Marie Curie. In the city of Schneeberg, most of the houses were built directly on these waste materials. Furthermore, the whole subsoil is crossed by a system of numerous pits and adits; therefore, radon gas from the soil can easily penetrate into the houses. The radon exhalation rates from the soil and also from ground floors in this region increase to values of about 1 Bq/m^2.s. In some houses in Schneeberg, ^{222}Rn concentrations of about 50 kBq/m^3 were found in the living rooms. Maximum values in the cellars were about 200 kBq/m^3. From the inhalation of its short-lived radon daughters, an effective dose of about 1.2 Sv/y may result. Mitigation methods like insulation and subsoil ventilation are proposed, and the efficiency of the different measures are discussed.

INTRODUCTION

The attractive landscape of the Saxon Erzgebirge is affected by former mining activities. The towers of the engine pits, the big tailings, the industrial plants, and the destruction by the open-cast characterize the region. The history of mining in the Erzgebirge is impressive and reaches from the Middle Ages to the presence. Over centuries, the rich deposits of metallic ores in the highlands of Saxony and Thuringia were mined intensively. Besides silver, copper, cobalt, bismuth, and other ores, the geological formations of the Erzgebirge contain a relatively high amount of uranium. At the end of the 18th century, a chemist of Berlin identified this isotope in a mineral derived from a mine near Johanngeorgenstadt. By systematic and partially unintentional working of the natural uranium ore in this region, damages arose to the environment and population whose removal now turns out to be a problem.

THE SILVER EXPLOITATION

At the end of the 14th century, mining of the silver deposits began in Schneeberg and Joachimsthal. The silver to coin the "Joachimsthaler" was found in the mines of Joachimsthal. From this, the German "Taler" and the english word "dollar" developed. In the following centuries, the silver treasure of the region had been exploited so intensively that innumerable shafts and mine adits pass through the underground. According to old records, particularly small people were settled here to do the hard work with pick and shovel in the narrow mine adits. Many

inhabitants of this region had tiny silver mines of their own in the cellars of their houses to raise the income of their families.

When working the silver deposits, the miners often found useless material, which was piled up on the tailings. This black, slightly shimmering rock brought bad luck to the workers (the German expression is "pech"), because it always occurred when the silver deposits in the mine adits ran short. For this reason, the miners called that material "pechblende", without foreseeing that this "pechblende" was a high-grade uranium ore that would bring bad luck in a second sense. These uranium enriched ores were deposited on special tailings or simply anywhere in the environment. Later on, houses were built on this waste material without sufficient insulation, such as on the Kirchberg in Schneeberg, a thoroughly undermined hill covered with large tailings. Also one of the biggest churches of Saxony, the St. Wolfgangskirche, is situated in the middle of that contaminated area.

In former times, a disease common to the silver mine workers was noted, which led oftenly to an early death. At that period, there was no possibility of diagnosing this disease accurately, but the connection between the disease and the mining was sensed. For 75% of the miners in the region around Schneeberg and Joachimsthal, this disease "Schneeberger Krankheit" caused death (Schüttmann 1983).

THE URANIUM MINING

Henri Becquerel discovered the natural radioactivity in 1896 (Franke 1969) by examining the "pechblende" which originated from Joachimsthal. Also Marie and Pierre Curie performed their investigations on radioactivity with the uranium ore from Joachimsthal. They extracted the element radium out of the residues of paints which also contained uranium mined in Joachimsthal. The high value of radium, one gram cost about 500 000 Marks in the year 1911, led to a radium-fever. Many places in Saxony competed for recognition as radium spas.

Then in 1938, the discovery of the nuclear fission by Otto Hahn and Fritz Straßmann forced the exploitation of the uranium. In August 1945, when the atomic bombs were dropped on the cities of Hiroshima and Nagasaki, Soviet geologists began to investigate the deposits of uranium ore around Schneeberg and Johanngeorgenstadt. Only one year later, the Soviet joint-stock company *Wismut* was founded, and thus the uranium from Saxony and Thuringia was destined for the Soviet Union exclusively. In the following years, the company expanded the uranium exploitation. The hectic activities recalled the gold rush in America. Uranium miners earned more money, the supply of goods was better, and the hospitals were more generous and more modern than usual. So the *Wismut*, later called *Soviet-German Joint-Stock Company Wismut*, slowly became a state within a state (Beleites 1988).

Until last year, no details about the organization and productivity of *Wismut* were available. After the German unification, it became public that between 1946 and 1989 about 220 000 tons (or Mg) of uranium had been produced and exported to the USSR. East Germany was thus one of the three biggest uranium-producing countries in the world. In peak years, about 150 000 workers were employed, managed by 5 000 soviet employees. Altogether, approximately 400 mine shafts and up to 15 milling plants were run in these mining districts (Fig. 2).

ENVIRONMENTAL IMPACT

As a result of the uranium mining more than 8 000 tailings, deposits, and shafts remained in Saxony. In 34 regions with an expansion of 1500 km, the amount of radium in the soil of more than 0.1 Bq/g was determined by aeronautical gamma-dose-rate measurings. The biggest part of the "suspected areas" are about 6500 tailings with a radium content which exeeds the standard value of 0.2 Bq/g for free utililization. In many cases, the overburden of mining was used for the building of houses and roads, so it is too small to be detectable by aeronautical measurements. Meanwhile, it is supposed that about 170 km of soil are so heavily contaminated with radioactive material that these areas have to be cleared as soon as possible.

In 1879, the disease "Schneeberger Krankheit" was diagnosed as lung cancer. At the beginning of the 1950's, the correlation between the lung cancer risk and inhalation of the short-lived radon daughters and the resulting relatively high radiation dose in the lung could be explained. Unfortunately, this realization came too late for many uranium miners. It may be considered as an ominous sign, that the badge on the caps of the Schneeberg miners was identical with today's radiation warning symbol.

EXPERIMENTAL METHODS

The indoor concentrations of ^{222}Rn were measured by different methods, i.e., by time-integrating passive radon monitors exposed for about 3 months and by only 2- or 3-d exposed charcoal dosimeters (Pensko and Wicke 1988). The radon concentrations in the air and the soil were also determined by point and con-

tinuous measurements with the method of electrostatic deposition and subsequent alpha spectroscopy. These measuring arrangements were used in a similar way for the determination of the radon diffusion coefficients in different materials and also for the investigations of the radon exhalation rates from soil and building materials (Fig. 1). The exhaled radon passes through an insulated intermediate area and reaches the measuring chamber consisting of the positively-charged metallic hemisphere and the grid. After the alpha decay of radon, the polonium ion is deposited on the negatively-charged detector surface. The polonium decays further upon emission of alpha particles, and the alpha spectra are measured for various successive periods of time. The rate of radon exhalation from the soil is computed from the increase in polonium or radon concentration, respectively, in the measuring volume. The integral rise in concentration is determined every 15 min over a period of 2 h. The resulting lower limit of detection under these conditions for the radon exhalation rate is about 0.3 $mBq/m^2.s$, the median error being approximately 10% (Keller et al. 1984).

RESULTS OF MEASUREMENTS

The results of some measurements of the indoor radon concentration in Saxony are shown in Table 1 (Jacobi 1992).

In Schneeberg, the median radon concentration in dwellings amounted to values of about 300 Bq/m^3; that means a sevenfold increase compared with the median value of about 40 Bq/m^3 in dwellings in West Germany. In about 60 houses in Schneeberg, radon concentrations of more than 10 kBq/m^3 were determined, the maximum values even reached 80 kBq/m^3. The radon exhalation rates from the soil ranged from about 50 to 1000 $mBq/m^2.s$ in this mining district, whereas in regions without an elevated radium content, values of 1 to 10 $mBq/m^2.s$ are common.

HEALTH RISKS IN THE SAXON ERZGEBIRGE

The Saxon Erzgebirge, similar to the Black Forest, is a region where the inhabitants are permanently exposed to an elevated level of natural radioactivity. Additionally, the radiation exposure was increased, sometimes drastically, by mining and processing the uranium ore.

Apart from the deformation of the landscape and the pollution of the environment with chemical poisons, the mining activities in that region especially caused health problems to the inhabitants. The sink basins for uranium-enriched mud are a special source of risk, because radium may enter the ground water from the waste dumps. The water here may contain up to 40 Bq/L uranium and 3 Bq/L radium (Richter 1990). In the neighbouring rivers and lakes, the radium

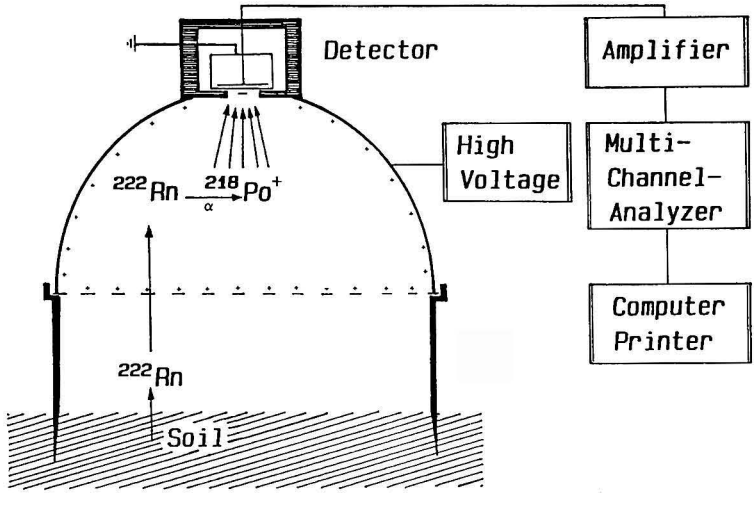


Fig. 1. Measuring arrangement for the determination of radon exhalation rates from the soil and building materials.

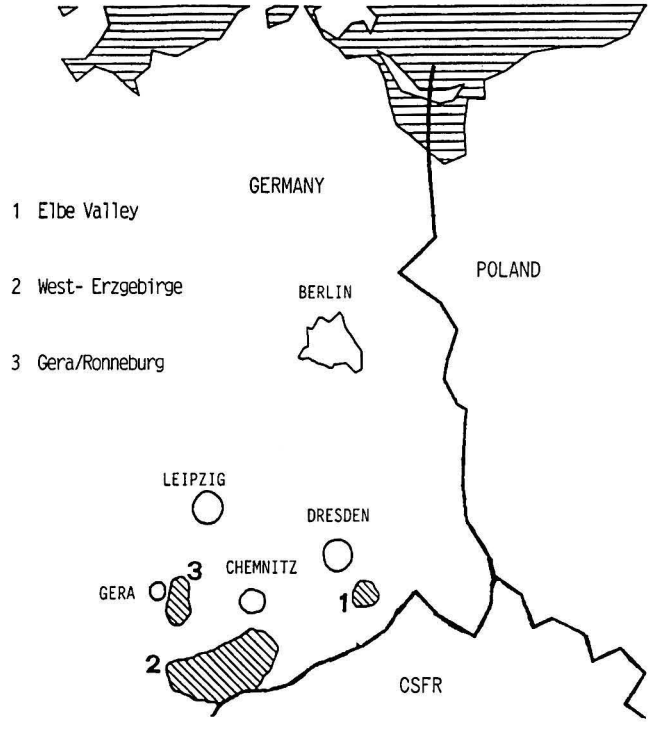


Fig. 2. Uranium mining districts of the *Wismut AG* in 1990.

Table 1. Measurements of indoor radon concentrations in Saxony.

District	50 % of all measured values have a radon concentration of more than	1 %
Ronneburg	48 Bq/m^3	3400 Bq/m^3
Johanngeorgenstadt	150 Bq/m^3	3000 Bq/m^3
Schneeberg	280 Bq/m^3	15000 Bq/m^3
compared to West Germany (about 6000 dwellings)	42 Bq/m^3	200 Bq/m^3

concentrations range between 0.04 and 0.1 Bq/L. The radionuclide content of the surface soil near sink basins or tailings are also elevated above ambient levels. Particularly in dried mud basins contaminated dust is deposited in the adjacent area by erosion. Likewise, depositing waste material in the environment contributes to the increase of the terrestrial irradiation.

The soil is burdened by deposits of contaminated dust and water, and, thereby, agricultural products may be affected. The most important contribution to the burden caused by the mining might be the inhalation of the short-lived radon daughters.

The radioactive noble gas radon decays to its short-lived progeny, which attaches itself to aerosols. The inhalation of these particles and their deposition in the lungs leads to an inhomogenous irradiation of the respiratory tract. The resulting average effective dose by inhalation is approximately one order of magnitude higher than the terrestrial radiation exposure in houses. The level of radon concentration in some houses, e.g., in Schneeberg, is comparable to the values in uranium pits, thus, increasing the risk of lung cancer for the population.

Since 1952, about 6 800 cases of lung cancer amongst *Wismut* staff were reported. Of these cases, 5 200 were accepted as occupational disease by health insurers (Richter 1990). The basis for this decision was the level of the inhaled radon concentration, which until 1955 had to exceed 1.5 Sv/y. The highest lung cancer rate of the miners was found during those years, when the uranium production was quickly raised by primitive mining methods irrespective of the danger to the worker's health.

The occupants of the houses are also at risk, the same as the miners. The mean values of the radon concentrations indoors in that region may be estimated to be about 250 Bq/m^3. In an especially affected part of Schneeberg, values up to 10 kBq/m^3 in dwellings are not uncommon. The maximum concentrations measured in homes are about 100kBq/m^3. Here, the radon gas of the whole hill can diffuse quickly through the old silver mine adits to the surface. Some shafts which are directly connected to the mine adits of the pits are now used as cellars. Additionally, the natural stone used to construct the houses exhales radon into the rooms. Almost all of the affected houses are built on soil contaminated with pitchblende and often they have neither sufficient insulation against the ground nor a massive substructure, so, the radon of the tailings may directly enter the houses. Often the "shaft cellars" are connected to the house by stairs which lead through the deposited waste material. In the high radon area around Schneeberg, the elevated radon values are attributed to the silver mining in the Middle Ages and not directly to the uranium mining.

The main influence of the uranium mining on radon concentrations indoors is the use of waste material for the building construction. Only in cases where houses are situated directly on the slope of a dump is a strong influence by radon possible. In postwar times, this uranium mining waste was released as building material without any restriction. On account of the shortage of building materials, the inhabitants willingly accepted this offer. The average concentration of ^{226}Ra in waste material is about 0.8 Bq/g, but due to the inhomogenous composition, values up to the tenfold may occur. Since 1970, the use of waste material with radium activities of more than 0.2 Bq/g have been subject to official approval, and its use as a building material has been prohibited. As this material was available and very cheap, large amounts of the waste material were used to construct houses, despite the above mentioned restrictions. Until last year, about 750 000 tons of material from the dump near Crossen were allotted to the building industry, for use in road construction. However, this material was also used in home construction, for example as admixture to concrete and as embankment. In houses built with this material, radon concentrations of several thousands of Bq/m^3 may occur.

CONCLUSIONS

The exposure of the population of the Saxon mining region to increased indoor radon concentrations requires urgent attention. About 1% of the dwellings in Schneeberg have radon concentrations exceeding 15 kBq/m^3. With the standard conversion factors (ICRP dose conversion factor, equilibrium factor F = 0.35, breathing rate 12.5 L/min, occupancy factor in houses 80%), the effective dose (Jacobi 1991) for the inhabitants due to the inhalation of short-lived radon progeny can be estimated to be greater than 0.4 Sv/y. This considerably exceeds the limits for occupational exposure.

In 1989, the German Bundesminister für Umwelt made about 6 million Marks available for mitigation of the most seriously affected houses in Schneeberg. In the first period, the effectiveness and practicability of several mitigation methods were investigated at six houses with the highest radon concentrations. Sixty houses with radon levels of more than 10kBq/m^3 were identified and will be mitigated in this project. If it is not possible to reach an acceptable radon value of about 250 Bq/m^3 by mitigation methods or if the costs are too high compared with

the worth of the house, a resettlement of the occupants or a new building will be considered.

The mitigation of the whole radioecological detriments in the Saxon Erzgebirge caused by mining may be estimated to last more than 10 y with costs of about 10 to 15 billion Marks.

REFERENCES

Beleites, M. Pechblende, der Uranbergbau in der DDR und seine Folgen. Kirchliches Forschungsheim, Wittenberg, Germany; 1988.

Jacobi, W. Aktuelle Umweltsituation in den neuen Bundesländern, Teil 1, Band 9. GSF-Forschungszentrum für Umwelt und Gesundheit, Neuherberg/München; 1992. Available from: Gesellschaft für Strahlen- und Umweltforschung, Neuherberg, Germany.

Jacobi, W. Radon: Ein altes Problem mit neuen Dimensionen, GSF-Mensch und Umwelt, 7. Ausgabe, Neuherberg; 1991. Available from: Gesellschaft für Strahlen- und Umweltforschung, Neuherberg, Germany.

Keller, G.; Folkerts, K.H.; Muth, H. Special aspects of the ^{222}Rn and daughter product concentrations in dwellings and in the open air. Radiat. Prot. Dosim. 7: 151-154; 1984.

Pensko, J.; Wicke, A. Measurement techniques for the estimation of the radon-222 concentration and the exhalation rate indoors and outdoors using an activated carbon bed. Compact of the 4th European congress of IRPA, Salzburg, Austrian Assoc. for Radiation Protection, A-2444, Seibersdorf; 1988.

Richter, S. AG Wismut; 1990. (Personal communication)

Schüttmann, W. Die Strahlenbelastung der Atemwege durch die Radionuklide der Umwelt. J. Erkrank. Atm.-Org. 161: 248-256, Leipzig; 1983.

Franke, H. Lexikon der Physik. Stuttgart: Francksche Verlagsbuchhandlung; 1969.

Environment International, Vol. 19, pp. 455-465, 1993
Printed in the U.S.A. All rights reserved.

0160-4120/93 $6.00 +.00

THE SCIENTIFIC DEVELOPEMENT OF A FORMER GOLD MINE NEAR BADGASTEIN, AUSTRIA, TO THE THERAPEUTIC FACILITY "THERMAL GALLERY"

J. Pohl-Rüling
Division of Biophysics, University of Salzburg, A-5020 Salzburg, Austria

EI 9203-130 M (Received 15 March 1992; accepted 3 February 1993)

The air in the Thermal Gallery (22 000 m³) has a mean radon content of about 100 kBq/L, temperatures of up to 41°C, and relative humidities of up to 99%. Due to these unusual conditions and to the first observations of health effects (curing of colds and rheumatic diseases) on mine workers during World War II, the idea was born in 1946 to use that abandoned mine as a big radon inhalation room for therapeutical purposes. This paper describes the historical development of the vast and detailed investigations, the facilities available at that time, of the physical mine properties, and the biophysical and basic medical effects after inhalation of the mine's atmosphere. The physical investigations included the variation of temperature and humidity with artificial and natural ventilation and numerous point measurements of the atmospheric radon content, which continously changed from site to site and day to day. These measurements showed that radon is supplied from the fissures in the rocks in response to changes in barometric pressure. Furthermore, the relation of the amounts of radon to its decay products were determined for the various sites. The biophysical investigations covered the solubility of radon in blood at high body temperatures (hyperthermia) and the determination of the dose in the various tissues and organs in animals and humans after inhalation of radon and its daughters. Individual assessments of personal doses according to the dose and dose distribution studies enabled a detailed investigation of the nonlinear dose dependence of radiation-induced structural chromosome aberrations in the peripheral blood lymphocytes of workers entering the mine and of persons living and working in the spa house in front of the mine and in the area of Badgastein. The basic medical information was obtained from voluntary collaborators and patients, brought into the mine for experimental purposes. They included hematological and cardiovascular tests, observations of mobility enhancements, and personally recorded pain reliefs. Due to these studies, the commercial therapeutic use could be started in 1952. Today, more than 500 patients per day can be treated.

GEOLOGY

The Gastein valley was formed by tectonic movements in connection with five periods of glaciation. It crosses the mountain chain, Hohen Tauern, which is a tectonic window. This means that the oldest rock units of the Alps, consisting of various gneisses, are exposed to the surface by basement elevation and erosion. These gneisses, formed from igneneous rocks, were then covered by slates from Paleozoic and Mesozoic shales, which in younger times again underwent liftings and foldings. Therefore, the whole region has a very complex internal structure and

contains a wide variety of rock types. Warm solutions of minerals and ores moved up into the fissures, resulting in quartz- and gold-bearing veins that were visible in the highest levels of this area (Exner 1966; Oxburg 1968).

HISTORY OF THE MINE

The mining for gold started in pre-Christian times; their next heydays were in the Middle Ages (between 13th and end of 16th century). Then the mines were situated 1600 m above sea level, mainly on the mountain Radhausberg, which contains quite a system of North-East striking fissures. Some gold mines were still operating until the beginning of this century.

In 1940, during World War II, an attempt was made to renew the search for gold with the modern mining equipment of that time. In the search for the prolongation of the gold veins at about 1 280 m above sea level, a straight tunnel was cut in approximately perpendicular to the general strike of the ore-bearing veinlets.

At that time, this tunnel was called Radhausberg-Unterbau-Stollen and later renamed the Thermal Gallery. The straight and even tunnel extends 2 425 m. At 1888 m, a wide ore-bearing vein was found and the mining followed this main vein 620 m to the north and 520 m to the south. Up to 1944, the overall length of the mine was 4.8 km, with an air volume of about 22 000 m^3 (Fig.1).

No gold was found, but the gallery proved to have unexpectedly high rock temperatures, measured in shot-holes before the blastings. The highest temperature of 44°C was found in the straight tunnel at 1600 m. It was about 23°C higher than expected according to the geothermal gradiant, given by the vertical thickness of the overlying rocks. In spite of thicker rock overlayings, the temperatures become lower again at the end of the straight tunnel at 2 425 m. This excessive heat is a local phenomenon. Nothing similar was found at the nearby train tunnel crossing the same mountain. Its origin has not yet been explained with modern geological techniques. There is the speculative assumption of thermal water seams very deep beneath the mine, because the rocks themselves are dry, but the atmosphere reaches 99% relative humidity.

Karl Zschocke, the chief mining engineer at that time, found trace amounts of secondary uranium minerals. He was thinking of possible radioactivity in the air and hung up light tight-packed photographic plates which became black, if developed, after some time.

Zschocke also observed that in Winter the mine workers, then mainly war prisoners, lost their colds and rheumatisms, when they had to endure temperature differences of 60° C (ranging from -20°C outside to +40°C inside).

In 1944, towards the end of the war, the order was given to blast the mine, after removing all technical installations for use in other mines. Zschocke did that very slowly, in order to postpone the blasting from one week to the other, until the end of the war. In that manner, he saved the mine and a great part of the artificial ventilation and train rails. These were still intact up to 1 888 m and 340 m to the south.

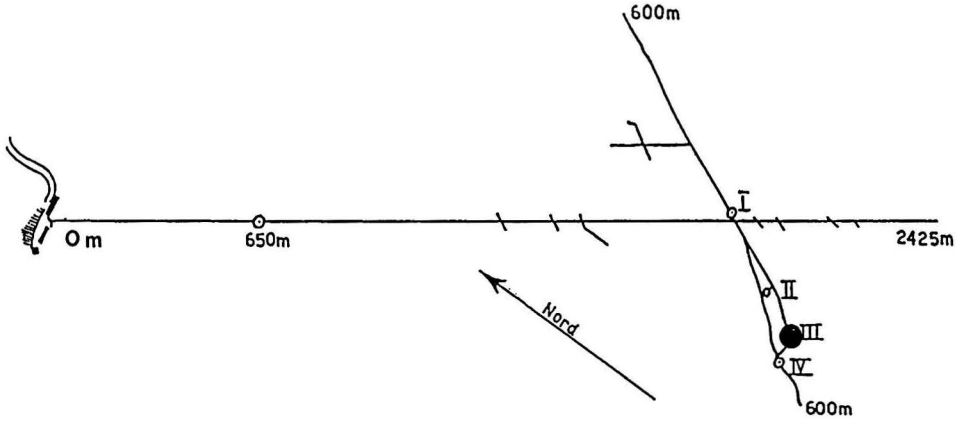


Fig. 1. Cross-section of the Thermal Gallery. The numbers I-IV designate the stations where the patients rest. Mean temperatures and relative humidities are taken from Scheminzky (1965). The black point is the place for measuring the daily radon contents, given in Fig. 3.

THE SPA BADGASTEIN, HISTORY, AND FACTS

The Thermal Gallery is situated about 8 km south of Badgastein, a town which has been a spa for many centuries. It is reported that the thermal water was discovered during the seventh century. The first documents containing the name Gastein were from 963 and 1350. The first book dealing with Badgastein as a spa was written 1497, and several books appeared in the past century, e.g., by Reissacher (1865/1964). The first medical doctor opened his practice in 1671. A book of rules for the innkeepers was written in 1688 (Gerke 1946). In this book, it was ordered that the beds of the guests have to be cleaned at least once a year! At that time, the patients spent many hours a day within the warm water, eating, and playing.

Nineteen hot water springs originate in the center of the town of Badgastein and supply together 5 million L of thermal water (up to 48°C) with a mean ^{222}Rn content of about 800 Bq/L. The radon content of this water was discovered by Curie. In the following years, it was measured in detail in the various springs by Mache and Bamberger (1914). Since then, the healing effect of this thermal water was attributed mainly to its radon content.

THE RESEARCH INSTITUTE GASTEIN

To investigate the medical effects of taking this thermal water and to measure its physical and chemical properties, the Research Institute Gastein was founded in 1936 (Scheminzky 1961). After being closed during the war, this institute again took up its scientific works in 1946, in provisionally installed laboratories in a cottage. The leader was Prof. Ferdinand Scheminzky, the Director of the Institute of Physiology at the University of Innsbruck. He died in 1973 and along with him died the main enthusiasms for further scientific medical investigations.

From 1952 to 1978, this institute was under the patronage of the Austrian Academy of Science. During this time, about 400 scientific papers were published by various scientists from different universities and research centers in Austria and from abroad. In 1979, the institute was reorganised and is now named the Research Institute of the Tauern Region.

Preliminary measurements of the radon content in the air of the Thermal Gallery, showed up to 200 kBq/m^3 (Hernegger 1946). In 1948, Scheminzky's team started to investigate scientifically the unusual phenomena in the Thermal Gallery, in order to decide whether it could be used therapeutically as a large radon inhalation room.

HISTORY OF SCIENTIFIC INVESTIGATIONS

Physical, biophysical, and medical investigations were necessary. It was no easy work at that time. The wooden hutments of the miners and war prisoners were provisionally adapted as physical and medical laboratories. A primitive road ended at the foot of the tailings from the mine. Heavy equipment could be brought up by the former mining elevator. But the staff had to walk about one hour up to the mine from the nearby village Böckstein. The necessary equipment for the measurements had to be improvised and built by the scientists. Still several years after the war, there were still shortages in everything including food. The only things that the scientists had plenty of were enthusiasm and long working hours (12 -14 h/d) during their usual two-week stay in this region. Changing the mine from mining purposes into a therapeutical facility required more than 100 000 working hours from the whole staff.

PHYSICAL MEASUREMENTS

Numerous measurements showed that the absolute and relative humidities unexpectedly did not conform with the temperatures. The reason was the air circulation in some areas instead of the expected continous heat convection to and from the mine entrance. This was observed from the direction of cigarette smoke (the only means for such investigations available at that time).

These results were necessary to answer the main questions, namely, what is the source of radon in the air; and will it remain constant or vanish when the mine is used daily as a therapeutical facility? The most obvious answer would have been that radon emanates from the rocks. Therefore, 140 rock samples from within the gallery were collected, pulverized, and their activity measured in an ionisation chamber by a static electrometer. The surprising result was that most of the samples showed a normal uranium and thorium content, only those of the gneiss in the first 340 m of the straight gallery were about three times higher (Exner and Pohl 1951). Thus, the rocks themselves could not be responsible for the high atmosperic radon content.

In 1948, numerous daily point measurements of the atmospheric radon content within the mine were started. (At that time neither accumulating radonmeters nor even electric calculators existed.) The single samples had to be filled into rubber balls and carried into the laboratories. Here, their activity was measured in a 10-L ionization chamber in connection with an electrostatic electrometer. Later, a double ionization

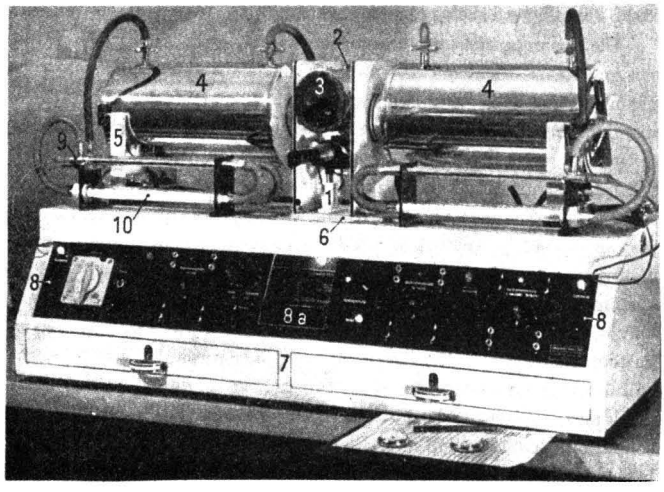


Fig. 2. Double ionization chamber device by E. Pohl (1953). (1): Electrometer; (2) Switch-gear, behind (3); (3) Change-over switch for electrometer; (4) Ionisation chamber; (5) U-holder for ionisation chamber; (6) Base-plate; (8) Switch-board; (9) Voltmeter; (10) Trap for ions; (11) Drier.

chamber device was built by Pohl (1953), allowing the measurement of about 30 samples a day (Fig. 2). It was found that the radon content within the mine varied not only from site to site, but also from day to day. It was confusing because the maximum would be found one day in the back above the main vein and on another day in the first meters of the gallery.

Finally, it was found that the variations were connected with the barometric pressure and the air streams, caused by the natural and artificial ventilation within the mine. Decreasing barometric pressures produce pumping effects for the gaseous radon from the rock fissures and from the inside of the main vein. This changing daily supply is much bigger than the constant emanation of radon from the free surfaces of rocks containing ^{226}Ra. During constant or increasing barometric pressures, the radon concentration is reduced by its radioactive decay and by removal due to natural and artificial ventilation and diffusion (Fig. 3). These components cause an effective half-life for radon in the Thermal Gallery of 1.83 d.

Similar investigations in other German mines were carried out and showed the same behaviour of the daily radon content. From these measurements, Pohl-Rüling and Pohl (1969) derived a formula to calculate the radon content according to the barometric pressure of the constant supply from the rock surfaces and the effective half-life of radon in the respective mine. They also suggested, for economy reasons in new mines, coordinating the necessary artificial ventilation rate with the barometric pressure.

In the Thermal Gallery, the daily radon supply from the surface was 13 Bq/L. The daily radon supply from the fissures of the rocks, however, was 48 Bq/L for a daily barometric decrease of 1 mm. Thus, the radon content within the mine will be supplied anew and always be at disposal for therapeutical use.

Besides the content of radon itself, the amounts of its short- and long-lived decay products were determined. At that time no working level meter was available; the decay products were measured at the various sites by sucking a certain amount of air through membrane filters. The deposited radionuclides including ThB (^{212}Pb) were then measured in the laboratory by alpha counters. Direct measurements within the gallery were not possible because of its high humidity. A special double filter device was constructed by Märk and Pohl (1972) to determine thoron (^{220}Rn) also (Fig 4). Typical amounts of the various radionuclides within the Thermal Gallery and the treatment house (spa hotel) built later at its entrance are given in Table 1 (Pohl-Rüling and Pohl 1950-1972).

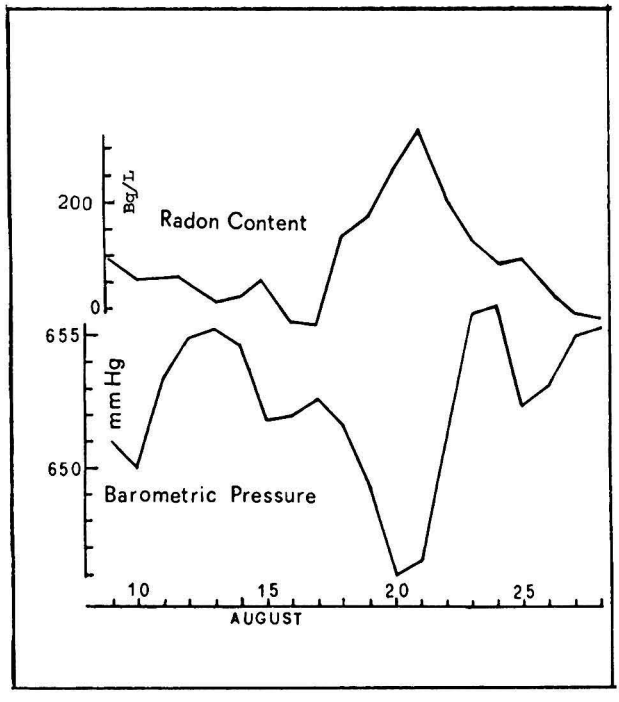


Fig. 3. Diagram showing the radon content in the air of the Thermal Gallery (Bq/L) compared with the barometric pressure (mm Hg). The point on which the measurements were carried out is given in Fig. 1.

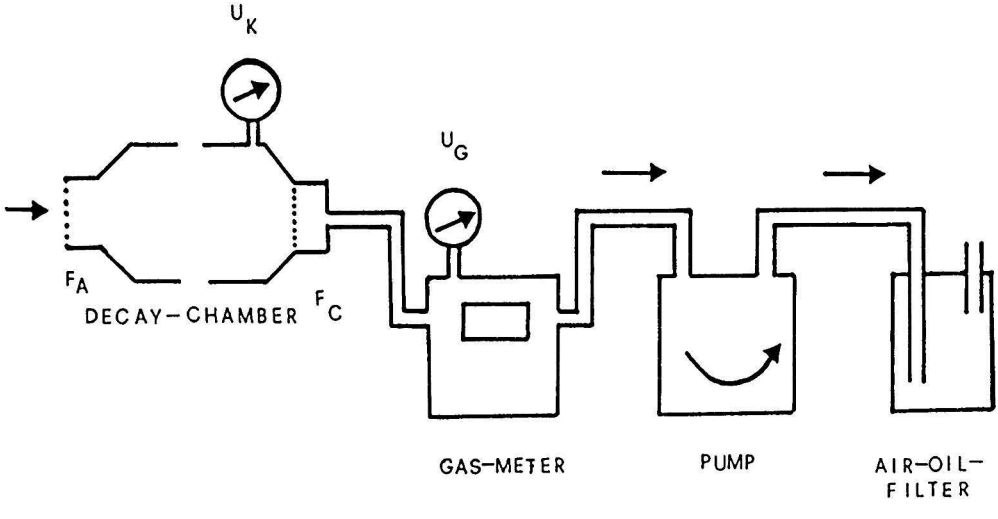


Fig. 4. Double-filter device by Märk and Pohl (1972). F_A: input filter supplies air content of ^{214}Pb, ^{212}Pb, and ^{210}Pb. Fc: output filter supplies air content of ^{222}Rn and ^{220}Rn.

Table 1. Mean activities of the various nuclides in the air of the Thermal Gallery and the spa hotel at their entrances.

	Radon 222 (Bq/m^3)	Lead 214 / Radon 222	Radon 220 (Bq/m^3)	Lead 212 / Radon 220
Thermal Gallery	111000	0.8	0.30	0.12
Spa Hotel	1110	0.45	--	--

BIOPHYSICAL INVESTIGATIONS

In 1950, the exhalation of radon by humans after a 2-h stay within the gallery was measured by Pohl and Pohl-Rüling (1950-1972) and Scheminzky (1965) and showed a biological half-life of 10 to 20 min (Fig. 5).

