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Bad to the Bone:

Analysis of the Federal Maximum Contaminant Levels for Plutonium-239 and Other Alpha-Emitting Transuranic Radionuclides in Drinking Water

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Main findings

The limit for gross-alpha contamination of drinking water is based on science that is over four decades old. It is an unsatisfactory basis for public health protection that is at variance with the content and intent of the safe drinking water regulations for radionuclides that were first promulgated in 1976. Specifically, the 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, has changed a great deal, beginning with revisions first published by the International Commission on Radiological Protection in the late 1970s. The United States Environmental Protection Agency (EPA) first officially 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.

EPA last reviewed its radionuclide standards in the year 2000 as part of a legally-mandated process. But despite the fact that it had been more than a decade since the publication of Federal Guidance Report 11, the EPA chose not to revise the maximum contaminant levels (MCLs) for alpha-emitting, long-lived transuranic radionuclides in that review. The next scheduled review of radionuclide MCLs in drinking water will occur in 2006.

This report provides an analysis of the changes in the dose estimates to the maximally exposed organ that have occurred since the MCL limits for radionuclides were first set in 1976. It presents the scientific underpinning for tightening the MCL for alpha-emitting, long-lived transuranic radionuclides by a factor of one hundred compared to the present gross alpha MCL of 15 picocuries per liter (pCi/L).

1. Drinking water maximum contaminant limits for plutonium-239 and other alpha-emitting, long-lived transuranic radionuclides are about a hundred times too lax.

The most recent science, as published by the 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 alpha-emitting, 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, long-lived 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 United States 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.

Since the vast majority of public water systems have alpha-emitting, long-lived transuranic radionuclide levels orders of magnitude below the proposed MCLs (from weapons testing). 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 one-time 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 (see below), 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, Tennessee, and Washington state. Further, 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 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.

Recommendations

The EPA is going to review the radionuclide standards for drinking water as part of a scheduled process in 2006. We urge the EPA to revise the drinking water regulations in regard to alpha-emitting, long-lived transuranic radionuclides. The Department of Energy should evaluate its cleanup and decommissioning efforts with a view to meeting the tighter standard.

1. The 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, long-lived 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 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 independent 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 continue 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 off-site 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 model that is used to assess the impact of residual radioactivity on food, water, and the environment.

Argonne National Laboratory developed a "family" of 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 re-suspended soil. Its source code 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 a source code paid for by taxpayer dollars secret, particularly as billions of dollars are being spent on cleanup decisions based on the results generated by the RESRAD program.

I. Introduction

The National Primary Drinking Water Regulations specify rules that will protect drinking water and will maintain it in a state that is safe to drink. In these regulations, 40 CFR 141.66 sets safe drinking water standards for radionuclides in public water supplies under the Safe Drinking Water Act.[1] These standards are set in two ways: by specifying maximum contaminant levels of drinking water or by specifying maximum allowable dose to the whole body or any organ as a result of ingestion of drinking water. However, as demonstrated below, the concentration limits currently in effect for alpha-emitting transuranic radionuclides in drinking water are grossly inadequate to protect public health. Achievement of reductions in concentration is necessary to protect public health.

The current maximum contaminant level (MCL) as set forth in 40 CFR 141.66(c) for gross alpha particle activity, including radium-226, but excluding uranium and radon, is 15 picocuries per liter. 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,[2] then the rule allows a maximum contaminant level for gross alpha of 10 picocuries per liter. For instance, if plutonium-239 were the only alpha-emitting, long-lived transuranic radionuclide present, the MCL for plutonium-239 in this case would be 10 to 15 picocuries per liter, depending on the concentration of radium-226.

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 and Measurements (NCRP), a United States agency, and published as ICRP Publication 2 and in abbreviated form in the U.S. by the National Bureau of Standards as NBS Handbook 69.[3]

But the science has changed since then. As a result of these changes, as well as changes in the dose conversion factors adopted by the EPA since that time, dose estimates to the most exposed organ, while complex to assess, are far greater than those implied by the limit of 10 to 15 picocuries per liter when evaluated according to the methods specified in NBS 69.

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[1] The text now published under 40 CFR 141.66 were formerly published under 40 CFR 141.15 and 141.16. (CFR = Code of Federal Regulations). See also SDWA.

[2] This assumes that no radium-228 is present. The radium MCL in the rule is set for the combined concentration of Ra-226 and Ra-228. The former is an alpha-emitter and the latter is a beta-emitter. Hence the latter is omitted from the gross alpha part of the rule.

[3] ICRP-2, 1959 & NBS 69. 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 (NCRP). Tables and scientific discussion are drawn from ICRP-2, 1959. NBS Handbook 69 was published in 1959 and then again, with an added table and errata, in 1963. We cite NBS 69 throughout this report. 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.

It is therefore necessary that the MCLs of transuranics in drinking water be changed in order that the MCL remain within the spirit and framework of the standards as promulgated in 1976. This can be done based on the dose conversion factors that the EPA has since adopted and published in Federal Guidance Report 11,[4] which are the basis for present EPA regulation and risk estimation. They were published in 1988. The EPA has since published Federal Guidance Report 13. This is the most recent EPA scientific publication relevant to safe drinking water standards. The scientific basis of this guidance (ICRP 72)[5] has been adopted for some federal dose calculation purposes, but not yet sanctioned for use in regard to assessing doses from drinking water. In this report, we will consider the changes in the drinking water standards for alpha-emitting, long-lived transuranic radionuclides.

The basis for the needed MCL change is the potential danger that residual radioactive pollutants remaining after cleanup of the Cold War nuclear weapons production sites will pose to individuals in this generation and future generations. Of particular concern are the long-lived transuranic radionuclides neptunium-237, plutonium-238, plutonium-239, plutonium-240, plutonium-242, americium-241, and americium-243. All of these are man-made radionuclides.

