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NATURAL RADIOACTIVITY IN NORTH CAROLINA GROUND WATER SUPPLIES

by

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ABSTRACT

In North Carolina over 2,000 public water systems have been monitored for radionuclides by the State Laboratory of Public Health as part of the Safe Drinking Water Act requirements. In this study, the results from 2,089 water systems monitored were examined. Of these systems, 51 or 2.4 percent of the systems tested exceeded standards for radionuclides in drinking water. Thirty-nine systems had combined radium in excess of the 5 pCi/l standard and an additional 12 systems had gross alpha concentrations in excess of the 15 pCi/l standard. Potential health effects on humans of these levels of radionuclides are examined. Types of water treatment needed for radionuclide removal are discussed. Recommendations are presented for treatment of water systems and for the evaluation of criteria for radium-228 monitoring and the need for a uranium standard.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

The Safe Drinking Water Act monitoring requirements for radionuclides in public water supplies initiated the largest radiological inventory of ground water ever before undertaken. In the State of North Carolina as of August 1, 1982, 2,089 water systems had been analyzed. Of these systems, 2.4 percent or 51 water systems were found in violation of federal and state standards. Thirty-nine systems or 1.9 percent of those systems tested had combined radium violations; an additional 12 systems or 0.6 percent had gross alpha violations and 14 of these systems or 0.7 percent had both combined radium and gross alpha violations. For systems in violation of the standards, the average concentration of gross alpha was 29.5 pCi/l, and the average concentration of combined radium was 12.2 pCi/l.

Using the average level of radium (12.2 pCi/l) and the percent of systems with radium violations (1.9 percent of those systems tested), and the present population of North Carolina using ground water (3 million), it can be calculated [using methods recommended by the Environmental Protection Agency (1976b) based on the linear extrapolation of the dose effect relationship] that one person may die every 4.8 years from cancer due to the ingestion of radium.

Based on the concentrations of radium found in ground water used for drinking purposes in North Carolina and the consequential health effects of this form of radionuclide, the State Department of Human Resources and/or the Environmental Protection Agency should initiate treatment of water high in radium.

An examination of the $^{228}Ra/^{226}Ra$ ratios for systems exceeding the radium standard indicates that in approximately a third of these systems,

radium-228 concentrations exceed the levels of radium-226. To be sure that all systems are identified which have combined radium in excess of 5 pCi/l, separate monitoring of both radium-226 and radium-228 would be necessary if the levels of gross alpha activity exceed 5 pCi/l. Consideration of the cost-benefit aspects of radium-228 monitoring is needed to evaluate the current criteria for monitoring.

More than half of the systems examined with a level of gross alpha particle activity in excess of 15 pCi/l were also shown to have levels of uranium in excess of 10 pCi/l. Examination of levels of uranium in ground water systems having elevated levels of gross alpha activity should continue. If levels of uranium in North Carolina in excess of 10 pCi/l continue to be found in as many systems as was found in this study (approximately 50 percent of those systems with gross alpha exceeding 15 pCi/l) and at concentrations averaging near that found in this study (27 pCi/l), standards for uranium should be considered.

Like chemical or biological contaminants, radiological contaminants are cause for concern. If this concern motivates study to learn as much as possible about radiological contaminants and their distribution, we can deal with them and improve our environment.

Recommendations

1. Treatment should be initiated on water systems with radium concentrations in excess of the standard.
2. Cost-benefit analyses of radium-228 monitoring should be performed to evaluate the adequacy of current monitoring requirements.
3. Examination of levels of uranium in ground-water systems should continue in order to evaluate the need for a uranium standard.

INTRODUCTION

Purpose

The Safe Drinking Water Act initiated the largest radiological survey of drinking water ever performed in North Carolina. All community public water supplies must monitor the radiological contents of their water according to federal and state regulations. The objective of this study was to examine the available 2,089 water system analyses for samples collected between June 24, 1979, and August 1, 1982. The results are compared with the regulations, and possible effects on humans are examined. The types of water treatment needed for radionuclide removal are discussed. Possible improvements to the Safe Drinking Water Act and improvements in monitoring procedures for radionuclides are addressed.

Federal and State Standards

Generally, large water systems utilize surface water, which on the whole contains very low concentrations of radionuclides unless there has been a man-induced discharge from an upstream facility. Small water systems, usually serving less than 500 people, commonly use ground water, which may contain naturally occurring radionuclides. Uranium and its decay products, radium and radon, are the radionuclides of principal interest.

On August 14, 1975, the Environmental Protection Agency (EPA) proposed National Interim Primary Drinking Water Regulations for radioactivity pursuant to the Safe Drinking Water Act, P. L. 93-523. Taking effect on June 24, 1979, in the State of North Carolina, these regulations made it mandatory that all community public water supplies (defined as having at least 15 service connections used by year-round residents or regularly serving at least 25 year-round residents) are required to have their water

analyzed for radioactivity. Compliance is based on the analysis of an annual composite of four consecutive quarterly samples or the average of the analyses of four separate samples obtained in consecutive quarterly intervals. This procedure is designed to minimize the chance of variation in the radionuclide content due to seasonal changes in hydrologic conditions.

The Safe Drinking Water Act directed the EPA to set standards for drinking water that "shall protect health to the extent feasible, using technology, treatment techniques and other means, which the Administration determines are generally available (taking costs into consideration)." The maximum contaminant levels are based on the assumption that there is no harmless level of dose from ionizing radiation and that any detrimental effects on health produced by the radiation will be proportional to the dose equivalent delivered. The EPA policy utilized the best available scientific knowledge and assumed a linear, non-threshold relationship between the magnitude of the radiation dose received and adverse health effects produced. The lack of information on effects of low doses of ionizing radiation caused the EPA to choose a conservative method, using a linear extrapolation from data for high doses (Cothern, 1981).

The Environmental Protection Agency has set a limit for man-made radionuclides of 4 mrem per year. This corresponds to 20,000 pCi/l for tritium or 8.0 p Ci/l for strontium-90. (Environmental Protection Agency, 1976a). Only communities with a population greater than 100,000 using a surface water source are required to monitor for man-made radionuclides. None of the systems in North Carolina exceed these limits; and therefore, man-made radionuclides are not monitored in North Carolina.

For naturally occurring radionuclides, the Environmental Protection Agency has set a limit of 15 pCi/l for gross alpha particle activity

(including radium-226, but excluding radon and total uranium), and 5 pCi/l for combined radium-226 and radium-228 (Environmental Protection Agency, 1976a).

The average dose to bone from the continuous ingestion of water with a radium-226 concentration of 5 pCi/l reaches an equilibrium value of 150 mrem per year. The radiotoxicity of radium-226 (an alpha particle emitter) and radium-228 (a beta particle emitter) is very similar when decay products are considered, giving similar doses to bone for equal concentrations.

The estimated risk of death from bone cancer (80 to 85 percent of radium is retained in bone material) and other forms of cancer such as leukemia (approximately 15 percent of radium is deposited in soft tissue) from consuming two liters of drinking water per day containing 5 pCi/l of radium is 1.5 cancer cases per year per million persons exposed (Environmental Protection Agency, 1976b).