At about the same time, the radon concentration ratio of blood to air was measured for the human body with and without hyperthermia. The mean values (7 and 18 experiments) were 31% and 74% with maxima of 34% and 224%, respectively (Pohl-Rüling and Scheminzky 1954).

The most important biophysical results of the later years were those enabling the calculation of dose distribution in the various organs and tissues of the human body after inhalation of a certain amount of radon and its decay products. For that purpose, many experiments with animals and human blood and urine within the thermal gallery and in artificial inhalation facilities were necessary (Pohl 1962; Pohl 1964; Pohl and Pohl-Rüling 1967). Table 2 gives the calculated dose distribution for patients after their 2-h stay in the gallery. For comparison, the doses received after one typical bath in the thermal water of Gastein are also given. There are big differences between the doses at these two treatment facilities.

At various times in further years, such dose calculations were also carried out in 122 persons living and/or working in the entire Gastein area, including the mine workers. From these, a nonlinear dose-response curve of chromosome aberrations at this low-dose level could be obtained, and the hypothesis of triggered repair enzymes at a certain damage of the DNA was established by Pohl-Rüling and Fischer (1979) (Fig. 6).

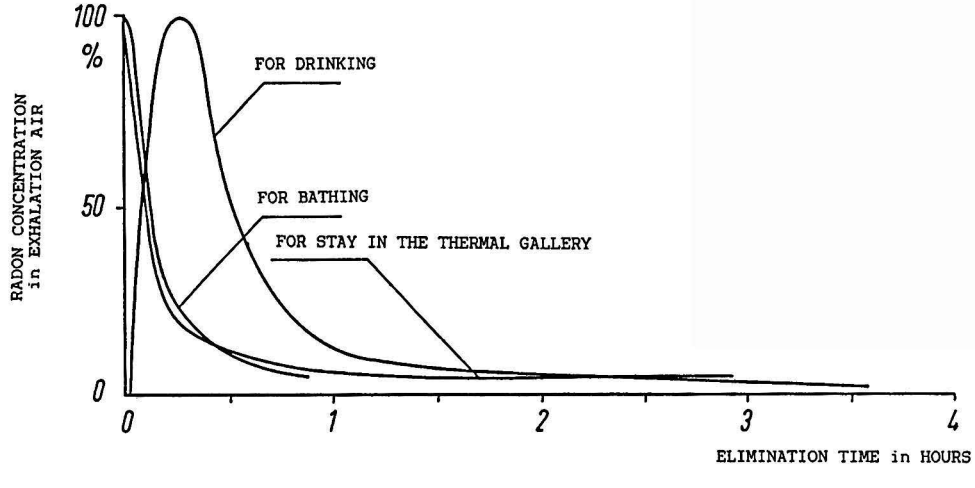


Fig. 5. Exhalation of radon after a 2-h stay within the Thermal Gallery showing a biological half-life of 10-20 min, measured by Pohl and Pohl-Rüling (1950-1972).

Table 2. Alpha-doses for a whole cure of 20 thermal baths or 12 Thermal Gallery treatments.

20 thermal baths	
Radon 222 content in water (mean)	555000 Bq/m^3
Radon 222 content in air (mean)	3330 Bq/m^3
Ratio: Radon 222 : Polonium 218 : Lead 214 : Bismuth 214 in air (mean)	1 : 0.8 : 0.4 : 0.3
Bathing time	20 min

Organ/Tissue	Dose in µGy*
Lungs, alveolar tissue	5.4
Blood, all influences**	1.2
Liver	1.0
Kidneys	1.8
Adrenal glands	2.0
Gonads	1.2
Bone marrow	1.0
Muscles	0.8
Bones	0.2

12 Thermal Gallery treatments	
Radon 222 content in air (mean)	111000 Bq/m^3
Radon 222 : Polonium 218 : Lead 214 : Bismuth 214 in air (mean)	1 : 0.9 : 0.7 : 0.6
Treatment time	2 h

Organ/Tissue	Dose in µGy*
Lungs, alveolar tissue	890
Blood, all influences**	84
Liver	52
Kidneys	200
Adrenal glands	41
Gonads	17
Bone marrow	30
Muscles	15
Bones	19

* As the RBE of alpha rays in this dose range is not known, the doses are given in µGy

** This value also includes the additional dose received by the blood on its way through the lungs.

MEDICAL USE OF THE MINE

Treatment proceedure

As soon as volunteer patients were available, they underwent the following treatment. From the main entrance (first outdoors and then indoors to normal room temperature), the patients reached tunnel-meter 600 in about 10 min on a small electric train, where it was warm enough to undress. After a second 10-min ride, they entered Therapy Station I (TS I, see Fig. 1) at tunnel-meter 1888, with a mean air temperature of 37.3°C and a mean relative humidity of 84%. During this time, the human body gets used to its higher surrounding temperature and starts to sweat because of the high humidity. Then, the patients remained lying for one hour either at TS I, without hyperthermia, or, after a further short train ride, at one of the Therapy, Stations II (40.5°C, 85%); III (40.5°C,86%); or IV (40.1-40.4°C, 95-97%), whereby their body temperatures increased up to

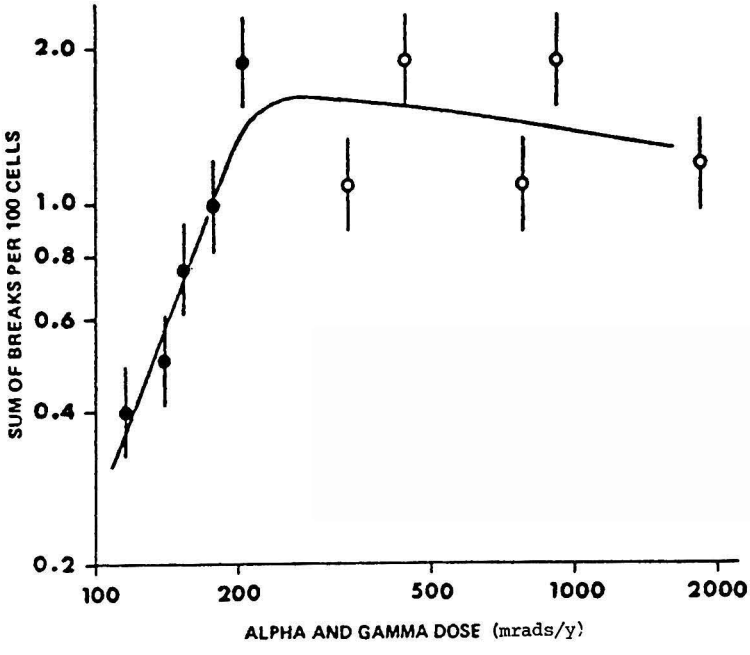


Fig. 6. Mean annual chromosome aberrations of persons living and/or working in the Gastein area (Pohl-Rüling 1979). Each point is based on 2 500 to 3 500 scored cells. The errors are calculated according to a Poisson distribution. Black points: from persons with continual burdens mainly due to gamma irradiation. Circles: from persons with occupational (fractionated) burdens mainly due to internal alpha irradiation.

38.6°C. The locus of their stay was prescibed by a medical doctor according to their illness, overall constitution, and heart capacity. Ten to twenty of such treatments made an entire cure. Such cures were first tentatively applied; after proving to be successful, they are still prescribed up to now.

Assumed healing factors

1) The increase of body temperature because of heat accumulation. Due to the high air and rock temperatures and high humidity, the body cannot lose heat as usual through heat conduction, radiation, or sweat evaporation. Heat is a well-known means to influence several rheumatic diseases. Due to the slow acclimatisation during the ride into the hot areas, the treatment heat is applied with more protection of heart and circulation than at normal sauna baths.

2) The high air humidity. During the one-hour lying within the Gallery, the mean loss of water through sweat was measured to be 0.6 kg for women and 1.2 kg for men (with a maximum of 4 kg). Sweat has several therapeutical effects: an overall vegetative stimulation; excretion of toxic substances from the body; immune system improvement; increase of skin circulation (which is poor especially in patients with rheumatism); and renewal of the skin protective mantle against bacteria and pathogenic microorganisms.

3) The radon content of the air. At the beginning of this century, after the discovery of radioactivity, the healing abilities of this new phenomena were tested. Until the mid-fifties, the short-lived alpha emitter ^{224}Th was even used as an immune-stimulatoric means at cancer and Morbus Bechterew (spondylarthritis ankylopoetica) and several successes were reported (Wilberg 1952; Buchhorn and Wilberg 1957). Also, the main healing factor of the waters in the spa Badgastein was then ascribed to its radon content. Therefore, the high radon content of the Thermal Gallery was the reason for all the investigations to use it as therapeutical facility. Various investigations were carried out to find medical effects which could be attributed to the incorporation of radon.

MEDICAL INVESTIGATIONS

From 1948 on, many kinds of hematological and cardiovascular tests were started with volunteers, first from the staff and then from Gastein patients. For a summary of these reports, see Scheminzky (1965).

During the early therapeutical trials at the Thermal Gallery, it was shown that heart frequency and minute volume increase less than during sauna treatment. The mean pulse rate after one hour lying in the hot areas reached only 101-103/min. At the same body temperature, the increase during a sauna bath to a mean of 150/min is usual. This was later proved in a laboratory experiment at the University of Innsbruck, simulating a Thermal Gallery treatment with the same temperature, humidity, and radon content (Scheminzky 1965; Scheminzky and Schröcksnadel 1961). The mean results from 15 volunteer students showed lower systolic and diastolic blood pressures and lower heart frequencies per minute when radon is added to heat and humidity (Fig. 7).

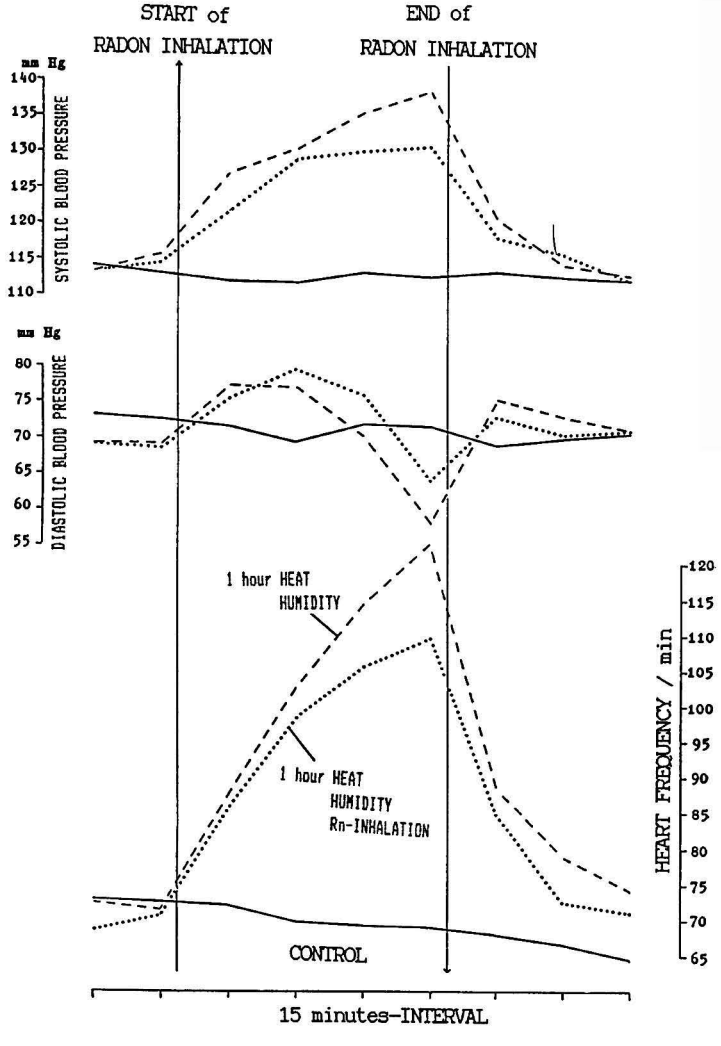


Fig. 7. Systolic and diastolic blood pressure and heart frequencies per minute in presence of heat and humidity, with and without radon inhalation (Scheminzky 1965; Scheminzky and Schröcksnadel 1961).

This lower circulation burden was attributed to an adrenaline reduction and an increase of noradrenaline under the influence of radon, as was observed by Wense (1951) for taking the bath in the Gastein thermal waters and later also for the Thermal Gallery treatment by Eigelsreiter and Schmidt (1959).

In 1951, a medical certificate was given by Prof. Scheminzky and Prof Hittmeier (Chief of the Medical Department of the Hospital of the University in Innbruck) confirming that for patients with various diseases of the rheumatic group, vascular diseases, disorders of endocrine organs, and metabolic disorders (e.g., gout), as well as gerontological complaints, the entrance and stay within the Thermal Gallery was a successful treatment. This certificate was then the basis for the declaration of the Thermal Gallery as a healing facility.

More recent papers on scientific medical studies are the following: Effect of radon and hyperthermia, in the Thermal Gallery as therapy for spondylarthritis ankylopoetica (Sandri 1973); DNA synthesis and DNA repair (Günther and Altmann (1978); DNA-Repair und Kortisolproduktion unter Radontherapie bei Gesunden und Rheumakranken (Günther et al. 1979); Reparaturprozesse in Lymphozyten beruflich strahlenexponierter Personen (Tuschl and Klein 1984); The influence of radon on the immune system and DNA metabolism (Tuschl and Altmann 1979); Immunologische Untersuchungen während Radon-Balneotherapie (Egg et al. 1984); Effect of low doses of ionizing radiation on peptide in blood and tissues (Bernatzky et al. 1989); Balneological treatment of patients with rheumatoid arthritis (Steiner et al. 1986), and The influence of radon inhalation and hyperthermia of the Thermal Gallery on bronchial asthma of children (Novotny et al. 1991).

COMMERCIAL CURES AND TREATMENTS

Commercial treatments in the thermal gallery started in 1952. A road was constructed, and the treatment house in front of the mine was built, enlarged and partly rebuilt in the following years.

By the sixties, more than 200 patients per day could be brought into the mine; today more than 500 patients can be treated per day. A statistical investigation of 18 195 patients taking a Thermal Gallery treatment between the years 1949 and 1963 showed very good success in 34.8% of the cases, a small success in 41.2% of the cases, and no success and the end of their treatment in 24% of the cases, Scheminzky (1965). This distribution of success is valid up to now.

REFERENCES

Bernatzky, G.; Saria, A.; Holzleitner, H. et al. Auswirkung niedrigdosierter ionisierender Strahlung auf regulatorische Peptide im Blut und in Geweben. Z. Phys. Med., Balneol., Med. Klimatol. 19(Sonderheft 2): 36-40; 1989.

Buchhorn, P.O.; Wilberg, C. Bericht ber die Erfahrung mit Thoros (perorales Thorium X-Präparat) in der Krebsbehandlung. Strahlentherapie 104: 306-313; 1957.

Egg, D. et al. Immunologische Untersuchungen während "Radon-Balneotherapie". Z. Phys. Med., Balneol., Med. Klimatol. 13 (Sonderheft 1): 56-60; 1984.

Eigelsreiter, H.; Schmidt, W. Die Beeinflussung der Haut-Kapillaren durch Adrenalin während einer Gasteiner Badebzw. Stollenkur. Z. Angew. Baeder-Klima-Heilkd. 6: 612-616; 1959.

Exner, C. Geology of Austria. Wien: Geologische Bundesanstalt; 1966: 77-84.

Exner, C.; Pohl, E. Granosyenitischer Gneis und Gesteinsradioaktivität bei Badgastein. Jahrb. Geol. Bundesanst. (Austria) 94(2): 2-57; 1951.

Gerke, O. Gasteiner Badebüchlein. Eine historisch-medizinische Studie. In: Wilh. Braumüller, ed. Wien IX/Leipzig: Universitäts-Verlagsbuchhandlung; 1946.

Günther, R.; Altmann, H. DNA-Synthese und DNA-Repair bei chronischem Rheumatismus unter Einwirkung kleiner Dosen natürlicher Radioaktivität. Z. Rheumatol. 37: 17-23; 1978.

Günther, R.; Egg, D.; Herold, M. DNA-Repair und Kortisolproduktion unter Radontherapie bei Gesunden und Rheumakranken. Z. Angew. Baeder- Klimaheilk. 26: 336-340; 1979.

Hernegger, F.; Unpublished results. Available from: Institute of Radium Research, Vienna. 1946.

Mache, H.; Bamberger, M. Über die Radioaktivität der Gesteine und Quellen des Tauerntunnels und über die Gasteiner Therme. Sitzungsber. Akad. Wiss. Wien, Math.-Naturwiss. Klasse, Abt. IIa, 123: 1-5; 1914.

Märk, T.D.; Pohl, E. Untersuchungen über den Thoron-, Radon- und Zerfallsproduktegehalt der Luft im Raum Badgastein mit einer Doppelfilterapparatur. Sitzungsber. Oesterr. Akad. Wiss. IIa, 180: 327-352; 1972.

Novotny, A. et al. Der therapeutische Einfluß von Radon-Inhalation und Hyperthermie im Gasteiner Heilstollen auf das Asthma bronchiale im Kindesalter (preliminary results). Z. Atemwege Lungenkrankh. 17(4): 2-24; 1991.

Oxburg, E.R. An outline of the geology of the eastern Alps. In: Proc. Geol. Assoc. 79: 1-46; 1968.

Pohl, E. Ein neues Emanometer für Präzisionsmessungen mit vielseitiger Verwendungsmöglichkeit. Sitzungsber. Oesterr. Akad. Wiss. IIa, 162: 435-450; 1953.

Pohl, E. Die Strahlenbelastung bei der Inhalation von Radium-Emanation. Strahlentherapie 119: 77-96; 1962.

Pohl, E. Dose distribution received on inhalation of radium-222 and its decay products. In: Proc. Radiological health and safety in mining and milling of nuclear materials; IAEA, Vienna, I: 221-236; 1964. Available from: IAEA, Vienna.

Pohl, E.; Pohl-Rüling, J. The radiation dose received by inhalation of air containing radon-222, radon-220, lead-212 (ThB) and their decay products. An. Acad. Bras. Ciencias (Mexico City) 39: 393-404; 1967.

Pohl-Rüling, J.; Fischer, P. The dose-effect relationship of chromosome aberrations to alpha and gamma irradiation in a population subjected to an increased burden of natural radioactivity. Rad. Res. 80: 61-81; 1979.

Pohl-Rüling, J; Pohl, E. Arbeitsberichte für das Forschungsinstitut Gastein, (mainly unpublished data, some in Badgasteiner Badeblatt, Badgastein); 1950-1972.

Pohl-Rüling, J.; Pohl, E. The radon 222 concentration in the atmosphere of mines as a function of the barometric pressure. Health Phys. 16: 579-585; 1969.

Pohl-Rüling, J.; Scheminzky,F.; Das Konzentrationsverhältnis Blut/ Luft bei der Radon-Inhalation und die Radon-Aufnahme in den menschlichen Körper im radioaktiven Thermalstollen von Badgastein/Böckstein. Stahlentherapie 95: 267-282; 1954.

Reissacher, K. Kurort Wildbad Gastein (Neudruck). Salzburg; 1964.

Sandri, B.; Die kombinierte Radon- und Hyperthermiebehandlung der Spondylarthritis ankylopoetica im Thermalstollen Böckstein-Badgastein. In: Prohaska, E. ed. Proc. Morbus Bechterev. Sonderheilanstalt für Erkrankungen des rheumatischen Formenkreises der Österr. Pensionsversicherungsanstalt der Angestellten; Wien-München-Bern: Verlag Maudrich; 1973: 75-90.

Scheminzky, F. Die Gasteiner Therme im Lichte der Wissenschaft. 25 Jahre Forschungsinstitut Gastein. Wien: Verlag Notring der Wissenschaftlichen Verbände Österreichs; 1961.

Scheminzky, F.; Schröcksnadel, H. Einfluß der Radium-Emanation auf Herzfrequenz und Blutdruck. Wiener Klin. Wochenschr. 73: 123-129; 1961.

Scheminzky, F. Der Thermalstollen von Badgastein-Böckstein, seine Geschichte, Erforschung und Heilkraft. Forschungen und Forscher der Tiroler Ärzteschule, Vol. V. Innsbruck: Verlag Tyrolia; 1965: 496.

Steiner, J.F. et al. Balneologische Behandlung von Patienten mit rheumatoider Arthritis. Z. Phys. Med., Balneol., Med. Klimatol. 15: 184-190; 1986.

Tuschl, H.; Altmann, H. Untersuchungen über den Einfluß von Radon auf Immunsysteme und DNA-Stoffwechsel. Z. Angew. Baeder- Klimaheilkd. 26: 391-397; 1979.

Tuschl, H.; Klein, W. Reparaturprozesse in Lymphozyten beruflich strahlenexponierter Personen. Z. Phys. Medizin, Balneol., Med. Klimatol. 13(Sonderheft 1): 44-49; 1984.

Wense, T. Adrenalin Inaktivierung durch das Gasteiner Thermalwasser und ihre Beziehung zur Gasteiner Kur. Wiener Z. Inn. Med. 32: 282-287; 1951.

Wilberg, C. Anwendung der radioaktiven Isotope, besonders der natürlichen Radioisotope Thorium X in Klinik und Praxis. Strahlentherapie 87: 430-436; 1952.

Environment International, Vol. 19, pp. 467-473, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

HISTORY AND CURRENT USES OF ^{224}Ra IN ANKYLOSING SPONDYLITIS AND OTHER DISEASES

R. R. Wick

Institut für Strahlenbiologie, GSF—Forschungszentrum für Umwelt und Gesundheit, Neuherberg, D-85764 Oberschleißheim, F. R. Germany

W. Gössner

Institut für Pathologie, GSF—Forschungszentrum für Umwelt und Gesundheit, Neuherberg, D-85764 Oberschleißheim, F. R. Germany

EI 9203-138 M (Received 28 January 1992; accepted 15 March 1993)

During and briefly after World War II, ^{224}Ra was used in a German hospital in combination with platinum and eosin (*Peteosthor*) for the treatment of tuberculosis and ankylosing spondylitis. The patients, primarily children and juveniles, received repeated intravenous injections of up to 2 MBq ^{224}Ra per injection twice a week for months, sometimes even for years. Injected amounts totalled up to 140 MBq. Following this therapy, an enormous increase in the incidence of bone tumors (56 cases among 900 patients), as well as other lesions was observed. The surviving patients are still under follow-up. Treatment of ankylosing spondylitis with drastically reduced doses of ^{224}Ra, however, was continued up to the recent present and over 1500 patients were so treated in West-German hospitals. This second cohort, exclusively adults, received much lower amounts applied in most cases as one series of 10 weekly injections of about 1 MBq of ^{224}Ra each. This would result in a cumulative alpha-dose of about 0.56 Gy to the marrow-free skeleton of a 70 kg man. These patients have been followed for several years, together with a control group of ankylosing spondylitis patients not treated with radioactive drugs or x-rays. By August 1991, three cases of malignant bone tumors have been observed among the exposed (0.7 - 2.4 cases expected) vs. one case among the controls. Diseases of hematopoietic tissue included leukemias (9 in the exposure group vs. 6 in the control group) and bone marrow failure (12 cases vs. 9). The increase of total leukemias among the exposed, compared to a standard population, is highly significant (9 cases observed vs. 2.8 expected, $p < 0.003$). Chronic myeloid leukemia, specifically, was elevated in the exposed group (3 cases observed vs. 0.8 expected, $p = 0.047$) but not in the control group.

INTRODUCTION

Radium-224 is one of the decay products of the natural thorium series, formally known under its historical name "thorium X" (Fig. 1). It was first separated in the supernatant remaining after precipitation of a thorium salt solution with aqueous ammonia by Rutherford and Soddy(1902). Radium 224 decays by alpha-disintegration, with a half-life of 3.66 d to ^{220}Rn (thoron) and then by three further alpha- and two beta-decays to the stable ^{208}Pb (Fig. 2). The daughters of ^{224}Ra are all short-lived; the longest half-life is that of ^{212}Pb (thorium B, 10.6 h), and altogether six different radionuclides are represented.

Radium 224 is an alkaline earth element which deposits preferentially in the bone when introduced into an organism. Due to its short half-life, most of the nuclide decays at the bone surface, thus, showing a similar distribution pattern and radiobiological

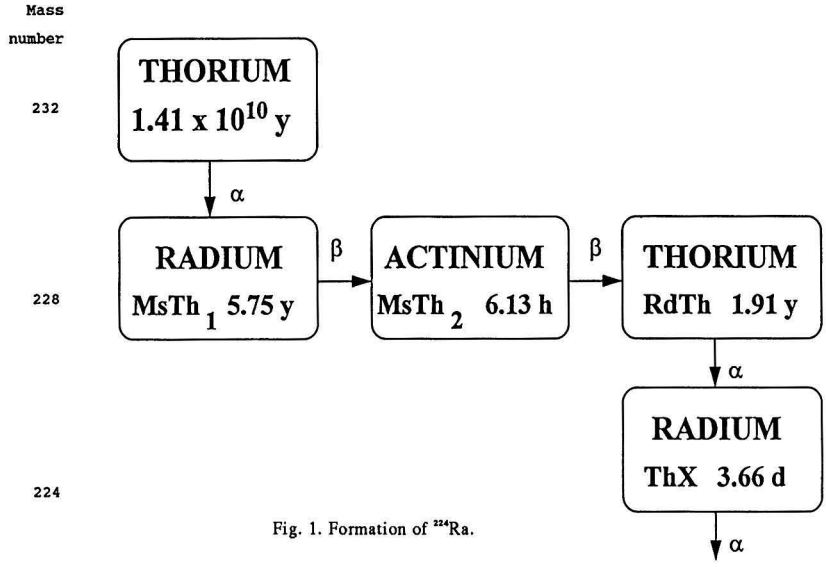


Fig. 1. Formation of ^{224}Ra.

behaviour to ^{239}Pu, which also is predominantly deposited on the bone surface. Internally applied, ^{224}Ra is mainly effective by its alpha-irradiation; the beta- and gamma-radiation is of minor importance. The decay products are partially accumulated in liver, kidney, spleen, and also in the eye. Investigations in humans on the distribution and excretion of ^{224}Ra have shown (Schales 1966), that about 50% of the

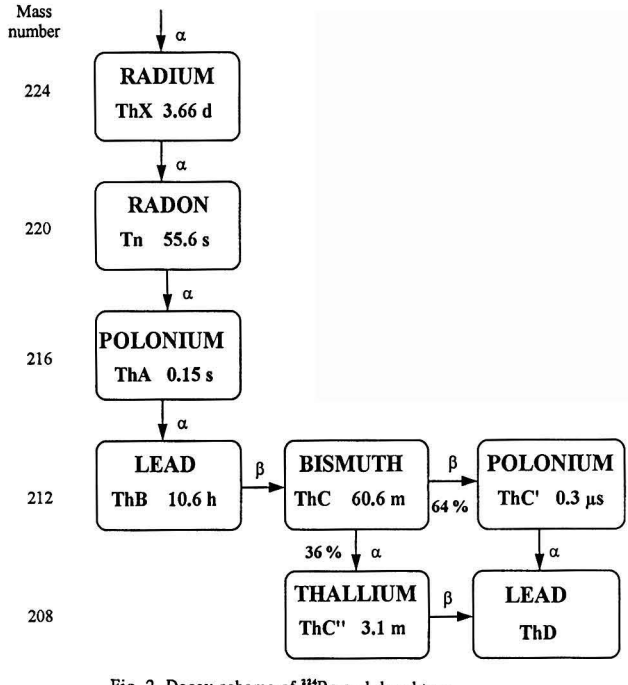


Fig. 2. Decay scheme of ^{224}Ra and daughters.

applied amount is excreted within the first week via feces and urine with a ratio of about 30:1. The biological half-life in the human body, excluding bone is 6 d, 10^4 d for kidney and liver, and 1.64 × 104 d for bone; giving effective half-lives of 2.3 d for total body, 2.7 d for kidney and liver, and 3.6 d for the bone (Seelentag and Schmier 1962). The resulting alpha-dose per 1 MBq of injected ^{224}Ra to the marrow-free skeleton of a 70-kg adult male is 0.56 Gy, or 0.67 Gy for a 63-kg adult female.

Soon after the first separation of ^{224}Ra in 1902, there was great interest in its potential therapeutic uses due to its radiochemical and biological properties. Numerous publications can be found in the medical literature before World War I, but there was no systematic search for medical uses and no animal experiments were reported from that period. Bickel (1913) was the first to report the treatment of ankylosing spondylitis with the new radionuclide.

Ankylosing spondylitis is a chronic inflammatory disease of the vertebral column which affects about 0.1% of the population. Onset is around the age of 20 y and is followed by a progressive stiffening of the whole spine caused by new bone formation. The disease was described at the end of last century independently by Bechterew in Russia, by Strumpell in Germany, and by Marie in France (Moll 1980).

Intravenous injection of ^{224}Ra (ThX), however, fell into disuse after several untoward incidents following internal application of large amounts of ^{224}Ra solution. Two patients actually died with acute radiation syndrome. After this experience, only external dermatological use in the form of ointments or lacquers continued. In France, intravenous injection of ^{224}Ra in patients with rheumatic diseases was reintroduced by Léri (1922), and was continued from around 1930 by Forestier (1951) with allegedly good success. In England, Hernaman-Johnson (1946) started with ^{224}Ra therapy of ankylosing spondylitis. In Germany, Troch (1943; 1949), relying on a historical hypothesis by von Wassermann (von Wassermann and von Hansemann 1912) about a reversed electrical polarity of diseased cells, developed a compound preparation of ^{224}Ra and traces of platinum stabilised by eosin which he called Peteosthor. When, after World War II, Troch became the head of a tuberculosis sanatorium, he introduced the Peteosthor treatment, mainly for the different forms of tuberculosis, into his hospital and later adopted it for ankylosing spondylitis also.

In the years between 1946 and 1950, about 2000 people, most of them children and juveniles, received repeated intravenous injections of Peteosthor, up to 2 MBq ^{224}Ra per injection twice a week for months or even for years, resulting in total injected quantities of up to about 140 MBq ^{224}Ra. This Peteosthor was used for the treatment of various diseases, such as tuberculosis of bone and soft tissue, ankylosing spondylitis, and others. The pretended improvement in tuberculosis treatment could not be verified by other clinicians, and use of the treatment did not spread to other institutions. Moreover, beginning in the early 1950's, objections to the treatment were raised—the primary one being that ^{224}Ra deposited in the growing skeleton of the children and juveniles would cause severe damage.

DAMAGE IN THE EARLY HIGH-DOSE PATIENTS

Rathke (1954) was the first to publish on the appearance of a bone tumor in a Peteosthor-treated juvenile coxitis patient; in 1955, he reported two osteosarcomas in juvenile TB patients. Spiess (1956) observed an alarming increase of bone tumors in the patients, among them mostly children and juveniles, treated with the high doses of Peteosthor by Troch. Thereafter, he made extensive efforts to relocate as many of those patients as possible, and to include them in a still ongoing follow-up study, with the late C. W. Mays as co-investigator from about 1970 until his untimely death in 1989. Their study generated numerous important contributions on the dosimetry and the biological effects of incorporated radionuclides.

Spiess and Mays (Spiess 1969; Spiess and Mays 1970; Mays et al. 1978; Mays et al. 1989) were the first to extensively investigate the epidemiologic consequences of exposure to the short-lived bone seeker ^{224}Ra in humans. Since the beginning of that study, they have reported 56 cases of bone tumors, mostly osteosarcomas among patients aged 2 - 56 y at beginning of injection. Among the children and juveniles studied, 38 cases (38/218, 17%) have been reported, whereas among the adults only 18 cases (18/682, 2.6%) have occurred. All, or virtually all, of these bone tumors can be ascribed to radiation, since only 0.3 cases would have been expected naturally, based on the general population rate of 2 bone sarcoma/y·100 000 persons. The distribution of tumor appearance times ranged from 3.5 to 33 y, averaging about 10 - 12 y. The last bone tumor appeared in 1985 after a latency time of 33 y. Based on present trends, few, if any, additional bone sarcomas are expected among the surviving ^{224}Ra patients (Chmelevsky et al. 1988; Chmelevsky et al. 1990).

Most data previously obtained on the induction of bone tumors by incorporated radioactivity have been

related to long-lived radionuclides (Vaughan 1973). The long duration of irradiation was believed to be necessary in order to produce those effects. A well-known example in human experience is the increased incidence of osteosarcomas following the intake of long-lived ^{226}Ra by luminous dial painters.

It is interesting to compare the distribution of appearance times of bone sarcomas induced by short-lived ^{224}Ra vs. those from the long-lived ^{226}Ra and ^{228}Ra (Fig. 3). The earliest appearance times are similar: 4 y in the ^{224}Ra patients and 5 y in the U. S. dial painter group. The important difference is that whereas the risk from short-lived ^{224}Ra seems to be exhausted after 33 y, tumors induced by the long-lived radium continue to appear throughout the remaining lifespan. This effect can be attributed to the continued production of new tumors by radiation received a long time after the initial deposition of the long-lived radium. For example, the bone sarcoma appearing after 63 y might have been induced by alpha-particles emitted 10 y earlier, some 53 y after the initial skeletal deposition of ^{226}Ra.

Bone tumors, however, were not the only lesion observed after injection of high amounts of Peteosthor or ^{224}Ra into mostly juvenile patients. Spiess and his co-investigators, as well as other study groups, have reported increased incidences of cataracts (Chmelevsky et al. 1988; Stefani et al. 1989; Honegger 1969; Taylor and Thorne 1988), tooth damages (Spiess 1969; Sonnabend et al. 1986; Haunfelder et al. 1977; Reichart 1979), exostoses (Spiess and Mays 1979), and growth retardation (Spiess et al. 1986; Koch 1957) after incorporation of high amounts of ^{224}Ra. Also, diseases of the liver (Spiess and Mays 1979) and kidney (Spiess and Mays 1979; Mays et al. 1989) have been described. In none of these treatments could a beneficial effect on the primary tubercular disease be verified.

RECENT TREATMENT OF ANKYLOSING SPONDYLITIS

Contrary to those uniformly negative observations for the various forms of TB and the other diseases, beneficial and long lasting results, mostly relief of pain, have been obtained in the treatment of ankylosing spondylitis. Treatment with ^{224}Ra was adopted by Pitzen (1949) for ankylosing spondylitis and subsequently therapeutic guidelines were established by Koch (Koch and Reske 1952; Koch 1978). Eosin and platinum, first omitted from the mixture for purely financial reasons, were soon shown in animal experiments to be completely without influence on distribution and effect.