II. National Primary Drinking Water Regulations - Radionuclides

In 1959, the National Bureau of Standards published its Handbook 69 (NBS 69), which established 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 most exposed organ, also called critical organ, for each pathway and solubility class.[6] As discussed below, a somewhat different method was used for bone-seeking radionuclides like radium-226 and plutonium-239. All these limits were established for radiation workers.[7]

ICRP 2 and NBS 69 also set forth the scientific approach for calculating these maximum permissible concentrations, with ICRP 2 providing significantly greater detail. A table adding data and correcting some errors in the 1959 version of NBS 69 was published in 1963, along with the original 1959 NBS 69 publication. In the text that follows, the term NBS 69 refers to this 1963 publication, since the EPA based its drinking water standards on it.

In March 1975, the EPA proposed, for the first time, National Primary Drinking Water Regulations for public water systems.[8] The proposed rules for radionuclides were published in August of that year.[9] The regulations for contaminants other than radionuclides were promulgated in December 1975;[10] the rules for radionuclides were promulgated in July 1976.[11] The MCLs and dose limits were

[4] FGR 11, 1988.
[5] ICRP-72, 1996.
[6] NBS 69.
[7] Until 1958 there were no separate radiation exposure limits for the public. They were the same as for workers. In 1958, the dose limits for the public were set at one-tenth the maximum allowable doses for workers (NBS 59 Addendum, page 5).
[8] Fed. Reg. 1975/03/14.
[9] Fed. Reg. 1975/08/14.
[10] Fed. Reg. 1975/12/24.
[11] Fed. Reg. 1976.

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.[12]

In the final rule of July 1976, the EPA promulgated Maximum Contaminant Levels (MCLs) for radionuclides in public water systems either by directly specifying the MCL values (in picocuries per liter) or by specifying dose limits, which implied MCLs for drinking water, based on an adult water intake of two liters per day. The science underlying the standards was published in NBS 69. The drinking water limit for alpha-emitting radionuclides excluding uranium and radon, but including radium-226, was set at 15 picocuries per liter. There was a separate sub-limit for radium-226 and radium-228 of 5 picocuries per liter. For beta and photon-emitters the dose limit was 4 millirem per year (mrem/year) to the most exposed organ. (For radionuclides that are approximately uniformly distributed in the body, such as cesium-137 and tritium, the most exposed organ is considered to be the whole body.) The MCLs for beta- and photon-emitters were set according to the 4 mrem/year criterion, with a slight variation from this being adopted for tritium and for strontium-90. The limits for these categories have remained the same since that time.[13] Detection limits and analytical methods for radionuclides were set forth in 40 CFR 141.25.

The rule as originally promulgated discusses natural and man-made radionuclides separately. However, it does not explicitly discuss the alpha-emitting transuranic radionuclides that are the subject of this report, but specifies only a gross alpha MCL. The gross alpha limit excludes only uranium and radon and it automatically includes the alpha-emitting, long-lived transuranic radionuclides of concern here, as these radionuclides are explicitly listed in the tables in NBS 69.

The following statement indicates the intent of the regulation that first established maximum contaminant limits for man-made radionuclides in drinking water:

> 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-*

[12] The changed numbering can be found in the 2004 edition of 40 CFR 141.

[13] The limits were first specified in 40 CFR 141.15 and 40 CFR 141.16. An MCL for uranium of 30 micrograms per liter was established on December 7, 2000, in 40 CFR 141.66 (e), based mainly on the heavy metal toxicity of uranium to the kidney. The revision to 40 CFR 141 was announced in Fed. Reg. 2000.

> *water will normally be small, the Agency believes that the risk of future contamination 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.*[14]

While this passage does not explicitly mention nuclear-weapons-related activities and facilities, their inclusion is clearly indicated, notably from the fact that fallout from nuclear weapons testing is discussed as the most important source of surface water contamination. It is also clear from the discussion of fallout that the intent was to consider the most important sources of contamination. The mention of industrial users also does not exclude weapons facilities (which handle radioactivity in considerably smaller amounts when compared to reactor core and spent fuel inventories in the commercial nuclear power sector). It is implicit, therefore, that there was no intent to exclude alpha-emitting man-made radionuclides from the vigilance and concern of the regulations.

The understanding of what is the most exposed organ for alpha-emitting, long-lived transuranic radionuclides has evolved.

The level of doses at which concern and vigilance were warranted in regard to man-made radionuclides was a few millirem per year. 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…[15]

This quote shows that the sum of the doses from military and civilian activities was considered in evaluating the limit of 4 millirem per year that was set for beta- and photon-emitters in 1976. In fact, fallout was the single most important component of the dose from man-made radionuclides evaluated by the EPA.

The cancer fatalities from whole body exposure to 4 millirem per year from man-made beta and photon sources of radioactivity were estimated at between 0.4 and 2.0 deaths per year per million people exposed. This was comparable to the exposure to natural radium-226 and radium-228 estimated at 0.7 to 3 fatal cancers per year per million persons at the level of 5 picocuries per liter selected as the maximum contaminant level. The slightly higher fatality rate for radium (a factor of 1.2 to 1.8) at the allowable limit of 5 picocuries per liter must be seen in the context that it is a ubiquitous, naturally occurring radionuclide, with considerable variation in drinking water concentrations (which the EPA estimated at the time to be between 0.1 and 60 picocuries per liter).[16] The EPA imposed considerable costs on public water systems by requiring remediation of those systems that had levels of radium greater than 5 picocuries per liter in order to bring them to the

[14] Fed. Reg. 1975/08/14, page 34324, emphasis added.
[15] Fed. Reg. 1975/08/14, page 34325, emphasis added.
[16] Fed. Reg. 1975/08/14, page 34325.

regulatory level. Further, the EPA mandated testing of water supplies and established detection limits (at the 95 percent confidence limit) that were considerably below the MCLs set forth in the regulation.[17] The detection limits were set in order to ensure that the mandated MCLs would not be exceeded. In considering the mandated MCLs and detection limits, the EPA took technical, health, and economic considerations into account.

