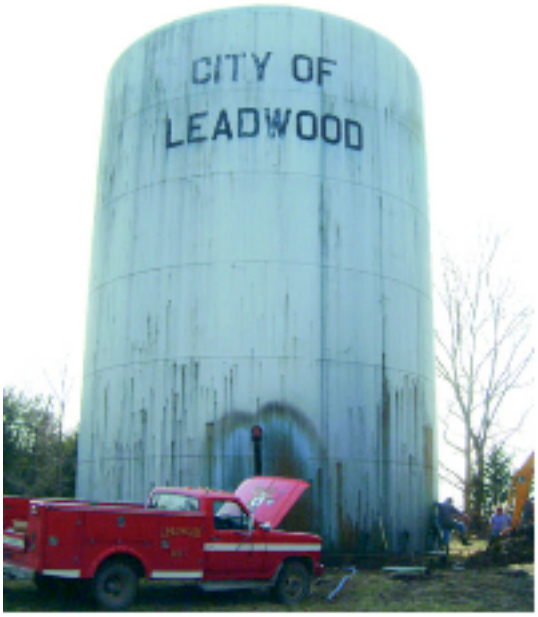
Leadwood Radionuclides Removal Technology Assessment

submitted

September, 2003



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Background

From the naturally-occurring radioactive decay products of uranium and thorium in the rocks and soils underlying Leadwood, Missouri, gross alpha emitters and radium ions are found in the groundwater at levels exceeding the USEPA Maximum Contaminant Levels (MCLs). In June of 1997, Leadwood completed construction on a water treatment facility designed to remove these radionuclides from their raw water source. In December of 2000, the final rule on radionuclides was published in the Federal register, which included MCLs of 5 pCi/l for combined Radium 226 + 228 and 15 pCi/l for gross alpha. These limits have been set in an effort to limit the long-term potential for the occurrence of bone and other cancers in humans.

The scope of this project is to analytically characterize the raw drinking water (including radionuclide content), ensure that the current treatment train meets manufacturer's specifications, analytically evaluate the effectiveness and efficiency of the current treatment train in removal of radionuclides, investigate possible enhancements of the treatment train to improve effectiveness and efficiency of removal, evaluate options for adequate disposal of waste residuals from the treatment process, and provide a final report specifying the effectiveness and efficiency of this technology in removal of radionuclides.

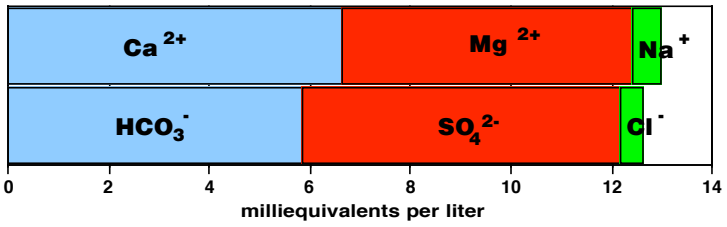
Characteristics of Source Water

In its history, Leadwood has utilized two deep wells as their water supply source. Well #1 is now only used as a supplemental or 'back-up' source. Water from this well is not filtered. Well #2 water is treated in the filtration facility constructed in 1997. This facility was primarily designed to reduce the concentration of radionuclides in the Well #2 water. However, the addition of chlorine also provides for disinfection of the water source and the propagation of a residual in the distribution system

Both well waters are very hard (620 mg/l as calcium carbonate equivalent or 12 meq/l hardness) and contain a significant amount of sulfate ion. The bar diagram, shown below, illustrates the general inorganic composition of the groundwater (based on MDNR finished water data; September, 1997). The inorganic composition is typical of hard (calcium plus magnesium ions), alkaline Missouri water derived from limestone formations. It is quite low in sodium and chloride, but high in sulfate ion. During treatment, pH has been observed to increase slightly, from 7.25 to 7.34.

The well waters contain little iron (<0.2 mg Fe/l) and virtually no manganese. It is because there is no iron or manganese naturally present in this water that ferric ion must be added as a supplement to form a ferric hydroxide precipitate.

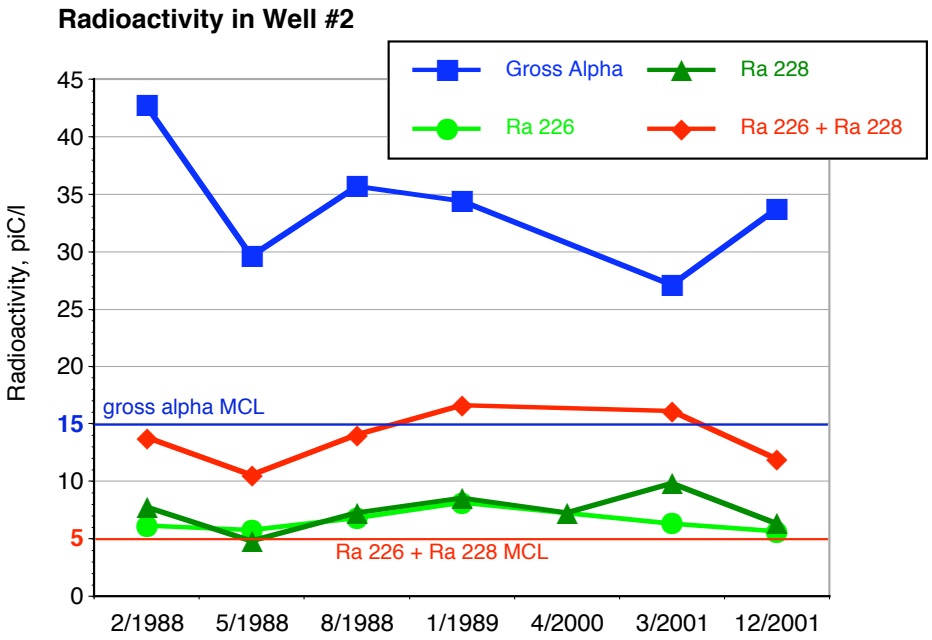
Aside from hardness and mineral content, Leadwood's well water does not appear to exhibit odors or an objectionable taste. Observation of a freshly drawn sample from Well #2, however, showed the presence of dense, black particulate matter similar in appearance to iron sulfides. These particles would be readily removed on filtration.



