Radon Emissions from Feed Materials Production Center K-65 Silos

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INTRODUCTION

At the September 3, 2014, Fernald (Feed Materials Production Center) Work Group (WG) meeting NIOSH agreed to address the question of radon release rates from the K-65 silos in light of Sanford Cohen &Associates' (SC&A) reports to the WG in 2008 and 2010. Those reports questioned the radon release values in the Fernald site profile. The K-65 radon release values in the Fernald site profile (Table 4-9 of ORAUT-TKBS-0017-4) are based on Radiological Assessment Corporation's (RAC) "Radionuclide Source Term and Uncertainties" section of The Fernald Dosimetry Reconstruction Project. That work was directed at estimating doses to neighbors of the Fernald facility, but the radon release quantities should be useful for estimating radon exposures on the Fernald site as well.

The Fernald operating contractor made a number of modifications to the K-65 silos that affected the rate of radon emission. Those were described in RAC-1995a and were reproduced in SCA-2008. They are presented in Table 1 below for convenience.

Table 1. Summary of Historical Changes to the K-65 Storage Silos
(Source: RAC 1995a, Appendix J)

Date	Repairs or Improvements
May 1964	Cracks in silo walls were patched, waterproofing sealant applied, and earthen berm constructed to counterbalance material inside silos
End of June 1979	Openings in silo domes, including the gooseneck pipe and other penetrations, were sealed, with gaskets installed, to prevent Rn emissions
June 1983	The earthen berms were enlarged to correct erosion problems
Early 1986	Dome covers added to protect the center sections of the silo domes; neoprene membrane layer applied to part of Silo 2
November 1987	Radon Treatment System* installed to treat displaced Rn during work on Silos (not continuously operated)
December 1987	Rigid, polyurethane foam layer and urethane coating applied to exterior of silo dome surfaces to weatherproof the Silos
November 1991	Addition of layer of bentonite on top of K-65 material in Silos

*The Radon Treatment System (RTS) is a system that pumps air in the headspace of the silos through a series of calcium sulfate and charcoal beds, which adsorb Rn-222.

EXECUTIVE SUMMARY

NIOSH spent considerable time comparing SC&A's estimate of potential radon releases (SCA-2008) to the radon release study performed by Radiological Assessments Corp. (RAC-1995a). There are some important points of agreement in the SC&A and RAC approaches:

- Both agree that the removal of the gooseneck pipe and sealing of other penetrations, which was done in June 1979, significantly decreased the rate at which radon was released to the environment.
- Both agree that most of the radon that entered the silo headspace prior to June 1979 was released to the environment, although RAC maintains that slightly more was retained in the silos than SCA does.
- Both compared direct radiation readings taken before 1979 when the silos were sealed to direct radiation readings taken in 1987 after the Radon Treatment System (RTS) was operated and to direct radiation readings taken after the silos were sealed but before RTS operation to evaluate the difference in radon headspace concentrations before sealing vs. after sealing. They interpret the direct radiation reading differences somewhat differently, though.

The key difference between the two approaches is the position they take on the fraction of the radon generated in the K-65 residues that diffuses into the silo headspace.

SC&A looks at the difference in the average concentrations of Ra-226 and Pb-210 that were reported from a 1991 sampling, and concludes the basis for this discrepancy is a large portion of the intervening Rn-222 diffused from the residues into the headspace and then environment. Once it diffused from the residues, it was no longer available to decay to Pb-210. SC&A-2008 estimates that 58% of the radon in silo 1 and 28% of the radon in silo 2 diffused into the silo headspace, and that about 97% of the radon that diffused into the headspace was released to the atmosphere. SCA-2008 estimates the radon release rate before the silos were sealed in 1979, but not for the period 1979-1987. It should also be noted that SCA-2008 uses Ra-226 and Pb-210 residue concentration values from ORAUT-TKBS-0017-5. Had it used the concentrations from the 1991 sampling, it would have estimated that roughly 60% of the radon generated in the residues in each silo would have diffused into the headspace air (63% for silo 1, 59% for silo 2), and would have calculated higher total releases of radon.