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 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[18]:

> 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 man-made radioactivity will be exceeded except in extraordinary circumstances.[19]

There is no explicit exclusion of alpha-emitting transuranic radionuclides from this statement. Also, the National Primary Drinking Water regulations explicitly mention strontium-90 in fallout. Hence, the regulations explicitly took into account a man-made radionuclide from a military activity – nuclear weapons testing – in protecting public water supplies from radioactive contaminants. Further, the critical organ listed in NBS 69 for strontium-90 and for the transuranic radionuclides that are the subject of this report was the same – the bone.

The language of the regulation indicates that the MCL in the range of 10 to 15 picocuries per liter for the alpha-emitting, long-lived transuranic radionuclides set at the time would have corresponded approximately to a bone dose of a few millirem per year according to then-prevailing estimation methods. We show in the next section, *A. Bone dose estimation in ICRP 2*, that was indeed the case. However, present-day methods result in far higher dose estimates, as discussed below in the section after next, *B. Bone dose estimation, present-day dose conversion factors*.

A. Bone dose estimation in ICRP 2

Bone dose was estimated in ICRP 2 (and NBS 69) as dose to the skeletal bone without the marrow. The reference bone-seeking radionuclide used by ICRP 2/NBS 69 was radium-226 and the reference amount was 0.1 microcurie of radium-226 in the skeletal bone. The amount of energy deposited in the bone each year corresponded to an absorbed radiation dose rate of about 3 rad per year, not accounting for relative biological effectiveness (RBE) of alpha particles. ICRP 2 used an RBE = 10, thus yielding an annual dose for a 0.1 microcurie body burden of radium-226 of 30 rem per year,

[17] Fed. Reg. 1976, page 28404.
[18] Of the nuclear weapons states, only China was testing in the atmosphere at the time. China conducted its last atmospheric nuclear test in 1980.
[19] Fed. Reg. 1975/08/14, pages 34325-34326, emphasis added.

according to the then-prevailing method of estimation.[20] Doses were calculated by estimating a whole-body or organ burden of the radionuclide assuming lifetime ingestion or inhalation at the MCL, for which values were given either in the workplace (40-hour workweek) or continuously (168 hours per week).

Some radionuclides, such the beta-particle-emitting strontium isotopes, were recognized even then to behave somewhat differently than radium-226 in the body in that they tended to concentrate in certain parts of the bone, while radium-226 is distributed less unevenly. Research since that time has validated that observation. For instance, the alpha-emitting, long-lived transuranic radionuclides tend to concentrate adjacent to the endosteal cells on the bone surface. Hence, these radionuclides deliver a considerably higher dose to the endosteal cells than would be indicated by an assumption of uniform distribution over a marrow-free skeleton.

In order to account for non-uniform distribution of several bone-seeking radionuclides, ICRP 2 suggested (and used) a factor of safety of 5 for such radionuclides when estimating maximum permissible levels of radionuclides in air and water for workers.[21] The effect of this safety factor was to reduce the maximum allowable dose for workers from alpha-emitting, long-lived transuranic radionuclides to 6 rem per year, compared to 30 rem per year for radium-226. Correspondingly, the maximum permissible concentrations were also reduced by a factor of five.

This intent to reduce the maximum permissible dose to the bone by a factor of about 5 can be confirmed by estimating the dose corresponding to the maximum permissible burden of plutonium-239 in the bone of 0.04 microcuries specified in NBS 69. Using a value of 5.15 MeV per alpha particle and an RBE = 10, the annual dose corresponding to a bone burden of 0.04 microcuries of plutonium-239 is about 5.5 rem per year. Since the whole body and organ burdens in NBS 69 are rounded, this is in close agreement with the figure of 6 rem inferred by applying the safety factor of 5 to the radium-226 dose of 30 rem.

The MCL for soluble plutonium-239 set in NBS 69 corresponding to the 6 rem per year bone dose would be 5×10^{-5} μCi/cc, or 5×10^{-2} μCi/liter, or 50,000 pCi/liter. The current drinking water limit of 15 picocuries per liter in the absence of radium-226 corresponds to a bone dose of about 1.8 millirem per year (or 1.2 millirem per year corresponding to 10 picocuries per liter, which is the MCL for plutonium-239 in the presence of radium-226 at its MCL of 5 picocuries per liter).[22]

The bone doses corresponding to 15 picocuries per liter for various alpha-emitting, long-lived transuranic radionuclides are shown in Table 1, estimated according to the method in NBS 69 which was the prevailing scientific understanding in 1976, when the EPA first promulgated the MCLs for radionuclides. All of these calculations follow NBS 69 in assuming soluble radionuclides when estimating doses to the bone from drinking water. An assumption of soluble forms of the radionuclides is reasonable (and in keeping with the regulation as originally promulgated) since it is likely that the radionuclides will be in that form if they are present in drinking water. The presence of insoluble colloidal forms is not excluded, but the likely presence of soluble forms makes it necessary to use the uptake coefficient for that form, which has been done throughout this report.

[20] ICRP-2, 1959, page 13 and FGR 11, 1988, page 18. The current value of the RBE, often called the quality factor in the regulatory context, for alpha particles is 20.
[21] FGR 11, 1988, pages 16-19.
[22] This assumes that no Ra-228 is present.