Radionuclides

The following plot of all available data on *gross alpha emitters* and *radium^{226} plus radium^{228}* show that Well #2 contains 33 picoCuries per liter (pCi/l) of alpha radiation. This is over twice the allowable maximum contaminant level (MCL) for gross alpha. The well water averages 14 pCi/l of *radium^{226} plus radium^{228}*.

From this limited data, it appears that the radionuclide of the well water is at steady-state and does not vary greatly. This would be consistent with the chemistry of many well waters that are at saturation equilibrium with the subsurface rocks and minerals.



The MCL for *radium^{226} plus radium^{228}* (combined) is 5 pCi/l. Water from Well #2 contains almost three times this level. Accordingly, to achieve compliance with state and federal regulations, treatment at Leadwood must consistently reduce *gross alpha* by, at least, 55% and *radium^{226} plus radium^{228}* by at least 64%.

The pilot plant studies that were conducted prior to the design and installation of the treatment facilities at Leadwood indicated that the treatment process being installed was capable of achieving up to 90% removal of both gross alpha and radium.

Existing Treatment Process

The process being used at Leadwood is a Filtronics system consisting of two contactors in series and two filters in parallel. This process requires the addition of a solution of ferric chloride and chlorine gas to the well water prior to the first contactor, and a feed of sulfur dioxide gas prior to the second contactor. In the presence of chlorine, *ferric ion* from the added ferric chloride salt solution is precipitated as ferric hydroxide. It is theorized that the in-situ formation of the orange-brown ferric hydroxide precipitate forms an active surface which has an adsorptive capacity for radium and other alpha emitters. The ferric hydroxide precipitate along with the adsorbed radionuclides can then be removed by filtration on proprietary Electromedia I media.

The treatment process currently in use has not been investigated by USEPA and is not currently listed as a "best available technology (BAT)" option for radionuclide removal. However, pilot studies indicated that this process has the potential for allowing Leadwood to meet the stringent standards set by USEPA on alpha-emitting radionuclides. Based on the results of the pilot studies, the treatment system was installed in 1997.

Filtronics' system provides for ferric chloride, chlorine, and sulfur dioxide feeds

Results of Pilot Studies of Radionuclides Removal

Based on the results of several pilot studies conducted by Metropolitan Engineering and Surveying (1995), the plant process design recommended the following chemical additions prior to filtration:

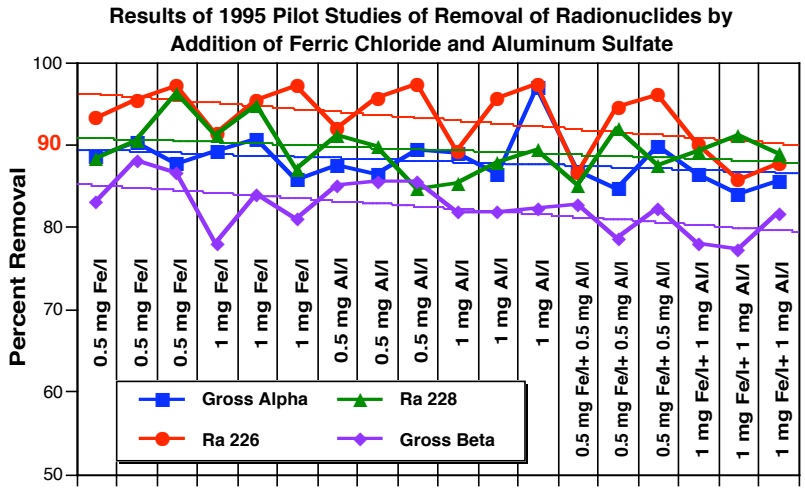
- *chlorine* (0.2 pounds per day, for breakpoint chlorination),
- *sulfur dioxide* (0.1 pounds per day, to reduce excess chlorine), and
- *ferric chloride* (0.5 mg Fe/l, for adsorption of radionuclides),

Pilot study results using the 0.5 mg Fe/l dosage showed an approximately 90% reduction in gross alpha, 95% removal of radium^{226} and 91% removal of radium^{228}. In addition, gross beta reductions of

approximately 85% were observed incidental to the removal of the alpha emitters. While somewhat improved reductions might have been expected, increasing the ferric ion dosage to 1 mg Fe/l resulted in similar reductions in each of these parameters.

Replicate studies were also conducted using aluminum sulfate (alum) as an alternative to the ferric chloride solution. Both 0.5 and 1.0 mg Al/l dosages yielded results comparable to those attained using the ferric chloride feed.