Since the 1950's, the total amount of ^{224}Ra administered during a course of treatment has been drastically reduced to about 10 - 12 MBq. The treatment with ^{224}Ra, mostly at this dose level, has been performed by a great number of orthopedic hospitals in Germany which, unanimously, have reported beneficial analgesic effects, ranging from pain-free to at least moderate reduction of pain in the vast majority of cases (Schneller 1950; Mahlo 1952; Wilde 1952; Lentz 1952; Titze 1952; Rütt 1952; Kaiser 1953; Herdt 1956; Kutz 1957; Weber-Böllhoff 1959; Best 1959; Härtling 1962; Diederich 1965; Kellermann 1968; Rudolph et al. 1980). Radium 224 has been

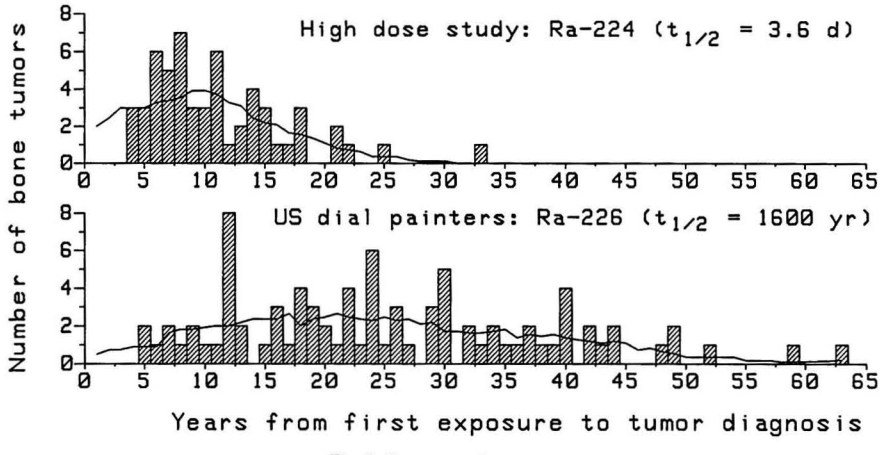


Fig. 3. Bone tumor latency.

used sporadically in neighbouring countries too (Erlacher 1953; Louyot et al. 1970; Bertrand et al. 1978; Roux et al. 1978; Roux and Mattei 1988). In France, it was injected also intra-articulary, as an alternative to other radionuclides, for the treatment of rheumatoid synovitis.

Dosages in prior times were usually calculated in electrostatic units (elektrostatische Einheiten, esE), an obsolete unit of radioactivity which was used until 1969 for the medical dosage of ^{224}Ra. This unit was based on an early experimental technique for the measurement of alpha-ray ionisation. Here, the geometry of the apparatus and the rapidity with which the measurements followed the separation of the ^{224}Ra sample from its parents both play an important role, and it is not possible to specify an universal conversion factor. For the preparations used in Germany for the treatment of ankylosing spondylitis, an equivalent of 28 μCi (1.036 MBq) for 200 esE of ^{224}Ra is generally accepted.

EPIDEMIOLOGICAL INVESTIGATIONS OF RECENT ^{224}Ra TREATMENT

One of the most severe late effects detected in the high-dose study of Spiess and Mays was the induction of bone tumors. The lowest dose associated with a bone tumor was found to be 0.9 Gy. Patients having received lower doses, however, were not sufficiently represented in that study. In order to extend the conclusions, a new study was started in 1971 (Schales 1978; Wick and Gössner 1983; Wick et al. 1986; Wick and Gössner 1989). The objective was the evaluation of the late effects risk to humans for bone tumors and other lesions potentially related to injected alpha-emitters below that lowest dose of 0.9 Gy.

This lower dose study includes most patients treated in West Germany for ankylosing spondylitis with low amounts of ^{224}Ra (Table 1). As of August 1991, the study includes around 1500 ankylosing spondylitis patients from nine hospitals. The majority of these patients, most of them treated in the years 1948-75, received one series of ten weekly injections, each of about 1 MBq of ^{224}Ra. In addition, there exists a control group of ankylosing spondylitis patients not treated with radioactive drugs or x-rays.

Up till now, causes of death have been ascertained in 542 patients of the exposure group and in 650 patients of the control group. Table 2 shows a summary of the cancers of bone and soft tissue observed so far in this study. In this table, we have considered only those diseases which are known or implied from the higher dose study to be associated with administration of

Table 1. Exposure and follow-up parameters.

	Exposure Group	Control Group
Observed patients	1471	1336
cause of death certified	542	650
av. inj. ^{224}Ra (MBq/kg)	0.17	–
av. skeletal dose (Gy)	0.67	–
av. inj. span (weeks)	10.2	–
av. follow–up time (yr)	19.0	20.4

Table 2. Cancers of bone and soft tissue.

	Exposure Group		Control Group	
	obs	*exp*	*obs*	*exp*
Total cancers	117	118–144	146	154–188
Liver	1	2.5–3.7	7	3.4–4.8
Urinary System	9	8.1–10.6	9	10.7–13.9
Female Breast	1	2.6–4.3	1	2.2–3.6
Skeleton	3	0.7–2.4	1	0.8–2.8
Leukemia	9	2.7–2.8	6	3.3–3.5
Chron. myeloid Leuk.	3	0.8	1	1.1

^{224}Ra. There is no significant difference between observed and expected cases for cancers of the liver, urinary system, or the female breast. Malignant primary bone tumors (according to the histological typing of bone tumours of WHO) have been observed in three cases in the exposure group: one fibrosarcoma of bone; one reticulum cell sarcoma (malignant lymphoma) of bone; and one medullary plasmocytoma (multiple myeloma) originally observed in the bone marrow of sternum and pelvis. The expected number of spontaneous bone tumors for the present follow-up time is estimated to be 0.7 - 2.4 cases, determined from the age-dependent spontaneous rates for bone tumors from the three cancer registries of the Federal Republic of Germany. Thus, an increased incidence for bone tumors in the exposure group of this lower dose study cannot be verified statistically.

Diseases of hematopoietic tissue among the study population included leukemias (9 in the exposure group vs. 6 in the control group) and bone marrow failure (12 cases vs. 9). The increase of total leukemias is, compared to a standard population, highly significant for the exposure group (9 cases observed vs. 2.7 - 2.8 expected, $p < 0.003$) and striking also for the controls (6 cases observed vs. 3.3 - 3.5 expected, $p = 0.14$). Possibly, the increase of leukemias in the control group may indicate an effect of the generally considerable intake of painkilling or other drugs for the treatment of the basic disease. Subclassification of the leukemias shows a clear preference for chronic myeloid leukemia in the exposure group (3 cases observed vs. 0.8 expected, $p = 0.047$), whereas in the control group (1 case observed vs. 1.1 expected) the observed cases are within the range of expectancy. Disorders of the hematopoietic system following treatment with ^{224}Ra have been reported earlier by other authors (Kutz 1963; Stieglitz et al. 1973) even at the same low dose level. Animal experiments with varying amounts of ^{224}Ra have also shown that mice given amounts of ^{224}Ra lower than those found to cause osteosarcomas may be at risk instead from myeloid leukemia (Humphreys et al. 1985).

EPILOGUE

Due to technical and commercial reasons, the production of ^{224}Ra for injection purposes was discontinued in spring 1990. It is uncertain whether it will ever be re-established.

Acknowledgment — This research has been supported by the Radiation Protection Programme of the Commission of the European Communities (CEC), Contract No. BI6-D-0083 D.

REFERENCES

Bertrand, A.; Legras, B.; Martin, J. Use of radium-224 in the treatment of ankylosing spondylitis and rheumatoid synovitis. Health Phys. 35: 57-60; 1985.

Best, G. Über die funktionelle Therapie bei der Spondylitis ancylopoetica in Verbindung mit Thorium X-Gaben. Arch. Orthop. Unfall-Chir. 50: 368-399; 1959.

Bickel, A. Weitere Beiträge zur ThX-Therapie bei Anämie, Leukämie und rheumatischen Erkrankungen. Berliner Klin. Wochenschr. 50: 346-348; 1913.

Chmelevsky, D.; Kellerer, A.M.; Land, C.E.; Mays, C.W.; Spiess, H. Time and dose dependency of bone sarcomas in patients injected with radium-224. Radiat. Environ. Biophys. 27: 103-114; 1988.

Chmelevsky, D.; Mays, C.W.; Spiess, H.; Stefani, F. H.; Kellerer, A.M. An epidemiological assessment of lens opafications with impaired vision in patients injected with radium-224. Radiat. Res. 115: 238-257; 1988.

Chmelevsky, D.; Spiess, H.; Mays, C.W.; Kellerer, A.M. The reverse protraction factor in the induction of bone sarcomas in radium-224 patients. Radiat. Res. 124: 69-79; 1990.

Diederich, O.; Klinik und Therapie der Spondylarthritis ankylopoetica -Behandlungsergebnisse mit Thorium X und Corticoiden. Bonn: Thesis, University, 1965.

Erlacher, P. Zur Behandlung des Morbus Bechterew mit Thorium X. Wien. Klin. Wochenschr. 65: 825-826; 1953.

Forestier, J.; Jacqueline, F.; Rotes-Querol, J. La Spondylarthrite ankylosante. Paris: Masson; 1951.

Härtling, S. Ein Beitrag zur Thorium-X-Therapie bei der Spondylitis ankylosans. Dresden: Thesis, Academy of Medicine; 1962.

Haunfelder, D.; Gessner, B.; Barckhaus, R.H.; Niemann, R.; Themann, H. Spätschäden am Odonton nach Behandlung mit Thorium X. Dtsch. J. Mund-Kiefer-Gesichts-Chir. 1: 191-201; 1977.

Herdt, L. Nachuntersuchungen an mit Peteosthor bzw. Thorium X behandelten M. Bechterew-Kranken des "Annastifts" Hannover. Hamburg: Thesis, University; 1956.

Hernaman-Johnson, F. Thorium X in spondylitis and chronic rheumatism. Rheumatism 3: 21-24; 1946.

Honegger, H. Strahlenkatarakt nach Peteosthorbehandlung (Thorium X). Monatsbl. Augenheilk. 155: 890-893; 1969.

Humphreys, E. R.; Loutit, J. F.; Major, I. R.; Stones, V. A. The induction by ^{224}Ra of myeloid leukemia and osteosarcoma in male CBA mice. Inter. J. Radiat. Biol. 47: 239-247; 1985.

Kaiser, G. Über die Behandlung des Morbus Bechterew, insbesondere mit Thorium X. Dtsch. Gesundheitswes. 8: 474-479; 1953.

Kellermann, P. Erfolge und Gefahren bei der Behandlung des Morbus Bechterew mit Thorium X. Berlin: Thesis, Karl-Marx-University; 1968.

Koch, W.; Reske, W. Die Ergebnisse der intravenösen Thorium X-Behandlung bei der Spondylarthritis ankylopoetica (M. Bechterew). Strahlentherapie 87: 439-457; 1952.

Koch, W. Strahlenbedingte Wachstumsstörungen der Gliedmassen. In: Proc. Septième Congrès de la Société Internationale de Chirurgie Orthopédique et de Traumatologie, Barcelona 16-27 septembre 1957. Brussels: Soc. Internat. de Chirurgie Orthopédique et de Traumatologie:1957: 457-466.

Koch, W. Indication for ^{224}Ra-therapy in ankylosing spondylitis (Morbus Strumpell-Bechterew-Marie). In: Müller, W. A.; Ebert, H. G., eds. Biological effects of ^{224}Ra-benefit and risk of therapeutical application. The Hague: Martinus Nijhoff Medical Division; 1978: 21-29.

Kutz, G. Zur Behandlung des Morbus Bechterew mit Thorium-X., Thesis, University Frankfurt; 1957.

Kutz, G. Zur Frage von Spätschäden nach der Behandlung mit Thorium-X. Z. Orthop. Traumat. 97: 474-482; 1963.

Lentz, W. Die Behandlung der Bechterew'schen Erkrankung mit Peteosthor. Med. Wochenschr. 7: 78-84; 1952.

Léri, A.; Thomas, M. Traitement des Rhumatismes Chroniques par le Thorium. Bulletins et mémoires de la Société Médicale des Hôpitaux de Paris 44: 10-13; 1922.

Louyot, P.; Mougin, G.; Legras, B.; Montet, Y.; Combebias, J.-F.; Pourel, J.; Simon, J.-M.; Diebold, P. La thérapeutique médicamenteuse de la spondylarthrite ankylosante. Rev. Rhum. 37: 281-309; 1970.

Mahlo, K.L. Orales Thorium X als Mittel der Wahl beim Morbus Bechterew. Dtsch. Med. J. 3: 472-474; 1952.

Mays, C.W.; Spiess, H.; Gerspach, A. Skeletal effects following ^{224}Ra injections into humans. Health Phys. 35: 83-90; 1978.

Mays, C. W.; Spiess, H.; Chmelevsky, D. Malignancies in patients injected with Radium-224. In: Taylor, D. M.; Mays, C. W.; Gerber, G. B.; Thomas, R. G., eds. BIR Report No. 21, Risks from Radium and Thorotrast. London: British Institute of Radiology: 7 - 12; 1989.

Moll, J.M.H. Ankylosing spondylitis. Edinburgh: Churchill Livingstone; 1980.

Pitzen, P. Über die Behandlung des Morbus Bechterew mit Peteosthor (Troch). Med. Klin. 44: 1111-1115; 1949.

Rathke, F.W. Schädigungen und Spätschäden nach Peteosthorbehandlung. Münch. Med. Wochenschr. 96: 884-886; 1954.

Rathke, F. W. Kritische Stellungnahme zur Frage der Schädigung nach ThX-Behandlung. Münch. Med. Wochenschr. 97: 952-953; 1955.

Reichart, P. A. Radiographic finding in ^{224}Ra-caused dental resorptions. Int. J. Oral Surg. 8: 467-473; 1979.

Roux, H.; Schiano, A.; Liprandi, D.; Serratrice, G.; Paulin, R.; Roux, F. Notre expérience du 224-Ra dans le traitement de la spondylarthrite ankylosante. In: Müller, W. A.; Ebert, H. G., eds. Biological effects of ^{224}Ra-benefit and risk of thera-peutic application. The Hague: Martinus Nijhoff Medical Division; 1978: 212-214.

Roux, H.; Mattei, J.-P. Traitement de la Spondylarthrite ankylosante par les radiations ionisantes. In: Simon, L.; Herisson, Ch., eds. La Spondylarthrite ankylosante, actualités nosolagiques et thérapeutiques. Paris: Masson; 1988: 142-147.

Rütt, A. Zur Therapie des Morbus Bechterew. Dtsch. Med. Wochenschr. 77: 649-652; 1952.

Rudolph, F.; Salewski, H.; Franke, J. Spätergebnisse nach Thorium-X-Behandlung des Morbus Bechterew (eine 15-Jahres-Studie). Beitr. Orthop. Traumat. 27: 29-37; 1980.

Rutherford, E; Soddy, F. The radioactivity of thorium compounds. Part I. An investigation of the radioactive emanation.Trans. Chem. Soc. London 81: 321-350; 1902a.

Rutherford, E; Soddy, F. The radioactivity of thorium compounds. Part II. The cause and nature of radioactivity. Trans. Chem. Soc. London 81: 837-860; 1902b.

Schales, F. Strahlenbelastung bei der therapeutischen Anwendung von Thorium X (Radium 224). Strahlentherapie 130: 420-425; 1966.

Schales, F. Brief history of ^{224}Ra usage in radiotherapy and radiobiology. Health Phys. 35: 25-32; 1978.

Schneller, H. Erfahrungen mit Peteosthor-Behandlung bei Morbus Bechterew. Med. Klin. 46: 142-144; 1950.

Seelentag, W.; Schmier, H. Inquiry into the treatment with Thorium-X, delivered by request of the Senator for Public Health of Berlin. 2. Auflage vom 7.5.1962. Berlin: Bundesgesundheitsamt; 1962 (in German).

Sonnabend, E.; Spiess, H.; Mays, C.W. Tooth breakage in patients injected with ^{224}Ra. Strahlenther. Suppl. 80: 60-64; 1986.

Spiess, H. Schwere Strahlenschäden nach der Peteosthorbehandlung von Kindern. Dtsch. Med. Wochenschr. 81: 1053-154; 1956.

Spiess, H. Radium-224-induced tumors in children and adults. In: Mays, C.W.; Jee, W. S.S.; Lloyd, R.D.; Stover, B.J.; Dougherty, Jean H.; Taylor, G.N., eds. Delayed effects of bone-seeking radionuclides. Salt Lake City, UT: University of Utah Press; 1969: 227-248.

Spiess, H.; Mays, C.W. Bone cancers induced by ^{224}Ra (Th X) in children and adults. Health Phys. 19: 713-729; 1970. + addendum Health Phys. 20: 543-545; 1971.

Spiess, H.; Mays, C.W. Liver disease in patients injected with ^{224}Ra. Environ. Res. 18: 55-60; 1979.

Spiess, H.; Mays, C.W. Exostoses induced by ^{224}Ra (ThX) in children. Eur. J. Pediatr. 132: 271-276; 1979.

Spiess, H.; Mays, C.W.; Spiess-Paulus, E. Growth retardation in children injected with ^{224}Ra. In: Gössner, W.; Gerber, G.B.; Hagen, U.; Luz, A., eds. Suppl. to Strahlentherapie Vol. 80. The radiobiology of radium and thorotrast. München-Wien-Baltimore, MD: Urban & Schwarzenberg; 1986: 45-50.

Stefani, F.H.; Spiess, H.; Mays, C.W. Cataracts in patients injected with a solution of radium-224, colloidal platinum, and the red dye eosin (Peteosthor). In: Taylor, D.M.; Mays, C.W.; Gerber, G.B.; Thomas, R.G., eds. BIR Report No. 21, Risks from radium and thorotrast. London: British Institute of Radiology; 1989: 12-21.

Stieglitz, R.; Thiele, M.; Stobbe, H.; Wegner, G. Schädigungen der Hämatopoese durch Thorium-X-Therapie. Folia Hämatol. (Leipzig) 100: 95-103; 1973.

Taylor, D.M.; Thorne, M.C. The potential for irradiation of the lens and cataract induction by incorporated alpha-emitting radionuclides. Health Phys. 54: 171-179; 1988.

Titze, H. Ergebnisse der Peteosthorbehandlung bei Bechterewscher Erkrankung. Die Medizinische (Stuttgart) 22: 763-765; 1952.

Troch, P. Über Peteosthor und seinen Einfluß auf bösartige Geschwülste des Menschen. Monatsschr. Krebsbekämpf. 11: 77-91; 1943.

Troch, P. Neue Wege des Heilens. Braunschweig: Vieweg; 1949.

Vaughan, J. M. The effects of irradiation on the skeleton. Oxford: Clarenton Press; 1973.

von Wassermann, A.; von Hansemann, D. Chemotherapeutische Versuche an tumorkranken Tieren. Berliner Klin. Wochenschr. 49: 4-10; 1912.

Weber-Böllhoff, H. Die Behandlung des Morbus Bechterew mit Peteosthor und Thorium X unter Berücksichtigung möglicher Spätschäden. Düsseldorf: Thesis, University; 1959.

Wilde, R. Peteosthor und Thorium X als Therapeuticum des Morbus Bechterew. Arch. Orthop. Unfall-Chir. 45: 329-333; 1952.

Wick, R.R.; Gössner, W. Follow-up study of late effects in ^{224}Ra-treated ankylosing spondylitis patients. Health Phys. 44 (Suppl.1): 187-195; 1983.

Wick, R.R., Chmelevsky, D.; Gössner, W. ^{224}Ra: Risk to bone and haematopoietic tissue in ankylosing spondylitis patients. In: Gössner, W.; Gerber, G.B.; Hagen, U.; Luz, A., eds. Suppl. to Strahlentherapie Vol. 80, The Radiobiology of radium and thorotrast. München-Wien-Baltimore, MD: Urban & Schwarzenberg; 1986: 38-44.

Wick, R.R.; Gössner, W. Recent results of the follow-up of radium-224 treated ankylosing spondylitis patients. In: Taylor, D.M.; Mays, C.W.; Gerber, G.B.; Thomas, R.G., eds. BIR-Report No. 21, Risks from radium and thorotrast. London: British Institute of Radiology; 1989: 25-28.

Environment International, Vol. 19, pp. 475-489, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

REVIEW OF RADIUM HAZARDS AND REGULATION OF RADIUM IN INDUSTRY

R. M. Pratt
State of New York Department of Labor, Division of Safety and Health, Brooklyn, NY 11201, U.S.A.

EI 9203-131 M (Received 17 August 1992; accepted 7 March 1993)

Radium usage in the United States peaked in the late 1930's and early 1940's, much of it centered in New York, in both medical applications for cancer therapy and industrial applications mainly for self-luminous paints and static elimination. While the production of radium shifted from America to Europe after 1920, the importation of radium salts and their incorporation into products peaked during the World War II years and continued by some U.S. firms into the 1970's. New York State Department of Labor (NYS DOL), Division of Industrial Hygiene, investigated radium facilities in the 1940's to evaluate radium hazards and make recommendations for worker protection, but these were not always followed by industry. In December 1955, Industrial Code Rule 38 was promulgated which set limits for external doses and air concentrations and required that all radiation facilities be registered with the Department. Many firms, with operations dating back to the 1930's, had major difficulties in revamping their operations to achieve compliance with these new standards. This paper will review NYS DOL investigations and enforcement activities toward Code compliance of three major radium corporations which were engaged in radium refining, reclamation, source manufacture, and radium dial painting. The summary includes review of worker dosimetry, external exposures, and air concentration data compiled from reports of on-site inspections and will briefly review several autopsy reports involving alleged radiation injury.

INTRODUCTION

During radium's heyday in the 1920's, 30's, and 40's, many firms which used, sold, or leased radium were located in New York State, a commercial and manufacturing center with a skilled work force. Approximately 100 firms were known to have possessed radium for use in dial painting and refinishing or distribution of luminous timepieces and instrument dials. Some of the larger companies used radium in the production of sources for medicine, industry, and self-luminous paints which were widely used for defense contracts during World War II (Morewitz and Stebbings 1985).

Industry utilized radium for such varied applications as static elimination, smoke detection, lightning arresters, well logging, radiography, electron tubes, electron capture detectors and gauges for vacuum determination, dewpointers and material thickness, and density and level gauges. Radium content of these devices ranged from 0.1 μCi to 10 mCi (3.7 kBq to 370 MBq) depending upon the use of alpha, gamma, or neutron interaction (Table 1) (Robinson 1968). Self-luminous paints were prepared with 10 to 70 μCi (0.370 to 2.590 MBq) radium per gram of zinc sulfide compound. The contents of radium needles and plaques for medical use ranged from 1 to 100 mCi (37 to 3700 MBq).

Table 1. Industrial radium devices.

Type of device	Radium content		
Gauges			
moisture (Ra-Be)	1-10	mCi	37-370 MBq
density	1-10	mCi	37-370 MBq
thickness	10-100	µCi	0.37-3.7 MBq
level	1-10	mCi	37-370 MBq
vacuum	100-400	µCi	3.7-14.8 MBq
humidity	6-200	µCi	0.22-7.4 MBq
Static eliminators			
industrial	3	mCi/in	0.044 MBq/cm Max
	25-50	µCi/in	0.36-73 MBq/cm
laboratory balances	6-20	µCi	0.22-0.74 MBq
Fire detection	0.1-40	µCi	0.004-1.50 MBq
Electron capture detectors	50-100	µCi	1.85-3.70 MBq
Electron tubes	0.1-10	µCi	0.0037-0.37 MBq

Ref: Robinson 1968

RADIATION PROTECTION GUIDELINES

Beginning in 1913, standardization of radium for medical purposes was done by the U.S. Department of Commerce, National Bureau of Standards (NBS) which measured nearly all radium offered for sale in the United States. The large amount of radium which passed through the NBS laboratory made it necessary to give considerable attention to the problem of radium protection (USDC 1938). Tragic manifestations of biological effects of ionizing radiation led to the formation of the Advisory Committee on X-Ray and Radium Protection in 1928 to make recommendations for the safe use and handling of radium. In 1946, the scope of the Committee was broadened and its name was changed to National Commission on Radiation Protection (NCRP) (USDC 1949). NBS Handbooks set forth voluntary radiation protection guidelines which emphasized good housekeeping techniques, medical examinations and dose limitation. As more information was obtained on biological effects, recommendations were periodically revised resulting in the downward trend of exposure limits as presented in Table 2. In 1941, NBS Handbook H27 (USDC 1941) set the maximum permissible body burden for radium at 0.1 µCi (3.7 kBq) which was equivalent to 1 pCi/L (0.037 Bq/L) of expired air in the breath.

In the 1930's, the State of New York Department of Labor (DOL) formed an Advisory Committee to develop a regulatory code for industrial uses of radium because their investigation of radiation hazards disclosed the need for more than voluntary guides (Kleinfeld and Abrahams 1964). A draft code was proposed in 1952 to include all ionizing radiation sources; it was promulgated in December 1955 as Industrial Code Rule 38 "Radiation Protection" (Kleinfeld 1964).

The Code required registration of radiation sources, competent personnel, instruction on radiation safety precautions, personnel monitoring, physical surveys, and record keeping. Safety criteria were established in terms of maximum permissible dose for external radiation exposure and limits for airborne contamination. Table 3 shows Code Rule 38 external dose limits from 1955 to the present. A major change to the Code in 1971 introduced contamination limits for release of vacated facilities for unrestricted use (Kleinfeld and Abrahams 1964; NYS DOL 1955).

Table 2. NCRP guidelines for external dose limitation.

Reference	Year	Daily (mGy)	Weekly (mGy)	Annual (mSv)
Handbook 59	1934	0.2 r (1.8)	1.0 r (8.8)	
Handbook 23	1938	0.1 r (0.9)	0.5 r (4.4)	
Handbook 27	1941	0.1 r (0.9)	0.5 r (4.4)	
Handbook 42	1949		0.3 r (2.6)	
			0.5 rep (4.7)	
Handbook 47	1951		0.3 r (2.6)	
			0.5 rep (4.7)	
Handbook 59 Addendum: April 15, 1958	1958			5 rem (50)
				3 rem/qtr (30)
				Lifetime limit
				5(N-18) rem (50(N-18)) mSv

r = roentgen
rep = roentgen equivalent physical (93 erg/g)
N = age in years at last birthday

Table 3. New York State Code Rule 38 dose limits.

Year Amended	External Dose
1955	300 mrem (3 mSv) per week
	3000 mrem (30 mSv) per 13 weeks
1962	5 rem (50 mSv) per year
	3 rem (30 mSv) per 13 weeks
	Lifetime limit 5(N-18) rem/y 50(N-18) mSv/y
1971	no change
1978	no change
1985	no change

N = age in years at last birthday

REVIEW OF RADIUM INVESTIGATIONS

In September 1959, New York State had 450 registered installations with 40 kCi (1480 TBq) of radionuclides and 3 000 pieces of radiation producing equipment (Kleinfeld and Udel 1959). Documented investigations by DOL representatives at radium facilities dating from the 1940's demonstrated the State's early efforts made in the interest of worker protection. Radiological health inspections, reinforced by Code Rule 38, continued and subsequent enforcement actions contributed to the dismantlement of the radium industry in New York. The following is a review of DOL files on U.S. Radium Corporation, Canadian Radium and Uranium Corporation, and the Radium Chemical Company, Inc.

UNITED STATES RADIUM CORPORATION

Background

U.S. Radium Corporation played a major role in American radium history. Prior to 1921, the company was known as Radium Luminous Materials Co. and occupied two New York locations for refining and radiochemical laboratories dating back to 1913 (Morewitz and Stebbings 1985). The firm was engaged in dial painting, reclamation of radium, and manufacture of self-luminous paint and finished products such as radium sources and neutron sources. In 1946, U.S. Radium developed the Ionotron static eliminator, a device for neutralizing static electricity by ionization. The Ionotron comprised an aluminum channel bar approximately 1.5 inches (3.8 cm) wide and of varying lengths from 1 to 8 feet (0.3 to 2.4 m) with up to 50 μCi (1.9 MBq) radium sulfate per linear inch (or 760 kBq/cm) (USRC). By 1952, there were approximately 400 plants using Ionotrons in the State (Mayers 1952a).

Dr. Victor Hess served as research director at the company's Orange, New Jersey plant from 1921 to 1923. In 1938, Dr. Hess joined the physics faculty at Fordham University following his return from Austria, where his work with cosmic rays earned him the Nobel Prize in 1936 (Landa 1987). He remained associated with the company and, in 1956, registered his Fordham laboratory with DOL in the name of U.S. Radium Corporation.

Company operations

From 1924 to 1954, the firm's luminous dial painting plant, located on Pearl Street in lower Manhattan, occupied five floors of a 10-story factory building. The site served as headquarters and center for distribution of radioactive products as well as radium processing activities. During World War II, the plant housed one of the largest dial painting and screening operations in the country and, at its peak, employed about 1000 workers. After the build up for the War effort, demand for radium dials decreased and the work force declined to 135 in 1945 (Kleinfeld undated; Mayers 1945). By 1947, hand painting was replaced by a screening machine in which application of luminous paint was entirely enclosed. One worker could print 2000 dials per day while 80 dials was a large number for hand painting. Parts of the equipment were cleaned each night in a tub of solvent, which was then taken to the laboratory where residual radium was recovered (Mayers 1947a).

The laboratory was located in a penthouse on Lafayette Street, another Manhattan location, where the firm engaged in preparation of radium luminous compounds and fabrication of radium-beryllium neutron sources, as well as reclamation of radium from old dials. Neutron sources, used in well logging, were fabricated in steel cartridges about 3 inches (7.6 cm) long. In a lead-lined exhaust booth, powdered beryllium was mixed with radium solution; evaporated and poured into steel shells; and then sealed with solder. The finished neutron source cost from $10 000 to $50 000 in 1947, but only five or six were manufactured up to that time (Mayers 1947b).

Inspections and worker dosimetry

Files dating from 1942 documented that DOL visited the U.S. Radium plant and recommended engineering controls, proper work techniques, safety precautions, and monitoring of workers' exposures by blood counts and breath radon determinations as suggested in the NBS Handbooks. Dial painting was performed in glass hoods with ventilation to avoid hazard from inhalation of radioactive dust and radon. Hazard from gamma rays was minimized by using small amounts of material, medical examinations, and supervised cleanliness. Hand washing was strictly enforced and ultraviolet lamps were employed to check hands and clothing for remnants of luminous paint (Mayers 1945; Heimann 1942).

A radon breath survey of the dial painters was conducted in April 1944 by Dr. Frank Hoecker of U.S. Public Health Service. Table 4 provides a sampling of results which ranged from 0.21 to 1.73 pCi/L (7.8 to 64 mBq/L) of radon in expired breath. Those workers whose breath analysis exceeded the tolerance level of 1 pCi/L (37 mBq/L) were removed from radium work (Hoecker 1944).

Table 4. Radon breath analysis for U.S. radium workers.

Worker	Period of Employment	Age	Radon Concentration	
	years	years	pCi/L	Bq/L
A	3	22	1.16 0.74	0.043 0.027
B	2	23	1.73	0.064
C	2	23	0.97	0.036
D	1.67	22	0.52	0.019
E	0.67	26	0.21	0.008

Ref: Hoecker 1944

In 1948, inquiry by DOL was made into the deaths of two U.S. Radium employees who died of aplastic anemia, potentially caused by exposure to radium. The cases were of particular interest because it was felt that their deaths resulted from external gamma exposure rather than internal deposition of radium in the body (Silson et al. 1955).

The first case (Worker F) was a 67-y-old man who worked for U.S. Radium from January 1944 until November 1944 in the shipping department as a packer of finished luminous buttons. Thereafter, he worked as assistant chemist in the laboratory engaged in reclamation of radium from spent luminous compound until he died of aplastic anemia on July 11, 1946. Breath analysis done by Dr. Hess on Worker F in November 1945 and June 1946 gave results of 1.2 and 1.4 pCi/L (44 to 58 mBq/L), respectively. Study of the bones, liver, and kidney at autopsy revealed radioactive substances were absent as reported by Dr. Alexander O. Gettler. The possibility of his anemia being due to internal radiation was ruled out by Dr. Harrison Martland at the Workmen's Compensation hearing in 1947 when he reported that there was no radium detected by geiger counter and the illness was not a radium induced anemia. The radiation injury claim was denied (Anonymous undated; Anonymous 1948).

The second case of aplastic anemia (Worker G) was that of a 24-y-old man who died on February 27, 1948 following 3.25 years as an assistant chemist in the compounding department. He was employed in the preparation of luminous compounds and screening work. The Workmen's Compensation case for Worker G was decided in favor of the claimant in 1949 (Anonymous undated; Silson 1949b). Autopsy revealed wide-spread hemorrhage as reported by Curphey (1948) The anatomical summary is given in Table 5. Study of tissue ash and bone for radioactivity reported by Dr. Gettler estimated that the deceased had a total body content of radium close to the accepted tolerance limit of 0.1 μCi (3.7 kBq). The findings were confirmed by Dr. Martland who concurred that insufficient radioactive material was present to have caused death (Anonymous undated; Silson 1948a). This man's radon breath analysis, performed by Dr. Victor Hess at his Fordham University laboratory in January 1948 showed 3.16 pCi/L (0.12 Bq/L) of radon (Table 6) (Silson 1949a). Gamma ray testing indicated a total of 0.358 ± 0.03 μCi (13 ± 1) of radium in the body (Silson 1948b).

The death of Worker G in 1948 caused renewed attention to the 1946 case of Worker F. That both men died from aplastic anemia with apparently insufficient radium body burdens to have caused fatality sparked the interest of Dr. John Silson, DOL Senior Industrial Hygiene Physician. He sought the opinion of Dr. Robley Evans of Massachusetts Institute of Technology (MIT) who advised that estimated gamma exposures of the deceased should be obtained (Evans 1948; Evans 1949; Silson 1948a; Silson 1949a).

In November 1948, Dr. Silson performed a survey of the U.S. Radium screening room and found a general level of 25 mR/h (0.22 mGy/h), although no radium operations were being performed at that time. Inside the hood, readings were above 120 mR/h (1.1 mGy/h), even though no radium was handled or stored (Silson 1948c). While it was not possible to make direct measurements of exposure rates at the

Table 5. Anatomical summary of U.S. Radium Worker G.

SKIN:	Diffuse petechial hemorrhage with areas of confluence.
CONJUNCTIVA:	Solitary petechial hemorrhage, left.
LYMPH NODES:	Diffuse lymphoid hypertrophy. Hemolymph nodes.
HEART:	Epicardial petechial hemorrhage. Endocardial hemorrhage.
DIAPHRAGM:	Interstitial hemorrhage, left leaf.
LUNGS:	Sub-pleural hemorrhage, bilateral. Pulmonary edema, right upper lobe.
SPLEEN:	Marked splenomegaly (390 grams).
KIDNEY:	Petechial hemorrhage. Perirenal hemorrhage, right.
BRAIN:	Intracerebral hemorrhage. Hemorrhage into wall of fourth ventricle.
BONE:	Hyperplasia of bone marrow.
GENERAL:	Massive intra-muscular hemorrhage, right gluteal region.
FINAL DIAGNOSIS:	Panhemocytopenia probably secondary to exposure to radioactive material.

Ref: Curphey 1948

Table 6. U.S. Radium Worker G radon breath analysis.

Date	Breath Radon Concentration	
	pCi/L	Bq/L
July 1945	3.11	0.115
December 1945	3.74	0.138
June 1946	3.80	0.141
November 1946	1.35	0.050
April 1947	1.00	0.037
January 1948	3.16	0.117

Ref: Anonymous

time of employment of both men, data was collected from a general study of the radium industry. Exposure rate from packaged radium dials was on the order of 400 mR/h (3.5 mGy/h) at contact with the case. Boxes of radium powder were found to emit rates of 750 mR/h (6.6 mGy/h) at contact. Estimated doses received by the deceased individuals were in the vicinity of 2 rem/week (20 mSv/week), about four times the tolerance limit of 500 mrem/week (5 mSv/week), but still insufficient to have caused the observed autopsy findings (Anonymous undated; Silson et al. 1955).

