A CENTURY'S CHALLENGES
HISTORICAL OVERVIEW OF RADIATION SOURCES IN THE USA

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The failure to properly control, account for and dispose of radioactive sources and devices can lead to their entering the public domain in an uncontrolled manner. Once there, lost and unwanted sources can cause safety problems such as radiation exposures of the public and radioactive contamination. The prevention of radioactive sources from entering the public domain in an uncontrolled manner has become an international challenge to authorities responsible for regulating the safe use and disposal of radioactive sources. The problem, however, has historical antecedents dating to the earliest days of radium usage in the 20th century.

EARLY USE OF RADIUM IN THE UNITED STATES

The use of radium sources in the United States predates the US Atomic Energy Act, as amended. Further, radium sources are not covered by the Act and, thus, are not subject to regulation by the United States Nuclear Regulatory Commission (NRC). The potential medical benefits of radium were recognized shortly after its discovery in 1898 and resulted in a demand for radium sources. Information on the extent of early usage of radium in the USA is scant but that which is available indicates a slow growth in usage up to World War II when it increased dramatically followed by a gradual decline. *(See graph, page 50.)* In 1921, between 35 and 40 grams of radium were in use in the US while the number of medical users was between 400 and 500. In 1932, the US Bureau of Mines estimated that there were 710 medical radium users in the US using 124.7 grams. Usage of radium had expanded during World War II, mainly as a result of its use for industrial radiography when 200 grams of radium were used for this purpose. Another 190 grams were consumed during the war in the manufacture of radium luminous paint.

In 1964, the United States Public Health Service (PHS) concluded that radium usage had probably peaked in the immediate post-World War II years and estimated that there were 4500 radium users in the US using between 300 and 700 grams of radium as identifiable sources. The majority of these, 3500, were medical users. Thereafter, usage declined mainly as a result of other radioactive materials becoming available and the increased regulatory oversight of radium by the states, which caused many radium users to discontinue its use.

In 1975, there were 3600 radium users in the US. The number of radium users is certainly less today. Even at its probable peak after World War II of 5000 to 6000 users, this population is much smaller than the number of US licensees now using byproduct, source and special nuclear materials -- an estimated 22,000 specific licensees and 135,000 general licensees.

The extraction of radium from ores was difficult and in the early part of the century was expensive. In 1923, radium cost US $120,000 per gram. Thus, when radium sources were lost or stolen, avoiding the cost of replacing the sources became a strong

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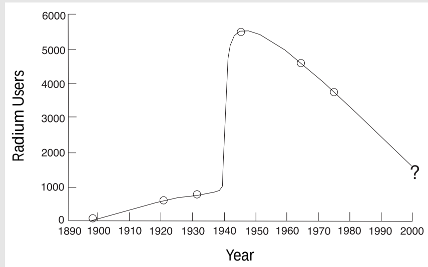
incentive to search for and recover the radium.

RADIUM INCIDENTS

In 1968, the PHS published a summary of known radium incidents in the US based upon a review of the literature and the *New York Times* for the period 1913 to 1964. A total of 396 incidents was tabulated which included 261 losses and 25 thefts. The remaining incidents involved contamination, overexposures and miscellaneous events. The vast majority of the 396 incidents, 331 or 84%, involved medical sources. The recovery rates were 71% (170 of 240 cases) for lost medical sources, 53% (9 of 17) for lost non-medical sources, 60% (15 of 25) for thefts and 50% (2 of 4) for transportation losses. The earliest incidents for which dates are known occurred in the 1911-20 period, totaling nine losses. Losses and thefts steadily increased, peaking in the 1961-67 time frame. The PHS felt that this increase reflected the rising use of radium up to the 1950s and the greater availability of reports of incidents within the most recent years covered by the survey. The greater availability of reports probably reflected compliance with newly issued state requirements for reporting of losses and thefts of radioactive materials not covered by the US Atomic Energy Act, as amended.

Many radium incidents, however, probably escaped public scrutiny. For example, consultants acting in their private capacity to search for lost or stolen radium sources often did not publish or report their work publicly. In the case of state radiation control programmes -- which responded when losses and thefts of radium sources occurred and to requests for assistance in disposing of unwanted radium -- their written reports frequently went straight to the files. Fortunately, some individuals involved with searches for lost and radium sources left public records of their experiences.

RADIUM TRENDS



The use of radium in the United States grew slowly until the advent of World War II, when it increased dramatically. After the war, use declined and is believed to continue to decline.

