HOW NOT TO DISPOSE OF NORM/TENORM-BEARING WASTES: A CASE STUDY

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ABSTRACT
The Ashtabula River in northern Ohio contains a large amount of sediment containing quantities of NORM and TENORM from previous industrial activities at nearby mineral processing plants. Due to PCB contamination, these sediments were to be dredged and isolated in a landfill to be constructed by the responsible parties. Unfortunately, the State of Ohio has determined that these wastes may not be disposed of in this manner, and this determination has resulted in delaying the remediation project.

Computer models performed using the RESRAD computer code indicate that isolating these wastes in this manner will reduce dose to the nearby population because the NORM/TENORM will be safely buried beneath a compacted clay cover and isolated from all sources of exposure. In fact, radiation doses (including radon emanation) from these wastes in a properly maintained landfill are significantly lower than in the present condition, and the reduction is even more marked for NORM/TENORM in tailings piles. This suggests that, in many cases, disposal of NORM/TENORM wastes in on-site landfills may be a cost-effective and dose-conscious method of disposal, if regulatory issues can be resolved.

REGULATORY MATTERS
A company in northern Ohio[1] uses natural and synthetic titanium ores to produce titanium dioxide, which is then used for a variety of other purposes. Many sources of titanium ore also contain elevated levels of uranium, thorium, and associated progeny nuclides. Titanium is removed from the ore minerals during processing, resulting in a de facto increase in radionuclide concentrations. This is viewed as technological enhancement of the existing naturally occurring radioactive materials (NORM), and the resulting filtercake materials are viewed as TENORM by the Ohio Department of Health.

According to Section 3701-39-021 of the Ohio Administrative Code, Technologically Enhanced NORM (TENORM) "…means the chemical properties or physical state of natural sources of radiation have been altered or the potential exposure pathways of natural sources of radiation to humans have been altered to increase the human radiation exposure." Paragraph B of this section of the OAC states that Naturally Occurring or Accelerator Produced Radioactive Materials (NARM) are exempted from licensure if Ra-226 concentrations are "…27 picocuries per gram[2] (1 Bq g^{-1}) or less of technologically enhanced Radium 226 or radium 228 in soil…or in media other than soil…" NARM is defined to include TENORM.

The company would like to place filtercake waste into a sanitary landfill along with other materials resulting from removal actions elsewhere in Ashtabula. This landfill will be constructed to safely contain wastes containing PCBs and will include a 1.8 meter clay and soil cap. Calculations described in following sections indicate that this method of disposal will reduce radiation exposure pathways to the public and will not increase radiation exposure to the public compared to the raw ore materials or compared to filtercake that is left to accumulate on company grounds. Therefore, under the definition of TENORM provided in the OAC section quoted above, filtercake disposed of by burial in a properly-designed landfill cannot be considered TENORM and is exempt from regulation by the State of Ohio.

According to Section 3701-39-021 (B)(1)(a)(i and ii), radium concentrations of up to 27 pCi/gm (1 Bq/gm) are permitted in the top 15 centimeters of soil or matrices other than soil provided radon emanation rates are less than 20 pCi/m^2-sec (0.74 Bq/m^2-sec). According to Section 3701-39-021 (B)(1)(a)(iii and iv), radium concentrations of up to 5 pCi/gm (0.185 Bq/gm) are permitted in the top 15 centimeters of soil or matrices other than soil if radon emanation rates are greater than 20 pCi/m^2-sec (0.74 Bq/m^2-sec). Because the U-238 and Th-232 decay series appear to be present in approximate secular equilibrium, it is highly unlikely that any other decay series nuclides are present in concentrations exceeding 20 pCi/gm (0.74 Bq/gm) and it is certain that no decay series nuclides exceed concentrations of 135 pCi/gm (5 Bq/gm) in these materials. The company analyzed radon emanation rates on contact with filtercake materials and found them to be less than 20 pCi/m^2-sec (0.74 Bq/m^2-sec).

Based on these results, the company felt that the filtercake materials meet the exemption criteria noted above. The company also felt that the Ra-226 concentrations found in the filtercake materials are similar to or less than Ra-226 concentrations found in nature and in exempted TENORM materials. Accordingly, the company notified ODH that the filtercake materials are exempt from regulation by the State of Ohio as specified above.

Other portions of this section of the OAC exempt fertilizers, flyash, zirconium sands, and other similar materials from regulation as NARM. Some of these materials can have higher concentrations of U, Th, and progeny nuclides than is noted in the filtercake in question (Eisenbud and Gesell, 1998; EPA, 1990). In addition, language in this section of the OAC exempts “…materials in the recycling process contaminated with scale or residue not otherwise exempted or other equipment containing NARM with a radiation exposure level that does not exceed 25 microrads per hour (0.25 μGy) above background at any accessible point.”

