Proceedings of the American Association of Radon Scientists and Technologists 2008 International Symposium Las Vegas NV, September 14-17, 2008. AARST © 2008

Disposal of Granular Activated Charcoal used for the Treatment of Radon-222 in Well Water.

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Abstract

Though not yet a major issue, the disposal of spent granular activated charcoal (GAC) filters used in the application of residential radon reduction from well water may one day become a concern. Residual radioactivity collected on the filter media, such as natural uranium, radium, and lead need to be accounted for in order to minimize problems at the disposal site. Well water is known to contain small quantities of uranium, radium, and radon, and the GAC filter has varying collection efficiencies for each. Lead, a decay product of radon will also build up on the filter. Various disposal concerns need to be considered such as, U.S. DOT transportation issues, U.S. NRC regulations, and state TENORM guidance. This paper addresses these issues, in particular as they relate to Pennsylvania.

Thesis

Can a granular activated charcoal (GAC) filter used for the treatment of radon-222 (radon) in residential well water be legally disposed of in a municipal waste landfill?

Introduction

The current use of GAC filters in the Commonwealth to treat elevated radon in public well water is very limited, with the number of systems unknown. Additional GAC filters are in use for the treatment of various chemical contamination problems associated with well water, such as for the treatment of trichloroethylene. However, it would be expected that with the final determination of the maximum contaminant level (MCL) by the U.S. Environmental Protection Agency (EPA) for radon-222, the use of the GAC filters would

increase both in the public water systems and in private wells. This increased use would obviously necessitate the increased demand for disposal of these filters, and would exacerbate issues associated with said disposal.

After reviewing the available literature, no definitive study could be found that actually looked at the disposal issue from cradle to grave. Numerous authors (Martin, 1990, Kiner, 1993 , Lowry, 1987a , Lowry, 1987b, Graves (ed.), 1987, Cothern, 1990 (ed), and Reid, 1985) mention the fact the disposal could be a problem, but none investigate the actual state and federal regulations in place to prohibit or allow GAC disposal. Marc J Parrotta (Parrotta, 1991) of the U.S. EPA Office of Drinking Water does provide a good EPA perspective on radioactivity in water treatment wastes; however, the emphasis of the paper is on public drinking water supplies.

The primary issue at hand is that the GAC bed will accumulate, over time, various radioactive materials, Rn-222 and its decay products, Natural Uranium, Natural Thorium and Ra-226/228. Hess et al (Hess, 2000) observed Pb-214, Bi-214, Pb-210, Ra-226, U-235, Th-234, Th-231, Tl-208, and Ac-228 on carbon filters in use for reducing radon in water. However, some of these filters were in use for up to 10 years, and in a very high radon in water area, Maine. Each of these radioactive species has different adsorptive properties toward the GAC bed, some with high affinity and some with very low affinity. Additionally, all of these radioactive substances are found in ground water in varying concentrations. One author (Martin, 1990) calculated that if the influent radon concentration was 10,000 pCi/L it would take in excess of 12 years for the GAC bed to be considered low level radioactive waste, with the presumption that it would the need to be disposed of in a low level radioactive waste site, of which there are only three in the country, Richland, WA., Clive, UT., and Barnwell, SC. This author's (Martin) definition of low-level radioactive waste used the 2000 pCi/g (Pb-210) limit which is now outdated.

A material that contains radionuclides that are present naturally in rocks, soils, water, and minerals and that have become concentrated as a result of human activities such as manufacturing, water treatment, or mining operations, is known as **T**echnologically **E**nhanced **N**aturally **O**ccurring **R**adioactive **M**aterial (TENORM). The uranium, radium, and radon that accumulate on a GAC bed are an example of TENORM (US EPA, 2000).

Regulation

It is not unreasonable to think that the regulations that cover hazardous and radioactive waste can be overwhelming (See Table 2). For instance the disposal of a GAC bed contaminated with radioactive isotopes could be covered under regulation by the U.S. EPA and the Resource Conservation and Recovery Act (RCRA), the Nuclear Regulatory Commission (NRC) and the Atomic Energy Act (AEA) of 1954, the U.S. Department of Transportation regulations for shipment of radioactive materials, and finally Commonwealth of Pennsylvania regulations on Municipal Waste Landfills (Comm. of PA., 1988).

U.S. EPA

The Resource Conservation and Recovery Act (40 CFR 239 to 282) establishes programs for regulating non-hazardous solid waste (Subtitle D), hazardous waste (Subtitle C), and underground storage tanks (Subtitle I). There are now two aspects to consider, is the GAC bed considered hazardous or non-hazardous waste, and is it considered a mixed waste? Mixed wastes are wastes that contain a hazardous waste component, regulated under RCRA, and a radioactive component, regulated under the NRC. According to 40 CFR 261.4(b)(1) The following solid wastes are not hazardous wastes: Household waste, including waste that has been collected, transported, stored, treated, disposed, recovered or reused. "Household waste" means any material (including garbage, trash, and sanitary wastes from septic tanks) derived from households. According to a letter of February 28, 1995 by Michael Petruska (US EPA, 1995), Chief Regulatory Development Branch, U.S. EPA, household waste must fulfill two criteria to be considered exempt from regulation under RCRA: first, household waste has to be generated by individuals on the premises of a household and, second, "the waste stream must be composed primarily of materials found in waste generated by consumers in their homes." The waste on the GAC filter is generated by the homeowners on the premises of the household, and the waste stream is via the well water used by the consumers in the home. From this we come to the conclusion that the GAC filter would not be considered hazardous waste and is exempt from regulation under RCRA. We now turn to the consideration of a mixed waste, from which we have already eliminated the hazardous aspect. Since the TENORM on the GAC filter is not regulated under the Atomic Energy Act of 1954, and since we have already concluded that it is not a hazardous waste, it is therefore not considered a mixed waste either.

