Biokinetics and Dosimetry of Inhaled Tritiated Aerosols

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INTRODUCTION

Tritium could be released to occupational and ambient environment in tritiated aerosol forms. The major categories of tritiated aerosols include (1) volatile tritiated organics such as tritium-gas-contaminated pump oils and tritiated methane, and (2) stable metal tritides or tritiated dusts (Hill and Johnson, 1993). The deposition, retention, and clearance of inhaled tritiated aerosols can be quite different from tritiated water (HTO) and tritium gas (HT), depending on the dissolution rate of the metal tritides and the chemical nature (e.g., reactivity, diffusivity etc.) of the tritiated organics.

The operating experience with vacuum pumps in tritium handling facilities have shown that the saturated hydrocarbon mineral oils in these pumps require frequent changes because of vapor pressure increases (off-gassing) and liquid viscosity increases. The specific activities of tritium in pump oils can easily range from few kBq L^{-1} to tens of kBq L^{-1} (Krasznai et al., 1995). The wide range in specific activities can result from variations in the tritium inventory and total throughput of tritium in facilities. Depending on the history of the operating pumps, tritium could be present as some combinations of HT, HTO and organically bound tritium (OBT). While the chemical forms of tritium in pump oils are not fully characterized, quantitative radio-gas chromatography techniques to measure tritiated organics have identified the chemical species of tritiated hydrocarbons in the head space of various samples of waste vacuum pump oils and in the work-place atmosphere during a vacuum pump oil change-out procedure (Krasznai et al., 1995). Since tritiated pump oil, by their nature, are relatively volatile, the potential is always present for creating a workplace hazard by inhalation of vaporized aerosols of contaminated oils, especially during pump maintenance. The tritiated pump oils also present, to a much lesser extent, a radiological hazard to the public from their waste by-products. No metabolic data and dose models are currently available concerning the inhaled pump oil aerosols or vapors.

Stable tritiated materials are stable chemical compounds formed through adsorption of the tritium into metals, carbon or other dusts. They are widely used in nuclear engineering facilities for research; in purification, compression, and storage of tritium (Ortman et al., 1990); in fusion technology (Skinner et al., 1999); and in neutron generators (de Ras et al., 1980). Respirable size particles (<10 μm) of metal tritides can be released as aerosols during fabrication, assembling, and application of components or in accidental or fugitive releases. As a results, workers may be exposed to these radioactive materials by inhalation.

The current health protection guidelines for tritiated aerosols are based on the assumption that the biological behavior of these compounds is similar to HTO exposures. Tritiated water is absorbed into body fluid following all modes of intake and instantaneously distributes uniformly over all soft tissues. Tritium retention in the human body is described by a sum of two exponential functions with an effective biological half-life of 10d and 40d, respectively; the later contributing about 10% of the dose (ICRP-56, 1989). However, studies of tritium adsorbed into titanium and zirconium tritides suggest that tritium in these materials is poorly soluble in aqueous solutions. The cumulative fractional release of tritium from macroscopic objects of titanium and zirconium tritides after 100 d varied between 0.9 to 6.7×10^{-5} (Miller and Boka, 1985). In a recent study, the dissolution rate of 100 and 1 μm titanium tritide particles in a simulated body fluid was measured. The dissolution of tritium from the particles could be expressed by an exponential function with a half-time of 1 year for 100 μm particles, and two exponential functions with a longer half life of 33 d for 1 μm particles (Cheng et al., 1997). Existing information suggests that the solubility of metallic complexes of tritium may be low.

To evaluate the radiological hazards from the inhalation of tritiated aerosols, the distribution of tritium in the lung and the body of male rats were examined after their lungs were contaminated with tritiated pump oils or stable tritiated particles. The dynamics of tritium clearance from the body is investigated from biokinetic data of tritium-in-urine and tritium-in-feces. Data from this study has provided information that the inhaled tritiated pump oils and metal tritides (titanium tritides) do not transfer instantaneously from the lung to body fluids. The inhaled aerosols fix and label tritium in the exposed area of the respiratory tract. It is likely that the inhalation

dose coefficient values recommended by the International Commission on Radiological Protection Publication 71 (ICRP-71, 1995) for tritiated organics (i.e., pump oil) and stable tritiated compounds can introduce sizable uncertainties in the dose estimates for these types of exposure. The presented results are useful for evaluating the dosimetry of inhaled ritiated aerosols.

