

# **Review of NIOSH's Current Approach to Reconstruction of Insoluble Tritium Particulate at the Pinellas Facility**

**Revision 0**

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## **National Institute for Occupational Safety and Health**

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## Background

During the Pinellas working group meeting on November 19, 2012, NIOSH indicated that they wanted to review the current approach to reconstruction of insoluble tritium exposures at Pinellas. While the approach described in section 5.7.1.2 of the Pinellas site profile (ORAUT-TKBS-0029-5) is similar to that used for the Mound facility, NIOSH wanted to make sure that the methodology and parameters used for the bounding intake calculation were appropriate. To this end, the following 5 issues were evaluated: 1) the resuspension value used; 2) the validity of the bounding contamination survey used; 3) the appropriateness of the tritide measurement technique; 4) a review of the extent of tritide contamination, and 5; the solubility types of tritides present at Pinellas. A review of each of these issues is provided below.

### **Issue 1: The appropriateness of the 1E-06 resuspension factor (RF) in light of the approach currently being adopted at Mound that uses an RF of 5E-05**

A review of the Mound facility and other sites where resuspension factors have been applied in active work areas was performed. An RF of 1E-06 is generally used for undisturbed areas; whereas, an RF of 1E-05 is generally used for areas with ongoing operations. As discussed in the sections below, Pinellas was aware of the impact of contaminated areas and worked to maintain a clean work environment when contamination was identified. However, one could argue that the type of tritium work and facilities at Pinellas are similar to those at Mound. Therefore, the more favorable to the claimant RF of 5E-05 will be used. This will result in an increase in the annual metal tritide (MT) intake rate from 6.184E+05 pCi/year to 3.092E+07 pCi/year.

### **Issue 2: The use of the highest contamination survey reported between 1957 and 1973 as the basis for the airborne contamination source term**

For the Mound approach the site profile calculated the 95th percentile value's surface contamination levels using the 60,264 contamination smear sample results that were captured from the site.

For the Pinellas Plant, little individual contamination smear data is available. The primary data source that we have for the Pinellas Plant's contamination survey results were the monthly Health Physics Summary Reports that reported the highest contamination levels measured for a given month.

An analysis of the available data of the monthly Health Physics Summary Reports (SRDB 27095) found the highest surface contamination level reported was 4.4E+06 dpm/100 cm<sup>2</sup>, which is 10,000 times their control limit. These reports also indicate that surface contamination levels above the control limits of 2E-05 μCi/in<sup>2</sup> (688 dpm/100 cm<sup>2</sup>), as early as 1959, and 440 dpm/100 cm<sup>2</sup> (reported in 1969) resulted in the initiation of decontamination efforts. This was confirmed

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in an interview of a past radiological control personnel (SRDB 127111) who indicated that metal tritide contamination was cleaned up fairly quickly. Radiological Control personnel would take wipes in the morning and, if contamination was identified, they would then mop up the area and re-survey. The likelihood of routine surface contamination level in the millions of dpm/100 cm<sup>2</sup> should be considered unusual and short in duration.

As a comparison, the assumption of an average contamination level of 4.4E+06 dpm/100 cm<sup>2</sup> is one to two orders of magnitude higher (depending on the year) than the surface contamination levels at Mound, which had a similar process as Pinellas. Therefore, the approach applied at Pinellas is considered bounding and favorable to the claimant.

