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# Bad to the Bone: Maximum Contaminant Levels for Plutonium in Drinking Water

BY ARJUN MAKHIJANI

**Editor's note:** The promulgation in the United States of safe drinking water regulations for toxic chemicals and radionuclides in 1975 and 1976 pursuant to the Safe Drinking Water Act was a historic first step for making water safer for the public. But time has shown that many chemicals and radionuclides pose greater risks and a larger variety of risks to public health.

This article on maximum contaminant levels (MCLs) for plutonium-239, and radionuclides akin to it, is part of a series of IEER initiatives that will call attention to the implications of scientific research for public health and environmental protection. IEER's work shows that the existing MCL for these radionuclides, known as alpha-emitting, long-lived transuranic radionuclides, needs to be tightened by about 100 times in order to maintain approximately the same goals in regard to radiation protection that were part of the rulemaking when the MCL was first promulgated in 1976.

We invite your participation in these initiatives. The next legally-mandated review of radionuclide MCLs in drinking water is scheduled for 2006. We invite SDA readers and the organizations with which they are associated to join IEER in sending the U.S. Environmental Protection Agency a strong and clear message that the drinking water limit for alpha-emitting, long-lived transuranic radionuclides must be strengthened to protect public health. Please see the box on page 4 for information on how to do that.

This article is based on IEER's scientific analysis of drinking water MCLs for alpha-emitting transuranic radionuclides, entitled Bad to the Bone: Analysis of the Federal Maximum Contaminant Levels for Plutonium-239 and Other Alpha-Emitting Transuranic Radionuclides in Drinking Water. A summary of the report's main findings and recommendations can be found on pages 10-11. References can be found in the report, which is on the Web at www.ieer.org/reports/badtothebone/.

he National Primary Drinking Water Regulations specify rules that will protect drinking water and will maintain it in a state that is safe to drink. These regulations set standards for radionuclides in public water supplies under the 1974 Safe Drinking Water Act.<sup>1</sup> The standards are set in two ways: by specifying maximum contaminant levels of radioactivity in drink-

ing water, or by specifying



Still safe from plutonium. Will it stay that way? The federally-allowed level of drinking water contamination by plutonium and similar radionuclides is 100 times too high. States that may be impacted by plutonium waste include Georgia, Idaho, Nevada, New Mexico, Oregon,

maximum allowable radiation dose to the whole body or any organ as a result of ingestion of drinking water.

The current maximum contaminant level (MCL) for gross alpha particle activity — which includes radium-226, plutonium-239 and all other alpha-radiation-emitting radionuclides together, except uranium and radon — is 15 picocuries per liter.<sup>2</sup> There is a sub-limit for radium-226 and radium-228, combined, of 5 picocuries per liter (including any naturally present radium-226 and radium-228). For instance, if water is contaminated with plutonium-239 alone, the level of contamination could reach as high as 15 picocuries per liter if no other qualifying alpha-emitting radionuclides were present. If radium-226 is present to the maximum allowable limit of 5 picocuries per liter,<sup>3</sup> then the rule allows a maximum contaminant level for gross alpha of 10 picocuries per liter.

This standard was set in 1976, based on scientific assessments done in the late 1950s by the International Commission on Radiological Protection (ICRP) and the National Committee on Radiation Protection

Help keep drinking water safe from plutonium contamination \* see page 4 \*

and Measurements (NCRP). Both are scientific advisory bodies. The assessments were
published as ICRP Publication 2, in NRCP Report No. 22, and in abbreviated form by
the U.S. National Bureau of Standards as

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NBS Handbook 69.<sup>4</sup> The modern versions of the NBS handbooks are published by the U.S. Environmental Protection Agency (EPA) as a series of federal guidance reports (FGRs).

The drinking water standards as they were promulgated in 1976 distinguished broadly between naturally occurring and man-made radionuclides in the environment. Specifically, the EPA limited doses from man-made beta- and photon-emitting radionuclides to 4 millirem per year to the critical (or most exposed) organ. At the time, the main radionuclide contaminants in the water were from fallout due to atmospheric nuclear testing. Contamination with plutonium and other alpha-emitting, long-lived transuranic radionuclides was not considered explicitly, presumably because at the time these radionuclides were not considered as threats to public water systems, since their disposal was mainly within the boundaries of nuclear weapon plants (though some was sent to commercial low-level radioactive waste disposal sites). However, despite the lack of explicit consideration, the MCL for the alpha-emitting, long-lived transuranic radionuclides was set at a level that would produce a dose to the bone of less than 4 millirem per year.

By contrast, no specific MCL was set for uranium or for radon, both naturally occurring radionuclides, which are present in water. The sublimit that was set for radium-226 (a maximum of 5 picocuries per liter in the absence of radium-228) was considerably higher than a 4 millirem dose limit would imply. Radium-226 occurs naturally in water (and soil) in varying amounts, from a fraction of a picocurie per liter to tens of picocuries per liter. The EPA's intent was to reduce the most serious risks in areas with high radium contamination, and at the same time limit the overall cost of water treatment and the number of systems to be treated for radium removal.

The intent to limit exposure from all man-made sources is clear from the text of the regulation. Military sources were clearly included, since fallout from nuclear weapons testing was explicitly considered:

Man-made radioactivity may enter the public water systems from a variety of sources. Such contamination is usually confined to systems utilizing surface waters. Past deposition of fallout materials from nuclear weapons tests, particularly strontium-90 and tritium, is probably the most important source of contamination. The dose equivalent to individual users of public water systems in some areas of the United States from this pathway is in the range of 1 to 2 millirem (mrem) per year. At present, the dose equivalent from public water systems contaminated by effluents produced in the nuclear fuel cycle is probably only a fraction of that due to fallout materials, though perhaps ranging up to 0.5 mrem per year. The dose equivalent from effluents released by medical, scientific, and industrial users of radioactive materials that enter the public water systems has not been fully quantified. Taken as a whole these users handle much smaller amounts of radioactivity than nuclear power facilities but (with the exception of tritium) their liquid releases and the resultant doses to man may be somewhat comparable.