In 1914, after graduation from Purdue University, Arthur L. Miller accepted an offer to work for the Standard Chemical Company, in Pittsburgh, Pennsylvania, which was then the largest producer of radium. There, he specialized in calibrating radium sources using an electroscope. Since he was familiar with the operation of electroscopes, he was frequently called upon to search for lost radium sources using that instrument. In 1923, he wrote about seven cases. His most intriguing story involved the unsuccessful recovery of 150 milligrams of radium lost by a hospital. As often was the case, the radium found its way to the hospital's coal-fired incinerator where Miller found evidence of contamination but not the ashes which would have contained the radium sources. Upon inquiry, he learned that incinerator ashes were sold to a nearby contractor who used them as aggregate when making concrete that was poured to make a sidewalk. Miller found the sidewalk and confirmed that the radium was embedded in it. Since the radium could not be easily recovered, the sidewalk was left in place and the search was terminated. Miller, unfortunately, did not say where that sidewalk was poured. At the time, the radiation hazards from the embedded radium were not considered. This case was later investigated by another radium searcher, Robert B. Taft, who contacted the insurance company that covered the hospital's loss but found that the company's records had since been destroyed. So, somewhere, probably in the eastern United States, there is, or was, a sidewalk in which is embedded 150 milligrams of radium.

Taft was a physician who frequently was called upon to search for lost radium. He initially wrote of his experiences, which began in 1933, in a paper presented to the American Roentgen Ray Society in 1935. He subsequently recounted his experiences in a book, *Radium - Lost and Found.* Taft's tools for searching for radium included willemite ore (which scintillated when exposed to radiation), electroscopes and early GM detectors. Taft reported 187 incidents, some of which he was personally involved in, and others which were reported to him. Most involved lost or stolen radium sources but some also involved contamination.

A number of cases involved lost medical radium sources that became mixed with hospital wastes that were disposed of at land disposal sites. A frequent practice of the time was to raise swine at these sites. Taft reported that on one occasion when searchers visited such a site to find a lost radium source, they found indications from their electroscope that the radium was nearby but could not pinpoint it. They noticed that a swine herd had walked by. The herd was captured and they confirmed that one pig was radioactive. It was slaughtered and the radium source was recovered.

In Philadelphia, Frank Hartman, a radium sales representative, left a written record in the form of personal notes of his searches for lost and stolen sources. Hartman's notes cover 120 cases from 1930 to 1958. Like Taft, he used willemite ore as well as ZnS, electroscopes and GM detectors. The 120 cases represented a total of 4.259 grams of lost or stolen radium. Of this, he was able to recover 3.806 grams or 89%, an amazing percentage considering the primitive nature of his radiation detection devices and a tribute to his thoroughness and tenacity. Also amazing were his "repeat customers," one of whom lost radium on eight different occasions!

Another category of incidents involved transportation. An intriguing example is the manner in which Standard Chemical Company transferred partially refined radium from its plant in Canonsburg, south of Pittsburgh, to its laboratory in Pittsburgh for final refinement. This was accomplished by carrying the radium on passenger trams operating between the two cities. In 1959, Miller provided details on this practice. The radium was packaged in corked glass bottles that were placed into bailed galvanized steel cans. These were carried by two messengers riding the trams to the Pittsburgh facility. Miller's account implied that one of the two messengers regularly made this trip, an individual by the name of "Tommie" Thomas who was also the head of the department in Canonsburg that performed the initial fractional crystallization of the radium from the chloride solutions. Nothing is mentioned about protective shielding and probably there was none. As much as "a couple hundred mg" were carried at one time. Based upon known tram transit times between the two sites, the annual dose to Thompson from this activity alone could have been as much as 1 Sv in Standard Chemical Company's peak production year, 1920, when 18.5 grams were produced. Nearby passengers and operating crews, of course, would also have been exposed.

Another incident, this involving the US Post Office, was reported by the *Associated Press* in 1921. In this case, a patient being treated with radium on an outpatient basis misunderstood the directions given to him and returned home with the radium still applied to him. At home, he removed the radium and put it away. The physician then advertised for the US $3500 source and the patient, upon reading the notice, placed the source in an envelope and returned it by postal service. Based upon the then current cost of US $120,000 per gram, the quantity of radium thus mailed was about 29 milligrams.

CONTAMINATED GOLD JEWELRY

The metal recycling industries are currently faced with the challenge of preventing radioactive sources which are lost, stolen or improperly disposed of from becoming mixed with metal scrap, or failing that, detecting the sources before the scrap metal is processed or melted to make new products. Interestingly, this problem has historical antecedents dating as early as 1910.