MATERIALS AND METHODS

Tritiated pump oils

The tritiated pump oil sample was prepared from the saturated hydrocarbon mineral oils (Duo-Seal vacuum pump oil) as mentioned earlier (Trivedi, 1995). The analysis of the pump oil, prior to animal experiments, showed that more than 95% of the tritium was fixed as non-exchangeable tritium in the oil. The different classes of tritiated species were not analysed in the stored oil.

Exposure methods

Four-month-old Sprague-Dawley male rats, weighing 450 ± 45 g, were used in this study as described elsewhere (Trivedi, 1995). The animals were mildly anaesthized prior to instillation of tritiated pump oil into the lung. Simplicity, rapidity, safety and dose accuracy were prime considerations in choosing the instillation method. About 0.1 mL (1.1 ± 0.4 MBq) of tritiated pump oil (10 MBq mL^{-1}) was deposited into the trachea bifurcation. The activity among the exposed animals differed due to variable instilled volume of high viscous oil.

Following exposures, the animals were returned to their cages. Selected animals were euthanized at 30 min, 1 d, 3 d, 5 d, 7 d, 14 d and 28 d post-exposure to collect a series of organs (e.g., brain, intestine, liver, lung, kidneys, stomach, testis) and carcass for tritium retention activity. Three animals in a group were euthanized post-exposure.

A group of animals were kept in metabolic cages to examine urinary and fecal excretion of tritium. The cumulative 24-h urine and fecal samples were collected at regular periods up to 28 d post-exposure to determine the HTO and OBT rate of excretion. Urine, feces and tissue samples from unexposed animals were collected to provide background tritium activities.

Activity measurements

All samples were stored frozen until the activity measurements were performed as described previously (Trivedi, 1995). All samples were analyzed within 3-4 months after sampling. The HTO and OBT concentrations in bioassay samples were corrected for possible cross-contamination (Trivedi et al. 1993).

Metal Tritides

The titanium tritide (Ti-T) was obtained from the Martin Marietta Pinellas Plant, Largo, Florida. These powders were further ground to fine powders in a ball mill at the Lovelace Respiratory Research Institute (LRRI). The fine powders were characterized as being respirable, with count median diameter (CMD) 0.95 μm, and geometric standard deviation (GSD) 1.93 (Cheng et al., 1997).

A 30 ml saline suspension containing 27.15 mg of the fine powder of the titanium tritide was made for the intratracheal instillation of rats. The specific activity of ^{3}H in the tritide was determined to be 2.23MBq/mg (60.25 μCi/mg). Each rat on study was intratracheally instilled with 0.5 ml of the suspension, which was 1.00MBq (27.11 μCi) of ^{3}H in 0.45 mg of the powdered tritide.

Animal Exposure

Thirty-six male F344/Crl rats of 11-12 weeks old were obtained from Charles Rivers Laboratories, Kingston, NY for this study. The rats were anesthetized with a mixture of halothane, nitrous oxide, and oxygen and intratracheally instilled with the titanium tritide suspension of 0.5 ml. An additional 0.5 ml of the saline was instilled into each rat lung immediately after instillation of the suspension of the tritide to help disperse the tritide particles into the deep lung.

Following instillation, the six rats assigned for excreta collections were transferred to individual

metabolic cages. Urine and feces were collected daily for 10 days, then for 5 consecutive days at 1, 2, and 4 months. Exhaled air from two of the metabolic cages was collected for the same times. Six rats from each group were sacrificed at 3, 14, 30, 61 and 121 d after intratracheal instillation to collect lungs and bronchial lymph nodes (BLNs) for analyses of their ^{3}H content. At the scheduled sacrifice times, the six rats were anesthetized with halothane and exsanguinated using cardiac puncture.

Sample Processing

The samples to be analyzed for ^{3}H included lung, BLNs, urine, feces, and exhaled air. These samples were segregated into three batches and each batch included its own set of quench correction standards. The samples to be analyzed for ^{3}H were prepared for liquid scintillation counting using a base digestion procedure as described previously (Cheng et al., 1999). Eight ml of liquid scintillation cocktail (Ready Value, Beckman Instruments Inc. Fullerton, CA) and 3 ml of deioned water were added to each vial. The prepared samples were stored at room temperature for about 48 hours to stabilize samples, then counted for tritium activity by liquid scintillation counting in a Packard 2500 TR Liquid Scintillation Analyzer (Packard Instrument Company, Downers Grove, IL). Quench correction standards were prepared along with each batch of tissue and excreta samples using non-radioactive lung tissue or feces samples from rats and the same base digestion procedure as what described above. Quench correction standards were spiked with known amounts of ^{3}H in tritiated water.

