Basis for a Bounding Estimate of Radon Exposure at Allied Chemical and Dye

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Introduction

As part of the 17th set of dose reconstruction reviews, the Advisory Board tasked Sanford Cohen and Associates (SC&A) with performing a blind review of a case with employment at the Allied Chemical and Dye Corporation. In their December 2014 report, SC&A raised a question regarding the internal exposures that NIOSH used in the dose reconstruction for this case. In particular, SC&A believed that the radon exposure estimate assigned for this case was not necessarily bounded by the data that was used by NIOSH. The radon levels uses for this dose reconstruction were based on 10% of the 95 percentile values contained in ORAU-OTIB-0043. This white paper evaluates the potential for radon exposure at Allied Chemical and Dye, given what is known about the nature of the radium source-term and the laboratory experiments that were performed there. In that light, the suitability of using the existing phosphate plant data in ORAU-OTIB-0043 to bound exposure to radon with sufficient accuracy is evaluated.

Background on Allied Chemical and Dye

According to a 1977 DOE letter, Allied Chemical and Dye Corporation's plant in Claymont Delaware was involved in research and development and small pilot scale operations in the recovery of uranium. This work, which was conducted in the early 1950s, took place under two Atomic Energy Commission (AEC) contracts.[1] DOE interviews with former workers indicated that the Claymont work focused on filtration development and that, less than 2 to 10 pounds of uranium were produced (SRDB 16503). The research under these contracts established the basis for the designation of Allied Chemical and Dye as a covered Atomics Weapons Employer facility under EEOICPA. Because the contracts could not be located, and the phosphoric acid plant was in existence until early 1970, the covered period on the DOE website is listed as the "early 1950s – late 1960s." For purposes of dose reconstruction, NIOSH has interpreted this to be January 1, 1950 through December 31, 1969. There is also a residual contamination period at Allied Chemical that covers 1970 through 1977.

[1] The AEC contract numbers were: AT (49-1)-610 and AT (49-6)-913.

At the time of the dose reconstruction, the information above was all that was available on the nature of the AEC activities at Allied Chemical and Dye. Recently, however, NIOSH has discovered an Allied Chemical and Dye progress report to the AEC Raw Materials Division (the sponsor of Allied's research) that sheds some light on type of research performed at Allied Chemical and the magnitude of that activity (SRDB 159354). The report, which is a summary of work accomplished through March of 1954 under AEC contract AT (49-6)-913, discusses progress in two areas: leach zone filtration studies and uranium extraction from phosphoric acid. Because the radium source term had been removed from the phosphoric acid, the leach zone filtration studies is the only work that could generate a radon exposure potential.

Evaluation of Radon Exposure Potential using Source-Term Information

The leach zone ore studies involved laboratory-scale research into the optimum conditions under which gypsum could be precipitated and quantitatively filtered. To accomplish this, a typical batch was reported to use 150 grams of leach zone ore[2]. Although the number of batch runs that were made are not reported in the study, the effect of 11 different factors or variables were reported to have been evaluated. Under future work, the report states that Allied Chemical and Dye intended to verify the reported results in 45 additional batch runs.

There are several possible approaches that can be used to estimate or bound the magnitude of the radon exposure associated with the amount of material used in the experiments described above. First, a simple source term model can be used to produce an estimate of the magnitude of the potential exposures.

Assuming that each experiment used 150 grams of leach zone ore containing 0.014% natural uranium by weight[3], it is possible to estimate the amount of radon that would be in a laboratory room at equilibrium. The only parameters required to place an upper bound of the estimate are the air ventilation rate and the size of the room. If we use an air turnover rate of 1 exchange per hour and a small room of dimensions 10 foot length and width and a ceiling height of 8 feet, the equilibrium concentration of radon in the room would be 0.002 pCi/L or 9.3E-06 WL, assuming 40% progeny equilibrium in indoor air. If we assume that all 45 batch runs are processed at the same time in the same room, the concentration would be 0.105 pCi/L or 0.00042 WL if there is 40% equilibrium between the radon gas and its progeny. We consider these calculated values to be upper estimates because:

- It is assumed that the amount of radon emanating from the leach zone ore is 100%;

[2] Leach zone ore is from the upper layer of a phosphate ore deposit that is not used in the production of phosphoric acid. It is normally scraped off the surface and placed in piles at the mine site. The uranium content of leach zone ore is not considerably different from the phosphate ore that is mined.
[3] For this analysis uranium and radium are considered to be in secular equilibrium.

