Limitations of Water Treatment Methods for Removing Radioactive Contaminants

By CONRAD P. STRAUB, Ph.D.

RADIOACTIVITY has been known since 1896, when Becquerel first observed the phenomenon by its effect on photographic plate. However, until the first controlled nuclear chain reaction was demonstrated, the supply of radioactive materials was limited, and the opportunities for large groups of the population to be exposed to significant amounts of radiation were small. Since 1942, many kinds and considerable quantities of radioisotopes, many of high specific activity, have been produced in nuclear reactors.

Increased use of radioisotopes in recent years has resulted in the discharge of these materials to the environment. A 1954 report, based on a questionnaire sent out in December 1952 to 1,027 users of radioisotopes, showed that about 41 percent of them disposed of radioactive wastes by dilution and discharge to the sewers (*1*). In all probability, the amounts of radioactive materials used will increase many fold over the amounts employed now, as additional applications are developed. An increase in the amounts used will, in turn, result in the release of a greater variety of radioisotopes, as well as greater quantities of radioactive materials. At present, considerable effort is being expended in developing nuclear energy as a source of power. These developments will aggravate the waste disposal problem and will add emphasis to the statement made in 1951 by Mark D. Hollis, chief engineer officer of the Public Health Service, that public health officials will be responsible for protecting the public against the harmful effects of ionizing radiations (*2*).

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Maximum Permissible Concentrations

Ionizing radiations, which result from the disintegration of unstable nuclei, are damaging to living tissues. The extent of damage from internal exposure to radiation depends on many factors, some of which are the quantity of radioactive material deposited in the body; the point of localization; the type of radiation (alpha, beta, or gamma); chemical state of the isotopes deposited in the body; energy of the radiation; and the physical and biological half-lives of the isotopes. These factors and others are considered in establishing the maximum permissible amounts of radioisotopes in the human body and the maximum permissible concentrations in air and water. Maximum permissible amounts in the body and maximum permissible concentrations in the environment are summarized and reported in a handbook published by the National Bureau of Standards (*3*).

The maximum permissible concentration

(MPC) values serve as a guide to the public health official in determining the levels of radioactive materials that may be discharged from various installations and the amounts of these materials that may be permitted in the air we breathe and the water we drink. They represent the best available information on permissible concentrations of radioactive materials in the environment. It should be noted that these values have been derived on the assumption that the contaminated supply is the sole source of supply during the lifetime of an individual (taken as 70 years). Further, it is assumed that these values are superimposed on the natural background radiation of the particular area under study.

Comparison With Other Contaminants

The Drinking Water Standards promulgated by the Public Health Service define limits for specific materials in waters used for drinking on interstate carriers. In table 1 are shown some of these limits and the maximum permissible concentrations in water of similar radioisotopes. It can be seen that the concentrations permitted on the basis of radioactivity are many orders of magnitude lower than safe or acceptable concentrations of the stable isotopes of the same element. These extremely low concentrations present a problem of identification, since ordinary qualitative and quantitative procedures are of little value in detecting and identifying the presence of such minute concentrations of specific radioisotopes. However, because these materials are radioactive, it is possible to use this characteristic to detect, measure, and identify them. The radiochemical procedures used to identify specific radioisotopes, however, become less accurate as the number of different radioactive constitutents present increases.

A characteristic of radioactivity that should be mentioned, in order fully to understand the difficulties in handling these materials and removing them from waters to make them safe for normal use, is that the rate of disintegration cannot be altered by any physical, chemical, or biological process. Other polluting substances can be removed or modified so that they are no longer noxious: For example, chromium or other heavy metals may be eliminated by precipitation; cyanides may be treated with chlorine to form the relatively nontoxic cyanate, which may decompose further to ammonium carbonate; or pathogenic bacteria may be de-

Table 1. Quantitative comparison of limits set by Public Health Service Drinking Water Standards and maximum permissible concentrations of radioisotopes in water