β-ray Self-absorption in Ti-T

Because the tritium betas are of such low energy (maximum 18.6 keV, and average 5.7 keV), beta self-absorption within a metal tritide particle is significant. Therefore, it was necessary to make a correction of the self-absorption of beta activity and its energy using a correction factor, defined as a self-absorption factor (SAF) that is the fraction of beta particles escaping from their absorber (Kropf et al, 1998). A numerical method was developed to evaluate the self absorption of tritium. Metal tritide particles were assumed to be spherical in shape as described in another paper (Kropf et al., 1998). The results indicated that the self absorption factor for titanium tritide particles with CMD of 0.95 μm (σ_g = 1.93), the SAF is 0.21. This means that about 80% of tritium beta particles are absorbed in the fine powders of titanium tritide. This is very close to the experimental value of 0.23± 0.05. The activity in the samples detectable by liquid scintillation counting was apparent activity. Apparent activity was corrected to total activity using the self absorption factor of 0.23. This factor was applied in this biokinetic modeling so that real tritium activity was estimated for lung, BLNs, and feces.

Biokinetic Modeling

Based on results of experiments with rats intratracheally instilled with titanium tritide particles, and on a self-absorption factor for beta particles determined by a numerical method, a biokinetic model was developed as described in Cheng et al. (1999). The simulation model was used to help evaluate the biokinetics of the instilled tritide and make prediction about the consequence of inhalation exposures to titanium tritide. The model included compartments of lung, bronchial lymph nodes (BLNs), gastrointestinal tract (GI tract), blood and carcass, urine, feces, and exhaled air. For particulate materials deposited in the lung, absorption into blood is depicted as a two-stage process. First, constituents of the particles elute from the particles or the particles dissolve, then the dissolved material passes into blood (ICRP-66, 1994). Hence the particulate and dissolved phases of ^{3}H in the lung , BLNs, and GI tract were placed into separate compartments. Compartment I denoted the particulate phase of tritium whereas Compartment II denoted the dissolved phases of tritium or tritiated water. In this way, the model could evaluate contributions to absorbed dose resulting from tritium in particles and tritium in soluble form.

Clearance of tritium originally bound with the metal tritide particles from the lung occurs as a result of three competitive processes: mechanical clearance via ciliary movement of particles from the lung into GI tract, mechanical clearance by the particle translocation to BLNs, and dissolution of the tritide particles in the lung. These processes are all described by time-dependent rate functions. Following both mechanical clearance processes, dissolution of the tritide particles also occurs in the GI tract and BLNs. Once the particles have been dissolved in lung, BLNs, and GI tract, tritium is absorbed into blood and then distributed over the carcass through the circulatory system. The tritide particles contained in GI tract are mostly excreted into feces. Tritium absorbed into blood is either excreted via urine or feces or deposited in the body. Tritium gas(HT) and tritiated water vapor evolved in lung and other tissues may be eliminated in exhaled air. The kinetics software, SAAM II^{TM} (University of Washington, Seattle, WA), was employed for this modeling purpose. Experimental data were

used to determine the best-fitted transfer rate functions or constants.

RESULTS AND DISCUSSION

Tritiated Pump Oils

Tritium in Urine

The tritium elimination in urine following contamination of tritium from instilled tritiated pump oil is shown in Figure 1. The average biological half-life, which are a function of tritium distribution and retention in the body, are 0.7 ± 0.2 d and 3.9 ± 0.8 d for the short-term component of OBT and HTO, respectively. The average biological half-life for the long-term component is 14.4 ± 2.2 for OBT and 18.4 ± 3.6 for HTO. The time-integrated amount of tritium in urine was about 12% of the instilled activity in the lung. The kinetics of tritium in urine also showed that more than 80% of the total tritium in urine was eliminated with a biological half-life of four days or less, indicating that the majority of tritium eliminated through urine clears quickly. The kinetics and pattern of tritium excretion are, in general, comparable for tritium contamination from percutaneous absorption of tritiated pump oil (Trivedi 1995).

