Health-Protective Considerations Regarding Measurement of Gross Alpha Particle Activity in Drinking Water

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SUMMARY

The Office of Environmental Health Hazard Assessment (OEHHA) has examined the practicality of proposing a gross alpha particle Public Health Goal (PHG). Health and Safety Code Section 116365(c)(1) directs OEHHA to "prepare and publish an assessment of the risks to public health posed by each contaminant for which the department proposes a primary drinking water standard." The risk assessment must contain "an estimate of the level of the contaminant in drinking water that is not anticipated to cause or contribute to adverse health effects, or that does not pose any significant risk to health. This level shall be known as the public health goal for the contaminant."

OEHHA has concluded that it would not be practical to develop a PHG for the category of alpha emitters, despite the fact that this category has a primary standard, or maximum contaminant level (MCL), which is currently set at 15 pCi/L under both federal and California law. There are several reasons for this conclusion; the most important is that the designation of gross alpha does not refer to a specific chemical contaminant, but rather to a group of radioactive elements. Furthermore, the MCL represents only a screening level for assay of radioactivity produced by alpha emitters. If excessive activity levels are found, further sample characterization is required, and selected alpha emitters are subject to regulation under specific MCLs. OEHHA is developing PHGs for the selected isotopes for which there are MCLs.

The use and choice of a screening level is in this case a risk management decision, based on consideration of cost and feasibility, and therefore to develop a PHG, based exclusively on health-based considerations, for the gross alpha measure appears to be inappropriate. OEHHA believes it is useful, however, to describe the risk associated with various levels of alpha emission, which could have an important role in the selection of a screening level by incorporating the latest health based considerations. This may allow re-evaluation of the use of the current screening standard. The issue is complicated by apparent inconsistencies in the derivation of the primary standard. The gross alpha particle MCL was originally intended by U.S. EPA to screen drinking water for an unacceptable amount of radioactivity from natural sources, primarily represented by radium-226 and present primarily in groundwater. However, it also addresses radium-228, a beta emitter. The companion screening standard for gross beta particle/photon activity addresses man-made sources of radionuclides, focusing primarily on surface water, but ignores natural beta emitters. Measurement of these natural beta emitters would be more appropriately conducted under the conditions specified for the gross alpha measurement scheme, which favors the measurement of groundwater rather than just surface water sources.

Although other health impacts are possible, cancer has been recognized as the major health effect of most studied radionuclides. Moreover, risk assessment procedures to estimate the cancer risk from radionuclides have been well developed. Thus cancer is the principal endpoint that will be used to evaluate the health risk from alpha particle emitters in the present discussion.

Cancer risk from alpha particle emitters varies depending on the manner in which various body organs respond to that energy because certain organs are more sensitive to alpha particle energy from some isotopes than from others. Therefore, a risk-based health-protective value would vary for the different isotopes measured in the gross alpha category. Based on current U.S. EPA cancer coefficients, all of the alpha-emitting radionuclides exceed a *de minimis* risk level of one in a million at the 15 pCi/L screening level. Our PHGs (to be developed) for radium-226, -228, and uranium will address the predominant health concerns represented by the gross alpha particle category. Other alpha particle-emitting radionuclides, including polonium-210 and radium-224, could be present in water supplies, but are not monitored under the established gross alpha screening method. While these radionuclides are generally not as prevalent as radium-226 and -228, their occurrence under rare circumstances might still be a concern for human exposure. This document discusses relative risk and exposure possibilities for various alpha emitters, with the intention of providing a perspective on risks associated with various screening levels for this radiation category. It does not suggest a specific screening level for (combined) gross alpha particles, because this involves analytical considerations. As the appropriate health-protective value varies for different chemicals, any "screening level" would not represent a consistent health-protective standard that would cover all radionuclides of this class. When alpha emitters in water are characterized individually rather than as a class, their risks can be estimated using the U.S. EPA cancer coefficients specific for each radionuclide.

INTRODUCTION

Elements that contain unstable nuclei are said to be radioactive or are called radionuclides. This instability is manifested as the potential to decay or fall into a lower energy state by releasing principally either alpha or beta particles, or gamma rays. An alpha particle is defined as a positively charged particle consisting of two protons and two neutrons. A beta particle is either a negatively charged negatron/electron or a positively charged particle (positron). Gamma rays are high energy, short-wavelength electromagnetic radiation. Radioactive emissions are measured by an activity unit called a Curie (Ci), representing $3.7x10^{10}$ disintegrations per second. For drinking water, the common representation of activity is the picoCurie (pCi), equal to 10^{-12} Ci. Another representation of radioactivity is the Becquerel (Bq), which is one disintegration per second (1 pCi = 3.7^{-2} Bq).

