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Appropriateness of using 1997 Gross Alpha Air Sampling Data as a Method of Bounding Thoron Intakes at the SRS H-Area Tank Farm Between 1972 and 1994	ORAUT-RPRT-0081	Rev. 00
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04/07/2017	00	New report for the analysis of Savannah River gross alpha air sampling data as a method of thoron intake bounding between 1972 and 1994. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by James M. Mahathy.

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ACRONYMS AND ABBREVIATIONS

CFR	Code of Federal Regulations
Ci	curie
cm^3	cubic centimeter
DOE	U.S. Department of Energy
dpm	disintegrations per minute
ft	feet
FTF	F-Area Tank Farm
hr	hour
HTF	H-Area Tank Farm
L	liter
NIOSH	National Institute for Occupational Safety and Health
ORAU	Oak Ridge Associated Universities
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
WL	working level
µCi	microcurie

1.0 INTRODUCTION

During a February 2014 meeting of the Advisory Board on Radiation and Worker Health‘s Work Group on the Savannah River Site (SRS), discussion was held regarding whether air sampling measurements at the H-Area Tank Farms would bound thoron exposures for thorium storage areas. The discussion centered on whether measurements made during the mid- to-late 1990s could be used to bound thoron exposures for the prior two decades, or whether there were modifications made to the tanks that would impact whether these later measurements would be bounding for workers in the Tank Farm area.

As an outcome of this discussion, NIOSH committed to collect a representative selection of air samples from the tank farm area over the time period under consideration. Initial counts, six-hour counts, and 24-hour counts would then be evaluated to determine the thoron component of the total alpha activity on the filter. Measurements from the 1970s and 1980s would then be compared to the measurements made by the site during the 1990s. The calculated airborne concentration would then be used to assign a dose to workers in the area of the tank farm.

1.1 BACKGROUND

Since the early 1950s, the primary mission of the SRS has been to produce nuclear materials primarily for national defense and deep space missions. A legacy of the SRS mission was the generation of liquid waste from chemical separations processes in both F and H Areas. Since the beginning of SRS operations, an integrated liquid waste system consisting of several facilities designed for the overall processing of liquid waste has evolved. Two of the major components of this system are the H-Area Tank Farm (HTF) and F-Area Tank Farm (FTF), which are near the center of the site. In H Area, neptunium, uranium, and other radionuclides were separated from irradiated fuel and target assemblies using chemical separations processes. The tank farms, which store and process the chemical separations waste, include waste tanks, evaporators, transfer line systems, and other ancillary structures (SRR 2015).

There are 29 waste tanks in HTF. The waste tanks are built of carbon steel and reinforced concrete, but the designs vary. There are four principal types of waste tanks in the HTF designated as Types I, II, III, and IV. Type III tanks are sub-categorized as Type III and IIIA, based on the cooling mechanisms installed. The waste tanks were constructed at different times, with design features improving over time. The HTF waste tank numbering along with their design type is as follows (SRR 2012):

- Type I. Tanks 9 through 12.
- Type II. Tanks 13 through 16.
- Type IV. Tanks 21 through 24.
- Type III. Tanks 29 through 32.
- Type IIIA. Tanks 35 through 43 and 48 through 51.

During the 1960s, the H-modified chemical separations process in 221-H was used to recover ^{233}U from irradiated ^{232}Th targets. Quantities of ^{232}Th are known to exist in Tanks 11, 12, 13, 14, 15, 40, 42 and 51 (Thames 2000). A consequence of the storage of thorium waste is an increase in the levels of thorium progeny, including ^{220}Rn (thoron) (Epperson 1995a).

In 1995, SRS noted that the radon/thoron emissions from Tank 15 were higher than normal because of a decrease in the cooling capacity of the in-tank condenser (Epperson 1995b). This observation prompted an in-depth study, conducted by SRS in 1997, that evaluated exposure potentials due to periodic venting releases from Tanks 12 and 15 by collecting samples from several areas around the two tanks. These samples were analyzed via gamma spectroscopy, making mathematical derivation of the thoron progeny concentration unnecessary (Sigg et al. 1997).

A series of results from air samples taken at the tank farm during 1974 to 1979 was retrieved from the site (DuPont 1974–1979). However, many of these records did not include an entry for a count taken 6 hours after sampling. Of 20 results specific to the tank farm, 9 had both the 6-hour counts and the 24-hour counts. This limited quantity of data was not sufficient to draw any statistical conclusions from its analysis.

