Draft Report

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ADVISORY BOARD ON RADIATION AND WORKER HEALTH

TASK 3: REVIEW OF NIOSH/ORAUT PROCEDURES AND METHODS USED FOR DOSE RECONSTRUCTION

Review of ORAUT-OTIB-0066
Calculation of Dose from Intakes of Special Tritium Compounds

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Prepared by

S. Cohen & Associates
1608 Spring Hill Road, Suite 400
Vienna, VA 22182

Saliant, Inc.
5579 Catholic Church Road
Jefferson, Maryland 21755

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Task Manager: _____ Date: _____ Stephen L. Ostrow, PhD	
Project Manager: _____ Date: _____ John Mauro, PhD, CHP	

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EXECUTIVE SUMMARY

INTRODUCTION

This report presents a technical review of *Calculation of Dose from Intakes of Special Tritium Compounds*, ORAUT-OTIB-0066, Rev. 0, April 26, 2007 (ORAUT 2007, hereafter referred to as "the OTIB" or "OTIB-0066"). The review is limited to those aspects of this Oak Ridge Associated Universities Team Technical Information Bulletin (OTIB) judged by SC&A to be potentially significant to dose reconstruction, and is not a line-by-line check of data sources and calculational results. This review also considered the availability of information and data to effectively implement this procedure during individual dose reconstructions.

The most commonly encountered exposure pathway to tritium in dose assessments is when the tritium is in the form of tritiated water (HTO), in which a tritium (H-3) atom replaces one of the light hydrogen (H-1) atoms. NIOSH presents its methodology to determine the dose from the intake of tritiated water in ORAUT-OTIB-0011, *Tritium Calculated and Missed Dose Estimates* (ORAUT 2004) and recapitulates it in OTIB-0066. Section 2.0 of OTIB-0011 states the following:

> *The model used for assessing effective dose (ED) from intakes of tritiated water is that of the International Commission on Radiological Protection (ICRP). The current model is described in detail in ICRP Publication 56* [ICRP 1989] *with the current dose conversion factor published in ICRP Publication 68* [ICRP 1994b].
>
> *Tritiated water is assumed to be completely and instantaneously absorbed into the systemic circulation whether taken in by inhalation, ingestion, or absorption through the intact skin. In a relatively short time, tritiated water equilibrates with the body water and thereafter the concentration in all body fluids, including urine, is assumed to be equal. For this reason, urine measurements of tritium are considered a direct measurement of the concentration of tritium in body water.*

Whatever the merits of the guidance provided by OTIB-0011, however, it does not, provide guidance for assessing dose due to intake of other tritium compounds that may be encountered in certain circumstances; these situations are treated in OTIB-0066, which "provide[s] guidance on how to use urine bioassay data to calculate the best estimates of the annual organ doses for intakes of bound to organic compounds (organically bound tritium - OBT) and tritium in a metal matrix (stable metal tritide - SMT)" (ORAUT 2007, pg. 5).

REVIEW METHODOLOGY

SC&A reviewed ORAUT-OTIB-0066 (ORAUT 2007) in a systematic manner, following the OTIB's organization and the guidance of SC&A's review procedure, *A Protocol for the Review of Procedures and Methods Employed by NIOSH for Dose Reconstruction* (SC&A 2004). Section 1 of this review report summarizes the methodology NIOSH presents in the OTIB sections, Section 2 presents SC&A's assessment and recommendations, and Section 3 contains a

checklist taken from SC&A 2004. The checklist has sections concerned with the following issues:

- The degree to which the procedure supports a process that is expeditious and timely for dose reconstruction
- Whether the procedure provides adequate guidance to be efficient in instances where a more detailed approach to dose reconstruction would not affect the outcome
- The extent to which the procedure accounts for all potential exposures, and to ensure that resultant doses are complete and based on adequate data
- Whether the procedure provides a consistent approach to dose reconstruction, regardless of claimant's exposures by time and employment locations
- Whether the procedure is fair and gives the benefit of the doubt to the claimant
- Whether the procedure adequately accounts for the uncertainty of dose estimates
- Whether the procedure strikes a balance between technical precision and process efficiency

This review also discusses the feasibility of implementing the procedure, since the process developed in OTIB-0066 serves as a supplement to dose reconstruction guidelines provided in individual site profiles and special exposure cohort (SEC) evaluation reports.

SUMMARY OF FINDINGS AND COMMENTS

SC&A's review of the OTIB is presented in Section 2; the review produced six Comments. SC&A divided these Comments into four Findings and two Observations, where the former are felt important by SC&A and should be addressed by NIOSH, while the latter comprise items of agreement and support for the OTIB.

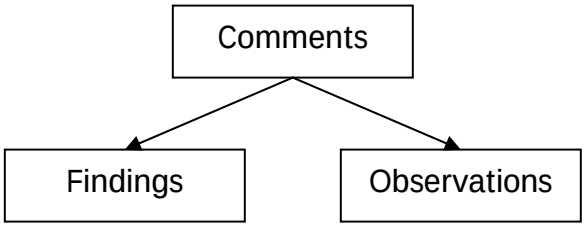

Table 1 collects the Comments, notes where they are found in this document, and indicates whether each is a Finding or an Observation.

