White Paper: Representativeness and Applicability of United Nuclear Corporation Air Sampling for Reconstructing Thorium Intakes

by
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1.0 Introduction and Purpose

United Nuclear Corporation (UNC), located in Hematite, Missouri, manufactured uranium metal and uranium metal compounds from natural and enriched uranium for use as nuclear fuel. The facility became operational in 1956, initially producing uranium products for use in the naval fuel program. For an approximately nine-month period in 1964, UNC also blended thorium dioxide powder with uranium dioxide powder to produce fuel pellets for use in fuel assemblies for the Elk River Reactor (Pellet Plant Data, 1964; Swallow, 1963; Swallow, 1964a). The facility processed un-irradiated uranium scrap for the AEC, recovering enriched uranium for use in the nuclear weapons complex. Under the Energy Employees Occupational Illness Compensation Program, the site is designated as an Atomic Weapons Employer from 1958 through 1973; the Residual Radiation period is specified as 1974 through October 2009 (DOE Facilities, 2011).

Although UNC maintained an active urinalysis program for operations workers during the nine-month Th/U blending period, the urinalysis samples were analyzed only for uranium (Bioassay Results, 1964; AEC, 1964). Instead, UNC-Hematite established a Maximum Allowable Concentration (MAC) for thorium work of 2E-11 µCi/ml and verified that exposures remained below this level through a routine general area and personal air sampling program (Darr, 1964a). Although these air samples are designated ThO_2 on the sample log sheets, there is no indication that thorium-specific assays were performed in addition to the gross alpha counting of the samples (Pellet Plant Data, 1964; Monitoring Surveys, 1964).

Questions arose during a meeting of the Advisory Board’s Work Group on Uranium Refining Atomic Weapons Employers as to whether the available air sampling data are representative of the air concentrations to which workers were exposed, and whether counting such data for gross alpha rather than specifically assaying for thorium provides a plausible upper-bound for thorium exposures. This white paper addresses the representativeness of air sampling measurements performed in the Pellet Plant (Building 255) during 1964. This paper specifically addresses the expressed concern about the ability to use such air sampling measurements to estimate worker intakes to thorium and its daughter products.

2.0 Process Considerations

Processing of UF_6 to UO_2 was accomplished in another area of the United Nuclear plant known as the Red Room (Darr, 1964b); ThO_2 was received from off site. While the thorium was of natural abundance, the uranium was enriched to 6.5% U-235, as approved by the U.S. Atomic Energy Commission (AEC) (Swallow, 1964a).