Table 1: Bone dose from alpha-emitting, long-lived transuranic radionuclides according to NBS 69 (ICRP 2)

Radionuclide	Bone dose at 15 pCi/L in mrem/y
plutonium-238	1.8
plutonium-239	1.8
plutonium-240	1.8
americium-241	1.8
neptunium-237	3.0

Note: These doses are estimated by proportionally reducing the doses for these radionuclides corresponding to the MCLs listed in NBS 69, which correspond to a bone dose of 6 rem per year. The figure of 6 rem for bone dose for alpha-emitting, long-lived transuranic radionuclides is derived by applying the safety factor of 5 to the bone dose of 30 rem for radium-226 (see text). NBS 69 lists the kidney as well as bone as the target organs for americium-241. We consider only bone-dose-related MCLs in this report. Plutonium-242 dose is the same as plutonium-239.

The NBS 69 (ICRP 2) calculations for bone dose are not directly comparable to present-day methods of dose estimation. NBS 69 specifies annual doses to the "bone," defined as the marrow-free skeleton. But Federal Guidance Report 11, which lays out methods of dose estimation that are the basis of EPA regulations at the present time, defines committed doses to two different parts of the bone – the "red marrow" and the "bone surface."[23] The latter is defined as the most exposed organ in Federal Guidance Report 11 for alpha-emitting, long-lived transuranic radionuclides because they concentrate adjacent to the endosteal cells, which are located on the bone surface. In other words, the understanding of what is the most exposed organ for alpha-emitting, long-lived transuranic radionuclides has evolved along with the methods of dose estimation since the MCLs were promulgated in 1976.

As shown in Table 1, the range of doses to the bone using a limit of 15 picocuries per liter for alpha-emitting, long-lived transuranic radionuclides estimated according to NBS 69 is approximately from 1.8 to 3 millirem per year. This is about the same as the doses estimated from man-made radionuclides, notably in fallout, in the safe drinking water regulation as promulgated in 1976. Hence we can infer that the intent of the rule was to limit the dose from drinking water to the maximum exposed organ, defined then as the bone, to approximately 2 millirem per year.

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 plutonium-239, the annual dose to the endosteal cells would be about 26 millirem per year.[24] The bone surface dose for the other radionuclides shown in Table 1 are about the

[23] There is more recent federal guidance on the subject in *Cancer Risk Coefficients for Environmental Exposure to Radionuclides*, Federal Guidance Report No. 13. Washington, D.C., Environmental Protection Agency, 1999 (hereafter cited as FGR 13). This report also uses the same two parts of the bone as the target organs for which doses are calculated.

[24] This estimate is derived by using a mass of 120 grams for the endosteal cells corresponding to an overall skeletal mass of 7,000 grams. Further, it is assumed that one-fourth of the energy is deposited in the 120-gram mass of the endosteal cells, with the rest being deposited in other parts of the bone. This mass of the endosteal cells is specified in Federal Guidance Report 11. This gives a ratio of dose to endosteal cells of (7000/120)*0.25 = 14.6. All calculations assume that the dose to the bone permitted under NBS 69 at the specified MCL was 6 rem per year. There is some imprecision

same, except for Np-237, for which the figure is about 44 mrem per year. These estimated doses, which take into account the evolution of scientific understanding in the years after 1976, are far higher than what the safe drinking water regulations allow. The implied dose to the endosteal cells is about a factor of 14.6 higher for plutonium-239. All of these calculations were done within the framework of NBS 69, which was (and continues to be) the scientific guidance for the safe drinking water regulation.

B. Bone dose estimation, present-day dose conversion factors

Scientific understanding of radiation doses and harm from intake of radionuclides has advanced considerably over the years. Regulations have also evolved to some extent, though at a slower pace. Specifically, in the 1970s, the International Commission on Radiological Protection (ICRP) published ICRP 26 and ICRP 30 followed by ICRP 48 in 1986. The scientific work in these publications was incorporated by the EPA into Federal Guidance Report 11 in 1988. The doses from alpha-emitting, long-lived transuranic radionuclides in the new guidance issued by the EPA are much higher than those estimated by NBS 69 methods. Federal Guidance Report 11 is the report that is the basis of current EPA regulatory dose estimation methods. We will estimate bone doses according to Federal Guidance Report 11 (FGR 11) in this section. Then we discuss the same problem using Federal Guidance Report 13 (FGR 13), which is the most recent EPA Guidance, but not yet in force for regulatory calculations for doses from air and water.

1. Bone doses according to FGR 11

As touched upon above, several major changes have transpired from NBS 69 to FGR 11 so far as this analysis is concerned:

- The quality factor, or RBE, was increased from 10 to 20.
- The bone was divided into two different target organs, the "bone marrow" and the "bone surface," as compared to a single organ, the marrow-free skeleton, in NBS 69.
- The division of the bone into two organs in FGR 11 allowed the omission of the safety factor of 5 that was used in NBS 69 to account for selective, non-uniform deposition in the bone of certain radionuclides.
- NBS 69 used annual doses, while FGR 11 provides the conversion factors for committed doses. [25]

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associated with the fact that the MCLs were rounded to one significant figure in NBS 69, but this is not significant in the present context.

[25] "Annual dose" corresponds to the amount of energy from ionizing radiation deposited in the target organ per unit mass of the organ in a single year. The dose in rem is then calculated by applying the RBE to the deposited energy. "Annual committed dose" corresponds to the amount of energy that would be deposited in the organ over the entire time that the radionuclide is present in the organ due to the intake of the radionuclide in a single year. If a radionuclide is eliminated rapidly from the body (say in a few days or weeks), as for instance is the case with tritium, 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 (the latter is the case for alpha-emitting, long-lived transuranic radionuclides, their target organ being the bone), the dose to the bone from an intake in any given year is delivered over a period of decades after that. With the annual committed dose, the intake is over a year but the dose is delivered over a different period of time – and, in the case of alpha-emitting, long-lived transuranic radionuclides to the bone, a much longer period of time. Hence, the actual dose delivered to the person in the case of an intake of an alpha-emitting, long-lived transuranic radionuclide late in life (say a

While these technical changes are complex, it is possible to estimate the effect of the changes from NBS 69 to Federal Guidance Report 11 on doses in several different ways, each of which raises some technical issues. The approaches and issues are set forth in Table 2 using plutonium-239 as the reference alpha-TRU radionuclide.

