ORAU Team **NIOSH Dose Reconstruction Project** Technical Basis Document: Basis for the Development of an Exposure Matrix for Tennessee Valley Authority, Muscle Shoals, Alabama, Period of Operation: 1951–1955	Document Number: ORAUT-TKBS-0020 Effective Date: 03/30/2004 Revision No.: 00 Controlled Copy No.: _____ Page 1 of 10
Subject Expert: Jeri L. Anderson Document Owner Approval: Signature on File Date: 03/29/2004 Jeri L. Anderson, TBD Team Leader Approval: Signature on File Date: 03/30/2004 Judson L. Kenoyer, Task 3 Manager Concurrence: Signature on File Date: 03/29/2004 Richard E. Toohey, Project Director Approval: Signature on File Date: 03/30/2004 James W. Neton, OCAS Health Science Administrator	Supersedes: None

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RECORD OF ISSUE/REVISIONS

ISSUE AUTHORIZATION DATE	EFFECTIVE DATE	REV. NO.	DESCRIPTION
Draft	01/05/2004	00-A	New Technical Basis Document for the Tennessee Valley Authority Site. Initiated by Jeri L. Anderson.
Draft	01/18/2004	00-B	Incorporates internal comments. Initiated by Jeri L. Anderson.
Draft	02/20/2004	00-C	Incorporates internal and NIOSH review comments. Initiated by Jeri L. Anderson.
03/30/2004	03/30/2004	00	First approved issue. Initiated by Jeri L. Anderson.

1.0 SITE DESCRIPTION AND OPERATIONAL HISTORY

The Tennessee Valley Authority (TVA) in Muscle Shoals, AL had a cooperative agreement with the Atomic Energy Commission (AEC) to research the recovery of uranium during the production of fertilizer from phosphate ore. The TVA performed this research and development from 1951 to 1955. The facilities involved in the project were laboratories 20 and 21 of the Research and Engineering Office Building and the Pilot Plant Building 411. Also, two laboratories (T-283 and L-275/276) at the National Fertilizer Development Center were used for analytical work associated with uranium production. The TVA owned and operated the laboratories and pilot plant and still maintains ownership of the facility (US DOE 1980, US DOE 1987).

In 1951, TVA began researching processes for producing fertilizer from leached-zone phosphate ore. This ore is a low-grade, high-alumina phosphate ore that contains very small amounts of uranium. The leached-zone ore is normally discarded as waste during phosphate mining processes, but TVA was interested in conserving this material and developing economical methods of producing fertilizer from it. A pilot plant was constructed and a process was developed to extract the phosphate with a mixture of nitric and sulfuric acid. The principal steps of the process were to calcine the ore at 2000° to 2100°F, and then extract the phosphorus pentoxide (P_2O_5) with a mixture of nitric and sulfuric acids. Over 90% of the uranium in the ore was extracted with the phosphate, making it available for recovery. The extraction slurry was then filtered and the filtrate concentrated by evaporation. The uranium was recovered at this point. The concentrated filtrate was ammoniated and granulated, and potassium chloride was added to produce a three-component fertilizer (nitrogen, phosphorus, and potassium). The product was then dried (Hignett et al. 1957).

The research and development on uranium recovery at TVA involved using solvent extraction to recover the uranium from the acid extract, a process that was originally developed by Dow Chemical Company of Walnut Creek, California (Stolz 1958). The process involved using alkyl pyrophosphoric acids to extract uranium from the phosphoric acid. Essentially, the phosphoric acid containing the uranium from the acid plant was reduced with iron. Then a pyrophosphate ester was added which complexed the uranium. The organic complex was then separated from the phosphoric acid solution, which was sent back to the plant for continuation of the fertilizer production process. Sulfuric acid was used to remove calcium, iron and other ions, and the uranium was recovered by reacting with hydrofluoric acid. The recovered uranium in the form of uranium tetrafluoride, UF_4, was dried and shipped to the AEC for further processing (Greek et al. 1957).

Very little actual work was done on uranium recovery from the acid extracts because it was first necessary to ensure that the fertilizer production by this method was marketable and economically feasible. It appears that only about 5.5 pounds of uranium concentrate was produced which contained an average of 2% (range of 0.3-25%) uranium by weight. No accountability records were retained so the disposition of the material and the laboratory and pilot plant equipment is unknown. There was also no information found concerning the radiological status of the facilities at the termination of the project (US DOE 1980, US DOE 1987).