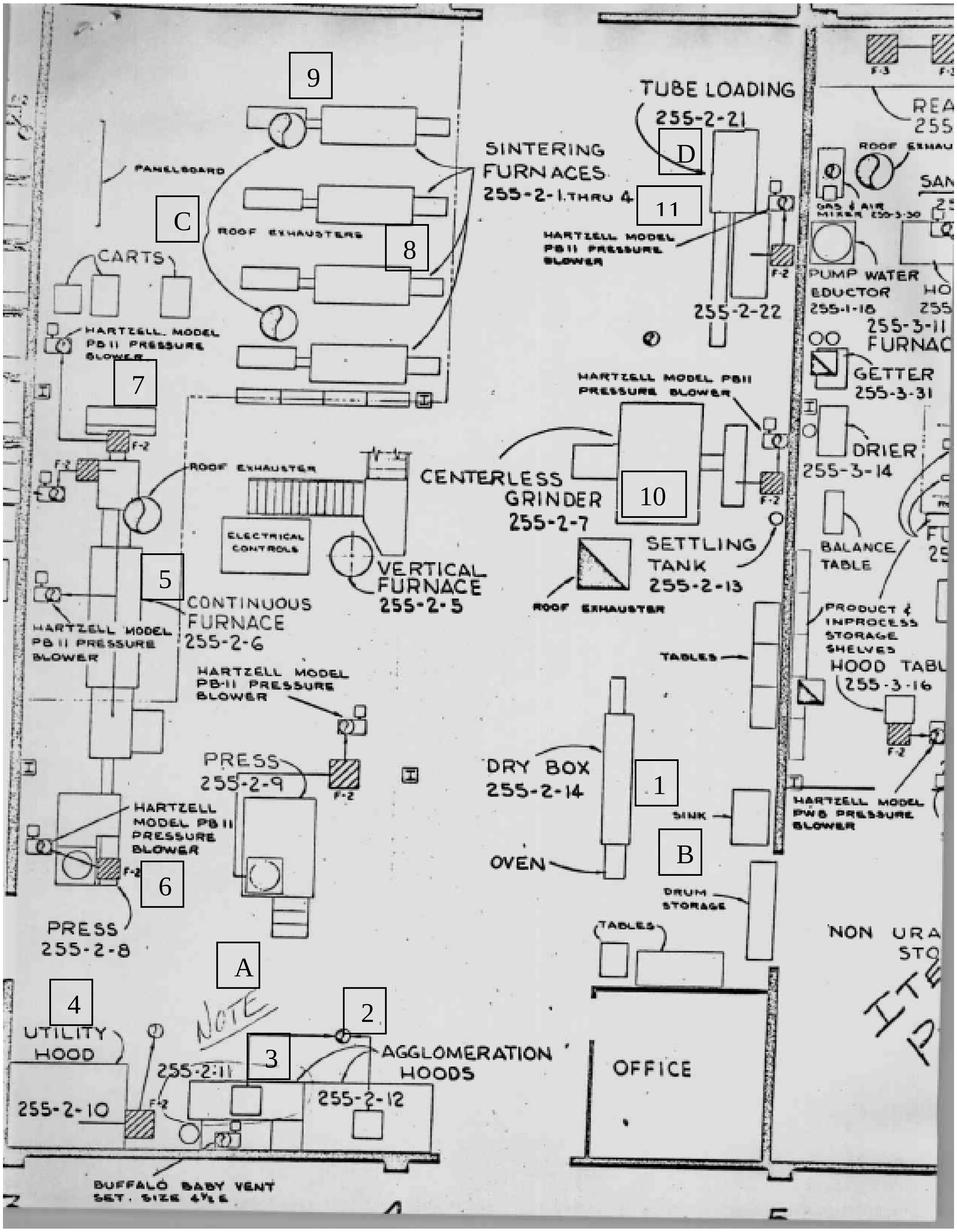
The process material was handled in small batches to limit criticality concerns (UNC Health Physics, 1964). Batches consisting of 3% UO_2 and 97% ThO_2 were blended, granulated to a specific size, and dried in a drying oven, all while under a dust-tight hood that exhausted air through two MSA filters and into the atmosphere. After drying and inspection, these mixtures would be transferred to a pellet press storage hopper under another dust hood. From there, the powders would be pressed into pellets and then heated twice to harden. The hardened pellets would be graded for size; those that were too large would be ground to the appropriate size on a centerless grinder under another dust hood, while those that were too small would be recycled back into the process (Leaders, 1958a; Leaders, 1958b).

3.0 Air Monitoring Details

An air sampling program was conducted to monitor exposures to airborne radioactive dust, to identify process areas that needed better engineering controls or designs, and to identify operations that required worker protection to minimize airborne dust exposures. Air samples were collected in the Pellet Plant by four general area samplers and 11 breathing zone samplers, as shown in Figure 1 (Darr, 1964b).

In all, 210 air sample measurements were taken between March 27, 1964 and September 23, 1964. Of these, sample logs indicate that 133 measurements were breathing zone (BZ) samples (BZ locations are boxed numbers in Figure 1); 75 were general area (GA) samples (GA locations are boxed letters in Figure 1). The remaining two samples were special samples taken in response to specific events, such as a spill that occurred during milling (Pellet Plant Data, 1964).

Although sample logs do not indicate which instrument was used for each sample, documentation indicates that UNC possessed a Filtronics, Inc., breathing zone sample head and rechargeable Ni-Cd battery, which is labeled “lapel sampler” in the list of health physics equipment and instrumentation (Swallow, 1964b). In addition, the UNC Health Physics Procedure Manual indicates that, for uranium operations, efforts were made to ensure that sampling heads were positioned near the breathing area of the operator (Swallow, 1964b). Given that the thorium pelleting operations also involved the use of uranium, there is no reason to believe that a deviation from this procedure would have occurred for thorium operations. Therefore, it is assumed that all samples listed as BZ were taken in the breathing zone, either with a lapel sampler or a sampling head placed in the breathing zone.



Source: Darr, 1964b

Figure 1: BZ and GA Air Sampling Locations in the UNC Pellet Plant in 1964

4.0 Determination of Sample Representativeness

In the absence of individual monitoring data, EEOICPA allows for the use of workplace area monitoring data to estimate personnel doses. In such a case, workplace area monitoring data are understood to include both general area air sampling results as well as breathing zone air sampling results (42 C.F.R. pt. 82). However, if air samples are to be used for determining intake, correct interpretation of the data is important and should include some analysis of the extent to which samples represent the air inhaled by the workers (NUREG-1400).