U.S. NRC

We now deal with the radioactive part. The NRC is authorized to license and regulate the receipt, possession, use, and transfer of "byproduct material," "source material," and "special nuclear material". The NRC regulations are found in 10 CFR 1-199. Title 10 CFR Part 20 Subpart K deals with radioactive waste disposal, Title 10 CFR Part 61 deals specifically with land disposal of licensed radioactive material. GAC filters are not licensed material nor are TENORM on the filter licensed material. Title 10 CFR 40 deals with the disposal of byproduct material. Byproduct material is material made radioactive by exposure to the radiation incident to the process of producing special nuclear material and the tailings or wastes produced by the extraction or concentration of uranium or thorium from ore processed primarily for its source material content. GAC filters are not byproduct material. Title 10 CFR 61.1 deals with the land disposal of byproduct, source and special nuclear material. GAC filters are also not special nuclear material (Plutonium, U-233, and U-235). Source material is uranium or thorium or any combination of uranium or thorium in any physical or chemical form or ores that contain by weight, 0.05% or more of uranium or thorium or any combination of uranium and thorium (10 CFR 20.1003). By this definition the uranium or thorium on a GAC filter could be considered source material. However, it is considered an "unimportant quantity of source material" (10 CFR 40.13(a)) if the uranium or thorium, or any combination of

uranium or thorium is by weight less than 0.05% of the mixture. Therefore, the uranium or thorium on a GAC filter would be exempt from regulation if it can be determined that the uranium or thorium concentration on the filter is less than 0.05% by weight or about 335 pCi natural uranium per gram of material or 110 pCi natural thorium per gram of material. This final caveat is yet to be determined. It will be examined in the modeling part of this paper.

Three other radioactive isotopes to consider are Ra-226, Ra-228, and Pb-210. There should be very little radium collecting on the GAC filter material. Kinner et al (Kinner, 1993) showed that very little radium adsorbs to the GAC bed due to its extremely hydrophilic nature. Additionally, a cationic ion exchange unit prior to the GAC bed would remove greater than 99% of the radium in the influent stream before it even reached the GAC bed. Neither the Radium-226, Radium-228, nor the Lead-210 is regulated by the NRC since they are not source material, byproduct material, or special nuclear material. (And while these constraints are for licensed materials it is prudent to recognize that agencies such as the NRC, etc. may act on GAC filters as they discover the limits to which GAC filters can become radioactive.)

U.S. DOT

From a U.S. Department of Transportation point of view the main concern for GAC filters is the transportation of radioactive materials over the highways. Fortunately, there are exempt quantities of radioactive material for DOT purposes. The 2000 version of 49 CFR defined radioactive material as "any material having a specific activity greater than 70 Bq per gram (0.002 microcurie per gram)." This equates to 2000 pCi/g, which is the value found in the US EPA Carbdose (US EPA, 2001) program for disposal issues. However, the DOT regulations changed concerning the definition of radioactive material. The 2006 version of 49 CFR 173.436 now provides isotope specific values for exempt quantities of radioactive material, see Table 1 below. The exempt activity concentrations for natural uranium, uranium-238, radium-226, and lead-210 are 27 pCi/g, 270 pCi/g, 270 pCi/g, and 270 pCi/g respectively. The limits for radium-226, natural uranium, and lead-210 are for the parent and assumes their progeny are in secular equilibrium. If it could be determined that the GAC filters have quantities less than the above values then DOT regulations would not apply. This is difficult, short of very expensive laboratory analysis of the GAC material. One paper by (Kinner, 1993) presented data of uranium-238, radium-226, and lead-210 coring data from GAC filters taken out of service after about one year. The GAC filter had received about 100,740 gallons of water over the year-long study. The average radon-222 concentration of influent water was 35,000 pCi/L. One GAC filter was preceded by an ion exchange filter for pre-treatment. The GAC was analyzed using high-resolution gamma spectrometry. Only the maximum values are presented here for comparison with the DOT exempt quantities. The uranium-238 concentration was 178 pCi/g, radium-226 was 0.7 pCi/g, and lead-210 was 246 pCi/g. In this case the analyzed isotopes all are considered exempt from DOT transportation regulation. The unknown in this study was the influent uranium and radium concentrations in the well water. Additionally, had the GAC filter been left in place for

more than the one year period the exempt activity concentrations may have been exceeded.