Table 2: Approaches for deriving an updated drinking water limit for plutonium-239 that account for changes from NBS 69 to FGR 11

Approach	Issues	Derived, updated Pu-239 MCL, pCi/liter
1. Compare the NBS 69 annual bone dose to the FGR 11 bone surface annual committed dose	**Advantage**: Uses the prevailing dose framework at the time. **Disadvantages**: (i) For alpha-emitting, long-lived transuranic radionuclides, which have a long biological half-life, committed dose is not equivalent to annual dose. The actual cumulative dose over a lifetime is considerably less than the product of the years and the annual committed dose. (ii) Target organ is different – bone for NBS 69 and bone surface for FGR 11.	0.04
2. Compare NBS 69 cumulative bone dose over a lifetime at 15 pCi/L to actual cumulative bone surface dose estimated from FGR 11	**Advantage**: Closest to the intent of the regulation to limit doses to the most exposed organ. **Disadvantage**: Changes the target organ from marrow-free skeleton to bone surface.	0.08
3. Compare cumulative bone surface dose imputed from NBS 69 to bone surface dose as per FGR 11	**Advantage**: Compares the same target organ. **Disadvantage**: Changes the framework from maximally exposed organ, as defined at the time by prevailing science, to comparing bone surface dose, which was not explicitly defined in NBS 69.	1.2

Notes: For Pu-239, it is assumed that 63 percent of the committed dose is delivered in 50 years. The values in the last two rows correspond to a 70-year intake. The estimate in Federal Guidance Report 11 for bone "surface seeking alpha-emitters" is a factor of 12, but a value for Pu-239 is not specified. We estimate the ratio of cumulative bone surface dose from FGR 11 to NBS 69 for Pu-239 is a factor of 12.3, which is about the same as the value in FGR 11. This validates the approach used for the calculations in the last row of the above table.

Of these approaches, the first one is the least persuasive scientifically because it compares cumulative annual doses to cumulative committed doses. Since plutonium is eliminated from the bone very

few years before death) is less than the full committed dose and less than the dose that would be delivered from the same intake early in life.

slowly (with a biological lifetime of several decades), most of the dose from intakes in the last years of a 70-year reference lifetime would be delivered after the full lifetime of even a long-lived person (even if one considers a ~100 year life, for instance). Hence, only the latter two approaches are scientifically reasonable. Both yield values for MCLs for alpha-emitting, long-lived transuranic radionuclides that are far below 15 picocuries per liter. However, they yield values also an order of magnitude different from each other – 0.08 picocuries per liter and 1.2 picocuries per liter. The approach shown in the second row is the most close to the intent of the drinking water regulation because it compares cumulative dose over a lifetime to the most exposed organ as defined in 1976 (marrow-free skeleton) and the most exposed organ as currently defined (bone surface). The last approach compares dose to the same organ (bone surface), which has scientific merit. However, it is not in accord with the intent of the regulation to limit dose in that the prevailing views of the most exposed organ (marrow-free skeleton in 1976 and bone surface in 1988) are no longer being compared. Hence, the most appropriate value to use for a new standard based on Federal Guidance Report 11 would be 0.08 picocuries per liter. However, since this is no longer the most recent scientific guidance published by the EPA, this factor would also need to be considered in the review of MCLs for alpha-emitting, long-lived transuranic radionuclides when they are reviewed in 2006.

2. Bone doses according to FGR 13

The most recent regulatory guidance for estimating doses is based on dose conversion factors published in ICRP 72. These have been incorporated into Federal Guidance Report 13, including the compact disk supplement, which has dose conversion factors for various ages published in a database.[26] The dose conversion factors are age-dependent and can be used to estimate committed doses for the remainder of life from the age of intake to age 70 years. This allows the estimation of total dose over a lifetime corresponding to a water contamination at 15 picocuries per liter.

The dose conversion factors in Federal Guidance Report 13 are generally somewhat lower than those in Federal Guidance Report 11. Therefore the total dose to the bone surface using the newer dose conversion factors in Federal Guidance Report 13 is roughly a factor of two lower than that estimated using FGR 11. In addition to the change in the dose conversion factors, water intake variation with age also needs to be considered. The current drinking water MCLs are based on an adult intake of 2 liters of water per day, excluding the water content of food. However, the water intake of children is smaller and there is also some gender variation. Further, children have a greater intake of fluids, notably in the form of milk. Therefore, we have done the Federal Guidance Report 13-based dose calculation using two sets of intake rates for various ages that are published in the literature. The first set corresponds to fluid intakes, including milk. The second set includes only water intake. These assumptions about intake rates are show in Table 3 below:

[26] FGR 13, 1999 and 2002 (the latter for the CD supplement, rev.1).

Table 3: Drinking water assumptions for FGR 13 dose calculations

Age range, years	Fluid intake, including milk, liters/day (Case 1)	Water only intake, liters/day (Case 2)
0 to 4	1.3	0.7
5 to 14	1.3	0.95
15 to 70	1.95	1.65

Note: For Case 1, the main reference is ICRP 23, 1975. The fluid intake rate of 1.4 liters per day for 10 year-olds has been changed here to 1.3 liters per day for ages 0 to 14 years. For Case 2 the main reference is Smith and Jones 2003, which provides the most recent recommendations of the British National Radiological Protection Board.

