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Radiochemistry and Removal Characteristics of Radium Isotopes in Iowa Well Waters

R. L. Morris[1] and J. W. Klinsky[1]

Abstract. Significant concentrations of radium –226, a naturally occuring isotope which has no relationship to fall-out, have been detected in a number of deep wells in Iowa. Wells that have significant radium concentrations are located in smaller communities mostly in southeastern part of the State. Several of the towns have municipal zeolite softening processes which remove radium down to low levels. In some cases where the raw well water shows relatively high concentrations, zeolite softened water distributed to the public has had approximately 90 percent of the radium removed. Wells tested so far show that the average radium values are in most instances within the range specified by the Federal Radiation Council.

Deep well waters from the Jordan aquifer in Southeastern Iowa yield levels of radium 226 often exceeding the present radiation protection guide recommended by the Federal Radiation Council in their Staff Report #2. The State Hygienic Laboratory in cooperation with the Iowa Geological Survey and Argonne National Laboratories has been investigating this situation in an attempt to delineate the various geologic, radiochemical and epidemiological factors involved.

Radium 226 and isotopes Ra^{228} and Ra^{224} have been existing in our Jordan aquifer for millions of years and have no relation

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whatsoever to present man-made fallout. They are isotopes with long half-lives both physically and biologically and are considered to be bone seekers metabolically. Radium 226 with a physical half-life of approximately 1600 years is produced by the following radiochemical degradation known as the Uranium series:

$$ {}^{238}_{92}U \ldots {}^{234}_{90}Th \ldots {}^{234}_{91}Pa \ldots {}^{234}_{92}U $$

$$ \ldots {}^{230}_{90}Th \ldots {}^{226}_{88}Ra \ldots {}^{222}_{86}Rn $$

This series finally decays to stable lead 206. The isotopes Ra^{228} and Ra^{224} are produced from the Thorium series by the following reactions:

$$ \ldots {}^{232}_{90}Th \ldots {}^{228}_{88}Ra \ldots {}^{228}_{89}Ac \ldots {}^{228}_{90}Th $$

$$ \ldots {}^{224}_{88}Ra \ldots {}^{220}_{86}Rn \ldots {}^{212}_{82}Pb \ldots {}^{208}_{82}Pb \text{ Stable} $$

$Radium^{228}$ and RA^{224} have 6.7 year and 3,6 half-lives respectively.

The maximum permissible concentrations based on the physical, biological and effective half-lives of the three pertinent isotopes of radium are stated in the National Bureau of Standards Handbook 69 to be:

Ra^{226}	10 uuc/L
Ra^{228}	30 uuc/L
Ra^{224}	2000 uuc/L

These values represent MPC for general population averages for life time usage.

It is also stated in the Federal Radiation Council Staff Report #2 that the total skeletal radium 226 concentration should be no greater than 0.001 micrograms in an average of suitable samples of average population. Assuming the body to retain that amount of radium contained in 50 liters of drinking water it follows that a concentration of 20 uuc/L of Ra^{226} is permissible. This corresponds to the top of Range II of the FRC radiation protection guides. These guides are predicated on average daily intakes for periods up to one year without demonstrable physiological damage.

The Federal Register of March 6, 1962 publishing the USPHS Drinking Water Standards on radioactivity states, "The effects of human radiation exposure are viewed as harmful and any unnecessary exposure to ionizing radioactivity should be avoided. Approval of water supplies containing radioactive materials shall be based upon the judgment that radioactivity intakes in such water supplies when added to that from all other sources aren't likely to result in an intake greater than the radiation protection guidance recommended by the Federal Radiation Council and approved by the President. Water supplies can be approved without further consideration of other sources of radioactivity intake of radium 226 and strontium 90 when the water contains these substances in amounts not exceeding 3 and 10 uuc per liter. When these concentrations are exceeded the water supplies shall be approved by the certifying authority after surveillance of radioactivity from all sources indicates that such intake are within the limits recommended by the Federal Radiation Council for control action. In the known absence of strontium 90 and alpha emitters the water supply is acceptable when the known beta concentration does not exceed 1000 uuc per liter. Gross beta concentration in excess of 1000 uuc per liter shall be grounds for rejection of the supply except when more complete analyses indicate the concentration of nuclides are not likely to cause exposures greater than the radiation protection guide as approved by the President on recommendation of the Federal Radiation Council." This figure of 3.0 uuc/L Ra^{226} is more conservative than the FRC guide limit in Range II and is designed to keep radiation levels as low as economically feasible.

Table I shows typical examples of radium 226 concentrations found in a few Iowa communities and demonstrates the fact that conventional cation exchange municipal softening is acceptably efficient for removal of elevated radium levels. It also shows the significant fluctuations of radium levels due to geologic and pumping variations.

It is common practice to blend back some unsoftened water into the effluent from a municipal cation exchanger in order to provide sufficient calcium ion for deposition of a corrosion protective $CaCO_3$ film on distribution piping. This practice reintroduces some radium concentration above the level of maximum removal efficiency for Ra^{226} and suggests the possibility that another source of Ca ion might be necessary where radium removal is the prime design criteria.

Massive epidemiological studies are now underway in an attempt to determine the authenticity of our presently accepted MPC values. If the levels remain the same or are reduced, it will obviously introduce a new concept into adequate water treatment design for water exceeding 20 uuc/L of radium 226. The design of treatment plants may eventually include zeolite

Table I. Radium 226

Town	*Source*	*Date*	Ra^{226}
Richland	Well #2	10/58	15.3
Danville	Distribution	6/60	9.0
Danville	Distribution	9/61	15.4
Danville	Distribution	9/61	13.5
Danville	Distribution	10/61	12.6
Donnellson	Well	9/5/61	54.0
Donnellson	Zeolite (Dist.)	9/5/61	7.1
Donnellson	Well	9/5/61	59.0
Donnellson	Zeolite (Dist.)	9/5/61	7.3
Donnellson	Well	9/12/61	38.0
Donnellson	Zeolite (Dist.)	9/12/61	5.4
Donnellson	Well	10/3/61	22.4
Donnellson	Zeolite (Dist.)	10/3/61	3.5
Donnellson	Well	10/31/61	30.4
Donnellson	Zeolite (Dist.)	10/31/61	4.2
Donnellson	Zeolite (Effluent)	10/31/61	1.0*
Eldon	Well	10/11/61	44.0
Fairfield	Well (2155)	9/58	19.1
Fairfield	Well	1/60	17.3
Morning Sun	Well	9/61	9.3
Morning Sun	Well	9/61	11.0
Morning Sun	Well	10/61	6.2
Morning Sun	Well	10/61	5.9
Mt. Pleasant	Well #3	9/58	9.7
Mt. Pleasant	Well #3	5/60	11.0
Mt. Pleasant	Well #4	5/60	11.1
West Point	Distribution	9/61	16.1
West Point	Distribution	10/61	16.4
West Point	Distribution	10/61	13.0
Winfield	Well	6/60	10.4
Winfield	Well	9/61	30.4
Winfield	Zeolite (Dist.)	9/61	10.4
Winfield	Well	10/3/61	16.9
Winfield	Well	10/3/61	26.6

* Zeolite effluent prior to any blend back. This indicates maximum radium removal of zeolite. Zeolite (Dist.) refers to the composite result after blend back for corrosion control. All values are in micromicrocuries per liter.

softening for radium control as well as for removal of hardness, iron and manganese.

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