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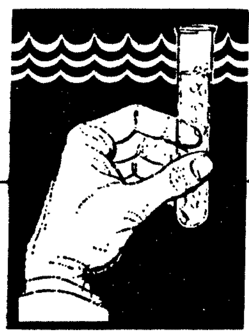
RADIUM REMOVAL FROM GROUNDWATER BY ION EXCHANGE RESIN



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Water Quality Association welcomes the responsibility of helping the public gain a better understanding of our world's water problems and the resources available for solving them.

TECHNICAL PAPER

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Introduction

For many years it has been known that ionizing radiation produces unusual and detrimental biological effects on humans. Many groups, both national and international, have studied the correlation of these levels in respect to the biological effects. The results of these studies and reports were used by the Subcommittee on Radioactivity in Drinking Water of the Safe Drinking Water Committee of the National Academy of Sciences to assess the probable effects on the human population. The results of their assessment were published in a manual entitled "Drinking Water & Health," 1980.

As a result of the information submitted by the National Academy of Sciences and others, the regulatory sector of our government established the maximum contaminant level for radionuclides which was reported in the *Federal Register* on July 9, 1976, Part II. This document reported the establishment of an MCL for Radium 226 and 228 at 5 pCi/L total. When this MCL was established, it became of utmost importance to determine what methods of treatment were available for reducing the high level of naturally occurring Radium 226 and 228.

The objective of this study is to demonstrate the effectiveness that point-of-use/point-of-entry strong acid cation exchange resin (sodium cycle) water softeners have in reducing the radiological contamination of a water supply to a level at or below the MCL of 5 pCi/L as required by the Primary Drinking Water Regulations.

A series of studies[1,2,3,4,5,6,7,8] have reported the effectiveness of ion exchange water softeners in reducing the radiological contaminates to an appropriate level, but none of them actually addressed the relation that ion exchange resins have between hardness capacity and radium reduction. Studies have indicated that some resins have a more selective capacity for Radium 226 and 228 than hardness, and that the removal effect for radium continued even after the resin had been saturated with hardness.[8] This phenomenon would be an additional advantage for public health protection when using ion exchange water softeners on radium contaminated water, a safety factor.

Acknowledgements

We wish to take this opportunity to thank Culligan International Company and Mr. Donald A. Mahlstedt, Vice President and Director of Household Marketing, for supplying the Culligan Mark 89 water softener used in this study. Also, we wish to thank Mr. Joe Bolsinger of Culligan Water Conditioning of Aurora, Inc., for allowing the WQA Laboratory personnel to perform this study at his facilities.

Experimental Procedure

The test site selected for this study was located in the city of Aurora, Illinois, which has its water furnished by the city from an existing groundwater supply. The hardness of the groundwater is approximately fifteen (15) grains per gallon (gpg) as calcium carbonate ($CaCO_3$) and has a natural occurring Radium 226, 228 contamination of approximately twelve (12) pCi/L.

The ion exchange water softener equipment selected for this study was a new, fully automatic time clock controlled, 5-cycle commercially available domestic water softener with a strong acid cation exchange resin. The resin volume contained in the unit was 0.7 cubic feet and was manufactured by Ionac Chemical Co./Division of Sybron Corporation (Trade Name: Ionac C-249) which is a commercially available resin. The unit has a minimum capacity of approximately 21,000 grains exchange when regenerated with 9.5 lbs. of sodium chloride (NaCl).

The ion exchange water softener unit was installed as shown in Figure 1. The influent piping contained a pressure gauge and sample valve. The effluent piping contained a sample valve, 8.0 gpm flow control device and a totalizing meter to record the throughput gallons treated.

Research Approach

As previously described, the ion exchange water softener equipment selected for this study was a new, fully automatic production unit. It has a minimum capacity of approximately 21,000 grains exchange when regenerated with 9.5 lbs. of salt (NaCl) and was allowed to regenerate automatically as determined by the control mechanism to simulate a normal in-house operation of the regeneration cycle. The unit was operated at a service flow rate of 6 to 8 gpm and exhausted and regenerated for three service cycles before collection of samples for laboratory testing. During the fourth cycle of exhaustion, laboratory samples were taken as follows:

one (1) composite influent sample

one (1) effluent sample every 150 gallons (10 samples) until hardness breakthrough (estimated at approximately 1,500 gallons)

one (1) effluent sample every 100 gallons (5 samples) after hardness breakthrough to total throughput of 2,000 gallons which represented a 33% overrun of an exhausted bed

The unit was shut down in the exhausted state for twenty-nine (29) days. On the completion of the twenty-ninth (29) day shutdown period, the unit was started up and operated for a total throughput of 3,000 gallons of water. During the final one thousand (1,000) gallon throughput, five (5) samples were drawn at approximately 200 gallon intervals.