The maximum contaminant level for gross alpha particle activity is based on the radiotoxicity of other alpha particle-emitting contaminants relative to radium. The 15 pCi/l gross alpha particle limit (which includes radium-226) is based on the assumption that if the radium concentration accounts for 5 pCi/l and the balance of the alpha particle activity is due to the next most radiotoxic alpha particle-emitting chain, the total dose to bone would be equivalent to less than 6 pCi/l to radium-226 (Environmental Protection Agency, 1976b). The maximum contaminant level for gross alpha particle activity excludes any uranium or radon that may be present in the sample. The monitoring of radon gas (^{222}Rn), with a relatively short half-life of 3.8 days, in the large number of systems involved and with the detailed sampling technique required would be expensive. The decision on whether to monitor for radon is a difficult one still being considered by

EPA. Uranium is also not limited in drinking water by the gross alpha contaminant limit, but the creation of a total uranium standard (combined uranium-238 and uranium-234) is presently being considered. The EPA is currently gathering data on the efficiency and cost of uranium removal by means of anion exchange. This will form the basis for the eventual decision as to whether to set a uranium limit and to make uranium monitoring mandatory (personal communication, C. R. Cothern, 1982).

In addition to the actual limits of activity of radionuclides in drinking water, the sequential determination of whether to test for a particular radionuclide is also contained in the National Interim Primary Drinking Water Regulations (Environmental Protection Agency, 1976a); see Figure 1. The first step in determining the concentration of radionuclides in drinking water is the measurement of the gross alpha particle activity. If the gross alpha particle activity is less than 5 pCi/l, the water supply is in compliance and the analysis need go no further. However, if the gross alpha particle activity is greater than 5 pCi/l, the activity of Ra-226 must be determined. If the concentration of Ra-226 is greater than 3 pCi/l, Ra-228 must then also be analyzed. If the total activity of Ra-226 and Ra-228 exceeds 5 pCi/l, a violation of the maximum contaminant level for radium exists. If the gross alpha particle activity exceeds 15 pCi/l, a violation of the maximum contaminant level for gross alpha particle activity exists. All measurements of the gross alpha particle activity are to exclude any activity from radon and uranium but include radium-226 activity (Environmental Protection Agency, 1976a); see Figure 1.

These standards have been formulated by use of judgments relating the presence of radioactive materials, the ionizing radiation which they emit, the absorption behaviorism of the radiation and the potential effects on

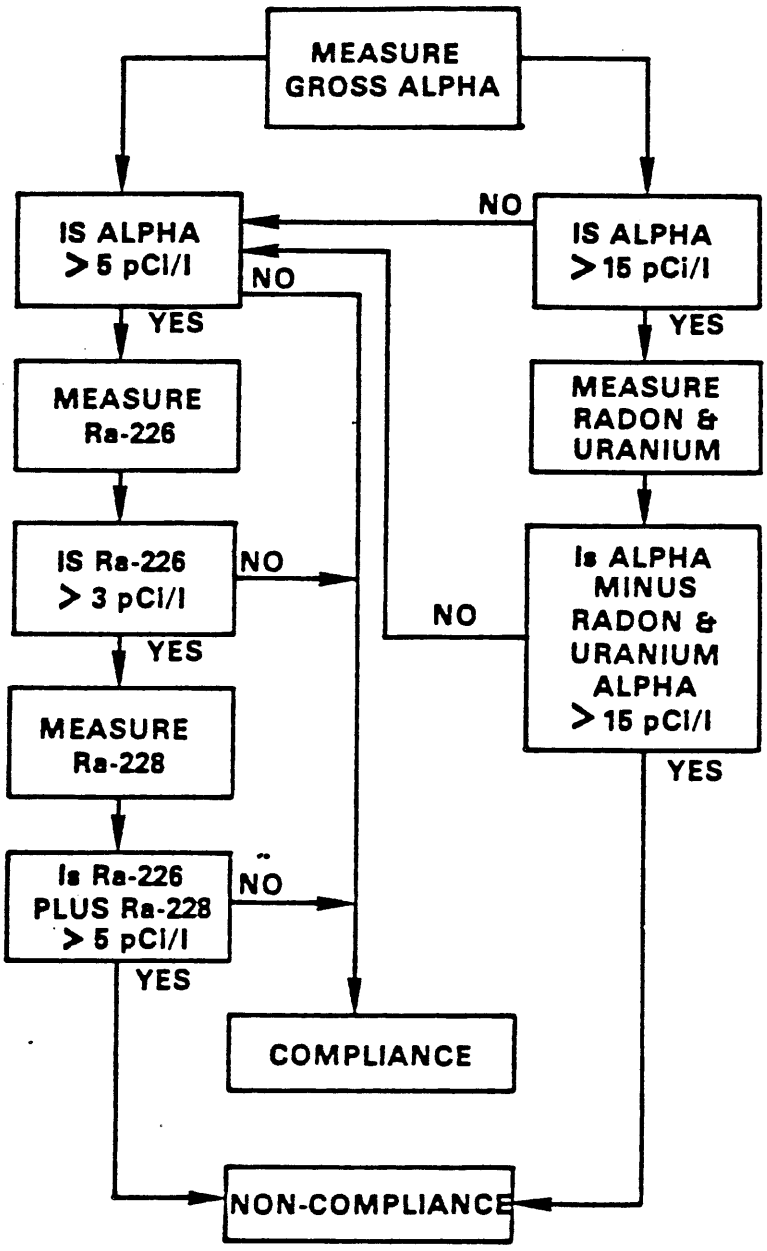


Figure 1. Flow Chart for Gross Alpha Particle Activity and Radium Monitoring

human health (Environmental Protection Agency, 1976b). The activity levels of radionuclides can be directly measured, and radiation doses can be calculated using models; but the relationship between doses to humans and health effects are derived from human epidemiological studies and research involving animals and other biological systems (Environmental Protection Agency, 1976b).

All radium cannot be removed from drinking water, but cation exchange should be able to remove sufficient quantities of radium from any ground water source to meet the limit of 5 pCi/l. Even at relatively high levels of 20 to 40 pCi/l a removal efficiency of approximately 90 percent from cation exchange will enable the water source to be within the 5 pCi/l standard. This is an important aspect on the determination of a feasible standard (Environmental Protection Agency, 1977).

PROCEDURES

With the issuance of the Safe Drinking Water Act and the Primary Drinking Water Regulations for Radioactivity, the EPA recognized the need for a comprehensive collection of analytical procedures for the measurement of radionuclides in drinking water. Considerations such as accuracy, analytical cost, reliability and practical time limitations were used to select these procedures. The procedures were selected from numerous radiochemical methodologies and are used as the prescribed method of analysis.

Water samples were measured for gross alpha particle activity, radium-226 and radium-228 using EPA's Interim Radiological Methodology for Drinking Water (Environmental Protection Agency, 1976c). Uranium was measured using the method "Microquantities of Uranium in Water by Fluorometry," as specified by the American Society for Testing and Materials Annual Book of ASTM Standards, Water and Atmospheric Analysis (ASTM, 1975) and analytical techniques specified in Osmond and Cowart, 1976 (Osmond and Cowart, 1976).