- It is likely that the laboratory room was larger than the dimensions used; and
- The filtration experiments were likely to have been done in a laboratory hood, given that the experimental protocol called for digestion of the sample in 140 to 208° Fahrenheit nitric acid.

As part of their review of the Allied Chemical dose reconstruction, SC&A prepared a source term calculation dated June 30, 2015. Their source-term analysis was significantly more refined that the simple, bounding analysis discussed above. It considered a number of additional parameters, including the possible production of ten pounds of uranium material per year, variation in room ventilation rates, and the emanation fraction of radon from dry phosphate ore, from wet phosphate ore and from sulfuric acid. The results of their analysis was a lower estimated radon concentration of 0.0042 pCi/L or 0.000017 WL assuming 40% equilibrium between radon gas and its progeny.

Radon levels in Phosphate Plants

As previously stated. NIOSH used the radon data in ORAUT-OTIB-0043 to bound exposures at Allied Chemical. ORAU-OTIB-0043 relied on the radon data that were contained in a Florida Institute of Phosphate Research (FIPR) report, which were collected a number of phosphate plants in Florida. The 130 applicable radon values in the report were found to fit a lognormal distribution with a geometric mean of 0.75 pCi/L and a geometric standard deviation of 1.99. The 95th percentile of the distribution is 2.33 pCi/L or 0.0093 WL if a 40% equilibrium factor is applied[4]. To account for the difference in scale between a phosphate production facility and the levels of radium that were present at Allied Chemical, the dose reconstruction used a value of 10% of this, or 0.00093 WL.

In 1983, ten radon measurements were taken at various locations at the Blockson Chemical plant in Joliet, Illinois, while the plant was still in operational mode. The highest value, which was taken at the Sodium Tri Polyphosphate (STPP) area, was reported to be 0.0042 WL (SRDB 30623). Although the Blockson plant and the Florida-based facilities that were included in the OTIB-043 analysis were different, with the Florida plants likely to have been more ventilated, the upper values are only about a factor of 2 apart (0.0042 vs 0.0093 WL). In fact, it of interest to note that the value measured at Blockson is approximately one-half of the 95th percentile value in the FIPR report. Reducing the measured levels at Blockson Chemical by a factor of 10, as was done for the FIPR data, results in a value of 0.00042 WL.

[4] The use of 40% equilibrium is consistent with literature values cited for indoor air.

Comparison and Conclusion

Table 1 provides a comparison of the estimated exposure radon exposure at Allied Chemical that were calculated above.

Basis of Estimate	Value (WL)	Comment
NIOSH Source-term (1 sample processed at a time)	9.3E-06	Upper bound using 150 gram samples and small room
NIOSH Source-term (45 samples processed at a time)	0.00042	Upper bound using 150 gram samples and small room
SC&A Source-term	0.000017	Allows for the annual production of 10 pounds of uranium
OTIB-043 adjusted by a factor of 10	0.00093	95th percentile of measured data
Blockson Chemical adjusted by a factor of 10	0.00042	Largest value measured

It can be seen in the comparison that a value of 10% of the FIPR production facility measurements provides a bounding estimate when compared to both the NIOSH and SC&A source term calculations. Given the levels of uncertainty in the above estimates, NIOSH believes that it is claimant favorable to continue the use of the adjusted OTIB-043 value in assessing radon exposures at Allied Chemical. While the difference between the source-term estimates and the measured plant data is on the level of an order of magnitude, this is not unreasonable given the low levels of exposure predicted by both.