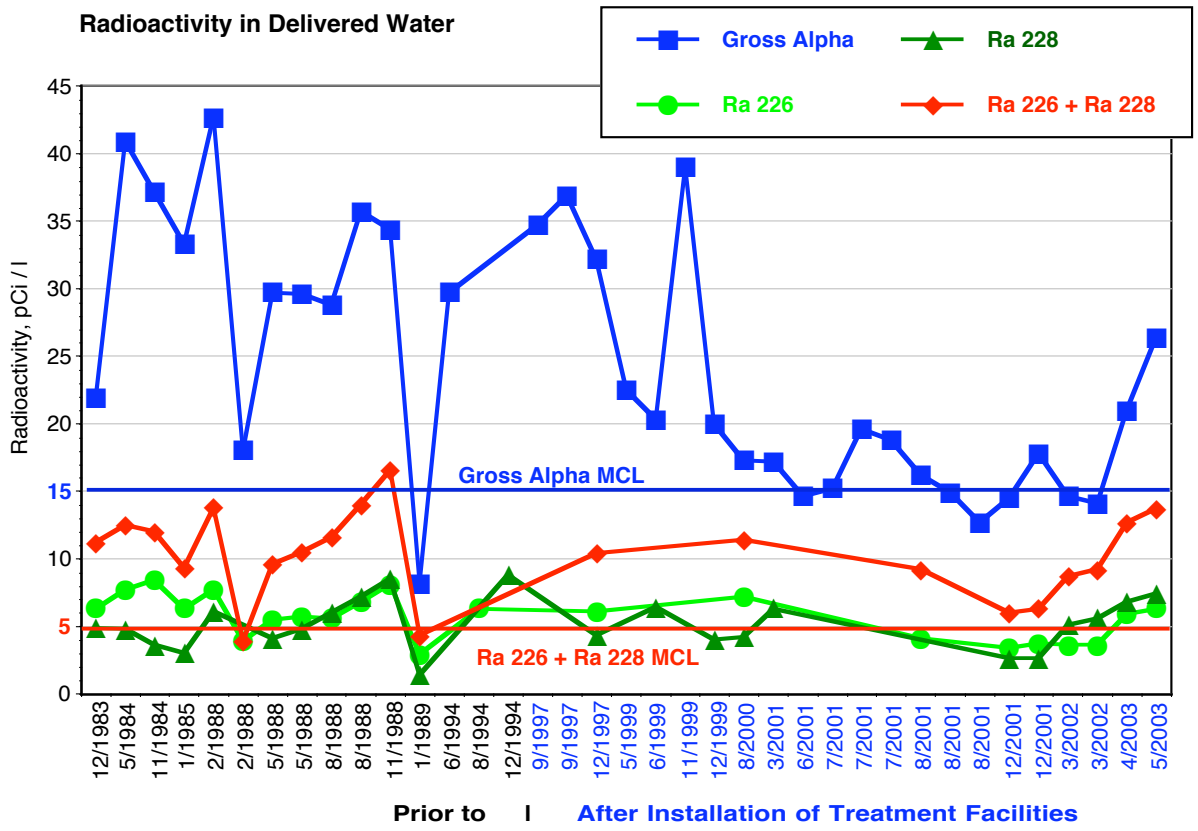
Finally, two studies were conducted using a combination of ferric chloride and aluminum sulfate feeds (both at 0.5 and 1 mg/l levels). Again, the results were very similar to all of the previous results. Since no marked improvement was observed either with increased chemical dosage or with iron and aluminum in combination, 0.5 mg Fe/l was recommended for plant-scale operation. Oddly, as indicated by the linear trend lines shown in the graph below, the efficiency of removal of radionuclides appeared to decline slightly during the progress of the testing program.



To verify the results of the pilot testing program, the current evaluation was undertaken to observe the effectiveness of this treatment process on a full plant-scale.

Radionuclide Removal Performance

Historical data on the concentrations of *gross alpha emitters* and *radium*^{226} *plus radium*^{228} in Leadwood's water supply, as delivered to consumers, is shown in the following plot. Although data taken prior to the installation of facilities in 1997 is extremely limited, it reflects the concentrations of radionuclides observed in Wells #1 and #2.



Based on all available historical data, the plant has removed an average of 35% of gross alpha, 22% of *radium*^{226}, and only 5% of *radium*^{228}. Gross alpha radiation in treated water averaged 19.4 pCi/l, and *radium*^{226} *plus radium*^{228} averaged 9.7 pCi/l over the period 1998 to 2002. On several occasions, finished water met the 15 pCi/l MCL for gross alpha, however, the finished water has not met the 5 pCi/l MCL for *radium*^{226} *plus radium*^{228} since the plant has been in service.

all units: pCi/l	Gross Alpha	Ra 226	Ra 228	Ra 226 + 228
pre-treatment avg.	30.0	6.2	5.3	10.8
post-treatment avg.	19.4	4.9	5.0	9.7
% decrease	35%	22%	5%	10%

Recent Trends

The most recent analyses conducted for the current study (April; May 2003) indicate a slight increase in the concentrations of radionuclides in Leadwood's delivered water. Gross alpha concentrations were recorded as 21 and 26.4 pCi/l while *radium*^{226} *plus radium*^{228} totaled 12.7 and 13.7 pCi/l. If these analyses are accurate, there appears to be an upward trend in both parameters.

When the sample was collected in April, no iron was being added to the well water influent. Alternately, when the filter effluent was sampled in May, 0.5 mg/l of ferric chloride was being added and oxidized prior to filtration. Even so, the May sample of finished water exhibited a higher level of radionuclides than the April sample.

The implication of this result is that the ferric hydroxide precipitate formed at a dosage of 0.5 mg/l is not effectively adsorbing the radionuclides. Instead, the May radionuclide levels in the finished water have again approached the levels observed before the installation of treatment in 1999. Minimum levels of radionuclides were observed in December 2001.