As stated in Section 3.0, 133 of the 210 air samples covering the thorium operational period were breathing zone samples. These samples were taken in breathing zones near hood openings or in other areas where airborne contamination was anticipated to be the highest. These samples are considered representative of maximum intakes for workers while performing operations in hoods or while handling thorium or thorium-contaminated equipment. The 75 general area air samples were taken at four locations throughout the process area. These samples would be representative of breathing air for workers while they were located in the process area, but not while actively performing operations in hoods or handling thorium or thorium-contaminated equipment. Therefore, all 210 samples together are representative of the overall distribution of exposure potential a worker may have received in the process area, where the upper portion of the distribution is used for routinely- exposed workers and the median would be representative of the non-routinely-exposed workers.

5.0 Evaluation of Gross Alpha Counting

The air samples were counted only for gross alpha activity rather than undergoing a specific radionuclide assay (Monitoring Surveys, 1964). Table 1 presents a comparison of the dose coefficients per unit of inhaled activity (ICRP, 1994) for 100% U-234, the United Nuclear fuel mixture, natural thorium, and freshly-separated thorium. The raw ICRP 68 dose coefficients for each of the respective isotopes are given in Table 2. For the 100% U-234 calculation, the ICRP suggestion of a Type S material for uranium and thorium oxides was taken. The fuel mixture coefficients assume that the fuel was 3% UO_2 and 97% ThO_2 by weight and that the uranium was enriched to 6.5% U-235 by weight. As a result of the enrichment process, it was assumed that the fraction of U-234 increased to 0.0555% by weight. Th-232 and Th-228 were assumed to be in secular equilibrium and the relative contribution of each isotope to the total alpha activity was determined. The calculated activity ratio was used to weight the dose coefficients for each radionuclide.

For natural thorium, an assumption of secular equilibrium was made and contributions from Th-232, Th-228, and Ra-224 are included in the dose coefficients. Finally, the dose coefficients for freshly-separated thorium assume that sufficient time has not elapsed to allow for the in-growth of radium and other daughter products. Therefore, the alpha contribution to total dose from this scenario would be exclusively the result of decaying Th-232 and Th-228.

In all cases, the uranium results in lower dose to each organ per unit intake than thorium. It is also important to note that uranium doses are assessed via urinalysis records. Therefore, the assumption that all alpha counts present on air samples are the result of thorium decay does not reduce the assessed dose due to uranium intakes. Therefore, the assumption that all of the detected alpha activity is due to the decay of thorium is claimant-favorable.

Table 1: Inhalation Dose Coefficient Comparison: Enriched uranium, UNC Fuel Mixture, Natural Thorium, and Freshly-Separated Thorium (Sv/Bq)

(ICRP 68)

Organ	Radionuclide			
	U-234	**UNC Fuel Mixture[a]**	**Natural Thorium[b]**	**Th-232/Th-238[c]**
Adrenals	8.68E-09	1.11E-07	1.50E-07	2.24E-07
Urinary Bladder	8.61E-09	1.08E-07	1.46E-07	2.17E-07
Bone Surface	2.68E-07	3.63E-05	5.09E-05	7.60E-05
Brain	8.61E-09	1.08E-07	1.46E-07	2.17E-07
Breast	8.68E-09	1.11E-07	1.50E-07	2.24E-07
Esophagus	8.69E-09	1.11E-07	1.51E-07	2.24E-07
Stomach	9.14E-09	1.11E-07	1.51E-07	2.23E-07
Small Intestine	9.91E-09	1.12E-07	1.52E-07	2.24E-07
Upper Large Intestine	1.69E-08	1.27E-07	1.77E-07	2.48E-07
Lower Large Intestine	3.20E-08	1.64E-07	2.37E-07	3.10E-07
Colon	2.30E-08	1.42E-07	2.02E-07	2.74E-07
Kidneys	9.91E-08	7.00E-07	9.12E-07	1.36E-06
Liver	3.66E-08	7.36E-07	1.01E-06	1.51E-06
Muscle	8.64E-09	1.11E-07	1.50E-07	2.23E-07
Ovaries	8.61E-09	3.38E-07	4.68E-07	7.00E-07
Pancreas	8.65E-09	1.11E-07	1.50E-07	2.23E-07
Red Marrow	2.78E-08	1.69E-06	2.37E-06	3.52E-06
Extrathoracic Airways	7.40E-05	1.15E-04	1.09E-04	1.60E-04
Lungs	4.03E-05	8.94E-05	1.03E-04	1.44E-04
Skin	8.62E-09	1.08E-07	1.46E-07	2.17E-07
Spleen	8.65E-09	1.11E-07	1.50E-07	2.23E-07
Testes	8.61E-09	3.38E-07	4.68E-07	7.00E-07
Thymus	8.69E-09	1.11E-07	1.51E-07	2.24E-07
Thyroid	8.63E-09	1.11E-07	1.50E-07	2.23E-07
Uterus	8.61E-09	1.08E-07	1.46E-07	2.17E-07

Source: ICRP 68, *Dose Coefficients for Intakes of Radionuclides by Workers* (ICRP, 1994)

[a] The UNC fuel mixture assumes 3% UO_2 and 97% ThO_2 by weight and assumes uranium has been enriched to 6.5% U-235

[b] Natural thorium dose coefficients are determined assuming a secular equilibrium between Th-232, Th-228, and Ra-224.