Table 1: Comment Summaries

Comment Number	Finding Number	Summary Description
1 (Sect 2.4.1)	O (a)	**Issue 1:** SC&A has found that ORAUT-OTIB-0066 is clear and consistent with International Commission on Radiological Protection (ICRP) Publications 56 (ICRP1989) and 67 (ICRP 1993), ORAUT-OTIB-0011, and open literature (Balonov et al. 1984, 1995; Cheng et al. 1997, 1999, 2002a, 2002b; Inkret et al. 2001; Zhou and Cheng 2003; Hodgson et al. 2004; and Zhou and Cheng 2004).
2 (Sect 2.4.1)	O	**Issue 2:** The procedure provides adequate and valid technical information and guidance to evaluate the dose due to intake of HTO, OBT, and SMT, applying the ICRP models in IMBA.
3 (Sect 2.4.2)	1	**Issue 3:** The recommendation given in ORAUT-OTIB-0066 to assess dose due to intake of OBT is not claimant favorable. The OTIB recommends the use of the methodology given in ORAUT-OTIB-0011 to calculate doses from intakes of OBT to all organs and tissues. The dose coefficient for OBT given in ICRP Publication 78 (ICRP 1997), which is 1.52×10^{-7} mrem/pCi (see Table 2); the one derived applying the biokinetic model for OBT (ICRP 1989; ICRP 1993; ICRP 1995; and ICRP 1997) using the AIDE computer code, which is 1.52×10^{-7} mrem/pCi (see Table 2); and the one derived using the methodology given in ORAUT-OTIB-0066 (ORAUT 2007), which is also 1.52×10^{-7} mrem/pCi (see Table 1), are 1.4 times higher than the one obtained applying the methodology given in ORAUT-OTIB-0011 (ORAUT 2004), which is 1.08×10^{-7} mrem/pCi.
4	2	**Issue 4:** Bounding techniques proposed in OTIB-0066 cannot be effectively developed and applied without some basic understanding of the special tritium compounds handled, the quantities of material, the locations and time periods of potential exposure, and the physical behaviors of tritium compounds in the environment (e.g., conversion to HTO, formation of rust) to correctly characterize tritium exposure.
5	3	**Issue 5:** OTIB-0066 does not ensure that resultant doses are based on adequate monitoring data.
6	4	**Issue 6:** The procedure provides no guidance on how to distinguish between intakes of STCs, elemental tritium, and/or tritiated water which occur simultaneously or overlap.

Note: (a) "O" denotes observation.

1.0 SUMMARY OF ORAUT-OTIB-0066 METHODOLOGY

The OTIB's development of the guidance given to dose reconstructors in estimating doses from exposure to tritium-containing compounds proceeds through a number of steps, which were examined by SC&A to determine whether they are based on accepted, sound scientific or engineering practice; reasonable; clearly stated; adequately documented; and claimant favorable. The following summarizes the OTIB section-by-section for convenience.

1.1 ORAUT-OTIB-0066 MODELING

OTIB-0066 treats three scenarios of tritium intake into the body and subsequent exposure, where the tritium taken in is either in the form of tritiated water (HTO), a stable metal tritide (SMT), or organically bound tritium (OBT). Section 3.0 discusses the biokinetic models employed by NIOSH in these cases. Figures 3-1, 3-2, and 3-3 of the OTIB display biokinetic block diagrams for the three situations respectively. The block diagrams are reproduced here and the models will be briefly described. It should be noted that in all three biokinetic models the water compartment (with a biological half life of about 10 days) feeds with a 47% split directly into the bladder for elimination as urine and with a 53% split (essentially, tritium in the blood) into a compartment marked "other." The latter represents, in effect, a feedback pathway to the transfer compartment for another pass through the body.

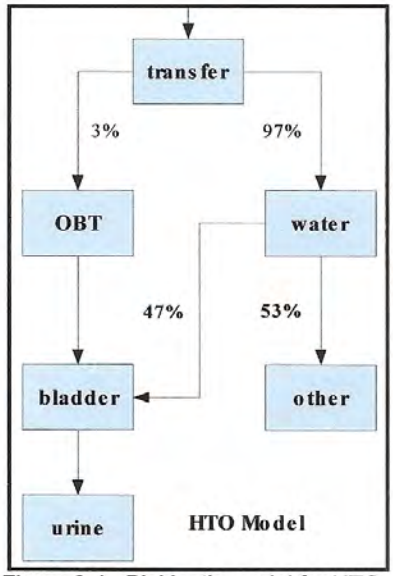

Figure 3-1. Biokinetic model for HTO.

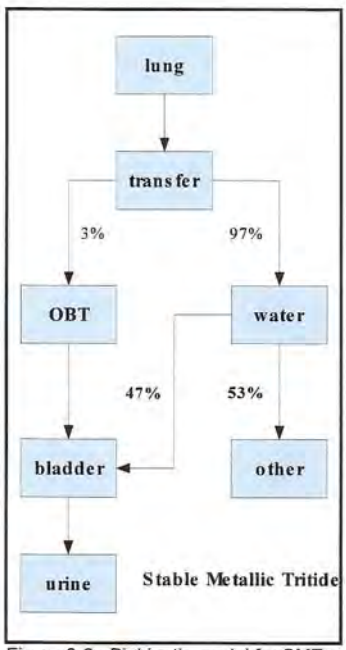

Figure 3-2. Biokinetic model for SMTs.

Tritiated Water - HTO

Figure 3-1 of the OTIB shows the compartmentalized NIOSH biokinetic model for tritiated water, which is implemented in IMBA (Interactive Modules for Bioassay Analysis) software. It should be noted, that the model includes a compartment for organically bound tritium (OBT), which "represents tritium that is incorporated into carbon compounds and is retained with a half-life of 40 d as described in the ICRP carbon biokinetic model [ICRP 1993]" (ORAUT 2007, pg. 5). The model assumes that 3% of the HTO that is taken in goes to the OBT compartment, 97% remains in the water compartment, and all of the tritium in the OBT compartment is eventually excreted from the bladder as HTO.

Stable Metal Tritide - SMT

Figure 3-2 of the OTIB incorporates a lung model to reflect inhalation of stable metal tritides, which are certain metallic compounds (containing, for example, titanium, hafnium,

palladium, lithium, uranium, or zirconium) that can bind and retain tritium interstitially in their crystalline structures (in fact, various metal hydrides have been considered to store hydrogen fuel for hydrogen-powered automobiles). An SMT may be inhaled into the lungs as an aerosol and slowly released there "as the particle of the SMT dissolves and the tritium diffuses out of the particle" (ORAUT 2007, pg. 6). The tritium is assumed to displace hydrogen in water molecules and form HTO, which then can be treated by the HTO biokinetic model. "The SMT biokinetic model shown in Figure 3-2 is therefore the HTO biokinetic model with an ICRP Publication 66 Human Respiratory Track Model feed compartment [ICRP 1994a]" (ORAUT 2007, pg. 6). As in the HTO model, it is assumed that 3% of the HTO goes to the OBT compartment.