Restoration of Plant Operating Condition

Prior to determining the radionuclide removal effectiveness of the existing plant, the treatment train was restored to original manufacturer's engineering design specifications. Based on the operations manual and recommendations from Mike Hoyer and Bill Webb of Filtronics, the following steps were taken.

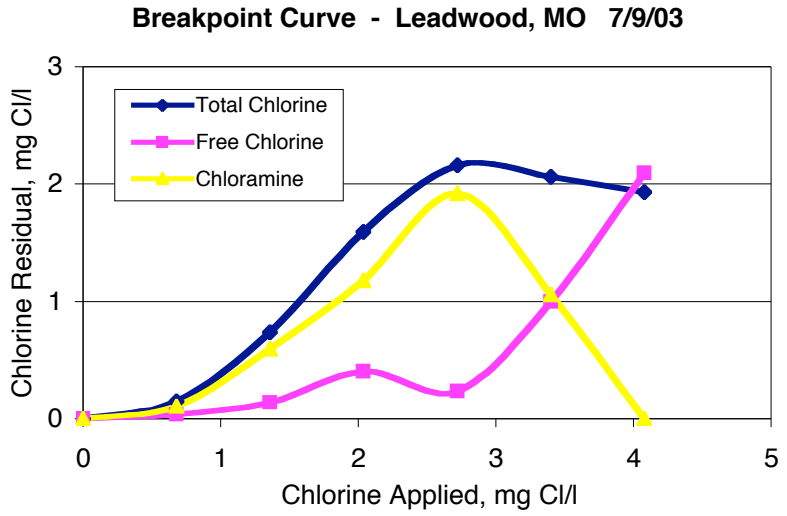
Backwash and Filter Flow Rates

The backwash rate setting was confirmed at approximately 510 gpm for a total of 4.4 minutes duration. The temperature of Leadwood's ground water, which remains fairly constant throughout the year, was about 62° Fahrenheit. Flow rates for both Filter #1 and Filter #2 were set at 100 gpm.

Chemical Feeds

At the time of resetting, the rate of ferric chloride feed was yielding an iron concentration of 0.66 mg Fe/l. The rate of iron feed was subsequently increased to 1 mg Fe/l for the new operational protocol.

The appropriate chlorine dose was determined by conducting laboratory tests to establish a breakpoint curve for Leadwood's well water. The results confirmed Leadwood's current chlorination practice that resulted in the formation of a 1 mg Cl/l free chlorine residual in the filtered water.



Based on the laboratory testing, the application of, at least, 4 mg Cl/l will satisfy the chlorine demand and result in a free chlorine residual of approximately 2 mg Cl/l.

The breakpoint curve plot indicates that the immediate chlorine demand was satisfied by 0.5 mg Cl/l. Approximately 1 mg Cl/l of chloramine and 1 mg Cl/l as free chlorine was formed at an applied chlorine dose of 3.5 mg Cl/l.

Ferric Chloride and Sulfur Dioxide Feed Systems

At the time of resetting, sulfur dioxide was not being fed post-chlorination. However, to conform with the manufacturer's recommendation, the sulfur dioxide feed was reestablished so as to feed 0.25 mg SO_2/l. According to Filtronics, this low sulfur dioxide dosage was considered to be sufficient for waters containing high concentrations of sulfate ion.

Filter Break-In Period

A number of filter backwashes were performed throughout the break-in period for the restored filter in order to restratify and condition the media as well as wash out filter media fines. Initially clear, the appearance of the backwash water turned orange-brown after a minute of backwash and cleared readily thereafter. There was no evidence of media loss at this time, although operators reported a slight loss upon initial backwashing a day earlier.

Filter Media Replenishment

Both filters were opened and inspected to determine media loss. At the outset of the project, this was suspected to be the cause of the plant's inability to achieve the degree of radionuclides removal exhibited by the pilot testing.

The filter media was core-sampled using a one-inch, thin-walled PVC pipe to obtain a vertical section through the filter depth. When initially installed, the proprietary filter media (Filtronics *Electromedia 1*) consisted of multiple layers. It had been suggested that the upper layer(s) of media had been lost as a result of excessively high rates of backwash when the plant was initially placed into service in 1997. However, the continuing reduction in radionuclides that reached a minimum in December 2001 suggested that radionuclide removal continued beyond 1997. Moreover, turbidity reductions have reportedly been consistently effective since the start of plant operations.

Based on their inspection of the filter media core sampling, Filtronics determined that the crushed anthracite coal (top) layer remained intact, but the soluble, white (middle) layer had been dissolved and/or fouled by iron deposits. Based on their analysis and recommendation, that portion of missing and fouled filter media was purchased from Filtronics and shipped to Leadwood in July, 2003.

The restoration of the filter media took place on July 8, 2003 with the introduction of the 1,500 pounds (3 drums) of Electromedia I media into the manway atop Filter #2. This was followed by several backwashings to restratify the filter bed. No media addition was made to Filter #1 so that a comparison could be made between radionuclide removals achieved by a restored and a depleted filter.

Two carbon steel,vertical filter vessels containing Filtronic's *Electromedia 1*

H_2O'C Engineers, John and Tom O'Connor, traveled to Leadwood on July 9, 2003 to meet with representatives of MDNR (Tim Robbs, Deana Cash) and USEPA Region VII (Robert Dunlevy) to review progress on the project. At that time, the engineers also worked to evaluate, restore, and confirm the initial Filtronics plant operating conditions in conjunction with Leadwood staff (Raymond Moore, Van Gilliam).