EPA recognizes that the national use of radionuclides in medicine and industry and the utilization of nuclear power to supply energy needs will unavoidably lead to some radioactivity entering the aquatic environment so that the quality of some surface waters is likely to decrease *slightly* in the future. *Even though the increase of radioactivity in drinking-water will normally be small, the Agency believes that the risk of future contamination* 

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warrants vigilance. It is the intent of the proposed monitoring and compliance requirements to provide a mechanism whereby the supplier of water can be cognizant of changes in the level of radioactivity in its water sources, so that the appropriate remedial measures may be taken.<sup>5</sup>

The maximum contaminant level for photon- and beta-emitters was set to 4 millirem per year because they were considered to be the most important sources of man-made radioactivity:

Considering the sum of the deposited fallout radioactivity and additional amounts due to effluents from other sources currently in existence, the total dose equivalent from made-made radioactivity is not likely to result in a total body or organ dose to any individual that exceeds 4 millirem per year...<sup>6</sup>

This quote shows that fallout was the single most important component of the dose from man-made radionuclides evaluated by the EPA at

that time. In looking to the future, the EPA did not anticipate that man-made radionuclides would result in a dose of more than 4 millirem per year to any organ from drinking water, because it believed that fallout would remain the main source and that this source would decrease with time due to the ban on atmospheric tests:

The 4 millirem per year standard for

man-made radioactivity was chosen

on the basis of avoiding undesirable future contamination of public water supplies as a result of controllable human activities. Given current levels of fallout radioactivity in public water supply systems and their expected future decline, and the degree of control on effluents from the nuclear industry that will be exercised by regulatory authorities, it is not anticipated that the maximum contaminant levels for manmade radioactivity will be exceeded except in extraordinary circumstances.<sup>7</sup>

#### **Evolving science**

The understanding of how radionuclides move within the human body once they are inhaled or ingested has improved greatly since the mid-1970s. One of the main changes has been the large increases in radiation dose to the bone estimated for the same intake of plutonium-239 and other alpha-emitting, long-lived transuranic radionuclides relative to those estimated using the method in NBS 69 on which the MCL for these radionuclides are still based. The other change has been how the "bone" is defined for the purposes of estimating doses and, hence, for radiation protection. As a result, the current MCL for alpha-emitting, long-lived transuranic radionuclides no longer reflects the intent of the safe drinking water rules promulgated in 1976 or the Safe Drinking Water Act (see Centerfold on pages 6 and 7). As we show below, the MCL of 15 picocuries per liter that is allowed for these radionuclides (in the absence of other alpha-emitting radionuclides<sup>8</sup>) is unsatisfactory as a basis for public health protection since it is at variance with the content and intent of the safe drinking water regulations for radionuclides that were first promulgated in 1976.

The first major changes in the official scientific understanding of how plutonium and other alpha-emitting, long-lived transuranic radionuclides behave in the human body, and of the magnitude of radiation dose they deliver to various organs, were recognized in by the ICRP in the late 1970s and early 1980s (that is, shortly after the promulgation of the U.S. safe drinking water rules). The EPA first adopted these changes for assessment of radiation doses in its Federal Guidance Report 11, published in 1988.

More changes have occurred since that time, which allow estimation of doses to people of various ages including infants, but these changes have not yet been incorporated into the drinking water standards. These changes were published by the EPA in Federal Guidance Report 13 in 1999 and in

# Evolving federal guidance : Late 1950s to 2002

an associated compact disk contain-

ing dose conversion factors in 2002.

In 1959, the National Bureau of

Standards published its Handbook 69 (NBS 69), which established the maximum permissible annual average concentrations of radionuclides in air and water calculated on the basis of a dose to the whole body or to the critical organ. Organs in radiation protection sometimes correspond to actual organs and sometimes they are more abstract, based on experiments and models of the movement of radionuclides in the body (called biokinetics). In the case of bone-seeking radionuclides, the long-established understanding that radium-226 behaves much like calcium in the body and concentrates in the bone led to the definition of the "marrow-free skeleton" as the critical organ, or the organ that would receive the maximum dose. NBS 69 published air and water concentration limits for radium-226, with a dose limit of 30 rem per year to the "bone" thus defined.

It was recognized at the time that other radionuclides that went to the bone did not behave in exactly the same way as radium-226. Specifically, they might not be as uniformly distributed in the bone as radium. Hence, for these radionuclides, a "safety factor" of 5 was es-

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The current MCL for alpha-emitting, long-lived transuranic radionuclides no longer reflects the intent of the safe drinking water rules promulgated in 1976 or the Safe Drinking Water Act

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tablished in estimating MCLs for air and water. This effectively reduced bone dose allowed for workers from these radionuclides to 6 rem. The critical organ was still said to be the marrow-free skeleton (the "bone"), but implicitly, by using a safety factor of 5, NBS 69 left the door open for revisions in how the distribution of plutonium and other alpha-emitting, long-lived transuranic radionuclides might be viewed in the future, given their differences from radium-226.

The dose to the bone, as then defined, for an MCL of 15 picocuries per liter works out to 1.8 millirem per year for all alpha-emitting, long-lived transuranic radio-nuclides of concern, except neptunium-237, for which it

is about 3 millirem per year. Hence, the gross alpha MCL set in 1976 so far as it concerned alpha-emitting, long-lived transuranic radionuclides was clearly set to produce doses less than the 4 millirem limit set for other man-made radionuclides.