The inclusion of materials such as flyash and fertilizer in this category make it evident that the exemption granted to materials meeting exemption criteria as listed in this section of the OAC applies to all uses and dispositions of exempt materials because of the obvious impossibility of attempting to regulate every source of fertilizer, flyash, and other exempted materials within the State of Ohio. Thus, the intent of the RAC, in approving this regulation, would appear to be that materials noted as receiving exemptions under this section of the OAC are intended to be exempt from both licensure and regulation by the State of Ohio.

In spite of this, the State of Ohio continued to insist that the filtercake materials were considered radioactive waste and must be handled appropriately. According to Ohio law, this meant that the materials could not be disposed of into a landfill, even a single-use landfill, in spite of the fact that Resrad modeling suggests that such a disposal option would result in a net *reduction* of dose to the public.

DESCRIPTION OF MATERIALS

Natural ore materials used in this process consist of minerals mined in South Africa, Australia, or other nations and transported to Ohio for processing. Synthetic ore consists of rutile (a titanium mineral) that is manufactured at another facility and transported to the company. Radiation levels from ore materials ranged from 9 to 45 μr/hr (0.09 to 0.45 μGy/hr). No radiation exposure above detection limits (10 mr or 0.1 mGy) was noted for any worker or area dosimetry in the vicinity of ore materials.

Synthetic Ore

The highest noted activity concentration of Th in synthetic ore was 376 ppm which corresponds to an activity concentration of about 41 pCi/gm (1.5 Bq/gm). The average Th concentration in synthetic ore was 33.8 pCi/gm (1.25 Bq/gm). Uranium concentrations were less than about 7 pCi/gm (0.25 Bq/gm). According to the OAC section cited above, NARM materials other than Ra-226 and Ra-228 present in concentrations less than 135 pCi/gm (5 Bq/gm) are exempted from regulation. The average concentrations of Ra-226 were less than 5 pCi/gm (0.185 Bq/gm). These levels are consistent with those noted in natural Ti ores such as ilmenite and rutile, which range from about 15 – 50 pCi/gm (0.56 – 1.85 Bq/gm).

Natural ore

Natural ore materials include ilmenite, rutile, and other ores of titanium. These are processed on-site, generating filtercake waste upon completion of processing. The process of removing titanium from these minerals results in concentrating the remaining elements in the filtercake, including U, Th, and other radioactive elements. The average concentrations of U and Th noted in natural ore materials were less than 5 pCi/gm (0.185 Bq/gm) for all isotopes noted. The average concentrations of Ra-226 were less than this value.

Filtercake

Following completion of processing, waste materials (filtercake) are generated and placed onto waste piles. The filtercake is fine-grained with properties similar to those of clays. The highest noted concentrations of Ra-226 in the filtercake were 7.6 pCi/gm (0.28 Bq/gm), associated with a U-238 activity of less than 5 pCi/gm (0.185 Bq/gm). Ra-226 concentrations were not noted for a number of samples, pending completion of the holding time. The highest noted U-238 concentration was less than 9 pCi/gm (0.33 Bq/gm). Assuming secular equilibrium through the U-238 decay series, this would correspond to a Ra-226 concentration of less than 9 pCi/gm (0.33 Bq/gm). Radiation levels from filtercake ranged from 15 to 25 μr/hr (0.15 – 0.25 μGy/hr). No radiation exposure above detection limits (10 mr, 0.1 mGy)) was noted for any worker or area dosimetry in the vicinity of filtercake materials.

RADIOLOGICAL MODELING

Three models were generated using version 5.82 of the RESRAD computer code developed by the Argonne National Laboratory. RESRAD is accepted by the Department of Energy and the Nuclear Regulatory Commission for determining radiation dose from residual radionuclides. RESRAD allows a variety of analyses of radiation dose under a number of site use scenarios. Three models were developed for this comparison; unprocessed ore, processed filtercake in aboveground waste piles, and processed filtercake placed into a landfill. Identical land-use scenarios and pathway analyses were used for all three scenarios to facilitate comparison. Site-specific data were used wherever possible. These models indicate the most significant exposures are those of direct exposure to gamma radiation and inhalation of suspended contaminated materials. Since parameters relevant to both of these exposure pathways have been measured directly by MIC, actual data were used to determine dose from these pathways rather than modeling data. Modeling data were used for exposure from soil ingestion and some radon pathways. The models used are described below. RESRAD default parameters were used for most site characteristics because they were found to have little importance to final calculated dose. Plots showing the results of sensitivity analysis on some parameters are appended to this report. Hard copy of the RESRAD summary printouts is appended to this report.