We have another method of relief from DOT regulation of GAC filters. In a letter from Edward T. Mazzullo (US DOT, 2004), the Director of DOT Office of Hazardous Materials Standards to David J. Allard, Director, PA DEP, Bureau of Radiation Protection, October 8, 2004. Mr. Mazzullo writes "Household wastes, including household wastes contaminated with short-lived radionuclides, are not subject to Hazardous Materials Regulation." This DOT "Special Permit" would then allow GAC filters (household waste) contaminated with radionuclides, even above 49 CFR 173.436 exempt limits to be transported over the highways.

There are two caveats to observe for Table 1 below; one is that the columns with the SI units are the columns to use, and second, in order for a shipment to be exempt from DOT regulation it needs only to be below the activity concentration value or the consignment activity value. This was done so that NORM could be transported on the highways without regulation. As one could imagine, there would be many cases where a truck load of soil would have a consignment activity well above 10,000 Bq (270,000 pCi), but it would be extremely unlikely that the soils specific activity would be greater than 10 Bq/g (270 pCi/g). Typical soil has a radium activity of about 1 pCi/g, well below the activity concentration limit.

Table 1

173.436 U.S. DOT Exempt Material Activity Concentrations and Exempt Consignment Activity Limits for Radionuclides.

Radionuclide	Activity Concentration		Consignment Activity	
	Bq/g	pCi/g	Bq	pCi
Pb-210 (b)	10	270	10,000	270,000
Ra-226 (b)	10	270	10,000	270,000
Th_{nat} (b)	1.0	27	1,000	27,000
U_{nat} (b)	1.0	27	1,000	27,000
U-238	10	270	10,000	270,000

(b) Limits are on the parent nuclides but assumes their progeny are in secular equilibrium.

Several definitions from Table 1 above are given below:

Consignment: A package or group of packages or load of radioactive material offered by a person for transport in the same shipment.

Natural thorium means thorium with the naturally occurring distribution of thorium isotopes (essentially 100% by weight Th-232).

Natural uranium means chemically separated uranium containing the naturally occurring distribution of isotopes (approx. 99.28% U-238 and 0.72% U-235 by mass).

Radioactive material means any material containing radionuclides where both the activity concentration and the total activity in the consignment exceed the values specified in the table in US DOT 49 CFR 173.436 or values derived according to the instructions in US DOT 49 CFR 173.433.

We will discover that since we need to stay below either the activity concentration limits, or the consignment activity limits, and not both, it is easier to stay below the activity limits alone. We will therefore use activity concentration limits when considering U.S. DOT limits in our modeling section of the paper.

PA DEP

Naturally occurring radioactive materials (NORM) are not specifically regulated in Pennsylvania. TENORM is not regulated unless resulting radiation doses exceed the limits set forth in Title 25, Chapter 219 of Pennsylvania Code (Comm. of PA., 1997). Chapter 219 deals with Standards for Protection against Radiation. As found in 219.51 the dose limit for members of the general public is 100 mrem/yr, and the dose limit in an unrestricted area is 2 mrem/hr, if the individual were continuously present in the area (PA Code 219.51). NCRP Report No. 116 (NCRP, 1993) also recommends a general public dose limit of 100 mrem/yr for man-made sources other than medical and natural background, for continuous exposure. For the specific case of a spent GAC filter disposed of in a solid waste land fill containing TENORM, dose modeling must show that the general public would be exposed to no more than 25 mrem/yr, a fraction of the total dose limit.

The Commonwealth can and does regulate discrete sources of radium-226, where the activity is above that stipulated in Chapter 217. The Appendix B Chapter 217, Exempt Quantities has an exempt quantity for radium-226 at 0.1 microcuries (100,000 pCi), and the exempt quantity for lead-210 is also 0.1 microcuries. Chapter 217 deals with the Licensing of Radioactive Material. For the specific case of land disposal of TENORM

material that exceeds Chapter 217 exempt quantity limits, dose modeling must show that radiation dose is maintained as low as reasonably achievable (PA Code 219.182).

Municipal waste landfill disposal of TENORM is specified in Chapter 273 (Municipal Waste Landfills) of Pennsylvania Code (Comm. of PA., 1988). Paragraph 273.201(m) says that TENORM may not be disposed of in a municipal waste landfill, "unless approved in writing by the Department, and the disposal does not endanger the environment, facility staff or public health and safety."

Where the problem arises is at the disposal facility. Though the radioactive material may be exempt from both Federal and State regulation, it may still cause a solid waste facility radiation monitor to alarm. Portal monitors are set to alarm at about 10 microR/hour above background, where background in PA is about 10 microR/hour. This is a most undesirable event, if not previously planned for. The truck must be surveyed, the radioactive material identified and a half-life determined, a DEP regional health physicist may need to be on site, the contents may be accepted or rejected, and the facility records of the incident must be logged. If the contents are rejected by the disposal facility the hauler must obtain a DOT Exemption Form prior to going back on the road (e.g. back to the originator of the waste.)