In 1988, the EPA adopted a different scientific understanding of the bone for the purposes of radiation protection. In Federal Guidance

Report 11, the EPA defined two organs related to the bone: the "bone marrow" and the "bone surface." This was because it was discovered that radionuclides like plutonium-239 had a tendency to disproportionately affect endosteal cells, which are located close to the bone surface. Also, the dose conversion factors (the values for dose per unit intake of radioactivity) for alpha-emitting, long-lived transuranic radionuclides published in Federal Guidance Report 11 were far greater than those implied in the MCL published in NBS 69. As a result of these two changes, the doses from ingestion of plutonium in drinking water estimated according to the more modern understanding increased dramatically. The estimated cumulative dose to the critical organ over a lifetime due to ingesting water contaminated with plutonium-239 at the MCL of 15 picocuries per liter increased about 180 times when Federal Guidance Report 11 was used compared to NBS 69. Both the change in the critical organ (marrow-free skeleton to bone surface) and the increase in dose per unit intake played a significant role in the larger Federal Guidance Report 11 estimate.<sup>9</sup>

The understanding of plutonium biokinetics has further evolved since 1988. In recent years, dose conversion factors for various ages have been published. This

The MCLs for alpha-emitting, long-lived transuranic radionuclides should be reduced from 15 picocuries per liter to 0.15 picocuries per liter. means that the lifetime dose can now be computed assuming an intake each year of a person's life of water contaminated at the MCL. Such calculations are possible using Federal Guidance Report 13 dose conversion factors, which the EPA published in 2002 on a compact disk.

Using these more up-to-date, age-specific dose conversion factors, we calculated that the dose to

the maximally exposed organ (the bone surface) using Federal Guidance Report 13 would be about 100 times greater than that to the bone as defined in NBS 69 (the marrow-free skeleton). This means that, according to the most current figures published by the EPA, the MCL for plutonium-239 and other alpha-emitting, long-lived transuranic radionuclides is too high by a factor of about 100. In other words, in order to remain within the intent and spirit of the 1976 regulations, the MCLs for alpha-

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# $\star$ Help keep drinking water safe from plutonium contamination $\star$

The Safe Drinking Water Act requires the U.S. Environmental Protection Agency (EPA) to review and revise, as appropriate, each national drinking water regulation at least once every six years.

On August 1, 2005, IEER and other groups sent a letter urging the EPA to incorporate IEER's scientific analysis of the drinking water standard for alpha-emitting transuranic radionuclides into its radionuclide review scheduled for 2006. The goal is to get the EPA to adopt the central recommendation: Set the combined Maximum Contaminant Limit (MCL) for alpha-emitting, long-lived transuranic radionuclides at 0.15 picocuries per liter.

We invite you, our readers, to endorse the report. Please encourage groups with which you are affiliated to endorse it, too. The more groups and individuals endorsing the report, the more likely the EPA will act to strengthen the drinking water standard.

**Endorse the report by visiting http://www.ieer.org/epaletter/** and clicking "sign the letter." Or call IEER at 301-270-5500. The letter will remain open for sign-ons until the EPA concludes its review, likely in fall 2006. IEER will forward additional signatures to the EPA periodically until that time.

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emitting, long-lived transuranic radionuclides should be reduced from 15 picocuries per liter to 0.15 picocuries per liter (annual average).

In considering this recommendation, IEER also looked at limits for plutonium-239 limit that exist in other standards. The surface water standard of the State of Colorado is the most relevant, since that state has been host to one of the most important plutonium handling and processing facilities in the United States, the Rocky Flats Plant near public wa

Denver. The statewide standard for plutonium-239 for surface water is 0.15 picocuries per liter.<sup>10</sup> See the box on page 8.

#### Conclusions

In conclusion, the MCL for alpha-emitting, long-lived transuranic radionuclides should be tightened by about a factor of 100 – that is, it should be reduced from 15 picocuries per liter to 0.15 picocuries per liter. The 15 picocuries per liter limit for transuranic radionuclides is obsolete, not protective of public health, against the spirit of the Safe Drinking Water Act, and not in accord with the intent of the initial regulation.

A combined standard for all alpha-emitting, longlived transuranic radionuclides will simplify the rule and reduce the cost of its enforcement. Moreover, since the plutonium isotopes among these dominate the total curie content of U.S. Department of Energy (DOE) waste<sup>11</sup> and since the dose conversion factors for Pu-238, Pu-239, Pu-240, Pu-242, and Am-241 are nearly the same, using Pu-239 as a reference for deriving the combined standard MCL is reasonable from a health standpoint as well as cost-effective.<sup>12</sup>

Corresponding to the change in the MCL for alphaemitting, long-lived transuranic radionuclides, there is also a need for a change in the detection limit. The regulation should be modified to include a separate detection limit of 0.01 picocuries per liter for each alpha-emitting, long-lived transuranic radionuclide. The current detection limit for these radionuclides is 0.001 picocuries per liter. The errors at such low levels can be large.

Our main findings and recommendations are on pages 10–11. The EPA should take up consideration of a tightened standard and the other recommendations in its upcoming 2006 drinking water radionuclide review.

#### Costs

Alpha-emitting, long-lived transuranic radionuclides are not ubiquitous in significant concentrations, unlike naturally occurring radionuclides like uranium-238, uranium-234, and radium-226. Almost all public water systems can therefore be exempted from routine monitoring requirements relating to such radionuclides. The monitoring requirements for these radionuclides should be applied to public water systems that draw water from aquifers or surface water that have potential hydrologic or hydrogeologic connections to areas or facilities with waste tanks, waste burial pits, and other potential sources of alpha-emitting, long-lived transuranic radionuclides in combined totals in excess of 100 curies.<sup>13</sup> Wastes disposed of at shallow and intermediate depths are included in this

No known contamination of public water systems exists above 0.15 picocuries per liter of alpha-emitting, long-lived transuranic radionuclides. definition. Alpha-emitting, long-lived transuranic radionuclides that are contained in secure buildings with institutional controls would be exempt from this limit and the associated monitoring requirements.

Public water systems are not at present contaminated at or near the requested MCL for alpha-emitting, long-lived transuranic radionuclides.

A strengthened drinking water standard is preventive rather than remedial. Only a small, one-time cost for an initial set of baseline samples is anticipated for those water systems that draw water from sources that include DOE sites with significant plutonium waste or soil contamination in drainage areas. We recommend that this one-time cost be borne by the DOE.