### **Issue 3: The ability of the method used to measure smear samples to detect tritium bound to particulate metal**

The primary focus of tritium contamination monitoring at the Pinellas Plant was monitoring for soluble forms of tritium. The three tritium contamination smear analysis procedures that are available (SRDB Ref 12806 p 80-82, SRDB Ref 12947 p 95-96 and 135-137) indicate that the Pinellas Plant used wetted cotton balls to collect smear samples for tritium contamination monitoring. The available procedures also indicate that the cotton ball smear samples were rewetted in a paper cup with a prescribed amount of deionized water (8 to 10 ml, depending on the procedure), and that a rinsate was squeezed from the cotton balls while they were still in the paper cup. These procedures indicate that the rinsate from the cotton balls was then filtered through a Whatman #1 filter. Two of the available procedures are undated, but are among a collection of other procedures that have dates range from the late-1960s into the 1980s (SRDB Ref 12947 p 95-96 and 135-137). The copy of the procedure found in an incident investigation report indicates that the rinsate was filtered through a Whatman # 1 filter as late as January 1976 (SRDB Ref 12806 p 80-82). Interviews with Pinellas Plant workers involved with the tritium contamination smear collection and analysis procedures indicate that the rinsate was no longer being filtered by 1977

In an interview with a Health Physicist that was at the Pinellas Plant during 1987–1997 (SRDB 127111), it was indicated that they had never used the Whatman #1 filter to filter the rinsate from the tritium smears. When they analyzed the smears, the rinsate was unfiltered. An additional interview with two of the Chemistry Department’s personnel that analyzed the tritium contamination smears during the years 1977–1997 (SRDB 129125) stated that they recall rinsing the cotton balls used to collect the tritium contamination smear samples. However, they did not recall filtering the rinsate through a filter. According to them, filtering the rinsate through a filter was not part of the routine analysis procedure when they were at the Pinellas Plant and all of the routine tritium contamination smears were analyzed by the same procedure. To their knowledge, there were not any routine or special procedures for analyzing the tritium contamination smears when metal tritide contamination was suspected.

One possible purpose of filtering a rinsate could be to limit the amount of dust and dirt ending up in the filtrate, which would minimize quenching issues. However, the chemistry department

personnel indicated that they could not remember any issues with the smears being so dirty that it would cause problems with the cocktail solution.

The Whatman #1 filter paper has a nominal pore size of 11 microns, and a maximum pore size of 12.7 microns. Therefore, the Whatman #1 filters would only filter out the non-respirable SMT particles (i.e. the particles greater than 10 microns), if they filtered out any SMT particulates at all. This may have been the reason why the Whatman #1 filters were chosen to be used as a filter.

The SMTs of concern at the Pinellas Plant were deposited as a vapor onto ceramic or metal target substrates. The SMTs on ceramic or metal target substrates were typically only dispersed from metal flaking off the substrates or from vacuum tube implosions (e.g. the accidental breaking of neutron tubes). A material data sheet for titanium-hydride (SRDB 12275, pg 28) indicated that average particle size ranged from 6 to 9 microns, which is less than the 11 micron nominal pore size for the Whatman #1 filters. Therefore, it is unlikely that there was any loss in particulate activity from filtering the rinsate (i.e. there would be no impact to an individual's assessed exposure).

In addition, a Health Physics paper (SRDB 12275, pp 9-11) titled, "A Study of the Particulate and Gaseous Emission of Tritium from Neutron Generator Targets", looked at impact on deuteron bombardment on the targets. The study looked at used and new targets. The study results for new targets (which what was handled at Pinellas) show that the gaseous emission rate was much higher than that of the particulate. This implies that any intake associated particulate tritium (SMT) would also be associated with a much higher intake of gaseous tritium (HT and HTO), which would have been identified based on the individuals urinalysis results. Since all contamination smears in the Pinellas methodology are treated as 100% particulate, even if the Whatman #1 filter paper filtered out the SMT particulates, the recorded tritium activity (which would be associated with just gaseous tritium alone) would provide a bounding estimate of SMT. However, given the Whatman #1 filter paper likely would not filter out the SMT particulate, the use of the total tritium results (particulate plus gaseous) as 100% Type S SMT particulate results in an bounding estimate of the particulate contamination levels.

#### **Issue 4: The magnitude and the extent of potential for tritide contamination at Pinellas**

At the Pinellas Plant, insoluble forms of tritium were handled only in areas where the more dispersible and more soluble forms of tritium were also present. A review of the available dosimetry records indicates that the Pinellas Plant routinely monitored workers with any potential for soluble tritium exposures. Therefore, SMT exposures only need to be assessed for workers were also monitored for soluble tritium exposures.