Three measurements of uranium-234 and uranium-238 were made by Ms. J. Michel at the University of South Carolina Geology Department Laboratories using analytical techniques outlined in Osmond and Cowart (1976). All other analyses followed procedures outlined in the EPA and ASTM methods previously mentioned and were performed at the North Carolina State Laboratory of Public Health by Ms. Shirley Bell. Annual composites of four consecutive quarterly water samples were analyzed.

RESULTS AND DISCUSSION

Natural Radioactivity in North Carolina Water Supplies

Gross Alpha and Radium Concentrations

Of the 2,089 systems analyzed, those that exceeded the Safe Drinking Water Act maximum contaminant level for gross alpha and radium concentrations are listed in Tables 1 and 2, respectively. The gross alpha values do not exclude uranium concentrations (as specified by the standard) because these data were not available at the time of this study for many systems.

The highest data obtained were 80.7 pCi/l for gross alpha particle activity (five times the limit), 17.2 pCi/l for radium-226, 22.7 pCi/l for radium-228, and 38.6 pCi/l for combined radium (over seven times the limit). Of the 2,089 supplies tested, 51 water systems or 2.4 percent of those systems tested were found to be in violation of federal and state standards by exceeding the maximum contaminant levels. Thirty-nine systems or 1.9 percent of those systems tested had combined radium violations; an additional 12 systems or 0.6 percent had gross alpha violations and 14 of these systems or 0.7 percent had both combined radium and gross alpha violations. Thus, a total of 65 violations of the standards was observed in North Carolina between June 24, 1979, the beginning of the monitoring program, and August 1, 1982. (For county distribution of violations, see Figure 2.)

A few systems had more than one analysis of the radiological content of their water performed. In these cases the higher analytical value was listed. However, in these cases all of the system values exceeded federal and state standards.

All data are displayed with the counting error in pCi/l for a 95-percent confidence interval. Because of the random nature of radiocative

Table 1

GROSS ALPHA PARTICLE ACTIVITY OF THOSE SYSTEMS
EXCEEDING FEDERAL AND STATE STANDARDS

No.	System Identification No.	Gross Alpha (pCi/l)**
1	02 91 110	25.7 ± 3.5
2	04 33 124*	34.6 ± 7.8
3	04 33 120	27.4 ± 5.4
4	04 33 108*	29.1 ± 6.6
5	03 92 261	80.7 ± 8.3
6	03 43 125*	18.0 ± 2.5
7	03 26 130	18.1 ± 2.1
8	01 18 158*	55.9 ± 4.5
9	03 26 659	19.9 ± 3.2
10	03 26 336	21.0 ± 1.6
11	04 33 105*	16.1 ± 3.7
12	03 92 171*	68.3 ± 7.4
13	03 26 216*	21.7 ± 2.1
14	04 96 117*	27.0 ± 3.1
15	03 92 175*	38.3 ± 3.7
16	04 64 045*	24.9 ± 5.6
17	03 26 255	18.3 ± 2.2
18	03 26 101	19.3 ± 2.0
19	03 26 214*	17.4 ± 2.5
20	03 92 205	16.9 ± 4.1
21	04 64 429	32.9 ± 7.5
22	02 35 025	28.8 ± 4.9
23	03 83 127*	16.8 ± 2.5
24	03 51 114*	17.6 ± 3.1
25	04 98 060*	47.3 ± 12.0
26	04 33 035	24.4 ± 12.9

* System also in violation of radium standard.
**Does not exclude uranium.