Since no known contamination of public water systems exists above 0.15 picocuries per liter of alpha-emitting, long-lived transuranic radionuclides, no further action would be required of public water systems and no further costs would be incurred provided there is sufficiently thorough monitoring by the DOE, coupled with remediation programs that are suited to free release of the sites in the long term. This will be sufficient to protect downstream surface waters and underground water systems. The DOE is supposed to carry out such monitoring in any case and therefore no additional, ongoing monitoring costs are anticipated.

The DOE, which is responsible for management of almost all the wastes and materials that pose risks of water contamination with alpha-emitting, long-lived transuranic radionuclides, is supposed to take adequate remedial action at sites like the Idaho National Laboratory, Hanford, the Savannah River Site, and Los Alamos National Laboratory. If it does so, no remediation costs for public water systems would be required under our recommended changes to the National Primary Drinking Water Regulations.

The costs of not tightening the standards would be to signal that remediation of nuclear weapons sites with large inventories of plutonium in the waste could proceed without adequate attention to safe drinking water health protection goals. DOE could then remediate these sites and declare them "cleaned up" without reference to a

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# SCIENCE FOR THE CRITICAL MASSES

# Plutonium and

## TIMELINE: PLUTONIUM DRINKING WATER STANDARDS IN THE UNITED STATES

# 1959

**NBS 69** and **ICRP 2** are published, setting forth the scientific approach for calculating maximum permissible concentrations of radionuclides in air and water. NBS 69 establishes the maximum permissible average concentrations of radionuclides in air and water calculated on the basis of a 5 rem dose to the whole body, and a 15 rem dose to the critical organ, with somewhat different methods used for bone-seeking radionuclides. NBS 69 defines the marrow-free skeleton as the critical organ for alpha-emitting, long-lived transuranic radionuclides.

## TERMINOLOGY

#### Alpha-emitting, long-lived transuranic radionuclides:

"Transuranic" radionuclides are radioactive isotopes that have atomic numbers greater than uranium (92), i.e., *trans*-, or beyond, uranium. "Long-lived" refers to the half-life of the radionuclide, or the duration that the radionuclide remains dangerous to human health. "Alpha-emitting" radionuclides emit alpha particles, which are positively charged particles consisting of two neutrons and two protons, when they undergo radioactive decay.

- **Committed dose:** the amount of energy from ionizing radiation deposited in an organ over the entire time that a radionuclide is present in the organ. Committed dose is different than annual dose in that annual dose is the amount of energy deposited in the organ in a single year. If a radionuclide is eliminated rapidly from the body, say in a few days or weeks, then annual dose and committed dose are usually the same. But if the radionuclide is slowly eliminated from the target organ, over years or even decades (decades being the case for alphaemitting, long-lived transuranic radionuclides), the dose to the organ from an intake in any given year is delivered over a period of decades after that.
- **Critical organ:** the organ or tissue that, when exposed to radiation, results in the greatest health hazard to the individual.
- **Gross alpha:** measurement of the total radioactivity caused by alpha particle emissions, regardless of specific radionuclide source. Also known as gross alpha particle activity.
- **MCL:** Maximum Contaminant Level, the level of pollutant concentration above which a public water system would be in violation of the U.S. Environmental Protection Agency drinking water regulation. The MCL is generally calculated as an annual average level.
- **MCLG:** Maximum Contaminant Level Goal, a non-enforceable level above which there are possible adverse health effects. The MCLG for all radionuclides is zero, since every exposure to radiation produces and incremental risk of cancer.

# 1974

Safe Drinking Water Act is enacted by Congress "to protect public health by regulating the nation's public drinking water supply," authorizing the U.S. Environmental Protection Agency (EPA) to establish regulations on contaminants in public drinking water systems. (see below)

## SAFE DRINKING WATER ACT

Congress passed the Safe Drinking Water Act (SDWA) in 1974 to "protect public health by regulating the nation's public drinking water supply." The SDWA was amended in 1986 and 1996 and authorizes the EPA to set national health-based standards for drinking water to protect against both naturally-occurring and man-made contaminants in drinking water. The primacy of the health goal (rather than numerical limits) is clear from the EPA's description of the SDWA:

US EPA sets national standards for tap water which help ensure consistent quality in our nation's water supply. US EPA prioritizes contaminants for potential regulation based on risk and how often they occur in water supplies. (To aid in this effort, certain water systems monitor for the presence of contaminants for which no national standards currently exist and collect information on their occurrence). US EPA sets a health goal based on risk (including risks to the most sensitive people, e.g., infants, children, pregnant women, the elderly, and the immuno-compromised). US EPA then sets a legal limit for the contaminant in drinking water or a required treatment technique. (Safe Drinking Water Act 30th Anniversary: Understanding the Safe Drinking Water Act, EPA Fact Sheet, at www.epa.gov/safewater/sdwa/30th/ factsheets/understand.html)

# **Drinking Water**

# 1975-1976

National Primary Drinking Water Regulations are proposed by EPA for the first time in March 1975. Rules for radionuclides are proposed in August 1975. Regulations for contaminants other than radionuclides promulgated in December 1975. Regulations for radionuclides are promulgated in July 1976. The MCLs and dose limits for radionuclides were originally codified in 40 CFR 141.15 and 40 CFR 141.16, both of which have since been renumbered and consolidated, without change, into 40 CFR 141.66. Detection limits and analytical methods for radionuclides were set forth in 40 CFR 141.25. (CFR stands for Code of Federal Regulations.)

# 1988

Federal Guidance Report 11 (FGR 11) is published by the EPA, forming the basis of many present-day EPA dose estimation methods. Compared to NBS 69, FGR 11 incorporated new advances in scientific understanding of radiation doses and harm from intake of radionuclides, namely using ICRP publications 26, 30, and 48. FGR 11 defines the bone surface as the critical organ for alpha-emitting, long-lived transuranic radionuclides.

# 1999

Federal Guidance Report 13 (FGR 13) is published by the EPA. FGR 13 is the most recent EPA guidance but not yet in force for regulatory calculations for doses from air and water. It is based on dose conversion factors published in ICRP 72. The dose conversion factors (that is, dose per unit intake of radioactivity) for alpha-emitting, long-lived transuranic radionuclides in FGR 13 are generally somewhat lower than those in FGR 11.