RAC was aware of the difference in Ra-226 and Pb-210 concentrations (the values are included in their 1995 Task 2/3 report), but chose a different method for estimating how much radon might have diffused from the residues into the headspace. RAC's estimate for radon release during the period 1979-1987 relies on using measured headspace radon concentrations and measured headspace air temperatures from 1987 to estimate the radon release rate due to thermal expansion of air in the silos; and calculating how much radon would diffuse through the concrete domes based on headspace concentrations and some

knowledge about the quality of the concrete in the K-65 silo domes. They then calculate the headspace concentrations for the pre-1979 unsealed silos. They do this by estimating the rate at which radon diffuses from the K-65 residues into the headspace, then using their estimate of radon concentration to calculate a total removal rate of radon from headspace air. After accounting for radioactive decay, the remaining removal rate is due to release to the environment. For the years before 1979, RAC-1995a estimates that 93% (median of the estimate's distribution) of the radon in the headspace was released to the environment. For the 1979-1987 period, RAC-1995a estimates that 16% of the radon in the headspace was released to the environment. RAC-1995a does not provide separate release estimates for each silo but rather averages values between the two silos and calculates a total release rate. For the 1979-1987 period, RAC-1995a estimates that 2.4% of the radon generated in the K-65 residues diffused into the headspace, and prior to 1979, 2.7% of the radon generated in the residues diffused into the headspace. The reason for the difference is that the radon concentration in the silo headspace constrains additional diffusion from the residues.

NIOSH has not found a technical error in either SCA's or RAC's approach, and cannot reconcile the widely disparate release values. If the RAC release estimates are correct, then NIOSH can only explain the relative Ra-226 and Pb-210 residue concentrations from the 1991 sampling is through a systematic error in the sampling and analysis of the samples. On the other hand, if SCA's pre-1979 release estimates are correct, then the sealing of the silos would have had to have reduced the rate of radon diffusion from the residues into the headspace by an amount that is not credible. Additionally, a radon diffusion rate into the headspace on the order of 60% is entirely inconsistent with the behavior of radon in various radium-bearing materials. Consequently NIOSH's view is that the release values in RAC-1998 constitute the best estimate of releases. However NIOSH does propose to adopt the 95th percentile estimate of release, rather than the median, as the value to be used in dose reconstructions.

SC&A EVALUATION OF RADON RELEASE

SC&A's analysis of radon emissions from the K-65 silos relies primarily on two types of data: 1) direct radiation surveys taken at the tops of the silos at different times; and 2) analysis results for Ra-226 and Pb-210 in the K-65 residues as reported from a 1991 sampling effort. Some of the direct radiation measurements were taken before the silos were sealed in 1979, some were taken between 1979 and 1987, and others were taken in 1987 immediately after a Radon Treatment System (RTS) had been used to remove almost all the radon in the silo headspaces. SCA compared the pre-1979 direct radiation readings with the post-RTS readings and concluded that there was essentially no difference in these values. From this SCA concludes that essentially no radon was retained in the silo headspace prior to 1979. The Pb-210 concentrations in the K-65 residues were considerably lower than the Ra-226 concentrations based on the 1991 sampling, and SCA concludes that the only mechanism to explain this is the diffusion of radon from the residues into the headspace. Since almost no radon was retained in the headspace, essentially all of radon that diffused from the residues was released to the atmosphere.

One piece of data that influences SCA's calculation of radon releases is unknown: the relative concentrations of Ra-226 and Pb-210 at the time the residues were initially placed in the silos. SC&A's analysis assumes that these nuclides existed in the same relative abundance at the time of residue placement as they did at the time they were sampled in 1991. This assumption neither maximizes nor minimizes the resulting estimate of radon emissions.