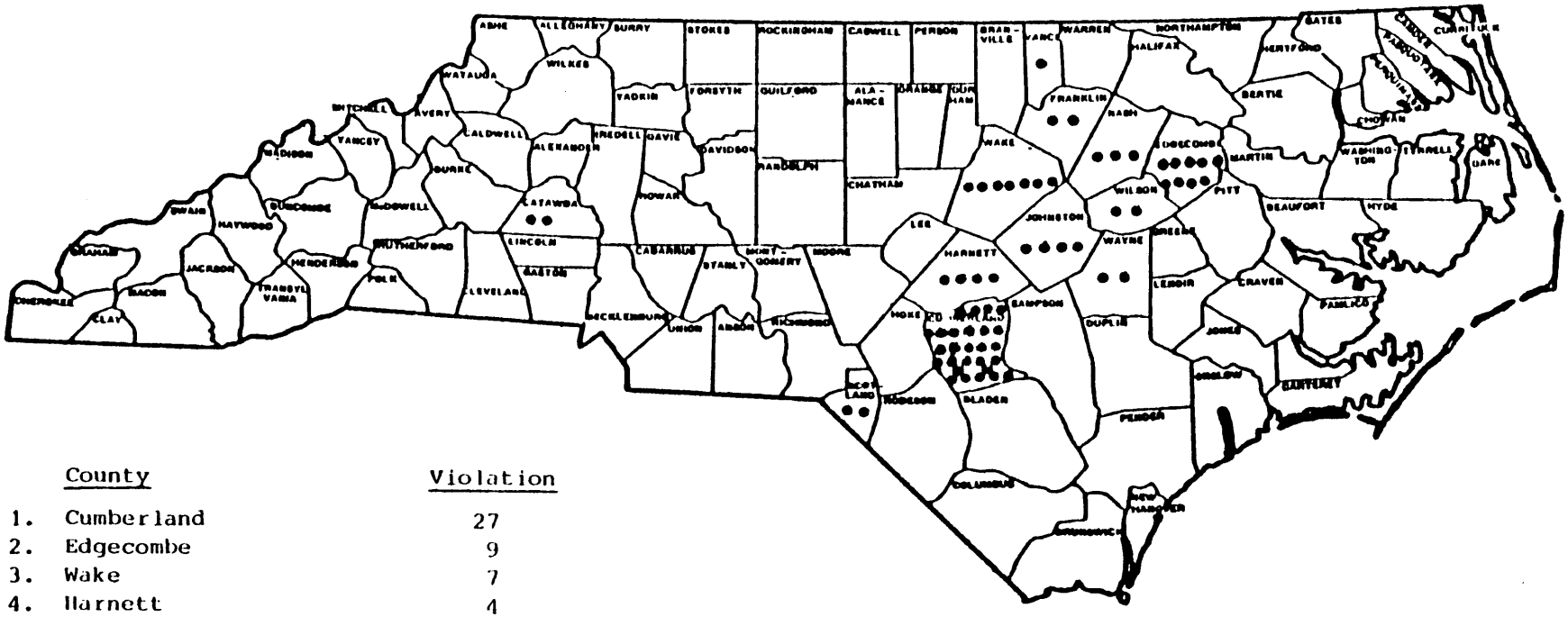
Table 2

^{226}Ra, ^{228}Ra AND COMBINED Ra OF THOSE SYSTEMS EXCEEDING FEDERAL AND STATE STANDARDS

No.	System I.D. No.	Radium-226 (pCi/liter)	Radium-228 (pCi/liter)	Combined Radium (pCi/liter)
1	03 26 194	8.9 ± 1.1	7.9 ± 4.5	16.7
2	04 33 124*	3.3 ± 0.3	5.1 ± 1.4	8.4
3	04 33 108*	4.4 ± 0.7	5.3 ± 1.5	9.7
4	03 51 422	3.3 ± 0.8	8.7 ± 0.6	12.0
5	03 43 125*	7.4 ± 1.5	8.8 ± 1.7	16.2
6	03 26 269	6.0 ± 2.8	4.7 ± 1.8	10.7
7	01 18 158*	7.5 ± 0.7	0.0 ± 1.3	7.5
8	04 33 040	8.3 ± 2.3	11.7 ± 1.6	20.0
9	03 26 313	7.6 ± 0.8	6.2 ± 2.1	13.8
10	03 26 346	7.6 ± 2.4	6.5 ± 1.3	14.1
11	03 26 183	3.9 ± 1.3	7.8 ± 0.2	11.8
12	04 33 105*	6.0 ± 0.5	0.0 ±	6.0
13	03 92 171*	8.3 ± 0.6	5.4 ± 0.3	13.7
14	03 51 149	6.4 ± 0.8	10.8 ± 0.3	17.2
15	03 26 239	3.5 ± 0.6	5.8 ± 0.6	9.4
16	03 26 200	6.4 ± 0.6	0.0 ± 1.9	6.4
17	03 26 125	7.0 ± 0.0	7.4 ± 0.4	14.3
18	03 26 160	8.7 ± 1.4	0.0 ± 2.0	8.7
19	03 26 294	12.2 ± 0.2	4.9 ± 2.9	17.1
20	03 26 162	4.7 ± 0.8	2.7 ± 1.7	7.4
21	03 26 216*	9.3 ± 1.6	2.4 ± 0.4	11.7
22	03 26 155	4.2 ± 0.3	1.9 ± 1.5	6.0
23	04 96 117*	17.2 ± 0.5	4.5 ± 1.5	21.7
24	03 26 179	4.1 ± 0.3	2.2 ± 1.7	6.3
25	03 26 147	4.1 ± 0.2	3.9 ± 0.8	8.0
26	03 92 306	5.8 ± 0.3	1.9 ± 1.7	7.7
27	03 92 175*	6.2 ± 0.7	1.2 ± 1.6	7.4
28	04 64 045*	11.8 ± 1.7	9.3 ± 0.8	21.1
29	03 26 180	6.8 ± 1.0	5.5 ± 0.3	12.4
30	03 26 102	5.1 ± 1.1	4.4 ± 1.8	9.5
31	03 26 163	4.2 ± 0.5	1.8 ± 1.3	6.0
32	03 26 250	3.0 ± 0.5	3.2 ± 1.0	6.2
33	03 26 214*	12.4 ± 1.0	9.3 ± 5.7	21.7
34	03 43 185	5.1 ± 0.5	3.4 ± 1.1	8.6
35	03 43 146	4.8 ± 0.5	1.6 ± 2.3	6.4
36	02 35 119	3.8 ± 0.6	3.5 ± 1.8	7.3
37	03 83 127*	3.8 ± 0.6	8.9 ± 1.4	12.7
38	03 51 114*	9.6 ± 1.0	4.4 ± 0.6	14.0
39	04 98 060*	15.9 ± 0.1	22.7 ± 1.3	38.6

*System also in violation of gross alpha particle activity standard.

NORTH CAROLINA



	County	Violation
1.	Cumberland	27
2.	Edgecombe	9
3.	Wake	7
4.	Harnett	4
5.	Johnston	4
6.	Nash	3
7.	Catawba	2
8.	Franklin	2
9.	Scotland	2
10.	Wayne	2
11.	Wilson	2
12.	Vance	1

Figure 2. County Distribution of Violations of Standards

disintegrations, there is an error associated with any measured count of these disintegrations. The variability of these measurements is indicated by the displayed counting error. Additional information on counting error is covered by Krieger and Whittaker's (1980) publication, "Prescribed Procedures for Measurement of Radioactivity in Drinking Water."

The average concentration of gross alpha particle activity for those systems in violation of federal and state standards was 29.5 pCi/l. The amount of whole body dose or organ dose cannot be estimated from the gross alpha particle activity, due to the lack of knowledge of specific radionuclide. The average level of radium for those systems in violation of the standard was 12.2 pCi/l which gives a human bone dose of 365 mrem/year and using the EPA linear extrapolation for dose vs. effect may cause 3.6 deaths per year from cancer per million people continuously exposed (Environmental Protection Agency, 1976b). At the present rate of occurrence of water systems which exceed the 5 pCi/l radium standard (1.9 percent of those systems tested), within the State of North Carolina using ground water (present population 3 million), one person may die from cancer every 4.8 years due to the amount of radium ingested in drinking water. This rate does not take into account other radionuclides in drinking water which the present regulations do not address such as radon and uranium.

These levels of exposure to radionuclides in drinking water will remain if nothing is done to change the concentrations of the radioactive contaminants or the source of water. When a water system is found to have levels of gross alpha particle activity or combined radium activity which exceed the standards, the system owner is responsible for notifying the North Carolina Department of Human Resources (DHR), notifying the households which consume the contaminated water by written notice; publishing in a local

newspaper a notification to the public for three consecutive days; notifying a radio and television station serving the area; submitting a plan of action for correcting the problem including a timetable for corrective action; and repeating quarterly the public notice and monitoring of the water system until two quarterly samples prove that the problem no longer exists (Environmental Protection Agency, 1976a).

Radium-228 to Radium-226 Ratios

Under present regulations the analysis of radium-228 is only performed if the concentration of radium-226 is in excess of 3 pCi/l. As Michel and Moore (1979) pointed out, the ratio of radium-228 to radium-226 ($^{228}Ra/^{226}Ra$) is not constant and can vary by as much as a factor of 3. This variability, therefore, makes it possible for a system with large amounts of radium-228 to exceed the combined radium limit of 5 pCi/l but not to be identified as such because the level of radium-226 did not exceed 3 pCi/l (Michel and Moore, 1979).

This variability is also evident in the radium data of Table 2, which has a maximum $^{228}Ra/^{226}Ra$ ratio of 2.6. Table 3 lists the $^{228}Ra/^{226}Ra$ ratios of the radium data (taken from Table 2). A statistical examination of the $^{228}Ra/^{226}Ra$ ratios yielded a mean of 0.85 and a standard deviation of 0.63, an indication of high variability. Of the 39 systems examined, 69 percent of the systems had $^{228}Ra/^{226}Ra$ less than one. In these cases the content of radium-226 was greater than the content of radium-228, which supports the present procedure. However, 31 percent of the systems had $^{228}Ra/^{226}Ra$ ratios greater than one. This demonstrates that approximately a third of these systems have radium-228 levels greater than their levels of radium-226, indicating that the present procedure may be inadequate in a third of the systems analyzed.

