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Progress Report on "Evaluation of Public Health
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Period Ending July 31, 1974

INTRODUCTION

The history of radioactive self luminous materials (RSLM) goes back to the discovery of radioactivity. The first reported RSLM seems to have been prepared by Giesel at the turn of the century (Curie 1903). He used a mixture of radium and a platino-cyanide compound. The material had a short useful life and was only of theoretical interest. The availability of zinc sulfide made it possible to produce RSLM in such large quantities that Berndt (1921) estimated that over 20g of radium were lost during the first World War as a result of loss of planes and other war equipment and materials.

Instrument panels, compasses, time pieces, and many exotic devices such as rings, door knobs, locks, and similar objects were painted with RSLM to improve their appearance and practicality. It was not until the pioneering work of Martland (1925) that the hazards of wide-scale application of radium were recognized. Even though these studies were available prior to World War II, the requirements of war necessitated a large scale application of radium in similar applications as those in World War I.

The availability of other radionuclides, subsequent to World War II, made it possible to consider radium substitutes. In a study conducted by the IAEA, tritium and promethium 147 along with radium 226 were recommended. Unfortunately, at the time of that study, little was known about the occupational and possible health implications of these three radionuclides on a comparative basis. The enclosed bibliography contains relevant references on the subject of RSLM. It is expected that this list will be revised and improved prior to the completion of the project.

DHS-22

TRITIUM GAS TUBES

Elemental tritium is used in the so-called gas filled tubes generally consisting of a capillary tube coated internally with zinc sulfide and filled with elemental tritium at pressures of less than half an atmosphere. Application of tritium in elemental form is potentially desirable and safe, provided certain requirements are met. As the biological absorption of elemental tritium is by a factor of 10,000 less than that of tritiated water, it is imperative to maintain a low water level in the container.

During the production process, separation of water from elemental hydrogen does not pose any significant problem. However, subsequent to the introduction of tritium into the tube, residual moisture as well as dissolved oxygen, or oxygen containing materials in the crystal and the glass wall, lead to production of tritiated water by oxidation or hydrogen exchange. Because carrier free tritium is used, minute quantities of oxygen or water may lead to significant quantities of tritiated water.

The actual measurements in regard to water content of gas filled tubes are scarce and contradictory. The values range from a few percent up to 50 percent. These contradictions may be based on inaccurate measurement techniques or reflect differences in production methods. Due to the fundamental importance of the water content of these tubes, it is desirable to develop standard methods for measurement of water, establish standards of performance, and enforce a quality control program for gas filled tubes. The major exposure from the gas filled tubes would occur during the breakage of the tube. An improper production technique could inevitably lead to the loss of the tritium and thus the product luminosity. Therefore, the economic requirements should lead to a management of the radioactive material in accordance with proper health physics practices.

Tritium release from gas filled tubes depends upon a number of factors. Niemeyer (1970) measured values corresponding to the diffusion rate of hydrogen through glass walls. However, values which exceed those produced by the diffusion rate have been measured indicating errors in the production process. Here again, the chemical form would be of significance because of the differences in biological absorption of the two chemical forms.

The entire field of gas filled tubes requires a careful study because of the apparent interest of the industry in this area and because of potential attractiveness and use of these tubes. Because of substantially larger quantities of radioactivity in these tubes, it is necessary that sufficient information is available before they enter the market. It is interesting to note that a committee of the European Nuclear Agency has recommended an exemption of gas filled tubes for a tritium content not to exceed 200mCi. No specified chemical requirements are associated with that exemption.

TRITIATED POLYMERS

The common application of tritium in RSLM is in the polymer form. Potentially all polymers containing hydrogen are available for this application. In practice, one is limited to reactions which permit an easy hydrogenation during the process. Polystyrene and silicone rubber are commonly used compounds. Although the final product may be the same, the mode of production has a significant influence on the stability of the tritium label. Many other compounds have been proposed for application in RSLM.

There is sufficient evidence to make at least a semiquantitative risk-benefit estimate for tritiated luminous paints. The risk consists of radiation dose to the producers of paint, dial painters, workers who assemble the watch

(or other devices), wholesalers, retailers, and the consumer.

Due to the significant quantities of tritium involved in preparation of RSLM, health physics rules are usually observed in that operation and licensing requirements are generally met by routine radiobioassay. Therefore, the man-rem/Ci of tritium used is small when compared to other operations.

The largest dose is received by the dial painters who mix the paint and apply it to the dials and hands. Table 1 indicates results from several dial painting operations. Because of the present dispute over the quality factor of tritium, all doses are expressed in rad. The values for rad/g.L are calculated based on the assumption that 0.2Ci of tritium is used to produce 1g of ZnS paint which if painted over a cm^2 surface would result in a luminosity of 40mL. Although the averaging of all values would have to include the number of individuals involved, in the absence of the necessary information the calculated average is indicative of the risk. It is of interest to note that the risk deviates by more than one order of magnitude among various plants.

No information is available on the radiation dose to workers during the assembling of watches. Efforts are underway to obtain this information in cooperation with the Texas Department of Public Health.

Bradley et al. (1971) evaluated several storage facilities where tritiated watches were kept. They measured tritium urine levels as high as 28.8mCi/l. The average air concentration divided by the total quantity of tritium on hand was 0.02 nCi/1.Ci. It is difficult to obtain man-rem data without some knowledge of the number of employees involved in the operation; Bradley et al. indicated one or two employees at each storage facility. If two employees are used for calculation of dose, the value for the dose would be 23 mrad/Ci. This value is comparable with the average of 18 mrad/Ci obtained for dial painting plants,

indicating the importance of surveillance in storage facilities. Caution should be exercised in using the value of 23 mrad/Ci because of the insufficient number of air data and several other unknown factors.

Tritium measurements in the retail stores are not known. It is probable that air concentrations of tritium in those facilities are small because of the rapid air change in summer and winter in the majority of retail stores.

Tritium release measurements from tritiated paint, although numerous (see bibliography), are not easily relatable to the body burden of users. Fitzsimons et al. (1972) report radiation doses of 0.1 to 1 mrad per year with an average of 0.35 mrad. The activity of the watch was unknown in their study.

Unpublished data from Moghissi et al. (1972) indicate radiation doses of 30 mrad/Ci. This corresponds to 0.75 mrad for a watch containing 25 mCi which is the maximum exempt quantity of tritium on a watch. If the values of Fitzsimons et al. are used to calculate the tritium content of watches, a value of 10-15 mCi/watch would be obtained. Bradley et al. report values ranging from less than 1 mCi to 5.6 mCi per watch, although they do not identify the source of their information.

For the purpose of dose estimation, the value of 10 mCi/watch is used which is probably the upper limit of an "average" watch. This value corresponds to about 0.3 mrad/year. It is estimated that 8×10^6 watches painted with tritium are imported annually to the U. S. and that 16×10^6 watches containing tritium are in use. If the 0.3 mrad/watch is used, this number corresponds to 4,800 man-rad to the user, corresponding to 1.6×10^5 Ci. The total known risk as a result of application of tritium is, therefore, as follows:

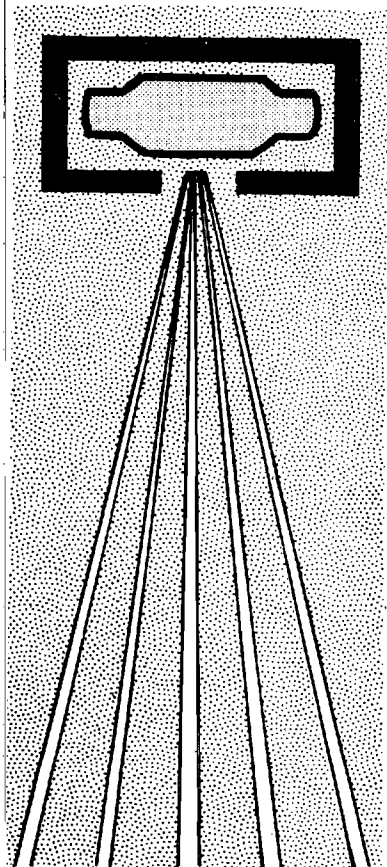
Painting:	1.6×10^5 Ci x 18 mrad/Ci	= 2.9 krad
Storage:	1.6×10^5 Ci x 23 mrad/Ci	= 3.7 krad
Usage:	1.6×10^7 watches x 0.3 mrad/watch	= <u>4.8 krad</u>

TOTAL 11.4 krad

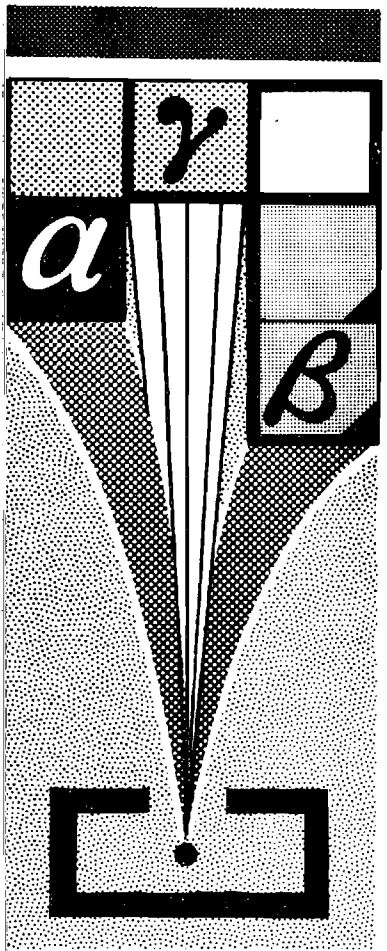
TABLE 1

<u>Location of Plants</u>	<u>mrad/Ci</u>	<u>rad/g.L</u>	<u>Reference</u>
U. S. A.	19.1	95.5	Moghissi <u>et al.</u> (1972)
Switzerland	1.4	7.0	Krejci (1972)
Switzerland	5.3	26.5	Krejci (1972)
Switzerland	5.4	27.0	Krejci (1972)
Switzerland	5.9	29.5	Krejci (1972)
Switzerland	1.1	5.5	Krejci (1972)
Switzerland	12.0	60.0	Krejci (1972)
Switzerland	21.8	109.0	Krejci (1972)