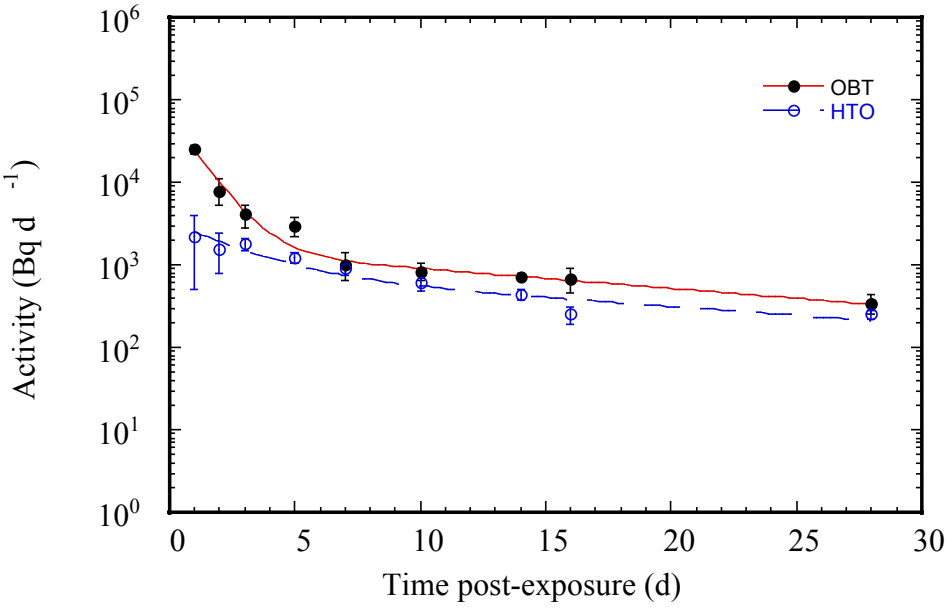

Figure 1. Urinary excretion of tritium from animals exposed to tritium-gas-contaminated vacuum pump oils.

Tritium in Feces

Similar to tritium-in-urine, the kinetics of tritium-in-feces is best described by the sum of two exponential functions (Figure 2). The biological half-life for the fast clearing component of tritium in feces is 1.0 ± 0.3 and 3.9 ± 1.1 for OBT and HTO. Here, the slow clearing component of tritium in feces is 8.3 ± 1.8 for OBT and 9.4 ± 1.6 for HTO. The calculated cumulative activity of tritium in feces is invariably higher than the tritium in urine. Of the total tritium instilled into the lung, about 20% of the total activity was accounted for by elimination in feces. About 95% of the excreted activity in the feces were in the form of OBT.

The cumulative excretion of tritium-in-urine and feces indicates that up to 30% of the instilled activity can only be accounted for by excretion from the body. Less than 10% of the activity excreted from the body would be in the form of HTO. This means that a large fraction of (70%) of the instilled activity was unaccounted for, and assumed to be exhaled as being highly volatile.

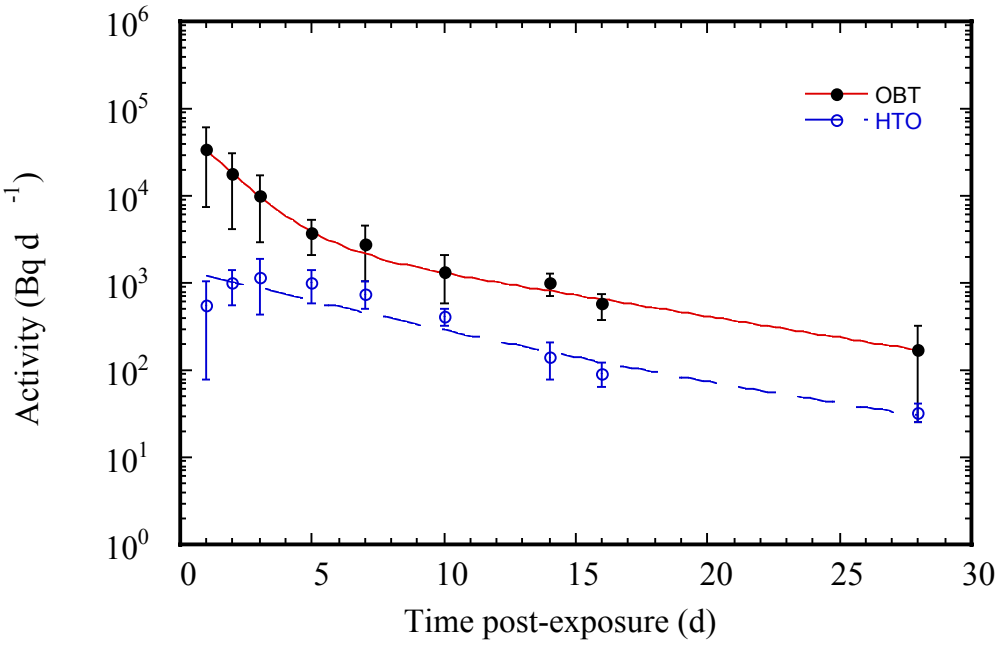

Figure 2. Fecal excretion of tritium from animals exposed to tritium-gas-contaminated vacuum pump oil.