Table 3

$^{228}Ra/^{226}Ra$ RATIO OF THOSE SYSTEMS EXCEEDING THE RADIUM STANDARD

1.	0.89	21.	0.26
2.	1.57	22.	0.46
3.	1.20	23.	0.26
4.	2.61	24.	0.55
5.	1.19	25.	0.96
6.	0.79	26.	0.32
7.	0.0	27.	0.19
8.	1.40	28.	0.79
9.	0.82	29.	0.80
10.	0.86	30.	0.86
11.	2.00	31.	0.42
12.	0.0	32.	1.06
13.	0.65	33.	0.75
14.	1.69	34.	0.67
15.	1.64	35.	0.33
16.	0.0	36.	0.93
17.	1.06	37.	2.33
18.	0.0	38.	0.46
19.	0.40	39.	1.42
20.	0.58		

69% < 1.0

31% > 1.0

Mean = 0.85

Standard deviation = 0.63

Thus, in order to be sure that the limit of 5 pCi/l for combined radium is not exceeded, radium-228 would have to be measured regardless of the concentration of radium-226, if the gross alpha particle activity exceeds 5 pCi/l. Consideration of the cost-benefit aspects of radium-228 monitoring is needed.

Uranium Concentrations

A limited amount of data was obtained on the total uranium concentration of twenty-three water supply systems in which the gross alpha concentration exceeded 15 pCi/l (see Table 4). These data are primarily for single samples collected from single wells rather than annual composites of four quarterly samples from the system. Thus, they generally are not for analyses of the same samples from which the radium data shown in Figure 2 were obtained. The total uranium content consists of the combined concentrations of uranium-238 and uranium-234. In those samples analyzed, the average level of uranium activity was 26.7 pCi/l. No regulatory standard currently exists for the concentration of uranium in drinking water.

Uranium is chemically toxic as well as radiologically toxic. The National Academy of Sciences has recommended to the EPA that a one-day and seven-day suggested limit for uranium of 3.5 mg/liter (2300 pCi/liter) and 0.2 mg/liter (140 pCi/liter), respectively, be used based on chemical toxicity only (Lappenbusch, 1979).

It is the judgment of the Environmental Protection Agency at this time that radiotoxicity should be given priority. The level of radiotoxicity of uranium is best calculated by comparing ^{238}U and ^{234}U with ^{226}Ra, since all of these radionuclides are bone seekers. It is the consensus of EPA's program that 20 pCi/day or 10 pCi/l of uranium would result in a dose rate to the bone of 150 mrem/year (Lappenbusch, 1979). Since the present standard

Table 4

TOTAL URANIUM CONTENT (^{234}U AND ^{238}U)

No.	System Identification Number	Total Uranium
1	01 18 125	21.7 ± 3.1
2	01 80 147	8.7 ± 1.4
3	02 17 107	72.1 ± 7.9
4	02 35 025	23.4 ± 3.2
5	02 35 119-	49.4 ± 4.6
6	02 79 115	17.2 ± 3.9
7	02 86 148	21.0 ± 2.1
8	02 91 015	20.0 ± 4.2
9	03 26 101	0.0
10	03 26 130	0.0
11	03 26 158	15.1 ± 1.8
12	03 26 214*-	7.7
13	03 26 659	0.3 ± 0.4
14	03 51 114-	0.4 ± 0.6
15	03 83 127-	0.1 ± 0.6
16	03 92 171	277.4 ± 10.7
17	03 92 205	0.4 ± 0.4
18	04 33 108-	3.2 ± 1.3
19	04 64 045*-	8.8
20	04 64 108	38.6 ± 2.6
21	04 64 429	10.7 ± 1.5
22	04 96 117*-	5.2
23	04 33 035	12.1 ± 0.3

*Analysis performed at the University of South Carolina Department of Geology Laboraotries.

-Systems also having a level of radium exceeding federal and state standards.

for radium limits the bone dose rate to 150 mrem/year, uranium's effect on the same organ system should not exceed this level. This opinion is also shared by the EPA (Lappenbusch, 1979).

The highest data value obtained was 277 pCi/l of total uranium or 28 times the 10 pCi/l limit discussed. This concentration of uranium would yield a bone dose of 4200 mrem per year. This high level of uranium demonstrates the vulnerability of present standards. Since uranium is omitted from being one of the constituents of gross alpha activity, drinking water could be legally supplied containing 277 pCi/l of uranium provided it does not exceed standards for other radionuclides. The average level of uranium was 26.7 pCi/l and would produce a bone dose of 391 mrem per year.

From the data obtained for systems exceeding standards or suggested standards, the average and highest concentrations of gross alpha particle activity, combined radium and total uranium, are listed in Table 5. These data give an indication of the need for consideration of a uranium standard. The small sample size tested for uranium prevented the extrapolation of the percent of systems in North Carolina having uranium concentrations in excess of 10 pCi/l.

An examination was made of the correlation existing between concentrations of radium-226 and total uranium. Total uranium is made up of uranium-238 and uranium-234, which are both in the uranium series, the same series as radium-226. Uranium data were available for eight water systems that exceeded the radium standard. These measurements were plotted, and a linear regression analysis was performed (see Table 6 and Figure 3). No correlation was found between concentration of radium-226 and total uranium.

Treatment of Water for Removal of Radionuclides

From the data listed in Table 2 we can ascertain that significant levels of radium exist in ground water. Violations for exceeding the radium limit

Table 5

LISTING* OF THE AVERAGE AND HIGHEST CONCENTRATIONS OF GROSS ALPHA PARTICLE ACTIVITY, COMBINED RADIUM AND TOTAL URANIUM AND THEIR STANDARDS (EXISTING AND DISCUSSED)

	Gross Alpha	Radium	Uranium
Limit (pCi/l)	15	5	10**
Average (pCi/l)	29.5	12.2	26.7
Factor times limit	2.0	2.4	2.7
High (pCi/l)	80.7	38.6	277
Factor times limit	5.4	7.7	27.7

*For systems exceeding standards.

**Equivalent to radium limit.

Table 6

LISTING OF RADIUM-226 AND TOTAL URANIUM

No.	System I. D. No.	Radium-226	Total Uranium
1	02 35 119	3.8 ± 0.6	49.4 ± 4.6
2	03 26 214	12.4 ± 1.0	7.7
3	03 51 114	9.6 ± 1.0	0.4 ± 0.6
4	03 83 127	3.8 ± 0.6	0.1 ± 0.6
5	04 33 108	4.4 ± 0.7	3.2 ± 1.3
6	04 64 045	11.8 ± 1.7	8.8
7	04 96 117	17.2 ± 0.5	5.2
8	04 33 035	2.7 ± 0.2	12.1 ± 0.3

Average Ra = 8.21
Average Uranium = 10.86
Y intercept = 18.63
Slope = - 0.95
Correlation Coefficient = - 0.37