Chemical constituent	Drinking water standards		Maximum permissible concentration		Radioactive form
	p.p.m. (mg./l.)	μg./cc.	μg./cc.	μc./cc.	
Grounds for rejection					
Lead (Pb)	0. 1	1×10^{-1}	---	---	---
Fluoride (F)	1. 5	1.5×10^{0}	---	---	---
Arsenic (As)	. 05	5×10^{-2}	---	---	---
Selenium (Se)	. 05	5×10^{-2}	---	---	---
Chromium (Cr^{+6})	. 05	5×10^{-2}	5.2×10^{-6}	0. 5	Cr^{51}
Recommended values					
Copper (Cu)	3. 0	3×10^{0}	2.1×10^{-8}	8×10^{-2}	Cu^{64}
Iron and manganese (Fe+Mn)	. 3	3×10^{-1}	1.8×10^{-6}	4×10^{-3}	Fe^{55}
			2.1×10^{-9}	1×10^{-4}	Fe^{59}
			9.3×10^{-9}	0. 2	Mn^{56}
Magnesium (Mg)	125	1.25×10^{2}	---	---	---
Zinc (Zn)	15	1.5×10^{1}	---	---	---
Chloride (Cl)	250	2.5×10^{2}	8.9×10^{-2}	2×10^{-3}	Cl^{36}
Sulfate (SO_4)	250	0.83×10^{2} (as sulfur)	1.2×10^{-7}	5×10^{-3}	S^{35}
Phenolic compounds (C_6H_5OH) [1]	. 001	[1] 1.5×10^{-4}	6.7×10^{-4}	3×10^{-3}	C^{14}

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[1] Assuming that only one of the carbon atoms is radioactive carbon-14.

stroyed by disinfection, pasteurization, or sterilization. All that can be done with radioactive wastes is to transfer the radioactive component from one phase to another: The soluble materials, for example, may be removed by precipitation, adsorption, and the like. The radioactive fraction remains for disposal. Thus far, the only real solution, and it is a tentative one, is storage, and this is the method now used to dispose of high-level wastes. The less hazardous and low-level wastes may be discharged into surface waters or into the ground. When discharged into the ground, the wastes may come in contact with the ground water and move slowly with it, and some of the radioactive materials may be removed by adsorption on the soils through which the waste stream and ground water flow.

Basic Principles

The efficiency of conventional water treatment processes for the removal of radioactive contaminants must be geared to the MPC values. Since these values define the accepted permissible levels of contamination, they may be used to calculate the degree of removal that must be provided to meet these levels when the nature and the amount of activity initially present are known. For example, if a water supply contains 10μc./ml. of promethium-147 and the MPC value for this substance is 1μc./ml., treatment which will result in removal of 90 percent of the promethium must be provided. Or, if the efficiency of a particular process and the MPC value are given, the maximum concentration of that component in the waters used for public water supplies can be stipulated. For strontium-89, for example, with an MPC value of 7 x 10^{-5} μc./ml. and a process that results in 90 percent removal of this substance, the highest concentration that may be present in the waters to be treated is 7 x 10^{-4} μc./ml. The difference in the initial concentrations of promethium-147 and strontium-89 varies by a factor of approximately 14,000 and is due entirely to the difference in their MPC values.

Conventional water treatment processes include coagulation, settling, filtration, and disinfection. In addition, auxiliary treatment, such as aeration, softening, and iron and manganese removal, may be used. Any discussion of treatment methods must take into account the fact that radioactive and stable isotopes of an element behave identically during chemical treatment. Thus, if the conventional process is not designed to remove a specific element, there is no reason to believe that the process will remove the specific radioisotope. Radioactive calcium, barium, or strontium, for example, will be significantly removed by softening, since the purpose of softening is to remove calcium. Barium and strontium will be removed with calcium, because of their similar chemical properties.