When total fluid intake is considered (i.e., Case 1 above) the cumulative lifetime dose to the bone surface from plutonium-239 over a 70-year period is about 15,500 mrem. For Case 2, water intake only, the lifetime bone surface dose is about 12,000 mrem. The corresponding dose to the maximally exposed organ under NBS 69 (the marrow-free skeleton) is 126 mrem. These doses are calculated by applying dose conversion factors specified in the relevant publications to the intake of plutonium in drinking water over a 70-year period. This last figure of 126 mrem can be viewed as the intent of the original regulation in terms of the dose to the maximally exposed organ from drinking water contaminated with plutonium to the maximum allowable limit of 15 picocuries per liter. If we compare the value of 126 mrem to the dose to the maximally exposed organ as estimated by the methods specified in Federal Guidance Report 13, we find that for drinking water intakes corresponding to Case 1, the MCL of 15 picocuries per liter is about 123 times too high and for Case 2, it is about 95 times too high. Therefore the most recent science would indicate a tightening of the current MCL for plutonium-239 (15 pCi/L) by about 123 times to about 0.122 picocuries per liter in the case of fluid intake case (Case 1) and by over 95 times to about 0.157 picocuries per liter for water intake only (Case 2). The results for the other alpha-emitting, long-lived transuranic radionuclides are similar, since the dose conversion factors are quite close to those of plutonium-239, with the exception of neptunium-237, for which the dose conversion factors are about a factor of two lower.

III. Conclusions

The analysis in this report shows that 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. A combined standard for all alpha-emitting, long-lived 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 DOE waste 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.[27]

The MCL for alpha-emitting, long-lived transuranic radionuclides should be reduced from 15 picocuries per liter to 0.15 picocuries per liter.

In considering what should be the optimal value for a drinking water standard for alpha-emitting, long-lived transuranic radionuclides radionuclides, we have also examined the values for a plutonium-239 limit that exists in other standards. Specifically, 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, namely, the Rocky Flats Plant, near Denver. The statewide standard for plutonium-239 for surface water is 0.15 picocuries per liter.[28] 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 2 liters of water per day for 30 years.[29]

The Colorado Department of Health, Water Quality Control Commission describes the background and the rationale for changing from 15 picocuries per liter to 0.15 picocuries per liter 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

[27] 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.

[28] Colorado Reg. 31, 2005. The State 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 and others of which result in more lax rules. We have simply used the State of Colorado's statewide surface water limit for Pu-239 as a guide for reference.

[29] CDPHE 2002.

> 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 1×10^{-6} risk level, based on residential use. This risk level is consistent with the Commission's policy for human health protection.[30]

This reasoning is based on CERCLA, the Superfund law, but is qualitatively in accord with the reasoning in this analysis. Specifically, 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. Further, the specific value for plutonium and americium recommended in the Colorado rule is just a factor of two lower than the geometric mean of the two values in the last two rows of Table 2 above.

In view of the complexities created by the change from NBS 69 to Federal Guidance Report 13, an MCL for alpha-emitting, long-lived transuranic radionuclides of 0.15 picocuries per liter is reasonable and justifiable. The action we are recommending is consistent with the intent of the National Primary Drinking Water Regulations as originally promulgated and is directly within the framework of the regulation as promulgated then and as it stands at present.

The primacy of the health goal (rather than numerical limits) is clear from the EPA's own description of the Safe Drinking Water Act, pursuant to which the radionuclide maximum contaminant limits are set. Its fact sheet on the Act states:

> 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.[31]

[30] Colorado Reg. 31, 2005, pages 138-139.
[31] EPA 2004.

By this standard, 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, as shown above, not in accord with the intent of the initial regulation. Because of this, the EPA should take up consideration of a tightened standard in its upcoming 2006 drinking water radionuclide review.

The 15 pCi/L limit for transuranic radionuclides is obsolete, not protective of public health, against the spirit of the Safe Drinking Water Act, and, as shown above, not in accord with the intent of the initial regulation.

Corresponding to the change in the MCL for alpha-emitting, long-lived transuranic radionuclides, there is also a need for a change in the detection limit. Table B in 40 CFR 141.25 should be modified to include a separate detection limit of 0.01 picocuries per liter for each alpha-emitting, long-lived transuranic radionuclide. This detection limit is well within the capabilities of present-day techniques. The current detection limit for these radionuclides is 0.001 picocuries per liter, according to Argonne National Laboratory. The errors at such low levels can be large however. The error at 0.01 picocuries per liter, the recommended detection limit, is estimated by Argonne National Laboratory to be 10 percent.[32]

We recognize that alpha-emitting, long-lived transuranic radionuclides are not ubiquitous in significant concentrations, unlike naturally occurring radionuclides like radium-226, thorium-232, and thorium-230. The vast majority of public water systems can therefore be exempted from routine monitoring requirements relating to alpha-emitting, long-lived transuranic 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 (see below).[33] Wastes disposed of at shallow and intermediate depths are included in this 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.

We recognize that the main recommendation of this report, to set a separate standard for alpha-emitting, long-lived transuranic radionuclides, requires that the present gross alpha limit be split up into two parts – one for alpha-emitting, long-lived transuranic radionuclides and the other for naturally occurring alpha-emitting radionuclides. However, this is not a departure from the content or intent of the present rule, for several reasons.

First, the present rule itself does not have a single standard for alpha-emitting radionuclides. There is a sub-limit for radium-226 and radium-228 of 5 picocuries per liter. Since radium-226 is an alpha emitter, there is in effect a separate sub-limit for an alpha emitter up to maximum of 5 picocuries per liter (depending on how much radium-228, a beta-emitter, is also present). Second, the gross alpha

[32] ANL 1995, Chapter 7, Table 7.1.