Historically, the dose contribution from thoron and its progeny has been considered negligible in comparison with other sources of dose. However, recent research has suggested that the deposition of thoron progeny within the respiratory tract may contribute to a non-negligible portion of the total dose (Haanes et al. 2016). There is no indication that an attempt was made to sample for thoron or thoron progeny at SRS before 1995 (Epperson 1995a; Epperson 1995b; Sigg, Ross, and Weber 1997). However, there are methods available for estimating airborne thoron concentrations based on routine air samples (Epperson 1995a).

2.0 METHODOLOGY

Because radon is a noble gas, it has no significant interaction with air sampling filter papers. However, the thoron progeny are solid matter that can aggregate to form larger particles, and can attach to airborne dust particles in the atmosphere. It is these particles that are collected during air sampling and analyzed through either spectroscopy or gross counting.

Air sample results available to NIOSH were analyzed for gross alpha activity. A challenge associated with the use of gross alpha results for estimating airborne thoron concentrations is the presence of ^{222}Rn and long-lived alpha-emitting radionuclides such as plutonium that could lead to overestimating thoron concentrations. At SRS air samples were counted for the first time 6 hours after the sample collection period, allowing for the alpha-emitting ^{222}Rn progeny to decay away. A second count was completed 24 hours after the filter was removed from the sampler. Decay and build-up of thoron daughters must be accounted for when calculating the concentration of thoron daughters on the filter at the end of collection period. Concentrations (µCi/cc) of ^{212}Pb on the filter at the end of collection were derived using the methodology given in ORAUT-RPRT-0084 (ORAUT 2017) for each set of gross alpha 6 and 24 hour gross alpha count results for the years 1982 to 1983, 1984 to 1985 and 1990. Only those data points that corresponded to Tanks 11 to 15, 21, 40, 42, and 51 were evaluated because these specific tanks were the ones that were identified as receiving significant quantities of thorium wastes (Thames 2000; O'Bryant and Weiss 2003).

Note the count data recorded on the air monitoring results forms are measured count rates. From the measured count rates, activities were calculated using an alpha scaling factor reported for each count by SRS and then converted to microcuries. The alpha scaling factor included the collection efficiency, including self-absorption on the filter, and geometry of the counter (Patterson 1957, DuPont 1986). Concentrations of thoron daughters were then derived using the methodology and equations given in ORAUT-RPRT-0084 (ORAUT 2017).

3.0 REPRESENTATIVENESS

3.1 DATA AND ANALYSIS

The position of workers relative to the location of sample collection must be considered when using air sampling to bound dose. However, the position of workers or the heights of catwalks is rendered inconsequential by basing the concentration to be assigned on data from the primary exhaust and annulus vents. It is unlikely that a configuration could exist that would place workers in an area with a higher thoron concentration than that found in the tank exhaust.

Derived concentrations for 1982-1983, 1984-1985, and 1990, as well as published thoron concentrations for 1997, were statistically analyzed. Derived values of 0.00 µCi/cc were censored at 0.2 µCi/cc, which correlates to the detection level that all samples were being assessed against. Summary statistics for each year are given in Table 3-1. Summary statistics for each year include the number of data points, geometric mean, 95th and 99th percentile values.

Table 3-1. Airborne thoron progeny concentrations (µCi/cm^3) by period.

Period	**N**	**n**	GM	**95th percentile**	**99th percentile**	**Reference**
1982–1983	184	183	1.24×10^{-12}	9.03×10^{-12}	2.05×10^{-11}	DuPont 1982–1983
1984–1985	184	174	3.97×10^{-12}	2.89×10^{-11}	6.58×10^{-11}	DuPont 1982–1983
1990	134	113	8.98×10^{-13}	6.95×10^{-12}	1.62×10^{-11}	WSRC 1990a, 1990b
1997	22	22	2.84×10^{-11}	2.67×10^{-09}	1.76×10^{-8}	Sigg et al. 1997

A significant source of variation in the datasets is not readily obvious from values given in the table: all of the available data points from 1984 to 1985 are direct measurements from either the primary vent of the tank, the annulus vent of the tank, or the tank purge exhaust stream itself. Of the 174 data points for that year, 110 were taken from the exhaust stream and annulus vent of Tanks 12 and 15, the two tanks holding the highest total thorium activities. Similarly, available data from 1982 to 1983 contain 90 data points from the annulus vent and exhaust stream. None of the available data points from 1990 indicate that they were sampled directly from the exhaust stream. Rather, the data points from 1990 are identified as having been taken from the annulus vent and the general air surrounding the tank, resulting in lower airborne concentrations as compare to the 1982-1983 and 1984-1985 values. Data for 1997 were recorded from the study conducted by SRS. Samples were collected from several areas around Tanks 12 and 15 during periodic venting of tank gases and are higher because of a decrease in the cooling capacity of the in-tank condenser (Sigg et al. 1997).