Overall, the operators appeared to be knowledgeable of the required operational protocols, familiar with the Filtronics operations manual, and eager to operate the plant so as to achieve the optimum removal of radionuclides.

With completion of the restoration of filter media and the resetting of chemical feed rates, the plant was considered ready for evaluation of its performance with respect to the removal of radionuclides.

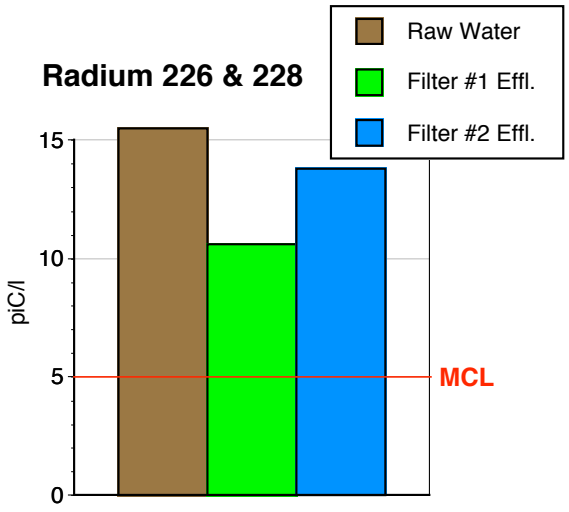
Confirmation of Plant Restoration Program

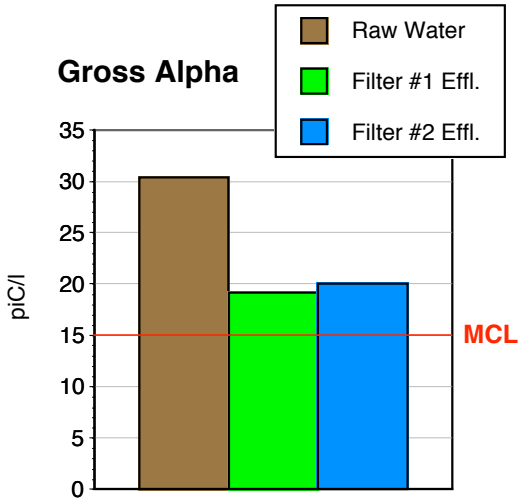
A final check was made with Filtronics (July 14; 22, 2003) on the restoration of proper operating conditions. The details described above and chemical feed dosages were considered appropriate for restoring the plant to its original operating condition.

With all settings made and chemical feeds in operation, preparations were made for the final radionuclides testing round. The final testing included three samples: Raw Water; Filter #1 (depleted media) effluent; Filter #2 (restored media) effluent. As before, each was analyzed for gross alpha emitters and radium^{226} plus radium^{228}.

Results

Analytical results showed modest removal of both gross alpha and radium comparable to plant historical data. Interestingly, the control filter (#1) performed slightly better than the filter with fresh media added (#2). This could possibly be due to radium being adsorbed to iron built up on the old media. While both filters removed approximately one-third of gross alpha, the control filter removed significantly more radium. However, in all cases, removals were again insufficient to meet the MCLs.





all units: pCi/l	Raw Water	Filter #1 Effl.	% removal	Filter #2 Effl.	% removal
Gross Alpha	30.4	19.2	37%	20.0	34%
Ra-226	6.7	5.2	22%	6.0	10%
Ra-228	8.8	5.4	39%	7.8	11%
Ra-226+228	15.5	10.6	32%	13.8	11%

Based on these results and Leadwood's experience, it does not appear that the existing treatment train in its present configuration is capable of consistently removing radionuclides from Leadwood's well water.

Treatment Process Modifications

Leadwood's radionuclide removal performance could be enhanced by modifying the existing treatment train. Leadwood's physical plant could be converted to one that utilizes one of the following treatment technologies.

1. Pre-formed Hydrous Manganese Oxide (HMO) Filtration
2. Greensand Filtration
3. Coprecipitation of Radium with Barium Sulfate
4. Ion Exchange

Pre-formed Hydrous Manganese Oxide (HMO) Filtration

This removal technology consists of a pre-formed HMO feed and filtration. This would be relatively easy to pilot test and to implement full-scale in Leadwood, due to the existing oxidation/filtration treatment configuration of the plant. In fact, the only modification could be the replacement of the ferric feed with the HMO. From an EPA compliance technology document (815-R-98-02, Sept. 1998):

> **Pre-formed Hydrous Manganese Oxide (HMO) Filtration**
>
> Although no municipal installations are currently reported in the literature, pilot studies have been completed and a full scale demonstration is planned (Clifford 1990a). Pilot studies conducted in Illinois consisted of a pre-formed MnO_2 (HMO) dosage of 2.0 mg/L, followed by a minimum of ten seconds of mixing and post-mixing diatomaceous earth filtration or multi-media filtration to remove the suspended MnO_2 (which contains the sorbed radium). Greater than 96% removals were consistently achieved over the five day run period (Clifford 1990a). Valentine et al. (1992) reported average removals of 83%, 79%, and 71% for HMO dosages of 1.0, 0.5, and 0.2 mg/L (as Mn). Where applicable, costs for HMO treatment were projected to be quite low if filtration is already in place (EPA 1994).
>
> HMO filtration is similar to oxidation/filtration in complexity and operator skill requirements. Proper dosages must be determined, and if water quality is variable, the dosage must be re-calibrated. But, since radium is typically a ground water problem, this is less of a concern for this application. Once the proper dose is determined, dosing is relatively easy, requires simple equipment, and is fairly inexpensive (NRC 1997). Filters must be backwashed, which may require intermediate operator skill. Radium containing wastes include HMO sludge, filter backwash, and sludge supernatant.