[33] For instance, the 100 curie limit is equivalent to 1,000 metric tons 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.

limit excludes uranium and radon. The limit of 30 micrograms per liter of uranium is set on the basis of heavy metal toxicity. However, this amount of uranium causes some amount of harm as a result of its radioactivity. Recent science indicates that the harm from the heavy metal aspects of uranium may be reinforced by its radioactivity. (See *Section VI. Other risks and radionuclides*, below). Hence, reconsideration of a variety of issues is warranted. In such reconsideration, it would be practical and less costly to separate out alpha-emitting, long-lived transuranic radionuclides. This is because the vast majority of water systems will not require any testing for alpha-emitting, long-lived transuranic radionuclides since they are not at risk.

IV. Costs

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

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

Since no known contamination of public water systems above 0.15 picocuries per liter of alpha-emitting, long-lived transuranic radionuclides exists, 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 Department of Energy, 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 science-based drinking water standard that corresponds to current understanding of plutonium movement and irradiation of the human body. Finally, some remediation actions could, in the long run, pollute the water above drinking water standards, and worse, be irremediable. No known technology could remediate vast bodies of water such as the Savannah

River or the Snake River Plan Aquifer if, once polluted, the aim is to reduce pollution from a few picocuries per liter to sub-picocurie per liter levels.

V. Estimating the impact of residual radioactivity

Vast areas of land and huge amounts of water remain contaminated with dangerous long-lived radionuclides from operations of nuclear weapons facilities.[34] 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.[35] 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. 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. DOE is grouting large amounts of plutonium in the tanks even though it has not yet obtained convincing evidence of the durability of grout. The tanks are buried underground in the watershed of the Savannah River, one of the most important rivers in the South Carolina-Georgia region. Experimental and field data leave room for considerable skepticism as to its performance. IEER's evaluation of the state of the research on grout indicates that the performance of grout remains highly uncertain. There is at present no sound basis, whether in experiment or in field data, to assume that leaving large amounts of grouted alpha-emitting, long-lived transuranic radionuclides in the tanks would be protective of the Savannah River.[36]

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 in waste so reclassified.[37] 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.

The consequences of the DOE cleanup policy on the concentrations of residual transuranic contamination in the soil and their potential effect on the health of individuals are discussed in a study by IEER entitled *Setting Cleanup Standards to Protect Future Generations: The Scientific*

[34] OTA 1991.
[35] Nichols 1996.
[36] Smith 2004 and Makhijani and Boyd, 2004.
[37] PL 108-375, 2004, Sec. 3116.

Basis of the Subsistence Farmer Scenario and Its Application to the Estimation of Radionuclide Soil Action Levels (RSALs) for Rocky Flats, December 2001.[38] In this study, IEER showed that the specific assumptions about future use have a major impact on what are considered acceptable residual radioactivity levels. A large part of this result is because different future site use scenarios have different assumptions about the use of water and food from the contaminated area in question. Since some radionuclides, including the alpha-emitting, long-lived transuranic radionuclides discussed in this report, are very long-lived, a basic assumption that there will be loss of institutional control over the long-term is essential to sound planning and cleanup.

However, even the adoption of a subsistence farmer scenario as the basis for cleanup cannot assure that levels for residual radioactivity on contaminated sites will be set in a manner that is protective of health and the environment. This is because the translation of residual levels into radiation dose and risk estimates requires the use of complex models and assumptions about the behavior of radionuclides in the environment. For instance, the amount of rainfall, the mobility of radionuclides in specific soil conditions, the porosity of the soil, the solubility of the radionuclides under various circumstances, and the rate of soil erosion are among the critical parameters that need to be known and characterized.

At present, remediation levels are typically assessed by the use of a model developed by Argonne National Laboratory called RESRAD (for residual radioactivity).[39] This computer code is complex and has, over the years, been developed to consider pathways for movement of radioactivity in a sophisticated way. Yet, it does not contain libraries of dose conversion factors for, and thus does not account for, infants or for young people at sensitive times in their hormonal development or for the fetus at various stages of fetal development. The estimation of doses to various segments of the population at sensitive periods in their lives may also require consideration of how the environmental pathways and the systems in the human body are represented in the model's source code.

The RESRAD source code is closely held by the U.S. government; it is not public. Ostensibly, the official rationale is that since RESRAD is used for regulatory decisions, such as those that are made in the context of cleanup at nuclear weapons sites, it should not be made public. However, we do not accept this rationale. The code can be made public and can be an open source code, available for modification in the same manner as the Linux operating system source code. That has resulted in its improvement and efficiency, without problems actually creeping into mass use of the code as an operating system. The U.S. government can surely retain its version of the code for regulatory purposes while making the source code publicly available for examination and improvement. If at a certain stage, the code is improved in a manner that regulatory bodies such as the EPA consider it useful for regulatory purposes, they will freely be able to adopt the changes but will be under no obligation to so.

[38] Makhijani and Gopal 2001.
[39] RESRAD.

VI. Other risks and radionuclides

New scientific work on radiation protection 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. However, these are still emerging areas of concern, where the risks are not quantitatively well established. How such risks are to be considered in the context of a review of drinking water MCLs will be considered in a future IEER report.