Probability plots of the tabulated data for each year are presented in Figures 3-1 through 3-4, below.

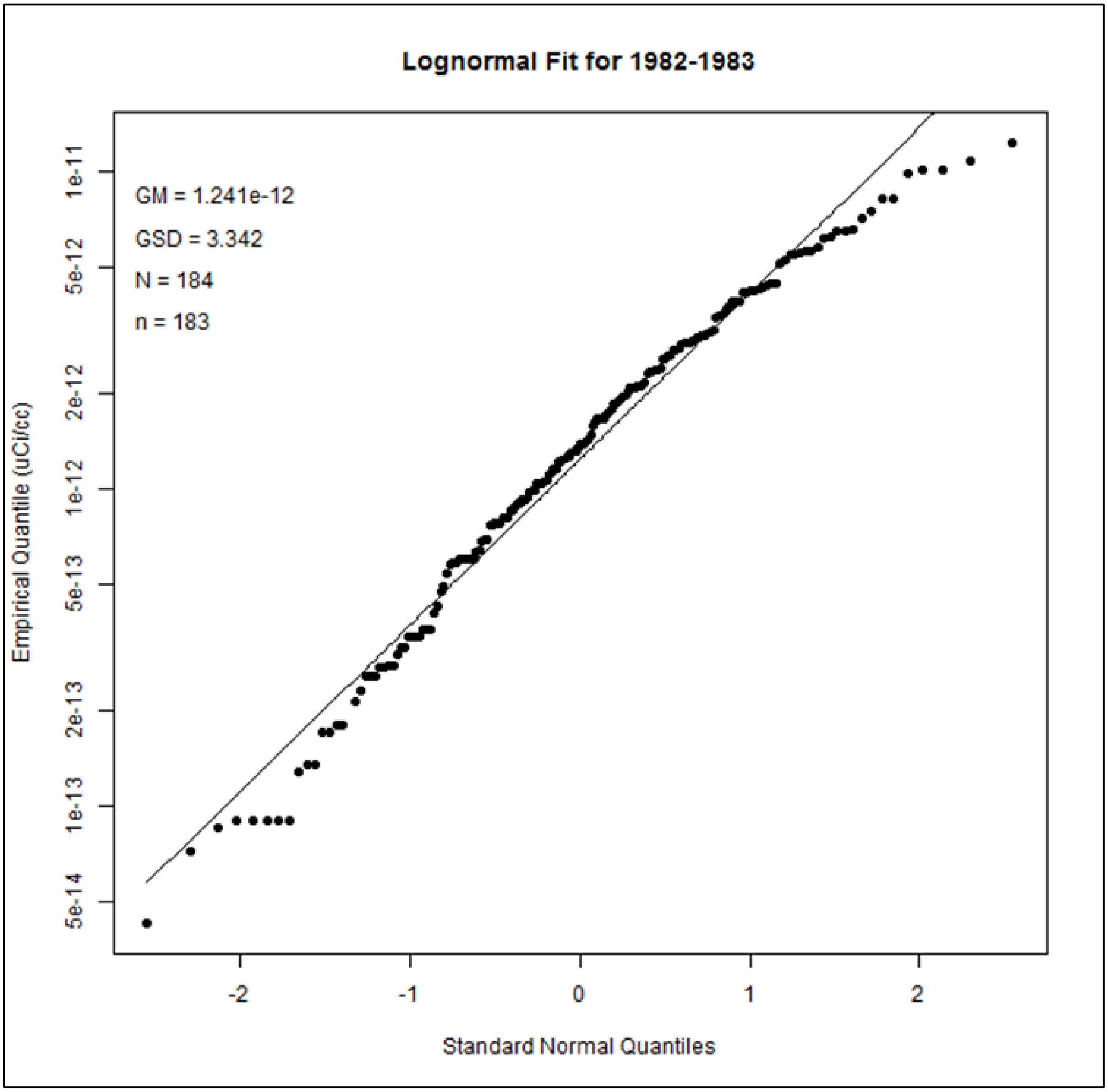

Figure 3-1. Lognormal probability fit of 1982–1983 air sample data.