[c] The Th-232/Th-238 mixture assumes thorium was very recently separated chemically and sufficient time has not elapsed to allow for the in-growth of radium or other decay products.

Table 2: Inhalation Dose Coefficient Comparison: U-234, Th-232, Th-238, and Ra-224 (Sv/Bq)

(ICRP 68)

Radionuclide >	U-234			Th-232		Th-228		Ra-224
Half Life >	2.445E5y			1.405E10y		1.9131y		3.66d
Type >	F	M	S	M	S	M	S	S
AMAD >	5			5		5		5
Organ f1 >	0.02	0.02	0.002	0.0005	0.0002	0.0005	0.0002	0.2
Adrenals	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.70E-08	4.19E-09
Urinary Bladder	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.00E-07	7.80E-07	3.40E-08	4.28E-09
Bone Surface	1.10E-05	2.70E-06	2.70E-07	1.50E-03	1.40E-04	2.80E-04	1.20E-05	7.27E-07
Brain	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.00E-07	7.70E-07	3.40E-08	4.13E-09
Breast	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.70E-08	4.17E-09
Esophagus	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.80E-08	4.20E-09
Stomach	3.90E-07	9.40E-08	9.20E-09	4.40E-06	4.10E-07	7.70E-07	3.60E-08	6.04E-09
Small Intestine	3.90E-07	9.50E-08	1.00E-08	4.40E-06	4.10E-07	7.80E-07	3.70E-08	8.29E-09
Upper Large Intestine	3.90E-07	1.00E-07	1.70E-08	4.50E-06	4.30E-07	1.10E-06	6.50E-08	3.52E-08
Lower Large Intestine	4.00E-07	1.10E-07	3.20E-08	4.60E-06	4.80E-07	1.70E-06	1.40E-07	9.22E-08
Colon	3.90E-07	1.10E-07	2.30E-08	4.50E-06	4.50E-07	1.40E-06	9.70E-08	5.97E-08
Kidneys	4.00E-06	9.80E-07	1.00E-07	2.30E-05	2.40E-06	7.30E-06	3.20E-07	1.76E-08
Liver	1.50E-06	3.70E-07	3.70E-08	2.30E-05	2.50E-06	1.20E-05	5.10E-07	2.30E-08
Muscle	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.50E-08	4.18E-09
Ovaries	3.90E-07	9.40E-08	8.70E-09	1.30E-05	1.30E-06	2.30E-06	1.00E-07	4.43E-09
Pancreas	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.60E-08	4.18E-09
Red Marrow	1.10E-06	2.80E-07	2.80E-08	5.70E-05	6.10E-06	2.20E-05	9.40E-07	6.96E-08
Extrathoracic Airways	3.90E-07	1.20E-05	7.50E-05	1.50E-05	1.10E-04	5.80E-05	2.10E-04	6.17E-06
Lungs	4.00E-07	1.60E-05	4.10E-05	1.60E-05	7.70E-05	1.30E-04	2.10E-04	2.12E-05
Skin	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.00E-07	7.70E-07	3.40E-08	4.13E-09
Spleen	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.60E-08	4.17E-09
Testes	3.90E-07	9.40E-08	8.70E-09	1.30E-05	1.30E-06	2.40E-06	1.00E-07	4.13E-09
Thymus	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.80E-08	4.20E-09
Thyroid	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.10E-07	7.70E-07	3.50E-08	4.15E-09
Uterus	3.90E-07	9.40E-08	8.70E-09	4.40E-06	4.00E-07	7.70E-07	3.40E-08	4.24E-09

6.0 Method for Assessing Doses from Intakes of Thorium

The 210 air samples collected in the Pellet Plant during thorium operations in 1964, which include a combination of breathing zone and general area samples, are believed to be a reasonable representation of airborne thorium concentrations inside the plant during that time. A method for assessing doses resulting from thorium intakes is described in DCAS-TKBS-0008. It employs the data from the 210 air samples to determine the parameters of a lognormal distribution. Operators who routinely handled thorium or operated thorium-processing equipment are assigned the 95^{th} percentile of this distribution as a constant. Supervisors, laborers, and other personnel who routinely entered the processing area but did not routinely handle thorium are assigned the full distribution (geometric mean with GSD as a lognormal distribution). Personnel who did not routinely enter the area are assigned the median value of the distribution as a constant. However, DCAS-TKBS-0008 should be modified to recommend the assignment of thorium assuming a thorium mixture.

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