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ESTIMATED RADIATION DOSE FROM TIMEPIECES CONTAINING TRITIUM*

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ABSTRACT

Luminescent timepieces containing radioactive tritium, either in elemental form or incorporated into paint, are available to the general public. The purpose of this study was to estimate potential radiation dose commitments received by the public annually as a result of exposure to tritium which may escape from the timepieces during their distribution, use, repair, and disposal. Much uncertainty is associated with final dose estimates due to limitations of empirical data from which exposure parameters were derived. Maximum individual dose estimates were generally less than 3 μSv/yr, but ranged up to 2 mSv under "worst-case" conditions postulated. Estimated annual collective (population) doses were less than 5 person-Sv per million timepieces distributed.

INTRODUCTION

An isotopic application of tritium (T) which increases the exposure of the general population to this isotope is its use in radioluminous timepieces. Presented here are the results of two studies (1,2) conducted to estimate potential radiation doses to the public due to the distribution, use, repair and disposal of such timepieces. These studies were initiated at the request of the Nuclear Regulatory Commission (NRC), for the purpose of identifying consumer products that pose a potential for unnecessary radiation exposure to the general public and of identifying products about which sufficient information is not available to estimate radiation exposures.

Tritium has been used in two different ways to illuminate portions of the dial or hands in timepieces. In one application, tritium and a phosphor such as zinc sulfide (ZnS) are incorporated into a polymer paint (3,4) which is applied to the hands and/or dials of the timepiece face. Types of timepieces available containing tritiated paint include wristwatches, pocketwatches, and clocks. Approximately 80 MBq (2 mCi) of tritium are used in each wristwatch, and approximately 20 MBq (0.5 mCi) are used in each pocketwatch or clock, according to NRC licensing files.

A more recently developed means of utilizing the phosphorescent properties of the tritium/phosphor combination is found in liquid crystal display (LCD) wristwatches. These timepieces contain approximately 4 GBq (100 mCi) of elemental tritium (T_2 or HT) in each of two sealed borosilicate glass tubes, the inside surfaces of which are coated with an inorganic phosphor. The tubes are bonded with a shock-absorbent silicone adhesive to a metal tray that is sealed to the display panel, and thus are well-protected and not easily accessible.

Because tritium readily exchanges with hydrogen atoms present in the atmosphere and in most materials with which it has contact, leakage of this isotope from timepieces can occur (5,6). The significance of exposure to tritium which may occur as a result of this leakage is the subject of this paper.

Levels of tritium in urine samples from individuals wearing wristwatches containing tritium have been found to be enhanced over samples from non-wearers (7,8), which indicates that internal exposures to tritium do result from use of the timepieces. In this paper, annual radiation doses potentially received by all individuals exposed to the products during distribution and marketing, repair, disposal as well as during use are estimated. The procedure followed to estimate doses and the doses so obtained are discussed below. The significance of exposure to tritium that escapes from timepieces is evaluated by comparing estimated doses with doses from natural background (including all naturally-occurring radioisotopes) and by considering the potential interference of escaped tritium in personnel monitoring, where it is desireable to detect body burdens of tritium due to sources other than timepieces.

PROCEDURE

Estimating Exposure

In order to estimate the magnitude of ex- posure of individuals to tritium, hypothetical exposure scenarios were developed to describe conditions under which typical exposures may occur during the various stages considered in the life span of timepieces containing tritium. Parameters describing leakage rates of tritium, ventilation characteristics of the locations in which exposures occur, and duration of exposures were defined for each scenario established. Values of these parameters were estimated from documented information and from informal interviews with individuals involved in occupations or activities pertinent to the distribution, use, repair, and disposal of consumer products such as timepieces. In most cases, parameter values were chosen to represent what are believed to be most probable values in a sometimes broad range of possible values. However, because the NRC must also consider exposures to individuals that could conceivable occur under "worst-case" conditions in order to fully analyze the impact of consumer products containing radioactive materials, a number of scenarios were developed to address this need.

Collective* exposures of the U.S. population to tritium which leaks from timepieces were also estimated, thus necessitating consideration of the number of individuals that might be exposed during the useful life of the timepieces. The number of individuals potentially exposed is a function of the number of timepieces distributed, used, repaired, and disposed of in a specified period of time. Due to the lack of a firm estimate of the numbers of timepieces distributed in the United States, collective exposures were estimated per million timepieces of each type distributed and disposed of each year. It was assumed that the useful life of each type of timepiece is ten years, and thus that 10 million timepieces of each type could be in use at any one time for each million distributed annually. Individuals not directly involved in activities pertinent to distribution, use, repair, or disposal of timepieces, but potentially exposed to tritium leaked from timepieces were also considered in estimating collective exposures.