All samples were analyzed for total hardness (gpg) as $CaCO_3$ and for combined 226/228 Radium.

Reporting

The information collected during the course of this study resulted in developing a series of curves which indicated the relationship of gallons throughput vs. hardness and radium reduction.

Results

The effective reduction of hardness and naturally occurring Radium 226/228 are tabulated in Table 1 and graphically displayed on Chart 1. At the hardness breakthrough of 1.0 gpg, the radium level was less than 0.3 pCi/L. As the ion exchange water softener was allowed to continue to complete exhaustion, influent and effluent hardnesses were equal, and the radium concentration was only 0.8 ± 0.4 pCi/L which is well below the PDWR of 5.0 pCi/L. This situation would allow a consumer to operate his water softener for at least 133% of rated capacity and still obtain protection from the contaminants Radium 226/228.

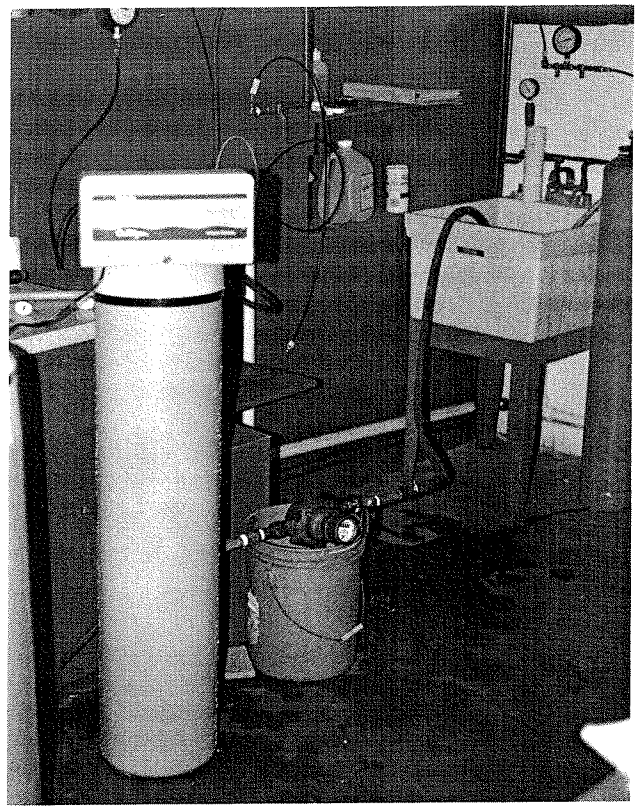


Figure 1

Chart 1

Hardness and Radium 226/228 Breakthrough vs. Total Gallons of Water

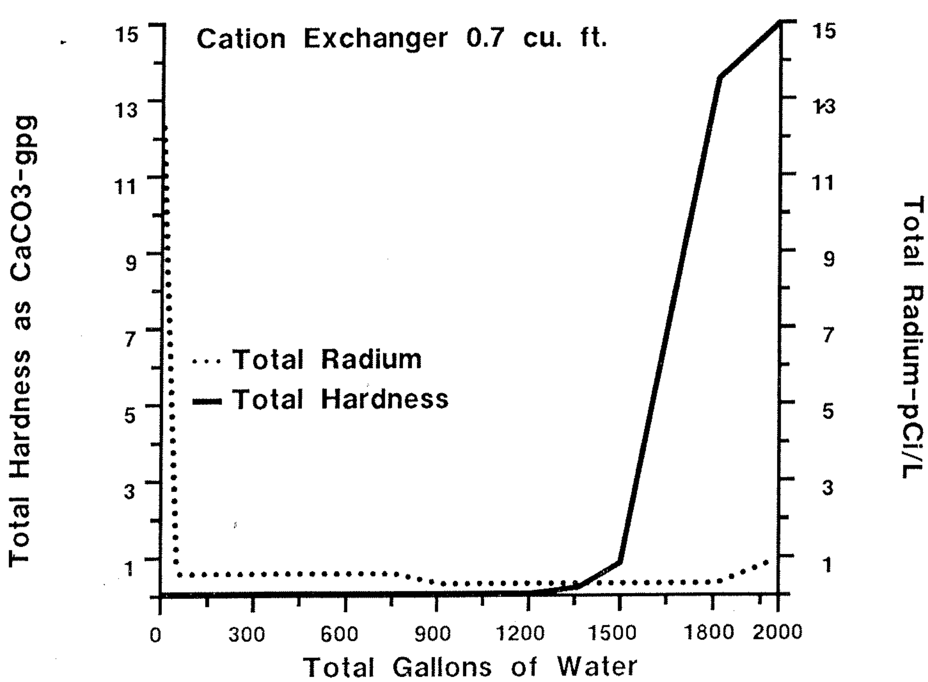


Table 1
Tabulation of Data Accumulated During Test Run

Sample Bottle Number	Total Radium 226/228 (pCi/L)	Influent Hardness grains per gallon (gpg)	Effluent Hardness grains per gallon (gpg)	Meter Reading	Volume of Water (gallons)	Remarks
0	0	0	0	675760		
1	12.3 + − 1.4	15.0				Influent Composite Sample
2	<0.5	15.0	0	675910	150	
3	<0.5	15.0	0	676060	300	
4	<0.5	15.0	0	676210	450	
5	<0.5	15.0	0	676360	600	
6	<0.5	15.0	0	676510	750	
7	<0.3	15.0	0	676660	900	
8	<0.3	15.0	0	676810	1050	
9	<0.3	15.0	0	676960	1200	
10	<0.3	15.0	0.15	677110	1350	
10a	-	15.0	0.3	677160	1400	
10b	-	15.0	0.7	677210	1450	
10c	-	15.0	0.9	677230	1470	
10d	-	15.0	0.95	677250	1490	
11	<0.3	15.0	1	677260	1500	Hardness Breakthrough