Table 2. Removal of radioisotopes by coagulation and settling: results obtained in jar-test studies

Radioisotope	Clay added [1] (p.p.m.)	Coagulant added [2] (g.p.g.)	Final pH	Percent removal
Cesium-137—barium-137	0	1	--------	0–6
	100	1	--------	35–65
Strontium-89	0	[3] 1.5	6.7–7.8	0–6
	100	0.5–6	6.7–10.7	0–51
Cadmium-115	0	1	--------	40–60
	100	1–5	--------	60–95
Barium-140—lanthanum-140	100	1–6	7.5–8.2	28–84
Scandium-46	100	1–6	6.5–8.2	66–98
Yttrium-91	0	[3] 1.5	6.8–7.1	83–93
	100	1–6	7.0–10.2	34–99
Zirconium-95—niobium-95	0	1–5	--------	70–98
	100	1	--------	95–99
Phosphorus-32	100	[3] 0.5–1.5	6.8–8.8	97–99
Chromium-51	100	1–6	7.6–8.8	73–98
Tungsten-185	100	1–6	7.5–8.4	5–91
Iodine-131	100	[3] 0.5–2	6.9–9.0	0–10
Cerium-144	0	[3] 1–1.5	7.2–7.8	81–94
	100	[3] 0.5–2.5	7.0–7.8	85–96
FPM-1 [4]	100	[3] 1–3	7.2–8.8	61–84
FPM-2 [5]	0	1–5	4.3–10.2	9–71
	100	1–5	4.3–10.2	12–73
FPM-3 [6]	0	10	9.9–10.0	46

[1] Local clay was added.

[2] Coagulant includes alum, ferrous sulfate, or ferric chloride; lime, soda ash, or sodium hydroxide; and sodium silicate.

[3] No sodium silicate added. Where added sodium silicate equals 40 percent primary coagulant dose.

[4] Iodine dissolver solution.

[5] Synthetic mixture containing fission products in the same concentrations assumed to be present 30 days after an underwater bomb blast.

[6] 3-year-old fission product mixture.

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

To show the limitations of conventional water treatment methods for the removal of radio-

Table 3. Removal of radioisotopes by sand filtration: results obtained in laboratory tests

Radioisotope	Initial activity (c./m./ml.)	Volume passed (ml.)	pH of effluent	Percent removal	
				Range	Average
Cesium-137—barium-137	800	500	8. 3	10–70	50
Strontium-89	2, 700	750	8. 3	1–13	4
Cadmium-115	1, 200	500	8. 1	60–99	95
Barium-140—lanthanum-140	1, 300	750	7. 6	39–99	74
Scandium-46	1, 500	750	8. 3	94–99	96
Yttrium-91	5, 700	750	7. 0	84–89	87
Zirconium-95—niobium-95	3, 400	500	7. 8	91–96	93
Tungsten-185	2, 200	750	7. 1	3–18	8

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

active materials, the results of experimental work at Oak Ridge National Laboratory, which has been carried out in cooperation with the Public Health Service and the Engineer Research and Development Laboratories at Fort Belvoir, Va., and of studies reported in the literature are cited.

Coagulation

In coagulation, certain soluble constituents are precipitated as insoluble hydroxides or are carried along with the heavy metal hydroxides that are formed when alum or iron salts are used. With the exception of most cations of valence 3, 4, or 5, including the rare-earths group, coagulation has not been very effective for the removal of radioactive materials from water (*4*). Phosphorus-32 has been removed from waters contaminated with this radioisotope (*4*, *5*), but considerable care had to be given to the coagulation step (*5*). Removals in excess of 98 percent have been reported. Iodine-131, which is in the form of an anion, cannot be removed by coagulation with alum or iron. However, the addition of small amounts of activated carbon (*4*), copper sulfate (*4*), or silver nitrate (*4*, *6*) has been found to increase the removal of this anion to about 96 percent. Other data are reported in table 2.