Because no monitoring records have been found for the AEC work at the TVA facility, exposures at the facility are estimated using the Blockson Chemical Company exposure matrix (Anderson 2003). The uranium recovery process at the Blockson facility was a much larger process that also involved recovery of uranium from wet phosphoric acid. The process was slightly different in that the uranium was precipitated using sodium hydrosulfite, and the uranium concentrate was a sodium uranous phosphate as opposed to uranium tetrafluoride (UF_4). Blockson produced 50,000 pounds of U_3O_8 per year compared to the 5.5 pounds produced by TVA from 1951 to 1955. For the purpose of dose reconstruction, the Blockson exposure matrix is scaled down proportionately using the ratio of the

estimated quantity of U_3O_8 produced daily at Blockson to the quantity of UF_4 produced daily at the TVA facility.

2.0 ESTIMATION OF INTERNAL EXPOSURE

The uranium produced at the TVA was the result of process research and development. The nature of research and development work is that it is not necessarily constant and ongoing as is the case with production work. Thus, potential intakes would most likely result from a series of acute exposures rather than a chronic exposure. However, because specific information regarding periods of exposure is unknown, to simplify the dose reconstruction, a single chronic intake is used to approximate multiple acute intakes.

The TVA uranium recovery process was a wet process. Therefore, the primary potential inhalation hazard would have been once the uranium had been recovered from the acid in the form of UF_4 and dried and packaged for shipment. Blockson produced an estimated 50,000 pounds of U_3O_8 per year, or 137 pounds (62.1 kg) per day. The total amount of uranium concentrate reported to be produced at TVA from 1951 to 1955 was 5.5 pounds (2.5 kg), which averages out to 0.003 pounds (1.37 g) per day. Thus, it is assumed that the quantity aerosolized at the TVA facility was a factor of 45,300 lower than the quantity aerosolized at Blockson.

Urinalysis data were available for 25 Blockson Chemical employees. These data were used to fit chronic intakes for each of the workers to determine exposure from inhalation. The resulting chronic intake rates were lognormally distributed with a median of 24 pCi/d and a geometric standard deviation of 1.6. Using the breathing rate of 1.2 m^3/h and an assumed 8-hour workday, the air concentration was estimated to be 2.5 pCi/m^3. The air concentration during operations at TVA is estimated by reducing the air concentration at the Blockson facility by a factor of 45,300, which results in an estimated air concentration of 5.5E-05 pCi/m^3. If the median chronic exposure rate at Blockson of 24 pCi/d is reduced by a factor of 45,300, the resulting chronic exposure rate is 5.3E-04 pCi/d. Assuming a continuous exposure to 5.3E-04 pCi/d for 5 years gives a total inhalation intake of 1 pCi. This level of exposure is considered insignificant and is not included in this dose reconstruction.

Ingestion intakes were estimated using guidance provided by NIOSH/OCAS (NIOSH 2004). The amount of uranium ingested daily is based on the average activity air concentration and is estimated to be 1.1E-05 pCi/d. This quantity is insignificant is therefore not included in this dose reconstruction.

3.0 ESTIMATION OF RADON EXPOSURE

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4.0 ESTIMATION OF EXTERNAL EXPOSURE

The primary radionuclides of interest for potential external exposure are U-238 and daughter radionuclides Th-234 and Pa-234m. For the purpose of this dose reconstruction, it is assumed that there was a potential for external exposure from five sources: submersion in air contaminated with

UF_4 dust, exposure from contaminated surfaces, exposure from contamination on the skin, exposure to drums of UF_4, and the assumed annual occupationally-related diagnostic x-ray.

For estimating external exposure due to submersion in air contaminated with UF_4 dust, the air concentration level was estimated to be 5.5E-05 pCi/m^3 (see Section 2.0). This estimated air concentration and an assumed 2000-hr work year were combined with dose coefficients for U-238, Th-234 and Pa-234m from Federal Guidance Report No. 12 (US EPA 1993). Table 1 shows external annual organ dose estimates due to submersion of workers in air contaminated with UF_4 dust. The cumulative dose from 1951 to 1955 is less than 1 mrem and is therefore not included in this dose reconstruction.

Table 1. Annual organ doses due to submersion in air contaminated with UF_4.