Tritium in the Body

The total accountability for tritium in the whole-body was examined to determine whether there is a significant amount of long-term storage of tritium in the body. The tritium activity balance is determined by summing the tritium activity in the lung, organs (brain, intestine, liver, lung, kidneys, stomach, testis), blood and carcass after an exposure. The tritium inventory in the body at 0 (30 min post-exposure), 1 d, 3 d, 5 d, 7 d, 14 d and 28 d post-exposure was investigated.

The distribution of tritium in the exposed animals following instillation of tritiated pump oil is shown in Figure 3. Tritium inventory in the body showed that, 30 min post-exposure, most tritium was retained in the lung (96%). The carcass and soft tissues had only 2% of the total tritium in the body. Up to 3 d post-exposure, most of the instilled activity (85%) was retained in the lung. The migrated activity during this period was retained mostly in the soft tissues (~12%) (Fig. 3). The majority of the activity was retained in the liver (60%), intestine (15%) and kidneys (10%), while remaining organs examined has low tritium retention.

The tritium activity balance showed that at 5 d post-exposure, the tritium activity in the lung dropped sharply (Fig. 3), and now most of the retained activity in the body was in the carcass (80%). This distribution pattern in the body was examined for the remaining period of the study (7-28 d post-exposure), indicating that the long-term storage of the tritium in the body resides in organs or tissues that have less significance for radiation protection.

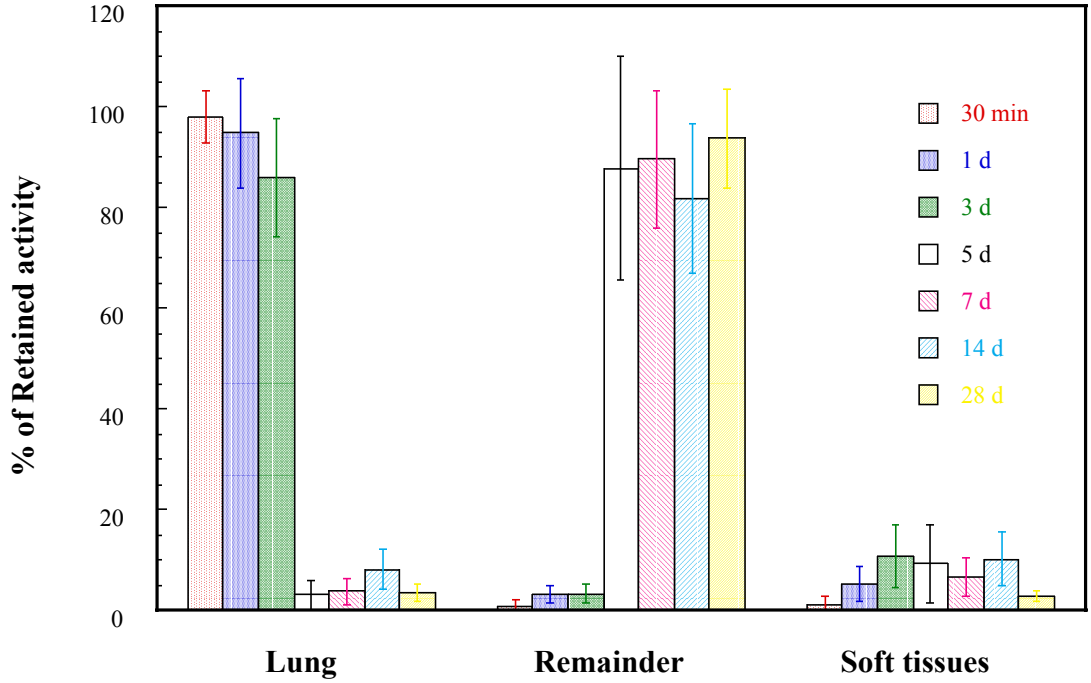

Figure 3. Distribution of tritium in the body after instilling tritium-gas-contaminated vacuum pump oil.