Solubility of the SMT in the lungs depends on the type, size, and shape of the metal particles. The OTIB states that, "of the SMTs discussed in the literature, titanium and zirconium appear to be best described as type M, whereas hafnium is best described as type S…Other tritides that consist of more reactive metals such as lithium and uranium are expected to be best described as type F" (ORAUT 2007, pp. 6–7).

Organically Bound Tritium - OBT

The biokinetic model for HTO (illustrated in OTIB Figure 3-1) contains an OBT compartment, where some (assumed 3%) of tritium in the HTO taken in by the person enters into carbon compounds in the body. In the biokinetic models for OBT (OTIB Figure 3-3), however, the tritium enters the body already in the form of a carbon compound. The OBT model follows ICRP guidance and "assumes that 50% of the OBT compounds taken into the body are immediately converted to HTO and the other 50% remain OBT" (ORAUT 2007, pg. 7).

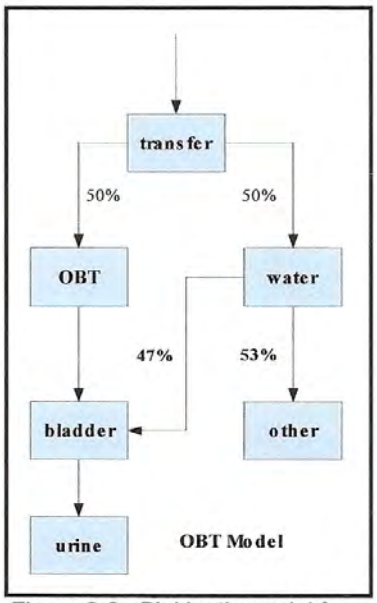

Figure 3-3. Biokinetic model for OBT.

1.2 ORAUT-OTIB-0066 DOSE CALCULATIONS

Section 4.0 of the OTIB discusses how doses are calculated by applying the three biokinetic models and estimates the accuracy of the methodologies. It begins by stating that, "intakes of HTO are usually evaluated using an isotopic dilution technique…, which is based on the following assumptions:"

1. *HTO is metabolized like H_2O.*
2. *The metabolic space of H_2O is the 42 L of body water.*
3. *The concentration of HTO in the urine is the same as the concentration of HTO in the body water* (pp. 8–9)

Hence, under these assumptions, "the quantity of HTO in the body at any given time is simply the concentration of HTO in the urine times the volume of body water, and the dose to the soft tissues of the body is proportional to the area under the excretion curve" (ORAUT 2007, pg. 9).

That area can be calculated in several different ways. The OTIB states that "the standard method for calculating the dose from intakes of tritiated water given in ORAUT-OTIB-0011, *Tritium Calculated and Missed Dose Estimates* (ORAUT 2004), uses three different techniques to calculate the area under the excretion curve" (ORAUT 2007, pg. 9). These techniques are called Type 1, Type 2, and Type 3.

A Type 1 area calculation is a linear approximation between two urinary excretion measurements when they are taken less than 40 days apart. A Type 2 area calculation uses an exponential extrapolation from one measurement point to the next, when they are more than 40 days apart. Application of the Type 2 method presumes that the fact that the two measurements were more than 40 days apart means that the person did not work with tritium; a Type 2 calculation gives a lower area (dose) than a Type 1 calculation. A Type 3 calculation extrapolates exponentially to infinity (in time) after the last measurement.

OTIB-0066 notes that adding the OBT compartment into the biokinetic model (shown in OTIB Figure 3-1) does not conform to assumption 2 listed above, since the tritium in the OBT compartment is not uniformly distributed in the body. The error, potentially resulting in a dose underestimation, is slight, however, since it is assumed that only 3% of the HTO goes into the OBT compartment; OTIB-0011 adjusts the dose conversion factor upward, rendering dose calculations to systemic organs conservative (an overestimate) for Type 1 area calculations. OTIB-066 extends its claim of conservatism for Type 1 calculations to the case where SMTs are present in the lungs, "because SMTs in the lungs are essentially a longer term feed compartment for HTO going to the systemic organs" (ORAUT 2007, pg. 9). However, the same is not true for intakes of OBT compounds, which are not uniformly distributed in the body-water space. In this case, "a Type 1 calculation underestimates systemic dose by approximately 30%" (ORAUT 2007, pp. 9–10). The OTIB maintains that this may not be very significant in practice as many "occupational sources of OBT (e.g., pump oil) tend to have a significant OBT component" (ORAUT 2007, pg. 10).

Such a claimant-favorable conclusion, however, cannot be reached for Types 2 and 3 calculations of the area under a tritium excretion curve, which assume a 10-day half-life, although "portions of the urinary excretion curves have half-lives that are significantly longer than 10 d" (ORAUT 2007, pg. 10). In these cases, calculated doses to systemic organs from OBT and SMT compounds will be underestimated. The OTIB states: "As an example of a worst-case scenario, if a single urine result is evaluated using a Type 3 calculation (i.e., assuming it is HTO) and is in fact from an intake of Type S tritide, the dose for the tail (not the total dose) will be underestimated by a factor of about 6" (ORAUT 2007, pg. 10).

> *The degree to which the total systemic dose is underestimated for OBT and SMT depends on the fraction of the systemic dose from Type 2 and 3 calculations. For example, if the entire systemic dose up to the time of diagnosis is determined with Type 1 calculations, the systemic dose can be taken to be accurate regardless of the material. On the other hand, if most of the systemic dose is determined with Type 2 and 3 calculations and the material is Type S SMT, the systemic dose could be significantly underestimated* (ORAUT 2007, pg. 10).

The OTIB cautions that the dose to the lung can be seriously underestimated (by several orders of magnitude) if ORAUT-OTIB-0011 methodologies are applied. Table 4.1 of OTIB-0066 (reproduced here as Table 2) presents calculated doses for several cases for an assumed 1-pCi acute intake of tritium compounds (these doses per unit source are sometimes referred to as dose coefficients). It is seen that while the OTIB-0011-calculated results agree fairly well with the IMBA soft tissue dose model and the IMBA lung dose model for HTO and OBT intake cases, they are very much lower than the corresponding IMBA lung dose results for SMT intakes.