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PUBLIC HEALTH IMPLICATIONS of RADIOLUMINOUS MATERIALS



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 FDA 73-8027 Course Manual for X-Ray Measurements (GPO 1715-00047, \$5.10).
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 FDA 73-8029 Health Physics in the Healing Arts - Seventh Midyear Topical Symposium (GPO 1715-00055, \$5.40), (PB 228 791/AS, mf only).
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 FDA 73-8036 The Chest X Ray as a Screening Procedure for Cardiopulmonary Disease - A Policy Statement (GPO 1715-00051, \$0.50) (PB 220 139, mf only).
 FDA 73-8038 Bureau of Radiological Health Research Grants Program, Fiscal Year 1972 (GPO 1715-00054, \$2.60) (PB 222 600, mf only).
 FDA 73-8039 Survey of Use and Performance of Ultrasonic Therapy Equipment in Pinellas County, Florida (GPO 1715-00056, \$0.90) (PB 223 660/AS, mf only).
 FDA 73-8047 Population Exposure to X Rays, U.S. 1970 (GPO 1715-00068, \$2.15) (PB 233 569/AS, mf only).
 FDA 74-8001 A Simple and Inexpensive Breathing System for Xenon-133 Ventilation Studies.
 FDA 74-8002 Quality Control in Medical X Ray (GPO 1715-00057, \$0.65) (PB 224 082/AS, mf only).
 FDA 74-8004 Report of State and Local Radiological Health Programs - Fiscal Year 1972 (GPO 1715-00059, \$1.10) (PB 228 716/AS, mf only).
 FDA 74-8006 Physics of Diagnostic Radiology - Proceedings of a Summer School (GPO 1715-00062, \$4.85) (PB 227 844/AS, mf only).
 FDA 74-8007 A Review of Determinations of Radiation Dose to the Active Bone Marrow from Diagnostic X-Ray Examinations (GPO 1712-00192, \$0.55) (PB 227 840/AS, mf only).
 FDA 74-8008 Fifth Annual National Conference on Radiation Control - Planning for Protection (GPO 1715-00061, \$3.25) (PB 228 972/AS, mf only).
 FDA 74-8009 A Fixed Format Lexicon for Nuclear Medicine Reports (GPO 1715-00060, \$1.10) (PB 227 782/AS), mf only).
 FDA 74-8010 A Review of Selected Bioeffects Thresholds for Various Spectral Ranges of Light (GPO 1715-00063, \$1.25) (PB 228 391/AS, mf only).
 FDA 74-8012 Scientific Seminars of Nuclear Medicine Laboratory Dedication (GPO 1715-00064, \$0.95) (PB 228 664/AS, mf only).
 FDA 74-8013 Method for Measurement of Frequency, Modulation Characteristics, and Timer Accuracy of Ultrasonic Therapy Equipment.
 FDA 74-8014 Status and Future Manpower Needs of Physicists in Medicine in the United States (GPO 1712-00198, \$0.95) (PB 228 383/AS, mf only).
 FDA 74-8016 A Limited Survey of Health-Related Laser Characteristics for Selected Applications in the United States (GPO 1715-00065, \$0.95) (PB 230 701, mf only).

**PUBLIC HEALTH
IMPLICATIONS of
RADIOLUMINOUS MATERIALS**

Prepared by:

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Under

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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
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Rockville, Maryland 20852

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FOREWORD

The Bureau of Radiological Health conducts a national program to limit man's exposure to ionizing and nonionizing radiations. To this end, the Bureau (1) develops criteria and recommends standards for safe limits of radiation exposure, (2) develops methods and techniques for controlling radiation exposure, (3) plans and conducts research to determine health effects of radiation exposure, (4) provides technical assistance to agencies responsible for radiological health control programs, and (5) conducts an electronic product radiation control program to protect the public health and safety.

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Readers are encouraged to report errors or omissions to the Bureau. Your comments or requests for further information are also solicited.

John C. Villforth
John C. Villforth
Director
Bureau of Radiological Health

PREFACE

This report contains a compilation of information relating to radioluminous materials and the impact of such radioactive materials on the public health.

Principally, the report deals with the use of radium, tritium, and promethium as the phosphor activator of the paint in the dial painting industry with particular emphasis on timepieces. It is noteworthy that radium (the most hazardous of these three radionuclides) is no longer used in the United States for the painting of watch dials. However, radium is still employed extensively on clock dials and other luminous instruments. Because of the useless and hazardous gamma emissions from radium, its role as a luminizer has been replaced largely by tritium and, to some extent, promethium.

It is intended to use the information derived from this contract (FDA 233-74-6093), as well as other information from similar studies, to prepare guidelines relating to the manufacture and use of radioluminous consumer products.



Peter Paras
Director
Division of Radioactive Materials
and Nuclear Medicine
Bureau of Radiological Health

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ABSTRACT

This study evaluates the safety, efficacy, and relative merits of commonly used radioluminous materials. A comparative risk benefit analysis is carried out for radium-226, promethium-147, and tritium. These materials have been used most extensively in the dial painting industry for the illumination of timepieces and other instruments where self illumination offered an advantage.

In general, occupational exposure to radium is higher than that to tritium. Occupational exposure to promethium-147 cannot be measured with any degree of accuracy and thus remains essentially unknown. The addition of tracer quantities of promethium-146 to promethium-147 should be given serious consideration to make a body burden measurement possible.

More recently tritium and to some extent promethium have replaced radium as the light activator in phosphors. Although only a few radium watches have been sold in the U. S. in the past 3 years, radium continues to be used in clocks. This is reflected by the 1973 estimates of the population dose--3,600 person-rem from tritium absorption, 2,500 person-rem from radium exposure, and an unknown commitment from promethium. These estimates are derived from the average activities per timepiece-- 5 mCi of tritium for 24 million watches and 0.5 μ Ci of radium for 8.4 million clocks. The dose from radium would have been about 10 times greater had radium been used in watches rather than in clocks. No data are available on the level of promethium activity used in timepieces. Similarly, only limited information is available on the public health aspects of tubes filled with tritium gas. Assuming a reasonable minimum dose of 6,100 person-rem from all these nuclides, a minimum average exposure of 0.03 mrem/year is estimated to the U. S. population.

The study concludes that radium should not be used for dial painting because tritium can be used to accomplish the same results with substantially less exposure. Before a large-scale application of promethium-147, methods must be developed to determine the body burden of workers as a result of occupational exposure.

ACKNOWLEDGMENTS

This report summarizes various studies carried out by the authors during the last 10 years. During this period, primarily in the last few years, we were assisted by numerous individuals and organizations. We would like to express our gratitude to all who helped us in this endeavor, especially the following individuals: R.F. Barker, R. Bischoff, H.W. Dooley, W. Holm, P.B. Jenkins, S. Kinsman, K. Krejci, B.E. Lambert, L.A. Leiker, R.C. McMillan, D. Moeller, K.Z. Morgan, M. Riley, G.D. Schmidt, W. Seelentag, R.E. Simpson, A.C. Tapert, O.W. Thueler, A.T. Vess, J.C. Villforth, C.W. Wallhausen, R.N. Waltz, M.C. Wukasz, and A. Zeller.

Particular appreciation is expressed to R.N. Waltz for providing us with the information contained in table 8. We would also like to express our appreciation to the library staff of the Georgia Institute of Technology for their assistance in preparing this report.

PUBLIC HEALTH IMPLICATIONS OF RADIOLUMINOUS MATERIALS

1. INTRODUCTION

1.1 HISTORICAL OVERVIEW

The phenomenon of luminescence has been known for many centuries. Becquerel (21), who discovered radioactivity, was in actuality studying luminescence which led to the discovery of radioactivity. The discovery of radium by M. Curie and P. Curie (50) at the turn of the century and the availability of radium-226, polonium-210, and a number of other naturally occurring radionuclides led to many applications including the preparation of radioluminous compounds.

It is a historical irony that the most efficient phosphor was one of the first ones to be synthesized. In the last century, Sidot (215) prepared zinc sulfide by reacting zinc chloride with hydrogen sulfide. He was not quite sure why some batches resulted in very efficient phosphors whereas others resulted in poor crystals. Gruene (83) in 1904 described the necessity of the presence of small quantities of trace elements such as copper and silver in zinc sulfide to obtain an efficient phosphor. It is a paradox that many years later these studies were repeated in this country with identical results (161) which amazed the investigators.

The first reported radioluminous material was prepared by Giesel (78) by coprecipitating radium with one of the then well-known luminous materials, namely barium hexacyanoplatinate. It was also Giesel (79) who showed the great potential of zinc sulfide as a detector for alpha-emitting radionuclides. The early history of radioluminous materials is cloudy and, as expected, associated with claims and counter-claims. Kunz claimed to have patented any combination of radioactive materials and phosphors, and a patent was granted to him in 1903. This claim was disputed by Viol and Kammer who claimed a patent of their own (238). It is surprising that the economic potential of radioluminescence was recognized so early. The literature on preparation techniques, properties, and beneficial uses of radioluminous materials is voluminous (2, 4, 6, 15, 18, 25-28, 32, 33, 40, 59, 68, 69, 74, 93, 95, 96, 98, 100, 103, 120, 121, 123, 126, 133, 134, 142, 155-160, 170, 176, 192, 193, 199, 200, 205, 214, 224, 228, 239-246, 252, 258, 260).

As in many other historical events, the First World War brought the actual economic motive to apply radioluminescent materials on a large scale. After the war the requirement of a new market led to the search for possible candidates such as door knobs, locks, light switches, and particularly watches and clocks. The "frivolous" use of radium was often regretted because of the limited supply of radium and its potential usefulness in medicine. Berndt (25), who estimated a loss of 20 g of radium as a result of the loss of planes during World War I,

felt that this loss was probably irreplaceable and similar instances should be avoided in the future.

The literature contains many reports on the "wonder element" and its enormous abilities (3, 5, 7-14, 19, 21, 25, 37, 42, 53, 101, 131, 135, 141, 143, 144, 168, 173, 174, 196, 217, 218, 232, 234, 239, 250, 251). Although early warnings on the radiation effects of radium in humans were expressed by coworkers of C. W. Roentgen and M. S. Curie, it was the tragic cases of dial painters which showed the deleterious effects of radium. Castle et al. (39) seem to have been the first to recognize these dangers. However, Martland et al., in a series of papers (17, 147-152), clearly demonstrated the devastating radiation effects caused by dial painting. The literature on the deleterious effects of radium and related topics is voluminous (23, 34, 35, 39, 77, 99, 102, 132, 136-140, 145, 146, 162, 185, 186, 195, 201-203, 223, 227, 257). A quantitative evaluation of the dose-effect relationship was attempted by Evans (61-66), who also suggested the first maximum permissible body burden for radium which, in effect, was equivalent to the maximum permissible dose.