Distribution of timepieces was assumed to occur via a parcel delivery system. Thus, persons employed in a number of occupations supportive of the parcel delivery network were assumed to be ex-

*The term "collective" is used throughout this paper to represent the population which is exposed to tritium from timepieces.

posed to the tritium which leaks from timepieces being distributed to wholesale or retail outlets. Exposures assumed to occur at the wholesale and retail levels involved warehouse and stockroom clerks, salespersons, and customers.

Persons exposed to tritium leaking from timepieces during use include wristwatch wearers, pocketwatch users, and clock owners. Individuals who work in the immediate vicinity of, live with, or otherwise associate with owners of such timepieces were also considered.

Individuals assumed to be exposed to tritium during repair of timepieces included only repairmen, since customers would likely spend only a short period of time in repair shops. Disposal of timepieces after an average 10-year useful life was assumed to result in exposures to members of the general population who reside within a 72-km (42-mile) radius around the disposal site. Storage of obsolete timepieces in owner's homes prior to disposal was also considered a source of exposure to residents.

Exposures to tritium that may occur under "worst-case" conditions were estimated for the following situations. Fires in both a home and warehouse where watches containing tritiated paint were present were assumed to cause volatilizations of all tritium in the watch (80 MBq per watch), leading to exposure of persons present prior to evacuation. Accidental ingestion of tritiated paint which might flake from a timepiece during repair was also considered. Finally, destruction of glass tubes and total release of elemental tritium contained (8 GBq) was considered a possible means of exposure during repair.

The rate of tritium leakage from watches by diffusion through tubes containing tritium or as it emanates from paint via exchange with atmospheric hydrogen has been measured by manufacturers of the timepieces, and others (5,6). Surprisingly, average rates of leakage from wristwatches containing elemental tritium and those containing tritiated paint are approximately equal. A leakage rate of 18 mBq/sec (30 pCi/min) was assumed for wristwatches of both types based on available empirical data. For pocketwatches and clocks containing tritiated paint, a leakage rate of 5 mBq/sec (8 pCi/min) was assumed, based on the lower total content of tritium in these types of timepieces.

The dispersion of tritium into the air surrounding the timepiece is likely to be quite irregular and not readily described with available dispersion models, since the source is not stationary in most instances. Therefore, equilibrium concentrations of tritium were assumed to exist within the boundaries of the exposure area during distribution, use, and repair; boundaries being defined by a room size or truck size in the exposure scenarios. This assumption might underestimate doses to watch wearers but overestimate doses to persons assumed exposed in the vicinity of wearers. The exception to this is the exposure scenario for disposal where it is assumed that timepieces discarded in municipal waste are either burned or buried. Here diffusion of tritium from the disposal site is represented by a Gaussian plume model (9), and assumed to be insignificant beyond 72 km (45 miles) from the site.

Ventilation rates were assumed to be one air change per hour in the exposure area for structures such as homes or office buildings, and two air changes per hour for trucks used in transport shipments. The assumed duration of exposure for all potentially exposed individuals varied from less than one hour per year for some persons employed in parcel delivery terminals, to 8760 hours per year (24 hours per day, 365 days per year) for owners of timepieces containing tritium.

Estimating Dose

Annual individual and collective dose commitments were estimated using the exposure scenarios and associated parameters summarized above. The CONDOS computer code (10) was implemented to calculate potential doses received during distribution, use, and repair of timepieces, and for accidental situations. This code provides a methodology to systematically estimate radiation dose to each of a number of groups of individuals, members of which are exposed to tritium in the manner designated by the exposure scenario which prevails for that group. The AIRDOS-II computer code (9) was used to estimate doses during disposal of timepieces.