# U.S. DRINKING WATER LIMITS FOR RADIATION AND CERTAIN RADIONUCLIDES

Radionuclide	Drinking water limit	Comments
Man-made beta and photon emitters	4 mrem/yr to the critical organ	Includes fission products like cesium-137, iodine-129, and technetium-99. Excludes strontium-90 and tritium. MCLs are calculated from the 4 mrem/yr dose limit. Presence of more than one radionuclide reduces the MCL for each so total dose does not exceed 4 mrem/yr to the critical organ.
Gross alpha	I5 pCi/L	Radon and uranium not included, includes radium-226 and alpha-emitting, long-lived transuranic radionuclides (including plutonium-239). Presence of more than one radionuclide reduces the MCL for each so total does not exceed 15 picocuries per liter.
Radium	5 pCi/L	Includes radium-226 and radium-228. The latter is a beta-emitter.
Uranium	30 µg/L	Promulgated as part of the year 2000 review. Regulated as a toxic metal, not a radionuclide. MCL on a radioactivity basis varies according to enrichment: 20 picocuries per liter for natural uranium, about 12 picocuries per liter for depleted uranium, and more than 20 picocuries per liter for enriched uranium.
Strontium-90	8 <sub>P</sub> Ci/L	
Tritium	20,000 pCi/L	

mrem/yr = millirem per year, MCL = Maximum Contaminant Limit, pCi/L = picocuries per liter, µg/L = micrograms per liter

#### - ANSWERS TO ATOMIC PUZZLER FROM SDA VOL. 13 NO. 1

(1) a. 2,400 to 2,500 kWh/SWU	(3) a. Approximately 60 kg	(5) a. 3 to 5 percent U-235
b. 50 to 60 kWh	b. 20-25 kg	b. 90 percent U-235
(2) a. Approximately 100,000 to	c. Between 10.6 and 13.1 metric tons	c. 0.7 percent U-235
120,000 SWU per year	natural uranium and between 11,600 to	(6) False. The required amount of natural
b. Roughly 3 to 4	13,700 SWU enrichment services	uranium and the required number of SWUs
c. Less than 0.1 percent	d. 1.6 tons / less than one tenth the amount	during enrichment change in opposite direc-
	(4) a. Two thirds	tions for a given level of enrichment.
	b. One-third	(7) True

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science-based drinking water standard that corresponds to current understanding of plutonium's harm to the human body. Some remediation actions could, in the long run, pollute the water above drinking water standards, and worse, be irremediable. Once polluted, no known technology could remediate vast bodies of water such as the Savannah River or the Snake River Plan Aquifer if the aim is to reduce pollution from a few picocuries per liter to sub-picocurie per liter levels.

#### The urgency of the recommendations

Vast areas of land and huge amounts of water remain contaminated with dangerous long-lived radionuclides from operations of nuclear weapons facilities. The DOE has been given the task to clean up these sites. It is therefore of great importance that the levels of residual radioactivity meet strict standards that will protect the health of individuals of this and future generations that will be exposed to the residual contamination.

In the early 1990s, the DOE embarked on a cooperative

process with the EPA to develop national cleanup standards, but the DOE pulled out of the process abruptly in 1996 without any plans for its resumption. Since then, the DOE has proceeded on a site-by-site basis that has led to a welter of proposals for cleanup using various scenarios.

At the Savannah River Site in South Carolina, the DOE is grouting high-level waste in tanks as if it were low-level waste. The tanks are buried underground in the watershed of the Savannah River, one of the most important rivers in the South Carolina-Georgia region. This waste contains significant amounts of transuranic radionuclides. For instance, the residual waste in Tank 19, which has been grouted, had a concentration of plutonium 14 times above the EPA 100 nanocurie-per-gram limit for transuranic waste, which is normally required to be disposed of in a deep geologic repository. DOE is preparing to grout significant amounts of plutonium and other radionuclides in the tanks even though it has not yet obtained convincing evidence of the durability of grout. IEER's evaluation of the state of the research on grout indicates that the performance of grout remains highly

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# COLORADO'S DRINKING WATER STANDARD FOR PLUTONIUM

The standard of the State of Colorado for plutonium-239 for surface water is 0.15 picocuries per liter. It is calculated on the basis of a 30-day rolling average – that is, 30 consecutive measurements are averaged; they may or may not be taken on consecutive days. Colorado's standard is based on the risk of one person in one million developing a cancer from consuming two liters of water per day for 30 years.

The Colorado Department of Health,Water Quality Control Commission described the background and the rationale for changing the standard from 15 to 0.15 picocuries per liter (pCi/L) as follows:

Background The Commission previously adopted a basic standard for plutonium of 15 pCi/L and had no basic standard for americium. A basic standard was considered in this hearing for americium because it is closely associated with plutonium and these two radionuclides generally occur together. The current basic standard of 15 pCi/L plutonium was calculated using methodologies in the 1976 National Interim Primary Drinking Water Regulations and was consistent with a goal of keeping exposures below 4 millirem per year. The Basis and Purpose indicated that it was necessary and important to restrict levels because of the difficulty of removing this radionuclide by conventional treatment procedures and because the potential adverse effect on human health suggests that extreme caution be exercised in its release to State waters. Since plutonium is predominantly an alpha emitter, the basic standard was made consistent with the 15 pCi/L alpha standard....

**Basis for Commission Decision** Since the previous basic standard was set, several changes have occurred: 1) a new methodology for assessing carcinogens has become the standard practice, 2) new data have resulted in periodic updates to the slope factors used in this methodology, and 3) a more refined Commission policy on appropriate levels of protection for carcinogens has been developed. This latter risk-based policy also parallels a national trend towards risk-based approach to environmental cleanup standards.

The 15 pCi/L dose-based approach was calculated using a "reference-man" and considered exposure during his working life. It was an approach designed to address questions related to occupational exposure. It did not consider sex, age and organ-specific factors over a lifetime. In contrast, the new slope factor methodology, used in EPA's 1989 Risk Assessment Guidance for Superfund Sites, is more complete, more applicable to a general population and has become the standard practice for calculating risk.