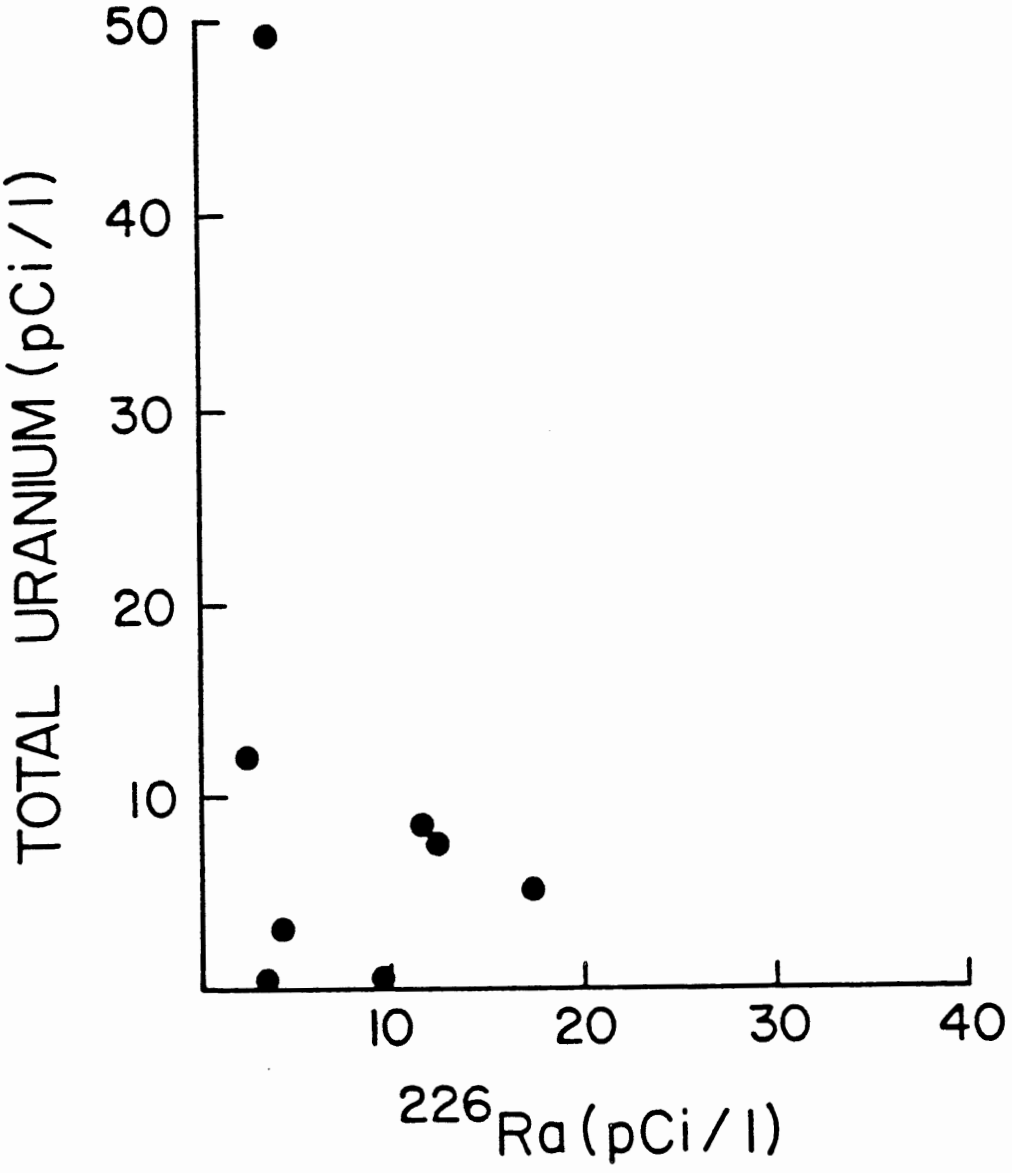


Figure 3. Correlation Between ^{226}Ra and Total Uranium

have been issued, and a few North Carolina water systems have found alternate sources of water. However, successful treatment of water containing high levels of radium has not been achieved. The Environmental Protection Agency (1977) has recommended radium removal techniques of lime or lime-soda softening (precipitative softening), ion exchange (cation exchange) and reverse osmosis. Extensive examples of radium removal for water systems have been published (Environmental Protection Agency, 1977; Brinck, et al., 1976; Singley, et al., 1977; Schliekelman, 1976; Bennett, et al., 1976).

Soluble radium, a divalent alkaline earth metal cation similar to calcium and magnesium, is removed along with other forms of hardness by a precipitative softening process (see Figure 4, Total Hardness Removal as a Function of Raw Water Radium Content). The lime-soda process is a precipitative softening process which uses the addition of lime (CaO-quicklime, or $Ca(OH)_2$ slated or hydrated lime) to convert the sodium bicarbonates of calcium and magnesium into insoluble calcium carbonate and magnesium hydroxide. In addition to this carbonate hardness, calcium and magnesium associated with the sulfate, chloride or other ions of non-carbonate hardness are removed by the addition of both lime and soda ash (Na_2CO_3) which provides the carbonate ion necessary for formation of calcium carbonate. Precipitative softening is usually employed to remove hardness and other impurities from surface waters. The chemicals added in the rapid-mix chamber are allowed to form a floc in the flocculation chamber and finally settle out in the settling chamber (see Figure 5). Plant efficiency of hardness removal will vary. In a study performed by the Environmental Protection Agency, radium removal efficiency averaged 80 percent (Singley, et al., 1977).

Precipitative softening in North Carolina is normally used in water systems with a large water demand utilizing surface water for its source.