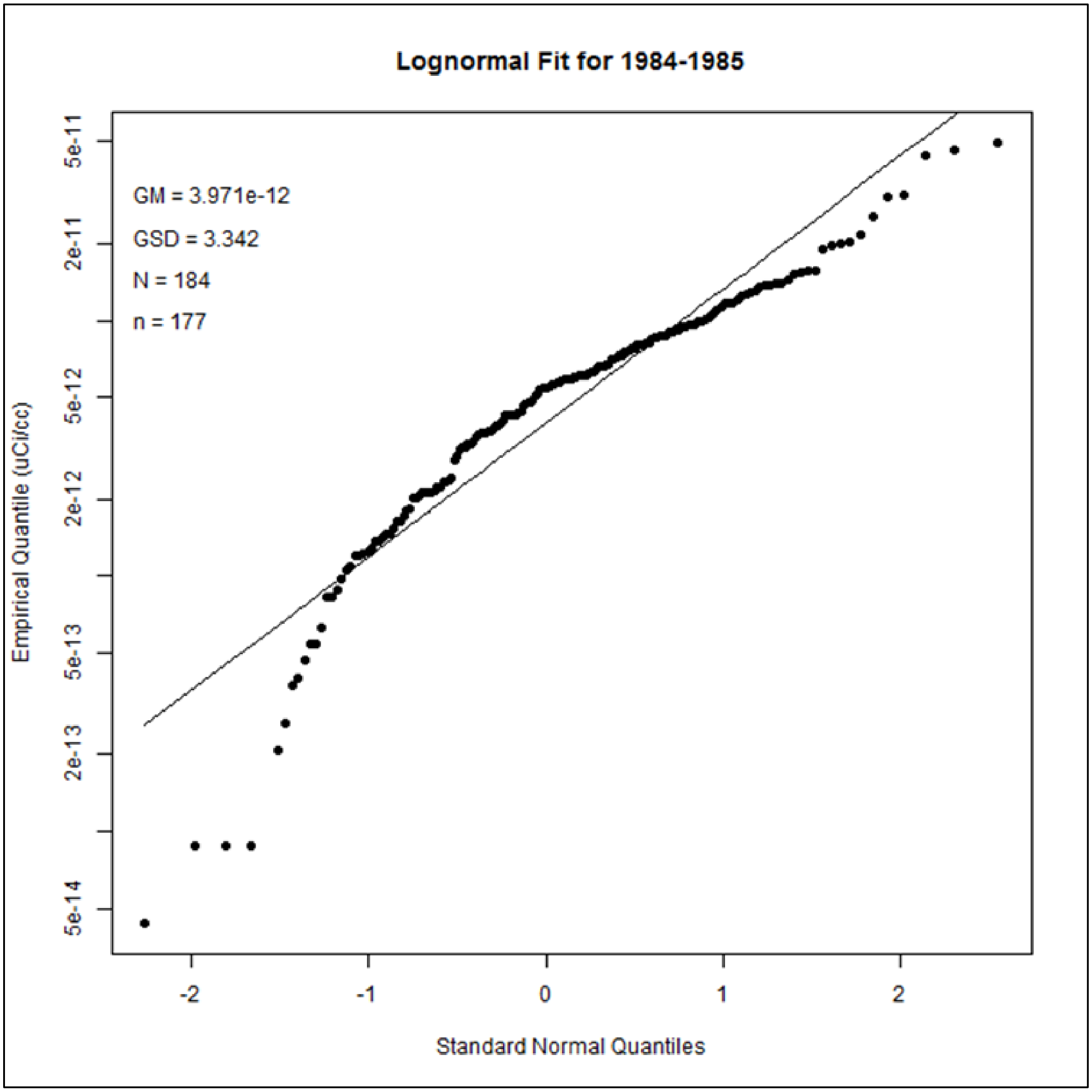

Figure 3-2. Lognormal probability fit of 1984–1985 air sample data.