However, if the incoming material that causes the alarm is determined to be NORM, then there are no disposal restrictions and the material can be accepted at the solid waste facility for final disposal. In the case where any available information would indicate that the suspect material is TENORM, as would be the case with a GAC filter, then the DEP Area Health Physicist may authorize immediate disposal. For small quantities the following conditions must be met: the volume of the waste must not exceed one cubic meter, the gamma radiation level at a distance of 5 cm from any source surface does not exceed 50 microR/hour, and the concentration of combined radium isotopes (226/228) does not exceed 5.0 pCi/g. Disposal of higher volumes, surface radiation values greater than 50 microR/hr, and higher radium concentrations may still be disposed of, however, with DEP approval, and environmental assessment and pathway analysis to demonstrate that the annual dose to any member of the general public is unlikely to exceed 25 mrem/yr (PA DEP , 2004). This analysis in fact has already been performed for the case of higher concentrations of radium. In our Pathway Analysis section we actually use 270 pCi/g of radium-226 as one of our inputs for the model. That model result showed a maximum dose of 4.62 mrem/yr, well below the 25 mrem/yr limit. This therefore shows that concentrations of radium-226 higher than 5 pCi/g can safely be disposed of. This also shows that we can exceed the Appendix B Chapter 217 exempt quantity of radium-226 of 100,000 pCi, since our GAC charcoal mass is 25,143 grams and we accumulate 0.01 pCi/g/day. After 399 days we have 100,149 pCi on the GAC filter assuming 100% removal efficiency. It is our desire to be able to keep the GAC filters in place for about two years. After two years the GAC filter would have about 100,149 pCi times two years or 200,298 pCi on GAC bed, well above the 100,000 exempt limit, but still acceptable based on pathway analysis.

Table 2, Federal and State Regulatory Limits

Agency	Uranium Nat	Thorium Nat	Radium	Lead
EPA	No Reg.	No Reg.	No Reg.	No Reg.
NRC	~335 pCi/g	~110 pCi/g	No Reg.	No Reg.
DOT	See table 1	See table 1	See table 1	See table 1
DEP			<5 pCi/g	
			(Ra-226/228)	
DEP	GAC Volume < 1 m^3			
DEP	<50 µR/hr at 5 cm from surface			
DEP	Annual does to Gen. Public < 25mrem/yr			

These criteria must be met for disposal and/or transportation.

An important note about Table 2 above. As published in the Federal Register on July 2, 1979, "On December 3, 1979, the NRC amended its regulations in 10 CFR 71 to require that all shipments of radioactive material by NRC licensees be made in accordance with DOT requirements." This would imply that for our purposes of this paper we should ignore the NRC limits and use DOT limits as found in Table 1which are more restrictive.

The GAC Filter

From a disposal point of view we need to know what would collect on the filter media over its time of use in the home. We certainly know its intended function is to reduce well water radon concentrations, thus it will collect radon-222 and its decay progeny up to and including lead-210. We will know the influent radon concentration and with several other assumptions can use Carbdose version 5.0 to calculate the build-up of lead-210 over time. We should try to assure that the lead-210 on the GAC bed does not exceed 0.1 microcuries, (Which turns out to be approximately 4 pCi/g in our GAC filter) the DEP Chapter 217 Appendix B Exempt Quantity limit for Pb-210. We can also calculate the gamma dose rate from the GAC tank. For dose rates we would like to be below 50 µR/hr, and preferably below twice background levels (~20 µR/hr), so as not to alarm portal monitors.

The other more difficult disposal issue to consider is the Ra-226/228 and uranium concentrations on the GAC bed. More difficult because, short of running a gamma spectrum on the GAC we can not predict how much radium or uranium will be on the

GAC. It is also very unlikely that any homeowner would have determined the radium or uranium concentrations in their private well water. And finally, we do know that both radium and uranium occur in all ground water. The one good thing about this fact is that their concentrations in ground water are generally very low. Data supplied by the PA DEP, Bureau of Water Standards and Facility Regulation (PA DEP, 2008) showed a three year (2005, 2006, 2007) average for natural uranium, radium-226 and radium-228 of 1.57, 0.15, and 0.27 pCi/L, respectively. These values represent samples at entry points (finished water) for public water systems within the Commonwealth. Private well water may show higher values. Through a literature search and several personal communications we have been able to determine some approximate efficiencies for the removal of uranium and radium from water by a GAC filter. This will be important to know when we try to calculate how much uranium and radium will accumulate on the GAC bed based on an assumed influent concentration of the two radioisotopes. Karadeniz et. al. (Karadeniz, 2000) found a uranium recovery efficiency for charcoal of 97%. However, this was under specific water quality conditions of 150 ppm uranium, 50 degrees Fahrenheit, and pH of 3. Phillip Egidi (Egidi, 2008) of the Colorado Department of Public Health claims that GAC is about 35% effective for removal of radium, and very effective (>90%) for uranium removal. Finally, Zakutevskii et. al. (Zakutevskii, 2007) conclude that carbon materials as sorbents have fairly high rate of uranium adsorption from a solution and sorption capacity of 1.5-2 mg-equiv g^{-1} (161 – 214 pCi U/g carbon, or 4,714,312 pCi/GAC unit.)