Coagulation is also helpful in the removal of radioactive material attached to or adsorbed on the natural turbidity found in many surface waters. Since coagulation will remove much of the turbidity, the activity attached to it will also be removed.

Sand Filtration

Except for removal by simple straining or sorption by the biological life contained in the *Schmutzdecke* sand filters, as shown in table 3, have not been effective for the removal of radioactive materials (*7–9*). Their major function is to remove the radioactivity previously incorporated in floc particles.

Softening

Softening with lime and soda ash has been studied (*9–12*) and, under proper conditions, effective removals of radioactive strontium, barium, cadmium, yttrium, scandium, and zirconium-niobium have been obtained (*9*). As shown in table 4, removals of 95 to 99 percent or better have been accomplished. For the most satisfactory removal of strontium, excesses of both lime and soda ash are required (*10*). Some recent studies have indicated that the strontium is removed by coprecipitation (*10*, *11*) with the calcium carbonate by the mechanism of mixed crystal formation (*11*). Data on the removal of stable strontium from municipal water supplies (*13*) are given in table 5.

Ion Exchange

Another method of softening that has been practiced for some time in municipal and industrial water treatment is ion exchange. Removal of radioactive materials by this process is technically possible (*12*, *14*, *15*). For most effective removal, mixed bed or cation and anion exchange beds in series are recommended. Results obtained with various types of synthetic exchange materials and with greensand (*9*), a material which has found considerable use in municipal softening installations, are shown in table 6. For adequate removals and economic operation, the waters to be treated should be low in dissolved solids. Experience in the laboratory has shown that certain radioactive mate-

Table 4. Approximate minimum combined dosage [1] of lime and soda ash to give stated percentage removal of radioisotopes

Radioisotope	50 percent removal		75 percent removal		90 percent removal		95 percent removal		99 percent removal	
	Lime (g.p.g.)	Soda (g.p.g.)	Lime (g.p.g.)	Soda (g.p.g.)	Lime (g.p.g.)	Soda (g.p.g.)	Lime (g.p.g.)	Soda (g.p.g.)	Lime (g.p.g.)	Soda (g.p.g.)
Barium-140—lanthanum-140	2	2	4	2	6	4	------	------	------	------
Strontium-89	4	3	5	5	7	9	20	20	------	------
Cadmium-115	2	2	3	3	4	4	4	4	6	4
Scandium-46	3	3	3	3	5	5	------	------	------	------
Yttrium-91	2	2	4	4	6	6	12	6	------	------
Zirconium-95—niobium-95	2	0	5	0	12	0	17	0	22	0
Cesium-137	48	48	------	------	------	------	------	------	------	------
Tungsten-185	48	48	------	------	------	------	------	------	------	------

[1] Minimum combined dosage is defined such that, of those variable dosages studied, the grains per gallon of lime plus the grains per gallon of soda ash is a minimum.

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

rials will continue to be removed (about 50-percent removal) even though the column has been saturated with calcium and the hardness is no longer reduced.

Modified Processes

Several modifications of existing water treatment processes have been studied in an effort to obtain better removals of specific radioactive materials. The most promising of these has been phosphate coagulation (*16*, *17*), as indicated by the data in table 7. For many of the radioisotopes of interest, including strontium-89, relatively high percentage removals have been obtained, but for strontium-89 the

Table 5. Removal of stable strontium by chemical treatment

Type of treatment	Percent removal		Number of cities
	Average	Range	
Alum or ferrous sulfate	12	10–31	7
Alum or ferrous sulfate, plus lime	37	10–75	11
Alum or ferrous sulfate, plus lime and soda ash	54	10–85	3
Alum or ferrous sulfate, plus lime and phosphate	42	10–70	5
Softening only (phosphate, ion exchanger)	73	69–76	2
None (except chlorine, fluoride, carbon, or ammonia)	10	------	8

Table 6. Percentage removal of individual radioisotopes with ion exchange materials