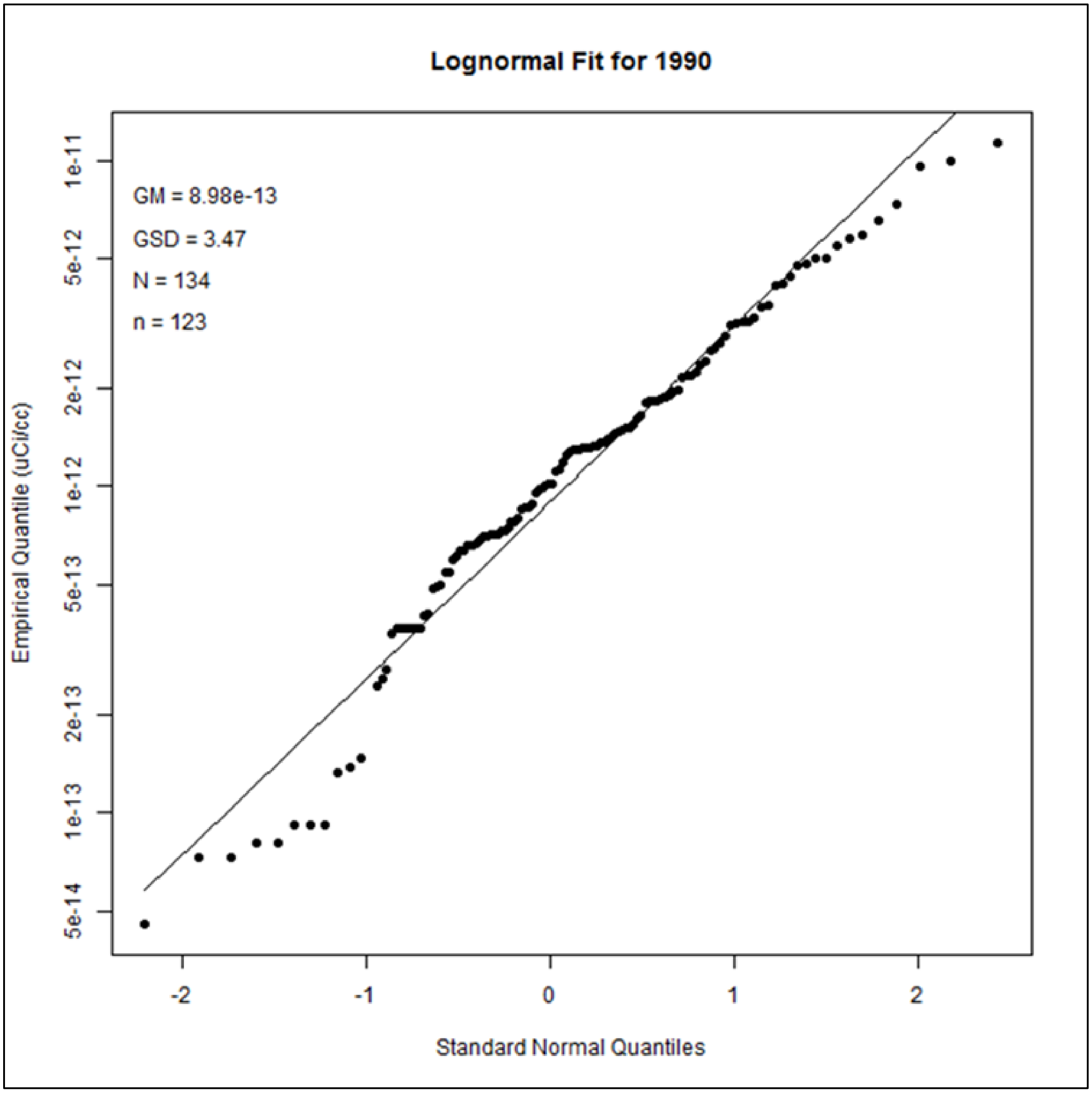

Figure 3-3. Lognormal probability fit of 1990 air sample data.