There are also various published values for radon-222 occurrence in the commonwealth, Jerry Rupert (Rupert, 1993) of the PA DEP, Bureau of Drinking Water Management, reported a population-weighted mean of 1,299 pCi/l with a median of 588 pCi/L, for community ground water system entry points. A U.S. EPA study found a mean of 756 pCi/l for finished water from public groundwater, and Dixon and Lee (Dixon and Lee, 1988) found a mean of 1,570 pCi/L.

MODELING

From Table 2 above, there are four areas of concern: Uranium, Thorium, Radium and Lead, all which may accumulate on the GAC bed.

Uranium

We will consider each in turn beginning with Uranium. There are two regulations of concern, one established by the DOT (27pCi/g.) and the other by the NRC (335pCi/g.). The quantities in parenthesis are the levels below which each of these agencies considers the material exempt from their regulations and rules.

As established by the U.S. EPA the best available technology(s) (BAT) for the removal of uranium from public drinking water is via anion exchange, lime softening, enhanced coagulation/filtration, or reverse osmosis, and not GAC. Nevertheless it was decided that it's likely some of the Uranium will be filtered out by the GAC unit, though the efficiency of removal is somewhat convoluted. In particular various references have tended to support a uranium removal efficiency by GAC in the 90 to 99% range, though this may be for a limited time after which breakthrough (the condition at which the uranium concentration on the GAC begins desorbing from the charcoal, causing the outflow water to have a higher uranium concentration than the inflow water) occurs. This time is reported to be between 3 and 6 months (Sorg, 1988, and Lowry, 1988).

The worst possible case is if the GAC unit was 100% efficient in removing the Uranium which would lead to the highest Uranium concentration in the charcoal. In the first few months of use we do find the efficiency of uranium removal by GAC to be almost 100%, and it is somewhat enlightening to investigate that case over a lengthy period.

During the balance of this work we make the following assumptions: The volume of the charcoal is 2.0 ft^3, the charcoal density is 0.44 gm/cm^3, a typical household uses 300 gallons (1134 liters) of water/day.

This leads us to recognize that there is 25,143 grams of charcoal in the tank. We will also use a natural uranium concentration in the water as 1.72pCi/l (PA DEP, 2008) (ground water sources only). This is a value in agreement with work done by Cothern and Lappenbusch (Cothern and Lappenbusch, 1983).

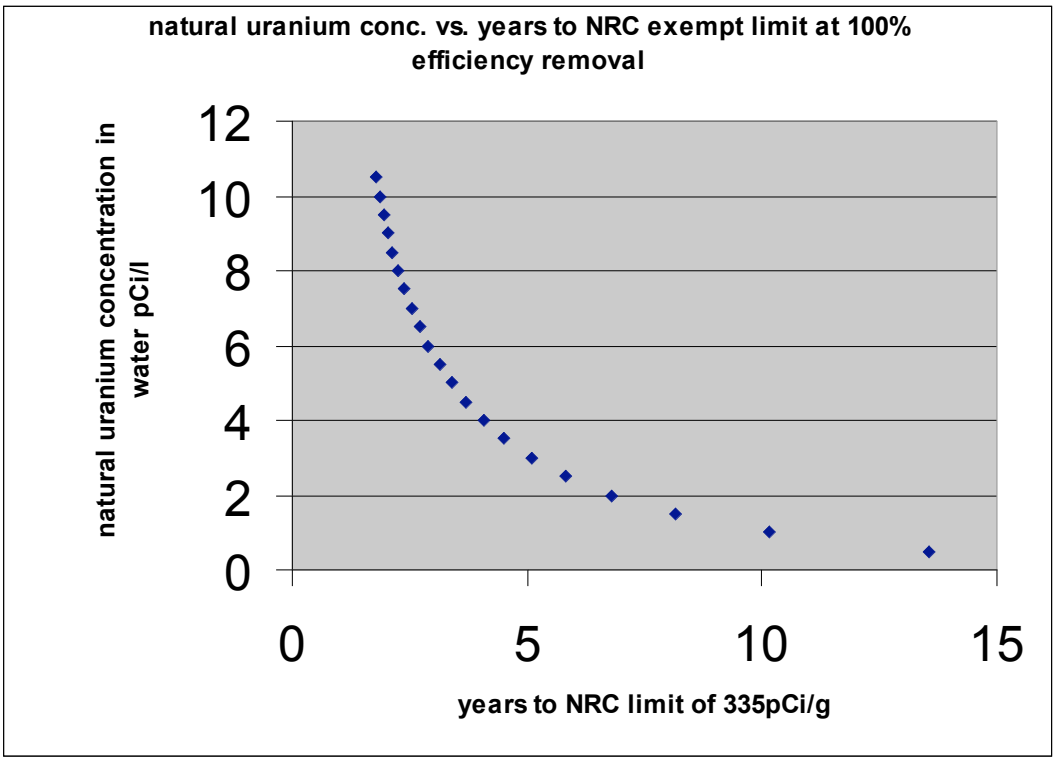
From these values we find 1950 pCi are added to the GAC filter each day (1.72pCi/l x 1134 l/day) or 0.078pCi/g/day (1950pCi/day / 25143g) which would result in the GAC filter reaching the DOT exemption limit of 27 pCi/g in 346 days or just about one year.