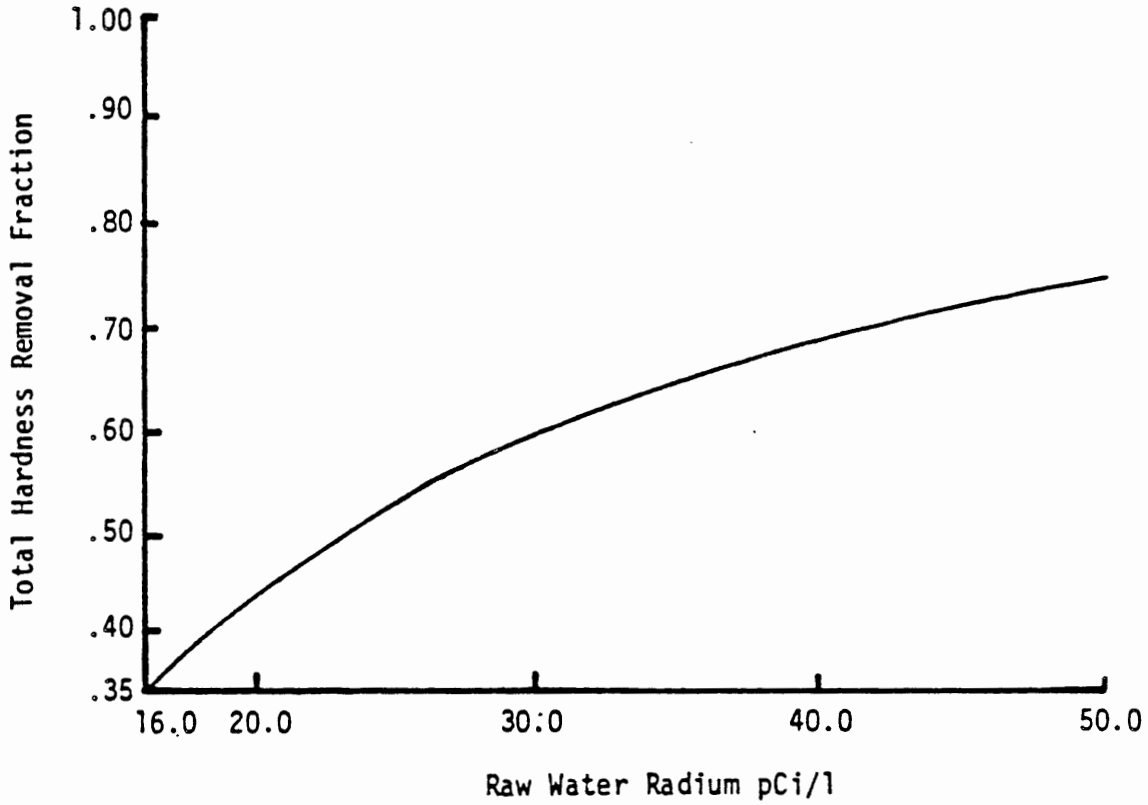


Figure 4. Total Hardness Removal as a Function of Raw Water Radium Content

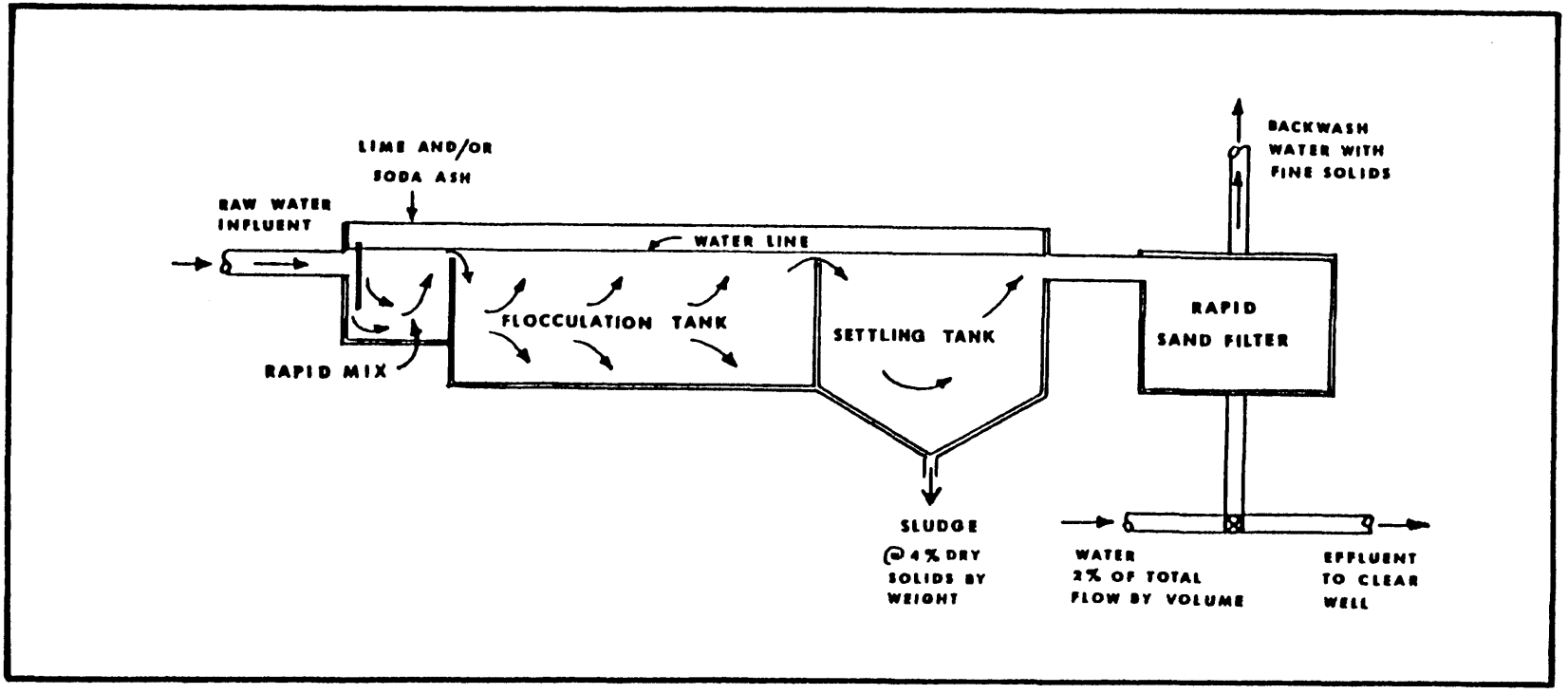


Figure 5. Diagram of Typical Lime-Soda Treatment Plant

Source: Singley, et al., 1977.

The level of radium in surface waters is normally low, unless uranium or phosphate mining activities are discharging effluents to a water course. No such occurrence upstream to a public water supply has been documented to exist in North Carolina. However, if precipitative softening is used for the removal of radium, the disposal of filter backwash water poses no particular problem since the activity levels in the waste stream have been found to be only slightly higher than levels found in the raw water. The majority of the radium activity removed appears to remain in the settling chamber waste sludge. The activity in the dry sludge approaches a level of 10^5 pCi/Kg, and special considerations must be given to safe disposal of the contaminated sludge (Singley, et al., 1977), such as shipment to a low-level radioactive waste burial ground.

Water softening by the ion-exchange process depends upon the ability of certain insoluble substances to exchange cations with other cations dissolved in water. When hard water is passed through a sodium cation exchanger, the calcium and magnesium in the hard water are replaced by sodium from the exchanger. After all of the readily replaceable sodium has been exhausted for calcium and magnesium from the hard water, the exhausted cation exchanger can be regenerated with a solution of sodium chloride. In the regeneration process, the calcium and magnesium of the exhausted cation exchanger are replaced with a fresh supply of sodium from the regenerating brine solution. The regenerated exchanger is then washed with treated water to free any excess salt and is ready to be used again to soften raw water.

Many materials with cation exchange properties such as polystyrene-type resins and mineral greensands are used for exchange media. Small water systems can utilize this means of treatment which can remove radium up to a 95-percent efficiency level, as well as hardness and trace amounts of heavy metals. Perhaps the greatest advantage of cation exchange is the completely

automatic operation of the treatment system (see Figure 6), making this form of treatment ideal for use by an untrained operator.

The repeated success of radium removal demonstrated by Singley, et al., (1977) by means of cation exchange makes this an attractive technique. Approximately 9 percent of the radium activity remains in the exchange medium; the remainder appears in the regenerant brine effluent at levels approaching 1,000 pCi/l. Disposal methods for spent brine may more often be limited by considerations of salinity rather than radium activity levels (Singley, et al., 1977). However, in North Carolina there are no examples of successful radium removal.