Organ	Annual organ dose (rem)
Adrenal	1.1E-12
U bladder	1.2E-12
Bone surface	3.4E-12
Brain	1.5E-12
Breast	1.8E-12
Esophagus	1.1E-12
Stomach wall	1.3E-12
Small intestine	1.1E-12
Upper large intestine wall	1.1E-12
Lower large intestine wall	1.1E-12
Kidney	1.3E-12
Liver	1.3E-12
Lung	1.4E-12
Muscle	1.4E-12
Ovaries	1.1E-12
Pancreas	1.1E-12
Red bone marrow	1.3E-12
Remainder	1.2E-12
Skin	8.0E-11
Spleen	1.3E-12
Testes	1.5E-12
Thymus	1.4E-12
Thyroid	1.5E-12
Uterus	1.1E-12

There was also the potential for external exposure from surface contamination in the area where the UF_4 was dried and packaged. To be claimant favorable it was assumed that a certain amount of UF_4-contaminated dust was allowed to build up between cleaning of the area. Although it was likely that the experimental work took place in a ventilation hood, to be claimant favorable, it was assumed that no hood was used. To estimate the extent of contamination on surfaces, the air concentration determined from the urinalysis results (5.5E-05 pCi/m^3) was multiplied by the indoor deposition velocity and the assumed deposition time. The indoor deposition velocity is dependent on the physical properties of the room (air viscosity and density, turbulence, thermal gradients, surface geometry) and the particles (diameter, shape, density). Because these characteristics are unknown, the terminal settling velocity was calculated for an aerosol with ICRP 66 default particle size of 5 μm activity mean aerodynamic diameter (AMAD) (ICRP 1994). The calculated terminal settling velocity of 0.00075 m/s was used as an estimate of the velocity of deposition to surfaces in the rooms. This value is within the range of deposition velocities (2.7E-06 to 2.7E-03 m/s) measured in various studies

(US NRC 2002) and is considered claimant-favorable. Also, room air exchange rates, ventilation, and facility housekeeping practices are unknown so it was assumed that there was a steady state air concentration and that surface contamination was the result of 365 days (1 year) of settling.

The estimated surface contamination is multiplied by the dose coefficients for U-238 and daughter radionuclides Th-234 and Pa-234m for contaminated ground surfaces from Federal Guidance Report No. 12 (US EPA 1993). Table 2 shows the external organ dose estimates due to exposure to ground surface contamination. The cumulative dose from 1951 to 1955 for each of the organs is less than 1 mrem and is therefore not included in this dose reconstruction.

Table 2. Annual organ doses due to exposure to ground surface contamination.

Organ	Annual organ dose (rem)
Adrenal	2.5E-10
U bladder	2.8E-10
Bone surface	7.1E-10
Brain	2.6E-10
Breast	3.6E-10
Esophagus	2.3E-10
Stomach wall	2.8E-10
Small intestine	2.6E-10
Upper large intestine wall	2.7E-10
Lower large intestine wall	2.7E-10
Kidney	2.8E-10
Liver	2.8E-10
Lung	2.9E-10
Muscle	3.4E-10
Ovaries	2.6E-10
Pancreas	2.5E-10
Red bone marrow	2.9E-10
Remainder	2.7E-10
Skin	1.3E-07
Spleen	2.8E-10
Testes	3.5E-10
Thymus	2.7E-10
Thyroid	3.0E-10
Uterus	2.6E-10

It was also assumed that there was a potential to receive a shallow dose from electrons due to skin contaminated with UF_4. The amount of skin contamination was calculated by using a measured deposition velocity for 4-μm particles to skin of 0.012 m/s (Fogh et al. 1999, Andersson et al. 2002). For simplification, it was assumed that the material deposited on the skin during an 8-hour period was deposited at the beginning of the shift and the worker took a shower at the end of the shift. The estimated amount of skin contamination was combined with electron dose-rate conversion factors for U-238, Th-234, and Pa-234m for skin in contact with radionuclides (Kocher and Eckerman 1987). The worker was assumed to receive exposure from skin contamination only during the hours worked. Based on these assumptions, the annual dose due to the skin from skin contaminated with UF_4 was estimated to be 1.9E-07 rem. Consequently, this skin dose is not included in this dose reconstruction.

There was also the potential for exposure to the container the uranium concentrate was stored in. It was assumed that all 5.5 pounds of the uranium concentrate (containing 0.3-25% uranium) produced at the site was stored in one container. NIOSH/OCAS provided results of a survey of measurements

of partially filled 55-gallon drums of UF_4 at the DOE facility at Fernald. These drums were assumed to be half-full, which suggests that a drum contains approximately 1500 pounds of UF_4. Measurements were taken at the center side and at the bottom side of the drum. For the Blockson Chemical exposure matrix, dose rates were also calculated for 55-gallon drums of yellowcake using MicroShield© (Grove Engineering 2003) and MCNP (LANL 2003). The mean measurements for the center and the bottom of the Fernald drums were averaged together to get a dose rate of 1.3 mrem/h at the surface. The drum measurements at 1 meter from the surface were below the instrument's detection limit of 0.5 mrem/h. So, to get an estimate of the dose rate at 1 foot and 1 meter from the UF_4 drums, the surface dose rate was divided by the average ratio of the surface-to-1-foot- and surface-to-1-meter-calculated dose rates obtained with MicroShield© and MCNP. Table 3 shows the results of the calculations for the UF_4 and yellow cake drums.