The current radiation protection guidelines emphasized prevention of ingestion and inhalation of radium and radon in the work place, but these deaths demonstrated that insufficient consideration was given to minimizing external gamma exposure. These cases refocused attention on external radiation hazards which led to requirements for personnel monitoring, use of meter surveys to identify major sources of exposure, lead shielding, and posting (Silson et al. 1955).

Decommissioning and decontamination

By 1954, U.S. Radium's dial painting operations had ceased in New York and it was engaged in transferring its operations to facilities in Morristown, NJ and Bloomsburg, PA. The firm registered the Pearl Street site with DOL in 1956 in compliance with the newly promulgated Code, but only the corporate office remained with a few demonstration sources; the plant no longer processed or stored radium. Later that year, the company vacated the space entirely and the building was then occupied by an envelope manufacturing firm.

In 1958, the subsequent occupant was instructed by DOL to commence clean up of radium contamination and complied to some extent. However, the occupant refused to perform a thorough decontamination because the building was scheduled for demolition, to be replaced by a new federal office building. The building was vacated in 1961, still contaminated, because there was no legal means to compel decontamination under the existing Code (Kleinfeld undated).

The U.S. General Services Administration (GSA) took title to the property and, in July 1962, the Pearl Street building was surveyed by a team of representatives from U.S. General Services Administration, U.S. Public Health Service, New York City Department of Health, and DOL. Hot spots, greater than 2 mR/h (18 μGy/h) were identified on several floors; the highest reading was 380 mR/h (3.3 mGy/h). DOL made recommendations which were implemented to remove all hot spots prior to demolition under supervision of a qualified radiation consultant. Salvaged scrap was surveyed to ensure that an acceptable level of 0.1 mR/h (0.9 μGy/h) at contact was maintained. Experts from Atomic Energy Commission's Health and Safety Laboratory in New York agreed to supervise the decontamination and required personnel monitoring for all workers (Harris 1962; Messite 1962).

By August 1962, all radium contamination had been properly packaged for disposal and authorization to proceed with demolition was granted by the New York City Health Commissioner (Baumgartner 1962). The project was an example of federal, state, and local cooperation in guarding the health and safety of workers and the general public. The experience led to amendment of Code Rule 38 in 1962 requiring decontamination of vacated installations, although contamination limits were not promulgated until 1971 (Kleinfeld and Abrahams 1964).

CANADIAN RADIUM AND URANIUM CORPORATION

Background

Eldorado Gold Mines Ltd., a Canadian company, claimed that all needs of the Western Hemisphere for radium and associated minerals could be supplied wholly from within the hemisphere with their successful development of rich-ore reserves at Great Bear Lake in the Canadian sub-arctic. In the 1940's, the company claimed to be the largest producer of radium in the world and sole producer in the Western Hemisphere with a radium refining facility at Port Hope, Ontario, and treating, servicing, and research facilities in the United States and Canada. In March of 1941, Eldorado Gold Mines Limited announced the appointment of Canadian Radium & Uranium Corporation (CRUC) of New York City as their exclusive sales representative in the United States.

CRUC operated a laboratory in Manhattan, under the name American Radium Industries, Inc., where radioactive luminous compounds were manufactured and radium sulphate was prepared for medical sources. Its activities included manufacture of radium solution, filling needles with radium, mixing radium ointment, and chemical processing from bromide to sulphate. Dial painting was also carried out at several other locations in Manhattan (McMahon 1944a).

Starting in 1942, CRUC operated a radium refinery in Mount Kisco, NY in Westchester County. The refinery, a two-story concrete factory building, extracted radium and other nuclides from the "white

cake" residue remaining after extraction of uranium from pitchblende and other ore mined in Canada. During World War II, the Mt. Kisco refinery was engaged in various activities connected with supplying radioactive material to the Manhattan Project in addition to supplying radium for industrial and medical uses. A separate calibration laboratory was located in Mt. Kisco in a commercial and residential building (Abrahams undated).

Company operations

The refining process consisted of the standard series of precipitations, leachings, and fractional crystallization used by Madame Curie, but it was commercialized in tanks, open vats, and large evaporation dishes (Abrahams undated). The raw material known as "white cake" was delivered in barrels weighing 450 lb (203 kg) and containing sludge taken from milled ore which had already gone through one step of the refining process at the mines. The sludge was boiled in tanks, and an acid was added once the radium was in solution.

In the upstairs crystallization lab, the radium solution was poured into tanks and hydrobromic acid was added. When mixed, it was filtered, poured into pails, and carried to evaporation tanks enclosed in glass with exhaust pipes to the roof. When evaporation was complete, crystals were shelled out on dippers and taken to the recrystallization room. On a long table, crystals were placed in quartz bowls with water and heated on gas rings. Nitric acid was added followed by a series of precipitation, evaporation, and filtering through which the crystals became finer and finer until like powder. Final crystallization was done in a fume cabinet by two chemists in the finishing laboratory. Finished radium bromide, in very fine crystals or powder form, was poured from small retorts into small glass tubes, sealed for shipping, and stored in a refrigerator. Metallic radium was kept in glass tubes in a circular lead safe for experimental use (McMahon 1944b).

According to a company spokesman, CRUC, in its early days, was capable of producing up to 10 g of radium monthly with approximately 40 workers (NYS DOL undated). The total amount produced declined to about 10 to 15 g of radium per year from 1944 to 1946. In 1946, uranium was declared a strategic material and the company was cut off from its source of "white cake". Subsequently, the firm engaged in reclamation of radium from watches, instrument dials, and medical applicators obtained from outside sources and production fell to about 4 to 5 g annually from 1947 to 1957 (Kleinfeld 1958). The work force dwindled to six employees and the operation continued until 1958 (Abrahams undated).

Inspections and worker dosimetry

Investigation by DOL in the mid 1940's found conditions at the Mt. Kisco plant to be unsatisfactory due to poor hazard control, and recommendations were made to reduce radiation exposures which included revamping the ventilation system. However, no action against the company was taken at that time due to the war emergency (Abrahams undated).

In 1952, DOL investigation intensified to document the deplorable conditions and to urge personnel monitoring and reduction of hazards. Instrument surveys found that exposure rates ranged from 400 mR/h (3.5 mGy/h) at the reception desk to 12 R/h (105 mGy/h) on the second floor. Radon air concentration averaged 410 pCi/L (15 Bq/L) and 346 pCi/L (12.8 Bq/L) on the first and second floors (Table 7). Surface contamination, on the order of 1 $\mu Ci/cm^2$ (37 kBq/cm^2), was widespread throughout the building (Mayers 1952b; Mayers 1952c).

During inspection in January 1954, clinical observation by DOL representatives revealed that two employees had radium burns of the hands; complete blood data revealed a mild to moderate anemia in four workers. Radon breath analysis revealed levels at tolerance or higher for five employees (Tabershaw 1954). Dosimetry records from 1957 revealed that personnel exposure exceeded the limits of 300 mrem per week (3 mSv/week) and 3 rem per quarter (30 mSv/quarter). Records for the thirteen-week period beginning March 3, 1957 and ending May 31, 1957 showed total external doses were as high as 7510 mrem (75 mSv) as given in Table 8 (Paul 1957a). Weekly doses for two periods in June and September 1957 are given in Table 9 showing that several workers exceeded the limit (Abrahams 1957).

Dr. Frank Hoecker, in cooperation with DOL, conducted radon breath studies in 1943, 1944, and 1945 which gave results for several workers as high as 45 times tolerance. Table 10 gives a sampling of the 1945 values. Workers Q through U all exceeded 1 pCi/L (37 mBq/L) (Hoecker undated). The highest radon analysis was recorded for Worker T who became chief chemist after Worker Q returned to Canada. In September 1957, Worker T died following illness and hospitalization. He had been employed by CRUC since 1944 (Kleinfeld 1958). His autopsy report listed cause of death as acute leukemia, anemia, and thrombocytopenia believed due to prolonged radiation exposure from radium. The anatomical diagnosis is given in Table 11 (DiMaio 1957).

Table 7. CRUC, Mt. Kisco plant gamma survey 1952.

Location	Exposure Rate	
	mR/h	mGy/h
First floor:		
Reception desk	400	3.5
Hallway	400	3.5
Locker room	60	0.53
Shower room	11	0.096
Second floor:		
General work area	100	0.88
Evaporation dish	5000	44.0
Finishing lab hood	1000	8.8
Finishing lab desk	75	0.66
North workroom, tanks	50	0.44
wooden boxes	1000	8.8
center	150	1.4
Evaporation dish	12000	110.0
Lab exhaust on roof	20	0.18
Outside:		
Incinerator building	200	1.8
Cans in yard	400	3.5
Fence	30	0.26
	Radon Concentration	
	pCi/L	Bq/L
Radon air sample location		
3 m from building	4	0.148
First floor	410	15.2
Second floor	346	12.8

Ref: Mayers 1952b; Mayers 1952c

Decommissioning and decontamination

On May 28, 1957, a court order was issued in the County of Westchester, directing CRUC to survey the installation and cease permitting employees to receive radiation exposures in excess of the legal limits. Investigations continued at the Mt. Kisco plant to assess compliance with the court's directive. Prosecution was initiated on June 14, 1957 against CRUC for failure to make a survey of the plant and failure to prohibit workers from receiving doses in violation of Code Rule 38. The firm stepped up efforts to decontaminate the plant and hired an outside consultant as radiation safety officer to perform surveys and conduct safety training. Lead shielding was supplied a fume hood was erected in the finishing lab, and workers were given respiratory protection. Exposure rates around the facility were reduced, but compliance efforts were a financial hard-ship for the firm. On July 25, 1957 the defendant withdrew the plea of not guilty and entered a plea of guilty to the charges. The president announced the decision to cease operations and all work with luminous compounds was transferred to Manhattan. Dismantlement of the lab and decontamination was begun by the remaining CRUC employees and DOL continued inspection of the project which continued into the 1960's (Paul 1957b; Beiser 1959).

Table 8. CRUC worker exposures for 13-week period, March 3 to May 31, 1957.

Worker	mrem	mSv	
H	7510	75.10	
I	7520	75.20	
J	1780	17.80	May 13 - 31, only
K	3256	32.60	
L	3090	30.90	Mar. 4 - Apr 26 only
M	4390	43.90	
N	3148	31.50	

Ref: Paul 1957a

Table 9. Film badge readings on CRUC workers during two periods.

	June 10-14, 1957				September 3-9, 1957			
Worker	Beta		Gamma		Beta		Gamma	
	mrep	mGy	mrem	mSv	mrep	mGy	mrem	mSv
K	40	0.35	310	3.1	---	---	---	---
M	210	1.80	580	5.8	195	1.71	290	2.9
O	50	0.44	470	4.7	---	---	---	---
N	30	0.26	520	5.2	---	---	270	2.7
P	390	3.40	880	8.8	270	2.37	290	2.9
H	340	3.00	700	7.0	145	1.27	270	2.7
I	1400	2.30	850	8.5	90	0.79	220	2.2

Ref: Abrahams 1957

In 1966, the Mt. Kisco Urban Renewal Agency purchased the site and the building was demolished. Contaminated equipment and debris were disposed of at a radioactive waste site in New York State and the building rubble and soil was carted to a local landfill. These efforts were carried out under surveillance of the State and County Health Departments, U.S. Public Health Service, and DOL (NYS DOH 1966).

CRUC consumed more time and effort by DOL than any other radiation operation in the State of New York at that time. Legal proceedings proved to be a labor-intensive and time-consuming method to achieve compliance with Code Rule 38. However, the case demonstrated the effectiveness of State regulations and provided an example for other firms using ionizing radiation.

RADIUM CHEMICAL COMPANY, INC.

Background

Radium Chemical Company was the sales division of America's largest radium producer, the Standard Chemical Company of Pittsburgh, established in 1909 by Joseph Flannery. It was one of the world's largest

Table 10. Radon breath analysis for CRUC workers.

Worker	Occupation	Date	Radon Concentration	
			pCi/L	Bq/L
Q	Chief chemist, Mt. Kisco	Feb. 10, 1945	13.5	0.500
R	Laborer, Mt. Kisco	---	10.7	0.396
S	---	---	10.2	0.377
T	Chemist, Mt. Kisco	Mar. 10, 1945	45.7	1.690
		Mar. 23, 1945	31.4	1.160
		Mar. 26, 1945	35.0	1.300
		Mar. 31, 1945	33.8	1.250
		Apr. 6, 1945	21.6	0.799
U	---	Mar. 21, 1945	3.2	0.120
	16th Street	Mar. 24, 1945	4.1	0.150

Ref: Hoecker undated

Table 11. CRUC Worker T autopsy report.

Purpuric hemorrhages of the body and viscera.

Hemorrhage in the ethmoidal sinuses and posterior nasal fossa.

Pulmonary edema and congestion with hemorrhage, confluent bronchopneumonia and bilateral fibrinous pleurisy.

Leukemic infiltration of the heart.

Minimal coronary arteriosclerosis.

Atherosclerosis.

Hepatosplenomegaly due to acute myelogenous leukemia.

Marked hypoplasia of bone marrow of the entire body.

Testicular atrophy.

Radium burns of left hand and fingers with atrophy.

Ref: DeMaio 1957

users of radium in the manufacture of radium needles and radon tubes for cancer therapy as well as for industrial gauges and radioluminous paints. In 1924, when radium from rich Belgian Congo ores dominated the market, Radium Chemical represented Union Miniere du Haut Katanga of Brussels, Belgium, for importation and distribution of radium in the Western Hemisphere (Landa 1987). The firm had several subsidiaries around the United States under various names which were engaged in radium dial painting.

The company's New York facilities were clustered in midtown Manhattan where the firm operated a radiochemical laboratory, a source manufacturing facility, and a dial painting plant (Morewitz and Stebbings 1985).

Company operations

The radiochemical laboratory operated from 1938 to 1954 housed in a two-story brick building in midtown Manhattan and employed 11 chemists. In the manufacture of radium needles, radium bromide (60-80 mg) contained in glass ampules was dissolved in distilled water and de-emanated for about 12 h in a vacuum desiccator which removed the radon gas. The solution was filtered and sulfuric acid added to convert the radium bromide into an insoluble sulphate. The sulphate was filtered out and dried in a small gas oven to be packed into needles (Mayers 1949).

To pack needles, the operator worked in a hood with 9 to 10 mg radium sulfate powder in crucibles which was transferred to a micro-sized funnel leading into the bore of the cell to be filled. Residual salt was brushed from the crucible into the funnel with a small camel hair brush. A fine wire was used to pack the radium salt into the cell after which a plug was driven into the opening with a small mallet. An average of eight cells were filled at one operation and then transferred to another worker who, beneath an exhaust hood, placed them into gold needles which were sealed and machine lathed to proper size. All equipment in this process was provided with exhaust hoods connected by vents to a large duct which was exhausted to the outside air. The needles, containing approximately 10 mg of radium salt, were stored in a shielded safe for 30 d after which they were calibrated on the second floor and stored for shipment. Radium needles were leased or sold to hospitals and physicians throughout the Western Hemisphere (Mayers 1949).

Radon gold seeds for implants were made on the second floor using an apparatus designed by Dr. G. Failla who was the consulting radiophysicist for the firm. By means of this apparatus, 0.75-mm diameter, 24-carat gold capillary tubing was filled with the required amount of radon liberated from 3 g of radium bromide solution stored in a lead-shielded safe. The tubing was then sealed and cut to size. Escaped radon gas was exhausted to the outside air (Mayers 1949).

In 1955, Radium Chemical Company moved to a larger facility in Woodside, New York in the County of Queens, and DOL continued routine on-site inspections. The building was a well constructed, one-story facility; however, the flow of operations constituted a potentially hazardous situation in that offices, locker room, and lunchroom were interspersed among operational areas (Kleinfeld 1955). The firm's registration, dated February 1956, listed 75 Ci (2.8 TBq) of radium as the maximum quantity on hand (NYS DOL 1956). At this site the company continued to prepare radon seeds for implants with the Failla radon apparatus. Radium sealed sources for medical and industrial uses and radium bromide powder for preparation of self-luminous paint were imported from Union Miniere in Belgium. The firm provided reconditioning services for the needles which included straightening, polishing, and sharpening.

Over the next two decades, the firm's radium operations contracted as physicians and industry turned to cobalt-60, iridium-192, and cesium-137 as substitutes for radium sources, and to gold-198 and iodine-125 instead of radon seeds. The firm processed tritium luminous paint as a substitute for radium when it was authorized by the U.S. Atomic Energy Commission in 1960 (NYS DOL 1963).

Eventually, the radon apparatus, the last remaining commercial radon plant in the United States, was ordered by DOL to be shut down and disposed of in 1981. The special glass in the Failla apparatus had been in use for approximately 25 years and had to be replaced. The firm suggested a steel radon system, but its approval by DOL was denied because of the firm's sloppy housekeeping.

Inspections and worker dosimetry

Inspection by DOL in 1949 at the Manhattan laboratory determined that potentially hazardous conditions existed because no radiation monitoring or personnel dosimetry had been conducted since the laboratory started functioning. Biological monitoring was done by means of monthly blood counts on workers who put in four seven-hours days per week. Heavy lead shielding was provided for all handling processes. Recommendations were made for the firm to survey the entire laboratory and provide a regular film badge program with wrist badges for the chemists. A program of periodic breath radon sampling was suggested for all workers (Mayers 1949).

With the advent of a more formal regulatory framework in 1956, all radiation workers at the Woodside plant were required to wear film badges, and management was required to keep exposures within prescribed limits. Radiation monitoring of workers required modification of work procedures and rotation of personnel, as workers continually approached the limit of 0.3 rem (3 mSv) per week and 15 rem (150 mSv) per year. Review of DOL files provided

an overview of dosimetry data for Radium Chemical Co. workers as given in Table 12. Seven workers out of 44 employed at the plant over a 33-y period exceeded 100 rem (1 Sv) lifetime dose and two workers exceeded 200 rem (2 Sv). Whole body counts were done on these workers at New York University, and all were found to be less than the permissible limit except for one value of 133 nCi (4.9 kBq); but a repeat body count revealed 65 nCi (2.4 kBq).

Decommissioning and decontamination

The beginning of the end came in September 1982 when DOL issued an Order to Comply to Radium Chemical which mandated that the company provide an inventory of contaminated gold radon seeds. When it was discovered that the company could not account for the contaminated gold, a second Order to Comply was issued in January 1983 which directed suspension of all activity involving radioactive

Table 12. Radium Chemical worker dosimetry.

Worker	Occupation	Years with firm	Whole body count	Lifetime exposure
			nCi ^{226}Ra (Bq)	rem (mSv)
V	Radon pump	26	58.8 (2175.6)	162.2 (1622)
W	Shipping	13	---	27.4 (274)
X	Radium paint	14	19 ± 0.6 (703 ± 22)	208.7 (2087)
Y	Radium sources	34	133 ± 2 (4920 ± 74)	170.8 (1708)
			65 ± 0.6 (2410 ± 22)	
Z	---	7	---	114.1 (1141)
AA	Glassblower	48	31 ± 0.6 (1150 ± 22)	244.9 (2449)
BB	---	--	---	155.9 (1559)
CC	---	--	83 (3070)	16.5 (165)
DD	Radon pump	6	24 ± 1 (888 ± 37)	35.1 (351)
EE	Radon pump/ Radium sources	35	84 ± 1 (3110 ± 37)	194.7 (1947)
			62 ± 1 (2290 ± 37)	
FF	Janitor/ driver	25	19 ± 0.5 (703 ± 19)	22.5 (225)

materials. In May 1983, the company was compelled to dispose of its inventory and decontaminate the plant.

Radium Chemical appealed and made attempts to clean up contamination in the building, to encapsulate leaking sources, and to lower radiation levels but their efforts, as assessed by DOL, were inadequate to permit resumption of business. In 1987, the State denied the firm's application for a new license and ordered the disposal of all radioactive sources by January 1988 and decontamination of the building. A task force of state, city, and federal agencies was formed to monitor the situation through weekly inspections.

The company was unable to find a buyer for its inventory of radium sources, and it did not have the financial means to pay the initial estimate of $1 000 000 for disposal at a licensed facility. The firm did dispose of 4 Ci (150 GBq) of radium mainly from nasopharyngeal applicators which were known to be poorly encapsulated. In July 1988, the owner of Radium Chemical agreed to a Consent Order, issued by the New York State Supreme Court, by which he formally abandoned the premises and all its contents. This action enabled the State to request the U.S. Environmental Protection Agency (EPA) to take remedial action under Superfund, the Federal program to clean up abandoned hazardous waste sites (NYS DEC undated).

In August 1988, EPA initiated Superfund Emergency Action to secure the site and to determine the extent of the radium problem. The radium storage vault held approximately 10 000 radium-filled needles contained in lead pigs and safes. The inventory included radium-beryllium neutron sources stored in the inner vault. Source removal began in June 1989, and 120 Ci (4.4 TBq) of radium were transported to Beatty, NV in 36 containers. Another 150 drums and steel boxes of contaminated debris were removed to a low level radioactive waste disposal site in Richland, WA. The final shipment, made in October 1989, ended phase one of the EPA project and cost $5 million (Kahn 1989). In November 1989, the Radium Chemical site was added to the Superfund National Priority List for remediation. After evaluation, EPA decided that the appropriate action was to decontaminate, demolish, and dispose of the entire building. This action commenced phase two in the spring of 1991 for which $20 million was appropriated by the federal government for demolition which was completed in 1992.

The story of radium in America began and ended with the Radium Chemical Company. Its source inventory was estimated to be about 10 % of the known radium that existed in the Western Hemisphere. Disposal of 120 Ci (4.4 TBq) of radium wrote the final chapter of the once thriving radium empire in America.

EPITAPH

The discovery of radium paved the way for present-day methods of cancer therapy and many industrial applications. As reactor-produced radionuclides came into common use, nuclides such as ^{60}Co and ^{192}Ir replaced radium for cancer therapy. Tritium and ^{147}Pm are now used in luminous paints. Industrial devices for radiography and level gauges contain ^{192}Ir, ^{137}Cs, and ^{60}Co among some of the more common nuclides in present use.

The radium era has seen its days come and go in New York from the welcome of Marie Curie in 1921 on her American visit to the disposal of the last large supply of radium in 1989. The experience with the radium industry gave rise to a body of radiation safety regulations, handling procedures, and decontamination standards which have permitted work with radionuclides to proceed with minimal hazard. Application of these regulations to former radium sites today necessitates, in some cases, decontamination which is expensive and labor intensive for both the contractor and regulator but is proving to be a growth industry.

REFERENCES

Abrahams, A. Unpublished report M-434-57. New York State Department of Labor (NYS DOL), CRUC file; December 11, 1957.

Abrahams, A. Outline of Canadian Radium and Uranium Corporation Story, Unpublished report. NYS DOL, CRUC file; undated.

Anonymous. External radiation exposure in the radium dial printing industry, Unpublished report. NYS DOL, U.S. Radium Corp. file; undated.

Anonymous. Unpublished summary of Workmen's Compensation Case Report No. 46058088. NYS DOL, U.S. Radium Corp. file; circa 1948.

Baumgartner, L. Commissioner, New York City Department of Health, letter to M. Pollack. NYS DOL, U.S. Radium Corp. file; August 7, 1962.

Beiser, A. Letter to M. Kleinfeld. NYS DOL, CRUC file; December 16, 1959.

Curphey, T. Autopsy Report Case No. 48-117. Meadowbrook Morgue, Medical Examiner's Office: NYS DOL, U.S. Radium Corp. file; February 27, 1948.

DiMaio, D. J. Autopsy Report Case No. 6702. City Mortuary New York; September 5, 1957.

Evans, R. D. Letter to J. Silson. NYS DOL, U.S. Radium Corp. file; June 3, 1948.

Evans, R. D. Letter to J. Silson. NYS DOL, U.S. Radium Corp. file; May 5, 1949.

Harris, S., U.S. Public Health Service, unpublished report. NYS DOL, U.S. Radium Corp. file; July 10, 1962.

Heimann, H. Unpublished report M-341-42. NYS DOL, U.S. Radium Corp. file; October 27, 1942.

Hoecker, F., U. S. Public Health Service, unpublished report on chemists in radium plants 1943, 1944 and 1945. NYS DOL, CRUC file undated.

Hoecker, F. U.S. Public Health Service, unpublished table of breath radon analysis. NYS DOL, U.S. Radium Corp. file; April 1944.

Kahn, M. Cleaning up a New York hot spot. U. S. EPA Journal; September/October 1989.

Kleinfeld, M.; Abrahams, A. Administrative experience with decontamination of vacated radiation installations. J. Occup. Med. 6: 255; June 1964.

Kleinfeld, M. Early regulatory effort (1930-1945). J. Occup. Med. 6: 179; April 1964.

Kleinfeld, M.; Udel, M.. The Industrial Code as a tool in radiation protection. A.M.A. Arch. Indust. Health 20: 199-201; Sept. 1959.

Kleinfeld, M. Unpublished report. NYS DOL, Radium Chemical Co. file; October 11, 1955.

Kleinfeld, M. Unpublished report C-40-58. NYS DOL, CRUC file; June 26, 1958.

Kleinfeld, M. Unpublished memorandum. NYS DOL, U.S. Radium Corp. file; undated.

Landa, E. R. Buried treasure to buried waste: the rise and fall of the radium industry. Colorado School of Mines, Quarterly Vol. 82, No. 2; 1987.

Mayers, M. Industrial radiation hazards in New York State. New York State Department of Labor Monthly Review, Vol. 31, No. 3; February/March 1952a.

Mayers, M. Unpublished report M-648-45. NYS DOL, U.S. Radium Corp. file; October 10, 1945.

Mayers, M. Unpublished report M-138-47. NYS DOL, U.S. Radium Corp. file; July 7, 1947a.

Mayers, M. Unpublished report M-151-47. NYS DOL, U.S. Radium Corp. file; September 5, 1947b.

Mayers, M. Unpublished report M-194-48. NYS DOL, Radium Chemical Co. file; January 27, 1949.

Mayers, M. Unpublished report M-2-51. NYS DOL, CRUC. file; April 15, 1952b.

Mayers, M. Unpublished report M-132-52. NYS DOL, CRUC file; July 22, 1952c.

McMahon, M. Unpublished report M-179-44. NYS DOL, CRUC file; April 29, 1944a.

McMahon, M. Unpublished report M-281-44. NYS DOL, CRUC file; June 12, 1944b.

Messite, J. Unpublished memorandum to M. Kleinfeld. NYS DOL, U.S. Radium Corp. file; July 16, 1962.

Morewitz, S.J.; Stebbings, J.M. . Inventory of radium sites in the United States. Human Radiobiology Section, Argonne National Laboratory. Unpublished report; 1985.

NYS DEC (New York State Department of Environmental Conservation). Radium Chemical Company background sheet, unpublished report. NYS DOL, Radium Chemical Co. file; undated.

NYS DOH (New York State Department of Health). Unpublished draft news release. NYS DOL, CRUC file; October 5, 1966.

NYS DOL (New York State Department of Labor), Division of Industrial Hygiene, Unpublished report Case No. 3525-53. NYS DOL, CRUC file; undated.

NYS DOL (New York State Department of Labor). Part 38 of Title 12 of the Official Compilation of Codes, Rules and regulations of the State of New York, Industrial Code Rule 38, Ionizing Radiation Protection; December 1955, Amended 1962, 1971, 1978 and 1985.

NYS DOL (New York State Department of Labor). Radioactive Materials License No. 0344-0120. NYS DOL, Radium Chemical Co. file; August 8, 1963.

NYS DOL (New York State Department of Labor). Radioactive Materials Registration, LR-0120. NYS DOL, Radium Chemical Co. file; February 7, 1956.

Paul, G. Unpublished report MR-340-57. NYS DOL, CRUC file; June 28, 1957a.

Paul, G. Unpublished report MR-537-57. NYS DOL, CRUC file; December 20, 1957b.

Robinson, E. The use of radium in consumer products. U.S. Department of Health, Education and Welfare, Public Health Service; November 1968.

Silson, J. Letter to R. D. Evans. NYS DOL, U.S. Radium Corp. file; May 18, 1948a.

Silson, J. Letter to R. D. Evans. NYS DOL, U.S. Radium Corp. file; April 25, 1949a.

Silson, J. Letter to T. J. Curphey. NYS DOL, U.S. Radium Corp. file; December 8, 1949b.

Silson, J. Unpublished notes. NYS DOL, U.S. Radium Corp. file; January 19, 1948b.

Silson, J. Unpublished report C-137-48. NYS DOL, U.S. Radium Corp. file; November 12, 1948c.

Silson J.E., Kleinfeld, M.; Harris, S.J. External radiation exposure in the radium dial painting industry. A.M.A. Arch. Indust. Health 12: 503-508; 1955.

Tabershaw, I. Unpublished report M-207-53. NYS DOL, CRUC file; April 1, 1954.

USDC (U. S. Department of Commerce). Radium protection. National Bureau of Standards Handbook H23; August 25, 1938.

USDC (U. S. Department of Commerce). Safe handling of radioactive isotopes. National Bureau of Standards Handbook 42; September 1949.

USDC (U.S. Department of Commerce). Safe handling of radioactive luminous compound. National Bureau of Standards Handbook H27; May 2, 1941.

USRC (United States Radium Corporation). Functioning, installation and maintenance of ionotron static eliminators, unpublished pamphlet. USRC, Morristown, NJ, NYS DOL, U.S. Radium Corp. file; undated.

Environment International, Vol. 19, pp. 491-501, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

THE ORIGIN AND EARLY DEVELOPMENT OF THE BELGIAN RADIUM INDUSTRY

A. Adams
University of Ghent, B-9000 Ghent, Belgium

EI 9301-108 M (Received 18 January 1993; accepted 8 April 1993)

Radium was an important export product of Belgium between 1922 and the second World War. The uranium extraction of the ore mined in the Belgian Congo and the production of pure radium were monopolised by two Belgian companies, the Union Minière du Haut Katanga (U.M.H.K.) and the Société Générale Métallurgique de Hoboken (S.G.M.H.). This work is based on secondary historical sources such as the annual reports of both companies, economic reports of the Belgian and foreign Governments, both Belgian and foreign specialised and popular press reports, and the technique of oral history. Particular emphasis was placed on the special relationship between U.M.H.K. and S.G.M.H. and their efforts to create a market to match the expanding radium production after 1922.

INTRODUCTION

The Belgian radium industry has never been studied in detail, not even by Belgian scholars. The fact that little research has been performed on this particular industry is odd, as no other Belgian export product equalled the importance of radium in the period between 1922 and 1938. In this period, marked by severe export restrictions and protectionism, the Belgian producers succeeded in opening up all frontiers with their product and supplied most of the world market.

Several reasons can be found to explain this apparent indifference of scholars to the subject, the most important one being the fact that no primary historical sources are available for a historical and economical analysis of this former Belgian production.

Both Belgian companies involved in the production of radium up until the 1960's have officially declared that they have not preserved any of the company archives dealing with their radium production. Their silence is the result of a gentlemen's agreement, concluded between all the governmental and nongovernmental parties involved in the former industry, not to release any information concerning a (by now) noncontroversial period like the 1920's and 1930's. This attitude of the Belgian producers is opposed to the attitude of radium producers of other countries such as France or the U.S., where the authorities or corporations always have been somewhat more willing to provide information on their former activities in this particular economic sector.

As a result of the policy of silence, Belgium's glorious pre-World-War-II radium monopoly is at present almost forgotten, and a description requires

a reconstruction using secondary historical sources: official Belgian and foreign governmental reports, annual reports of both industries, monographs, specialised and nonspecialised Belgian and foreign press reports, and interviews with former workers in the radium industry.

ORIGIN OF THE BELGIAN RADIUM INDUSTRY: THE COLONIAL CONNECTION

At the very basis of the Belgian radium industry lays the powerful colonial company Union Minière du Haut Katanga (U.M.H.K.), founded in 1906 on the advice of King Leopold II, to exploit the minerals in the Katanga region in the southern part of the Free State of Congo, shortly before it became the Belgian Congo colony. U.M.H.K. focused its activities on the exploitation of copper, tin, iron, tungsten, gold, silver, and coal. A first discovery of uranium ore was made accidentally in 1913 while prospecting for copper. This prospecting could not be completed as World War I broke out and the U.M.H.K. had to invest its full attention to copper production for the Allied war effort.

Nevertheless, new prospecting activities were initiated during the First World War and, accidentally again, U.M.H.K. discovered uranium ore in Shinkolobwe, a site situated in the huge copper concession zone of the company (AMC 1914-1916; Anonymous 1956a). A hastily performed laboratory investigation in the company's small laboratory in Lubumbashi and further investigations in Belgium proved the uranium ore to be of an exceptional richness (Buttgenbach 1920). In spite of the promising outlook of the mine, further prospecting had to wait as the company and the world economy were struck by a severe economic crisis. Until 1921, U.M.H.K. was not able to invest further capital or manpower in the exploration work.

It was clear, however, that the company considered the discovery of the uranium ore of the greatest importance as the mine during this whole period remained closely guarded and only a few people within the company were informed of its very existence.

New explorations and laboratory investigations in 1921 affirmed the quality of the ore (Schoep 1923). U.M.H.K. immediately considered the construction of a radium plant with a view to the commercialisation of the findings. A first twelve tons of uranium ore were shipped to Belgium and arrived in Antwerp in December 1921.

When U.M.H.K. began pit mining of the uranium ore, they encountered several problems. Firstly, they were confronted with the nonregular structure of the veins of uranium ore. The thickness of the ore veins varied from a few centimetres to several meters and contained ore of various grades (Thoreau et al. 1933). Due to the impressive quantity of ore available, U.M.H.K. transported only the richest pitchblende ore to Belgium. The uranium ore of lesser quality, mainly torbernite which was found everywhere in the mines, was dumped in the immediate surroundings of the mine (Anonymous 1957). Secondly, for the whole of its industrial production, the company had to face a serious shortage of native workers in the region of the Katanga. Various campaigns were organised to recruit workers from other provinces of the colony and even from outside the Belgian Congo. In spite of these recruitment efforts, the number of available Congolese workers could hardly keep track with the rising production figures and seriously undermined the industrial policy of the company during the 1920's and 1930's (Anonymous 1926).

As opposed to the copper mining activity, for which figures on the number of workers were regularly released, there are few figures available on the number of workers employed in mining less important minerals. No indications could be found for the Shinkolobwe uranium mine, except for the early years. In December 1921, at the outset of the uranium exploitation, 390 native workers were employed locally, under the supervision of four European agents. One year later, the number of workers had increased to 400 and the European staff increased to five. For 1923, the number of workers in the uranium production hardly differed from the previous year, being 325 and 5. Considering the fact that U.M.H.K. was primarily a large-scale copper producer and taking into account that the total workforce of the company in the Katanga region, for the same year, amounted to 7662 native workers and 760 nonnative employees, it is obvious that the Shinkolobwe mine was one of the smallest mining operations of the company (AMC 1922). Furthermore, on the basis of the exportation figures of uranium as illustrated in Fig. 1, there is no reason to believe that the workforce increased considerably at least until the middle of the 1930's, when uranium became important for strategic reasons. The biggest problem U.M.H.K. had to face concerning the new uranium production, however, was the geographical location of Shinkolobwe, situated in a remote and virgin tropical area, hundreds of miles away from any railway or road that could connect it to an industrial centre or a harbour. Especially during the first half of the 1920's, the geographical location of the mine seriously hampered full-scale uranium exploitation.