RAC CALCULATION OF RADON RELEASE, 1979-1987

RAC-1995a provides different release rate estimates for three different periods: 1) 1953-1958, before the silos were sealed; 2) 1959-1987, after the silos were sealed and before a layer of protective foam was sprayed onto the silos; and 3) after 1987, when the foam had been added. RAC began by evaluating the years 1979-1987, the period when the silos were sealed, before operation of the Radon Treatment System. During this period there are radon concentration measurements and temperature measurements in the silo headspace. RAC estimated release rates for two release mechanisms during this period, which they refer to as ventilation and diffusion. "Ventilation" refers to silo head space air being released to the environment; "diffusion" means the movement of radon from the headspace through the concrete of the silo domes. In RAC's estimate the only mechanism for silo headspace air to be release to the environment during the 1979-1987 period is daily heating of the silos and subsequent expansion of the silo headspace air. Silo headspace air temperatures were not available for the entire year, so RAC used the available data to develop a correlation between outdoor air temperature daily change and silo headspace air temperature daily change. They then used daily outdoor air temperature readings, which were available for the entire year to estimate daily silo headspace air temperature changes. RAC then used the ideal gas law to calculate how much the silo headspace air volume would expand based on daily changes in silo air temperature, and assumed that the daily expansion of headspace air would be released to the environment. Multiplying the expansion volume by the measured radon concentration (2.62×10^7 pCi/L) yielded the amount of radon released to the environment by ventilation.

RAC estimated the diffusion release using classic radon diffusion techniques and some knowledge and some assumptions about the diffusion characteristics of the silo dome concrete.

RAC-1995a rarely uses average values in calculations, but rather assumes a distribution of values and then uses Monte Carlo techniques to determine the distribution of the result. Consequently it doesn't report an average value for radon release due to ventilation or diffusion, but rather a distribution of values, giving the 5^{th}, 25^{th}, 75^{th}, and 95^{th} percentiles of the distribution along with the median. For simplicity, this paper will refer to median values of the various distributions. The median values for RAC's estimate of release to the environment during the 1979-1987 period are 810 Ci/yr via ventilation, and 130 Ci/yr via diffusion. RAC's methods for arriving at this value are described in Appendix J of RAC-1995a, starting on page J-27.

RAC CALCULATION OF RADON RELEASE, 1959-1979

RAC's calculation of radon releases for 1959-1979 begins with the same direct radiation readings that SCA utilized in its calculation: the pre-sealing readings; the post-sealing before RTS operation readings; and the post-RTS operation readings. RAC referred to the post-RTS readings as a sort of background reading, meaning the direct radiation that was strictly due to radioactive materials in the K-65 residues, without any contribution from radon and progeny in the silo headspace. RAC treated the range of measurements recorded for each instance ("background," pre-sealing, post-sealing) as uniform distributions and did Monte Carlo runs to determine the distribution of differences. For this paper we'll still use median values. In the list below median values are added to the direct radiation reading ranges, as reported by RAC:

"background" : 35.5 to 76 mR/hr; median = 56 mR/hr

Pre-sealing : 65 to 90 mR/hr; median = 78 mR/hr

Post-sealing: 168 to 400 mR/hr ; median = 284 mR/hr

RAC's median estimate for radon concentration in the K-65 silo headspace prior to sealing (i.e., pre-1979) was 2.6 X 10^6 pCi/L, one order of magnitude lower than the headspace concentration that was measured after the silos had been sealed. Using this concentration, and an estimate of the rate at which radon was diffusing from the K-65 residues into the silo headspace, RAC estimated a median release rate to the environment of 6,200 Ci/year. Details of the calculation are found in Appendix J of RAC-1995a starting on page J-41.

DIFFUSION OF RADON FROM K-65 RESIDUES INTO SILO HEADSPACE

1979-1987

During the period when the silos were sealed there is a measured radon concentration in the headspace and a relatively convincing estimate of radon release rate to the environment. This means that the rate of radon diffusion from the residues into the headspace can be calculated by a straightforward steady-state calculation, where the "production rate" (rate at which radon entered the headspace) would be equal to the "removal rate" (rate at which radon decayed combined with rate at which it was released to the environment). RAC estimated that 16% of the radon that entered the headspace was released to the environment with the remainder undergoing radioactive decay in the headspace. Therefore the estimated annual release of 940 Ci/yr indicates that 940/0.16 = 5,880 Ci/yr diffused from the residues into the headspace. This represents 2.4% of the radon generated per year in the silos, using the values from SCA-2008.