Seeds containing radon were developed as an alternative to the use of radium sources for medical implants. The most common technique involved pumping radon generated from a radium salt solution into a thin gold tube that was then cut and sealed into short segments (seeds). After calibrations, the seeds were shipped to hospitals and clinics for implantation.

Compared to radium, the radon seed technology was more versatile and, because of the radiation characteristics of the radon daughters, the seeds could be implanted permanently.

Lacking the tissue imaging technologies available today, therapists had to make their best estimate of the size of the tumor to determine the number of seeds that were needed. Since estimates of the tumor volume were normally on the high side, some of the ordered seeds were often unused. Excess seeds could be returned to the supplier for credit but some physicians kept the seeds and later sold them to gold recyclers. When melted, the metallic radon daughters, lead-210, bismuth-210, and polonium-210 (or Ra DEF in the radium decay chain nomenclature) became intermixed with the gold. Jewelry made from such gold became a source of radiation exposure especially if the jewelry was worn close to the skin. By the 1960s, reports of radiation injuries from wearing of such jewelry appeared in the literature. In 1981, the New York State Department of Health mounted a special campaign to find such jewelry and remove it from circulation. About 160,000 items were screened resulting in the collection of 133 radioactive items and the identification of another 22 pieces whose owners declined to surrender them. Most of the items were made or acquired in the 1930s and 1940s but one item, a plain gold ring, dated to 1910.

The last US radon generating plant was operated by Radium Chemical Company at its Queens, New York, site using apparatus deigned by Gioacchino Failla. It ceased operation in 1981, thus ending the possibility of new radon seeds entering the gold recycling stream. However, in 1982, when Radium Chemical Company was ordered to inventory its depleted gold seeds, it could not account for them and there was no anecdotal evidence of their showing up anywhere. One cannot help but speculate that the inventory had been disposed of in the gold recycling market.

The foregoing underscores the point that the known data on losses, thefts and unwanted or improperly disposed of radium sources are but the proverbial tip of the iceberg. The true picture will never be known.

US GOVERNMENT OVERSIGHT

Although information on losses, thefts and other safety problems with radium was fragmentary, there were sufficient reports in the literature to raise public and legislative concerns which lead to government oversight of radium users. By the 1960s, many states were developing, or had developed, regulatory control programmes for radium. The PHS provided direct assistance to the states in the forms of monetary grants and loans of personnel to develop their radiation control programmes.

By this time, many radium sources were no longer wanted and their owners could not, or were unwilling to, pay for disposal. Unwanted radium sources were found stored in unexpected places such as bank vaults. In response, the PHS began a radium disposal project in 1965 under which persons having unwanted radium could transfer the sources to the PHS. In most cases, state radiation control programme inspectors acted as transfer agents who shipped the sources to the Southeastern Regional Radiological Health Laboratory in Montgomery, Alabama, where they were stored. This laboratory, originally operated by the United States Food and Drug Administration Bureau of Radiological Health, is now a facility of the United States Environmental Protection Agency (EPA). In 1983, the accumulated inventory of 140 grams of radium was transferred to and disposed at the Hanford, Washington, low-level radioactive waste disposal site.

Subsequently, other large amounts of radium were disposed of elsewhere. In 1989, 120 grams of radium were removed from the former Radium Chemical Co. plant site in Queens, New York, and disposed of at the Beatty, Nevada, low-level radioactive waste disposal site.

In the 1990s, several states mounted campaigns to locate, recover and dispose of radium sources. A total of 4.2 grams was collected and disposed of by Oklahoma and Ohio. The Conference of Radiation Control Program Directors estimates that radium disposals amounted to 12 grams per year in the 1970s, ten grams per year in the 1980s and eight grams per year in the 1990s.

THE AEC'S GENERAL LICENSE PROGRAMME

In 1958, about the same time the PHS began assisting states in developing regulatory initiatives to improve control,

Photo: Lost or abandoned radiation sources have been found in scrap shipments, thus entering the public domain without regulatory control. Steps are being taken by US industry and government to prevent problems. *(Credit: NRC)*

accountability and disposal of radium sources, staff of another Federal agency, the United States Atomic Energy Commission (AEC), proposed extending the general license concept to "measuring, gauging and controlling devices" containing radioactive materials covered by the Atomic Energy Act of 1954, as amended. AEC staff noted that "[a]bout 1000 users would be affected."

This proposed change was approved in 1959 by the Commission. Ironically, the rule change eventually led to control, accountability and disposal problems for this population of radioactive sources that, in retrospect, are similar to those that were found with radium sources.