DESCRIPTION OF MODELS

Models were named F1 (landfilled filtercake), F2 (exposed filtercake), and F3 (exposed ore materials). All models share these input parameters:

Pathways:	Soil ingestion	
	Radon inhalation (water dependent and water independent)	
Cont. zone thickness:	30 meters	
Occupancy:	0.1 (about 17 hours per week for 52 weeks per year)	
Radionuclides:	U-238 plus progeny	30 pCi/gm (20 pCi/gm for ore)
		1.1 Bq/gm (0.74 Bq/gm for ore)
	Th-232 plus progeny	30 pCi/gm (20 pCi/gm for ore)
		1.1 Bq/gm (0.74 Bq/gm for ore)
	Deposited at time t=0 years	

The F1 model had the following cover parameters:

Cover density:	1.2 gm/cc
Cover thickness:	1.8 meters

Default parameters were used in all other cases. Using default parameters in all instances other than those directly affecting the most critical pathways facilitate intercomparison between these three models. Sensitivity analysis was performed for all default parameters to confirm the models accuracy was not dependent on site-specific information in those cases. In fact, sensitivity analysis indicates the use of default parameters in this manner did not change the end results significantly for any scenarios. Pathways for which site-specific monitoring data were available were not modeled. In addition, pathways which are not plausible under reasonable assumptions regarding future land use were not modeled. Specifically, no pathways that involved living on this land, placing potable water wells, farming the land, and similar scenarios were used for constructing these dose estimates.

Finally, it must be noted that many of these parameters were chosen to represent the "worst case". Specifically, a density of 1.2 gm/cc for the cover material is lower than is expected while using the default soil parameters for the disposed filtercake is similarly conservative. Also, the values chosen for radionuclide concentrations were well in excess of radionuclide concentrations noted except in some samples of synthetic ore. Since synthetic ore is blended with natural ore in varying proportions, use of the value of 20 pCi/gm (0.74 Bq/gm) is thought to be conservative and likely to produce a modeled dose higher than what would actually be the case. In all cases, the actual parameters in the field are expected to differ in such as way that actual radiation exposure will not exceed that modeled here.

RESULTS OF MODELING

In the case of ore and exposed filtercake, on-site measurements indicate the primary source of exposure to be inhalation of suspended contaminated particles. The secondary source of radiation exposure is direct exposure to gamma radiation. No other source of radiation exposure is expected to contribute more than 1 mrem annually to worker or public radiation dose.

It is noteworthy that radon provides an almost trivial dose in all scenarios. This reflects the fact that radon emanations are well-diluted by outside air and even personnel working nearly half their work week on top of exposed filtercake inhale very little radon. However, those parameters that serve to reduce dose from direct radiation exposure will also serve to reduce radon emanation rate because a thicker cap made of compacted soil or clay will retard radon diffusion through the cover material, causing it to decay before it emanates from the ground.

DISCUSSION OF MODELING RESULTS

These models clearly show that the lowest radiation dose to any persons at or near the site comes from filtercake properly disposed of into a landfill, covered with a properly designed cap. In fact, placing these materials into a landfill reduces radiation exposure from 48 mrem/yr (0.48 mSv/yr) (filtercake) or 33 mrem/yr (0.33 mSv/yr) (ore) to 25 μrem/yr (0.25 μSv/yr) (filtercake in landfill). This clearly demonstrates that filtercake disposed of into a landfill cannot be regulated as TENORM because it does not meet the definition presented in the OAC section cited above.

Calculated radiation doses

The radiation doses calculated under all scenarios are less than those permitted by law for non-occupational workers, although the doses calculated for ore materials and exposed filtercake exceed 25 mrem/yr (0.25 mSv/yr). However, it must be remembered that the radionuclide concentrations used are higher than those that exist in the bulk filtercake by a factor of 4. Since radiation dose from direct gamma exposure is directly proportional to radionuclide concentration, the actual dose from bulk filtercake under this scenario is about 12 mrem/yr (0.12 mSv/yr). The highest dose from landfilled filtercake is less than 1 mrem/yr (0.01 mSv/yr) as modeled.