12	<0.3	15.0	3.9	677360	1600	
13	<0.3	15.0	9.5	677460	1700	
14	1.1 + − 0.5	15.0	13.5	677560	1800	
15	0.6 + − 0.4	15.0	14.5	677660	1900	
16	0.8 + − 0.4	15.0	15	677760	2000	

Comments: The error given is the probable counting error at 95% confidence level. Less than values (<) are based on 3 sigma counting error for background.

The ion exchange softening unit was shut down in the exhausted state for twenty-nine (29) days at which time the unit was started up and operated for an additional one thousand (1,000) gallons for a total throughput of three thousand (3,000) gallons. As the ion exchange water softener was allowed to continue to excessive exhaustion, the radium concentration was in the range of 0.8 ± 0.4 pCi/L. The effective reduction of naturally occurring Radium 226/228 is tabulated in Table 2 and graphically displayed on Chart 2.

Table 2
Tabulation of Data Accumulated During Test Run

Sample Bottle Number	Total Radium 226/228 (pCi/L)	Influent Hardness grains per gallon (gpg)	Effluent Hardness grains per gallon (gpg)	Meter Reading	Volume of Water (gallons)	Remarks
0	6.4 + − 1.1	15.0				Influent Composite Sample
1a	0.8 + − 0.6	15.0	15.0	678060	2300	
2a	0.6 + − 0.6	15.0	15.0	678210	2450	
3a	0.6 + − 0.5	15.0	15.0	678360	2600	
4a	0.8 + − 0.6	15.0	15.0	678510	2750	
5a	0.8 + − 0.5	15.0	15.0	678760	3000	

The error given is the probable counting error at 95% confidence level.

Samples of the ion exchange resin used for the previous testing were then removed from the ion exchange resin tank and analyzed for total radium. The reduction of total radium concentration between an exhausted resin and regenerated resin would be an indication of the effective removal of Radium 226/228 from the resin during the regeneration cycle. Table 3 indicates the results of such sampling and testing.