The Commission adopted a basic standard of 0.15 pCi/L for plutonium and americium, calculated using a  $1 \times 10^{-6}$  risk level, based on residential use. This risk level is consistent with the Commission's policy for human health protection.\*

The central scientific point of the Colorado rule is that the science has changed, indicating greater risk than previously assumed from exposure to plutonium and americium; therefore the maximum contaminant limits should be adjusted accordingly.

\* Colorado Department of Public Health and Environment. Water Quality Control Commission. The Basic Standards and Methodologies for Surface Water (5 CCR 1002-31). Regulation No 31. Originally adopted in 1979 and last amended on November 8, 2004, with the amendments to be effective March 22, 2005. Link on the Web at http://www.cdphe.state.co.us/op/regs/waterqualityregs.asp. Viewed June 2, 2005. Pages 138-139.

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uncertain.<sup>14</sup> There is at present no sound basis, whether in laboratory experiments or in field data, to assume that leaving large amounts of grouted alpha-emitting, longlived transuranic radionuclides in the tanks would be protective of the Savannah River in the long-term.

A large part of the urgency that our recommendations be incorporated into EPA's forthcoming review of MCLs for radionuclides in drinking water derives from the fact that, in 2004, Congress passed a law allowing DOE to reclassify residual high-level waste as incidental waste at its South Carolina and Idaho sites. The law did not set any limits as to the residual radioactivity allowed to remain in this waste. Several long-lived radionuclides, including plutonium isotopes, strontium-90, and cesium-137, may be grouted in the tanks or disposed of in shallow saltstone vaults. A realistic framework to guide DOE's decision-making, so that it does not endanger crucial water resources, is therefore of urgent and immense importance.

#### Emerging science

New scientific work on radiation health effects is currently emerging, for instance in relation to (i) protection of the embryo/fetus and infant, (ii) non-cancer effects of exposure to certain radionuclides, (iii) potential synergistic effects of exposure to certain chemicals, such as hormonally active chemicals, and exposure to radiation, (iv) the need for protection of key non-human species and ecosystems, and (v) the synergisms indicated for certain effects between the heavy metal toxicity component of uranium and its radiotoxicity. These are emerging areas of concern, where the risks are not quantitatively wellestablished. Such risks need to be incorporated in radiation and health protection as a whole. This issue will be a major part of IEER's work in the next two years. Because these considerations are based on emerging science and will require a wide debate on protection of health from environmental pollutants, their incorporation into radiation protection standards is likely to take some time.

The matter of revising the MCLs for alpha-emitting, long-lived transuranic radionuclides is far more simple, as it is based on existing science and existing regulations. Since the EPA is legally mandated to review its radionuclide drinking water standards in 2006, accomplishing this change should be a straightforward matter. It is, moreover, urgent, since the DOE is now grouting large amounts of plutonium in the watershed of the Savannah River and since its approach to clean-up has deteriorated significantly in the past few years. This is putting crucial water resources at risk.

Tightening drinking water MCLs for alpha-emitting, long-lived transuranic radionuclides is important because it will ensure that DOE's cleanup approach and levels of residual radioactivity do not exceed levels that would endanger the health of future generations far more than indicated by present day science and the intent of drinking water regulations. A reduction of the alpha-emitting, long-lived transuranic radionuclides MCL to 0.15 picocuries per liter is indicated by the science, by the spirit and intent of the drinking water rules, and by the practical necessity to protect water resources near sites with significant amounts of plutonium waste.

- The National Primary Drinking Water Regulations are published under 40 CFR 141.66 (formerly 40 CFR 141.15 and 141.16), on the Web at http://www.access.gpo.gov/nara/cfr/waisidx\_04/40cfr141\_ 04.html. The Safe Drinking Water Act is on the Web at http:// www.epa.gov/safewater/sdwa/index.html. Also see page 8.
- 2 40 CFR 141.66(c). The radionuclides of particular concern here are the long-lived transuranic radionuclides – neptunium-237, plutonium-238, plutonium-239, plutonium-240, plutonium-242, americium-241, and americium-243 – all man-made radionuclides.
- 3 This assumes that no radium-228 is present. Radium-228 is a betaemitting radionuclide that is not included in the gross alpha MCL. The radium MCL in the rule is set for the combined concentration of Ra-226 and Ra-228 even though one is an alpha emitter and one is a beta emitter because small amounts of both are naturally present in essentially all water bodies along with other natural alphaand beta-emitting radionuclides (like uranium-238 and potassium-40, respectively).
- 4 NBS 69, which also bears the series title NCRP Report No. 22, is a recommendation of the National Committee on Radiation Protection and Measurements, which is now known as the National Council on Radiation Protection and Measurements. The dose conversion factors, the scientific content, and other details in NBS 69 are the same as those in ICRP 2. ICRP 2 was published by the International Commission on Radiological Protection in 1959. The NCRP was (and is) a participating organization in ICRP. We will refer to NBS 69 in this article for ease of reference.
- 5 "Environmental Protection Agency [40 CFR Part 141] [FRL 410-3] Interim Primary Drinking Water Regulations, Notice of Proposed Maximum Contaminant Levels for Radioactivity." *Federal Register*, v. 40, no. 158, August. 14, 1975, page 34324, emphasis added.
- 6 ibid., page 34325, emphasis added.
- 7 ibid., pages 34325-34326, emphasis added. Of the nuclear weapons states, only China was testing in the atmosphere at the time. China conducted its last atmosphere nuclear test in 1980.
- 8 Uranium is still not included in the gross alpha limits for drinking water. The EPA established a separate limit of 30 micrograms per liter for uranium, based on its toxicity as a heavy metal. IEER will, in future articles and publications, consider the issue of uranium and other radionuclides more broadly as they relate to health protection.
- 9 While the bone surface was not specified as a target organ for dose calculations in 1976 when the safe drinking water regulations were promulgated, it is possible to estimate the dose to the endosteal cells at a level of drinking water contamination of 15 picocuries per liter based on the NBS 69 dose conversion factors. For pluto-nium-239, the annual dose to the endosteal cells would be about 26 millirem per year. The bone surface dose for other alpha-emitting, long-lived transuranic radionuclides are about the same, except for neptunium-237, for which the figure is about 44 mrem per year. We have not used these estimates in the recommendation for change in the MCL because the drinking water regulations clearly require limitation of dose to the critical organ, however it is defined. In 1976 this meant the marrow-free skeleton. At present, it means the bone surface.
- 10 The State of Colorado estimated its limit based on risk limits in the Superfund law (CERCLA). We have used the dose limits and text of the drinking water regulation itself in arriving at our recommendation. Colorado also sets standards for other radionuclides and considers different limits for different watersheds. We have not considered these issues, some of which result in more stringent