Energetic atoms of radionuclides release their energy either through particles or electromagnetic radiation, which then may in turn interact with other atoms or matter, particularly to knock electrons from off their orbits around the nucleus. This process is defined as ionizing radiation. Ionizing radiation is a particular concern for living tissues as it could lead to changes in important constituents of the cell including DNA, and result in changes in structure and function of the cells or organ systems. Understanding the potential for ionizing radiation to effect changes to cells and tissues requires knowing how much

energy is deposited in the tissues as a result of these emissions. This concept is referred to as the absorbed dose and is represented by units of rad (radiation absorbed dose), which is the amount of energy (in units of 100 ergs) deposited in one gram of matter or tissue. In International Units, the Gray (Gy) is used for characterizing absorbed dose, representing one joule/kg of energy deposited and is equivalent to 100 rad.

However, the radiation particles or energy types differ in their ability to affect tissues, and thus an adjustment or quality factor can be used to compensate for the differences. For example, an alpha particle deposits its energy in a short range and rarely can penetrate the surface layers of tissues, while beta particles and gamma radiation deposit their energies over a greater range. The rem (roentgen exposed man) unit accounts for the difference in the type of radiation by multiplying the absorbed dose in rads by a quality factor, and can also be represented as sieverts (Sv), equaling 100 rem. Another fine-tuning of the absorbed dose is to adjust for the different types of organs affected by radioactive emissions; this is referred to as rem-ede (effective dose-equivalent).

CURRENT CALIFORNIA REGULATIONS AND GUIDANCE

The primary drinking water standards for the State of California include Maximum Contaminant Levels (MCLs) for natural and man-made radioactivity (California Code of Regulations Title 22, Division 4, Chapter 15, Article 5). Section 64441 of the California code addresses Natural Radioactivity and states the following:

> a) All community water systems shall monitor their water supplies for radium-226, radium-228 and uranium at least once every four years. Compliance with maximum radioactivity levels shall be based on the average of the analysis of four consecutive quarterly samples.
>
> (b) Gross alpha particle measurement may be substituted for measurement of radium-226 and radium-228.
>
> > (1) The supply is considered to be in compliance with maximum radioactivity levels if the gross alpha particle activity does not exceed 5 picocuries per liter (pCi/L).
> >
> > (2) If gross alpha activity exceeds 5 pCi/L, measurement of radium-226 shall be made.
> >
> > (3) If radium-226 exceeds 3 pCi/L, measurement of radium-228 shall be made.
> >
> > (4) The sum of the radium-226 and radium-228 shall not exceed 5 pCi/L.
>
> (c) If the average maximum contaminant level for gross alpha particle activity, total radium or uranium exceeds the levels shown on Table 4, the water supplier shall report this information to the Department within 48 hours.

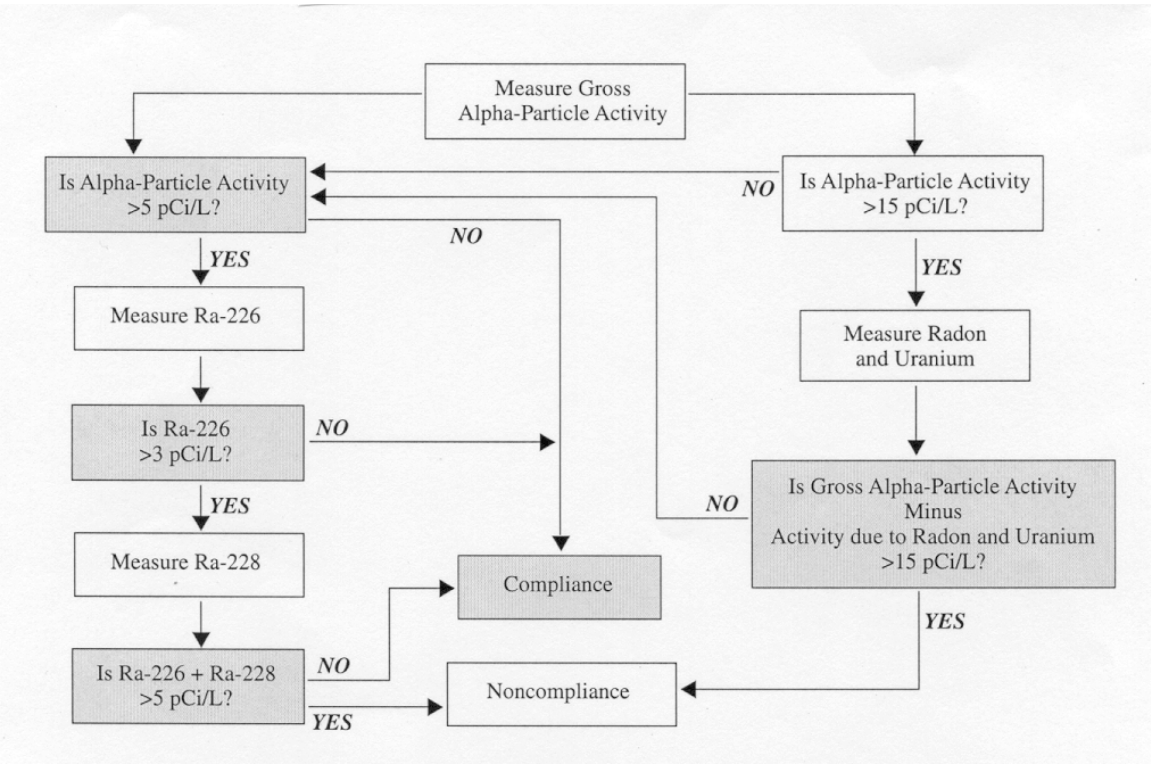
In Table 4 of section 64441, the MCLs for naturally occurring radioactivity are listed, including uranium, radium-226 and -228, and gross alpha particle radioactivity. This table is reproduced below as Table 1.