Radioisotope	Cation exchange	Anion exchange	Mixed bed	Greensand
Tungsten-185	12 –16	97. 2–99. 2	98. 9	9
Yttrium-91	86 –93. 1	94. 2–98. 5	97. 6 –98. 7	75
Scandium-46	95. 7–97. 2	98. 8–99. 0	98. 5 –98. 7	96. 4
Strontium-89	99. 1–99. 8	5 – 7	99. 95–99. 97	99. 8
Barium-140—lanthanum-140	98. 3–99. 0	36 –42	99. 5 –99. 6	96. 3
Cesium-137	99. 8	9	99. 8	------
Cadmium-115	98. 5	0	99. 2	------
Zirconium-95—niobium-95	58 –75	96. 4–99. 9	90. 9 –99. 4	------

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

Table 7. Removal of radioisotopes by phosphate coagulation

Radioisotope	Coagulant	Coagulant dose (mg./l.)	Percent removal
Cerium-144	KH_2PO_4	200	99. 8
	Na_3PO_4	120	99. 9
Strontium-89	KH_2PO_4	100	81. 3
	Na_3PO_4	240	97. 8
Yttrium-91	KH_2PO_4	100	99. 9
Antimony-124	KH_2PO_4	100	66. 1
	Na_3PO_4	120	67. 4
Zinc-65	KH_2PO_4	50	99. 6
Tungsten-185	KH_2PO_4	200	10. 7
Zirconium-95	KH_2PO_4	100	99. 5
Niobium-95	KH_2PO_4	100	99. 2

results were obtained under carefully controlled conditions of pH and the calcium-hydroxide/trisodium-phosphate ratio. Other methods that have been suggested for the removal of radioactive materials from water include the use of metal powders (*18*) and added clay (*4*, *19*). Results obtained with these materials are given in tables 8 and 9.

Evaporation

Although not a municipal treatment method, evaporation is included in this discussion because it is the most effective process available at present for producing waters low in radioactivity. High decontamination factors are possible, but the cost is great. However, under emergency conditions, evaporation should prove to be very satisfactory.

Removal of Mixed Fission Products

The discussion thus far has centered on the removal of specific radioisotopes from waste solutions. For a fission products mixture or any mixture of radioisotopes, the efficiency of removal is a function of the individual radioisotopes comprising the mixture. To date, removals of mixtures in the order of about 70 to 80 percent have been obtained with alum or iron coagulation (*4*). A series of studies on actual process waste waters at Oak Ridge National Laboratory disclosed that up to 90 percent of the contaminants could be removed with excess lime-soda ash softening or with phosphate coagulation when coupled with the addition of clay, which is specific for the removal of the cesium in the waste. Experimental results in removing fission products mixtures are summarized in table 10.

Table 8. Removal of radioactive contaminants by metallic dusts

[1,000 p.p.m. metal dust, 90 minute contact, filtered sample counted]

Contaminant	Stock solution pH	Initial activity (c./m./ml.)	Percent removal			
			Iron [1]	Zinc	Copper	Aluminum
Ruthenium-106—rhodium-106	8. 0–8. 2	1, 975– 3, 950	99. 6	98. 4	93. 7	92. 8
Yttrium-91	7. 9–8. 0	1, 610– 3, 230	98. 0	98. 0	--------	--------
Zirconium-95—niobium-95	7. 2–8. 3	1, 210– 4, 470	99. 1	97. 9	99. 1	99. 8
Phosphorus-32	7. 8–8. 4	128–12, 000	99. 8	98. 1	--------	84. 2
Iodine-131	6. 5–8. 2	4, 288– 6, 689	37. 2	45. 7	42. 2	23. 2
Cerium-141, -144—praseodymium-144	7. 9–8. 1	3, 915– 4, 491	99. 9	99. 9	99. 5	99. 8
Barium-140—lanthanum-140	7. 9–8. 2	4, 644–10, 205	94. 8	74. 5	65. 7	73. 8
Cobalt-60	8. 0–8. 2	1, 571– 4, 792	[2] [3] 92. 2	--------	47. 8	[3] 30. 2
FPM-6 [4]	7. 2–8. 0	4, 730–10, 375	85. 8	76. 6	92. 1	89. 4
FPM-5 [5]	2. 0–7. 2	2, 890– 4, 220	55. 5	39. 6	49. 3	8. 2
Cesium-137—barium-137	8. 0	13, 085	8. 6	--------	--------	--------