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### MAIN FINDINGS

1. The drinking water maximum contaminant limit (MCL) for plutonium-239 and other alpha-emitting, long-lived transuranic radionuclides is about a hundred times too lax.

The most recent science, as published by the U.S. Environmental Protection Agency (EPA), indicates that the radiation dose to the most exposed organ, the surface of the bone, from drinking water contaminated to the maximum allowable limit is about a hundred times greater than the dose to what in 1976 was regarded as the maximally exposed organ (the marrow-free skeleton). This indicates that the drinking water standards are about a hundred times too lax, as measured by the intent of the regulations when they were first promulgated. The current MCL for each alphaemitting, long-lived transuranic radionuclide separately is 15 picocuries per liter.

- 2. Drinking water regulations, when they were first set, explicitly included military sources of radionuclides — specifically, fallout from testing.
- 3. A much tighter MCL for alpha-emitting, longlived transuranic radionuclides is needed to prevent lax approaches to cleanup of weapons sites.

Once drinking water is polluted to a few picocuries per liter, which is many times the indicated MCL by current science, it will be essentially impossible to remediate it. A stringent MCL is therefore needed as a guide to the U.S. Department of Energy (DOE) in its cleanup and as a preventive measure for protecting public water supplies.

#### 4. The vast majority of public water systems will incur no costs from the proposed change and a few would incur a one-time monitoring cost.

The vast majority of public water systems have levels of alpha-emitting, long-lived transuranic radionuclide that are orders of magnitude below the proposed MCL. They are not at risk for further contamination. No sampling, monitoring, or remediation is needed for these systems. For public water systems that are hydrologically or hydrogeologically connected to DOE sites where large amounts of plutonium waste were dumped or were disposed of, a onetime initial sampling and analysis should be done. If found clean, further sampling need not be conducted provided the DOE maintains a thorough water sampling program for surface and ground waters on site and reports the results publicly. It is presently mandated to do that, so no additional expenses would be incurred in this regard.

5. The relaxation of DOE goals in regard to cleanup and the lack of national cleanup standards necessitates an urgent revision of MCLs for alpha-emitting, long-lived transuranic radionuclides, if critical drinking water systems are to be protected for the long-term.

The timing and urgency of the main recommendation of this report — that MCLs for alpha-emitting, long-lived transuranic radionuclides be tightened by one hundred times derives largely from the very large inventories of alpha-emitting, long-lived transuranic radionuclides at several DOE nuclear weapons sites. Some wastes containing these radionuclides (both low-level and transuranic wastes) were dumped in unlined trenches in cardboard boxes and similar non-durable packaging in the early decades of the Cold War. The primary sites are in Idaho, Nevada, New Mexico, South Carolina (at the Georgia border), Tennessee, and Washington state (upstream from Oregon). The combined plutonium-238, -239, and -240 inventory contained in DOE high-level waste tanks at Savannah River Site is over a million curies. In 2004, Congress gave DOE the latitude to reclassify some of this waste. DOE can now grout high-level waste in place by reclassifying it as waste incidental to reprocessing. Congress set no limit on the total residual radioactivity content of the grouted waste. Since grouting the tanks is essentially irreversible, it is imperative the DOE implement the law in a manner that is compatible with the protection of the Savannah River, which is increasingly used by more people as a source of drinking water in South Carolina and Georgia.

# Thanks.

IEER is grateful to our superscribers (people who have donated at least \$100 to IEER), hyperscribers (at least \$250), and Dr. Egghead's Financial Angels (\$1,000 or more).

**Dr. Egghead's Financial Angels** since January 2005: Kay Drey, Elizabeth Shafer, Cathie Sullivan **Superscribers** since January 2005: Gerhard Bedding, Henry S. Kahn

Your support allows SDA to remain the valuable resource it's been for more than a dozen years.

Thanks also to our foundation funders, listed on page 2.

#### RECOMMENDATIONS

1. The U.S. Environmental Protection Agency (EPA) should reduce its maximum contaminant levels for all alpha-emitting, long-lived transuranic radionuclides, combined, by one hundred times to an MCL of 0.15 picocuries per liter during its 2006 review of radionuclide standards for drinking water.

EPA should set a combined maximum contaminant level for alpha-emitting, long-lived transuranic radionuclides of 0.15 picocuries per liter. If only one of the radionuclides in question were present, then the limit for that radionuclide would be 0.15 picocuries per liter. The radionuclides included are: neptunium-237, plutonium-238, plutonium-239, plutonium-240, plutonium-242, americium-241, and americium-243. These changes should be made as part of the EPA's review of radionuclide standards in drinking water that is scheduled for 2006.

#### 2. The DOE should fund a one-time baseline sampling and analysis for public water systems that are hydrologically or hydrogeologically connected to DOE sites with major plutonium wastes or dumps.

DOE sites with wastes buried underground or in tanks containing more than 100 curies of alpha-emitting, longlived transuranic radionuclides should be considered to have potential risks to drinking water. These sites include the Savannah River Site, Hanford, Idaho National Laboratory, Los Alamos National Laboratory, Oak Ridge, and the Nevada Test Site. Testing of downstream water for the purpose of providing a baseline level of contamination is desirable and should be funded by the DOE since the tiny amounts of alpha-emitting, long-lived transuranic radionuclides in current water supplies are due to military-related atomic energy activities (fallout from nuclear weapons testing).