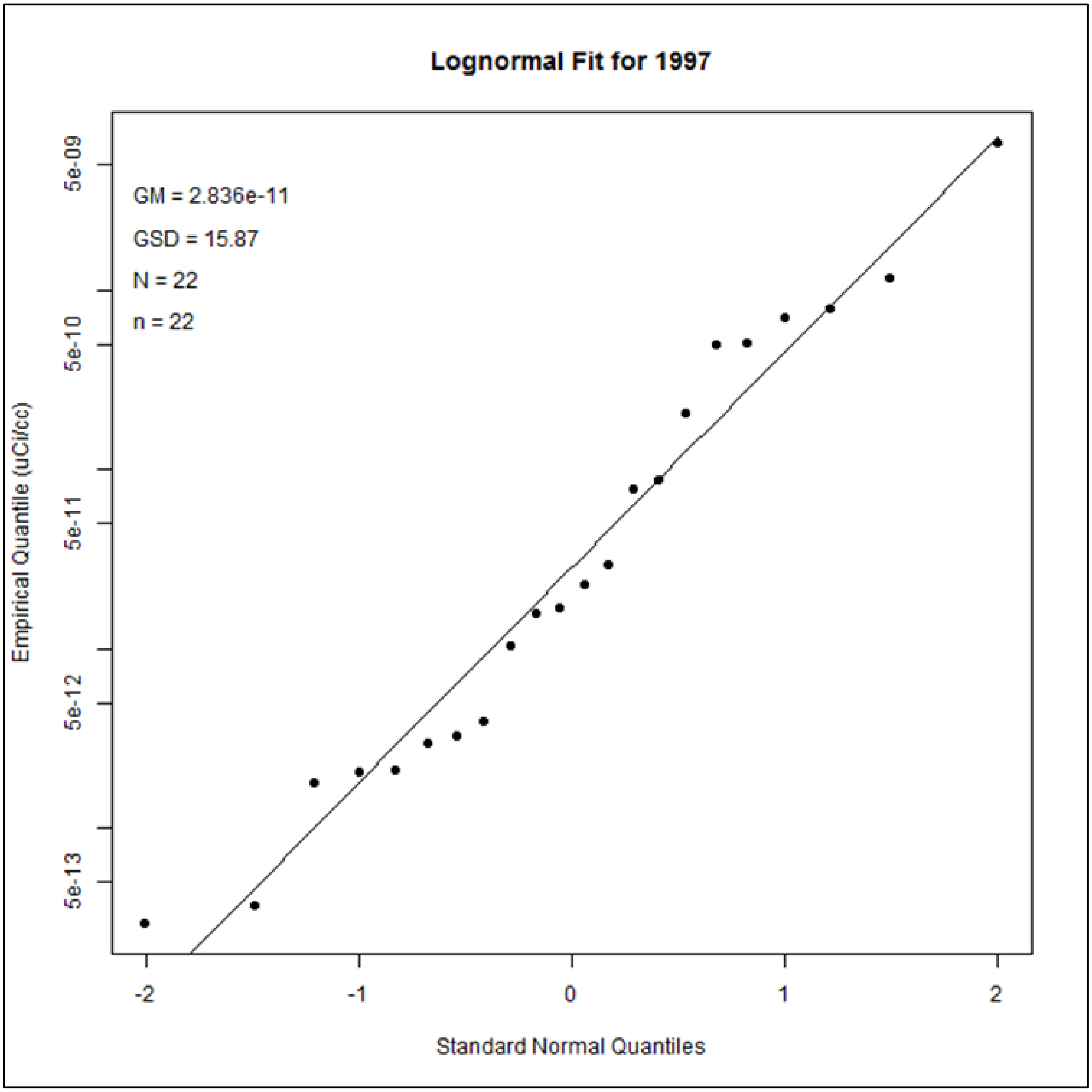

Figure 3-4. Lognormal probability fit of 1997 air sample data.

4.0 CONCLUSIONS

Air concentrations of thoron measured at H Area tanks in the 99th-percentile for the periods 1982-83, 1984 to 1985 and 1990 are less than 1% of the derived air concentration for thoron when its progeny are present (9.0×10^{-9} µCi/cm^3). The majority of the data used to arrive at these values are from measurements taken at the point of exhaust, maximizing the concentrations compared to open air. Both tanks with the greatest concentrations of thorium (Tanks 12 and 15) had exhaust stacks that rose 10 ft above ground level. In addition, Tank 15 had a catwalk at a height of 15 ft above ground level (Sigg et al. 1997). Such configuration makes it unlikely that a worker would be positioned in such a way as to be exposed to thoron progeny concentrations approaching those at the tank exhaust. However, the exhaust values represent a conservative upper bound of potential thoron progeny exposures. Further, measurements taken at the emission point ensure that assigned upper bound doses are independent of the worker location within the tank farm.

As previously discussed, the radon/thoron emissions from Tank 15 measured in the later 1990s were higher than normal because of a decrease in the cooling capacity of the in-tank condenser (Epperson 1995b). This can be seen in the concentrations of thoron daughters derived during the 1997 study, which are two to three orders of magnitude higher than concentrations derived from air monitoring conducted during the previous decades. NIOSH concludes that concentrations of thoron daughters reported in the 1997 study are sufficient to bound thoron exposures of workers in thorium storage areas.

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