Table 2. Comparison of Calculated Doses for a 1-pCi Acute Intake of Tritium Compounds (mrem)

	ORAUT-OTIB-11 Soft tissue dose	IMBA Soft tissue dose	IMBA Lung dose
Type S SMT	6.58E-10	6.30E-10	3.86E-06
Type M SMT	7.28E-09	6.93E-09	5.52E-07
HTO	7.11E-08	6.76E-08	6.76E-08
OBT	1.08E-07	1.52E-07	1.52E-07

Source: ORAUT 2007, Table 4.1

1.3 ORAUT-OTIB-0066 RECOMMENDATIONS

Section 5.0 of the OTIB presents recommendations for determining doses from tritium intakes via different pathways; these are summarized as:

(1) "Selection of the appropriate tritium compound in an intake evaluation must usually be based on process knowledge of the source terms in the workplace."

(2) "The methodology in ORAUT-OTIB-0011 (ORAUT 2004) can be used without modification to calculate doses from intakes of HTO and OBT to all organs and tissues."

(3) The OTIB-0011 methodology can be used to calculate systemic doses from intakes of SMT "if the majority of the dose is calculated with the Type 1 method… On the other hand, if the majority of the systemic dose is calculated with Type 2 or 3 methods, IMBA should be used to calculate the dose."

(4) "If the observed urinary excretion of tritium is deemed to be the result of intakes of SMT and the dose to the lung or gastrointestinal (GI) tract is needed, the urinary excretion data must be evaluated with IMBA in the same fashion as any other radionuclide."

(5) "If the metal substrate of the SMT is not known, type S solubility should be assumed."

2.0 SC&A ASSESSMENT OF ORAUT-OTIB-0066

2.1 INTRODUCTION

ORAUT-OTIB-0066 recommends that the dose reconstructor assume solubility classes of Type M or S for metal tritides (SMT), depending on the compound. This guidance is consistent with recent data available in the literature (Balonov et al. 1984, 1995; Cheng et al. 1997, 1999; 2002a, 2002b; Inkret et al. 2001; Zhou and Cheng 2003; Hodgson et al. 2004; Zhou and Cheng 2004).

The OTIB provides guidance to assess the dose applying ICRP systemic models and lung retention parameters for HTO, OBT, and SMT, using the IMBA computer code. Section 4.0 calculates and compares doses obtained using its approach to that determined using the ORAUT-OTIB-0011 model (ORAUT 2004), which was developed to be applied to doses resulting from intake of HTO. Table 4.1 of OTIB-0066 (reproduced here as Table 1) presents calculated doses for an assumed 1-pCi acute intake of tritium compounds. The OTIB recommends the use of the methodology in ORAUT-OTIB-0011 to calculate doses from intakes of HTO and OBT to all organs and tissues and for systemic organ doses due to intake of SMT. However, the dose to the lung or gastrointestinal (GI) tract must be evaluated with IMBA as for any other radionuclide.

2.2 REVIEW OF DATA AVAILABLE IN THE OPEN LITERATURE

2.2.1 Tritiated Water (HTO)

ICRP Publications 56, 67, 71, and 78 (ICRP 1989; ICRP 1993; ICRP 1995 and ICRP 1997) assume that for tritiated water there is 100% deposition in the respiratory tract, with instantaneous (Type V) absorption over the total body. Those publications recommend a two-component model for predicting the behavior of tritium that enters the human body as HTO, in which 97% of the tritium is assumed to be eliminated, with a biological half-life of ten days and that 3% became organically bound and is eliminated with a biological half-life of 40 days. However, it is recognized that a small fraction of the activity might be retained for much longer periods. This third compartment would represent tritium incorporated into structural or other tissue components with very slow turnover. The second compartment, plus any possible third compartment, is considered likely to contribute about 10% to the committed dose (ICRP 1989).

2.2.2 Organically Bound Tritium (OBT)

ICRP Publications 56, 67, 71 and 78 (ICRP 1989; ICRP 1993; ICRP 1995 and ICRP 1997) assume for organically bound tritium (OBT), 100% deposition in the respiratory tract, with instantaneous (Type V) absorption. Those publications recommend a default model for all unknown compounds containing OBT, which assumes that 50% of the OBT entering the systemic compartment would be metabolized to HTO and lost with a biological half-life of ten days, and that the remaining 50% would bond with carbon and be removed with a biological half-life of 40 days. The concentration in urine is assumed to be the same as in total body water. Urinary excretion will not result in a significant additional dose to the bladder wall, which is assumed to receive the same dose as other tissues.

2.2.3 Metal Tritides (SMT)

ICRP Publication 71 (ICRP 1995) states that:

> *(91) Tritium could be released to the environment in particulate form, and studies of the solubility of solid tritiated compounds have been conducted. For example, the results of an in vitro study of the dissolution of 1 μm (count median diameter) titanium tritide powder in serum ultrafiltrate were consistent with the assignment to Type M (Cheng et al., 1994). Preliminary results following intratracheal instillation of titanium tritide into rats indicate that the dissolution rate is similar to that observed in vitro (Cheng et al., 1995). It is assumed that for inhalation of inorganic particulate material, the biokinetics of tritium absorbed into body fluids follow that of HTO.*

There are additional studies, in the recent literature, showing that the dissolution of solid tritiated compounds follow the same behavior as Type M and Type S classes of solubility described in ICRP Publication 66 (ICRP 1994a).

Animal studies have shown that titanium tritide presents a slow lung clearance, and hence should be assigned to ICRP Publication 30 Class Y. It is suggested that titanium tritide should be assigned to Types M or Type S (Balonov et al. 1984, 1995; Cheng et al. 1997, 1999). The lung retention parameters for zirconium tritide are consistent with assignment to Type M and Type S in animals (Zhou and Cheng 2004). For carbon tritide, studies have suggested consistency with ICRP Type M and Type S classes of solubility (Cheng et al. 2002a, Hodgson et al. 2004). Experiments with animals have shown that hafnium tritide is consistent with ICRP Type M and Type S classes of solubility (Zhou and Cheng 2003; Inkret et al. 2001; Cheng et al. 2002b). In summary, the published studies on tritides suggest that the lung retention parameters are similar to the ICRP Type M and S compounds.