Table 1. The concentrations of OBT and HTO in the body and contributing radiation dose-rate from tritium.

Body/organ	Days post-exposure (d)	Tritium concentration❁ (Bq g^{-1} wet tissue)		Dose-rate✿ ($nGy\ s^{-1}$)
		Total tritium	OBT	Total (OBT)*
Lung	30 min	9.3×10^5	8.9×10^5	846 (96%)
	3 d	4.2×10^4	2.9×10^4	38 (69%)
	28 d	6.5×10^3	1.6×10^3	6 (25%)
Carcass	30 min	2.2×10^2	1.8×10^1	0.2 (8%)
	3 d	5.9×10^4	3.7×10^4	54 (63%)
	28 d	9.2×10^3	2.9×10^3	8 (32%)
Soft tissue	30 min	4.1×10^1	1.8×10^1	0.04 (44%)
	3 d	4.9×10^4	3.2×10^4	45 (65%)
	28 d	8.8×10^2	3.3×10^2	0.8 (38%)

❁Average of three independent animals.

✿The dose-rate is computed by the multiplication of activity concentration of tritium in tissues with 9.1×10^{-4} nGy per Bq g^{-1} s^{-1}. The factor, 9.1×10^{-4} nGy s^{-1} is for the absorbed dose-rate per unit activity concentration of tritium in tissue (Bq g^{-1}). The effective average energy for tritium beta-ray is 0.0057 MeV per decay. The homogeneous distribution of tritium is assumed.

*OBT contribution is expressed as a percentage of total dose, which is shown in parentheses.

Tritium Concentrations

The tritium concentrations in the body are summarized in Table 1. Also shown are the dose-rates to the lung, soft tissue and the carcass from the retained tritium. The maximum tritium concentration (~1 MBq) was at the point of instillation in the lung immediately after exposure (30 min post-exposure). About 96% of the tritium concentration in the lung were in the form of OBT. The uptake of tritium from the exposed lung into the body

continued up to 3 d post-exposure, and then tritium concentrations in the body declined gradually. At 3 d post-exposure, the tritium concentrations in the lung, soft tissue and carcass were about the same (0.01 MBq), and between 60% and 70% of the activity was as OBT. By 28 d post-exposure, the tritium concentration in the lung was less than the carcass. The tritium concentrations in the soft tissue was much lower (Table 1). Now OBT concentration was between 25% and 40% of total tritium concentration. The lung had the highest dose-rate 30 min post-exposure (Table 1). The ratio of dose-rate between the lung and the body was 4000-times.

Titanium Tritide Particulates

Retention of Tritium in Organs

Comparison of experimental retention and model prediction of ^{3}H in lung as a function of time is shown in Fig. 4. The figures include the simulation curves that resulted from biokinetic modeling for the total calculated activity and apparent activity. Apparent activity consists of two parts, tritium in the particles (Compartment I), and dissolved tritium present in the organ (Compartment II). Apparent activity corresponds to the experimental data from scintillation counting. Total activities on the lung and BLNs are higher than apparent activities because of self-absorption of beta in the particulate phase with a factor of 0.23. The clearance of tritium from the lung could be described as the sum of two exponential terms. The rapid clearance of ^{3}H after instillation was dominated by the first component with the half-life of 0.81 d. Seventy percent of the initial lung burden was removed from lungs by this faster term in the first 5 days. The initial solubility of the particles in the lung and mechanical clearance from the lung to the GI tract contributed to the rapid clearance during the first five days. Then the biokinetic process was dominated by much slower clearance, with half-life of 66 d. The rest of the lung burden would eventually be cleared by the slower clearance process.