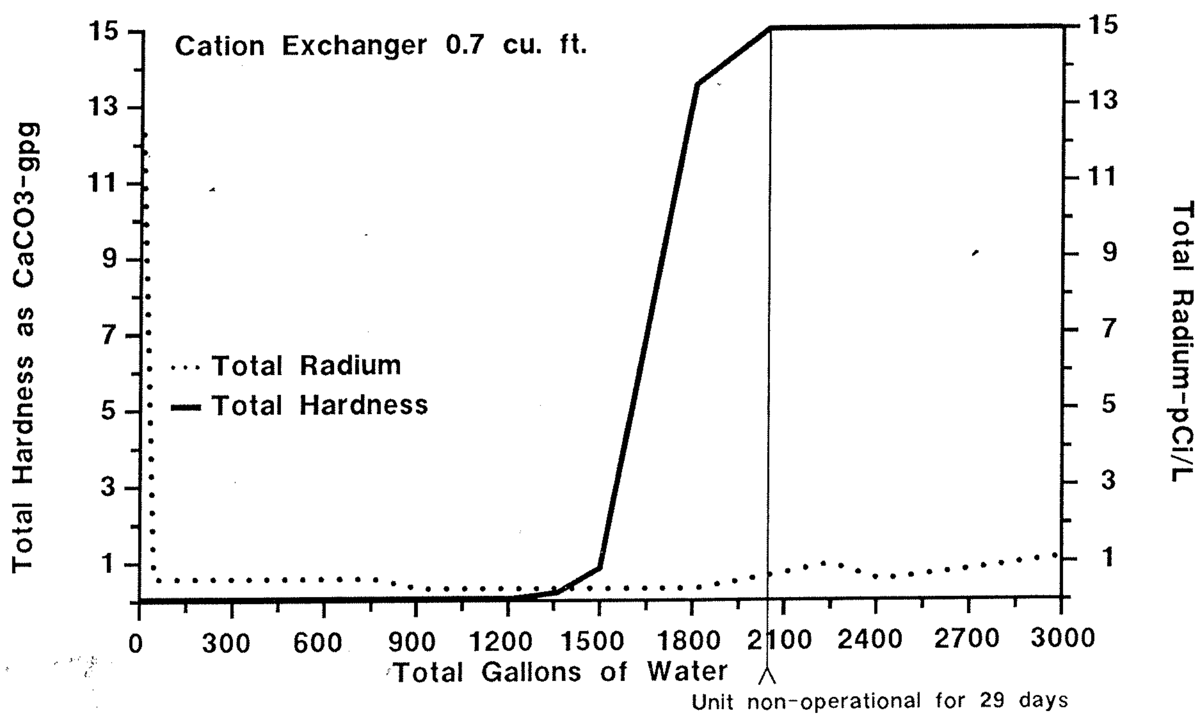
Table 3
Ion Exchange Resin—Total Radium

Sample Description	Lab Code	Concentration (pCi/G wet) Total Radium
Non-Regenerated (never used)	SPW-2552	0.006 ± 0.004
Exhausted Resin	2553	5.440 ± 0.090
Regenerated Resin	2554	0.020 ± 0.006

The error given is the probable counting error at 95% confidence level.

Chart 2

Hardness and Radium 226/228 Breakthrough vs. Total Gallons of Water



Conclusions

The results of this study indicate the effectiveness of a point-of-use/point-of-entry strong acid cation exchange resin (sodium cycle) water softener has in reducing the radiological contamination of a water supply to very low levels. It also demonstrated that an exhausted ion exchange bed has an excellent ability to reduce these contaminants even when its resin bed has exceeded its effective hardness reduction capabilities.

Recommendation For Future Study

Further studies should be undertaken to demonstrate the effectiveness of a point-of-use/ point-of-entry strong acid cation exchange resin softener (in the sodium cycle) to reduce Radium 226 and 228 in a contaminated water supply, utilizing a resin bed that has been in use in a domestic application for five (5) to ten (10) years. The ion exchange water softener equipment selected for such a study should be done on a random basis. Appropriate on-site radiation monitoring of the selected unit should be done prior to implementing the testing program. This unit should be allowed to regenerate automatically as determined by the control mechanism to simulate in-home operation of the regeneration cycle. The unit should be operated at a service flow rate appropriate for the unit's design and should be exhausted and regenerated for at least three (3) cycles before samples are collected for laboratory testing. During the fourth cycle of exhaustion, a laboratory sample should be taken as follows:

one (1) composite influent sample every 1,500 gallons

one (1) effluent sample every 150 gallons until hardness breakthrough (less than 1 gpg as $CaCO_3$)

one (1) effluent sample every 500 gallons after hardness breakthrough for a total gallonage flow of 15,000 gallons

All samples will be analyzed for total hardness (gpg) as $CaCO_3$ and for combined 226/228 Radium.

The ion exchange resin bed should be sampled to determine the amount of Radium 226/228 retained by an exhausted bed and regenerated bed.

References

1. *Radioactivity in Drinking Water*, pages 892 through 903, from the volume entitled DRINKING WATER AND HEALTH, published by the National Academy of Sciences, 1977.

2. Technical Letter 19 entitled *Removal of Water Supply Contaminants—Radium*, published by Illinois State Water Survey, February 1977.

3. *Determination of Radium Removal Efficiencies in Water Treatment Processes*, U.S. Environmental Protection Agency, December 1976.

4. *The Efficiency of Water Treatment Processes in Radium Removal*, AWWA Journal, December 1978.

5. *Point-of-Use Treatment for Compliance With Drinking Water Standards*, Temple, Barker & Sloane, Inc., May 16, 1983.

6. *Methods of Radioactivity Removal,* Report to the Atomic Energy Commission, 1946, William J. Lacy.

7. *Treatment Technology to Meet the Interim Primary Drinking Water Regulations for Inorganics: Part 5*, Thomas J. Sorg and Gary S. Logsdon, AWWA Journal, Research and Technology, July 1980.

8. *Barium and Radium Removal from Groundwater by Ion Exchange*, Vernon L. Snoeyink, Julie L. Pfeffer, David W. Snyder, and Carl C. Chambers, September 1984.