2.3 WORKPLACE CHARACTERIZATION

The Department of Energy (DOE) and its predecessor agencies have undertaken missions involving work with hazardous and/or exotic materials and agents, and byproducts of their production, storage, and use. Included in these exotic materials are metal tritides (MTs) and organically-bound tritium (OBT), referred to collectively as special tritium compounds (STCs) (DOE 2004). Sites identified as handling STCs directly include, but are not limited to, the Mound Plant, Lawrence Livermore National Laboratory (LLNL), Los Alamos National Laboratory (LANL), the Pinellas Plant, Sandia National Laboratory (SNL), and Oak Ridge National Laboratory (ORNL). Other sites came in contact with STCs as a byproduct of the work they conducted on components received from these facilities. Other STCs have been introduced to sites as a byproduct of handling large quantities of tritium gas and tritiated water, and its subsequent absorption into materials (e.g., rust, tritiated oil, tritiated dust, etc.). STCs differ from tritium oxide and elemental tritium (HT) in their physical, chemical, and radiological properties, which may make their detection, characterization, and subsequent assessment of exposure effects difficult.

Examples of processes where STCs may create a potential for exposure include (DOE 2004):

- Tritium targets for neutron generators
- Reactor operations
- Fusion experiments
- Extrication of tritium from fuel elements
- Isotope separation
- Storage of tritium
- Operations with tritium-labeled compounds
- Waste treatment and storage
- Decontamination and decommissioning activities
- Weapons testing

There are any number of locations where these materials may be encountered, such as glove boxes, fume hoods, ventilation systems, weapons components, and fuel storage basins.

STCs are categorized by physical form (e.g., liquid/vapor, particulate, or large solid form) and by solubility (e.g., insoluble, partially soluble, soluble). At some facilities, they refer to metal tritides as stable or unstable, which is a measure of how easily the tritium is released from the metal in air or aqueous material. Insoluble metal tritides (i.e., stable metal tritides) include hafnium, titanium, europium, and zirconium tritide. Soluble forms of MTs include uranium, lithium, and palladium tritide (DOE 1994, DOE 2004). While providing examples of soluble and insoluble metal tritides, this list is not inclusive of the metal tritides handled by the DOE complex. Tritiated metals can also be produced as a byproduct of handling large amounts of HT or HTO.

> *Tritium as HT or HTO will readily adsorb onto the surface of most metals (e.g., stainless steel, copper, aluminum), plastics, and rubbers. The tritium will remain fairly close to the surface unless the metal is heated to high temperatures. At room temperature, permeation into these metals is extremely slow.* (DOE 1994)

Furthermore,

> *Metal surfaces exposed to high pressures of HT or HTO for extended periods, especially at high temperatures, may allow enough penetration to cause structural damage to metal.* (DOE 1994)

Organically Bound Tritium (OBT) is formed when tritium forms a chemical bound with organic materials, typically by creating carbon-tritium bonds. Under some conditions, this is done intentionally, such as radioactive labeling of biological molecules, and, in other cases, it is a byproduct of organics adsorbing elemental tritium or tritium oxide. Solid particulate OBTs are largely formed by incidental contamination of environmental dust and other materials found in most tritium contamination areas. DOE-HDBK-1079-94, *Primer on Tritium Safe Handling Practices* (DOE 1994), describes the interaction of tritium with hydrogenous materials.

> *If adsorbed onto hydrogenous material, the tritium will easily permeate into the material. The HTO will move much more rapidly into the bulk material than will HT. The permeation rate varies with the type of material and is accelerated by increasing the temperature. As a result of this movement, plastics and rubbers exposed to tritium (especially as HTO) are readily contaminated deep into the bulk material and are impossible to decontaminate completely.*

Insoluble OBTs include tritiated pump oil, dust, rust, pump oil droplets, tritiated flyash, and tritiated nylon. Soluble OBTs include tritiated solvents and methane (DOE 2004). Particulates can be generated during dispersal mechanisms from material such as plastic, nylon, organic dust, or large molecule components of OBT oil. Insoluble OBTs present the same challenges in detection and measurement that other insoluble tritiated particulates present.

Because of the low energy beta particle emitted by tritium, area monitoring efforts are limited to surface contamination and airborne radioactivity monitoring. Surface contamination for tritium is determined by taking either a wet or dry smear and counting it in a liquid scintillation counting (LSC) system. Although LSC is effective for detecting the presence of tritium, this technique does not differentiate between types of tritium. Stable metal tritides present a unique challenge when attempting to monitor for surface and skin contamination. Since these materials are in particulate form, many of the tritium atoms associated with the particle are internal to the surface of the particle. As a result, the counting technique will show a tritium activity that is too low, since the beta particle from tritium within the metal tritide particle cannot escape the particle. As the tritium is released from the metal, the concentration of tritium in the scintillation cocktail will increase, and thus the concentration will appear to increase. To date, there is not a simple and reliable method for identifying STCs. Some work has been conducted using an Energy Dispersive X-ray/Scanning Electron Microscopy technique; however, this method has only been applied since the 1990s. The method is used to establish the presence, size and shape of insoluble tritium particulates, but does not provide information on the tritium concentrations.

Typical continuous air monitoring techniques used flow-through ionization chambers as the major monitoring technique for identifying tritium in air. These instruments can measure tritium in its elemental form (HT), oxide form (HTO), or any other gaseous compound form (tritiated methane, ammonia, etc.). However, filters and electronic precipitators are often used in conjunction with these air monitoring systems to prevent particulates from entering the ionization chamber and thus being detected.

While soluble STCs can be detected via urine bioassay, insoluble STCs present substantial bioassay challenges. Insoluble STCs can be difficult to detect in urine because of low dissolution rates of some compounds. For example, studies reported by Cheng, et al. (Cheng 2002b) point out that for hafnium tritide particles in simulated lung fluids, less than 1% of the tritium was dissolved after 215 days, and the long-term dissolution half-time was 4.28×10^{5} days. Dissolution rates are also size and shape dependent, and although dissolution rate data are available for several materials, these data do not include all the possible combinations of materials and particle sizes which might be encountered in the workplace. In vivo analysis is not a viable means of personnel monitoring, because the low-energy beta particle cannot penetrate through the body to the outer surface, and detection of bremsstrahlung x-rays is not likely unless

an STC uptake is very large. While fecal sampling is a possibility, a suitable fecal sampling program for tritides has not been developed.