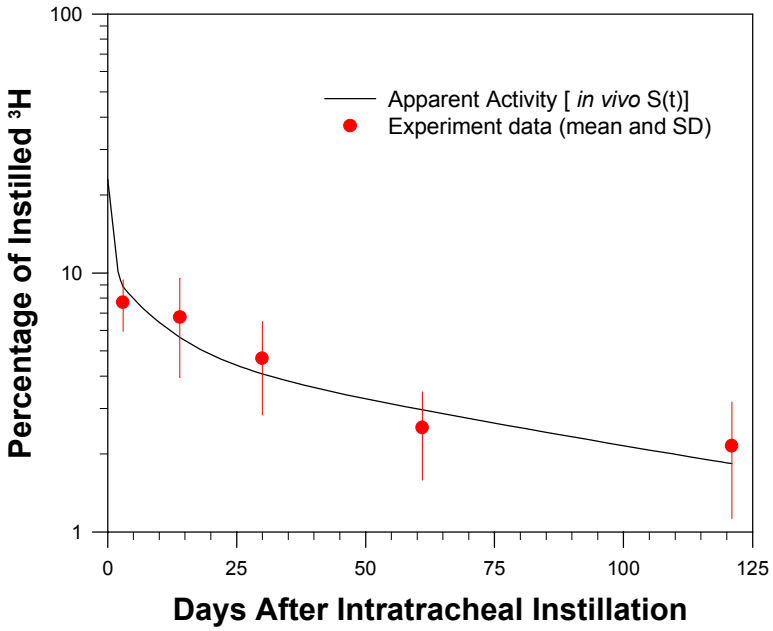

Figure 4. Retention of tritium in the rat lung after instilling titanium tritides

As a result of absorption of tritium into blood following dissolution or elution of the tritide particles in the lung, tritium was distributed in blood and redistributed to soft tissue. The retention of tritium in blood and carcass could be described in three phases. First, the amount of tritium increased rapidly due to the uptake of tritium. As tritium was excreted into urine and exhaled through air, tritium in blood and carcass reached its maximum, 37% of the initial lung burden at 3 days after instillation. Then, from 3 to 50 days, retention curves was characterized by a faster clearance that resulted from urinary excretion. Thereafter, a slower clearance process occurred in the remaining time period of the study.

Excretion of Tritium

The excretion of tritium in the urine and feces is shown in Figs. 5 and 6, respectively. Tritium in the

exhaled air is not shown. Experimental data for the exhaled air were average values over the collection times. Its activity was a sum of tritium evolved as tritiated water and tritium gas.

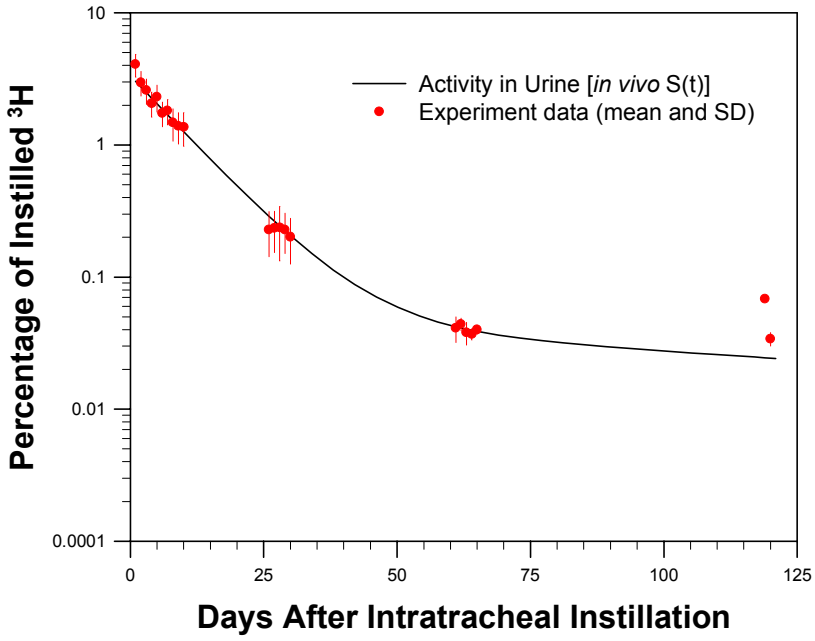

Figure 5. Urine excretion of tritium from inhaled titanium tritide particles.

The urinary excretion rate reflects the dissolution-absorption rate of tritium in the lung and the blood level of tritium. The fecal excretion rate indicates the mechanical clearance from the lung to the GI tract. The excretion of tritium in urine and feces showed similar patterns and could be described by a sum of two exponential components which were combined together to represent the kinetic process of uptake and excretion. Both excretion rates reached their maximum for the first samples obtained in the first day after instillation, decreasing monotonically thereafter. Although the excretion rates showed no significant difference between urine and feces, the rate through urine was slightly slower than that via feces. The former was characterized by the half-lives of 7 days for the fast term and 115 days for the slow term; these values for feces were 3 and 63 days. The modeling result of the activity in exhaled air could be expressed as two terms with half-lives of 6 and 80 days, respectively. Therefore, a common patterns seems to represent tritium excreta through urine, feces, and exhalation. All three excretion pathways after instillation of titanium tritide into rat lungs had similar half-lives ranging from 3 to 7 days for the fast term and from 65 to 115 days for the slow term.