3. The DOE should evaluate its on-site water monitoring from the point of view of the proposed standard and intensify it, if necessary. Resources for state and local verification should be provided by the federal government.

The DOE currently carries out extensive surface and ground water monitoring. This may be sufficient for the purposes of providing assurance that downstream water resources con-

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- and others of which result in more lax rules. We use the State of Colorado's statewide surface water limit for Pu-239 as a guide
- because our concerns are related to nuclear weapons plants and
- Colorado's limit for plutonium was specifically aimed at protection
- of surface water from contamination at the Rocky Flats nuclear weapons plant (now dismantled, but with considerable residual
- radioactivity still left in the ground in parts of the site).
- 11 The DOE is responsible for management of almost all the wastes and materials that pose risks of water contamination with alphaemitting, long-lived transuranic radionuclides.
- 12 The dose conversion factor for Np-237 is lower than those of the other alpha-emitting, long-lived transuranic radionuclides by about a factor of two.
- 13 For instance, the 100 curie limit is equivalent to 1,000 metric tons

tinue to be protected from contamination with alpha-emitting, long-lived transuranic radionuclides. If not, the existing programs should be intensified.

The federal government should also provide states and public water system authorities that are hydrologically or hydrogeologically contiguous to DOE sites with the funds to conduct independent checks on DOE's on-site and offsite water monitoring. Such funds would better be provided through the EPA, rather than through the DOE, in order to assure the independence of the monitoring and the continuity of the funding.

- 4. A separate limit of detection of each alpha-emitting, long-lived transuranic radionuclide of 0.01 picocuries per liter should be set.
- 5. The DOE should make public the source code for the computer model that is used to assess the impact of residual radioactivity on food, water, and the environment.

Argonne National Laboratory developed a "family" of computer software programs to assess the radiological impact of environmental contamination by radionuclides. The main one, called simply RESRAD, is used to assess the impact of residual radioactivity in the soil on human beings by estimating radiation doses by a variety of pathways, such as food and water and resuspended soil. Its source code, or the textbased set of instructions that make up the program, is not public. It does not incorporate dose conversion factors for children, infants, or fetuses at various times in their development. Its internal structure and its effects on the resulting estimates of doses and risks are not available for independent scrutiny. We strongly recommend that the RESRAD source code be made public, so that it can be examined and improved in the manner of the operating system Linux. The government, of course, need not adopt any changes that are made by the public unless it finds them useful for implementing environmental regulations. But there is no reason for holding secret the RESRAD source code, which is paid for by taxpayer dollars, particularly as billions of dollars are being spent on cleanup decisions based on the results generated by the RESRAD program.

of transuranic waste containing alpha-emitting, long-lived transuranic radionuclides at the lower limit of 100 nanocuries per gram. It would be equivalent to a larger mass of low-level waste, since the concentration in such waste (by definition) is less than 100 nanocuries per gram.

14 See Brice Smith, What the DOE knows it doesn't know about grout: serious doubts remain about the durability of concrete proposed to immobilize high-level nuclear waste in the tank farms at the Savannah River Site and other DOE sites (IEER, updated October 18, 2004), on the Web at www.ieer.org/reports/srs/grout.pdf. Also see Arjun Makhijani and Michele Boyd, Nuclear Dumps by the Riverside: Threats to the Savannah River from Radioactive Contamination at the Savannah River Site (IEER, March 11, 2004), on the Web at www.ieer.org/reports/srs/index.html.

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# Alpha-emitting, long-lived transuranic radionuclides

- a. Greek literary radio broadcasters who live to a ripe old age.
- b. Derogatory slang for people who won't stop talking about nuclear waste.
- c. "Transuranic" radionuclides are radioactive isotopes that have atomic numbers greater than uranium (92), i.e., *trans-*, or beyond, uranium. "Alpha-emitting" radionuclides emit alpha particles, which are positively charged particles consisting of two neutrons and two protons, when they undergo radioactive decay. "Long-lived" refers to the half-life of the radionuclide, or the duration that the radionuclide remains
  - dangerous to human health.

# Committed dose

- a. A dose that has promised faithfulness, fidelity, and love to another dose.
- b. A rare disorder characterized by incessantly yelling "D'oh!," an expression made popular by the TV cartoon character Homer Simpson.
- c. The amount of energy from ionizing radiation deposited in an organ over the entire time that a radionuclide is present in the organ. Committed dose is different
- than annual dose in that annual dose is the amount of energy deposited in the organ in a single year. If a
- radionuclide is eliminated rapidly from the body, say in
- a few days or weeks, then annual dose and committed
- dose are usually the same. But if the radionuclide is
- slowly eliminated from the target organ, over years or
- even decades (decades being the case for alpha-emit-

## ting, long-lived transuranic radionuclides), the dose to the organ from an intake in any given year is delivered over a period of decades after that.

## Critical organ

- a. The part of the body, usually the gall bladder, that is the most disapproving of others.
- b. The musical instrument in Dracula films.
- c. The organ or tissue that, when exposed to radiation, results in the greatest health hazard to the individual.

## Gross alpha

- a. Bioengineered alfalfa that tastes very bad.
- b. The flatulent cousin of Dr. Egghead's dog Gamma.
- c. A measurement of the total radioactivity caused by alpha particle emissions, regardless of specific radionuclide source. Also known as gross alpha particle activity.

# MCL

- a. Macho-Chauvinist League, a men's organization formed in reaction to the feminist movement.
- b. Minimum Closeness Limit, the shortest distance at which two people in conversation comfortably stand from each other. MCL is usually dependent upon culture.
- c. Maximum Contaminant Level, the level of pollutant concentration above which a public water system would be in violation of the U.S. Environmental Protection Agency drinking water regulation. The MCL is generally calculated as an annual average level.

Answers: c, c, c, c, c

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