In more recent times the availability of many potentially useful radionuclides, notably tritium and promethium-147, made it possible to replace radium by these radionuclides. Although strontium-90 was used for a short time, it was quickly abandoned after its occupational problems were recognized. Also, krypton-85 and tritium in elemental form are used as activators for luminous materials. Application of krypton-85 activated light sources has been limited because of its gamma radiation of 0.51 MeV. This external radiation problem is magnified by the production of bremsstrahlung from the 0.69 MeV beta radiation which constitutes a substantial shielding requirement unless the light source is used in remote locations. An example of the application of krypton-85 is radioluminous highway signs in remote areas where electricity may be unavailable.

1.2 REGULATIONS RELATIVE TO RADIOLUMINOUS MATERIALS

In the U.S. there are no Federal regulations limiting radium content of radioluminous materials. The following is a summary of maximum quantities which are permitted according to 10CFR 30, 10CFR 31, and 10CFR 32 (45).

1. Tritium

Air craft safety devices	10 Ci
Time pieces	25 mCi
Hands each	5 mCi
Dial	15 mCi
Automobile lock illuminators	15 mCi
Automobile shift quadrants	25 mCi
Precision balances	1 mCi
Any part of balance	0.5 mCi
Marine compasses (tritium gas)	750 mCi
Other navigational instruments	250 mCi
Thermostat dials and pointers	25 mCi

analysis can be carried out only in exceptional cases. Accidental intake of radium for persons who are otherwise not exposed to radium can be quantitated by urine and feces analysis during the early period following exposure. People who are occupationally exposed to radium cannot be monitored satisfactorily by using radium analysis because their excretion cannot be correlated with body burden.

According to ICRP 10 (107), "At early times after radium enters the blood much of it is in blood and soft tissues from which more than half disappears in the first day. At times beyond this a larger proportion of the radium remaining in the body is in bone where it is retained for long periods, declining with time according to a power function or a series of exponential functions."

Radium decay in the body leads to radon production of which approximately 70 percent is exhaled. This exhaled radon can be used to estimate body burdens because it is in equilibrium with the radium content of the body.

The body burden can also be measured by whole body counting using the remaining 30 percent of the radon and daughters, many of which emit gamma radiation. According to ICRP 10 (107), a body burden of 1 μCi of radium results in a radon concentration of 13 pCi/l in the exhaled air. The corresponding dose commitment to the bone over a 50-year period from an uptake of 1 μCi by blood is 100 rems.

2.2 OCCUPATIONAL EXPOSURE TO RADIUM

As mentioned previously, the first dramatic deleterious effects of internal radiation exposure were observed as a result of radium dial painting. Although the first observation seems to have been reported by Castle et al. in 1925 (39), systematic studies were done by Martland (17, 147-152) who pointed out the essential features of radiation effects of radium and its daughter products if taken orally. They particularly described the necrosis of the jaw which was caused by the specific habit of painters who formed a point on the brush by turning it in their lips. Occupational aspects of radium have been of considerable interest to numerous investigators (1, 23, 31, 34, 36, 60-66, 71, 97, 108, 111, 169, 177, 198, 216, 255, 256). These investigations have led to improvements in health and safety conditions of dial painting operations, although the improvements lag behind those of normal radiochemical laboratories. Evans (61-66), who for many years studied occupational exposures, working conditions, and many other factors related to dial painting operations, suggested many improvements in dial painting operations. Radiological monitoring of the workers has been made possible by the measurement of radon in the breath and whole body counting (82, 90, 204, 207, 208, 222, 225, 230, 231).

At present, as in the past, an estimation of radiation exposure to the workers is difficult because many dial painting operations do not keep records and in some instances are not obligated to monitor the workers. Many studies, however, have been carried out by investigators for short periods of time. All but one of these studies date back to the time of rather lax conditions, and thus such numbers would represent an overestimation of present exposure levels.

Duggan and Godfrey (57) recorded extensive data from the U.K. Their thorough study includes whole body counting and radon in breath measurements, and indicates a high degree of care and accuracy. Table 1 summarizes their data. The notation of these authors has been retained in columns one, three, and five to enable intercomparison between their presentation and the present modification. Only those data which were complete were included; i.e., operations with incomplete data were not considered. The third column is a reproduction of weekly working hours as given by the authors. The fourth column

Table 1. Summary data on occupational exposure to radium according to Duggan and Godfrey (57)

Estab- lishment	No. of workers	Exposure hours per week	Exposure hours per year	Processed radium (mCi/year)	Activity processed (μ Ci/hour)	Average body burden (nCi)	nCi body burden/mCi per year
A	4	30	3375	70	20.7	73 \pm 32	4.2
B	5	40	5625	200	35.6	34 \pm 18	0.9
C	3	40	3375	70	20.7	33 \pm 24	1.4
D	2	25	1406	85	60.5	10 \pm 10	0.2
E	3	40	3375	135	40.0	13.3 \pm 3.3	0.3
F	1	8	225	40	178	20	0.5
G	1	2	56	3	53.6	20	6.7
H	3	10	884	4	4.8	6.7 \pm 6.7	5.0
J	3	30	2531	20	7.9	6.7 \pm 3.3	1.0
K	3	40	3375	104	3.1	3.3 \pm 3.3	0.1
L	1	40	1125	80	71.1	10.0	0.1
M	3	1	84	1.5	17.8	3.3 \pm 3.3	6.6
N	2	2	113	3	26.5	0	0
R	3	20	1688	10	5.9	16.7 \pm 3.3	5.0
U	3	40	3375	30	8.9	3.3 \pm 3.3	0.3
W	5	10	1406	14	10.0	0	0
X	<u>1</u>	15	422	<u>10</u>	23.7	0	0
Total	46			879.5			
Average					35 \pm 10	14.9 \pm 4.4	2.0 \pm 0.6

was calculated from these weekly working hours as given by the authors using 50 weeks/12 months and 9 months/year operation as described by them. In addition, in discussions with many operators we found that the actual exposure during an 8-hour working day was only 6 hours because of coffee breaks, the time the workers use to wash their hands, and other interruptions. Therefore, the exposure time was accordingly reduced. Column five is reproduced from the values reported by Duggan and Godfrey. Column six is the ratio of columns five and four. Column seven (average body burden in nCi) was calculated from the data using values of <10 nCi as zero. Column eight is the result of combining the number of workers, average body burden (nCi), and processed activity (mCi/year). Errors associated with the last three columns are given whenever available.

As described in section 2.1, according to ICRP an uptake of 1 μ Ci of radium to blood corresponds to 100 rem to the bone. Using Duggan and Godfrey's average value of 2.0 nCi body burden/mCi of radium per year, a dose commitment to the bone of approximately 200 person-mrem is calculated.

In addition to the bone dose from radium use, there are two other types of exposure which must be considered; these are external radiation and radon exposure. According to the data of Duggan and Godfrey, presented in table 1, 46 workers processed 879.5 mCi of radium annually. This value of 19.1 mCi per worker is in close agreement with our observation that at present, in a well operated plant, 50 workers process approximately 1 Ci of radium in a year. However, it is our experience that more working hours are now required for painting (the present rate is 14 μ Ci/hour versus a past rate of 35 μ Ci/hour, as reported in table 1) because of the currently more stringent health and safety measures. For the purpose of calculation, a 40-hour week and 48 weeks/year are assumed for each worker, whereas the operation continues for 50 weeks/year. This difference of 2 weeks relates to the annual leave of workers. During this time, the operation continues although selected individuals are absent. Again, it is assumed that the workers are subjected to radiation exposure for only 6 hours per 8-hour day. These assumptions result in $6 \times 5 \times 48 = 1,440$ hours/year exposure.

An estimation of the exposure would require two additional assumptions; namely, the average turnover rate of radium in the plant and the average working distance of workers from radium. If it is assumed that radium paint is processed 2 to 3 days after its arrival, which is roughly equivalent to 15 hours of exposure time (6 hours of exposure per 8-hour day), the average activity "on hand" can be calculated to be 2.5 days/(50 weeks per year \times 5 days per week) = 0.01 of the annual volume of 1 Ci which is 10 mCi. In other words, each worker is exposed to an external radiation of 10 mCi of radium and its radon emanation. The average distance of workers from radium during the 6-hour exposure per 8-hour day is assumed to be 1 m. The external dose rate to the whole body from 1 mCi of open and unshielded radium is 0.84 mrem/hour (180). Therefore, each worker would receive a dose of $10 \times 1,440 \times 0.84 = 12,100$ mrem, and the entire work crew of 50 would receive 600 person-rem in processing 1 Ci of radium.

Another source of exposure is radon. Measurements of this radionuclide in radium dial painting operations are scarce. Again, the only useable data stem from Duggan and Godfrey who report an average of 10 pCi/l with highest values around 80 pCi/l. These values can be verified by the following calculations.

If one assumes that each worker is allocated an average space of $2 \times 2 \times 2.4$ m in a 50-worker plant, the total space requirements would be $2 \times 2 \times 2.4 \times 50 = 480$ m³. In a well ventilated plant, six to eight air changes per hour are common, and thus the total air for dilution would be about 3,000 m³/hour or 50 m³/minute.

Radon accumulation A (mCi) at time t from B (mCi) can be calculated from the following equation:

$$A = B(1 - e^{-0.693t/T_{1/2}})$$

If t is used as 1 minute and $T_{1/2} = 3.84 \text{ days} \times 1,440 \text{ (minutes/day)} = 5,530$ minutes, radon-222 accumulation will be about 125 nCi at the end of the first minute. It is assumed this quantity is being diluted by 50 m³ of air, resulting in a concentration of 2.5 pCi/l. This concentration should be multiplied by approximately three because air pumps generally operate during the 8-hour working day whereas radon is produced continuously.

Considering the many assumptions associated with the above calculation, the value of 10 pCi/l as reported by Duggan and Godfrey seems to be realistic and valid. This value will thus be used for dose estimations.

The dosimetry is based on the work by Harley and Pasternack (91) as used by Carter and Moghissi (38). This model uses Weibel's lung model and assumes an 8 percent unattached RaA with a ratio of Rn:RaA:RaB:RaC of 1:0.7:0.6:0.5. According to this model, a continuous exposure of 168 hours/week of 1 pCi/l of radon corresponds to 1,520 mrem/year to the basal cell nucleus of segmental bronchioles, or 250 mrem for the duration of exposure of 1,440 hours in the plant as described above. The individual exposure would then be 250 mrem x 10 pCi/l = 2.5 rem/pCi/l, whereas the total dose is about 125 person-rem/Ci of radium.

2.3 EXPOSURE TO RADIUM IN THE ASSEMBLY PLANT

No information is available on the worker's exposure in the assembly plant. However, it is conceivable that the exposure will be significant and, in terms of person-rem, comparable to the exposure during the painting operation.