The general license concept enables persons with minimal training in radiation safety to possess and use licensed devices with minimal risk to the users or to the public while the devices are in use. Robust design and manufacturing criteria applied to the devices enable this unique approach.

Persons using such devices do not need to apply for a specific license but possess and use the devices under the general license and its conditions which are provided in the regulations. Inherent in the concept was the notion that general licensees will exert appropriate control and accountability of the devices while they possess them and will properly dispose of them when they are no longer needed.

Because the requirements for robust design of general licensed devices provide assurance that they can be used safely, there is no routine inspection programme or other regulatory mechanism to contact most general licensees periodically. Most general licensees are exempted from user fees. As a result, most of the members of this group of licensees, presently consisting of about 135,000 using 1,800,000 devices, rarely have contact with the regulatory agencies.

In the absence of such contacts, some general licensees' programmes to control, account for and properly dispose of the devices deteriorate. As time passes, warning labels and signs on generally licensed devices often became obliterated as a result of exposure to adverse environments and improper maintenance. Also, personnel knowledgeable about the devices retire, are discharged or otherwise leave the licensee's plant.

The predictable consequence of these developments is that generally licensed sources are entering the public domain in an uncontrolled manner, most frequently by being discarded with scrap metal. Specifically licensed devices are also mistakenly discarded with metal scrap. However, the number of devices under specific licenses is smaller and their users are subject to routine regulatory contacts as a result of fee charges and routine inspections.

The similarity of these general licensees to the pre-1960s radium users is this: Neither group was universally subject to periodic contacts by regulators to remind them of the need to maintain control and accountability of their sources, to properly dispose of them when no longer needed and to use them safely.

A significant difference, however, is the size of the two populations. As noted, the number of radium users probably peaked in the 1950s at about 5000 to 6000 users. This was a fraction of the total US general licensee population using radioactive devices, which grew from 1000 in 1958 to 135,000 forty years later.

As early as 1981, the states expressed concern to the NRC about the general license programme. In 1986, an outside panel of experts that reviewed the NRC licensing and inspection programme for fuel cycle and radioactive materials facilities recommended that the NRC give higher priority to an ongoing review of general license policies and procedures because of problems with devices being abandoned, disposed of in unauthorized ways, malfunctions and lack of accountability.

In the 1990s, the scrap metal recycling industries expressed concern as well, reflecting their experiences with licensed radioactive sources and devices becoming mixed with scrap metals destined for recycling, and developed informational and guidance references.

A 1996 report by a joint Working Group of the NRC and its agreement states expressed similar concerns and recommended changes in the NRC general license programme.

The Working Group also discussed another problem, "orphan sources." These are sources or radioactive devices that are found in the public domain, most often by metal recyclers. When these are reported, the finders are often asked to take control of and secure the source or device temporarily, thus removing the potential hazard to the public. This is done because provisions to accept or arrange transfer of licensed radioactive material are not generally available to the regulatory agencies unless there is an immediate threat to the public health and safety.

If the owner of the source or its manufacturer can be identified, arrangements are usually made to return the source or pay for its disposal. On the other hand, if the owner or the manufacturer cannot be identified or is no longer in existence, the source is considered to be an "orphan source" and the unlucky finder may be held responsible for long-term security and eventual disposal of the unwanted source. Obviously, this is unfair and probably serves as a disincentive to some persons to report discoveries of radioactive sources. The Working Group recommended that this problem be addressed.

In 1998, forty years after the expansion of the AEC general license programme, the Commission directed NRC staff to make changes in the general license programme. The purpose was to improve control and accountability of general licensed devices and take steps to assure proper disposal of unwanted licensed sources.

Additionally, the states -- through the Conference of Radiation Control Program Directors with support from the EPA and NRC -- established a committee on unwanted radioactive materials which will attempt to tackle the problem of orphan sources.

APPLYING THE LESSONS LEARNED

In conclusion, an important lesson to be learned from the operational experience with radium users is that periodic contacts by regulators with users of radioactive sources serve as reminders to them of the need to maintain control and countability of the sources, to properly dispose of the sources when they are no longer needed, and to otherwise provide for their safe use.

This lesson has been reinforced by the experience following the rule change by the AEC to extend the general license programme to include users of radioactive devices. Again, the lack of periodic contacts by regulators led to control, accountability and disposal problems for this group of users. Periodic contacts by regulators with users of radioactive materials are an essential element of a regulatory programme.

Given this historical perspective, perhaps another lesson in this is that when dealing with radiological protection issues, we should not ignore the knowledge learned from past experiences. Otherwise, as George Santayana wrote, "Those who cannot remember the past are condemned to repeat it". ❑