Inherent in these codes is the use of a dose conversion factor (DCF) that estimates 50-year dose commitments for inhaled or ingested radionuclides. Thus, all doses reported are 50-year dose commitments for an annual exposure period. Doses received as a result of exposure to tritium are highly dependent on the chemical form of the airborn tritium. Exposure to airborne tritiated water vapor (HTO or T_2O) will result in an internal exposure that is approximately 10^3 times greater than that resulting from exposure to the same amount of tritium in elemental form (11, 12). Therefore, the DCF's will differ by a factor of approximately 10^3. In this assessment, it was assumed that tritium which leaks from timepieces results in exposure to tritiated water vapor alone. In other words, complete conversion of any elemental tritium to tritiated water vapor was assumed to occur before inhalation and absorption through skin takes place. For scenarios in which tritium is released to the atmosphere following destruction of the timepiece in fires, complete conversion is also assumed. However, in the case of mechanical destruction of glass tubes containing elemental tritium, only 20% conversion is assumed to occur. This latter assumption is based on measurements which indicate that, in the absence of a catalyst such as fire, conversion is a slow process (half-time > 96 hr) for concentrations much higher than those encountered in this study and that conversion rates decrease with concentration (13,14). A more exact representation of the percent conversion was not possible due to fluctuations in available data at lower concentrations which prohibit extrapolation to a value to be used in this study, although it is likely that this 20% value is probably an overestimate of the actual percentage.

A dose conversion factor of 0.34 pSv/Bq (0.13 mrem/μCi) for inhalation of tritiated water vapor (15) was used in this assessment for calculating whole-body dose commitments. This factor assumes that 1/3 of the total dose is received via absorption through skin. The contribution to dose of elemental tritium was neglected, since it is insignificant with respect to the dose produced by tritiated water vapor in all scenarios.

RESULTS AND DISCUSSIONS

Estimated annual maximum individual and collective dose commitments are presented in Table 1. The largest maximum individual doses calculated are associated with the exposure scenarios devised to account for accidental breakage of tubes containing elemental tritium (2×10^3 μSv) and accidental ingestion of tritiated paint (6×10^2 μSv for wristwatches; 1.5×10^2 μSv for clocks or pocketwatches).

The scenario in which a person was estimated to receive 2 mSv (200 mrem) involved the breakage of two glass tubes from one watch during repair, complete release to the atmosphere in the repair shop of 200 mCi of tritium of which an average of 20% is in the form of tritiated water vapor, a shop ventilation rate of 5.0×10^3 cc/sec (one air change per hour), and an exposure duration of 10 hours. The scenario describing accidental ingestion of tritiated paint also involved a repairman, who might be exposed to flaking paint from a timepiece during repair. The amount assumed ingested was 40 MBq (1 mCi) from wristwatches, or 10 MBq (0.5 mCi) from pocketwatches or clocks, representing one-half of the total activity originally contained in each timepiece. Tritium ingested was assumed to be completely soluble. As was mentioned earlier, these latter scenarios are based on "worst-case" assumptions, and thus their likelihood of occurrence is unknown, and at most, very low.

The maximum individual doses calculated as a result of disposal of timepieces are also on the upper end of the range of doses estimated for all stages assessed. These maximum doses were estimated by assuming that an individual remained for an entire year at a distance of 100 m from a municipal disposal site where 16,000 timepieces are assumed to be destroyed annually by ground-level burning. The number of watches destroyed at this site was estimated by assuming that residents in a large city such as New York City, where approximately 4% of the United States population resides (16), were serviced by only one municipal disposal site, and that 4% of the total number of timepieces disposed of annually in the United States by ground-level burning were present at this site. This scenario for the maximally-exposed individual is consistent with scenarios used to estimate maximum individual doses as part of the environmental impact assessment for nuclear reactors (17). The assumptions used in this scenario are highly unlikely in the context to which they are applied here.

Annual maximum individual doses for all other scenarios ranged from 0.5 μSv (.005 mrem) to 3 μSv (0.3 mrem), and the total collective doses for each type of timepiece ranged from 0.6 person-Sv (60 person-rem) to 5 person-Sv (500 person-rem). In interpreting the estimated doses listed in Table 1, it should be remembered that a large amount of uncertainty is associated with such values due to uncertainties inherent in assumptions concerning ventilation and dispersion characteristics, leakage rates of tritium, and exposure duration. It should also be noted that the estimated maximum individual doses for all scenarios, except the few unlikely accident situations discussed here, constitute less than 1% of the average annual whole-body radiation dose of 1250 μSv (125 mrem) received by individuals from exposure to naturally-occurring radiation (18).