The time requirements for painting a dial are probably comparable to the time necessary to assemble a watch--at least that phase of it which relates to assembling the hands and the face of the watch. The external radiation per Ci of radium therefore will be of the same order of magnitude as that in painting operations. Radon release from the painted materials, however, will be substantially lower than from the dry paint because of the sealing ability of many materials such as those used in paints (22).

For the sake of simplicity, it is assumed that the radiation dose from external radiation for each Ci processed is similar to the external radiation dose during painting. This possible overestimation is offset by the assumption that essentially no radon escapes from the painted materials and thus the radon dose is neglected. The external radiation dose thus would be 600 person-rem to process 1 Ci of radium.

2.4 EXPOSURE TO RADIUM DURING STORAGE

An estimation of the radiation dose during the storage period is even more difficult than during the assembly. Therefore, no estimate is being made. It should be mentioned that if the dose is significant, it probably is primarily as a result of external radiation.

2.5 EXPOSURE TO THE USER

One of the major disadvantages of radium is its inherent emission of a great deal of radiation which is not useful for production of light and yet exposes the users of objects painted with radium.

Radon emissions from watches and clocks have seldom been measured. In the past it was assumed that because of the containment properties of components of paint (22), little radon should escape from the time piece. In addition, because more expensive watches usually are luminous, it was expected to find many of them water resistant, and thus they would contain radon, should it be released from the paint. The fallacy of these assumptions becomes evident if one considers that they are also valid to some extent for tritium. In the case of tritium, the watches are also generally water resistant. Again, all but a few percent of the tritium should be contained in the paint, the fraction released being that which has become solubilized and is uniformly mixed in the lacquer and thus exposed to air. This fraction, however, is too small to account for the continuous releases several years after the painting and, subsequently, the presence of tritium in body water of the users of objects painted with tritiated paint. One would expect, therefore, that certain radiation-chemical mechanisms are involved which decompose the lacquer and crack its surface, enabling the underlying surfaces to be exposed. These mechanisms which cause tritium release could conceivably also lead to radon release from the watches. Radon exposure is important because of the comparatively significant biological effect of alpha radiation.

However, in the following calculation the contribution of alpha radiation is neglected because of the enormous difficulties associated with the exposure estimates and because, as can be seen in the following pages, the whole body exposure by gamma radiation is large enough to make comparative evaluations. Joyet (118, 119), who made comprehensive measurements and dose estimates from radium-containing time pieces, suggests a genetically significant annual dose of 65 to 70 mrem/ μ Ci of radium. These classical measurements were carried out under realistic conditions. Twenty-four human subjects were given a "mock watch" consisting of a plexiglass device containing 100 μ Ci of radium each. Small ionization chambers were placed on specific areas of the body and affixed with adhesive tape. Ionization chambers were placed on the left shoulder at about eye height; others were placed in close proximity of the ovaries or testes. After 3 days of exposure, the ionization chambers were removed and read. From these data Joyet calculated an annual gonadal dose of 70 ± 8 mrem for men and 65 ± 10 mrem for women per μ Ci of radium. Several other studies deal with this subject (58, 73, 154, 175, 188, 213, 226), including those by Haybittle (93) who measured a dose of 0.275 mrem/ μ Ci per hour at the back of a watch. He used a measurement technique similar to that used by Joyet (118, 119). Assuming the watch is worn 24 hours/day, an annual dose of 2.4 rem/ μ Ci to the surface of the wrist can be calculated from his data.

Paul (190), who made a survey of New York storage and import houses, pointed out the importance of pocket watches as a source of increased genetically significant dose to the male population. He made the observation that pocket watches are worn in closer proximity to the gonads than wristwatches and thus the dose would be accordingly higher.

In the absence of measured values, the exposure can be calculated if it is assumed the pocket watch is worn 16 hours/day at a distance of 25 cm with an exposure rate of 0.84 mrad/hour·mCi·m. The dose can be calculated to be:

$$H = 16 \text{ hours} \times 365 \text{ days} \times 0.84 \times 10^{-3} \times (1 \text{ m}/0.25 \text{ m})^2 = 78 \text{ mrem}/\mu\text{Ci per year}$$

This value compares with a gonadal dose of 65 to 70 mrem/ μ Ci as measured by Joyet for wristwatches. The reason for the agreement between these two values is the differences in the exposure time. Joyet assumes 24-hour/day exposure, whereas pocket watches are not worn 24 hours/day. This reduction in exposure time is compensated by the smaller distance between the pocket watch and the gonads as compared to wristwatches.

The dose to the user of an alarm clock can be estimated by assuming an 8-hour/day exposure at a distance of 2 m.

$$H = 8 \text{ hours} \times 365 \text{ days} \times 0.84 \times 10^{-3} \times (1 \text{ m}/2 \text{ m})^2 = 0.6 \text{ mrem}/\mu\text{Ci per year}$$

2.6 EXPOSURE IN THE REPAIR SHOP AND OTHER OPERATIONS

Large numbers of devices that were painted with radium-activated radioluminous paint are available. There are several reports available which treat various aspects of the occupational exposure to workers in aircraft instrument shops (76, 197, 209, 210, 219, 220). These reports indicate that air and surface contamination is significant. Also, external radiation is substantial. It is difficult to make a risk-benefit evaluation because of the lack of availability of the quantity of radium processed by these operations. In addition, it is difficult to assign any benefit to the old radium except that it may have been useful at the time of its preparation.

The exposure to members of certain aircraft crews must have been enormous. Seelentag (210), who made measurements in various planes including the Visc 802, DC-6B, and DC4, reports values up to 0.5 mR/hour for external radiation in the pilot's and copilot's seat. The design of present planes has made the application of radioluminous materials in planes unnecessary except for exit signs which are advantageously prepared with promethium or tritium gas.

3. TRITIUM

3.1 PHYSICAL AND BIOLOGICAL PROPERTIES

Tritium is an isotope of hydrogen. It is radioactive and decays with a half-life of 12.3 years to helium-3 which is stable. The maximum beta energy of tritium is 18 keV, whereas the average beta energy is about 5.7 keV. Because of the significance of tritium, several books and monographs have been published on this subject (60, 67, 72, 105, 166, 171). Introduction of tritiated water into the human body results in an even distribution of tritium approximately 12 hours subsequent to exposure. Therefore, any body water sample is representative of tritium concentration in body water.

Tritium, if introduced as water, is essentially completely absorbed regardless of the mode of introduction; i.e., injected, ingested, inhaled, or taken up by the skin. Elemental tritium (T_2 , HT) is much less absorbed than tritiated water, and thus the body can be subjected to much larger quantities of elemental tritium as compared to tritiated water (10^4 per volume). In addition, elemental tritium dissipates in air much more rapidly than tritiated water (154), and thus for the same quantity of tritium release the relative hazards are even larger than 10^4 . Tritiated compounds are also absorbed by the body. However, the relative absorption depends upon their nature. Absorbed tritium is excreted in accordance with the following equation:

$$A = s e^{-0.693t/T_s} + w e^{-0.693t/T_w} + i e^{-0.693t/T_i} + l e^{-0.693t/T_l}$$

where A represents the total absorbed tritium; T_s , T_w , T_i , and T_1 are biological half-lives of tritium excretion with values of 1 day, 10 days, 30 days, and 1 year, respectively; and s, w, i, and 1 are their respective coefficients.

The physiological reasons for these half-lives are beyond the scope of this report. However, it should be mentioned that the half-life of 1 day represents the turnover rate of the GI tract, and thus unabsorbed tritiated compounds are excreted at this rate. Tritium concentration in body tissues follows a more complicated pattern which, although subject to numerous investigations, is not well understood.

Coefficients s, w, i, and 1 depend upon the chemical form of tritium. Because of the complexity of chemical transformation of tritiated compounds in biological systems, it has not been possible to determine these coefficients with any degree of certainty.

Metabolism of tritiated luminous compounds, which has been the subject of many investigations (121, 194, 253), is summarized by Warwarna (249). From his evaluation it was concluded that biological behavior of tritiated luminous compounds varies considerably. One interesting finding relates to the percutaneous absorption of tritiated compounds. This kind of exposure may result in a delayed excretion of tritium from its exposure site to the body water.

Another significant finding was the presence of organic compounds in the urine of rats which were fed with tritiated compounds, causing significant loss of tritium subsequent to the distillation of urine. This finding led to the recommendation that, in operations where tritiated compounds are used, radiobioassay should be carried out using undistilled urine.

Because of the uncertainty associated with the fraction of tritium which ends up in the organic fraction of the tissue, Moghissi et al. (167) suggest a uniform distribution of tritium in all hydrogen-containing materials of the body. This assumption is probably conservative at early times subsequent to exposure but under occupational exposure conditions becomes more realistic at a later date. After prolonged continuous exposure of varied concentrations, the uncertainties in the concentration of tritium in body tissue exceed those in body water. This consideration is disputed by Lambert and Vennart (130), who suggest that little or no tritium is introduced into the organic fraction of the tissue under occupational conditions and thus that dosimetry based on body water alone is sufficient.

Another important consideration relates to the quality factor of tritium. Originally this factor was 1.7. At a later date ICRP reduced the quality factor of tritium to 1, and this value was also adopted by NCRP (181). This value is in contradiction with a recent review by Johnson (115) and new experiments by Moskalev et al. (172) who suggest a quality factor of 2 for tritium. 585

Although the evidence appears to be in favor of a quality factor of 2, in this report a quality factor of 1 is used in conformity with ICRP and NCRP. Because of an even distribution of tritium in body water and possibly in entire body hydrogen, the somatic dose from tritium nearly equals the genetically significant dose, the difference being caused by differences in hydrogen concentration in various organs. The differences in hydrogen concentrations amount to only a few percent and thus are small as compared to other inaccuracies associated with the present study.

From the foregoing, it becomes clear that occupational radiation dose from tritium exposure can be estimated using the equation, $H = 0.1C$, where H is the

annual radiation dose in mrem, and C is the concentration of tritium in body water in nCi/l. This equation was developed by Moghissi et al. (167) using data from ICRP for Reference Man (109). An interesting study by Colvin and Everts (48) indicates the necessity of care in evaluating hazards of radioluminous materials. These authors observed chromosomal aberrations in kidney and also in lung tissues subsequent to cutaneous absorption of tritiated luminous compounds. Although these aberrations were not excessive for the quantity of tritium used, the lung tissue aberrations were particularly important because the lung is generally not expected to be the site for concentration of absorbed tritiated compounds.