[1] Iron samples were centrifuged before counting.

[2] 500 p.p.m. metal dust.

[3] 60-minute contact.

[4] Iodine dissolver solution aged 20 days.

[5] Mixed fission product waste containing mainly cesium-137—barium-137, and ruthenium-106—rhodium-106.

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

Table 9. Effect of added clay on removal of radioisotopes[1]

Radioisotope	Percent removal		
	100 p.p.m.	750 p.p.m.	5,000 p.p.m.
Barium-140—lanthanum-140	41	58	85
Cadmium-115	3	30	64
Cerium-144	70–80	86	--------
Cesium-137	38	87	98. 0
Scandium-46	53	91. 7	96. 9
Strontium-89	2–12	14–22	49–52
Tungsten-185	0	4	49
Yttrium-91	22–45	56–70	93. 6
Zirconium-95—niobium-95	93. 5	99. 0	98. 0

[1] Local clay added to produce turbidity.

NOTE: In pairs of radioisotopes, the first is the parent element, and the second, the daughter. Both are radioactive.

Table 10. Removal of mixed fission products

Mixture	Treatment process	Percent removal
FPM-1[1]	Coagulation and settling plus clay.	61–84
FPM-2[2]	Coagulation and settling	9–71
	Coagulation and settling plus clay.	12–73
	do	46
	Coagulation and settling plus filtration plus clay.	70–73
FPM-3[3]	Coagulation and settling	46
FPM-4[4]	do	89
	Mixed cation-anion exchange slurry.	83–99
FPM-5[5]	Coagulation and settling	51–59
	Mixed cation-anion exchange slurry.	38
	Metallic dust slurry	8–56
FPM-6[6]	do	77–92

[1] Iodine dissolver solution.

[2] Synthetic mixture.

[3] 3-year-old fission products mixture.

[4] Iodine dissolver solution aged 30 days.

[5] Mixed fission product waste containing mainly cesium-137—barium-137 and ruthenium-106—rhodium-106.

[6] Iodine dissolver solution aged 20 days.

Summary and Conclusion

Laboratory studies have shown that conventional water treatment processes may remove varying percentages of radioactive contaminants from water. However, the percentage removal values have little meaning unless they are related to the initial concentration of the radioactive materials present and to the maximum permissible concentration value prescribed for the individual radioisotope. Generally speaking, unless the initial concentration is very low, certainly several orders of magnitude below the microcurie-per-milliliter range, water treatment processes will be ineffective for reducing radioactive contaminants to acceptable safe limits.

Radioactive materials should be retained as close to their source of production as possible. At present, the practice is to store the materials in tanks. The transfer of radioactivity from the liquid to the solid phase does not eliminate the problem, since suitable storage facilities still must be provided for the radioactive sludges, but it does reduce considerably the area of potential contamination.

To protect large centers of population that depend on sources of water which may become contaminated with radioactive materials, plans should be made now for supplying water from noncontaminated sources. For this purpose, existing ground water supplies may be used; other interconnected water supplies may be piped in; or special provisions should be made to bring in water from outside the area. These precautionary measures are applicable, however, only when radioactive contamination is not great enough to constitute an external hazard to man.

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PHS exhibit

A new exhibit, Social Workers Help Heart Patients, prepared by the Public Health Service in cooperation with the American Heart Association, is now available.

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