Table 3. Results of calculations of the dose rates from a drum of UF_4 and a drum of yellowcake.

	Dose rate		
	Surface (side)	**30 cm (1 ft.)**	**1 m**
MicroShield (mR/h)	5.5E-01	8.4E-02	2.2E-02
MCNP (mrem/h)	5.6E-01	1.3E-01	3.6E-02
UF4 (mrem/h)	1.3E+00	2.4E-01	6.8E-02

The container of UF_4 at the TVA was assumed to contain a maximum of 5.5 pounds, so using the dose rate from a 55-gallon drum containing around 1500 pounds of UF_4 to estimate annual dose is very claimant favorable. It was assumed that the worker was 1 meter from the container of UF_4 for 1 hour per day, 5 days per week, and 50 weeks per year. The resulting annual dose due to exposure to the container of 1500 pounds of UF_4 was calculated to be 0.017 rem.

The organ doses were calculated by multiplying the estimated annual dose of 0.017 rem by the "Ambient Dose Equivalent (H*(10)) to Organ Dose (HT)" photon dose conversion factors found in Appendix B of the NIOSH External Dose Reconstruction Implementation Guideline (NIOSH 2002a). The exposure geometry was assumed to be anterior-posterior (AP) and the dose rate was divided equally between photons with E=30-250 keV and photons with E>250 keV. Table 4 below shows the annual organ doses due to the potential exposure to the container of UF_4.

Table 4. Annual organ doses due to exposure to the container of UF_4.

	Annual organ dose (rem)		
Organ	**Photons E=30-250 keV**	**Photons E>250 keV**	**Total**
Bladder	8.0E-03	7.7E-03	1.6E-02
Red bone marrow	4.1E-03	6.3E-03	1.0E-02
Bone surface	7.8E-03	6.7E-03	1.4E-02
Breast	8.1E-03	8.2E-03	1.6E-02
Colon	6.8E-03	7.4E-03	1.4E-02
Esophagus	4.4E-03	6.5E-03	1.1E-02
Eye	8.0E-03	7.7E-03	1.6E-02
Ovaries	6.2E-03	7.2E-03	1.3E-02
Testes	9.2E-03	8.3E-03	1.7E-02
Liver	6.8E-03	7.5E-03	1.4E-02
Lung	6.3E-03	7.3E-03	1.4E-02
Remainder	5.7E-03	6.9E-03	1.3E-02
Skin	5.7E-03	7.3E-03	1.3E-02
Stomach	8.1E-03	7.8E-03	1.6E-02

Thymus	9.0E-03	7.8E-03	1.7E-02
Thyroid	9.2E-03	8.5E-03	1.8E-02
Uterus	6.5E-03	6.9E-03	1.3E-02

The organ doses in the second and third columns of Table 4 are entered into the NIOSH IREP program assuming a chronic exposure and a constant distribution. The organ doses in the second column are attributed to photons with E=30-250 keV and the organ doses in the third column are attributed to photons with E>250 keV.

The assumption was also made that workers received an annual occupationally related diagnostic X-ray. The exposure geometry was assumed to be posterior-anterior (PA) (NIOSH 2002a). Table 5 below shows the annual organ doses due to the assumed annual diagnostic chest X-ray (Kathren et al. 2003). The values in Table 5 are entered into the NIOSH-IREP program as the annual dose due to an acute exposure to photons (E=30-250 keV). The distribution is assumed to be normal with a standard deviation of +/- 30%.

Table 5. Annual organ doses due to the assumed annual diagnostic chest X-ray.

Organ	Annual dose (rem)
Thyroid	3.48E-02
Eye/Brain	6.40E-03
Ovaries	2.5E-02
Liver/Gall Bladder/Spleen	9.02E-02
Urinary Bladder	2.5E-02
Colon/Rectum	2.5E-02
Testes	5.0E-03
Lungs (male)	8.38E-02
Lungs (female	9.02E-02
Thymus	9.02E-02
Esophagus	9.02E-02
Stomach	9.02E-02
Bone Surfaces	9.02E-02
Remainder	9.02E-02
Breast	9.80E-03
Uterus (embryo)	2.5E-02
Bone Marrow (male)	1.84E-02
Bone Marrow (female)	1.72E-02
Skin	2.70E-01

5.0 Estimation of Exposure to Residual Radioactivity

According to the NIOSH residual radioactivity report, there is very little potential for radioactive contamination at the TVA facility beyond the period of AEC operations (NIOSH 2002b). Therefore, exposures due to residual radioactivity were not included in this dose reconstruction.

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