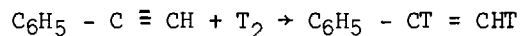
The tragic history of dial painting with radium seems to have been repeated in at least two cases of fatality as a result of tritium exposure (211). One of these cases deals with tritium dial painting. Although the subject was exposed to radium and strontium-90 as well as to tritium, the predominant exposure was tritium which, according to Seelentag (211), was the cause of death. Seelentag (211) suggests that overexposure was caused by lack of appropriate regulations and controls in Switzerland where the exposure occurred. Present Swiss regulations are in conformance with international guidelines (124, 125), and thus the reoccurrence of overexposures of the magnitude which led to the fatal case is unlikely.

3.2 TRITIATED POLYMERS

The common application of tritium in radioluminescent materials is in a polymer form. Potentially, all polymers containing hydrogen are available for this application. In practice, one is limited to reactions which permit easy hydrogenation during the process.

Krejci (124, 125), who reviewed various proposed polymers, concluded that polystyrene was probably the most suitable polymer. He based his recommendation on its ease of preparation, solubility properties, and its radiation resistance because of its predominant aromatic character.

At least in terms of radiation resistance, polystyrene--as it is produced for radioluminescent materials--is only slightly superior to the aliphatic polymers such as polyethylene. The common synthesis method is as follows:



Because the tritium label is at the side chain rather than the aromatic ring, it behaves almost as a normal aliphatic molecule. If the label had been in the ring, the radiation resistance obviously would have been improved. Aside from polystyrene, the only other compound with commercial applicability is a silicon polymer prepared by Evans and Maynard (59).

3.2.1 Occupational Exposure

Occupational aspects of tritium exposure have been subject to numerous investigations (20, 116, 117, 124, 125, 130, 167).

Vennart (234) reports that when tritium was introduced in the dial painting industry, it was anticipated that no tritium would be released from the tritiated paint. This expectation is somewhat unjustified as radiation decomposition of organic materials was well known, and at specific activities in which these materials were used self decomposition could have been calculated (60).

Radiobioassay of workers is easy and consists of measurement of tritium in urine at frequent intervals. Lambert and Vennart (130) point out the inaccuracies associated with the radiobioassay of tritium. They suggest that because of the short biological half-life of tritium of 10 days, even if weekly samples are taken, the dose may be underestimated by 60 to 70 percent if the exposure occurred precisely subsequent to the previous sampling.

One should, however, take into account the peculiarities of exposure of the workers. Although single large intakes occasionally occur in a dial painting operation, the contamination of air and surfaces is continuous, and thus the workers usually are subjected to almost continuous exposure. In addition, if the dose is occasionally underestimated by sampling too long after a significant intake, this underestimation is probably offset by the overestimation of the dose if a measurement is made shortly after a significant intake. This is particularly true for operations with many employees who are randomly subjected to exposure. There is, however, no question that a more precise dose measurement of the workers would be desirable, and thus a radiobioassay program with more frequent sampling than monthly may be advisable. Table 2 contains occupational exposure as measured by Moghissi et al. (167) and Krejci (124, 125). The values reported by Krejci are revealing because they indicate differences exceeding one order of magnitude.

Table 2. Average occupational exposure to tritium according to Moghissi et al. (167) and Krejci (124,125)

Location of plants	Average activity in paint (mCi/g)	Processed tritium (Ci/person-yr)	Average urine activity (μ Ci/l)	Risk (person-mrem/Ci)	Reference
U.S.A.	150	104.3	20.4	19.1	Moghissi et al.
Switzerland	150	193.4	2.57	1.3	Krejci
Switzerland	227	64.9	3.43	5.3	Krejci
Switzerland	102	140.8	7.64	5.4	Krejci
Switzerland	164	222.2	13.1	5.9	Krejci
Switzerland	262	67.6	4.86	7.2	Krejci
Switzerland	354	79.6	9.57	12.0	Krejci
Switzerland	453	65.3	14.2	21.7	Krejci
Average				9.1	

In order to obtain the total person-rem associated with tritium dial painting operations, either the number of employees or the total activity processed each year must be known. Moghissi et al. (167) include both values in their paper. Krejci (124, 125) does not include either value. Therefore, only average risk in mrem/Ci can be calculated from the data of Krejci. Dose estimates were carried out using the urine values (μ Ci/l) according to the method described in section 3.1.

Both Moghissi et al. and Krejci include the average specific activity of the paint in their data. Although it seems that higher specific activities are associated with higher dose, this relationship is weak and inconsistent and thus is not considered further.

3.2.2 Exposure to Tritium in the Assembly Plant

No information is available on the exposure of the workers in assembly plants where watch hands, faces, and mechanical parts are assembled. It is, however, conceivable that the exposure is significant. In the absence of measured values, radiation dose in assembly plants may be roughly estimated by comparing it to the painting operations. In the painting operation the paint is mixed, applied, and subsequently dried, whereas in the assembly plant the operation is somewhat comparable to the actual painting. Therefore, the expected exposures probably would be of the same order of magnitude. However, it is probable that, even though the residence time of tritium in these two operations may be comparable, tritium release in assembly plants will be comparatively smaller than in painting operations. This is caused by the increased tritium release from dry pigment as compared to paint and a reduction of tritium release with time in newly painted objects (106, 165, 167). The exact difference in exposure between these two operations is difficult to estimate.

In the absence of measured values, it is assumed somewhat arbitrarily that the radiation dose per Ci of tritium in the assembly plant is one-half of that in the painting operations. Obviously, this assumption is subject to substantial error. Based on this assumption, the average radiation dose to workers would be 4.5 mrem/Ci.

3.2.3 Exposure to Tritium During Storage

Bradley et al. (30) evaluated several storage facilities where tritiated watches were kept. They measured tritium urine levels as high as 28.8 μ Ci/l. The average air concentration divided by the total quantity of tritium on hand was 0.02 nCi/l per Ci. It is difficult to obtain person-rem data without better knowledge of the number of employees involved in the operation; Bradley et al. indicated one or two employees at each storage facility.

According to ICRP (109), an air concentration of 5 nCi/l corresponds to 5 rems for a 40-hour per week exposure when a quality factor of 1.7 is used. As mentioned previously (section 3.1), the quality factor is reduced to 1. Therefore, a tritium concentration of 1 nCi/l corresponds to a dose of 0.59 rem.

The average value of 0.02 nCi/l per Ci as measured by Bradley et al. (30) corresponds to an annual dose of 12 mrem/Ci for each worker. Assuming an occupancy of one worker per storage facility, 12 person-mrem/Ci would be obtained.

3.2.4 Exposure in the Repair and Refinishing Shops

Measurements in the repair shops are sketchy and limited. Bradley et al. (30) report average body burdens of 50 nCi/l corresponding to 5 mrem/year. They indicate 12 to 18 workers handling 10.5 Ci of tritium annually. If the average number of workers is assumed to be 15, an average of 7 person-mrem/Ci per year can be estimated. It should be noted that this dose is not delivered for every Ci of tritium processed in the dial painting of new watches because only a small fraction of watches is repaired.

3.2.5 Exposure to the User

Tritium release measurements, although numerous (41, 46, 52, 80, 84, 89, 104, 140, 184), are not easily relatable to body burdens of users of watches. Fitzsimmons et al. (70) reported tritium concentrations in body water ranging from 0.5 to 11 nCi/l above background corresponding to a radiation dose of <0.1 to 1.1 mrem/year with an average of 3.2 nCi/l corresponding to 0.3 mrem/year. Schell and Payne (206) report an average tritium concentration of 2.6 nCi/l above background in urine of people who are using pocket and wristwatches for an average of 18 hours/day.

Moghissi et al. (165) studied human exposure to tritium for radioluminous watches under controlled conditions. In their studies, 30 subjects were divided into six groups. Three commercially available tritiated compounds were used. The watches contained 15 mCi or 25 mCi and were worn 24 hours/day. In addition, a controlled group of unexposed individuals was included in the study. Urine samples were collected periodically from each individual and analyzed for tritium. Subsequent to an exposure time of 30 to 40 days, the watches were removed and the urine sampling and analysis was continued for an additional 30 days. The equilibrium tritium concentration was calculated from the urine data, and the dosimetry was carried out according to procedures described in section 3.1. The average dose per mCi of tritium paint was 0.03 mrem. The range of the values was 0.012 to 0.044 mrem/mCi. Considering differences in the chemical composition of the paint, variations in the manufacture of watches, and differences in the tritium turnover rate in the subjects, the relatively small range of these values seems to be remarkable.

Based on the value of 30 mrem/Ci reported by Moghissi et al. (165), the data reported by Fitzsimmons et al. and Schell and Payne can be used to estimate tritium content of a commercial watch. If the subjects studied by Schell and Payne had used their time pieces for 24 hours/day rather than 18 hours, as reported by those authors, their body water concentrations would have increased to 3.4 nCi/l, which is in agreement with 3.2 nCi/l as reported by Fitzsimmons et al. and corresponds to 13 mCi per watch. It should be noted that tritium content of watches is subject to considerable variation depending upon market requirements. Bradley et al. (30) who had access to a large number of watches suggest values ranging from 1 to 5.6 mCi per watch. Based on these data, it is assumed that the average activity of a time piece is 5 mCi, corresponding to an annual radiation dose of 0.15 mrem to the user.

3.3 TRITIUM GAS TUBES

Light sources can be produced using capillary tubes coated internally with zinc sulfide and filled with elemental tritium at pressures of less than one-half of an atmosphere. Application of tritium in elemental form is potentially desirable and safe provided certain requirements are met. As the biological absorption of elemental tritium is 10,000 times less than that of tritiated water, it is imperative to maintain a low water level in the container.

During the production process, separation of water from elemental hydrogen does not pose any significant problem provided proper manufacturing conditions are maintained. Subsequent to the introduction of tritium into the tube, however, residual moisture as well as dissolved oxygen or oxygen containing materials in the crystal and the glass wall lead to production of tritiated water either by oxidation or by hydrogen exchange. Because relatively carrier-free tritium is used in these tubes, minute quantities of oxygen or water may lead to significant quantities of tritiated water.

The actual measurements in regard to water content of gas-filled tubes are scarce and contradictory. Knapton and Comer (122) report values for T₂O in T₂ of up to 50 percent. These values are disputed by Guthrie and Coats (85), who suggest 1 to 2 percent T₂O in T₂ under proper manufacturing conditions. McNelis et al. (154), who exposed experimental animals to ruptured tritium-gas-filled tubes, found T₂O levels corresponding to 2 to 3 percent of the total activity. Also, Niemeyer (183) reports T₂O values of 2 to 3 percent in T₂. These contradictions may be based on inaccurate measurements or reflect differences in production methods.

The occupational exposure during production of these tubes could be significant if tritium is not properly contained. This is particularly important because elemental tritium is usually absorbed on titanium, zirconium, or uranium, and the biological implications of inhalation of these tritides are unknown. Although there is an economic motivation to contain tritium gas, the low price of tritium, which is one of the reasons for its application, reduces this motivation.