Manganese oxide suspensions have been shown to absorb large quantities of various aqueous metals (Murray, et al., 1968). Radium extraction studies (Moore, 1975 and 1982), performed by Moore on well water having elevated levels of radium (^{226}Ra) measured at approximately 115 pCi/l resulted in removal efficiency rates of 95 to 87 percent using 40 grams of Mn fibers. Acrylic fiber impregnated with 12 to 15 percent manganese acted as a cation adsorption filter in the removal of radium cationic radionuclide (Moore, 1982). In this study Moore estimated filter preparation cost for one kilogram of manganese filter media to be fifteen dollars. This amount of filter media could treat 10,000 liters of water while removing 100 pCi/l of radium (Moore, 1975) or 2.5 million liters of water removing 12 pCi/l, the average level of radium found in those systems exceeding the standard in North Carolina. A one-kilogram filter would be capable of providing adequate treatment for an average family for a number of years at a cost of $10 per family per year and, therefore, is a potential cost-effective means of radium removal for public water systems. This is, however, a new technology. Although promising, it is not yet fully understood nor has it been thoroughly

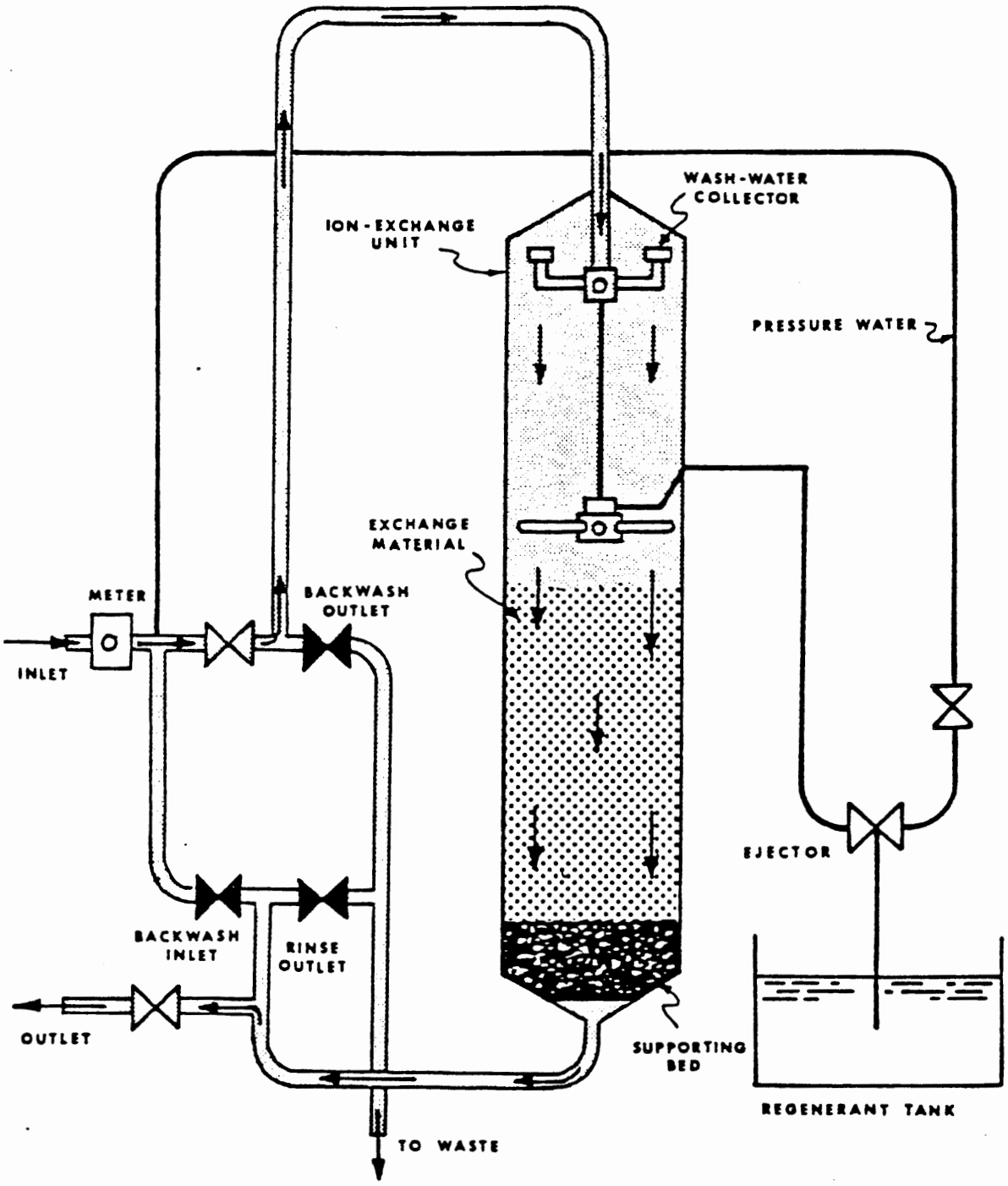


Figure 6. Diagram of Typical Ion Exchange Unit

and consistently documented to remove radium from public water systems to a level below the Safe Drinking Water Act Standard (Environmental Protection Agency, 1976a). Until further study has proved this method successful and a source of prepared filters is available to the public, radium removal with manganese filters is not a viable form of treatment.

The lack of a drinking water standard for uranium has been directly attributed to the lack of documented instances in which it was successfully removed (personal communication, C. Richard Cothern, 1982). The valence states of uranium (^{238}U, ^{235}U, ^{234}U) are +2, +3, +4 and +6 and are most commonly found in the hexavalent and tetravalent states (Cothern and Lappenbusch, 1983). It has a tendency to form uranyl di- and tri-carbonate anions which are both soluble and mobile. In a hydrologically reducing condition the uranium would not be mobile and, thus, would not be a problem until decaying to ^{234}Th or ^{230}Th (Osmond and Cowart, 1976).

Under laboratory conditions anion exchange has been successfully employed as the preferred means of separation of uranium from water and other radionuclides (Osmond and Cowart, 1976). There is no reason to believe that anion exchange would not also work for large-scale drinking water supplies, as it has for the removal of other anions in drinking water supplies, and radionuclides in nuclear reactor coolant process waters (Stranb, 1964).

The Environmental Protection Agency is collecting data on uranium removal efficiency rates (personal communication, C. Richard Cothern, 1982), but at the present time no publications of results are available. In addition, due to the nature of anion exchange and the strong acid resin rejuvenation requirements involved, it is not expected to be a practical alternative for small water system treatment or use by untrained operators. Until the creation of a uranium standard and adequate monitoring to identify those

water systems with high levels of uranium, there will be no need to create innovative methods of uranium removal.

Studies by Aldrich, et al., (1975) and Sasser and Watson (1977) have shown that high levels of radon-222 in ground-water supplies are not uncommon in North Carolina. Radon-222, a fairly insoluble gas, can be easily removed by aeration. The radioactive gas can then be vented to the atmosphere. However, no standard exists to limit the concentration of radon-222 in drinking water; and therefore, little is expected to be done for the removal of radon in North Carolina.

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