About 30% of the instilled tritide were eliminated from the body within the first 5 days. In this time period, the cumulative excretion from the urine was 13% of the initial lung burden due to dissolution-absorption of tritium. Similarly, 10% of the tritium was excreted in the feces mainly by means of mechanical clearance from the lung into the GI tract. At 25 days after exposure, approximately 63% of the initial lung burden had been cleared. At 121 days after instillation, 82% of the initial lung burden of ^{3}H had been eliminated, of which 37% was excreted via urine, 29% via feces, and 16% through exhaled air. Thus, tritium excretion through urine and feces were about equal for the titanium tritide. Although tritium elimination by exhalation was less than in urine and feces, its effect on total clearance cannot be neglected.

The dose conversion factor of inhaled titanium tritide particles was calculated based on the ICRP Publication 66 respiratory tract model using LUDEP computer code. A conservative estimate gave 4.5 Sv /Bq assuming AMAD of 1 μm and geometric standard deviation of 2.5. The dissolution rate was assumed to be type M (moderate) for titanium tritide.

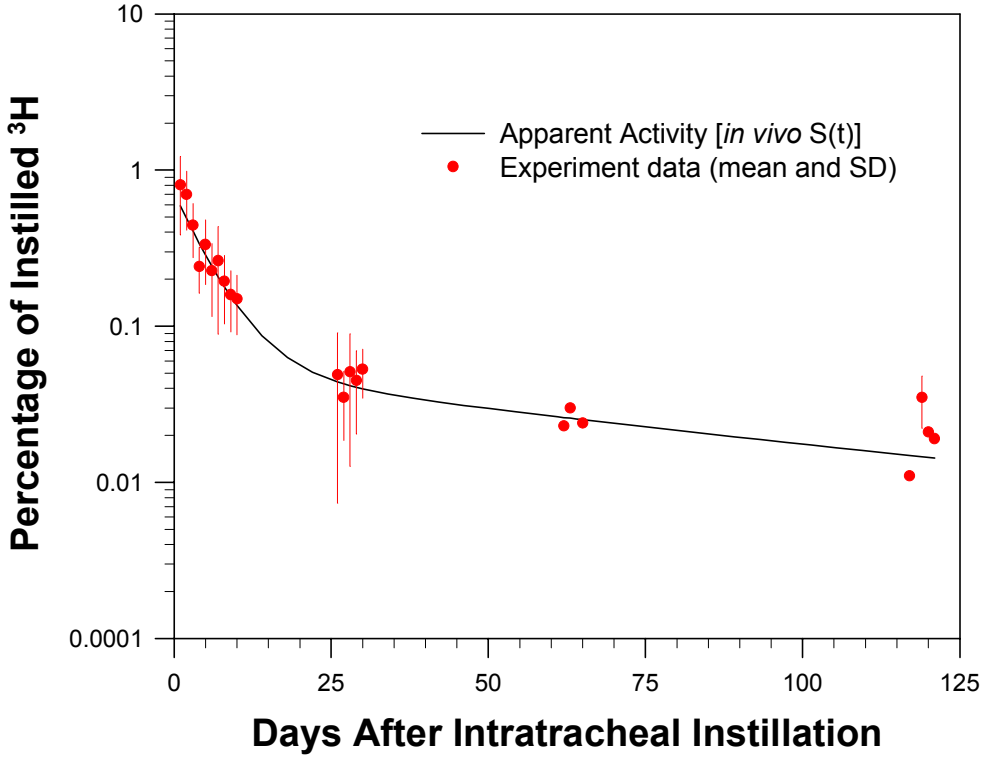

Figure 6. Fecal excretion of tritium from inhalation of titanium tritide.

CONCLUSIONS

Our biokinetic and distribution data for tritium for the inhalation of tritiated aerosols (i.e., metal tritides and tritiated pump oils) are significantly different from the dynamics of tritium following HTO intakes. The tritium distribution in the lung and body was chiefly in the form of OBT for tritiated pump oils exposure; whereas for metal tritides, tritium was retained in the lung as particulate aerosols. In both cases, tritium excretion in the urine, feces, and expired air were important routes. For radiological protection purposes, the knowledge of the long-term retention of tritium in the lungs and parts of the body is useful, and might be useful in developing better dose models for inhalation of tritiated aerosols. The radiation dose will be a function of not only particle size and dissolution rate, but also on knowing the chemical forms of the tritium in respired aerosols.

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