Coats (44) evaluated health implications of gas-filled tubes. Also, Doda (56) evaluated public health implications of the manufacture of these sources. It is generally accepted that the major route of exposure would occur during the breakage of the tube.

Because of the complete lack of available information on the occupational exposures and release rates from these tubes, it is impossible to estimate a dose associated with the application of these light sources.

Tritium release from gas-filled tubes depends upon a number of factors. Niemeyer (183) measured values corresponding to the diffusion rate of hydrogen through glass walls. However, values which exceed those produced by the diffusion rate have been measured, indicating errors in the production process. Here again, the chemical form would be of significance because of the differences in biological absorption of the two chemical forms.

Although several relevant studies have been reported (86, 182), the entire field of gas-filled tubes requires a careful study because of the apparent interest of the industry in this area and because of the potential attractiveness and use of these tubes. Because of substantially larger quantities of radioactivity in these tubes, it is necessary that sufficient information be available before they enter the market on a large scale. It is interesting to note that a committee of the Nuclear Energy Agency has recommended an exemption of gas-filled tubes for a tritium content not to exceed 200 mCi. Unfortunately, no specified chemical requirements are associated with that exemption.

4. PROMETHIUM-147

ISO.

4.1 PHYSICAL AND BIOLOGICAL PROPERTIES

Promethium is a rare earth element. Promethium-147 is produced abundantly in the fission process which accounts for its availability at reasonable cost. Promethium-147 decays with a half-life of 2.62 years to samarium-147 by beta decay with a maximum energy of 224 keV. Samarium-147 is an alpha emitter with a half-life of 1.05×10^{11} years. For evaluation of radiation hazard, the presence of samarium-147 can be neglected because a decay of approximately 1 Ci of promethium-147 would result in an activity of about 25 pCi of samarium-147. Various aspects of promethium-147 have been discussed in several publications (49, 75, 189, 221).

The uptake of promethium-147 through the GI tract is small--about 0.001 percent (189). This extremely low absorption is an added advantage for application of promethium-147 in radioluminous materials. Data by Moghissi et al. (164) indicate that GI tract absorption for promethium-147 in luminous compounds is reduced primarily as a result of high temperature firing of promethium on zinc sulfide which leads to the formation of insoluble promethium oxide. This reduction in solubility and the associated reduction in GI tract absorption is particularly pronounced if promethium is used in the form of microspheres such as those produced by the 3M Company.

Injected promethium quickly disappears from blood and is deposited in the liver and in bone. The biological half-life of promethium in humans is about 1,000 days (188) and agrees with measurements in large animals such as dogs and pigs. Experiments by Palmer et al. (189) in humans indicated a fraction going from blood to liver as 0.4 to 0.5 which is in disagreement with the ICRP (109) value of 0.06, calculated from rat data. According to ICRP (109), the maximum permissible body burden for promethium-147 is 300 μ Ci of which no more than 60 μ Ci may be deposited in the bone. The maximum permissible concentration of promethium-147 in air for occupational exposure is 1 pCi/l of air.

Promethium excretion in humans is complex in that in the first few days subsequent to intake, the excretion is predominately through urine, whereas at a later date fecal excretion dominates (189). It may be possible to estimate a promethium-147 body burden by combining urine and feces and attempting to relate their promethium-147 content to body burden, provided only a single intake occurred. The situation here is somewhat analogous to radium in that there exists a dynamic and complex relationship between intake and excretion.

The major route of intake of promethium-147 under occupational conditions is probably by inhalation, which would amount to an uptake of 25 percent versus 0.001 percent through the GI tract. No experiments are known on the possible cutaneous absorption of promethium-147, although this route may be as significant as inhalation due to the specific habits of dial painters.

From the foregoing it is evident that excretion data cannot be used to estimate the body burden of the dial painters. Whole body counting is not possible because of the nature of the promethium-147 emission, which is a beta particle of low energy. Attempts to use the bremsstrahlung of promethium-147 for whole body counting have failed because of the presence of natural radiation in the body which produces significant bremsstrahlung of a continuous nature, at least at the low end of the spectrum.

Promethium-147 is often accompanied by promethium-146 at levels ranging from 0.5 to 3 ppm. Promethium-146 decays with a half-life of 4 years: 35 percent by a beta decay with a maximum energy of 0.78 MeV and 65 percent by electron capture with cascading gamma emissions of 0.453 and 0.75 MeV, respectively. Because these two gamma photons are in coincidence, they are ideally suited for whole body counting. The similarity between the half-lives of promethium-146 and promethium-147 (4 versus 2.6 years) and identical chemical properties are added advantages. Therefore, it may be desirable to use a small amount of promethium-146 in promethium-147 to permit whole body counting of the workers using promethium-146 as a tracer. The level of promethium-146 in promethium-147 should be carefully considered to avoid a significant increase in external radiation, both at occupational and environmental levels.

4.2 OCCUPATIONAL AND COMMERCIAL EXPOSURE

Because of the lack of available data, the exposures to the dial painters and workers in the assembly plant and during storage cannot be estimated. By far the most important exposure should be that of internal dose commitment. As mentioned previously, there is no simple relationship between promethium excretion and the body burden. In a dial painting operation where exposure is usually continuous, there is essentially no conceivable way to estimate the body burden of the workers by urine and/or feces measurement. Specific aspects of promethium as related to dial painting operations, as a source of energy for activation of radioluminous materials and certain devices painted with promethium paint, have been discussed by various investigators (29, 47, 127-129, 233, 246-248).

In the U.S. there is only one plant which uses promethium-147 on a scale comparable to that used by other dial painting operations. This plant manufactures special devices and, because of its specific market conditions, is capable of maintaining occupational control conditions more comparable to radiochemical operations than to dial painting plants.

Data from this plant indicate that the air concentration had a range of 0.006 to 5 pCi/l, with the average being on the order of 0.01 pCi/l. The external dose was on the order of 5 mrem/Ci processed or less. As mentioned, the plant from which these data are reported is, however, an exceptionally well operated plant and is not representative of general dial painting operations. Therefore, these data will not be used.

4.3 EXPOSURE TO THE USER

Measurements of radiation dose are scarce and questionable. The problem is associated with the low energy beta emission of promethium-147 and the difficulties associated with TLD measurements at such low dose rates. In a measurement series in association with an Apollo spacecraft (229), a promethium-147 source of 1 mCi was measured with a GM counter and a value of approximately 3 μ rad/hour was reported from a distance of 30 cm. Measured rates at 15 cm and 60 cm were 10 and 0.8 μ rad/hour, respectively.

Obviously, the usual design of a GM counter is not appropriate for measurement of bremsstrahlung of 30 to 50 keV. If one accepts the methodology used by Joyet (118, 119), a 50-cm distance can be assumed, and assuming a 24-hour/day usage, a genetically significant annual dose of 8.8 mrem/mCi is calculated.

Unpublished data by Moghissi et al. (164) indicate that this value is too high. However, for reference purposes, 5 mrem/mCi per year will be used in the following calculations.

5. EVALUATION OF TOTAL RISK

Table 3 summarizes risks from processing 1 Ci of radium, promethium-147, and tritium as described in previous sections of this report.

Table 3. Total risk in person-mrem from various radionuclides per Ci processed

Occupation	Risk per Ci processed (person-mrem)		
	Radium	Tritium	Promethium-147
<u>Dial Painting</u>			
Bone	200,000	NA	Unknown
Whole body	600,000	9.1	5*
Lung	125,000	NA	Unknown
<u>Assembly</u>			
Whole body	69,000	4.5*	Unknown
<u>Storage</u>			
Whole body	Unknown	12*	Unknown
<u>Environmental (user's dose from wristwatches)</u>			
Whole body	(65-70)10 ⁶	30	5000

*Estimated values with limited usefulness

Occupational exposure is appropriately expressed as dose commitment. Obviously, radiation exposure caused by processing a certain quantity of a radionuclide does not necessarily cease after the worker leaves the plant. This is particularly valid for bone seeking radionuclides such as radium and promethium-147. However, environmental exposure (user's dose) is expressed in terms of annual dose.

Occupational skin exposure is not included in this evaluation. Evaluation of skin exposure to various radionuclides has been the subject of several studies (17, 92, 254), including investigations relative to radium dermatitis (113, 114). It has been extremely difficult to estimate even roughly the skin exposure in dial painting operations which presumably would be the area where the major exposure would occur. This exclusion, although regrettable, would not change the conclusions of this evaluation because it is acknowledged that although skin is as radiosensitive as most other body tissues in terms of carcinogenesis, skin cancers respond favorably to treatment and with proper care are unlikely to be fatal. A more detailed description of this subject is beyond the scope of this report, and the reader is referred to reports of various national and international organizations (106-110, 181).

The evaluation of the total risk should be based on benefits of the application of radioluminous materials. Although economic gain is certainly a consideration in evaluating the benefit, it cannot be used as the sole criterion. Also, the convenience of having a continuous light available at places where one otherwise would have to use other less convenient means must be

taken into account. Unquestionably, however the main benefit from the production of radioluminous materials is the light that is available on a continuous basis. Because the light production has a finite half-life, the usefulness of the paint must take this decay into account. If the light emission is below the limit of visibility, the light is of no use. Similarly, if exit signs are prepared from radioluminous materials, as soon as they decay below the user's visibility level, they are of no benefit in terms of risk-benefit analysis.

Table 4 is taken from an IAEA report (106) and contains relevant data on the relationship between the spot size and minimum brightness in μL . It is evident that the values deviate greatly because of the necessary assumptions that are required to make appropriate calculations from the data. It is obvious from the data in table 4 that a wristwatch which has decayed below 0.24 μL is below its useful life as far as its luminosity is concerned. In actuality, the subject is more complicated because a dark-adapted eye is capable of detecting the lower levels of luminosity than an eye which is not dark-adapted. In addition, as shown in figure 1, the optimum wavelength of light shifts to shorter wavelengths as a result of dark adaptation of the eye. All these facts must be taken into account if various radionuclides with different half-lives are being compared.

Another subject which must be considered is the property of the receptor of energy to receive the energy and convert it into light. By far the most efficient material for this conversion is zinc sulfide containing small quantities of copper or silver. This scintillator is often referred to as phosphor because it emits part of the energy promptly and another part after a certain length of time. This latter property is particularly desirable because phosphors are usually also excited by light having wavelengths equal or shorter than that of their emission. This added advantage, however, will not be considered here because it is independent of the light produced by the action of ionizing radiation, although often this excitation by light parallels that by ionizing radiation.