1959-1979

The diffusion rate of radon from the K-65 residues into the silo headspace would be different for this period than for the sealed silo period. That is because the concentration of radon in the silo headspace constrains the diffusion of radon into the headspace. Since the headspace concentration was lower in the pre-sealed silos than it was in the sealed silos, the diffusion rate during the unsealed period would be larger. Details of RAC's calculation or radon production rate, or diffusion rate from the residues into the headspace air, are found in Appendix J of RAC-1995a, starting on page J-38. This calculation does use characteristics of radon diffusion, but since it's being calculated in comparison to the reasonably well known value for 1979-1987, many of those characteristic values cancel out in the calculation. RAC calculates an environmental release rate of 6,200 Ci/yr during this period, and estimates that 93% of the radon that diffused into the silo headspace was released to the environment. This indicates that 6200/0.93 = 6,670 Ci/yr diffused from the residues into the headspace. This represents 2.7% of the radon generated in the residues.

NAS REVIEW

In earlier exchanges about this issue, NIOSH commented that the National Research Council (NRC) review of RAC's work should suffice to indicate that the work is technically appropriate. In SCA-2010, SC&A responded that the NRC review (NRC-1994) that is available in the Site Research Database (SRDB) contains language that hardly endorses the RAC approach. The SRDB contains several different RAC reports, generated at different times in their project. It appears to contain only one NRC review report, which reviewed one of the RAC reports which is in the SRDB. The RAC reports in SRDB are:

> RAC-1993, Task 4,"Environmental Pathways – Models and Validation."
>
> RAC-1995a, Task 2/3, "Radionuclide Source Terms and Uncertainties."
>
> RAC-1995b, Task 5, "Review of Historic Data and Assessments for FMPC."
>
> RAC-1998, Task 6, "Radiation Doses and Risk to Residents from FMPC Operations from 1959-1988, Volume 1.

The NAS review in the SRDB, NRC-1994, is a review of Task 4. It did not review the detailed radon source term calculations contained in RAC-1995a, Task 2/3. There may have been NRC reviews of other RAC reports, but NIOSH has not obtained those. RAC-1998 does include a discussion of review by the National Research Council though, starting on page 108. The essence of this discussion is that RAC resolved any NRC comments in this 1998 version of their report.

RADON BEHAVIOR IN RESIDUE MATERIAL

RAC-1995a contains an extensive discussion of what they call "an alternative calculation of unconstrained radon releases" starting on page J-73. This approach calculates the rate at which radon would diffuse out of the K-65 residues if the silos were not covered. This was done in order to calculate a bounding diffusion rate to check the reasonableness of the report's main diffusion estimates. Table J-40 on page J-81 contains the various parameters that are required for this calculation and the range of values for each parameter that RAC used in their calculations. The definitions of these parameters are on pages J-73 andJ-74, and the bases for the values that are used for these parameters starts on page J-75. Some of the parameter values are selected because of knowledge of the K-65 residues; others were selected from literature values because measurements were not available for the K-65 residues. RAC uses a range of values for most parameters, so their resulting calculation provides a distribution of possible results. The report lists the 5^{th}, 25^{th}, 75^{th}, and 95^{th} percentiles, as well as the median, for the distribution of diffusion values. NIOSH's view is that we cannot discount the body of knowledge about radon behavior in estimating a radon release rate.

REFERENCES

RAC-1993, "The Fernald Dosimetry Reconstruction Project , Task 4, Environmental Pathways – Models and Validation," Killough, G.G., et. al., March 1993 (SRDB Ref ID 075602).

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RAC-1995b, "The Fernald Dosimetry Reconstruction Project, Task 5, Review of Historic Data and Assessments for the FMPC," Shleien, B., et. al., March 1995 (SRDB Ref ID 038043).

RAC-1998, "The Fernald Dosimetry Reconstruction Project, Task 6, Radiation Doses and Risk to Residents from FMPC Operations from 1951-1988, *Volume 1*,"Killough, G. G., et. al., September 1998 (SRDB Ref ID 014175).

SCA-2008, "An Alternative Assessment of Radon Releases from K-65 Silos, an SC&A White Paper," Behling, U. H., November 2008.

SCA-2010, "Response to NIOSH's Statements Regarding the RAC Model for Estimating Radon Releases from K-65 Silos," Memo from Hans Behling to the Fernald Work Group and others, January 26, 2010.