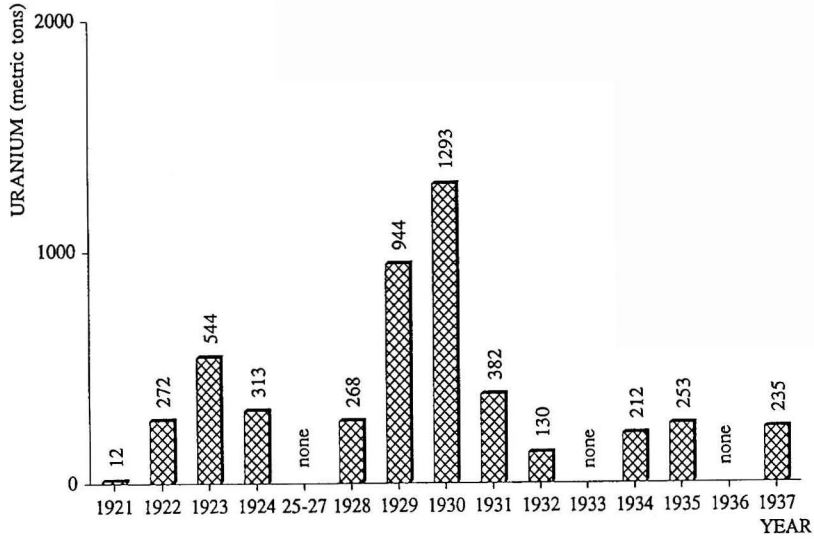


Fig. 1. Uranium exported from Belgian Congo by U.M.H.K. (1921-1937).

Until 1922, when trucks first became available, the uranium ore was carried by native workers from Shinkolobwe to the then small mining centre Jadotville, or to Kakontwe where U.M.H.K. exploited a limestone quarry (CSK 1923). In Jadotville or Kakontwe, both situated along the railroad, the ores were then loaded on the train. For several years, all the ores mined by U.M.H.K. were transported by rail to the port of Beira in Mozambique some 7 700 km distant. A map of the area is shown in Fig. 2.

Near the end of the 1920's, the construction of a national Congolese railroad was completed, connecting the north of the Katanga province with Port Francqui and later with the Congolese capital of Leopoldville. From Leopoldville, the ores were then shipped by barge to the Congolese port of Matadi (RF 1928). Although still covering the considerable distance of 2 251 km, this latter railroad had a major advantage: there was no other colonial territory crossed, and hence, there was no foreign taxation of the shipments. Although U.M.H.K. participated financially in its construction, the company did not regularly use the railroad for the ores mined in the southern part of Katanga province, including the Shinkolobwe ore. Although severely criticised, U.M.H.K. preferred, from 1931 on, to have these ores transported to the port of Lobito on the coast of Angola, using the Angolian (Benguela) railroad (UMHK 1932). This transfer was shorter than the national Congolese option (1860 km), cheaper although subjected to foreign taxation, and had the advantage that the ores did not have to be transshipped during the journey.

THE RADIUM PROCESSING INDUSTRY IN OOLEN, BELGIUM

The actual radium production plant had to be built in Belgium, as only in Belgium in that time, could a highly technological production process such as that for radium extraction be envisaged. The choice of the Belgian partner to extract the radium was quickly made. The radium factory became integrated within the structure of the Société Métallurgique de Hoboken (S.G.M.H.) in the company's division of Oolen in the province of Antwerp. The reasons were obvious. S.G.M.H depended, like U.M.H.K., on the financial holding Société Générale de Belgique. Both companies mutually owned a large amount of shares. Finally, S.G.M.H. already worked on colonial ores of U.M.H.K (RF 1918) and had ample experience in extraction of rare metals from minerals. The actual production process for radium was designed in the laboratories of the company between December 1921, arrival date of the first 12 tons of uranium ore in Antwerp, and March 1922. It was no simple operation as practical information on the extraction of

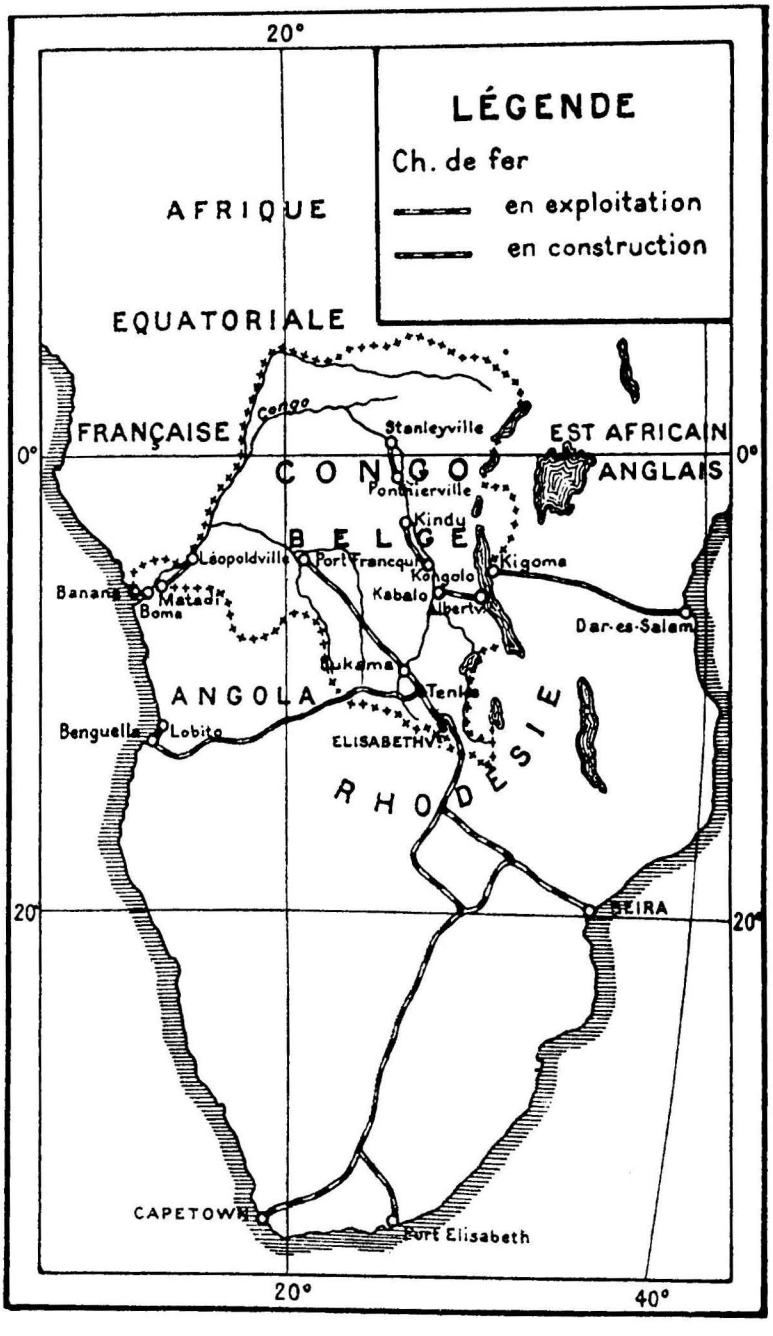


Fig. 2. Map of Central Africa in 1931.

uranium and the production of radium was not available to them, and the process had to be adapted specifically to the peculiar characteristics of the Congolese ore. Mineralogically, the Congolese ore consisted of a mixture of carnotite and an intricate mixture of many varieties of uranium ores from pitchblende to torbernite (a uranium phosphate) and included a series of different uranium silicates. Moreover, the composition proved to be highly variable as a function of place and depth of mining.

In the meantime, the construction of the radium factory was started with remarkable speed, even skipping the usual phase of semi-industrial production, going directly from the laboratory work to full-scale industrial production. The entire mechanical equipment of the new factory was manufactured in the workshops of the company's division in Hoboken. The construction of the first part of the factory was completed in July 1922 in such a hurry that the building architecturally showed a number of shortcomings. The ceiling of the large extraction hall, for example, reached halfway through a row of windows. Exactly one year after the first uranium arrived in Antwerp in December 1921, the first gram of Belgian radium was produced and photographed using the light originating from its radioactivity (Anonymous 1923).

Originally, the raw uranium ore was sent to Oolen via the port of Antwerp. After a few years, this bulk transport was eliminated when the ore was purified to a uranium/radium concentrate in a plant in Shinkolobwe. Further purification then took place in Oolen.

The production process can be divided in two stages: a chemical process aiming at separating Ra/Ba salts from metallic impurities (i.e., iron, copper, and lead), but also from anions such as phosphates, silicates, and aluminates; and a physical process (as the firm called it—it was in reality a physicochemical coprecipitation) that aimed to separate the radium from barium by way of a repeated fractionated crystallisation process. The entire process is poorly documented in the literature. It was based on the production process designed by Mme. Curie and A. Debierne. The following description is summarised from available reports issued in French by U.M.H.K. (Clerin et al. 1922) and supplemented by the information resulting from interviews with retired technicians from the radium plant (Adams 1988).

The chemical process

During the chemical process, the uranium ore was dissolved, precipitated, filtered, and washed to obtain a pure Ra/Ba solution with about 1 part of Ra to 120 000 parts of Ba. This process took place in two consecutive stages termed the major treatment and the minor treatment. This chemical process took place in the "buildings of the first treatment" (les batiments du premier traitement), mainly the large extraction hall, a two-story building.

The physical process

The physical process based on fractionated crystalisation was considered to be technologically refined for that time and included a "major cycle" (le grand fractionnement) containing 60 steps of recrystallisation in recipients of enamelled steel and a "minor cycle" (le petit fractionnement). The first step of the procedure was housed in one building, the second step in another. The distinction between the two cycles consisted in the scale of the operation and in the safety measures taken, the minor cycle being discontinuous batch type laboratory scale work on amounts of 2 to 3 g radium. The end result of the process was radium with a purity of 97% to 98%. The solid wastes remaining, including the uranium, were dumped in the immediate surroundings of the plant. Apart from these two buildings, the radium plant also included a small laboratory for analytical chemistry and radiation measurements that was located at a distance of 1 km of the actual factory, obviously in an attempt to prevent contamination. This laboratory is the only building remaining of the old radium factory, which was completely dismantled in the 1970's. Together with the production units for bismuth and cobalt, the radium production formed an entity called BIRACO (bismuth, radium, cobalt), which operated entirely separated from the copper smelter which was located on an adjoining site. Both factories were even separated by a brick wall for a long time.

Production campaigns

The radium factory did not function continuously but in distinct production campaigns. The length of such production campaigns was determined by the demand for radium but generally lasted for several months, leaving the plant inactive for a certain amount of time. Regularly, engineers of the radium factory went to Brussels to discuss the production campaigns with the management of U.M.H.K. When a production schedule was terminated, the radium workers were transferred to other production units of the Oolen factory. Before leaving the Oolen plant, the radium was sealed (as bromide salt) in small glass tubes and sent back to U.M.H.K., the actual owner of the product in Brussels. Before it could be commer-

cialised, the radium generally was worked up in therapeutical equipment such as needles. This final step of the long production process was performed in U.M.H.K.'s workshops in Brussels (de vereenigde werkhuizen). All the medical equipment was produced by these workshops as well. In the first half of the 1920's, the filling of the radium needles was performed by technicians that were only protected against radiation by means of gloves and a thin shield made of glass. Later, this operation took place in fume hoods and additionally arm protections were worn (UMHK 1925a; UMHK 1931).

Working conditions

From our interviews, it appeared that workers generally preferred to work in the radium factory as it appeared to them as a relatively clean and modern production site. The interviews also revealed that before the Second World War, few, if any, protective measures were taken to protect the workers against exposure to radiation and contamination with the element except for those working in the minor cycle of the physical process. The labourers wore the same clothing as those working in the other production units. They did not wear gloves, except when they came in direct physical contact with the solutions. There were no special medical examinations and no blood testing, and decontamination measures after work, such as taking a shower, were not obligatory. It was also clear that the workers were badly informed about the potential dangers of industrial radium production. Only at the end of the 1940's were serious measures against radiation initiated, and workers started to become concerned about their working conditions in the radium plant. Not until the end of the 1940's was ventilation introduced within the radium factory. Special bathing facilities for the radium workers were only available from the 1950's; before this period, radium workers had to take their showers together with the workers from the other production units. As noted above, protection measurements were only introduced in the minor cycle of the physical process, where the work was carried out by two engineers and one technician. The total number of workers involved in work in the radium factory is unknown; by the end of the 1930's, they must have been around a hundred. Before the second World War, there was no detailed information available on cases of radium poisoning in Oolen, but at the American Conference on Radium in Industry, held in 1928, German toxicologists did report radium poisoning cases in the Oolen plant (AANL 1928). Nevertheless, the company itself claimed in 1927 that "in spite of the considerable amount of radium produced, no accidents occurred since the fabrication had started" (Anonymous 1927).

COMMERCIALISATION OF BELGIAN RADIUM

The Belgian Government played a discreet but substantial role in the development of the country's radium industry. The interest of the Government in the exploitation of radioactive minerals was first expressed in the colonial regulation, presented by the Governor of the province of Katanga on the 8th of March 1923.

The regulation ordained that owners of concessions of diamond and radioactive minerals should consign the exploitation and commercialisation of their production to a consortium to be created by the Government. Such a state-controlled organization would ensure centralised control over the industrial production and the pricing policy of the companies involved. A similar organization already applied in the concessions for the exploitation of diamonds in the Kasaï province.

The Government defended the idea stating that the extraction of uranium and the exploitation of diamond were to be considered "exceptional industries", as they implied rare mineral resources that had a very limited market. Especially for the sale of radium, a similar coordinating/commercialisation structure was considered to be extremely important. The sale of the product was said not only to require smooth functioning marketing channels, but also considerable capital immobilisation to keep the price at a certain level (BOCB 1929).

The Decree, although severely criticized for its vagueness, was approved by the Colonial Council and published in the Congolese Official Journal of 15 September 1923. The law that resulted from this Decree stipulated that "if the government should decide to create an organization to assure without any profit the centralised exploitation or sale of uranium, radium, thorium, actinium, vanadium, niobium, titanium and tantalum, each holder of a concession would have to hand over the exploitation and the sale of these ores" (BOCB 1923).

The motives of the Government in providing for the possibility of creating such an organization are not completely clear. During the debate in the Colonial Council, the Minister of Colonies stressed on several occasions that the Government did not consider taking a direct hand in the exploitation or sale of radium. The issuance of the Decree would be a pro-forma measure, so that the legislation on radioactive minerals would conform the existing legislation

on diamonds applicable in the rest of the Colony. It is, however, difficult to match this motive with another assumption of the Government, that the establishment of such an organization would protect the Belgian diamond industries and those involved with radioactive minerals against severe economic competition. This assumption certainly did apply to the diamond producers but not to the producers of radium. In March 1923, the Belgian radium producers did not have to fear any competition on the world market and had already won their first commercial battle, striking a serious blow to the American radium industry by cutting the market price considerably.

The Belgian Government, holding a participation of 40% in the capital of U.M.H.K. and thus directly participating in the success of the new radium industry, might have wanted to safeguard its favourable position by means of the new law.

The actual commercialisation of the Belgian radium was taken care of by the commercial department of U.M.H.K. which was named "Radium Belge". Radium Belge assured 24-h service for the sale of radium packed in glass tubes. Radium worked up in medical equipment had to be ordered. In general, clients had to wait for 3 to 4 weeks for delivery. In comparison to former waiting periods, U.M.H.K. considered this period "remarkably short". Furthermore, Radium Belge assured its clients of the quality of their product by means of a certificate, guaranteeing its purity and contents. A purity of not less than 99.5 % of radium was guaranteed. The measurements were made by the Institut de Mesures des Substances Radioactives of the University of Brussels or by U.M.H.K. itself, but clients were free to request a measurement by an independent institution (UMHK 1925b). Numbers of the production batches and names and addresses of the buyers were carefully registered. The company still keeps these files, obliged to do so by the government, as these records serve to identify radium needles that still show up from time to time. A spokesman of Union Minière declared in 1987 to the author that these files are the only original documents U.M.H.K. has preserved on activities in the radium business (Adams 1988).

Marketing of a new product such as radium was an operation that was not well known in 1923, especially in Europe. The marketing techniques successfully applied by Radium Belge were largely inspired by the strategy of the American radium producers. U.M.H.K. position on the world market was also favoured by the foreign contacts of the Belgian Government, partner in the company's capital.

A serious problem for U.M.H.K. was the fact that radium therapy in the first half of the twenties was still quite new and experimental. This meant that U.M.H.K. not only had to sell a product but also had to actively promote its therapeutical uses. Secondly, almost all of the radium used originally in the medical world was privately owned and generally used in the treatment of wealthy patients, making radium therapy a very expensive technique to apply. The company immediately understood that if they were to build a lucrative production, radium therapy not only needed to be promoted but had to be democratised as well.

Several promotional activities were undertaken. Radium Belge published brochures on a regular basis demonstrating the latest models of medical radium instruments. Literature was distributed to universities, cancer institutes, individual doctors, and radiologists (Laude 1933). The company also launched the idea to organise specialized courses (although our research could not establish if such courses did indeed take place). Publicity on the Belgian radium could be found on a regular basis in several national and international medical magazines. Also, in a remarkably short span of time, U.M.H.K. succeeded in building up a commercial network with sales offices all over the world. The sale of the Belgian radium was delegated to exclusive sales agents appointed in different countries inside and outside Europe.

In view of democratizing the use of radium and thus increasing its sale, the Belgian producers stimulated the centralised use of larger amounts of radium in universities, cancer institutes in larger hospitals, and medical associations. They initiated this democratisation by donating the first grams of produced radium to the four existing university hospital centres of Belgium. Several hospitals of medical institutions offered low cost or even free treatment of the poor. U.M.H.K. introduced special payment facilities for hospitals and institutes or granted long-term loans of radium. With these initiatives, the company gathered a lot of positive publicity, but was severely criticised in the Belgian medical press by individual medical doctors who were the owners of small amounts of radium, and were afraid of losing their patients to the better supplied and equipped hospitals and cancer institutes. There are also several reflections in local medical journals that doctors tried to compensate for their limited individual financial capacity by starting up a cooperative movement, jointly procuring and using the radium they needed to keep up with the booming development of radium therapy. Although it would be unfair not to recognize the presence of a

certain altruism in U.M.H.K.'s motives, it may not be forgotten that the company realised that the development of an established network of medical institutions, well equipped in radium and offering a fair-price therapy to the public, was an essential condition to make their radium production a successful economical operation. Furthermore, the equipment of the Belgian medical sector was of U.M.H.K.'s prime concern, especially during the 1920's, as Belgium's infrastructure for radium therapy was to serve as an example for potential buyers abroad.

EVOLUTION OF PRODUCTION FIGURES AND PRICE STRUCTURE OF BELGIAN RADIUM BETWEEN 1922 AND 1938

To reconstruct temporal data on the production, exports, and prices of the Belgian radium and uranium, several sources were compared. Preference was given to the data provided by the Belgian Government or U.M.H.K. itself. Unfortunately, these institutions only sporadically released figures concerning the uranium and radium production. As a result, these data were supplemented, when possible, by estimates of foreign, primarily American sources. It is, however, difficult to judge the accuracy of those figures, the American estimates appearing in general to be too high.

Production figures for uranium and radium are practically unavailable. The figures that could be traced concern the amount of uranium exported from the Belgian Congo to Antwerp and the amount of radium that was sold by Radium Belge and are presented in the graphs (Figs. 1 and 3). Not even the annual reports of the company give production figures on a regular basis. The reason why U.M.H.K., even in this early period of production, chose to pursue this policy of secrecy on uranium and radium production is difficult to understand. For other branches of the company's production, regular figures were made available. Probably, this posture was dictated by purely commercial concerns, not wanting possible foreign competitors to have an idea on the company's radium stocks so that they would be able to criticize Belgian radium prices or influence the pricing structure.

The 1920's were a period of rapid expansion for the Belgian radium industry. In only a couple of years, the company succeeded in conquering the world market, and radium sales figures regularly increased until 1929.

Already by 1923, S.G.M.H. was believed to be able to attain a production level that could satisfy the total worldwide demand for radium (RF 1924). From 1923 on, during production campaigns, the factory produced about 4 g of radium monthly, supplying two thirds of the total world consumption (Anonymous 1925). In 1925, when the last American radium producer was forced to shut down production, the company estimated their sales to represent 80% of

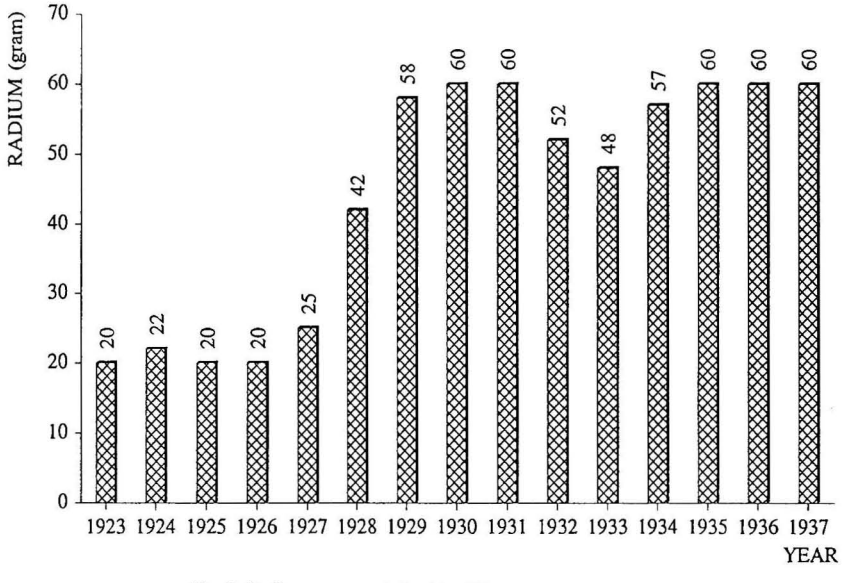


Fig. 3. Radium commercialised by U.M.H.K. (1923-1937).

world consumption (UMHK 1925 c). The English Radium Subcommittee estimated the monthly production of the Oolen factory not to be higher than 2 g or 3 g a month (CCR 1929). Even this was an impressive production rate never before attained.

During the entire 1920's and 1930's, U.M.H.K. was confronted with a production capacity considerably exceeding demand. This could explain the discontinuous production campaigns in the Oolen factory. The Shinkolobwe mine was also not in continuous production.

By 1924, radium sales lagged after production and stockpiles of radium were built. Stocks were estimated to have risen to an amount between 50 g and 134 g in 1924. American authorities calculated that the factory easily could have produced as much as 180 g of radium between 1922 and 1925. This figure came close to an estimate published in the Belgian medical press, with an estimated production of 150 g (USDC-BM 1925). During the last months of 1924, exploitation of uranium ore Shinkolobwe was suspended until the second half of 1925. Uranium concentrate exports were resumed only in 1928 (BP 1925).

When the Belgians entered the market in 1923, the price of radium dropped from about US $120 000, the official price for 1 g of American radium, to $90 000/g; it dropped further to about $70 000/g in the second half of the 1920's. The Belgian radium price was heavily criticized abroad for being exorbitantly high in comparison to the production costs. Although U.M.H.K. never released any information on production costs, they can be estimated to be about US $20 000/g. Numerous articles were published accusing U.M.H.K. of making huge profits at the expense of thousands of cancer victims (Anonymous 1929 a). The British were especially discontented with the price policy of U.M.H.K., and the Minister of Public Health even considered organizing the purchase of radium through the Health Department of the League of Nations, "that ought to bear upon the Belgian producers in order to bring them to reason" (Anonymous 1929b). When the same Minister was questioned on this issue one year later, he declared that propositions had been made within the League of Nations to start an inquiry on radium pricing and its supply by U.M.H.K. (Anonymous 1930). It is not known whether such an inquiry eventually did ever take place, but the fact is that U.M.H.K. reduced its prices in the beginning of the 1930's. Prices reached a level of $55 000/g in 1933 (USDC-BM 1930; USDC-BM 1934). The introduction on the market of the Canadian radium supplies and the general economic crisis of the period also help to explain this price reduction.

Beginning in the second half of the 1920's, fear rose that the development of radiation therapy with x-ray tubes, especially successful in the United States, might have a negative effect on the Belgian radium industry. As a matter of fact, in the United States, roentgenotherapy supplanted the use of large amounts of radium in so-called radium bombs (Quimby 1948). But this development did not seem to provoke a decrease in the sale of Belgian radium to the U.S. according to the regularly reported data of the US Bureau of Mines. Compared to the production of copper and tin, in serious trouble since the end of the 1920's, the industry of radium flourished until 1932, when the radium market finally became affected by the paralysing effects of the world economic crisis. In 1929 and 1930, U.M.H.K. reached a peak in uranium exports and radium sales.

Shinkolobwe was mined the whole year through, and in the radium (and cobalt) factories in Oolen, new installations were put into use (UMHK 1928). In the beginning of the 1930's, underground mining in Shinkolobwe was started (Anonymous 1956b), and electricity was introduced, replacing steam power. In the same period, discoveries of various precious metals (i.e., gold, palladium, and platinum) were made in the now underground mine. When exploitation of uranium was stopped in 1932 except for "a small amount", the mining of gold, platinum, and especially palladium continued. Uranium mining was to be suspended indefinitely to maintain price levels. The medical establishment had no need to fear a shortage of radium because of the considerable existing stocks in Brussels (AMC 1932). Nevertheless, U.M.H.K.'s radium industry did not seem to have been severely struck by the crisis as radium sales were stabilised in 1933.

Although Canadian radium production was launched the same year, Belgian radium sales did not seem to be affected, and the industry flourished during the second half of the 1930's. In Katanga, mining activities were started up again in 1933, and a considerable amount of uranium was mined (CSK 1933). One year later, a plant was established in Shinkolobwe for the mechanical and chemical treatment of uranium ore containing the precious metals mentioned above. After the ore was treated here, further extraction of precious metals was executed in Oolen (CSK 1934). If we take into consideration the uranium ore production figures that are available for the 1930's and the corresponding export figures (Fig. 4), it is clear that huge amounts

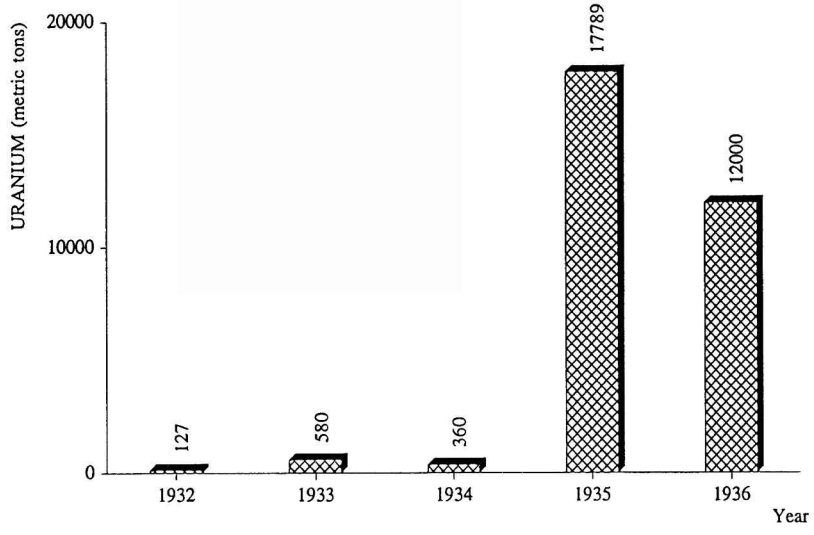


Fig. 4. Uranium mined by U.M.H.K. (1923-1937).

of uranium were stocked in Shinkolobwe, stocks accumulated through the 1920's. Unfortunately, no more information is available on radium sales, uranium production and export after 1937. From then on, attention would be focused primarily onto the strategic use of uranium ore. Although radium therapy remained quite popular during the 1940's and 1950's, radium for medical use would be regularly replaced by artificial radioisotopes.

CONCLUSIONS

Only fragmentary information is available on the industrial development of the radium industry in Belgium. Immediately after the second World War, it was widely realised that radiation in general and radium in particular is hazardous to humans. In the 1950's, radium was identified as a significant environmental pollutant, first in Colorado, then in Czechoslovakia, and finally at Oolen. It is understandable in these circumstances that the major actors in the development of the industry in Belgium, even during the uncontroversial period between both wars, were reluctant to provide information for an historical study.

The existing information in the literature, however, points to several interesting aspects of the development of the Belgian industry, which can be summarised as follows.

1) The industrial-financial complex in Belgium was quick to grasp the potential of an important mineral deposit and to foster the conditions for the development of an industry which dominated and, for some time, monopolised this scarce resource. The Société Générale de Belgique with its controlling interest in both U.M.H.K. and M.H.O. was undoubtedly vital in both the planning stage and the exploitation as a running concern of the industry.

2) The role of the Belgian Government in the development and consolidation of the Belgian radium monopoly cannot be underestimated. Even more than today, due to the smaller scale of action in the period considered, close links existed between the political and the economical-industrial establishments, many of these links being based on personal and friendship relations. Especially in the creation of a firm internal market for the new product and the democratization of the radium therapy, U.M.H.K. definitely had an advantage in its governmental relations. The Belgian Government also assisted considerably in commercializing the product internationally.

Acknowledgment — This work is based on the material available in a Master dissertation made at the Seminar of Contemporary History of the University of Ghent. Thanks are due to Prof. Dr. J. De Belder of this Seminar, and to Dr. G. Eggermont of SCK-CEN, Mol, Belgium.

REFERENCES

AANL (Archives of Argonne National Laboratories). In: Proc. conference on radium in industry, December 1928.

Adams, A. Het ontstaan en de ontwikkeling van de radiumindustrie in België. Master thesis, Rijksuniversiteit Gent; 1988.

AMC (Archives Ministère des Colonies). Fonds R.F. Dossier 4110. U.M.H.K. Mission des délimitation des mines 1914-1916.

AMC (Archives du Ministère des Colonies). Fonds U.M.H.K. Documentation 1918-1930. Rapport Economique du Haut Luapula. 2nd semester 1922.

AMC (Archives du Ministère des Colonies). Fonds RA/CB. Dossier (235) 1. Province d'Elisabethville, Service Provincial des affaires économiques. Rapport annuel 1932.

Anonymous. La Belgique Maritime, Coloniale et Economique (newspaper), 26 July 1923.

Anonymous. Le Radium Belge domine le marché du monde. Bruxelles-Méd. 13 September 1925.

Anonymous. La Belgique maritime, coloniale et économique (newspaper). 7 March 1926.

Anonymous. Quelques industries Belgo-Congolaises. Chim. Ind. (Société Chimique Belge), special issue 92; 1927.

Anonymous. The radium monopoly. Ind. Eng. Chem. 21: 615; 1929a

Anonymous. Tidings of the House of Commons, 24-7-1929; Lancet 3 October 1929b: 929.

Anonymous. Tidings of the House of Commons, 10-12-1930; Lancet 20 December 1930: 1375.

Anonymous. Union Minière du Haut Katanga 1906-1956. Bruxelles: Editions L. Cuypers; 1956.

Anonymous. U.M.H.K. 1906-1956. Evolution des techniques et des activités sociales. Bruxelles: Editions L. Cuypers 91; 1956b.

Anonymous. Belg. d'Outremer. 264 (February): 113; 1957.

BOCB (Bulletin Officiel du Congo Belge). Rapport du Conseil Colonial sur un projet de décret approuvant les ordonnances-lois des 8 mars et 9 avril 1923 du Gouverneur du Katanga, relatives à la législation minière de ce territoire. Bruxelles; 15 September 1929.

BOCB (Bulletin Officiel du Congo Belge). Ordonnance-loi du 8 mars 1923. Bruxelles; 15 September 1923.

BP (Belgian Parliament). Rapport annuel des activités dans la Colonie. Bruxelles; 1925, 1926 and 1927.

Buttgenbach, H. Note préliminaire sur des minerais d'uranium et de radium trouvés au Katanga. Ann. Soc. Géol. Belg. 24: 5-9; 1920-1921.

CCR (Committee of Civil Research). Radium Subcommittee Rep. Cmd. 3303. London; March 1929.

Clerin, J.; Leemans, J.; Sengier, E. Communications faites en présence de S.M. le Roi à la Manifestation Coloniale de Tervueren, le 13 novembre 1922.

CSK (Comité Spécial du Katanga). Rapport de l'Exercise 58. Bruxelles; 1923.

CSK (Comité Spécial du Katanga). Rapports et Billans, 1932-1933, 70. Bruxelles; 1933.

CSK (Comité Spécial du Katanga). Rapports Annuels 1934-1936; Bruxelles.

Laude, N.; Le radium congolais et canadien. Bull. Soc. Roy. Géogr. d'Anvers. 53: 80; 1933.

Quimby, E. Fifty years of radium. Am. J. Roentgenol. Rad. Ther. 723-730; 1948.

RF (Recueil Financier). Bruxelles; 1918-1923 (yearly reprints)

RF (Recueil Financier). Société Générale Métallurgique de Hoboken, Bruxelles; 1924.

RF (Recueil Financier). Cie. du Chemin de Fer du Bas-Congo au Katanga, Bruxelles; 1928.

Schoep, A. Le radium au Congo. Bull. Soc. Roy. Belg. Géogr. 2: 91-92; 1923.

Thoreau, J.; Du Trieu de Terdonck, R. Le gîte d'uranium de Shinkolobwe-Kasolo, Vol. 2. Institut Royal Colonial Belge, Bruxelles; 1933.

UMHK (Union Minière du Haut Katanga). Sales catalogue (Radium Belge), Photographic document N10. Bruxelles; 1925a.

UMHK (Union Minière du Haut Katanga). Sales catalogue (Radium Belge). Bruxelles; 45; 1925b.

UMHK (Union Minière du Haut Katanga). Rapport du Conseil d'Administration et du Collège des Commisaires, présenté à l'Assemblée Générale Ordinaire du 13 juillet 1925. Bruxelles; 1925c.

UMHK (Union Minière du Haut Katanga). Rapports du Conseil d'Administration et du Collège des Commissionaires. Bruxelles; 1928 and 1929.

UMHK (Union Minière du Haut Katanga). Sales catalogue (Department Radium), Photographic document fig. 15. Bruxelles; 1931.

UMHK (Union Minière du Haut Katanga). Rapport du Conseil d'Administration. Bruxelles; July 1932.

USDC-BM (U.S. Department of Commerce - Bureau of Mines). Mineral Resources of the United States. Rep. 471, Washington DC; 1924.

USDC-BM (U.S. Department of Commerce-Bureau of Mines). Mineral Resources of the United States. Rep. 616, Washington DC; 1925.

USDC-BM (U.S. Department of Commerce - Bureau of Mines). Mineral Resources of the U.S. Rep. 144. Washington, DC; 1930.

USDC-BM (U.S. Department of Commerce - Bureau of Mines). Mineral Yearbook U.S.A. Rep. 495, Washington DC; 1934.

Environment International, Vol. 19, pp. 503-508, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

A BRIEF HISTORY OF THE AMERICAN RADIUM INDUSTRY AND ITS TIES TO THE SCIENTIFIC COMMUNITY OF ITS EARLY TWENTIETH CENTURY

Edward R. Landa
U.S. Geological Survey, 430 National Center, Reston, VA 22092, U.S.A.