> *The ultimate shortfall of fecal bioassay for tritiated particulate is that it is not currently available. Until the technique is available, tested, and evaluated, fecal bioassay for tritiated particulate cannot be implemented* (Mound 2004).

Particulate air monitoring has been chosen as the method of choice for estimation of intakes in lieu of adequate bioassay techniques. According to *Mound Technical Basis Document for Stable Tritiated Particulates and Organically Bound Tritium* (Mound 2004):

> *The expected uncertainty range in the assigned dose from air sampling is <2 orders of magnitude, which in fact represents a range of overestimation. This is much less than the expected uncertainty range, from urine of 3-4 orders of magnitude.*

Particulate air monitoring shortfalls for tritiated particulates are:

(1) The self-absorption factor varies as a function of material and particle size by a factor of approximately 10.

(2) The activity collected and measured on filters under-represents the actual activity available for deposition in the lungs.

(3) The measured activity overestimates intake because of capture of non-respirable particulates on the filter.

(4) The air sample must represent the workers' breathing zone. Personnel air sampling is preferred over general area air sampling, because it is more representative of the worker breathing zone.

Uncertainties with the use of air monitoring, particularly general air monitoring, would be substantial and methods of air monitoring and the availability of appropriate air monitoring techniques in areas handling STCs is uncertain.

The potential exposure to STCs is significant in facilities that either directly handled these compounds, or in facilities that handled high quantities of elemental tritium and tritium oxide. Difficulties in area monitoring can inhibit the ability to measure STCs in the workplace. Furthermore, lack of adequate bioassay techniques limits the ability to identify uptakes which may have occurred. Potential exposures are not limited to hands-on workers, but also affect decontamination and decommissioning and maintenance workers.

2.4 DOSE ASSESSMENT BASED ON AVAILABLE ICRP MODELS

As part of its assessment of OTIB-0066, SC&A reviewed the available data in the open literature and derived dose coefficients based on ORAUT-OTIB-0011 (ORAUT 2004), with biokinetic models given in ICRP Publications 56 and 67 (ICRP 1989, ICRP 1993) considering Type M and Type S solubility classes for metal tritides. The dose coefficients are presented in Table 3 for soft tissue, lung, and colon; considering inhalation of vapor (HTO and OBT) and particles (SMT)

with AMAD = 1μm and 5 μm. The table also presents the dose coefficients from ICRP Publication 78 (ICRP 1997) for HTO and OBT.

Table 3. Comparison of Calculated Doses for a 1-pCi Acute Intake of Tritium Compounds

Methodology (organ)	Dose Coefficients (mrem/pCi)			
	SMT (Type S)	SMT (Type M)	HTO	OBT
ORAUT-OTIB-11 (Soft tissue)	6.58E-10	7.28E-09	7.11E-08	1.08E-07
AIDE[c] (Soft tissue)	6.30E-10[a]	6.93E-09[a]	6.76E-08[b]	1.52E-07[b]
AIDE[c] (Lung)	3.86E-06[a]	5.52E-07[a]	6.76E-08[b]	1.52E-07[b]
AIDE[c] (Colon)	3.16E-07[a]	2.87E-07[a]	6.76E-08[b]	1.52E-07[b]
ICRP 78 (Soft tissue)	-	-	6.66E-08[b]	1.52E-7[b]

Notes: a. Dose coefficients derived considering AMAD = 5μm
b. Dose coefficients derived considering vapor
c. Computer code for internal dose calculation

2.5 SC&A COMMENTS

2.5.1 Agreement with ORAUT-OTIB-0066

Issue 1 (observation): SC&A has found that ORAUT-OTIB-0066 is clear and consistent with International Commission on Radiological Protection (ICRP) Publications 56 (ICRP 1989) and 67 (ICRP 1993), ORAUT-OTIB-0011, and open literature (Balonov et al. 1984 and 1995; Cheng et al. 1997, 1999, 2002a, and 2002b; Inkret et al. 2001; Zhou and Cheng 2003; Hodgson et al. 2004; and Zhou and Cheng 2004).

Issue 2 (observation): The procedure provides adequate and valid technical information and guidance to evaluate the dose due to the intake of HTO, OBT, and SMT, applying the ICRP models in IMBA.

This observation is not withstanding implementation difficulties discussed in Section 2.4.2.

2.5.2 Disagreement with ORAUT-OTIB-0066

Issue 3 (Finding): The recommendation given in ORAUT-OTIB-0066 to assess dose due to intake of OBT is not claimant favorable. The OTIB recommends the use of the methodology given in ORAUT-OTIB-0011 to calculate doses from intakes of OBT to all organs and tissues. The dose coefficient for OBT given in ICRP Publication 78 (ICRP 1997), which is 1.52×10^{-7} mrem/pCi (see Table 3); the one derived applying the biokinetic model for OBT (ICRP 1989; ICRP 1993; ICRP 1995; and ICRP 1997) using the AIDE computer code, which is 1.52×10^{-7} mrem/pCi (see Table 3); and the one derived using the methodology given in ORAUT-OTIB-0066 (ORAUT 2007), which is also 1.52×10^{-7} mrem/pCi (see Table 2), are 1.4 times higher than the one obtained applying the methodology given in ORAUT-OTIB-0011 (ORAUT 2004), which is 1.08×10^{-7} mrem/pCi.

Issue 4 (Finding): The types of special tritium compounds, the quantities handled, the time periods of potential exposures, and the physical behavior of the tritium compounds in the environment must be known to effectively develop and apply OTIB-0066.

Bounding techniques proposed in ORAUT-OTIB-0066, cannot be effectively developed and applied without some basic understanding of the special tritium compounds handled, the quantities of material, the locations and time periods of potential exposure, and the physical behaviors of tritium compounds in the environment (e.g., conversion to HTO, formation of rust) to correctly characterize tritium exposure. This is partially confirmed in ORAUT-OTIB-0066 by the following recommendation (ORAUT 2007).