Greensand Filtration

With a potassium permanganate feed and greensand filtration, studies have indicated radium removals of 56-90%, depending on detention time prior to filtration. Again, this would be relatively easy to pilot test and to implement full-scale. From EPA:

Greensand Filtration for Radium Removal

Greensand filtration technology consists of a conventional filter box with the traditional filtration medium replaced by manganese greensand. Manganese greensand has enhanced sorptive abilities that allow it to remove certain dissolved ions in addition to normal filtration. The water is pre-treated with potassium permanganate to oxidize dissolved materials into more insoluble chemical species, which consequently are removed much more efficiently by the greensand filter. This is a continuous process, except when interrupted for filter backwashing. Studies indicate that up to 56% of radium is also removed by greensand filtration. In a pilot plant study, the radium removal efficiency was improved to 90% by passing the potassium permanganate pretreated water through a detention tank before filtration.

Wastes generated by this process include sludge and supernatant from the filter backwash, and eventually the greensand media must be disposed of. Radium concentrations in the filter backwash have been found to range from ca. 65 to 170 pCi/L. Dry weight concentrations of radium in the greensand medium have been found to vary from ca. 30 to 46 pCi/g at one plant and from 75 to 250 pCi/g at another (EPA 1994).

Co-Precipitation with Barium Sulfate

Waters high in sulfate ion concentration may be treated with barium chloride to precipitate barium sulfate. Up to 95% of the radium in mine wastewaters has been observed to co-precipitate with barium sulfate. Removal is then achieved by filtration through granular filter media. From EPA:

Coprecipitation of Radium with Barium Sulfate

Addition of a soluble barium salt such as barium chloride to water contaminated with radium and sulfate will result in the coprecipitation of a highly insoluble radium-containing barium sulfate sludge. This process has been primarily used for wastewater treatment. Mine wastewater treatment data indicates that up to 95% of radium is removed by this process. A full-scale drinking water treatment plant in South Dakota currently uses this process to treat a radium influent level of 16 pCi/L down to < 1 pCi/L.

Radium containing wastes generated by this process include the barium sulfate precipitate sludge, filter backwash, and sludge supernatant.

Ion Exchange

Radium ion is effectively removed (up to 97%) by cation exchange softening along with hardness (calcium and magnesium). Although a departure from the general oxidation/filtration configuration of the previous three options, it is possible that the existing Leadwood plant could be modified to accommodate an ion exchange treatment train, converting the existing filters to ion exchangers.

Ion exchange would have the added benefit of also softening that portion of water treated for radium removal. However, it would markedly increase the sodium ion concentration of the softened water.

Wastes generated include rinse and backwash water, and, eventually, the ion exchange resin itself.

Alternative Technologies

Lime Softening

Precipitation of metal carbonates and oxides by the addition of lime to raise pH above 10 also results in the co-precipitation of radium carbonate and uranium oxide. Field and laboratory studies indicate radium removals in the range of 75 to 90%.

Lime softening would require lime storage and slaking, mixing, clarification, filtration, sludge removal and final pH control (acidification, recarbonation). In addition, off-site sludge dewatering and storage (lagoon) facilities would be required.

Although requiring far more operational skill and attention, lime (precipitative) softening would reduce the hardness by 40 to 50% and result in the production of a calcium carbonate sludge containing a trace quantity of radium. Lime treatment is reportedly very effective in the removal of both radium and alpha emitters as well as a wide range of dissolved metals, including iron, manganese, copper, lead, zinc, nickel, etc. Accordingly, lime softening has been awarded BAT status by USEPA.

This option is not particularly attractive to Leadwood due to initial capital cost, land requirements, the complexity of operations, and the diseconomy of small-scale lime softening. However, a regional lime softening plant owned by a wholesale water commission may be feasible.

Reverse Osmosis

Reverse osmosis reduces radium concentrations by 70 to 97%. Reported costs of implementation for small utilities have been at least twice the costs of other treatment alternatives at around $2 to $4 per 1,000 gallons produced.

Reverse osmosis would reduce hardness and total dissolved solids in proportion to the amount of radionuclides removed. It would require comparatively little operator attention, but semi-permeable membrane cleaning (every six months) and replacement (every six years) would add to both costs and operational requirements. The rejected concentrate (perhaps 30% of the inflow) would represent a water production loss and a liquid waste stream slightly enriched (~4x) in radionuclides.

Removal on Radium Selective Complexer Resin

Dow Chemical company manufactures Dowex RSC (Radium Specific Complexer) resin for the removal of radium and barium from water. This ion exchange media has substantial capacity for the specific removal of radium, as opposed to hardness, and should be capable of treating millions of bed volumes of groundwater over several years before exhaustion. Since it is not currently in use in the State

of Missouri, a pilot study would be required to demonstrate the effectiveness of the methodology.

The exhausted resin is not regenerated, but must be disposed of in a low-level radioactive waste disposal facility. There are only three such facilities in the US, the nearest being in Utah.

The Dowex RSC is sold by Layne Christensen of Bridgewater, NJ. A similar product, Z-88, is sold by Water Remediation Technology of Arvada, CO.

This system is relatively expensive. Capital costs may approach $1,000,000, with annual operating costs near $50,000.