EI 9205-148 M (Received 7 May 1992; accepted 20 February 1993)

Federally funded remedial action projects are presently underway in New Jersey and Colorado at sites containing ^{226}Ra and other radionuclides from radium-uranium ore extraction plants that operated during the early twentieth century. They are but the latest chapter in the story of an American industry that emerged and perished in the span of three decades. Major extraction plants were established in or near Denver (CO), Pittsburgh (PA), and New York City (NY) to process radium from ore that came largely from the carnotite deposits of western Colorado and eastern Utah. The staffs of these plants included some of the finest chemists and physicists in the nation, and the highly-refined radium products found a variety of uses in medicine and industry. The discovery of high-grade pitchblende ores in the Belgian Congo and the subsequent opening of an extraction plant near Antwerp, Belgium, in 1992, however, created an economic climate that put an end to the American radium industry. The geologic, chemical, and engineering information gathered during this era formed the basis of the uranium industry of the later part of the century, while the tailings and residues came to be viewed as environmental problems during the same period.

INTRODUCTION

In February 1979, the people of Denver, CO, learned of radioactively contaminated sites in the city. These bore the wastes of an American industry with its roots in Colorado and Utah which had been born, blossomed, and passed into obscurity in a period of about 10 y more than half a century earlier. Ordinarily, such a brief existence might be taken as a sign of insignificance. This, however, was no ordinary commercial enterprise; it was an industry intimately tied to some of the leading scientists of the early twentieth century. These wastes, residues of the radium extraction industry, whose remediation will require the outlay of millions of dollars, are but one legacy of a remarkable substance and an exciting era in the scientific and technological life of America.

Most attention to radium today focuses on it as an environmental contaminant. Radium processing sites in Denver (from various companies); Orange, NJ (U.S. Radium Corporation); and Canonsburg, PA (Standard Chemical Company/Vitro Manufacturing Company), have recently, or are presently undergoing remedial action to minimize human exposure to radiation. Available stocks of radium are more than adequate to satisfy the market, and with its long half-life and diminishing demand, no radium extraction from ores for medical or industrial uses will probably ever occur again. Indeed, the only extraction operations even discussed today—as alternative uranium mill-

ing methods—would classify radium as a waste, and seek to concentrate and fix it in a form suitable for geologically long-term burial. Thus, in less than a century, it has moved from buried treasure to buried waste.

PRECURSORS TO THE EXTRACTION INDUSTRY

The phenomenon of radioactivity was discovered in 1896 by Henri Becquerel, who noted a light-struck effect on sealed photographic plates placed in proximity to samples of uranium salts. Early in 1898, Marie Curie found that certain uranium-bearing mineral specimens possessed a greater ability to ionize air than recently prepared uranium metal and compounds. The results pointed to the presence of at least one hitherto unknown radioactive substance in these minerals, and by the end of the year, the new element radium had been discovered. In order to isolate the large quantities of radium required to study its properties, Marie and her husband Pierre obtained about eight tons of tailings from a uranium extraction operation at Joachimsthal, Austria, which used rich pitchblende ores from the local mines. At that time, uranium was produced to be used largely as a pigment in coloring glass and ceramics. From these wastes, Marie Curie began extracting and purifying radium. By the middle of 1902, about 100 mg of the element had been isolated and its atomic weight determined.

The details of the extraction and refining process were published in Marie Curie's Doctor of Science thesis at the Sorbonne in 1903—no attempt was made on her part to patent the process. The scientific community was eager to obtain radium for study, and with the Curies' technical assistance, French industrialist Armet de Lisle established a commercial ore processing plant near Paris in 1904. During the next two decades, other radium plants were established in Germany, Austria, Great Britain, France, Sweden, Russia, and Australia using ores from these countries and from Madagascar, Portugal, Indochina, and the United States. For the years prior to 1913, the beginning year for large-scale production in the United States, an estimated 50% of radium produced in Europe was extracted from carnotite ores shipped from Colorado and Utah.

The ability of radium to spontaneously release heat, the deflection of its various radiations in a magnetic field, and the presence of radium and its decay products in water and rocks attracted considerable scientific attention. However, radium was more than a scientific curiosity; it could be used in cancer therapy and in the production of luminous paints. For both the lay and scientific communities, it was truly the miracle element of the new century. As the weight ratio of radium to uranium in typical ores was about one part radium to three million parts uranium, the extraction of radium from Colorado-Utah carnotite ores containing only about 1-2% uranium was an equally remarkable technological achievement.

Around the turn of the century, the American deposits of most interest to foreign buyers as a source of uranium and radium were pitchblende-bearing veins mined near Central City, CO. However, the highly localized nature of these deposits made profit risky, and attention soon shifted to the extensive carnotite deposits of western Colorado and eastern Utah. Large-scale, domestic production of radium from these ores began around 1913. From 1913 to 1922, most of the world's radium supply came from Colorado uranium ores.

The processing of these ores to extract trace quantities of radium was a complex task carried out at plants in or near Denver (CO), Boulder (CO), Pittsburgh (PA), Philadelphia (PA), Chicago (IL), and New York (NY). The discovery of higher-grade ores in the Belgian Congo and their processing for radium began around 1922, and was the death blow to the radium extraction industry of the United States (Adams 1993). However, the mining and milling practices of that era served as the forerunners of the uranium industry that provided materials for the first atomic bomb and, later, for peaceful uses of nuclear power. The use of radium in medicine during the first half of this century provided the basis for modern radiation therapy, and early, often tragic, experiences with occupational exposure to isolated radium salts served as a foundation for the modern science of radiation protection.

The greatest demand for radium came from physicians and surgeons. Armet de Lisle had fostered this interest by underwriting the establishment of a medical clinic for the study of radium in 1906, and later by publishing the first text on radium therapy. With growing demand came rapidly increasing prices. In 1913, Nobel-laureate Ernest Rutherford would complain that the high cost of radium, then about US$120 000/g, was artificially inflated and bore no relation to the cost of production. Even at about $40 000/g (the cost of production estimated in 1915 by a U.S. government study using Colorado carnotite ores) radium was still the most expensive substance known to man. As a point of comparison, the famed Hope diamond sold in 1909 for $80 060 or $8 779/g. When hard times hit in 1921, "thimbleful" quantities

of unsold radium salts represented million-dollar inventories.

That period holds a diverse story of metallurgy and medicine, of Nobel Prize scientists and luminous-dial painters, of radiochemistry and national policy debates. Here, an overview of the American radium industry is presented emphasizing the ties of the industry to the American and European scientific communities. A more comprehensive, fully-referenced treatment of the American radium industry, including chapters on the medical and commercial uses of radium, and the early status of uranium as an extraction by-product of minimal value, is presented an earlier monograph (Landa 1987). A discussion of the metallurgical and chemical processes used in extraction and refining of radium is given in Landa (1981).

THE PRE-1913 PERIOD

In 1898, near Richardson, UT, a miner named Welsh discovered a fracture zone containing yellow, orange, green, and blue minerals. Assay indicated that it contained significant quantities of uranium. Welsh and another miner, James H. Lofftus, seemingly disappointed with the negligible precious metal content, did not stake any claims. A young Buffalo, NY, attorney, Stephen T. Lockwood, who was traveling through the West in 1900, had an interest in the natural sciences and evidently was aware of the commercial market for uranium. He met the miners, apparently by chance, and convinced them of the economic potential of the deposit. The Welsh-Lofftus Uranium and Rare Metals Company was established with Lockwood as president and general manager. Claims were staked and development work began at the Richardson site, while Lockwood made plans for an extraction plant near Buffalo.

By early in 1902, Lockwood was experimenting on the extraction of radioactive components from the ore. He began corresponding with Pierre Curie, who examined the activity of a sample sent by Lockwood and advised as to extraction processes which might remove radium from the ore. In September 1902, Lockwood published a radiograph of a key and a Chinese coin, obtained by using an impure sodium uranate salt extracted from the Richardson ore. The report by Lockwood in the Engineering and Mining Journal made no mention of radium; Lockwood attributed the penetrating radiation from the ore and the uranium salt to the presence of actinium, for reasons that are not clear.

Within weeks of his September 1902 report, Lockwood apparently realized a need for technical expertise and sought out help at his alma mater, Princeton University. Professor Alexander Hamilton Phillips, a mineralogist, was given 25 lbs. (11.4 kg) of Richardson ore and began chemical extraction and fractional crystallization experiments which, in conjunction with radioactivity measurements made by physicist George B. Pegram at Columbia University, soon demonstrated the presence of radium in the ore. Experimental work by Phillips continued through the spring of 1904, on both the radium isolated from the Richardson ore, and another carnotite ore sample from Montrose County, CO, used to study methods of separating uranium, vanadium, and radium in anticipation of commercial production. The end products of these experiments also served as part of an exhibit on radioactive ores, sponsored by the U.S. Geological Survey at the 1904 Louisiana Purchase Exposition in St. Louis, LA, which earned Lockwood a silver medal.

An experimental plant for the processing of carnotite ore was built at Lackawanna, NY, in October 1903, and was expanded in the next few years. The plant, operated by Lockwood as the Rare Metals Reduction Company, produced uranium oxide and iron vanadate salts. The market for uranium was limited; the vanadium salt was converted to ferro-vanadium alloy in the electric furnaces of the Niagara Research Laboratories, a newly formed research and development consulting company which was soon to become the research unit of Union Carbide Company. Several lots were sold, beginning in 1906, to a Japanese firm. By 1906, plant capacity was 30 tons of ore per month. However, an insufficient supply of acceptable-grade ore at that time restricted commercial-scale production, and by March 1908, operations at Lackawanna had apparently ceased.

While primary attention focused on vanadium, Lockwood maintained an interest in the ores as a source of radium. A radium-barium sulfate concentrate was recovered as a by-product, and radioactivity measurements were made on select samples by Pegram at Columbia and by chemist Bertram Boltwood at Yale. The concentrate was not, however, refined to a marketable, high-purity product, as the cost of fractional crystallization did not allow for any profit. Portions of these concentrates were donated by Lockwood to Columbia University, and to the U.S. National Museum of the Smithsonian Institution.

LARGE SCALE PRODUCTION

The extraction of radium involved complex chemical processing. The European chemical community had about a decade of laboratory- and commercial-scale experience in this area when interest in a domes-

tic industry in the United States arose again around 1911, and this expertise was sought to get the fledgling enterprises started. Standard Chemical Company, which went on to become the largest American radium producer, brought W. A. Schlesinger from Heidelberg, and Otto Brill from the Austrian radium works at Joachimsthal to develop an extraction process at its plant near Pittsburgh, PA. The Radium Company of America brought Austrian chemist Siegfried Kohn to its Bucks County, PA, headquarters around 1912 to head its technical department.

Despite this initial reliance on imported expertise, academic and on-the-job training soon enabled American scientists and engineers to assume technical leadership roles in the industry. In 1913, when Otto Brill returned to Austria after one year with Standard Chemical, he was succeeded by Charles H. Viol who joined Standard Chemical in 1912, immediately after receiving his Ph.D. in radiochemistry at the University of Chicago; he served as the director of the company's research department from 1913 until his death in 1928. Dr. Herbert N. McCoy, Viol's mentor at Chicago, who had been unsuccessfully approached earlier by Standard Chemical, became associated in 1915 with the Carnotite Reduction Company which operated until 1920 with mining claims in Colorado and an extraction plant in Chicago.

The Denver-based, National Radium Institute was a cooperative venture of a federal agency (the U.S. Bureau of Mines), a Baltimore physician who wanted radium for his practice, and a mining engineer with philanthropic ties to New York's Memorial Hospital. The Bureau of Mines laboratory in Denver gave practical experience to young chemists and physicists during the period 1913-1916. These individuals often went on to leadership roles in the private sector: Charles F. Whittemore became the chief physicist of the research laboratories of the Radium Company of Colorado. Howard H. Barker served as the chief chemist of the Chemical Products Company, a Denver-based radium company which operated from 1916 to 1920, and the U.S. Radium Corporation. Barker was hired by the U.S. Radium Corporation (prior to 1921 called the Radium Luminous Material Corporation) to replace Victor Franz Hess, formerly of the Institute for Radium Research of the University of Vienna, who served as director of the company's research department at its Orange, New Jersey, plant from 1921 to 1923 (Seeger 1993). Hess returned to the University of Graz in his native Austria and continued research begun in 1911 on cosmic rays and in 1936, shared the Nobel Prize in Physics for his cosmic ray work. Hess came back to the United States in 1938 to join the physics faculty at Fordham University in New York City, where he remained until his death in 1964. While Hess survived to 81, he did suffer severe radiation damage to one of his thumbs, which was amputated in 1934. When this injury occurred is not clear, but during his years at Fordham, he maintained an interest in the medical problems unique to radium workers.

Hess, in turn, had come to the U.S. Radium Corporation in 1921 to replace Sabin A. (von) Sochocky, an Austrian who had studied medicine at the University of Moscow and later studied physics at the University of Manchester under Ernest Rutherford. Sochocky was a part owner of the company and, in his role as the chief physicist, supervised the extraction and refining of about 30 g of radium between 1913 and 1921. On at least four occasions, tubes containing high-purity radium or mesothorium salts exploded in his presence. Such explosions, due to pressure buildups associated with the radiolysis of water contained in the salts, were often reported by radium chemists and therapists. In addition to these "typical" modes of radiation exposure in the radium refinery, Sochocky's exposure was increased by practices described in a U.S. Department of Labor (1929) report on "Radium Poisoning."

"He [Sochocky] was fascinated by the qualities of radium, and is said to have played with it, taking the tubes of radium salts out of the safe and holding them in his bare hands while watching the scintillations in the dark. He is also said to have immersed his arm up to the elbow in solutions of radium or mesothorium."

In an article for a popular magazine, Sochocky (1921) wrote:

"For my own use, I have made radium oil paints, and because of my interest in art, I have made some paintings with them. Pictures painted with radium look like any other pictures in the daytime, but at night they illuminate themselves and create and interesting and weirdly artistic effect. This paint would be particularly adaptable for pictures of moonlight or winter scenes, and I have no doubt that some day a fine artist will make a name for himself and greatly interest us by painting pictures which will be unique, and particularly beautiful at night in a dark or semi-darkened room."

He also foresaw a world where the conductor's baton and the keys of a concert hall piano would be highlighted with radium luminous paint and indeed where such paint would be applied to the walls and ceilings of rooms in homes to provide a nonelectric source of illumination.

In 1920, he received severe burns to both of his thumbs, resulting in the destruction of part of the bone in one of them, from handling an apparatus containing one or two grams of radium. By 1925, he was suffering from extensive radium dermatitis of the fingers of both hands, anemia, and the "jaw rot", typically seen in the radium dial painters. (A New York Times reporter who interviewed Sochocky in June 1928 noted his fingers to be blackened up to the second knuckle). He spent part of his remaining years helping Dr. Harrison S. Martland, the Chief Medical Examiner of Essex County, NJ, in the measurement of radioactivity in persons presently or formerly employed in the watch- and clock-dial painting industry. Sochocky died at 46 of aplastic anemia in November 1928.

The previously mentioned National Radium Institute (NRI) produced about 8.5 g of radium in Denver between 1914 and 1916. From this supply, 4g went to Dr. Howard A. Kelly of the Johns Hopkins School of Medicine in Baltimore. With this large quantity of radium, Kelly decided to use radon seeds rather than radium tubes and applicators in therapy—the seeds being more versatile, and the design of a radon plant minimizing the chance of loss of radium by fire, theft, or accidental disposal, etc. As early as 1911, Kelly had experimented with this approach, placing 30 mg of radium bromide procured from Vienna in a small radon collection apparatus constructed for him under the direction of Bertram Boltwood of Yale. With a letter of introduction from Boltwood, Kelly's associate Dr. Curtis Burnam visited Nobel-laureate Ernest Rutherford, then Professor of Physics at Manchester University, in 1912. As a result of this visit, the staff of Ernest Rutherford's laboratory fabricated for the Kelly Hospital several specialized pumps for use in a radon plant. Early in 1915, one of Rutherford's technicians, Walter Lantsbury, came to Baltimore to design and establish a full-scale radon plant at the Kelly Hospital. All of the NRI radium and most of that purchased earlier was used in this apparatus, which was operated by physicist Fred West, a former student of Rutherford. The assistance which Sir Ernest Rutherford rendered to the Kelly Hospital was reciprocated about 15 y later. At the request of Professor James Chadwick, of Cambridge University's Cavendish Laboratory, headed by Rutherford, Drs. Burnam and West provided a quantity of old radon seeds. From these, Chadwick extracted the radon daughter product ^{210}Po. Chadwick was interested in research findings published by other physicists between 1930 and 1932 in which bombardment of such low atomic weight elements as beryllium and boron with alpha particles from polonium had resulted in emission of radiation having far greater penetrating power than that of any known gamma radiation. Preliminary experiments by Chadwick to elucidate the character of this radiation were hampered by the weakness of available polonium sources. Using the Kelly Hospital seeds in which the short-lived radon had long since decayed away, Chadwick prepared a polonium source that was successfully used in his 1932 experiments which revealed the existence of the neutron. Thus the NRI radium from which the polonium was derived played a significant role in this landmark discovery, which earned Chadwick the 1935 Nobel Prize for physics.

The U.S. Bureau of Mines chemists who developed the nitric acid extraction process used at the NRI went on to do pioneering work in radiochemistry and related fields. Richard Moore, who headed the research group, became involved in helium production for World War I airships, and developed the first commercial scale operation for the extraction of helium from natural gas. (Prior to this, the world supply of helium was very small and derived largely from the dissolution of radioactive minerals, the helium therein originating as alpha particles.) Samuel Lind, Moore's chief scientist, used the 0.5 g of radium which the Bureau retained from the NRI operation to pursue basic research on the effects of alpha particles on the chemical reactions of gases. These investigations led to his suggestion in 1931 that petroleum may originate from alpha-particle bombardment of hydrocarbons in sediments. This hypothesis, which received considerable experimental investigation by geochemists at the Massachusetts Institute of Technology in the 1940's and 1950's, is still being analyzed and discussed. Henry A. Doerner, a junior chemist during the NRI period, succeeded Lind in the mid-1920's as the Bureau's chief radium chemist. Following up on the 1921 work of Frank E. E. Germann of the University of Colorado, Doerner and colleague William M. Hoskins did theoretical and experimental work on coprecipatation and replacement reactions occurring at radium-barium sulfate crystal surfaces. The removal of radium from solution by barium sulfate crystals which Germann had attributed to adsorption, was shown to be a heterogeneous solid-solution phenomenon. The Doerner-Hoskins model which emerged from this study has been shown by others to be applicable to a variety of relatively insoluble solids of geological and industrial interest. Sixty years after its publication, it is still widely used.

Germann's work focused on the adsorption of radium from hydrochloric acid solutions by barium

sulfate, a question of considerable interest in the radium production industry. Sulfate-free hydrochloric acid was prepared by the radium producers to dissolve radium-bearing carbonate residues by precipitating any soluble sulfate in commercially available hydrochloric acid with barium; any subsequent sorption of radium by the barium sulfate crystals present in the acid would be detrimental to the extraction yield. Evidence suggests the research was done with the sponsorship, or at least the cooperation, of the Tungsten Products Company, a radium-extraction company based in Boulder, CO. The company was headed by chemist Warren F. Bleecker who began his career at Standard Chemical. The products of economic value typically isolated from carnotite ore were radium, vanadium and uranium. However in 1919, presumably in hopes of some economic gain, Bleecker isolated 60 g of a thorium oxide mixture containing about 4% ionium (^{230}Th) oxide from 60 gal (227 L) of radium refining waste solutions which came from the processing of several hundreds tons of carnotite ore.

Around 1922, Bleecker approached Bertram Boltwood regarding his possible interest in this ionium-bearing material. Boltwood had established the identity of ionium as the parent of ^{226}Ra in 1907, but by the early 1920's, his research took a back seat to administrative duties associated with the design and construction of the Sterling Chemistry Laboratory at Yale. Boltwood indicated a lack of time to devote to the study of the material, and early in 1923 Bleecker wrote to 1921 Nobel laureate radiochemist Frederick Soddy at Oxford regarding his possible interest in the examination and purchase of the sample; Bleecker estimated its value at US$2 500. Soddy indicated an inability to purchase it himself, but did some analyses of the sample and at Bleecker's invitation kept 5 g for future research. The remainder of the sample was returned to Bleecker during the summer of 1924. The subsequent fate of this unique material in Oxford and Boulder is not presently known.

The Congo pitchblende deposit discovery put an abrupt end to the U.S. radium extraction industry in the early 1920's. Vanadium became the only constituent of economic interest in the carnotite ores. In the late 1920's, the mining properties and concentrating mills of the Standard Chemical Company passed to the hands of the U.S. Vanadium Corporation. Despite vigorous research and development and marketing efforts during the radium era, focused mainly on metallurgical applications, uranium remained a product in search of a use. Things would change dramatically in 1938. Late that year, Otto Hahn and Fritz Strassmann, working at the Kaiser Wilhelm Institute in Berlin, discovered that uranium would fission when bombarded with low-energy neutrons. The quantity of energy released by such a reaction was soon calculated by Lise Meitner and her nephew Otto Frisch. Their results were published in Nature in February 1939. By the spring of 1939, Frederic Joliot (Marie Curie's son-in-law) and his colleagues in Paris had demonstrated the chain reaction phenomenon.

The military possibilities of such an energy release were realized almost immediately in war-poised Europe. These scientific breakthroughs did not go unnoticed by the American minerals industry. In 1939, construction began at the U.S. Vanadium Corporation's vanadium extraction mill at Uravan, CO (site of the former Standard Chemical Company's concentration mill) on a unit to permit the recovery of uranium, which was up to this time being discharged with the carnotite tailings. During that year, there was also a dramatic increase in the importation of uranium oxide and salts into the United States. Imports fluctuated around 200 000 to 400 000 pounds per year (90 000-180 000 kg /y)during the period 1928-1938. This figure increased to about 1 400 000 pounds (640 000 kg) in 1939, undoubtedly caused by interest in the fission process and its military applications. The linkage of the industry to the fore-front of science, forged during the radium industry, would persist in the modern nuclear industry.

REFERENCES

Adam, A. The origin and early development of the Belgian radium industry. Environ. Int. 19(5): 491-501; 1993.

Landa, E.R. A historical review of the radium extraction industry in the United States (1906-1926)—its processes and waste products. In: Proc. symposium on uranium mill tailings management, Colorado State University/Ft. Collins. 1981: 3-32. Available from: Civil Engineering Department, Colorado State University, Ft. Collins, CO.

Landa, E.R. Buried treasure to buried waste—the rise and fall of the American radium industry. Colorado School of Mines Quarterly 82(2): 77; 1987.

Seeger, K. Editorial: Victor F. Hess. Environ. Int. 19: 421-423; 1993.

Sochocky, S.A. Can't you find the keyhole? Amer. Mag. 91(1): 24-27, 106, 108; 1921.

U.S. Department of Labor (Bureau of Labor Statistics). Radium poisoning. Dept. of Labor, Washington, DC 75 pp.; 1929.

Environment International, Vol. 19, pp. 509-512, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

WASTES FROM THE FORMER URANIUM PAINT FACTORY AT JOACHIMSTAL (JÁCHYMOV) USED IN DWELLINGS

J. Thomas and D. Drábová
National Institute of Public Health, Praha, Czechoslovakia

EI 9203-137 M (Received 4 March 1992; accepted 20 May 1993)

Wastes resulting from the production of uranium paints in the K.u.K. Uranfarbenfabrik at Joachimstal became famous as the raw material for isolation of radium in great amounts by M. and P. Curie in 1898. In the past century, these wastes with radium concentrations up to 300 Bq/kg were also used to a great extent in mortars and plasters instead of sand in dwellings at Joachimstal. Radiological impacts of this unintentional abuse are given. The relationships between radium concentration of the building material, dose rate of gamma radiation, superficial radon exhalation rate, rate of radon entry, and radon concentration in air are shown.

THE SOURCE TERM

Joachimstal is the cradle of atomic age; this epithet can be read in advertising brochures of the late mining town Jáchymov, Joachimstal, and in treaties about the famous history of the town. The town had a silver period in the 16th century, a cobalt period, two uranium periods, a short radium period, and now has a long and successful radon spa period. This epithet expresses in a stilted way our national pride, that Czechoslovakia (at that time Bohemia in the Austrian monarchy) contributed to the great discovery of radioactivity. Joachimstal contributed with waste material from a factory which began producing uranium paints in 1853 for use in the glass and ceramic industry. M. and P. Curie assumed that after the extraction of uranium from the pitchblende, radium must be concentrated in the tailings. They ordered 100 kg of tailings in 1898, then 1000 kg in 1899, and 5000 kg in 1900; and isolated 0.1 g of radium (Těšínská 1989). With such dangerously high levels of radium, the basic experiments could be done easily and quickly. Concentration of about 800 kBq/kg of ^{226}Ra in the waste can be calculated. In a later paper (Markl 1927), concentrations of 10 MBq/kg are given, comparable to pure uranium ores (e.g., U_3O_8).

This waste material, as a crushed rock, was a high quality sand and was therefore commonly used in plasters and mortars during the whole second half of the 19th century. As fire and conflagrations were common hazards at this time, sand for plasters and mortars was a highly desirable building material. As no source of good sand existed in the vicinity, the tailings were used. Before the discovery of radium, the waste was worthless. Later, it became a valuable

and requested material, not only for the separation of radium, but also for self-therapy. It could be obtained only by the nobility or other persons of power and influence (Seidlerová 1972).

THE RADIOLOGICAL IMPACT

During the postwar uranium mining period, radiological prospecting occasionally revealed high activity in the walls of some houses. It was interpreted that some houses in Joachimstal were built of pitchblende ore. The story goes that several houses were demolished and the material transported to the uranium mill for processing. Recent radiological surveys of homes in Joachimstal show that sometimes only a stone with some ore can be found in the walls. Homogeneously distributed radium in the plasters or within the wall is more frequent.

The identification and localisation of these active building materials by a gamma dose rate meter are simply and reliably done; the dose rates are significantly higher than the background. But the external irradiation of the inhabitants is not the only risk. Exhalation of radon from the walls also contributes to the radon concentration in the air of the rooms. As the exhalation rate is constant in time compared with the very variable entry rate of radon from the soil, it is obvious to look for a relation between the relevant quantities in model situation.

a_m (Bq/kg) = concentration of radium (^{226}Ra) in the material;

$\dot{D}$ (μGy/h) = gamma radiation dose rate measured on the surface of the wall or ceiling;

a_s (Bq/m^{2}s) = superficial exhalation rate of radon from the surface of the wall;

a_v (Bq/m^3) = concentration of radon in the air of the contaminated room.

Typical parameters are used for the air exchange rate k, the volume V, and exhaling surface S of the room. The relations between these quantities can be easily derived (Thomas and Moučka 1990); but more important is the experimental verification. Of utmost interest is a graphical demonstration enabling one to estimate the more complicated radon concentration from the simple measurable dose rate. Also one can estimate the significance of the contribution to the average concentration of radon measured by an integral detector. In other words, one can decide if the contamination of the walls completely explains the radon comcentration or if another source of radon and its entry rate have to be searched for. Such a graph is given in Fig. 1.

These model parameters are used:

$$k = 0.3\ h^{-1};\ V = 60\ m^3;\ S = 60\ m^3;\ \text{and}\ F = 0.5.$$

Completely contaminated plaster on walls and ceiling or mortar in the walls is assumed. The relationships are linear. For reduced volume of contamination, the equivalent equilibrium concentration of radon has to be reduced proportionally. Great accuracy is not required for these measurements and estimations, as the removal of contaminated plasters is always ordered as a part of the mitigation actions. For comparison, in a new building material, a maximum radium concentration of 120 Bq/kg is permitted by a Decree (MH 1991).

The similar radiological impact of the usage of waste in plasters or mortar is evident. For contaminated mortar, the dose rate is nearly two times lower compared to plaster with the same concentration of radium. Therefore contaminated mortar cannot be detected if contaminated plaster is present resulting in a high background. Only after complete removal of the contaminated plaster can the presence or absence of contaminated mortar be confirmed.

PROPERTIES OF THE SOURCE TERM

First, it must be ascertained waste from the uranium paint factory is indeed that used in the houses. The factory was demolished in 1942. The rest of the waste was transported to the uranium mill during the postwar uranium period. The area of the former factory was completely excavated and landscaped; it is now a park of the spa area. It is peculiar that the only remaining contaminated ground is near the Curie memorial. Excavated material from this place, a mixture of tailings and normal soil, showed a radium concentration of 300 kBq/kg. Characteristic of this material is the nearly fivefold deficit in uranium (^{238}U), 60 kBq/kg. The observed Ra/U ratio is a result of the efficiency of extraction of uranium and of the sorption of radium on the crushed rock. (No original sample with concentration of 10 MBq/kg is reserved; perhaps some could be found in old stocks of the Technical High School in Prague which was engaged in the study of their properties (Markl 1927). Some results of concentrations in plasters and mortars are given in Table 1. It can be seen that the waste is differently diluted and that the Ra/U ratio is in some cases, surprisingly close to one, indicating natural radioactive equilibrium, i.e., not affected by a chemi-

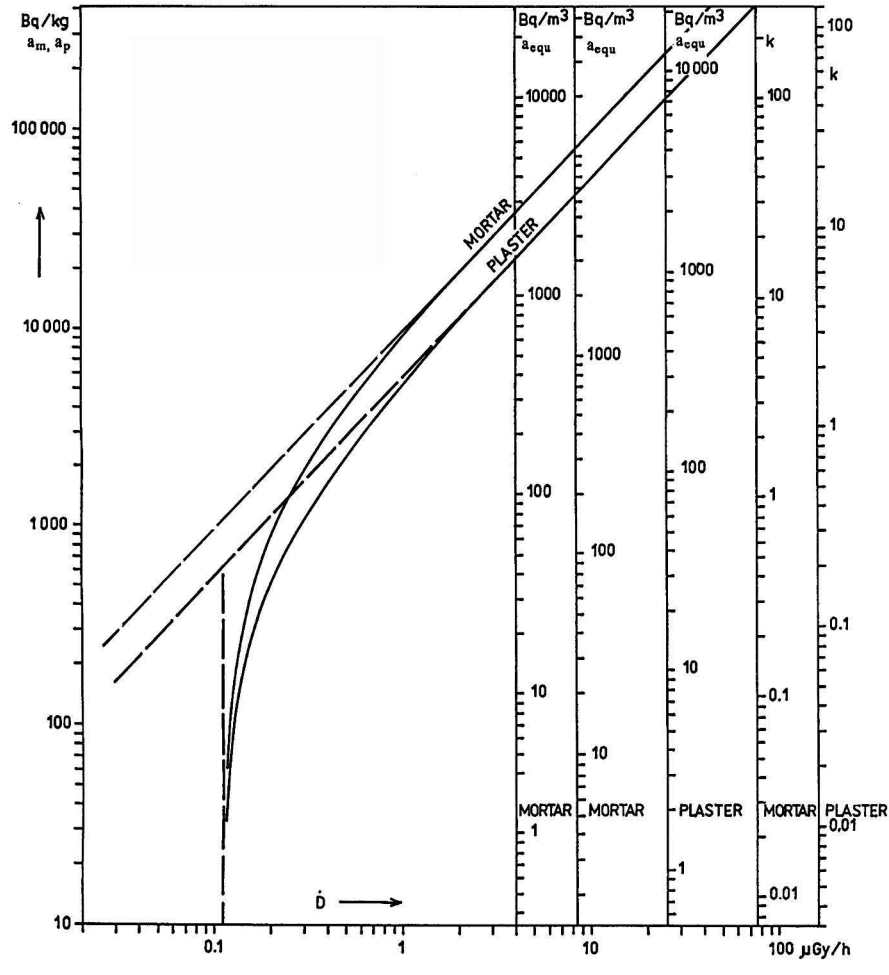


Fig. 1. Graph for the determination of the radon source. Note that a_m, a_p is the concentration of ^{226}Ra in mortar (m) or plaster (p) (in Bq/k); D is the gamma ray dose rate in contact with the contaminated wall (in µGy/h); a_{act} is the multiple of the relative remedial action level (summing the fractions of external and internal exposure relative to action levels 2µGy/h and 200 Bq/m³).

cal processing. It is unlikely that large quantities of crushed ore prepared for extraction would be found as waste. Perhaps this material represents wastes from the silver metallurgy period, because previously the uranium paint factory had been a silver, as well as cobalt smelting workshop.

The occurence of frequent fires and conflagrations in the 19th century also coincided with the uranium paint production period. The maximal production rate of the factory was only several tons of waste per year. Considering this with the fact that several hundreds kg of sand are needed for plastering one room, indicates that normal sand had been added to the tailings and that contaminated plasters and mortars are not found with great frequency in Joachimstal, a blessing in disguise.

Attempts at petrological analysis of the waste material were unsuccessful, and no uranium minerals were found. This is consistent with the assumption of uranium extraction, i.e., the main minerals were dissolved. Due to its very low concentration, radium typically does not form discrete minerals in rocks. It can be assumed that during the dissolution of the crushed ore (with rest of the rock), radium is also

Table 1. Concentration of ^{226}Ra and ^{238}U and the Ra/U ratio in some samples of plaster and mortar in Joachimstal.

	Concentration		Ra/U ratio
Material	Ra 226	U 238	
plaster	90 ± 1 kBq/kg	26 ± 10 kBq/kg	3.5 ± 1.3
	24	7	3,4
	16	4	4.0
	28	10	2.8
	125	25	5.0
	94 ± 1	115 ± 20	0.8 ± 0.2
	99 ± 1	98 ± 5	1.0 ± 0.1
mortar	80	-	-
	200	-	-

dissolved and then adsorbed on the surfaces of the undissolved rock grains. This is assumed to be the form of radium (as well as the rest of the uranium) in the waste. This is supported by results of chemical desorption analysis of plaster samples. During the separation process in the paint factory, a roasting process is also included; something as slag or charcoal could be separated from crushed plaster with significantly adsorbed activity.

CONCLUSIONS

The use of wastes from the uranium paint factory in dwellings as plasters and mortars is a special type of indoor radon source in Joachimstal. The gamma exposure and radon daughter concentration in air associated with various levels of radium contamination in mortars and plasters have been determined for homes in Joachimstal. When such contamination is found in a house, this leads unequivocally to the decision for its removal. Plasters can be removed easily, but contaminated mortar generally requires the demolition of the house. Nevertheless, this source facilitates "radiological archeology" in the houses which were built and rebuilt during four centuries; documentation about building activities are scanty. Some people lived for dozens of years in dose rates of hundreds of mGy/year, very near to the non-stochastic limit for workers. While no conspicious health effects have been reported, no extensive epidemiological studies exist to demonstrate or refute this apparent lack of health effects.

REFERENCES

Těšínská, E. The pitchblende of Jáchymov and the beginning of the research into radioactivity. Acta Historiae Rerum Naturalium Necon Technicarum 20 (special issue): 199-230; 1989.

Markl, J. Über Pechblende und Pechblendenrückstände von St. Joachimstal und deren Emanationsabgabe. Phys. Z. 28: 10-12; 1927.

Seidlerová, I. Joachimstal uranium ore in the beginnings of radiology. Ces. Cas. Fyz. A22: 198-201; 1972. (In Czech)

Thomas, J.; Moučka, L. Plasters and mortars with extremely high concentrations of radium in Joachimstal. Int. conf. high levels of natural radioactivity. Ramsar, Iran; 1990.

MH (Ministry of Health). Decree No.76 on requirements to reduce the exposure to radon and other natural radionuclides. Collect. Acts (Sbirka Zakonu) 1991.

Environment International, Vol. 19, pp. 513-518, 1993
Printed in the U.S.A. All rights reserved.