Recent developments in radiobiology and health effects research have increased understanding of radiation doses during fetal development. They indicate that non-cancer health effects resulting from fetal exposure to radiation could be very important. For instance, ICRP 90 emphasizes that the central nervous system is especially vulnerable during a certain period of fetal development:

> …[B]iological systems with a high fraction of proliferating cells show high radiation responsiveness. High rates of cell proliferation are found throughout prenatal development….Development of the central nervous system starts during the first weeks of embryonic development and continues through the early postnatal period. Thus development of the central nervous system occurs over a very long period, during which it is especially vulnerable. It has been found that the development of this system is very frequently disturbed by ionising radiation, so special emphasis has to be given to these biological processes.[40]

A variety of end points (disease outcomes) are at issue, from central nervous system development to cancer to birth defects to increased risk of miscarriages. Further, these end points raise the issue of the combined effects of other pollutants with radiation more insistently that ever before. For instance, one might ask about the potential for non-linear effects caused by exposure to both lead and radiation or mercury and radiation. One might also ask about the combined effects of exposure to endocrine disrupting chemicals and radiation in relation to a number of end points. These are areas still in a relatively early stage in the science compared to the understanding of radiogenic cancer induction. For these areas, which concern non-cancer end points as a result of fetal exposure, for instance, the conversion of the scientific data in publications such as ICRP 88 and ICRP 90 into regulations for health and environmental protection will take considerable time.[41] The EPA has not even published the necessary guidance documents as yet.

Recent research, much of it done at the Armed Forces Radiobiology Research Institute, pursuant to concerns about the health effects of depleted uranium, points to a surprising variety of harmful health effects of uranium. A recent literature survey by IEER summarized the situation as follows:

> The understanding of the risks of cancer due to radiation exposure from depleted uranium and kidney damage due to its heavy metal properties has expanded greatly in recent years. In addition, evidence is amassing that raises serious concerns regarding the impact of

[40] ICRP-90, 2003, page 9.
[41] ICRP-88, 2002; ICRP-90, 2003.

> chronic exposure to DU in relation to a number of other health issues. Studies in humans and animals have shown that uranium can concentrate in the skeleton, liver, kidneys, testes, and brain. In addition, rats implanted with DU pellets have also shown uranium concentrating in the heart, lung tissue, ovaries, and lymph nodes among other tissues. Research, primarily but not exclusively conducted since the 1991 Gulf War, indicates that exposure to uranium may be
>
> Mutagenic
> Cytotoxic
> Tumorigenic
> Teratogenic
> and Neurotoxic, including in a manner analogous to exposure to lead.
>
> Additionally…some research has also provided indications that there may be a synergistic effect between the heavy metal aspect of exposure to uranium and its radioactive effects….Current research conducted at the Armed Forces Radiobiology Research Institute (AFRRI) indicates that "[i]n the case of DU, cells not traversed by an alpha particle may be vulnerable to radiation-induced effects as well as chemically-induced effects." Additional work at the AFRRI has also shown that depleted uranium can cause oxidative DNA damage and thus provides the first indication that uranium's radiological and chemical affects might potentially play both a tumor initiating and a tumor promoting role. [42]

In other words, uranium may be a kind of radioactive lead, with serious health effects arising both from its heavy metal toxicity and its radioactivity. Should these risks be proven to be substantial, there may be a need to include new limits in the National Primary Drinking Water Regulations relating to the combined radioactive and heavy metal toxic effects of uranium.

There are also a variety of other issues associated with the potential interaction of hormonally active chemicals with radiation, and particular certain radionuclides, like iodine-129, which concentrates in the thyroid and crosses the placenta. The development of certain cancers, like breast cancer, is linked to hormonal systems, possibly to hormonally active chemical pollutants, and to radiation. Hence the issues associated with health protection in regard to certain cancers are likely to be much more complex.

Finally, there are issues that were once recognized but that appear to have been forgotten or ignored in the context of protection of public health from radiation. Consider the following passage from ICRP 2 that occurs in the context of a discussion of bone doses and the calculations that are the subject of this report:

> Certainly, if a major portion of the hematopoietic system were irradiated, e.g., concurrently from the spleen-seeking Po^{210} and from the bone-seeking Ra^{226}, the biological damage would be greater than if only a part of it were irradiated. *It has been shown that in some cases a synergistic effect results when several organs of the body are irradiated simultaneously*.[43]

Some of these synergistic effects are already implicit in the estimates of risk made from Hiroshima/Nagasaki survivors (since they received whole body radiation – i.e., all organs were

[42] Makhijani and Smith 2005, pages 9-10. Typos corrected.
[43] ICRP-2, 1959, page 14, emphasis added.

irradiated). However, others involving internal deposition and that selectively target certain organs may have more complex effects. This indicates that it is important to maintain regulations in the form of dose limits to maximally exposed organs in regulations relating to protection of public health, such as the National Primary Drinking Water Regulations (40 CFR 141), *Environmental Radiation Protection Standards for Nuclear Power Operations* (40 CFR 190), and *Environmental Radiation Protection Standards For Management And Disposal Of Spent Nuclear Fuel, High-Level And Transuranic Radioactive Wastes* (40 CFR 191). At the present time, there is still a significant amount of scientific work that remains to be done in a variety of areas before this framework can be changed into a better one from the point of view of health, environment, future generations, and the economy.

Consideration of changes in radiation protection in the medium- and long-term, that would take into account emerging scientific and risk issues such as those discussed in this section, is needed for a variety of reasons, some of which are mentioned above. However, this will be a complex and difficult task which must be done with due deliberation. It will also likely go far beyond safe drinking water standards. At the present time, the safety and protection of water resources from irreversible contamination with alpha-emitting, long-lived transuranic radionuclides as a result of ongoing activities by the Department of Energy cannot be allowed to be deferred to the longer, more comprehensive social, economic, and health discussion related to the protection of health from radioactive and toxic pollution. It must be considered as part of the EPA's 2006 review of standards for radionuclides in drinking water. A maximum contaminant level for plutonium that is 100 times too lax based on the intent and letter of the Safe Drinking Water Act must not be allowed to persist.

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