Reported Costs of Treatment for Radionuclides Removal

(USEPA: Radionuclides Notice of Data Availability Technical Support Document, March 2000)

Table VII-9

CASE STUDIES: PRODUCTION COSTS FOR REMOVAL OF RADIONUCLIDES FROM COMMUNITY WATER SYSTEMS
(Dollars per thousand gallons treated)

Treatment to Remove Radium from Ground Water [1]				
	Small Systems[2]		Large Systems[3]	
	Range	Average (No. of Studies)	Range	Average (No. of Studies)
Cation Exchange	$0.08 - $3.69	$1.10 (7)	$0.27 - $1.58	$0.89 (3)
Lime Softening	$2.91	NA (1)	$0.15 - $1.80	$0.97 (2)
Reverse Osmosis	$0.54 - $4.34	$2.19 (6)	NA	NA
Greensand Filtration	$0.63 - $1.47	$1.03 (3)	NA	NA
Other Oxidation/Filtration[4]	$0.01 - $2.40	$0.82 (9)	$0.04 - $1.83	$0.63 (5)

Notes:
1. Data source is EPA 1998a, "Actual Costs of Compliance with the Safe Drinking Water Act Standard for Radium-226 and Radium-228", otherwise stated otherwise.
2. Small systems are defined as those serving 10,000 persons or fewer.
3. Large systems are defined as those serving greater than 10,000 persons.
4. Data source is EPA 1998b.

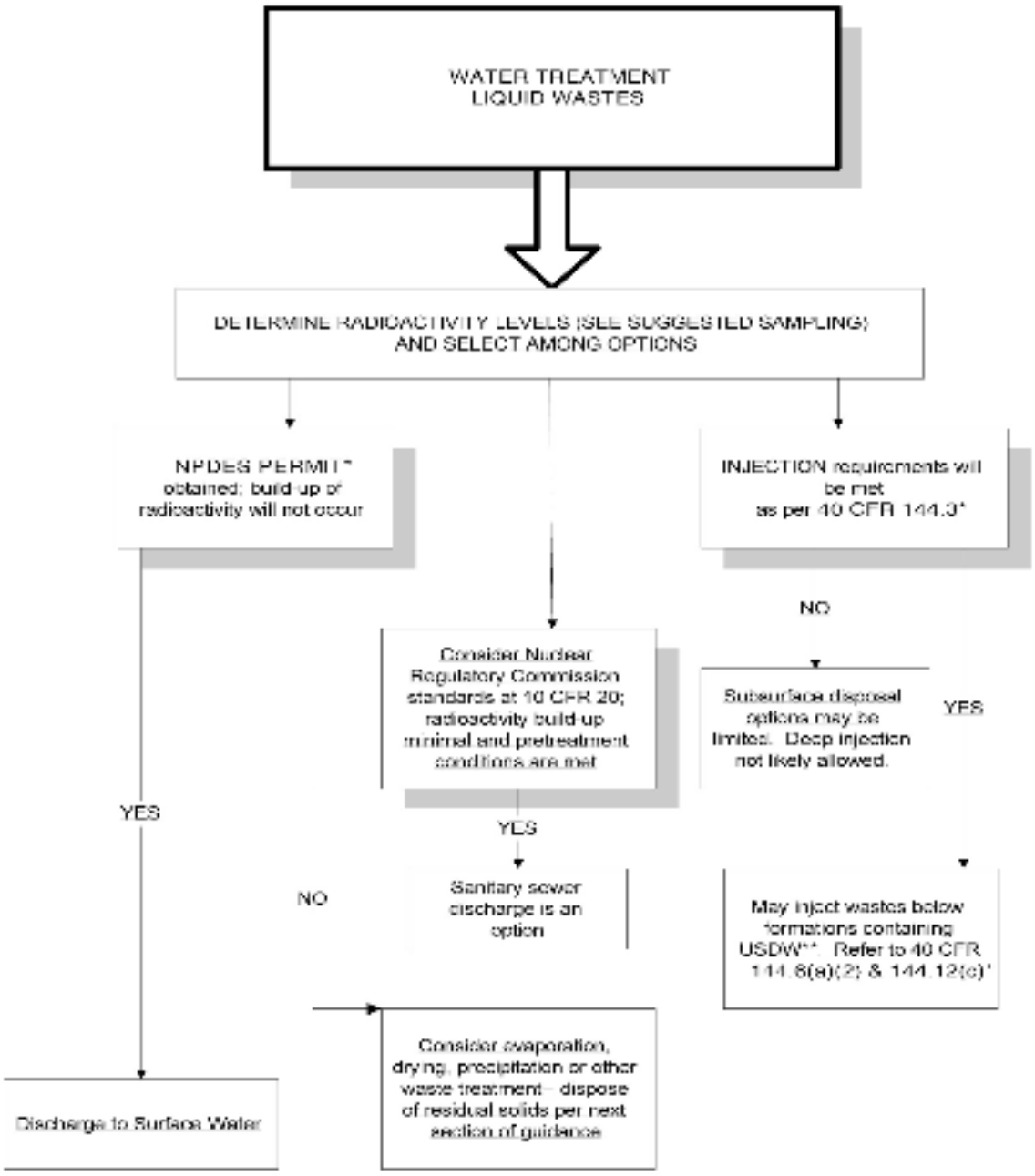
Disposal of Treatment Residues

The feasibility, cost and difficulty of disposing of treatment residues (sludges, waste brine, rinse waters; RO concentrates; radioactive resins) will be a determining factor in the selection of the optimum treatment option for Leadwood.

USEPA has prepared the following flow charts to illustrate disposal options for treatment process liquid and solids/sludge wastes. While it appears possible that a brine waste could be disposed of economically, solid wastes (such as dewatered sludges and radium-selective resin) become ineligible for basic landfilling at very low levels (3 pCi/g).