The results of two separate investigations involving the monitoring of tritium levels in humans indicate a correlation between the use of wristwatches containing tritium and urine levels of the isotope. In one case where personnel in a low-level tritium laboratory were monitored routinely (8), detectably enhanced levels of tritium in urine were correlated with use of wristwatches which leaked tritium at a rate greater than 3 mBq/sec (5 pCi/min). In the second case, persons residing near a natural gas stimulation experiment were monitored to obtain baseline data on urine concentrations of tritium prior to the experiment (7), and, again, enhanced levels were found to be correlated with the use of wristwatches containing tritium. Doses calculated from urine concentrations of tritium found to exceed background levels in these experiments range between 0.2 μSv (.02 mrem) and 30 μSv (3 mrem), assuming the specific activity of tritium in the urine is indicative of that throughout the body (19). These results indicate that doses estimated by procedures described throughout this paper may slightly underestimate doses received as a result of wearing wristwatches containing tritium, since a maximum individual dose of 0.2 μSv was predicted. However, the limited amount of empirical data collected to date [a total of 26 subjects were monitored in the two investigations (7,8)] precludes a more definitive statement regarding the accuracy of the theoretically-derived dose estimates. The empirical data do suggest, however, the potential for interference in personnel monitoring posed by timepieces which contain tritium.

CONCLUSIONS

The purpose of this paper has been to elucidate the potential significance of timepieces containing tritium with respect to radiation doses received by users, distributers, and other members of the general public who might be exposed to the radioactive contents of these consumer products subsequent to their manufacture. Annual individual and collective dose commitments were estimated for persons potentially exposed during the distribution, use, repair, and disposal of wristwatches, pocketwatches, and clocks that contain tritium. Per million of each type of timepiece distributed annually, estimated individual dose commitments were generally less than 3 μSv (0.3 mrem), but ranged up to 2 mSv (200 mrem) under "worst-case" assumptions. The degree of confidence which may be placed in such estimates is severly hindered by uncertainties associated with leakage rates of tritium from timepieces, aerosol dispersion of tritium released, and conversion rates of elemental tritium to tritiated water vapor. It was found, however, that some of the estimated doses associated with use of wristwatches fall within the range of doses calculated from empirical data obtained on urine levels of tritium in persons wearing wristwatches containing tritium.

Most of the maximum individual dose commitments constituted less than 1% of average annual individual doses received from all natural background radiation. The highest maximum individual dose estimated was approximately 160% of the average natural background dose, although the likelihood of such a dose is unknown and, is probably very low. The absolute significance of the collective doses calculated with respect to background collective doses is not known, since the number of timepieces in circulation is not known. It can be estimated, however, that for every million timepieces containing tritium of any type distributed annually, the maximum collective dose commitment would be less than 5 person-Sv (500 person-rem), which constitutes less than 0.01% of the average annual collective dose (approximately 4×10^5 person-Sv) received by the U.S. population from natural background radiation.

Thus, it seems that doses potentially received by the general public from timepieces containing tritium are generally insignificant with respect to those received from natural background radiation. However, the importance of individual doses potentially received under abnormal conditions, such as those described here, should not be overlooked and it is recommended that further data be collected to evaluate the assumptions upon which these "worst-case" dose estimates were based. Furthermore, the potential interference of tritium leaked from timepieces in personnel monitoring should be recognized.

Table 1. Estimated annual maximum individual and collective whole-body dose commitments for all stages considered in the life span of timepieces containing tritium (per million of each type distributed)

Timepiece	Distribution	Use	Repair	Disposal	Accidental
Wristwatches with elemental tritium					
Maximum individual[a]	3.0	0.2	0.6	200.	2000.
Collective[b]	0.09	2.3	0.01	2.5	*c*
Wristwatches with tritiated paint					
Maximum individual	3.0	0.2	0.6	2.0	600.
Collective	0.09	2.3	0.01	0.5	*c*
Pocketwatches with tritiated paint					
Maximum individual	0.8	0.06	0.2	0.5	150.
Collective	0.02	0.5	<0.01	0.1	*c*
Clocks with tritiated paint					
Maximum individual	0.8	0.05	0.2	0.5	150.
Collective	0.02	1.6	<0.01	0.1	*c*

[a]Individual dose commitments given in units of μSv/year (1 μSv = 0.1 mrem).

[b]Collective dose commitments given in units of person-Sv/year (1 person-Sv = 100 person-rem).

[c]Probability of occurrence unknown, but probably very low.

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