If we consider only the NRC limit of 335 pCi/g, this would allow 4295 days, or 11.8 years before the GAC unit arrives at the NRC exemption limit and this seems to be a respectable time period.

Naturally as the Uranium concentration in the water increases, the time to arrive at exempt values will decrease and this prompts us to consider under these conditions (100% removal efficiency) what the maximum natural uranium concentration would be that would lower the time to two years to arrive at the NRC exemption value of 335 pCi/g. One can easily show that the value is 10.2pCi/l, which is well above median uranium in ground water values. It is interesting here to note that according to the PA DEP the range of natural uranium concentrations found in ground water in PA is as high as 57 pCi/l. and as low as equipment detection limits (~0.03 pCi/L).

So it appears that under the worst possible conditions (Without breakthrough), a natural uranium concentration of 10.2 pCi/l will require removal of the charcoal every two years, or probably slightly less. At least for the case of a continuous uranium removal efficiency of 100%. In fact under these conditions it's simple to produce a graph of the time needed to arrive at the NRC exempt limit versus the natural uranium concentration in the water, leading to Figure 1.

Figure 1



Two studies, one performed by (Lowry, 1988), the other by (Sorg, 1988) have shown that Uranium collects in the GAC filter at almost 100% removal efficiency but only for a period of 3 to 6 months after which breakthrough occurs.

Another analysis using alpha spectrometry of a uranium in well water sample performed by the PA DEP Bureau of Laboratories in 2008 showed an influent natural uranium concentration of 3.635 pCi/l and an outflow natural uranium concentration of .017 pCi/l, leading us to a 95% reduction for a GAC unit that was in use for approximately only one month.

This tempers the analysis of Figure 1, since those results are based on 100% removal efficiency continuously over the time periods mentioned. This underlines the result that the analysis above is clearly leading to maxima which are very conservative.

We may also consider the case where the uranium removal efficiency is 100% but for a period of only six months at which point the uranium concentration on the GAC bed will be a maximum because of breakthrough. For these conditions we can then calculate the uranium in water concentration in the influent water that will produce a uranium concentration on the GAC bed of 335 pCi/g. The result is 41pCi/l and this concentration would be an anomalously high value. This suggests that in almost all cases the uranium concentration will not be a disposal issue.

If the GAC filter is to be transported, then the 27pCi/g limit is the constraint. In this case, using a 100% uranium removal efficiency for six months we easily show from:

$$27\frac{pCi}{g} = conc.ofU(pCi/l)\times 1134\frac{l}{d}\times 180d/25143g$$

Which leads us to the maximum allowed uranium in water concentration to be 3.3pCi/l

Lead

We'll now consider the Pb^{210} problem. This comes in two parts. First is the issue of how much radioactivity (pCi/g) will collect in the GAC tank, and second is the dose that radioactivity provides to its surrounding.

We deal with the first part of this problem here. As Radon decays the only daughters which produce a problem are those with half lives at least a few days or longer which lead us to the Pb^{210}, Bi^{210} and Po^{210} with corresponding half lives of 22.3y, 5.01d, and 138.4d. EPA has created a computer program titled "CARBDOSE" which is ideally suited to the calculation needed to answer the question, "How many years must pass for a given radon in water concentration to deposit 270 pCi/g of radioactivity from Pb^{210} in the GAC tank?" The results are the same if we include the daughter concentrations of Bi^{210} and Po^{210} since they are presumed to be in secular equilibrium with the Pb^{210}.

Again the assumptions of 300 gallons/day, 100% efficiency for the removal of radon by the GAC, 2 ft^3 of charcoal and a density of 0.45g/cc. (This last value for the density of the carbon is approximately 2% higher than that used earlier. It is the value used by the CARBDOSE program. This last value is slightly different than what was used in the above calculations but the difference is negligible.)

Running the program for different radon concentrations in the water yields Figure 2 below.

Figure 2

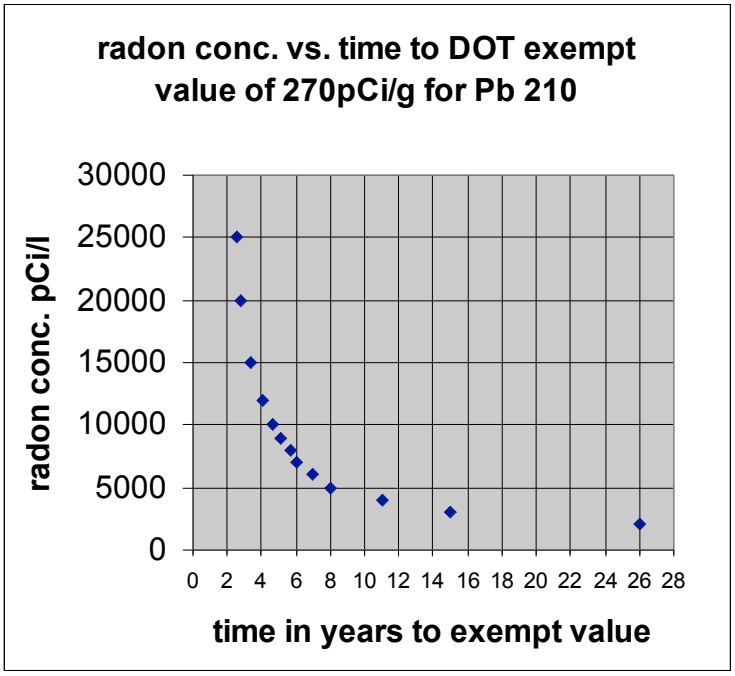


Figure 2: A graph of the time it takes to accumulate 270 pCi/g of Pb^{210} on the GAC filter as a function of the influent radon in water concentration.