The available information on the Pinellas Plant indicates that only a relatively small portion of the Pinellas Plant's workforce had the potential to be exposed to dispersible forms of SMTs, and those scenarios were typically limited to accidents. The available incident information in the monthly health physics reports (SRDB 13358, 27095, 133577, 133579, 133580, 133581, 133583,

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133586, and 133591) indicate that when the contamination levels exceeded the control level, the affected areas were cleaned up immediately. There is no indication from these reports that routine contamination levels were close to the bounding levels assumed in the Pinellas approach.

In a lot of cases, the source of the contamination from these reported incidents (SRDB 13358, 27095, 133577, 133579, 133580, 133581, 133583, 133586, and 133591) are not associated with SMTs. Rather, they are associated with loaded glass tubes of tritiated gas that were dropped releasing the tritium gas or from maintenance activities (which would likely be associated with tritiated pump oils, i.e. OBTs). Exposures associated from these types of releases would be also captured in the analysis of the tritium bioassay.

In cases (SRDB 27095) where contamination was associated with actual SMTs releases, it was normally associated with titanium tritide, which is a more soluble SMT (Type M). The assumption that 100% of all intakes are Type S SMTs is not a likely scenario, but is a favorable-to-the-claimant assumption. Scandium tritide was the only known Type S insoluble tritium compound present at the Pinellas Plant.

The highest reported tritium surface contamination level was  $4.4\text{E}+06$  dpm/100 cm<sup>2</sup> (10,000 times the control limit). This was reported in a 1970 health physics summary report (SRDB 27095) and was associated with maintenance operations, not a SMT incident. The highest tritium contamination level associated with a SMT incident (SRDB 27095) was  $1.89\text{E}+05$  dpm/100 cm<sup>2</sup> (about 4% of the surface contamination level used to estimate the SMT exposure in the TBD). In most cases, tritium surface contamination level associated with a SMT incident was at least an order of magnitude less. This means that the use of the highest reported tritium surface contamination level assumed to have occurred for 2000 hours/year to estimate the SMT exposure rate likely results in a SMT exposure that is orders of magnitude larger than the actual potential SMT exposure.

### **Issue 5: The relative solubility of the various metal tritides present**

The Pinellas Plant had SMTs representing all three lung absorption types. The SMTs at the Pinellas Plant were primarily erbium tritide (Type M), scandium tritide (Type S), titanium tritide (Type M), and uranium tritide (Type F), but other less prevalent SMTs were also at the site.

From 1957 through 1967, glass tritide storage beds that utilized titanium metal were used (SRDB 12026). Elemental target material (typically erbium, scandium, or titanium) was deposited as a vapor onto a ceramic or metal target substrate (SRDB 12945 & 88797), after which the targets were loaded in the neutron tubes. The nearly completed neutron tubes were then attached to a glass manifold vacuum system under an exhaust hood. The glass encased titanium-hydride storage beds were replaced with stainless-steel encased depleted uranium-hydride storage beds in 1968 (SRDB 13125).

Therefore, the source term for SMT exposures are typically not Type S SMTs, rather the more moderately insoluble and soluble Type M and F SMTs. The assumption that 100% of all intakes are Type S SMTs is not a realistic scenario, but it is a favorable to the claimant assumption.

## **Conclusions**

Given the ongoing nature of operations at Pinellas, NIOSH believes that the resuspension value of 1E-06 used in the current TBD should be increased to 5E-05. A review of the filtration methodology employed to measure smears at Pinellas indicates that the technique is capable of quantitatively assaying particulate tritium contamination. Because the majority of the smear surveys (and in particular the highest ones) were likely to represent soluble or slightly insoluble tritium compounds, NIOSH concludes that the use of the highest measured contamination survey result will result in claimant favorable doses to workers at Pinellas.