It is generally accepted that zinc sulfide is damaged by the action of ionizing radiation--the very radiation which produces light. Wallhausen (245) suggested a half-life of 10 years for zinc sulfide which is also accepted by the IAEA expert group (106). This decay must be considered in evaluating the useful luminosity of the radioluminous materials.

Because of the long physical half-life of radium, the effective half-life of radium-activated paint will be 10 years. The effective half-life of promethium-147 activated paint (Te) is calculated by combining the physical half-life of promethium-147 of 2.62 years with the 10-year half-life of zinc sulfide. Thus, $1/\text{Te} = 1/2.62 + 1/10$; $\text{Te} = 2.1$ years. The effective half-life for tritium gas can also be calculated to be $12.3 \times 10 / (10 + 12.3) = 5.5$ years. The actual value is rarely that long because of tritium release from the tubes.

A calculation of effective half-life for tritiated paint is somewhat more difficult because of differences in decay rate of various tritiated polymers. The IAEA report (106) assumes no tritium release and thus obtains the 5.5-year value calculated for tritium gas. This assumption is obviously invalid in light of data from many recent studies of both paint and users of luminous paints.

It is well known that tritium release is elevated during the first few weeks following application. We shall disregard this early part of the curve and consider tritium release subsequent to this time. Chollet (41) suggests an annual tritium release of 10 to 20 percent. Lorenzer and Born (140) report 16 percent release per year, whereas Guenther (84) suggests a 25 percent release

Table 4. Minimum brightness required for various types of ordinary timepieces (106)

Type of timepiece	Spot diameter (mm)	Reading distance (cm)	Minimum brightness (μL)
Ladies' wristwatches	0.4 - 0.8	25	0.38 - 1.4
Mens' wristwatches	0.6 - 1.0	25	0.24 - 0.65
Pocket watches	0.8 - 1.0	25	0.24 - 0.38
Travel alarm clocks	1.0 - 1.5	50	0.44 - 0.97
Large alarm clocks	1.5 - 3.0	50	0.11 - 0.44

Note: Luminosity is measured in units of brightness. Brightness is measured by the flux emitted per unit emissive area as projected on a plane normal to the line of sight. The unit of brightness is that of a perfectly diffusing surface giving out 1 lumen per square centimeter of projected surface and is called the lambert (L).

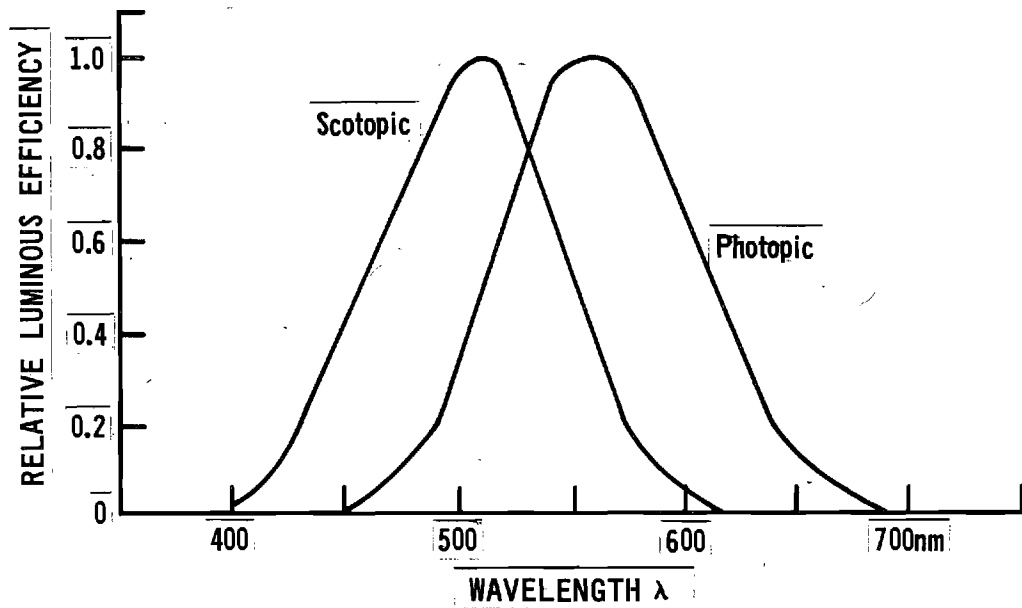


Figure 1. Standard relative spectral luminous efficiency functions for photopic and scotopic vision.

rate. Coenen (46) reports tritium release rates of 0.08 to 0.35 $\mu\text{Ci}/\text{mCi}$ per day with an average of 0.1 $\mu\text{Ci}/\text{mCi}$ per day. If the highest value is disregarded, this average corresponds to 3.6 percent per year, whereas his highest value relates to about 13 percent. From these data an average tritium release half-life of 4 years can be estimated. The effective luminosity half-life must take into account the physical half-life of tritium of 12.3 years, the stability half-life of zinc sulfide of 10 years, and the tritium release half-life: $1/T_e = 1/4 + 1/12.3 + 1/10$; T_e (tritium) = 2.3 years.

To properly compare these radionuclides, it is assumed that in every case sufficient activity is used so that at the end of the useful life of the object the minimum luminosity is above the level of effective visibility. In addition, the increased luminosity at the beginning is given credit by comparing the midpoint of the useful life of the object. This approach results in values equal to the integral of light during the useful life of the object. Figure 2 shows the details of the approach. Table 5 contains tritium and promethium activities corresponding to 1 Ci of radium. The conversion factors are taken from the IAEA report (106) and are 5.0×10^3 Ci for tritium and 1.7×10^2 Ci for promethium-147. These quantities produce a luminosity corresponding to that produced by 1 Ci of radium. Although newer paints produce light more efficiently than these factors would imply, particularly that for tritium, the above factors are used.

The IAEA report suggests a useful life of 10 years for time pieces. Increasing repair costs have surely reduced the useful life from 10 years to 5 years or less. In addition, the 10-year value was suggested by a panel consisting largely of representatives from countries where repair costs are lower than in the U.S. In the following evaluation values have been calculated for useful lives of 3, 5, and 10 years.

The evaluation of the total risk on a comparative basis is complicated by the differences in organs of reference. It is somewhat difficult to compare bone exposure with whole body exposure. In addition, many of the necessary data are unknown. This is particularly true for promethium. A closer look at table 3 indicates that for promethium occupational exposure is small as compared to environmental exposure. For tritium occupational exposure of about 25 person-mrem is about the same as that for environmental exposure of 30 person-mrem. For radium the user's dose of 65 to 70 person-krem is clearly dominant as compared to 600 person-rem each for dial painting and assembly plants. To facilitate the comparative evaluation of these three radionuclides, occupational and environmental (user's) exposures are discussed separately.

Table 6 contains radiation dose to various organs as a result of processing 1 Ci of radium and equivalent quantities of tritium and promethium-147. The values for tritium and promethium-147 have been calculated from data in table 3 using coefficients of table 5 for useful lives of 3, 5, and 10 years. Table 6 shows that radiation dose from tritium in every case is smaller than 600 person-rem whole body dose for radium.

Table 6 also indicates that for radium a large dose is delivered to the bone. As mentioned previously, it is somewhat difficult to compare bone dose with whole body dose. The comparison between bone dose delivered by radium and the whole body dose from tritium may be made possible if one considers that tritium is evenly distributed in the entire body hydrogen. Therefore, every organ is subjected to a dose proportional to the hydrogen content of that organ, provided T/H ratio is constant in the entire body. As discussed in section 3.1, the even distribution of tritium in the hydrogen pool of the body can be safely assumed. Unfortunately, bone is that organ of the body with a hydrogen concentration deviating more than that of any other organ from average hydrogen

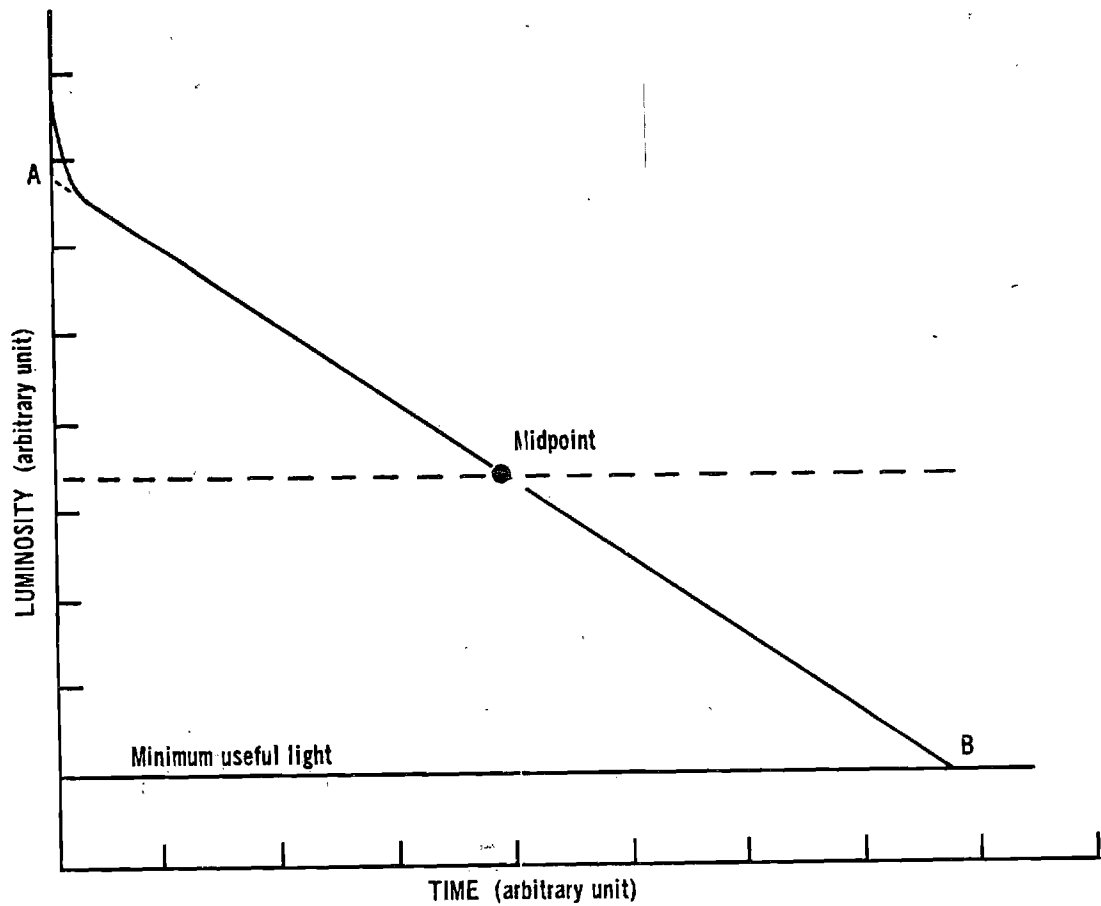


Figure 2. Decrease of luminosity with time; the average luminosity from A to B is expressed by the horizontal line passing through the midpoint.