We'll note that a GAC unit may have a radon concentration flowing into it as high as 25000pCi/l and it will arrive at the DOT exempt value of 270 pCi/g in 2.5 years. This should give guidance for how frequently the GAC needs to be changed out to avoid having radioactivity above the DOT exempt value of 270 pCi/g for Pb^{210}

Radium

We now consider the issue of radium. There are two radium isotopes of concern for our purposes, radium-226 with a half-life of 1600 years and radium-228 with a half-life of 5.84 years. The U.S. EPA MCL for radium is 5 pCi/L for the combined two isotopes. The EPA MCL applies to public water systems and not private wells; however, we will consider the question of how long radium in water takes to accumulate on the GAC cylinder in a private residence, to a concentration of 5 pCi/g. The 5 pCi/g value is a

disposal limit as defined by the PA DEP beyond which disposal will be problematic. So we consider an influent concentration of Ra 226 and 228 at the EPA MCL of 5 pCi/l, and we ignore the fact that some Ra 228 will decay during the calculated time period. This will be justified by our results and in any case will provide us with a conservative result.

The initial approach used is to assume a 100% removal rate to identify the time to 5pCi/g on the charcoal bed assuming a homogeneous distribution of the isotope throughout the bed.

With these assumptions we find it would take only 22 days for a GAC unit to reach the 5pCi/g limit. Turning this around we can ask what efficiency of Radium removal is needed by the GAC unit to take 2 years to reach the 5pCi/g limit when the influent concentration is 5 pCi/l, and find this to be 3%.

Next we consider data provided by the PA DEP, Bureau of Water Standards and Facility Regulation. They collected data that showed an average radium-226 concentration of 0.15 pCi/L and an average radium-228 concentration of 0.27 pCi/L in public drinking water supplies, for a sum of radium-226/228 of 0.42 pCi/l. Other information (Egidi, 2008) suggests that GAC may have a removal efficiency of about 35% or less for radium.

Again assuming none of the radium decays, we find (0.42 pCi/l x 1134 liters/day x .35 / 25143 g.) equals 0.007 pCi/g/day deposited onto the GAC bed. This leads us to a time of 2.1 years to reach the 5 pCi/g. limit.

Thorium

Next, we look at thorium. Ninety nine percent of thorium is Th-232 which has a half life of 14 billion years. Thorium is not regulated by EPA in public drinking water. However, since EPA has an MCL of 15 pCi/l for gross alpha activity, the maximum thorium activity would be 15pCi/l. (This assumes the water is treated to this limit of course.) We can again assume a 100% removal efficiency. We have found no references to describe the removal efficiency of thorium in ground water by the GAC column.

Using 15pCi/l, we find the number of days to the U.S. DOT limit of 27 pCi/g (The smaller of the two exemption limits, DOT and NRC) is 40 days at 100% efficiency.

Lowry (Lowry, 1988) points out that "Thorium levels…containing more than 0.1 pCi/l would be unusual, and one with more than 1.0 pCi/l would be extremely rare in ground water.

Another group (Jia, 2008) have measured the concentration of three thorium isotopes (232, 230, and 228) in drinking water in Rome, Italy and found their concentrations to range from 1.9×10^{-5} pCi/l to 3.6×10^{-2} pCi/l.

Using the largest of these values, 1.0 pCi/l as the thorium concentration in the water, we find that the time to reach the exempt limit of 27 pCi/g becomes 1.6 years. And this would be an extremely rare case. Of course if the value of 0.1 pCi/l is used this time to the exempt limit rises to 16 years, and this is the more likely case.

Thorium is not considered a disposal issue due to its extremely low concentration in ground water.

GAC Dose Rate

We now consider the issue of the gamma exposure rate and its limits as given by the PA DEP, Bureaus of Radiation Protection and Land Recyling and Waste Management, which requires that the exposure rate be below 50 uR/hr at 5 cm (see Table 2) from the surface of the GAC tank for the purposes of disposal. Only gamma emissions penetrate the tank so we don't need to consider the issues of betas and alphas.

It is interesting to note that for waste disposal, twice the background level of 10uR/hr, or 20 uR/hr is used to set off the alarms at the waste disposal site. In those cases, then the 50 uR/hr at 2 inches from the surface of the tank is applied.