Summary of Suggested Disposal Alternatives for Liquid Water Treatment Wastes

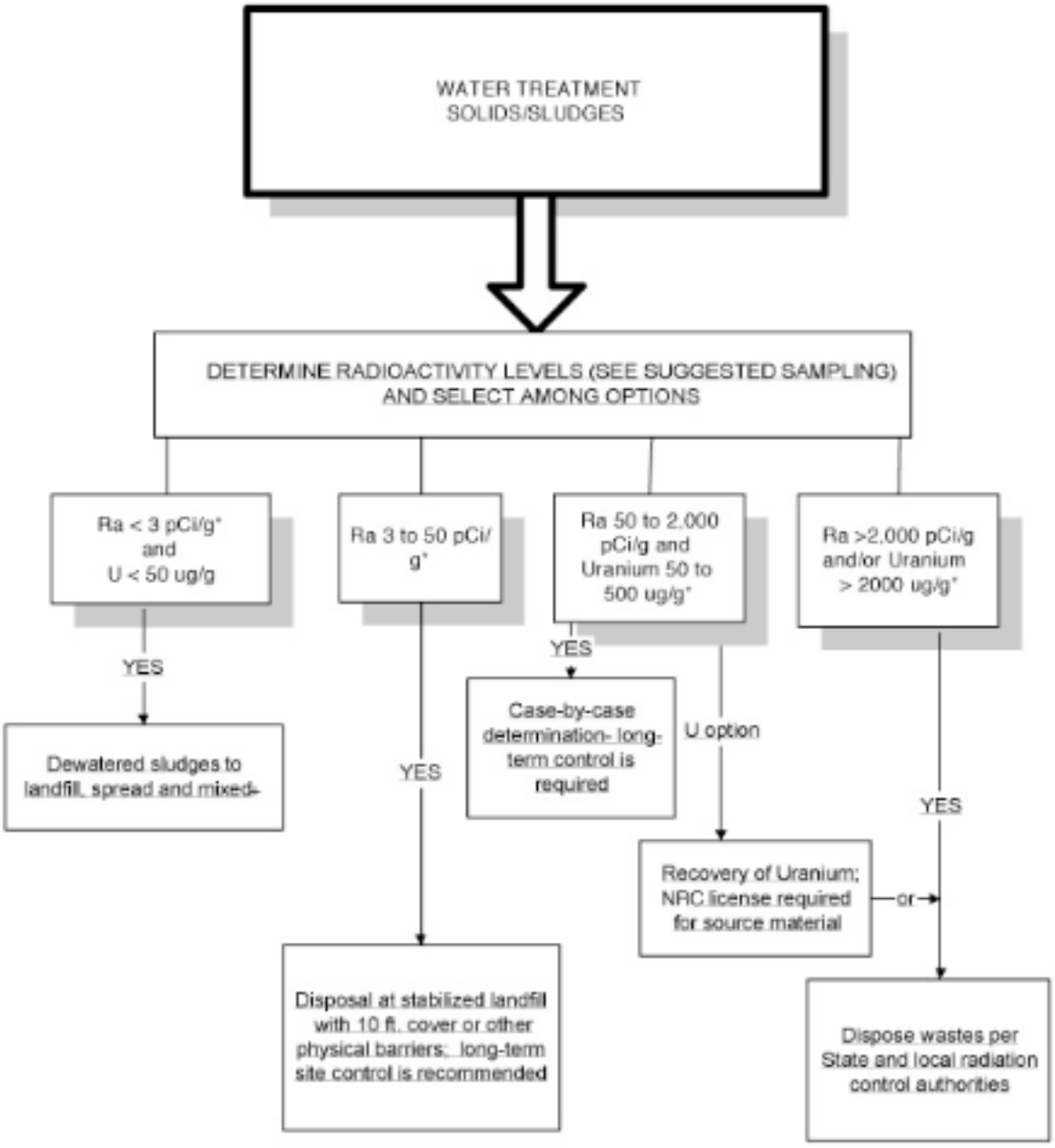
(USEPA: Radionuclides Notice of Data Availability Technical Support Document, March 2000)



* Bold type indicates actual regulatory requirements; other options are EPA suggested disposal methods.
**USDW is underground source of drinking water

Summary of Suggested Disposal Alternatives for Solids/Sludge Wastes From Water Treatment

(USEPA: Radionuclides Notice of Data Availability Technical Support Document, March 2000)



+ Provisions under 40 CFR 257, 258 and 260 thru 266 may apply.

* Bold type indicates Federal regulatory requirements; other options are EPA suggested disposal methods.

Conclusions and Recommendations

Restoring Leadwood's treatment train to original engineering design specifications and replenishing filter media did not have a beneficial effect on radionuclide removal. Based on these results, Leadwood's experience, and historical data, it does not appear that the existing treatment train in its present configuration is capable of consistently achieving sufficient radionuclide removals from Leadwood's well water to maintain compliance with the Radionuclide Rule.

However, options for treatment modification were identified that could improve the system's radionuclide removal. Preliminary engineering and, as warranted, basic pilot testing should be performed for the following three radionuclide removal technologies in the order listed below.

1. Pre-formed Hydrous Manganese Oxide (HMO) Filtration
2. Greensand Filtration
3. Coprecipitation of Radium with Barium Sulfate

Although these three options would best fit the existing plant configuration, it is likely that the conversion of the plant to ion exchange would not require major modifications. This option might be investigated as well.

Additionally, non-treatment alternatives should be investigated. Options such as purchasing water or forming a wholesale water commission can remove many of the ever-increasing burdens that the operation of a water treatment system places on a community.