Table 5. Required quantities of tritium and promethium-147, in Ci, which produce an average luminosity equal to the average luminosity of 1 Ci of radium for objects with useful lives of 3, 5, and 10 years, respectively

Useful life (years)	Tritium (Ci)	Promethium-147 (Ci)
0	5,000	170
3	8,000	280
5	11,000	390
10	23,000	880

concentration of the body. However, in the following it is assumed that bone contains "average" hydrogen concentration. This assumption introduces significant errors but makes it possible to compare bone dose with whole body dose, at least on a semiquantitative basis.

Table 6 shows that for useful lives of 3 and 5 years radiation dose to bone from tritium in every case is smaller than 200 person-rem. For a useful life of 10 years whole body dose for tritium in dial painting plants is about 209 rem as compared to the 200 person-rem for bone in radium dial painting plants.

Because of the similarities of chemical properties of radium and promethium, it can be expected that a major fraction of radiation from promethium decay would be delivered to the bone. Unfortunately, no data for promethium are available and none can be expected to become available because of the extreme difficulties in measuring the body burden of the workers. 1268

From the foregoing it can be seen that the occupational exposure is generally higher for radium than it is for tritium, although for longer useful lives the difference becomes smaller.

Table 7 shows the relative environmental risk (user's risk) for tritium and promethium-147 as compared to radium. This table is based on values taken from table 3 using coefficients of table 5. It is evident that radium exposes users of the objects by many orders of magnitude more than tritium or promethium, although the values for promethium could be inaccurate by as much as one order of magnitude.

It should be mentioned that these estimates are for wristwatches. Pocket watches would result in similar doses for radium as indicated in section 2.5. The gonadal dose in this case would be about 78 mrem/ μ Ci per year, which is similar to 65 to 70 krem/Ci per year. Pocket watches containing tritium would probably result in less exposure than wristwatches because of the possible absorption of tritiated water by fibers of the clothing. The estimates for promethium are missing.

The risk associated with alarm clocks would be smaller for all three radionuclides than the risk associated with wristwatches. This decrease would be comparatively more pronounced for promethium with its soft bremsstrahlung as compared to radium. Undoubtedly, the tritium release would be reduced because of the lower specific activities required for clocks.

If it were assumed that all radium is used to prepare alarm clocks, the user's exposure would be reduced by two orders of magnitude as described in section 2.5. However, at least a proportional reduction in tritium and promethium should be observed, and thus values in table 7 would not change in favor of radium. The comparison between promethium and tritium is not possible at present because of the lack of available data.

Tritium-gas-filled tubes are potentially the least hazardous of all radioluminous materials. Unfortunately, little pertinent data are available. Manufacturing conditions affect population exposure more in this case than in any other method used to prepare radioluminous consumer products.

Table 6. Occupational dose in person-rem as a result of application of 1 Ci of radium and equivalent quantities of tritium and promethium-147; the values have been normalized for objects with useful lives of 3, 5, and 10 years, respectively

Operation	Organ	Useful life (years)	Radium (person-rem)	Tritium (person-rem)	Promethium-147 (person-rem)
<u>Dial painting</u>	Bone	3	200	NA	Unknown
	Bone	5	200	NA	Unknown
	Bone	10	200	NA	Unknown
	Whole body	3	600	73	1.4*
	Whole body	5	600	100	2.0*
	Whole body	10	600	209	4.4*
	Lung	3	125	NA	Unknown
	Lung	5	125	NA	Unknown
	Lung	10	125	NA	Unknown
<u>Assembly</u>	Whole body	3	600	36*	Unknown
	Whole body	5	600	50*	Unknown
	Whole body	10	600	105*	Unknown
<u>Storage</u>	Whole body	3	Unknown	96*	Unknown
	Whole body	5	Unknown	144*	Unknown
	Whole body	10	Unknown	276*	Unknown

*Estimated value with limited usefulness.

Table 7. Relative radiation risk to the user of a wrist-watch painted with radioluminous material containing tritium or promethium as compared to radium; the values have been calculated for indicated useful life of the watch.

Useful life (years)	Tritium	Promethium*
3	3×10^{-3}	2×10^{-2}
5	5×10^{-3}	3×10^{-2}
10	10×10^{-3}	6×10^{-2}

*Numbers for promethium are of limited usefulness.

6. POPULATION DOSE

Radioluminous materials are being used in many applications. It has been impossible to obtain sufficient information to make a reasonable estimate relative to the types, numbers, and activity contents of the objects or the exposure data. This information is, however, fundamental in estimating the population dose and person-rem data. It is known that radioluminous materials are being used in exit signs, compasses, gun sights, and many other objects. The description of types and numbers of these objects is in many cases guarded by manufacturers and occasionally by users of these objects.

Because of the limited information available, in the following presentation only time pieces are considered for estimation of person-rem data, realizing that this estimate represents a minimum of the actual exposure which is probably higher.

Table 8 contains data obtained from manufacturers and importers of watches and/or clocks. The estimation of average specific activity is somewhat difficult and subject to considerable variation. An example is the values related to radium. According to table 8, no radium wristwatches and pocket watches have been sold in the U.S. in the last 3 years. However, there are many radium watches in use. The following calculations are therefore subject to considerable inaccuracies.

All data are calculated for January 1, 1974, and assuming an average useful life of 3 years, it can be seen that a total of 24×10^6 watches and clocks painted with tritiated luminous materials have entered the market. It is assumed that the exposure in every case is from wristwatches. This calculated overestimation is probably offset by unreported imported watches and other objects painted with radioluminous materials such as compasses. If the average activity of watches is assumed to be 5 mCi, the total activity can be calculated to be 1.2×10^5 Ci. The corresponding environmental dose is calculated to be 3600 person-rem.

Table 8 indicates 2.3×10^6 watches activated with promethium-147 for the 3-year period ending January 1, 1974. In addition, 3.8×10^6 clocks containing promethium have been imported. There are no data available on the average promethium-147 activity for a watch. Hence, no person-rem data are calculated for promethium-147.

The recordkeeping requirements for tritium and promethium-147 do not apply to radium. As a naturally occurring radionuclide, radium is not covered by rules and regulations promulgated by the Atomic Energy Commission. Although certain State rules and regulations include radium, the lack of uniform legislation has led to certain inaccuracies in estimating radium exposure.

Table 8 shows a total of 8.4×10^6 clocks painted with radium were sold during the period ending January 1, 1974. Using an average of 0.5 μ Ci per clock, a total of 4.2 Ci of radium can be calculated. This is probably a minimum and the actual value could be much higher.

Table 9 contains data relative to the quantities of time pieces, radioactivity, and the population dose. The reason for the comparatively small dose from radium is that its use is limited to alarm clocks. The same quantity of radium in wristwatches would have resulted in substantially higher values.

Table 8. Luminous timepieces distributed in United States

	1971	1972	1973
<u>Wristwatches</u>			
a) Tritium activated			
Made in U.S.	2,710,000	2,330,000	1,800,000
Imported	5,670,000	6,540,000	3,600,000
b) Promethium activated			
Made in U.S.	Negligible	Negligible	Negligible
Imported	620,000	770,000	900,000
c) Radium-226 activated	Negligible	Negligible	Negligible
<u>Clocks</u>			
a) Tritium activated			
Made in U.S.	18,000	10,000	20,000
Imported	500,000	190,000	240,000
b) Promethium-147 activated			
Made in U.S.	Negligible	Negligible	Negligible
Imported	1,470,000	970,000	1,370,000
c) Radium-226 activated	2,800,000	2,800,000	2,800,000

Table 9. Evaluation of population dose in U.S. to radioluminous time pieces

	Tritium	Promethium-147	Radium
Number of timepieces	24×10^6	6×10^6	8.4×10^6
Average activity of timepiece	5 mCi	Unknown	0.5 μ Ci
Total activity	120 kCi	Unknown	4.2 Ci
Population dose (person-rem/year)	3600	Unknown	2500

7. CONCLUSIONS AND RECOMMENDATIONS

Radiation exposure to the U.S. population as a result of application of radioactive luminous compounds, although relatively small, is significant. Radiation dose estimates indicate about 3600 person-rem from tritium, 2500 person-rem from radium, and an unknown quantity from promethium to the population of the U.S. during the calendar year of 1973. If one assumes 6100 person-rem as a reasonable minimum from all radionuclides, an average exposure of 0.03 mrem/year for the entire population of the U.S. of about 2×10^8 people can be calculated. The dose would have been significantly higher if radium had been used to produce wristwatches rather than clocks. The inclusion of occupational exposure would increase this value by at least a factor of two and probably more.

The occupational exposure in the U.S. for producing radioluminous devices is comparable for tritium and radium. No assessment of the occupational aspects of promethium is possible at this time due to lack of relevant information.

Population dose from radium would be higher by several orders of magnitude as compared to tritium and promethium for producing the same quantity of light for objects with useful lives of 3, 5, or 10 years. Radium offers no advantage as compared to tritium.

Like tritium, promethium-147 emits soft beta radiation which makes it suitable as an activator for radioluminous materials. External radiation from these radionuclides is small and in the case of tritium almost nonexistent. Unlike tritium, promethium-147 is not evenly distributed in the body and follows a dynamic excretion pattern. Therefore, it is impossible to estimate body burden of dial painters by any known method.

The conclusions thus are obvious: radium should not be used for dial painting because tritium can be used to accomplish the same results with substantially less exposure to the user. Before a large-scale application of promethium-147, methods must be developed to determine the body burden of workers as a result of occupational exposure.

One possibility consists of increasing the promethium-146 content of promethium-147. However, this increase must be carefully studied to avoid an unnecessary increase in population exposure to gamma radiation from promethium-146. A cursory look at the tables clearly indicates the need for evaluation of promethium-147. There is insufficient information to make all but the roughest estimates relative to this radionuclide.

In the case of tritium, exposure during assembly and to some extent during storage are insufficiently known. Further studies in this area are needed.

Radioluminous tubes filled with tritium gas are potentially useful. However, the chemical composition of tritium in these tubes and the mechanism of tritium release must be carefully studied prior to their wide-scale application.

During this study much information was given to the investigators with the understanding that the source would not be revealed. Furthermore, it was indicated several times that certain information would be released if similar information were also released by others. The desire to participate in a meeting to reveal, discuss, and evaluate the useful data and experience was repeatedly expressed. Such a meeting is therefore recommended.

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