> *In the vast majority of occupational exposures to tritium it is not possible to identify the tritium compounds taken into the body based on the observed excretion. Therefore, the selection of the appropriate tritium compound in an intake evaluation must usually be based on process knowledge of the source terms in the workplace* (pg. 10).

There are several input parameters critical to the application of OTIB-0066 that must be determined prior to application of the model at particular DOE sites:

- Compounds of tritium handled
- Quantities of material handled
- Particle size and shape of insoluble STCs
- Ratios of tritium to metal
- Locations where STCs were handled
- Time periods of potential exposure
- Engineering controls used for processing at a particular site
- Production of compounds from adsorption of HT and HTO
- Which workers came in contact with these materials

This is critical information because of the inadequacies in personnel and area monitoring programs prior to the 1990s. In many cases, characterization efforts to identify and locate STCs were limited. Some thought should be given to the chemical behavior of tritium, especially as it related to the decontamination and decommissioning of aging tritium facilities and other facilities where tritium was a byproduct of operations.

Characterization of STCs in the work environment is difficult because information may not be easily assessable and, in some cases, does not exist; process descriptions and documentation can be difficult to obtain; the CATI process does not allow for disclosure of key information from the energy employee unless special arrangements are made; and, in many cases, claimants are not aware of the STCs handled at the facility.

Identification of who were exposed, when they were exposed, and to what compounds they were exposed, can be difficult. This is compounded by the potential exposure to STCs formed in work environments that handled large quantities of elemental tritium and HTO (e.g., formation of rust, particulate dust). These sources of STCs can affect a larger number of workers in

facilities where tritium was present. There was also an inadequacy of early monitoring techniques to detect some STCs.

Issue 5 (Finding): OTIB-0066 does not ensure that resultant doses are based on adequate monitoring data.

Several factors contribute to the adequacy of monitoring data used as input to the proposed models. The purpose of OTIB-0066 is (ORAUT 2007):

> *.... to provide guidance on how to use urine bioassay data to calculate the best estimates of the annual organ doses for intakes of tritium bound to organic compounds (organically bound tritium; OBT) and tritium in a metal matrix (stable metal tritides; SMT)* (5).

Although urinalysis is the basis for application of the models outlined in OTIB-0066, there is no discussion on the practical interpretation on urinalysis results and the technical shortfalls associated with this technique for insoluble tritiated particulates. For insoluble tritiated particulates, effective methods for personnel and area monitoring were not implemented during periods of production and decontamination and decommissioning in many cases. As previously mentioned, insoluble STCs can be difficult to detect in urine because of the low dissolution rates of some compounds. The rates for the same compounds can vary by particle size and shape, making it difficult to predict how insoluble tritiated particulates will behave in the urine. Suitable methods for detection of tritium in fecal samples are currently not available (Mound 2004). In-vivo counting is not feasible, because the beta particle will not escape the body. Furthermore, Mound (2004) states that it may not be possible to accurately monitor for surface and skin contamination when dealing with stable metal tritides. The method of choice for personnel monitoring is particulate air monitoring; however, there are multiple issues with the use of these data.

The procedure does not provide information to the dose reconstructors on the characteristics of STC excretion in urine in practical situations enabling them under optimal circumstances to distinguish patterns of excretion from STCs. For example, McConville and Woods (1995) demonstrated, with individual excretion data following tritide uptakes, that tritium excretion curves for particulate tritides do not follow a simple exponential curve, as is the case with HTO. In the case of these individuals, tritides build up for a few days followed by a more traditional elimination curve.

McConville & Woods 1995 go on to raise the following concerns about dose assessment for uptakes of metal tritides:

> *Tritium in the form of metal tritides particles presents a peculiar problem for the calculation of internal dose. Standard calculations indicate that just a few 3 to 5 micron sized particles appears to lead to a very large dose. There are very few data on which calculations can be based.*

Furthermore,

> *As the dissolution rate for an SMT approaches zero, the calculated intake (and dose rate to the lung) becomes very large, and small uncertainties in urine measurements can lead to significant uncertainty in the resulting (calculated) dose* (Mound 2004).

However, because dissolution rates vary by orders of magnitude, including some very slow rates, choice of a very slow dissolution rate can lead to intake (and dose) estimates which are overestimated by orders of magnitude when derived from urine data. This overestimate is compounded when intakes of HTO accompany particulate intake and increase the concentration in the urine. Biokinetic information is not available in the open literature or through other sources for all compounds of special tritium compound handled at DOE sites. In some cases, the dissolution factor for compounds must also be determined.

Issue 6 (Finding): The procedure provides no guidance on how to distinguish between intakes of STCs, elemental tritium, and/or tritiated water which occur simultaneously or overlap.

Circumstances exist where workers may come in contact with multiple forms of tritium during the course of their work. Tritiated water, tritium gas, and soluble STCs can be readily evaluated via urine bioassay. As such, they can mask uptakes of insoluble STCs.

> *In urine bioassay, the excreted tritium from an HTO intake is indistinguishable from the excreted tritium from a tritiated particulate intake, largely obscuring the bioassay results from tritiated particulate. However, tritium from HTO and particulate could be distinguishable in fecal analysis..... Presence of OBT in fecal samples might not be distinguishable from tritiated particulate, however, and would serve to obscure particulate results (Mound 2004).*

In practice, the sites have assumed all tritium uptakes were from elemental tritium or tritium oxide; thus, dose calculations (sometimes the only values available) incorporate the default parameters for tritium oxide. OTIB-0066 proposes the use of Type S solubility when the SMT is unknown. When soluble forms of tritium are measured in the urine, and Type S SMTs are assumed, this procedure can grossly overestimate the exposure, giving values which are implausible.

It is important that the dose reconstructor chooses claimant-favorable forms of tritium for the dose estimation if there is a lack of process knowledge about the form that may have been involved with exposures. The dose to the lung from intakes of SMTs can be underestimated by orders of magnitude if it is incorrectly assumed that the dose can be calculated with the methods used for HTO (ORAUT 2007). Furthermore, doses obtained in a bounding technique must be plausible.