Measured radiation doses

Using the models presented above, the highest dose rate (from exposed filtercake) is about 2.4 μSv/hr. The dose rate from ore materials is about 1.7 μSv/hr. By comparison, the highest direct dose rate measured anywhere in the facility was 0.45 μSv /hr (from ore materials), the highest dose from filtercake materials was 0.25 μSv /hr, and the highest dose noted in any TLD measurements was less than 0.10 mGy during the monitoring period. Under all scenarios it was assumed that workers spend about 10% of the year in the vicinity or ore or filtercake materials (about 16.8 hours weekly). Measured radiation dose rates were multiplied by 16.8 hours per week and 52 weeks per year to arrive at the final values for radiation exposure from this exposure pathway. Measured and calculate doses are summarized in the following table.

Table I. Measured Radiation Doses

Item	**Ore** (mSv/yr)	**Filtercake** (mSv/yr)	**Comments**
Direct gamma	0.39	0.22	based on actual radiation measurements
Inhalation	41.6	0.92	based on average of DAC_{eff} measurements
Radon	0.000025	0.000022	based on ICRP 60 submersion dose factors for measured radon levels
other	0.0065	0.0095	from ResRad modeling
total	**45.6**	**11.4**	
Radon + other	**0.006525**	**0.009522**	

Table II. Dose rates measured in other mineral processing industries are summarized below.

Industry (minerals processed)	**Nuclide and concentration (Bq/gm)**	**Annual radiation dose to workers (mSv)**
Ore	***Th-232 (1.25)*** ***U-238 (<0.5)***	***0.33 (876 hrs/yr) calculated*** ***0.24 (2000 hrs/yr) measured***
Tin smelting	Th-232 (0.3) U-238 (1)	0.25 (2000 hrs/year)
Niobium ore	Th-232 (80) U-238 (10)	4.0 (50 hrs/year)
Ilmenite (Ti ore)	Th-232 (1) U-238 (1)	Not stated
Zircon ores	Th-232 (0.8) U-238 (2.6)	0.66 – 10.3 (2000 hrs/year)

FOOTNOTES

[1] For reasons of confidentiality this company cannot be identified by name
[2] Radioactivity concentrations are quoted from the Ohio regulations, which are given in terms of US units. SI units are noted in parentheses based on the author's calculations. 1 Bq is equal to about 27 pCi.

CONCLUSIONS

1. Filtercake materials disposed of into a properly designed landfill do not meet the OAC definition of TENORM because exposure pathways have been altered to ***reduce*** radiation dose to workers and to the public.

2. Calculated dose rates, under the most conservative parameters, show that neither workers nor the public are expected to receive regulatorily significant radiation from landfilled filtercake materials.

3. The calculations performed for ore and filtercake materials are within reason when compared to measurements reported in the literature for ore processing industries.

4. The most critical parameters to be considered for the purposes of radiation dose reduction are the thickness and density of the material covering the filtercake.

5. The only significant exposure pathways under any scenario are those of inhalation and direct gamma radiation from U-238 and Th-232 series nuclides.

6. Based on the above information, filtercake materials disposed of into a capped landfill are not considered TENORM and are not regulated by the State of Ohio.

7. In the absence of actual measured radiation dose from external gamma radiation and dust inhalation, the calculated radiation exposure from other pathways is minimal and the calculated differences cannot be distinguished from each other by measurement or expected biological effects.

Table III. Comparison of filtercake with ODH requirements for exemption as TENORM
(Measurements given in US units to facilitate comparison with regulatory standards)

Parameter	ODH standard	Ore materials	Exposed filtercake	Landfilled filtercake
Human radiation exposure (*)	Cannot increase	40 (456) mrem/yr	23 (115) mrem/yr	< 1 mrem/yr
Ra-226/Ra-228	< 27 pCi/gm if Rn emanation < 20 pCi/m^2 – sec	7 (assuming equilibrium with U-238)	7.2/22	7.2/22
Ra-226.Ra-228	< 5 pCi/gm if Rn emanation > 20 pCi/m^2 – sec	7 (assuming equilibrium with U-238)	7.2/22	7.2/22
Radon emanation rate (+)	20 pCi/ m^2 – sec	1.35 pCi/l	1.23 pCi/l	N/A
Other NARM	135 pCi/gm	33.8 pCi/gm	22 (max noted)	22 (max noted)
Radiation levels	0.025 mrad/hr	0.01 mrad/hr	< 0.015 mrad/hr	<$3x10^{-5}$ mrad/hy

(*) Values for calculated human radiation exposure are for, respectively, cases in which respiratory protection is and is not worn by workers. In both cases, ore materials provide a greater dose than exposed filtercake, based on actual and calculated measurements.

(+) Radon emanation rates are not available for the specific materials in question. Instead, area radon measurements were used to calculate dose based on immersion coefficients provided by the International Council on Radiation Protection.

REFERENCES

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