0160-4120/93 $6.00 +.00

EARLY USES OF RADIONUCLIDES IN MEXICO

N. Segovia, A. Tejera, S. Bulbulian, and F. Palma
Instituto Nacional de Investigaciones Nucleares, Ap. Post. 18-1027, 11801 México D.F., México

EI 9203-135 M (Received 18 March 1992; accepted 15 February 1993)

Mexico is traditionally a mining country and the first information about the presence of uranium is related to mine exploitation. Around 1945, when uranium became economically important, a rumor spread that large amounts of Oaxaca's black ceramics were being purchased and sent abroad because of their assumed high uranium content. It was not until 1949 that minerals containing thorium and uranium were declared by law as "national reserves". In the 40's, a radium emanation plant was installed at the "Hospital General" in Mexico City with the main purpose of carrying out radon seed implantation in tumors. In the 50's, a radium dial-painting facility was operating in the city of Toluca. In 1955, the National Commission of Nuclear Energy (CNEN) was founded by a government decree with two main activities: a training program on "Radioisotope Techniques and Nuclear Instrumentation" and the creation of specialized laboratories. In this paper, a general description of these events and undertakings spanning the decades from 1940 to 1970 is given.

INTRODUCTION

Mexico officially entered the radionuclide era when the National Commission of Nuclear Energy (CNEN) was founded by a government decree in 1955, several decades after the European countries. This institution regulated and controlled all available radioactive sources in the country, radionuclide importation, and exposure rates of occupationally exposed personnel as well as of the general population.

In this paper, a general description of radionuclide applications in medicine and industry and of the institutional undertakings spanning the decades from 1940 to 1970 is given. Some of the information was obtained from documents describing the early uses of uranium, radium, and related nuclides in Mexico. However, the main information was obtained directly through personal communication from professional staff that worked, from 1940 to 1970, with the available radioactive sources in the country.

URANIUM MINING

The first information on the presence of uranium in Mexico is related to mine exploitation. Mexico has been a mining country throughout its history. Several centuries before the Spanish conquest, in pre-Colombian times, gold, silver, and copper were used for ornamental purposes. Four centuries ago, the conquistadores were also interested in the exploitation of gold and silver mines, and they started mining operations in Zacatecas in 1546.

The University of Mexico, founded in the 16th century by the Spaniards, dealt basically with humanistic branches such as law, philosophy, and literature. The sciences were represented by medicine and, to a smaller degree, chemistry and engineering. However, being a colony, some subjects of study were influenced by the Spanish economy. Since mineral exploitation was important, this branch of study was promoted. In 1792, the first mining school in the American continent was founded in Mexico City (Escuela de Mineria) by Fausto Elhuyar, discoverer of tungsten, and Andres Manuel del Rio, discoverer of vanadium (Espíndola 1986).

In 1810, Mexico obtained its independence from Spain. In the early years of the 20th century, while Marie Curie was conducting research on radium and polonium, and radium enterprises were operating in Europe and USA, Mexico was dealing with a revolution. The political situation of the country was quite unstable until 1930; thereafter, the political situation stabilized and significant changes were made in the field of education in the country. Libraries and schools were founded, and branches such as engineering were substantially reinforced. At that time, the exploitation of oil was in the hands of foreign companies. The oil industry was expropriated by the government in 1936 and, since then, has been the main industry in Mexico.

At the end of the 30's, the situation in Europe was critical. The civil war in Spain was at its end, and Mexico opened its doors to all those Europeans wishing to leave Europe. The Spanish immigrants arrived in Mexico between 1939 and 1940 together with some immigrants from central Europe. For the professionals, the doors of the University were also opened, and physicists, chemists, mathematicians, and physicians were integrated into the scientific groups already formed at the universities.

Documents dating from the end of the 19th century dealing with uranium in the country were found. In 1868, a gold mine was discovered in the locality called *Placer de Guadalupe*, 45 km from the city of Chihuahua. The gold was mixed with a black and heavy mineral which was later found to be uraninite. In 1944, Gonzalez Reyna reported that the uranium in this mine was not commercially important. He also reported having found radioactive minerals in the state of Guerrero; however, this statement was not corroborated at that time. During that period, an American laboratory reported the finding of a highly radioactive material in a mercury sample extracted from the mine El Sotolar in Chihuahua.

Around 1945, when uranium became economically important, a rumor spread that large amounts of Oaxaca's black ceramics were being purchased and sent abroad because of their assumed high uranium content. The clay from Oaxaca is formed by volcanic rock decomposition within a strong mineralization system, and probably has only a slightly higher uranium content than other clays; therefore, uranium content in the ceramic manufactured with this mud could not be very high. In fact, radioactivity measurements in these ceramics have shown mean values of 5 ppm (μg/g) of uranium, only 2 ppm (μg/g) higher than the most common uranium content in clays (Iturbe 1991).

MEDICINE AND INDUSTRY

In the 40's, one of the main hospitals in Mexico City, Hospital General, received 1.5 g of ^{226}Ra as a donation from Memorial Hospital in New York, to be employed in a radium emanation plant. The goal of the plant, which worked until the 50's, was the preparation of radon seeds, capillary tubes containing radon, to be used for implantation in tumors. In the early 60's, the plant was closed and the radium was encapsulated in needles and pills which were, in turn, used for tumor irradiation (Diaz Perches 1991; De Garay 1991; personal communications).

After the Second World War, Luminous Processes Inc. from New York installed a radium dial-painting plant in the city of Toluca. This facility probably started operation between 1954 and 1955 using radium at a rather low concentration (1-4 μg/g paint). This was a typical range of concentration for alarm clock dials. No documents were found concerning radium contamination in this facility. In 1964, the plant changed to tritium as the main luminous agent (Rundo 1991; Stebbings 1991; personal communications).

INTERNATIONAL ACTIVITIES

Following World War II, when the USA and the European countries were devoted to the study of peaceful uses of nuclear energy, the government of Mexico undertook some actions concerning radioactive materials.

In 1949, a law declared those minerals containing thorium and uranium as "national reserves", thus bestowing upon three separate government agencies authority on the use, exploitation, and research of radioactive minerals in the country. These agencies were the General Court of Justice, the Ministry of

Water Supply, and the Ministry of Education through its agency, the National Institute of Scientific Research (INIC). During that time, the General Court of Justice appropriated, for reasons of national security and by an act of law, all chemical reagents containing ^{235}U, such as uranyl nitrate and acetate. The Water Supply Ministry, while dealing with exploration programs in Oaxaca, obtained uranium and thorium minerals at the so-called *Mina del Muerto*. The chemicals appropriated by law and a large amount of uranium and thorium minerals from Oaxaca's pegmatite were given to INIC. With these materials and thanks to the presence of enthusiastic Mexican scientists who had previously visited the Enrico Fermi laboratory, a project for the construction of a nuclear demonstration reactor was started at INIC (1951-1956). Unfortunately, only some aspects of the project were carried out. The planning of the whole project is shown in Fig. 1, where the dashed lines indicate the unfinished activities and the continuous lines show the activities which were completed. For the latter, the Inorganic Chemistry Laboratory of INIC started operations in 1951. The laboratory was devoted to the treatment of uranium mineral and its conversion to metallic uranium. As a part of this laboratory, a pilot plant for extraction and concentration of uranium, using alkaline hydrometallurgy, and an analytical control section were installed. Uranium tetrachloride was the product obtained in the early period of the pilot plant. Metallic uranium was obtained by UCl_4 reduction with metallic calcium. It was only after the First International Atoms for Peace Conference, held in Switzerland in 1955, when uranium tetrafluoride by a dry method was used for metallic uranium obtention (Palma et al. 1968).

The participation of INIC in geological studies for uranium prospection started at El Sotolar, Chihuahua, where the above-mentioned American laboratory had found highly radioactive material in mercury samples.

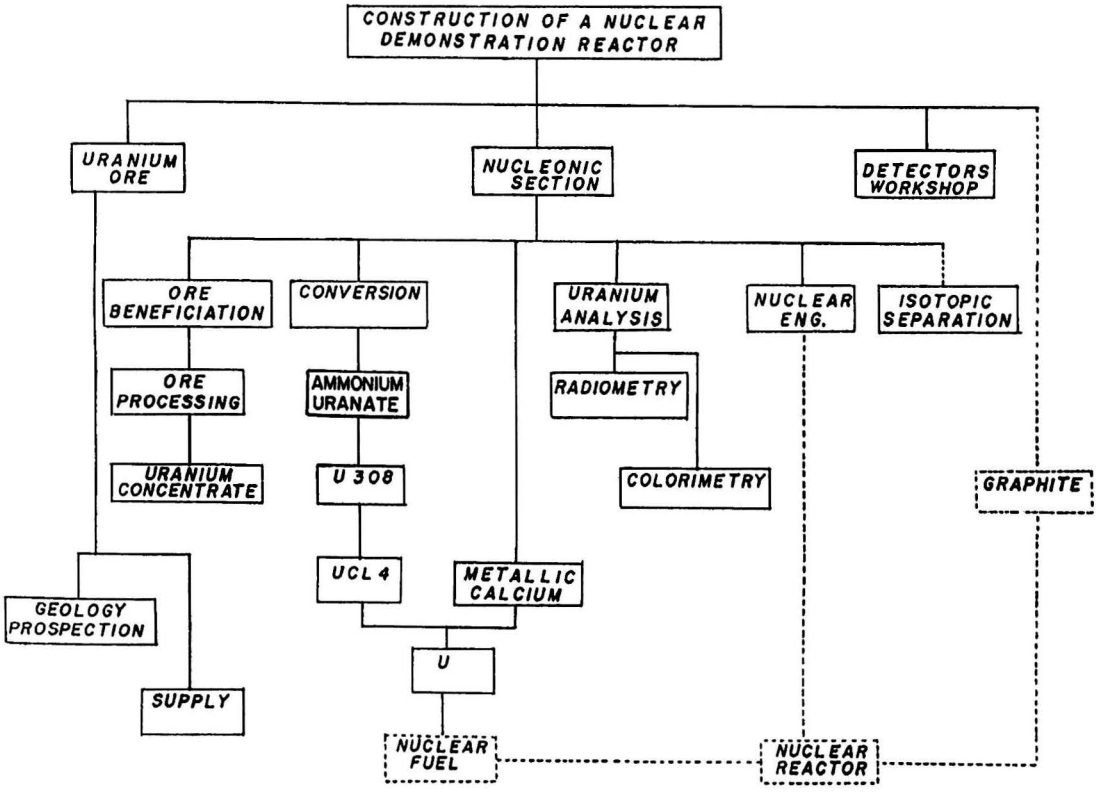


Fig. 1. Sketch of the project for the construction of a natural uranium nuclear demonstration reactor, INIC (1951-1956).

THE INSTITUTIONS AFTER 1955

In May 1955, two young Mexican physicists attended the First International Course in Radioisotope Techniques for Non-U. S. Citizens held at the Oak Ridge Institute of Nuclear Studies, in Tennessee, USA. In August of the same year, Mexico participated in the First International Atoms for Peace Conference held in Switzerland.

With the establishment of the National Commission of Nuclear Energy (CNEN), by a government decree on December 19, 1955, two main activities were established in the country: a training program on Radioisotope Techniques and Nuclear Instrumentation; and the creation of specialized programs and laboratories. The nuclear science laboratories of INIC were incorporated into the new institution.

Some of the CNEN projects were related to uranium exploration and extraction, radionuclide applications, radiological security, radioactive standards, radionuclide dilution, genetic effects of radiations, electromagnetic radiation, plasma physics, and reactor physics.

The CNEN Uranium Exploration program promoted uranium prospection by lending Geiger counters to all interested prospectors in return for purchasing their findings. The results of this program showed that uranium was found associated to clays, shales, and rhyolites in the north of the country, in Baja California, Coahuila, Sonora, and Chihuahua, four of the states on the USA border. The geological units of these regions are formed by sedimentary rocks of Cambrian, Devonian, Mississippian, Pennsylvanian, Triassic, Jurassic, and Cretaceous origin. In the south of the country, particularly in Oaxaca, uranium and thorium minerals were also found in pegmatites. In 1958, Antúnez already estimated that these rocks were highly favorable for uranium content (Antúnes 1958). Figure 2 shows a map of Mexico published in 1958 where regions with radioactive mineralizations were found; these zones were recommended for prospection and exploitation of uranium.

The CNEN programs devoted to radionuclide applications in medicine, radioactive standards, and radionuclide dilution worked together very closely in the early 60's.

In 1962, tellurium was irradiated for the first time in a nuclear research reactor on exhibition in Mexico within the Cooperative Mexican - United States Atoms at Work Exhibit. On this occasion, ^{131}I was obtained in small amounts by distillation (Bulbulian 1963).

By this time in Mexico, scientists specialized abroad were already working in fields such as nuclear reactors, nuclear medicine, and industrial applications.

The first ^{131}I sample for medical use, which was imported from Canada in February 1962, was diluted and sent to the hospitals. During the first part of this year, the average ^{131}I amount bought by private and public hospitals was 30 mCi per month. The amount required at the end of the year was already 115 mCi per month and in 1967, 2 Ci per month. Fifteen differently labeled molecules were also systematically imported for medical uses in that year. The CNEN's laboratories had been operating in rented houses in Mexico City, but in 1964, the construction of a Nuclear Center was begun and laboratories specially constructed for the needs were planned. In 1967, the facilities at the dilution laboratory were not adequate to satisfy the requests of the hospitals, so the laboratory moved to the Nuclear Center of Mexico as part of a program called Radioisotope Production.

The main industrial application of radionuclides in the early 60's was radiography, using gamma emitters (García-Moreno 1963).

The CNEN Standards Laboratory was in charge of the importation of radioactive standards, and of the calibration of detection devices and radioactive material. For this purpose, ^{226}Ra standards, and kits of several gamma and beta emitters were also acquired for national use. At the end of 1967, secondary standards were offered on loan to the laboratories using radioactivity detection devices.

THE NUCLEAR CENTER OF MEXICO

The CNEN laboratories moved to the Nuclear Center of Mexico between 1966 and 1970. The Nuclear Center occupies an approximate area of 1.50 km^2, 36 km west of Mexico City. Its main equipment included a Triga Mark III nuclear reactor and a tandem accelerator. In addition to the projects already working in Mexico City, new projects were added to the activities of CNEN at the Nuclear Center, such as Radioisotope Production, Reactor Physics and Engineering, Solid State Detectors, Accelerator Design for Industrial Applications, Experimental Nuclear Physics, and Nuclear Fuels, etc. The projects related to the exploration and exploitation of uranium were carried out near the mines, mainly in the northern part of the country. All the workers in the mines were under radiological control with occupationally exposed personnel status.

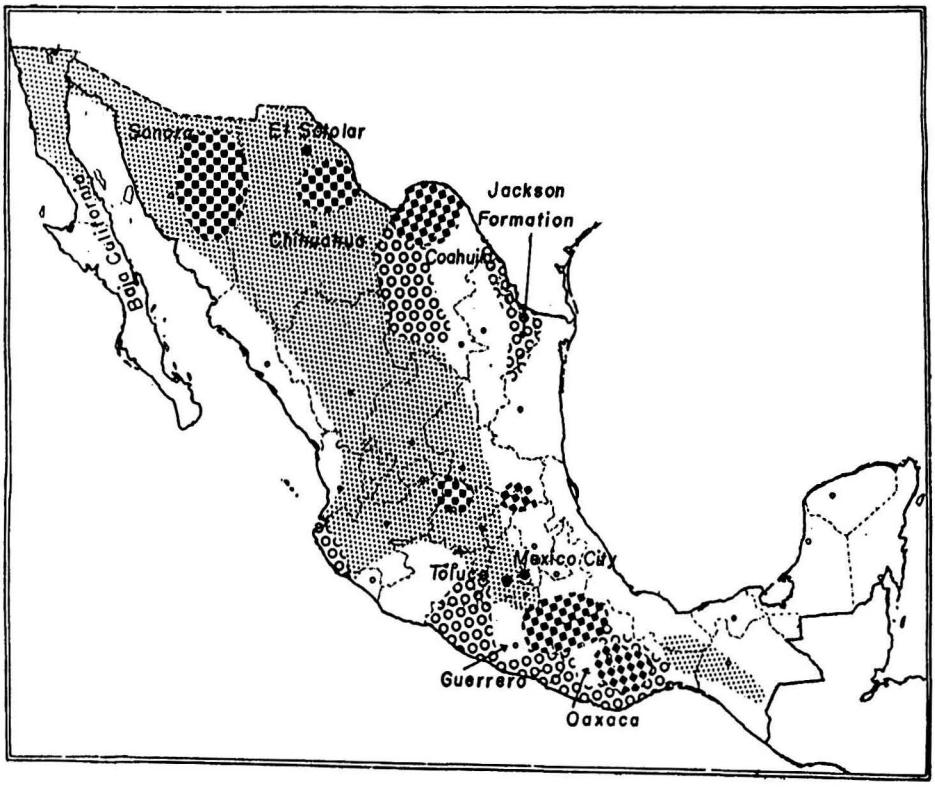


Fig 2. Localization of radioactive minerals in Mexico:

Under exploration by CNEN;

radioactive minerals known but not explored by CNEN;

recommended for exploration (Antúnez 1958).

CONCLUDING REMARKS

The activities performed from 1940 to 1970 laid the basis for the laboratories and for the development of the human resources available today.

The exploration and exploitation of uranium ores realized by CNEN, renamed Instituto Nacional de Energia Nuclear in the 70's, was one of its most important projects (Tejera and Rodriguez Soto 1977). However, since the early 80's, prospection and exploitation of uranium on a national scale have been substantially reduced.

The use of radium-based paintings for industrial purposes has evolved. Presently, under a radiological control program, some enterprises produce bulbs with thorium mantles, metal gauze structures coated with thorium and used in gas/petrol lanterns.

For medical purposes, radium has been replaced by other radionuclides, but radium standards for calibration purposes are still used.

The nuclear fuel program for the Laguna Verde nuclear power plant in the State of Veracruz today represents one of the principal projects of the Instituto Nacional de Investigaciones Nucleares (formerly Instituto Nacional de Energia Nuclear).

Radionuclide importation for medical and industrial purposes has continued in larger amounts than in the

60's. Short-lived radionuclides for medical applications and research are produced in Mexico, mainly using the irradiation facilities of the Triga Mark III nuclear reactor located at the Nuclear Center.

As a last remark, it is worth noting that the Treaty of Tlatelolco was signed by several nations in Mexico in 1967. The nations signing this document agreed, among other things, to develop the peaceful uses of nuclear energy.

Acknowledgment —— The authors are grateful for the personal conversations with pioneers in the uses of radioactivity in Mexico. Special mention is given to Dr. Leon De Garay, Dr. Rodolfo Diaz Perches, and I. Q. Gustavo Barrera Echeverry. We also acknowledge the personal communications from Dr. James H. Stebbings and Dr. John Rundo. We thank E. Tamez and P. Peña for collecting information; R. Noriega for language revision; and C. Martinez and G. Zenteno for technical assistance. We acknowledge financial support from CONACyT, Mexico.

REFERENCES

Antúnez, F. Manual para la Exploración del Uranio. CNEN (Comisión Nacional de Energía Nuclear), Mexico City; 1958.

Bulbulian, S. Produccion de ^{131}I. Thesis. Universidad Nacional Aútonoma de México, Mexico City; 1963.

Espíndola, J.M. Minerales y Rocas. Mexico City: SEP-UNAM; 1986.

García-Moreno, C. Técnicas de Gammagrafía Industrial. Thesis. Universidad Nacional Autonoma de México, Mexico City; 1963.

Gonzalez Reyna, J. Mineria y Riqueza Minera de Mexico. Monografias Industriales del Banco de México S.A., Mexico City; 1944.

Iturbe, J.L. Determinación de Uranio en Cerámica de Oaxaca. Tech. Rep. ITQ 0391, ININ (Instituto Nacional de Investigaciones Nucleares), Mexico City; 1991.

Palma, F.A.; Herrera, M.; Ríos Nieto, A. Obtención de Uranio Metálico. Rev. Soc. Quím. Mex. XII(4): 173A-176A; 1968.

Tejera, A.; Rodriguez Soto, E. La Mineria de las Materias Primas Radiactivas en México. INEN (Instituto Nacional de Energía nuclear), Mexico City; 1977.

Environment International, Vol. 19, pp. 519-526, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

HISTORY AND DEVELOPMENT OF WHOLE BODY COUNTING IN BRAZIL

A.S. Paschoa, C.A. Nogueira de Oliveira, M.C. Lourenço, J.L. Lipsztein, and O.Y. Mafra Guidicini
Comissão Nacional de Energia Nuclear, Rio de Janeiro, Brazil

I.M. Antunes*
Indústrias Nucleares Brasileiras, Rio de Janeiro, Brazil

EI 9203-128 M (Received 4 March 1992; accepted 25 July 1993)

The first whole body counter (WBC) built in Brazil used sugar as shielding material, and for this reason became internationally known as the "Sugar Bowl". The main purpose of building that first WBC was to detect natural gamma emitters other than ^{40}K in the inhabitants of Guarapari, then a small fishing village with a population not greater than 6 000 people, suspected of having long-lived contamination with natural radionuclides of the ^{232}Th and ^{238}U series. However, the Sugar Bowl was also used to whole body count the workers of a gas mantle factory. This paper reviews the history behind the construction and uses of the Sugar Bowl, as well as presents a brief view of the basic characteristics of the subsequent WBCs built in Brazil. A total of 12 WBCs have been in existence in this country until today.

INTRODUCTION

Various investigations suggested the possibility that long-lived products of the thorium series might be present in the body of some Guarapari residents, resulting in high internal alpha doses. This was the rationale behind the construction of the first whole-body counter (WBC) ever built in Brazil. Sugar was used as shielding material. Accordingly, this WBC became known worldwide as the "Sugar Bowl".

In 1965, in an open area known as the "pilotis" at the building Cardeal Leme at the Pontifícia Universidade Católica do Rio de Janeiro (PUC-Rio), the Sugar Bowl was assembled. However, it proved to be short-lived, as the sugar shielding absorbed moisture from the highly humid air of Rio de Janeiro in the summer of 1967, and became syrup. The data yielded by the Sugar Bowl, however, proved the need for a longer-lasting installation.

Table 1 lists the WBCs and all alike installations that ever existed in Brazil. One can notice in Table 1 that there was a gap between 1969 and 1983 in building WBCs in the country. The reason for this gap is not well understood at this writing. A likely reason,

Permanent affiliation: Pontifícia Universidade Católica, Rio de Janeiro, Dept. de Física, Cx. Postal 38071, RJ 224530-900, Brazil.

Table 1. Whole body counters and alike installations in Brazil.

UNIT	TIME	TYPE	PLACE
A	65 - 66	"Sugar Bowl"	PUC-Rio
B	66 - 67	"Iron shield" (IEA I)	IPEN
C	68 - 69	WBC "Iron Room"	PUC-Rio
D	68 - 69	WBC (IEA II)	IPEN*
E	68 - 69	Mobile WBC	PUC-Rio
F	79 - 80	"Shadow Shield"	MB°/FURNAS*
G	83 - 85	WBC	IRD/CNEN
H	85 - 86	"People Mover"	FURNAS*
Ia,b	87	Improvised WBCs**	Goiânia
J	89 - 90	WBC (Cuba)	NUCLEP***

° MB = Marinha do Brasil
* Unit bought from foreign commercial suppliers
** Emergency building system to monitor highly contaminated persons
*** NUCLEP = Nuclebrás Indústrias Pesadas

however, could be the discontinuity of the studies on natural radioactivity, plus a change in priorities concerning the radioprotection of workers in the nuclear and nonnuclear industries. However, from the historical viewpoint, it is worth investigating the motives that resulted in such a gap.

At the time that the Sugar Bowl was turning into syrup at PUC-Rio, the Instituto de Energia Atômica (IEA), located in the State of São Paulo, was building an iron shield to measure hospital patients. However, the iron shield was installed far from hospitals, at the campus of the Universidade de São Paulo (USP), creating discomfort for ambulatory patients to be measured. Soon, the need for a WBC in a hospital environment became clear.

The substitute for the Sugar Bowl at PUC-Rio was a WBC using slabs of steel, lead sheets, and bricks as shielding material. The steel was obtained from the Companhia Siderúrgica Nacional (CSN, the National Steel Co.) and the lead was bought with funds from the Comissão Nacional de Energia Nuclear (CNEN, the Brazilian Nuclear Energy Commission). The shielding was set up in the basement of PUC-Rio, almost under the place where once the Sugar Bowl was installed. The iron shielding was separated from the walls of the room so that further shielding made of iron, lead, and water could be added at a later time.

The CSN executives at the time were cooperative, but they could not quite understand the request to deliver steel sheets of 1-cm thickness (to be added to the ceiling), when it was easier to deliver a slab 10-cm thick. Nor could they understand why a physicist would be interested in such obscure technical problems as, for example, whether cobalt entered the process of making the steel to be used as shielding. The shielding was built in the years 1968 and 1969. Simultaneously, an attempt was made to build a portable or mobile WBC, because it was not easy to convince people from Guarapari to spend three days away from home to go to Rio de Janeiro and be whole body counted. The success of the mobile WBC would depend on the size of the body burdens of the Guarapari people. It was understood that a mobile WBC would never equal in sensitivity a well-mounted permanent WBC system. Thus, it was decided that if there was enough reason to believe that one could find body burdens higher than 10 nCi ^{208}Tl (about 400 Bq ^{208}Tl), an attempt to measure people in Guarapari would be justifiable. Unfor-

tunately, the electronic equipment associated with the mobile WBC became damaged beyond repair on the first trip to Guarapari.

The WBC IEA II was also built in the years 1968 and 1969. The mechanical and electronic equipment were similar to those that can be found in any WBC of a major hospital that uses radionuclides. The metallic support of the NaI(Tl) crystal could be moved, and a 2π-collimating geometry was used to improve the performance of the system.

After the nuclear agreement between Brazil and the Federal Republic of Germany was signed in 1975, it became clear that more sophisticated WBCs were needed to supplement the radiation protection program associated with the large nuclear program soon to be implemented. Although a great deal of attention was given to training personnel in a variety of engineering specialties, the field of radiation protection was not well contemplated either with material or human resources. As a consequence, most of the existing and operating WBCs in Brazil became obsolete due to time and lack of proper maintenance, and were not readily updated or substituted. Soon, there was no operating WBC with the necessary reliability to support the nuclear program being implanted.

The Hospital Marcílio Dias, a naval hospital located in Rio de Janeiro, and Furnas Centrais Elétricas S. A. (FURNAS), a Brazilian utility that operates the Central Nuclear Almirante Alvaro Alberto (CNAAA 1), better known as Angra 1 nuclear power plant, became associated in 1979 to import and operate a shadow shield bought from a foreign commercial supplier. This WBC (MB/FURNAS) was the most modern installation of its kind in the country at that time.

The Instituto de Radioproteção e Dosimetria da Comissão Nacional de Energia Nuclear (IRD/CNEN) built a WBC in the mid-eighties. The construction and training of personnel lasted from 1983 to 1985. The motivation to build this WBC was the increasing need for individual monitoring of Brazilian radiation workers at a routine basis to provide the national regulatory body, CNEN itself, with body burdens and dose estimates concerning a wide range of occupational activities in and out the nuclear fuel cycle. Today, this WBC unit is still the most modern installation of this type in existence in Brazil.

In 1985, FURNAS bought a "People Mover" from a foreign commercial supplier for its ambulatory, to be used in the case of emergencies or in conjunction with a decontamination facility for its personnel, to support the Angra 1 nuclear power plant. The People Mover consists essentially of a vertical Pb shielding 5 cm thick with extra shielding around the detector column, made of 10 cm Pb plus Cd and Cu, the latter two to shield from the Pb x-rays. The detector system consisted of two 10 cm x 10 cm x 40 cm NaI(Tl) with two photomultipliers. This facility is strategically located near CNAAA 1, and it is permanently attended by trained medical staff of the ambulatory.

Few weeks after the Goiânia accident, that occured in September 1987, an improvised WBC was built at the Hospital Geral de Goiânia (HGG, Goiânia General Hospital) to attend the need to monitor the decontamination efforts, including those of the most contaminated victims. After three attempts, a decision was made concerning the equipment for the measurement of the subjects. The main problem at that time was to overcome the high electronic dead time of the counting system that could not respond adequately to such high counting rates resulting from unusual levels of internal contamination of some subjects being whole body counted.

Subsequently, an improved WBC was built in Goiânia to attend the dual purpose of monitoring the workers involved in building the final waste repository (Paschoa at al. 1992) and to follow up the earlier measurements of those subjects that became contaminated during the accident (Melo et al. 1993).

SELECTED EXPERIMENTAL INFORMATION

The shielding of the Sugar Bowl consisted of 10 Mg of sugar, contained in 166 sacks of sugar, each 60 kg, obtained from the Instituto do Açúcar e do Alcool (IAA, the Brazilian Institute of Sugar and Alcohol). This sugar was supplemented with 0.5 Mg of iron from the CSN (Cullen 1966). Sugar was used as shielding material, because of its chemical purity and the then low price of this commodity in Brazil. Furthermore, one needs to observe the graphs of mass attenuation coefficient of lead, iron, and sugar as a function of gamma ray energy, as shown in Fig. 1. Details of the determination of the mass attenuation coefficient for sugar can be found elsewhere (Paschoa 1967). Data on the Sugar Bowl are scarce. However, one can reminisce that the sugar sacks were piled in the form of a bowl giving the equivalent of 15 cm of iron shield below and 10 cm on the side walls. The person sat in a small cave with the detector close to the chest, and the iron and lead packed closely around the detector. Figure 2 shows an external view of the Sugar Bowl adapted from an old photograph.

A background index, defined as the total counting rate from 0.4 to 2.0 MeV divided by the volume of

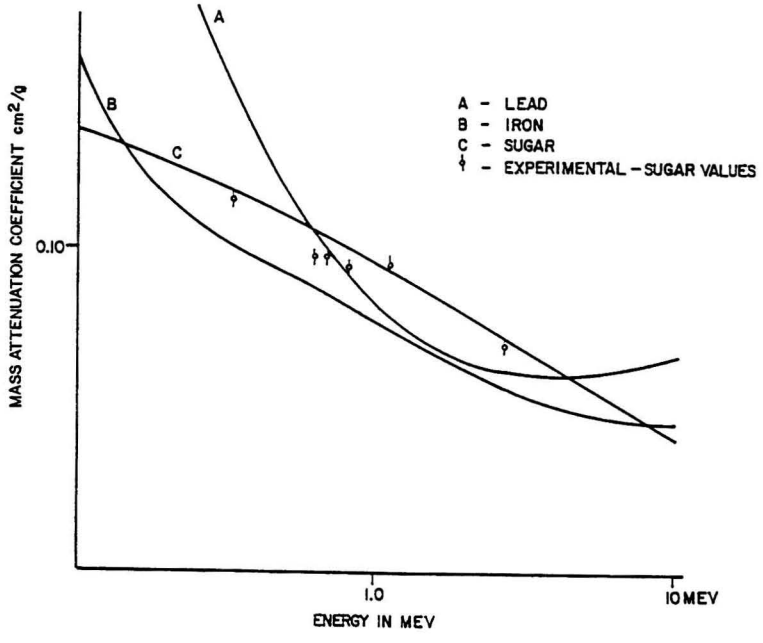


Fig. 1. Mass attenuation coefficients of sugar, lead, and iron as a function of photon energy.

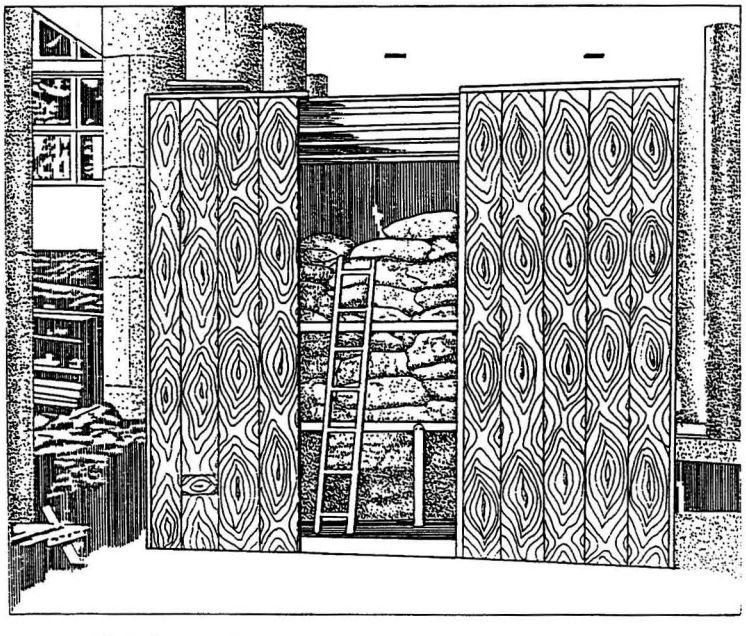


Fig. 2. External view of the Sugar Bowl, adapted from an old photograph.

the NaI(Tl) crystal, was found to be 0.92 cpm.cm^{-3}. This value was considered fairly good for a provisional counter. The counter was calibrated with standard solutions of ^{226}Ra and ^{228}Ra. The solutions were sandwiched between two 10-cm thick plastic bottles filled with distilled water to represent a phantom of the chest region. The standard error in the calibration experiment can be used to estimate the lower limit of sensitivity. However, for the Sugar Bowl, the minimum significant activity (MSA) and the minimum detectable activity (MDA) were calculated for each subject on the basis of two measurements of the background, taken immediately before and after the actual measurement of the subject. These quantities were calculated using the following expressions:

$$(MSA) = \gamma \cdot K \cdot (2B)^{1/2} \quad (1)$$

where
γ is the calibration factor in Bq.counts^{-1} (or pCi.counts^{-1}) for 50-min counting time; B is the number of counts due to the background in the channels of interest, based on 50-min counting time; and K is the constant corresponding to a 90% confidence level.

In addition, the MDA was defined as

$$(MDA) = 2 \cdot (MSA) \quad (2)$$

The Sugar Bowl MDAs for ^{224}Ra and ^{228}Ra were reported by Cullen (1966) to be 1.2 nCi (44 Bq) and 1.9 nCi (70 Bq), respectively.

The IEA I, made at IPEN, unit B in Table 1, had 15-cm iron walls, a 20-cm diameter NaI(Tl) cylindrical detector with appropriate electronics, and facilities to measure in two geometries: chair and arc.

The basic information on the WBC "Iron Room", unit C in Table 1, has been reported elsewhere (Paschoa 1990). However, it is worth summarizing some of the characteristics of unit C. The interior volume dimensions of unit C were 2 x 1 x 1.5 m^3. The ceiling and the floor were made of iron 0.20 m in thickness; and the side walls were 0.10 m thick. The detector was a 10 cm x 15 cm NaI(Tl) cylindrical crystal that could be moved horizontally and vertically, and could also rotate to face the ceiling, the floor, or a few intermediate angles. The typical counting time was 50 min, and the chair geometry was used. The minimum detectable activities ranged from 63 Bq (1.7 nCi) for ^{208}Tl to 333 Bq (9.0 nCi) for ^{137}Cs, as shown in Table 2.

The IEA II, unit D in Table 1, was a WBC with a small 7.6 cm x 7.6 cm NaI(Tl) cylindrical detector provided with a 1.7-cm lead collimator to measure the subjects in suppine and prone positions. The typical detection limits of IEA II, for 30-min counting time and the detector at 65 cm from the subject, ranged from 16 nCi (590 Bq) at 120 keV up to 520 nCi (19 kBq) at 1.86 MeV.