2.5.3 SC&A Recommendations

(1) The ORAUT-OTIB-0011 methodology should be applied just for tritiated water (HTO).

(2) The ICRP biokinetic model for OBT must be applied in IMBA to assess the organ doses due to intake of OBT.

(3) The ICRP biokinetic model for HTO, along with the lung retention parameters for Type M or Type S, must be applied in IMBA to assess the organ doses due to intake of SMT.

(4) Characterization of the potential tritium exposure at a facility including STCs (produced or byproducts) is critical to the application of models in OTIB-0066 and must be documented more fully. Claimant favorable assumptions cannot be made in the absence of this information.

(5) OTIB-0066 should consider the availability and adequacy of data used in the model and how the shortfalls of these data will be addressed.

3.0 PROCEDURE CHECKLIST

SC&A reviewed the OTIB in accordance with its procedure, *A Protocol for the Review of Procedures and Methods Employed by NIOSH for Dose Reconstruction* (SC&A 2004). Table 4 is taken from that procedure. Since the table has general applicability, not all of its items are germane to the review of the OTIB.

Table 4: Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
1.0	**Determine the degree to which procedures support a process that is expeditious and timely for dose reconstruction**		
1.1	Is the procedure written in a style that is clear and unambiguous?	See Comment	5- Description of the available models 2- Application of the model to DRs
1.2	Is the procedure written in a manner that presents the data in a logical sequence?	5	
1.3	Is the procedure complete in terms of required data (i.e., does not reference other sources that are needed for additional data)?	2	In order to apply this proposed model, one has to rely on OTIB-0011.
1.4	Is the procedure consistent with all other procedures that are part of the hierarchy of procedures employed by NIOSH for dose reconstruction?	5	Consistent with ORAUT-OTIB-0011.
1.5	Is the procedure sufficiently prescriptive in order to minimize the need for subjective decisions and data interpretation?	2	The procedure provides a model, but does not discuss the practical application of the models. Furthermore, it does not provide guidance on distinguishing exposures to STCs in the presence of other tritium compounds.
2.0	**Determine whether the procedure provides adequate guidance to be efficient in instances where a more detailed approach to dose reconstruction would not affect the outcome**		
2.1	Does the procedure provide adequate guidance for identifying a potentially high probability of causation as part of an initial dose evaluation of a claim?	2	The procedure provides insufficient guidance for identifying potential exposures and applying the OTIB.
2.2	Conversely, for claims with suspected cumulative low doses, does the procedure provide clear guidance in defining worst-case assumptions?	2	OTIB-0066 does not consider the difficulties associated with adequate personnel monitoring data and how this will impact the application.

Table 4: Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
3.0	**Assess the extent to which procedures account for all potential exposures and ensure that resultant doses are complete and based on adequate data**		
3.1	Assess quality of data collected via interviews:	N/A	
3.1.1	Is scope of information sufficiently comprehensive?	N/A	
3.1.2	Is the interview process sufficiently flexible to permit unforeseen lines of inquiry?	N/A	
3.1.3	Does the interview process demonstrate objectivity, and is it free of bias?	N/A	
3.1.4	Is the interview process sensitive to the claimant?	N/A	
3.1.5	Does the interview process protect information as required under the Privacy Act?	N/A	
3.2	Adequacy and use of site specific data pertaining to:		
3.2.1	Personal dosimeters (e.g., film, TLD, PICs)	N/A	
3.2.2	In-vivo/in-vitro bioassays	2	The procedure does not consider the problems associated with in-vitro monitoring for insoluble STCs.
3.2.3	Missing dosimetry data	2	The OTIB fails to consider the availability of raw tritium bioassay data. Tritium was often calculated in terms of dose and considered a part of the whole-body exposure. At some sites, dose data is the only readily available monitoring data.
3.2.4	Unmonitored periods of exposure	2	The concern over STCs in the work environmental and their unique characteristics has only recently been raised. Prior to the late 1990s, it is unlikely mechanisms were being implemented for the specific measurement of insoluble STCs.

Table 4: Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
4.0	**Assess procedure for providing a consistent approach to dose reconstruction regardless of claimant's exposures by time and employment locations**		
4.1	Does the procedure support a prescriptive approach to dose reconstruction?	2	OTIB-0066 presents a model for application which is based on international standards, and does not consider DOE site-specific information including types of special tritium compounds, the quantities handled, the time periods of potential exposures, and the physical behavior of the tritium compounds in the environment.
4.2	Does the procedure adhere to the hierarchical process as defined in 42 CFR 82.2?	4	
5.0	**Evaluate procedure with regard to fairness and giving the benefit of the doubt to the claimant**		
5.1	Is the procedure claimant favorable in instances of missing data?	3	In some cases, the procedure will overestimate dose while in other cases it will underestimate dose. There is still a great deal of information unknown about the biokinetic behavior of some STCs, so in some cases the proposed procedure cannot be evaluated.
5.2	Is the procedure claimant favorable in instances of unknown parameters affecting dose estimates?	3	
5.3	Is the procedure claimant favorable in instances where claimant was not monitored?	3	
6.0	**Evaluate procedure for its ability to adequately account for the uncertainty of dose estimates**		
6.1	Does the procedure provide adequate guidance for selecting the types of probability distributions (i.e., normal, lognormal?)	N/A	
6.2	Does the procedure give appropriate guidance in the use of random sampling in developing a final distribution?	N/A	

Table 4: Procedure Review Outline/Checklist

No.	Description of Objective	Rating 1-5*	Comments
7.0	**Assess procedures for striking a balance between technical precision and process efficiency**		
7.1	Does the procedure require levels of detail that can reasonably be accounted for by the dose reconstructor?	2	
7.2	Does the procedure avoid levels of detail that have only limited significance to the final dose estimate and its POC?	4	
7.3	Does the procedure employ scientifically valid protocols for reconstructing doses?	3	

* Rating system of 1 through 5 corresponds to the following: 1=No (Never), 2=Infrequently, 3=Sometimes, 4=Frequently, 5=Yes (Always), N/A indicates not applicable

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