The computer program CARBDOSE is the tool of choice to perform this calculation. We make the very good approximation that the daughters, namely Bi^{214} and Pb^{214} are in secular equilibrium with the radon as the radon decays

CARBDOSE allows us to calculate the gamma exposure rate verse the influent radon concentration, see Table 3.

Table 3

Influent Rn Conc. Vs. Gamma Exposure rate		
Inflow Rn conc. (pCi/l)	**Exposure 2in. from tank wall uR/hr**	**Time for exposure to decrease to 50 uR/hr. (days)**
1000	244	8.8
5000	1220	17.7
10000	2440	21.4
15000	3660	23.7
20000	4880	25.3
25000	6090	26.5

The last column assumes the daughters decay at the same rate as the radon after the tank is removed from the inflow. Table 3 also assumes that after 30 days of operation the

GAC has attained almost 100% equilibrium with radon and its daughters, and 30 days is used in the above calculations.

From this table we conclude that for those cases where GAC is a viable treatment for radon in water, that is, radon concentrations in water of less than 25,000pCi/l at least as far as exposure concentrations are concerned, setting the tank aside for one month will prevent the exposure from the tank as measured two inches from the tank wall from being greater than 50uR/hr. This Carbdose calculation looks only at the radon daughters. Other isotopes such as Uranium, etc. which may be in the influent water and collect on the GAC filter will also contribute to the gamma exposure rate. The authors' recommend that all installers of GAC filters purchase and use an appropriate instrument for measuring the gamma exposure rate from the GAC tank and if the exposure rate is greater than 50uR/hr, store the tank at a safe location until such time as the exposure rate is significantly below 50 μR/hr and preferably below 20 μR/hr.

Pathway Analysis

A solid waste facility must have in place a plan to describe the potential exposure pathway for dealing with radioactive material in the waste stream. One example of this radioactive material for our purposes would be the used GAC filter. One way to model these exposure pathways would be to use basic and conservative regulatory computer codes such as the EPA's CAP88 or the DOE/NRC's RESRAD codes. To meet DEP disposal requirements the modeling must show that the annual dose to the general public does not exceed 25 mrem/yr, and for this demonstration we use RESRAD, RESidual RADioactivity (RESRAD, 2001).

The input parameters for the model used the maximum values as found in our Table 2 for uranium, thorium, radium, and lead, 335, 110, 270, and 270 pCi/g respectively. Due to the complex nature of the parent-daughter relationship in ground water systems we did not run the dose calculation with all daughters in full equilibrium. If a precise gamma analysis of the GAC material could be run this pathway analysis could be refined with those specific isotopes. It was assumed that 7.4 square meters (80 square feet) of ground was contaminated to a depth of 0.01 meters (0.4 inches). This coverage would be approximately what the 2 cubic feet of GAC material would disperse to. The model then assumes that a farmer lives on top of the contaminated site, raises crops and livestock, and drinks the ground water. There are nine environmental exposure pathways considered in the model. The model calculates doses out to 1000 years. The model also provides for a soil cover over the contaminated area of 0.6 m (24 inches). A maximum dose of 4.62 mrem/yr occurs at 599 years, and this is primarily due to ground shine. Ground shine is radioactivity deposited on the ground that provides a pathway for external exposure. This computer run show's that this particular GAC filter would not be subject to the 25 mrem/yr limit, and from this aspect would be safe for disposal.

Conclusions

It is very possible that the issue of TENORM, i.e. the radionuclides on a GAC filter, will become more of an issue in the near future.

We found the EPA does not regulate TENORM, nor does the NRC unless it becomes Source Material. We found that of the three U.S. Government agencies, the U.S. DOT regulations would be the most likely to have an impact on the disposal of spent GAC filters, via transportation of the GAC over the highways. However, with the clarification from the Edward T. Mazzullo letter to the PA Department of Environmental Protection, we see that household waste, which GAC is considered to be, is exempt from the US DOT Hazardous Materials Regulations.

We have come to the conclusion that the issue of disposal of TENORM wastes will most likely be handled at the state level, this is certainly the case as it applies to the Commonwealth of PA.

In the Commonwealth of PA, waste containing NORM may be disposed of in solid waste disposal facilities with no restrictions. If available information indicates that incoming waste contains TENORM, then various PA DEP criteria must be met before the waste can be accepted for disposal.

We researched and found various concentration values for uranium, thorium, radium, and lead in private and public water supplies, and found out how efficient the GAC material is at removing said radioisotopes.

We have concluded that any thorium on the GAC bed will not a disposal issue and that uranium, radium, and lead on the GAC can be safely dealt with by disposal approximately every two years.

We have not examined the issue of the decreasing efficiency of the GAC bed for radon removal over time. As this issue is clarified the two year disposal recommendation may need adjustment.

It is our conclusion that the spent GAC filter will be able to be picked up by curb side waste removal services, transported over the highways and safely disposed of in the local solid waste disposal facility, without encumbering any regulatory oversight, assuming the recommendations of this paper are followed.

All mitigators should use appropriate meters for monitoring exposures near the GAC tank and record such information prior to carbon disposal.

Influent concentrations of radon as high as 25,000 pCi/l would require the tank be set aside for approximately 30 days.

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