The poorer technical performance of IEA II as compared with the Iron Room at PUC-Rio, for example, can be easily traced to the smaller detector volume used in IEA II *vis-a-vis* the larger and low-background detector, provided by the IAEA for the Iron Room, plus the fact that the shielding of the latter unit was carefully built by CSN engineers with pre-World-War-II steel to avoid contamination with weapon-tests-produced radionuclides.

The attempt to measure some Guarapari workers and inhabitants *in loco* with a mobile WBC, denoted as unit E in Table 1, resulted from the need to test the hypothesis that if there were measurable body burdens in those workers and inhabitants they were, quite likely, due to radon 220 inhalation. In such case, the body burdens would be short lived, decaying with the 10.6 h half life of ^{212}Pb, making it difficult to detect such body burdens considering the 63 Bq ^{208}Tl MDA of the Iron Room at PUC-Rio. Thus, if the ^{208}Tl body burden of a subject in Guarapari would be 8nCi (300 Bq) at the time, he (or she) was leaving that village to be measured (i.e., at about 24 h prior to be actually whole-body counted), the body burden at the time of the measurement would be less than the ^{208}Tl MDA. This was the rationale behind the 400 Bq ^{208}Tl (10 nCi) body burden hypothesis that justified the attempt to build the mobile WBC to screen the subjects to be brought from Guarapari to be measured in Rio de Janeiro, far away from home.

Unit F in Table 1 was, as mentioned earlier, a commercial shadow shield for use in emergencies and isolated cases of radionuclide contamination at Angra I nuclear power plant. It was part of a decontamination facility built by Furnas at a location a few kilometers away from the nuclear power plant.

Unit G in Table 1 is the WBC built at IRD/CNEN. This is the only Brazilian facility able to detect and identify internal photon emitters in the energy range from 10 keV up to 3 MeV (Oliveira et al. 1989). Most of the equipment used in this WBC was obtained with IAEA assistance through a series of coordinated research projects (CRPs) and technical cooperation programs (TCPs). The first of those IAEA CPRs dates back to 1985 when a Phoswich Detector and associated processing electronics were acquired, twenty years after the Sugar Bowl and the Iron Room at PUC-Rio were conceived and built with the same

Table 2. Minimum detectable activities (MDAs) for the WBCs at PUC-Rio (unit C) and IRD/CNEN (unit G).

NUCLIDE	ENERGY (MeV)	COUNTING TIME (min)	MDA Bq(nCi) PUC-Rio	IRD/CNEN
^{241}Am	0.0169†	60††(lungs)	–	1.6(0.043)
	0.0596(36)			
^{239}Pu	0.017*	30††(*skull*)	–	915(25)
^{57}Co	0.122	15**••	–	146(39)
^{131}I	0.364	5**•• (thyrold)	–	80(2.2)
^{137}Cs	0.662	50**• 15**••	333(9.0)	152(4.1)
^{212}Bi-^{228}Th	0.727	60**•• (lungs)	–	42(1.1)
^{54}Mn	0.835	50**••	307(8.3)	
^{228}Ac	0.911	60**•• (lungs)	–	20(0.54)
^{65}Zn	1.115	15**••	–	310(8.4)
^{60}Co	1.173	50**	89(2.4)	–
^{22}Na	1.271	50**	159(4.3)	–
^{40}K	1.460	50**	189(5.1)	–
^{214}Bl	1.764	50**	118(3.2)	–
^{88}Y	1.836	50**	104(2.8)	–
^{208}Tl	2.615	50**60**••(lungs)	63(1.7)	26(0.7)

† A combination of the Lx-ray peaks at 13.9 keV (13.3%) and 17.8 keV (19.4%).
†† Phoswich
* Taking into account that most gamma transitions from the excited states of the daughter nuclides are converted to orbital electron emissions followed by the Lx-ray emissions of uranium ($L\alpha$ at 13.6 keV - $L\beta$ at 16.4 and 17.2 keV - $L\gamma$ at 20.2 and 20.8 keV).
** NaI(Tl)
• PUC-Rio
•• IRD/CNEN

helpers, consisting of the IAEA for equipment and the New York University (NYU) for training of personnel. The IRD/CNEN system of detectors was constituted of two NaI(Tl) cylindrical crystals 20 cm x 10 cm and 7.6 cm x 7.6 cm, plus three 13-cm Phoswich Detectors. The latter were donated by the Kernforschungszentrum Karsruhe (KfK). The monitoring room was 2.5 cm x 2.5 cm x 2.62 cm made by 15-cm steel plates, internally lined with 0.3-cm thick lead, 0.15-cm thick cadmium, and 0.5-cm thick copper sheets. The detectors were placed in such a way that they could be positioned with three degrees of freedom. Anthropomorphic phantoms of the Lawrence Livermore Laboratory (LNL) type were used for calibration purposes.

It has been assumed by some investigators that the steel used in the construction of the IRD/CNEN's WBC was free from radioactive contamination, because it was manufactured in the Southern Hemisphere. Such assumption, however, needs further clarification because the Southern Hemisphere, in general, and the southern cone of South America, in particular, received radioactive fallout from weapon tests in the atmosphere. However, samples of the

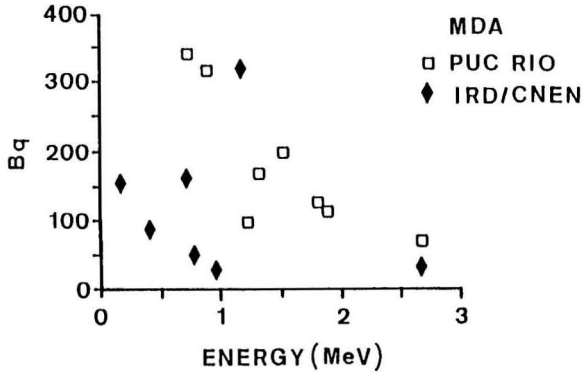


Fig. 3. Minimum detectable activities of the PUC-Rio and IRD/CNEN WBCs, without consideration of counting time, as a function of the energy of gamma emissions of selected radionuclides.

steel used in the IRD/CNEN's WBC were tested for radioactivity at NYU's gamma spectrometric facility, and proved to be adequate for the purpose of building a WBC.

The evolution of whole-body counting in Brazil is fairly well illustrated by the experimental data on the MDAs that appear in Table 2. The MDAs in Table 2 are those of the WBCs built at PUC-Rio and at IRD/CNEN, which are denoted as units C and G in Table 1. One can observe from Table 2 that the MDA for ^{137}Cs is 60% better at IRD/CNEN WBC, for a 10-min counting time, than that at PUC-Rio, for the same radionuclide, counted for a time five times longer. In addition, the IRD/CNEN WBC has the capability of measuring ^{239}Pu and ^{241}Am through their low energy photopeaks detected by the Phoswich Array. Further details of the IRD/CNEN WBC, including its operational characteristics are reported by Oliveira (1991). Figure 3 shows the MDAs (in Bq), ignoring differences in counting time, for the WBCs of PUC-Rio and IRD/CNEN, as a function of the energy of the gamma emissions of selected radionuclides. Figure 3 shows the superiority of performance of the IRD/CNEN's WBC as compared to the one at PUC-Rio. This reflects the evolution of whole body counting in Brazil from the late sixties to the mid-eighties.

As it is well known, in September 1987, a ^{137}Cs teletherapy source was stolen from an abandoned building in Goiânia, and then breached. This act resulted in high internal ^{137}Cs contamination of the individuals involved and the persons close to them. Initially, improvised WBCs were built *in loco*, taking advantage of the facilities of the Hospital Geral de Goiânia (HGG), where the contaminated persons were first being treated. Later, a partiallly decontaminated house was used. These WBCs are identified in Table 1 as units Ia and Ib. The basic detection system of both units consisted of a 20 cm x 10 cm NaI(Tl) cylindrical crystal detector, collimated with 5 cm lead wrapped around it. The reason for using such an unnecessarily large and efficient detector for whole body counting highly contaminated individuals was the fact that it was the only one readily available at the time. The detector was positioned at 2.2 m from the floor, and 2.05 m from the center of the chair used to approximate the configuration of an arc geometry for whole body counting of the contaminated subjects. The dead time under this configuration was 8% for the subject with the highest ^{137}Cs contamination. The MDA of unit Ia for ^{137}Cs was 9.2 kBq (247 nCi) for a 2-min counting time. Unit Ia was in operation from the first week of November 1987 until 15 January 1988, when the detecting system was moved to the Goiânia 57th Street, near the very place where the source was breached. It then became unit Ib. At that time, all contaminated patients had already been released from HGG. The radiation background, however, was four times higher than that at the HGG. Thus, it became necessary to identify the origin of such a high counting rate at the building. After a detailed search, the roof of the house was identified as the area contaminated with ^{137}Cs.

The top side of the detector was then shielded to overcome the problem. The extra shielding allowed a reduction of 20% in the MDA, obtained earlier at

the HGG for unit Ia, resulting in a specific MDA for ^{137}Cs of 7.3 kBq (197 nCi), for 2-min counting time in unit Ib.

A total of 356 subjects were monitored in Goiânia in units Ia and Ib from 8 November 1987 until 13 April 1988. From those, 40 had body burdens above the MDAs of the detecting systems.

Unit J of Table 1 is the WBC ordered by the IAEA, projected by CNEN, and built by NUCLEP, that was sent to Cuba in 1992, and quite recently installed in a building dedicated to whole body counting. The technical characteristics of this WBC will be published by Cuban scientists in due time.

The objective of this paper is to preserve the memory of the efforts made by past and present generations of scientists in the field of radiation protection, so future generations can better appreciate their inheritance.

Acknowledgment — The inspiration of persons like Drs. F. X. Roser, s.j., T. L. Cullen, s.j. (both deceased), E. Penna Franca and M. Eisenbud rendered possible to write this article.

REFERENCES

Cullen, T.L. Dosimetric measurements in Brazilian regions of high natural radioactivity leading to cytogenetic studies. In: Proc. first international congress of the International Radiation Protection Association, Rome; 1966.

Melo, D.R.; Lipsztein, J.L.; Oliveira, C.A.N.; Bertelli, L. The effect of pregnancy on Cs-137 metabolism. In: Bioassay, analytical and environmental radiochemistry; 1993. (In press)

Oliveira, C.A.N.; Lourenço, M.C.; Dantas, B.M.; Lucena, E.A.; Bertelli, L. The IRD/CNEN whole-body counting facility: background and calibration results. Rad. Prot. Dos. 29: 203-208; 1989.

Oliveira, C.A.N.; Lourenço, M.C.; Dantas, B.M; Lucena, E.A. Design and operation of a whole-body monitoring system for the Goiânia radiation accident. Health Phys. 60: 51-55; 1991.

Paschoa, A.S. Determination of the coefficient of gamma ray attenuation in sugar. In: A study of natural radioactivity in Brazil. Pontifícia Universidade Católica, Rio de Janeiro, NYO - 2577-2578; 1967: 38-43.

Paschoa, A.S. Nuclear and atomic analytical techniques in environmental studies in South America. In: Zeisler, R.; Guinn, V., eds. Nuclear and analytical methods in the life sciences. 1990: 647-660 or Biol. Trace Elem. Res. 27: 647-660; 1990.

Paschoa, A.S.; Heilbron Filho, P.F.; Malamut, C.; Tranjan Filho, A. Waste management in Brazil: basis to build the final repository for the radioactive wastes from the Goiânia accident. In: Post, R.G., ed. Waste management; Arizona Board of Regents. 1992; 133-138.

Environment International, Vol. 19, pp. 527-530, 1993
Printed in the U.S.A.

0160-4120/93 $6.00 +.00

BOOKS

Acid Soil and Acid Rain. 2nd Edition. I.R. Kennedy. Research Studies Press Ltd., Somerset, England; 1992. 254 pp. (ISBN 0-86380-124-2) hardcover.

According to the author, the primary objective of this book is to examine the fundamental chemical processes involved in the acidification of ecosystems. Because the book was written before the completion of the National Acid Precipitation Assessment Program (NAPAP), the conclusions of the book are somewhat outdated. However, the chemical processes continue to be useful and valid.

Carbon Adsorption for Pollution Control. Nicholas P. Cheremisinoff and Paul N. Cheremisinoff. PTR Prentice Hall, Englewood Cliffs, NJ, 1993. 216 pp. (ISBN 0-13-393331-8) hardcover.

This book resulted from the collaboration of an engineer employed in a major industry and a university professor and thus provides the perspectives of both academic and industrial user. The comprehensive coverage includes basic adsorption processes, adsorption of liquids and gases, and regeneration. An important part of this book is safety and environmental controls. This book is highly recommended as desk copy and as a library reference.

A Dictionary of Environmental Quotations. Barbara K. Rodes and Rice Odell. Simon & Schuster, New York, NY, 1992. 343 pp. (ISBN 0-13-210576-4) $35.00 hardcover.

This book contains over 3700 quotations in 143 categories from acid rain to zoo. The authors are associated with the World Wildlife Fund and the Conservation Foundation and thus the quotations are primarily from those sources.

Electrochemical Engineering and the Environment 1992. P.J. Mitchell. Institution of Chemical Engineers. Hemisphere Publishing Corporation, Bristol, PA, 1992. 308 pp. (ISBN 0-85295-280-5) $79.00 hardcover.

This volume contains papers presented at a symposium held in April 1992 at Loughborough University. The three major topics covered in the meeting were energy efficient cell and process design, water treatment and energy conversion, and management. Although several papers covered fundamental aspects of electrochemical processes, there were others devoted to subjects that are of interest to environmental scientists. These included production of ozone, efficient and rechargeable batteries, electrochemical processes for removal of heavy elements from effluent streams, wastewater treatments, and fuel cells.

Energy Options for Africa. Stephen Karekezi and Gordon A. Mackenzie, eds. Zed Books, London, U.K., 1993. 184 pp. (ISBN 1-85649-205-2) $55.00/£32.95 hardcover.

The United Nations Environment Programme Collaborating Centre on Energy and Environment consists of the Risø National Laboratory, the African Energy Policy Research Network, and the Foundation for Woodstove Dissemination. The center organized an African Energy Expert Meeting which was held in Nairobi, Kenya in May 1992. The objective of the meeting was to assess the applicability of alternative energy sources. The papers resulting from that workshop are published in this volume.

Environmental Quality. Dale Curtis and Barry Walden Walsh, eds. Council on Environmental Quality, U.S. Government Printing Office, Washington, DC, 1993. 451 pp. (ISBN 0-16-0412-4) softcover.

This is the 23rd Annual Report of the (US) Council of Environmental Quality. As in previous years, this CEQ report contains a wealth of data on pollutants, governmental actions, production of chemicals, and much other useful information. This report is highly

recommended as a desk copy and as a library reference.

Grasslands and Grassland Sciences in Northern China. James Ellis, Chairman. Committee on Scholarly Communication with the People's Republic of China. National Academy Press, Washington, DC, 1992. 214 pp. (ISBN 0-309-04684-X) $31.00/£37.25 softcover.

This report resulted from a cooperative effort between a committee of the (US) National Academy of Sciences (NAS) and its Chinese counterpart. It demonstrates that scientific cooperation can continue despite political tensions. The report is divided into four parts. Part I presents an overview of the ecology, society, and land use practices in the grasslands of northern China, based on published sources and direct observations by members of the panel. Part II contains reviews of recent Chinese literature on seven topics: scientific research on the grasslands of northern China, scientific research on the grasslands of each of five subregions—the Northeast, Xilingele League of Inner Mongolia, central Inner Mongolia, Gansu and Qinghai, and Xinjiang—and social science research on the region as a whole. Part III consists of a description of Chinese institutions for grassland studies. The final part describes key issues. This report is highly recommended as library reference and as a desk copy.

The Holes in the Ozone Scare. The Scientific Evidence that the Sky isn't Falling. Rogelio A. Maduro and Ralf Schauerhammer. 21st Century Science Associates, Washington, DC, 1992. 356 pp. (ISBN 0-9628134-0-0) $15.00 softcover.

This book counters the popular notion that there is an ozone hole. It identifies uncertainties and inaccuracies associated with the hypothesis that chlorofluorocarbons (CFCs) are harmful. The authors are accomplished scientists who write in a style understandable to the general public. The authors argue that a decision to ban CFCs was premature and the benefits of CFCs should have delayed their ban until there was sufficient knowledge. They suggest that the likelihood of any impact of CFCs on the stratospheric ozone is small. This book is recommended as desk copy.

Impact of Acid Atmospheric Deposition on the Biogeochemistry of Moorland Pools and Surrounding Terrestrial Environment. H.F. van Dobben, J. Mulder, H. van Dam and H. Houweling. Pudoc Scientific Publishers, Wageningen, Netherlands, 1992. 231 pp. (ISBN 90-220-1072-4) $61.00 softcover.

This research report contains the result of an extensive study. It includes the data from six years of biogeochemical monitoring of oligotropic ecosystems in the Netherlands. The report is most useful for those who are involved in ecosystem analysis as impacted by solute additions. Given the originality of the data and the impact of the results of the study, the more common method of dissemination of this material would be publication in a refereed journal. This report is recommended for those working in the related scientific fields.

Marine Ecosystem. Enclosed Experiments. C.S. Wong and P.J. Harrison, eds. International Development Research Centre, Ottawa, Canada, 1992. 439 pp. (ISBN 0-88936-543-1) softcover.

This book includes nine review papers describing various marine enclosures as well as research papers from six experiments in the People's Republic of China and Canada. The reviews describe benthic and pelagic enclosures and report the effects of pollutants and results of studies of phytoplankton blooms, uptake and release of dissolved organic materials, and effects of pesticides. The experiments in China and Canada examined the effects of contaminated sediments, primarily heavy metals, on bacteria, phytoplankton, and zooplankton; the pathways and fates of these metals; and the chemistry and biological effects of chemically dispersed oil. Although the symposium upon which this volume is based was held in May 1987, given the scarcity of this information from China, this book continues to be useful. It is recommended as a library reference.

Pollution Prevention Technology Handbook. Robert Noyes, ed. Noyes Data Corporation, Park Ridge, NJ, 1993.683 pp. (ISBN 0-8155-1311-9) $98.00 hardcover.

This book is largely based on several reports by the U.S. Environmental Protection Agency (USEPA). It is a comprehensive document which covers essentially the entire field of source reduction and waste minimization as seen by the EPA. Despite this shortcoming, the collection of information in one volume makes it a valuable document. Over 30 in-

dustries and other sources of waste are included in this book. This book is recommended as a library reference.

Process Engineering and Design for Air Pollution Control. Jaime Benítez. PTR Prentice Hall, Englewood Cliffs NJ, 1993. 466 pp. (ISBN 0-13-723214-4) hardcover.

This volume is intended as an introductory textbook on air pollution control engineering. It covers the design of controls of air pollutants from stationary sources (organic compounds, flue gas desulfurization, nitrogen oxides, and particulate emissions). It also includes the U.S. regulatory requirements and economic aspects of the topic. This book is recommended as a desk copy.

Soils and the Environment. An Introduction. Alan Wild. Cambridge University Press, Cambridge, U.K., 1993. 287 pp. (ISBN 0-521-43280 4) £40.00 hardcover, (O-521-43859-4) £14.95 softcover.

This book provides an overview of soil as an environmental medium on equal footing with water and air. This introductory description includes an introduction to soil properties and processes. It also describes the role of soil in plant growth, flux of atmospheric gases, and cycling of various elements including nutrients. This book is highly recommended as a desk copy.

Stormwater. Best Management Practices and Detention for Water Quality, Drainage, and CSO Management. Ben Urbonas and Peter Stahre. PTR Prentice Hall, Englewood Cliffs, NJ, 1993. 449 pp. (ISBN 0-13-847492-3) hardcover.

This book discusses various types of storm water storage facilities, including inlet control facilities, open ponds, concrete basins, storage in sewer networks, pipe packages, tunnel storage, and storage at sewer treatment plants. It examines flow regulation and the basic principles, precipitations data needs, and calculating methods for estimating storage volumes. Coverage concludes with discussion of topics that affect stormwater quality enhancement such as stormwater pollutants, fundamentals of sedimentation, design of water quality basins for stormwater design of wetland detention basins and channels, and best management practices for stormwater quality. This book is recommended as a reference for libraries and as a desk copy.

Synoptic Climatology in Environmental Analysis. A Primer. Brent Yarnal. Belhaven Press, London, U.K., 1992. 195 pp. (ISBN 1-85293-117-5) £37,50 hardcover.

Synoptic climatology deals with the relationship between the atmospheric circulation and the surface environment of a specific area. This book introduces methods used in synoptic climatology and indicates the significance of synoptic climatology in understanding environmental systems. The book is intended as a primer for environmental scientists and a text in applied climatology. This book is recommended as a desk copy.

Urban Waste Waters. Treatment for Use in Steam and Power Generation. K.M. Abdullaev, I.A. Malakhov, L.N. Poletaev, and A.S. Sobol. Ellis Horwood, West Sussex, England, 1992. 254 pp. (ISBN 0-13-929977-7) hardcover.

This most interesting book was written by three authors from the Azerbaijan Republic and one author from Russia. It represents a unique source of information on the approach used by the former Soviet Union for water treatment and refuse. The availability of environmental information provides interesting comparisons between the technologies available and used in the former Soviet Union. This book is recommended as a desk copy and as a library reference.

Water Treatment and Waste Recovery. Advanced Technology and Applications. Nichola P. Cheremisinoff and Paul N. Cheremisinoff. PTR Prentice Hall, Englewood Cliffs, NJ, 1993. 310 pp. (ISBN 0-13-285784-7) hardcover.

This introductory text describes fundamental aspects of waste water treatment. The book includes methods for separation of pollutants from waste water, biological treatment, thermal treatment, adsorption processes, and ion exchange. This book is highly recommended as a desk copy.

The Chernobyl Accident: Updating of INSAG-1. Safety Series No. 75-INSAG-7. A Report by the International Nuclear Safety Advisory Group. International Atomic Energy Agency, Vienna, Austria, 1992. 135 pp. (ISBN 92-0-104692-8) $50.00 softcover.

Does Moderate Alcohol Consumption Prolong Life? R. Curtis Ellison, M.D. American Council on Science and Health, New York, NY, 1993. 23 pp. $3.85 softcover.

Phantoms and Computational Models in Therapy, Diagnosis and Protection. International Commission on Radiation Units and Measurements. ICRU Publications, Bethesda, MD, 1992. 194 pp. (ISBN 0-913394-45-9) $55.00 softcover.

The Politics of Hazardous Waste. Charles E. Davis. Prentice Hall, Englewood Cliffs, NJ, 1993. 156 pp. (ISBN 0-13-683202-4) softcover.

Radioaktivität der Umwelt in der Schweiz.Bericht des Bundesamtes für Gesundheitswesen, Fribourg, Suisse, 1992. (ISBN 3-905235-04-8) softcover.

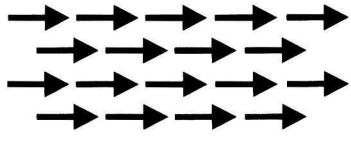
Report on Radioactive Waste Disposal. International Radioactive Waste Management Advisory Committee. International Atomic Energy Agency, Vienna, Austria, 1993. 104 pp. (ISBN 92-0-100393-5) $45.00 softcover.

Unproven Allergies": An Epidemic of Nonsense. Stephen Barrett, M.D. American Council on Science and Health, New York, NY, 1993. 14 pp. $3.85 softcover.

Vital Signs 1993. The Trends that are Shaping our Future. Lester R. Brown, Hal Kane, and Ed Ayres. Worldwatch Institute, Washington, DC, 1993. 150 pp. (ISBN 0-393-31024-8) $10.95 soft-cover.

AIR FILTRATION

An Integrated Approach to the Theory and Applications of Fibrous Filters



R. C. BROWN

Health and Safety Executive,
Research and Laboratory Services Division, Sheffield, UK

The need for clean air has probably never been greater. Many industrial employees need to avoid exposure to airborne particulates, while certain industrial processes demand clean air of exceptional quality. Medical and biological fields rely on sterile air, while filtration can improve air quality in offices, homes and all types of transport.

Filters act in a variety of complex and subtle ways. Each chapter introduces its subject matter simply, before tackling the problem in greater depth. The book therefore provides an excellent basis for detailed study, but is also invaluable to anyone needing an overview of the subject.

Readership: *For industrial hygienists, filter manufacturers and users, respirator manufacturers and users, and air conditioning engineers. Suitable for undergraduates.*

Contents

250pp 150 illus approx August 1993

0 08 041274 2 (H) **£55.00/US$88.00***

BOOKS: SAVE POSTAGE AND HANDLING CHARGES If you send full payment with your order Pergamon will pay postage.
Prices and proposed publication dates are subject to change without prior notice.
*Sterling prices quoted are definitive and apply worldwide, except in the Americas. US dollar prices quoted apply in the Americas only.

EN3A11 6/93

PERGAMON PRESS *A member of the Elsevier Science Publishing Group*

North America: Pergamon Press Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, USA Tel: +1-914-524-9200 Fax: +1-914-333-2444

UK & all other countries: Pergamon Press Ltd, Headington Hill Hall, Oxford OX3 0BW, UK Tel: +44 0865 743685 Fax: +44 0865 743946

Electrochemical Methods in Soil and Water Research

T. R. Yu *and* **G. L. Ji**
Institute of Soil Science, Chinese Academy of Sciences, PRC

This book deals with the principles and practices of electrochemical methods as applied to soil and water research, particularly those that can be carried out in the field. Beginning with the basis of potentiometric methods, including electrode potential, principles of potentiometric methods, reference electrodes, liquid-junction potential and characteristics of ion-selective electrodes, the author then proceeds to describe the properties and applications of various types of potentiometric electrodes, including glass, solid-state membrane, liquid-state membrane, oxidation-reduction and gas sensors. A special chapter devoted to commonly encountered problems will aid readers not familiar with potentiometric methods. Voltammetric methods, conductometric methods and electrochemical instruments are also discussed.

Readership: *For soil scientists and scientists engaged in water research, earth and environmental sciences and biological sciences.*

Contents

230 x 155 mm 477pp 146 illus
788 lit refs June 1993
0 08 041887 2 (H) **£65.00/US$105.00***

Prices and proposed publication dates are subject to change without prior notice.
*Sterling prices quoted are definitive and apply worldwide, except in the Americas.
US dollar prices quoted apply in the Americas only.

EA3A79 7/93

PERGAMON PRESS *A member of the Elsevier Science Publishing Group*

North America: Pergamon Press Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, USA Tel: +1-914-524-9200 Fax: +1-914-333-2444
UK & all other countries: Pergamon Press Ltd, Headington Hill Hall, Oxford OX3 0BW, UK Tel: +44 0865 743685 Fax: +44 0865 743946

Concise Encyclopedia of Environmental Systems

Edited by **P. C. Young,**
Professor of Environmental Systems and Director, Centre for Research on Environmental Systems and Statistics, Institute of Environmental & Biological Sciences, Lancaster University, Lancaster, UK

The *Concise Encyclopedia of Environmental Systems* provides a concise overview of the current state of the art in the study of environmental systems. The subjects covered include: agricultural systems; atmospheric processes and air quality; ecosystems; environmental chemistry; geology, soil processes and geophysics; hydrology, fluid dynamics and water quality; marine processes; meteorology; and climatology. In addition, many of the articles cover the methodological procedures used in environmental systems analysis, with contributions on automatic control and management; computers in modelling and management; environmental planning; environmetric methods, including time-series analysis; mathematical modelling, including data-based, physically based and simulation modelling; remote sensing and image processing; uncertainty in environmental systems; and sensitivity analysis. The encyclopedia is extensively cross-referenced on two levels - to articles of direct relevance as well as to other articles which will provide the reader with more general background information.

For those working on environmental systems, or undergraduates and postgraduates studying environmental science (including ecology), engineering, or mathematics.

Contents include:

Series: ASCI Advances in Systems Control and Information Engineering

248 x 184 mm 750pp
400 illus approx 2000 lit refs approx
Due July 1993

ISBN 0 08 036198 6 (Hardcover)

Pre-publication price £155.00/US$250.00

Post-publication price £195.00/US$310.00

(Pre-publication offer applies to orders received before 30 September 1993)

PERGAMON PRESS
USA, Central & South America: Pergamon Press, Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, USA
UK & all other countries: Pergamon Press Ltd, Headington Hill Hall, Oxford OX3 0BW, UK
A member of the Elsevier Science Publishing Group

FV 205/03

ENVIRONMENT INTERNATIONAL

Aims and Scope

Environment International, published bimonthly, is a multidisciplinary forum for the publication of original environmental literature. Vital data, causes of pollution, and methods for protection are all featured. Covering the entire field of environmental protection, *Environment International* includes contributions from the following areas

- Concentration of elements and compounds, notably pollutants
- Transport of pollutants in the environmental media
- Control technologies
- Public policy alternatives including legislation
- Release rates of pollutants from various sources
- Health and ecological effects of pollutants
- Description and interpretation of laws, regulations, and standards
- Information which will contribute to the understanding of environmental behavior of pollutants or will promote environmental protection
- National and international recommendations and practices to help bring about lasting improvement in environmental protection

From time to time *Environment International* will publish issues devoted exclusively to special topics, e.g., specific pollutants. These issues will discuss such problems as environmental concentrations, health and ecological effects, environmental kinetics, production and release rates, legislative and regulatory aspects, control technologies, and other pertinent data. Other special issues will be devoted to pollution problems in selected geographical areas. Authors and readers are encouraged to send in suggestions to the Editor-in-Chief recommending topics for these topical issues

Information for Contributors

Since the journal will serve a multidisciplinary audience, authors are urged to write for nonspecialists. In particular, they are discouraged from using expressions which are understandable only to a select audience. Clarity should be the guide when preparing manuscripts. All the contributions will be subjected to peer reviews. Owing to the broad coverage of *Environment International,* no single format can accommodate all contributions. Manuscripts must be submitted in English. Mail manuscripts to the Editor-in-Chief, *Environment International,* P.O.Box 7166, Alexandria, VA 22307, U.S.A. The following categories of contributions will be considered for publication in the journal

Articles—Previously unpublished information including data from routine monitoring operations will be published in this category. Papers dealing with new analytical techniques should contain data indicating their usefulness in environmental measurements. Similarly, basic biological effects data should be related to environmental pollutants.

Discussions—This section is intended as a forum for the readers to express their views on environmental problems on a global or national basis. Letters to the editors, editorials, contributions containing personal opinions and comments on articles published in the journal or elsewhere will be included in this section.

Book Reviews—Solicited and unsolicited reviews on relevant books will be considered for inclusion in this section.

News—This section will contain newsworthy items in the scientific, technological, governmental and political areas relating to the environment. Readers are encouraged to suggest subjects for inclusion in this section.

Preparation of Manuscripts—The journal is produced using a computer-based desktop publishing system. Accordingly, the processing time of manuscripts submitted in a computer-compatible form is greatly reduced.

Manuscripts should be typed double-spaced on one side of bond paper with wide margins. Each paper should include a title page with the title of the paper, name(s) of author(s), affiliation(s) and mailing address. In the case of co-authors, it should be clearly indicated which is to receive correspondence and proofs for correction.

All tables and figures must be camera-ready. Use a new ribbon for typing tables. Photocopies, blue ink or pencil are not acceptable. Use blue ink for lettering figures. The lettering should be large enough to be legible after the diagram has been reduced in size for printing. In the interest of conserving space, it is desirable that figures are drawn so they will reduce to the single column width of 8.5cm (3.35 in). Type figure legends double-spaced on a separate sheet of paper. Write article title and figure number lightly in pencil on the back of each piece of artwork.

Submissions in a computer-compatible form require a disk, one original copy on paper containing the original figures and tables, and a second paper copy. The system requires compatibility with IBM computers and can accept 13.5 cm(5.25 in) and 9 cm (3.5 in) disks. The WordPerfect word processing system prepared with an IBM-compatible (MS-DOS or PC-DOS) computer is the preferred submission. If WordPerfect is unavailable, WordStar, MultiMate, Revisable-Form-Text (IBM DCA Format) can be used. If none of these is available, use ASCII (American Standard Code for Information Interchange) or DOS text file. Indicate which program was used. In the absence of a computer, one original and two copies are required.

References—The journal generally follows the referencing style of the *Council of Biological Editors.* In the text, refer to the author(s) and the year. Examples for one author, two authors and more than two authors are: Wolf(1984); Repace and Lowrey(1985); Smith et al.(1979). If there is more than one listing of the same author and year, use a, b, c, etc. Prepare a list in alphabetical order containing all literature cited. In this list of references, include initials and surnames of the authors, title of the paper, name of the journal using the *World List of Scientific Periodicals* abbreviations, volume number and page number(s). Note that et al. is not acceptable in the reference list unless the publication has more than six authors. The following examples should be used as a guide:

Article: Repace, J.L.; Lowrey, A.H. A.quantative estimate of nonsmokers' lung cancer from passive smoking. Environ. Int. 11:3-22; 1985.

Report: NIH (National Institutes of Health). Report of the National Institutes of Health ad hoc working group to develop radioepidemiological tables. NIH 85-2748. Washington, D.C : U.S. Department of Health and Human Services; 1985.

Book: Henderson, P.M. Inorganic geochemistry. New York, NY: Pergamon Press; 1982.

Regulation: USEPA (U.S. Environmental Protection Agency). National primary drinking water regulations: fluoride. 40 CFR Parts 141, 142 and 143. Fed. Reg. 50:47142-48933; 1985.

Proceedings: Swedjemark, G.A.; M.jönes, L. Exposure of the Swedish population to radon daughters. Berglund, B.; Lindvall, T.; Sundell, J., eds. Proc. 3rd international conference on indoor air quality and climate. Vol. 2. Stockholm: Swedish Council for Building Research; 1984:37-43.

Reprints can be ordered on a Reprint Order Form which will accompany proofs. Unless instructions to the contrary have been received, only the first named author will receive this order form with proofs of the article. The author filling in the order form should make sure that all co-authors' needs for reprints are incorporated in the total order. Reprint order forms must be received before printing in order to qualify for lower prepublication rates. The original manuscript and diagrams will be discarded one month after publication unless the publisher is requested to return original material to the author.

It is a condition of publication that manuscripts submitted to this journal have not been published and will not be simultaneously submitted or published elsewhere. All authors must sign the Transfer of Copyright agreement, available from the editors, before the article can be published. (U.S. and U.K. government officers or employees whose submitted work was prepared as part of their employment are exempt from the transfer requirement, but must certify their status as government writers.) This transfer agreement enables Pergamon Press to protect the copyrighted material for the authors, but does not relinquish the authors' proprietary rights or rights to use their work as they please in the future. The copyright transfer covers the exclusive rights to reproduce and distribute the article, including reprints, photographic reproductions, microform, or any other reproductions of similar nature and translations.

ENVIRONMENT INTERNATIONAL

CONTENTS Volume 19 Number 5 19

SPECIAL ISSUE

HISTORY OF RADIUM, URANIUM, THORIUM, AND RELATED NUCLIDES IN INDUSTRY AND MEDICINE

INDEXED IN Current Contents, BIOSIS Database, Eng. Ind. Monthly and Author Ind., Environ. Per. Bibl., CABS, Energy Re Abstr., Energy Data Base, Excerpta Medica, Geo. Abstr., Sci. Cit. Ind., and Biology and Environmental Sciences

ISSN 0160-41 (3

0160-4120(1993)19:05;1-L