Table 1. Maximum Contaminant Levels for Natural Radioactivity

Constituent	MCL, pCi/L (Bq)
Combined Radium-226 and Radium-228	5 (0.19)
Uranium	20 (0.74)
Gross alpha particle activity (including Ra-226, but excluding uranium and radon)	15 (0.58)

The gross alpha particle measurement is a relatively easy and quick laboratory technique originally developed to screen water samples for the presence of some key naturally-occurring alpha particle-emitting radionuclides, although the method also screens for radium-228, which is a beta particle emitter. The scheme below illustrates how the gross alpha-particle activity measure is used to evaluate compliance with the MCLs.

Figure 1. Overview of compliance monitoring scheme based on gross alpha particle measures (USGS, 2001)



REGULATORY BACKGROUND

One of the main intentions of the U.S. EPA's 1976 MCL for gross alpha particle activity (15 pCi/L), which includes radium-226 but excludes uranium and radon, was to limit the concentrations of other naturally occurring alpha emitters relative to radium-226. For naturally occurring radioactivity, health professionals are particularly concerned about the alpha-emitting radionuclides from the uranium, thorium and actinium decay series that have relatively longer half-lives. These radionuclides exist long enough to travel through water distribution systems and reach people and are particularly damaging to humans when ingested or inhaled. Fourteen alpha-emitting radionuclides with half-lives longer than one hour are included in this group. Table 2 lists these radionuclides. To ensure that the drinking water levels of these 14 radionuclides are safe, the U.S. EPA promulgated MCLs for total uranium, radium-226, and gross alpha radioactivity. The MCLs for uranium and radium include the most prevalent alpha-emitting radionuclides found in drinking water. In addition to uranium and radium-226, the gross alpha MCL procedure includes a step to screen for radium-228, a beta particle emitter, which is generally not as prevalent in drinking water as uranium and radium-226.

Table 2. Alpha-Particle Emitters from the Uranium, Thorium and Actinium Decay Series with Half-Lives Longer than One Hour

Decay Series	Radionuclides
Uranium	^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{222}Rn, ^{210}Po
Thorium	^{232}Th, ^{228}Th, ^{224}Ra, ^{212}Pb
Actinium	^{235}U, ^{231}Po, ^{227}Th, ^{223}Ra

The U.S. EPA based the gross alpha particle activity MCL on a number of toxicity and exposure considerations. According to U.S. EPA (2000), continuous consumption of drinking water containing polonium-210, the next most radiotoxic alpha particle emitter in the uranium-238/radium-226 decay chain, might cause a total bone dose equivalent to 6 pCi/L of radium-226 at a polonium-210 concentration of 10 pCi/L. The 15 pCi/L limit, which includes radium-226 but excludes uranium and radon, was based on the conservative assumption that if the radium concentration is limited to 5 pCi/L and the balance of the alpha particle activity is due to polonium-210, the total bone dose would be less than the dose associated with an intake of 6 pCi/L of radium-226.

The U.S. EPA was concerned about the analytical costs and the capabilities of state laboratories to analyze individual alpha-particle-emitting radionuclides, and concluded that an MCL for gross alpha was more in keeping with costs and capabilities for most state laboratories.

In 1991, the U.S. EPA proposed excluding radium-226 from the adjusted gross alpha particle activity. In the final 2000 rule, U.S. EPA decided to retain the 1976 standard based on the most current risk analysis using Federal Guidance No. 13 (U.S. EPA, 1999) cancer risk coefficients. These new cancer risk coefficients indicated that the risk from radium-226 was too significant to remove from the gross alpha activity without an appreciable loss of health

protection. Excluding those radionuclides that have specific limits, the U.S. EPA also showed that the concentration that represents a $1x10^{-4}$ lifetime cancer risk (an acceptable risk level by U.S. EPA standards) for most alpha-emitting radionuclides, while varying widely, generally falls below the present MCL of 15 pCi/L (U.S. EPA, 2000).

OCCURRENCE OF ALPHA PARTICLE ACTIVITY IN DRINKING WATER

Of all the naturally occurring alpha-emitting radionuclides of concern for drinking water (Table 2), the U.S. EPA considers those from the actinium decay series to be unimportant because the abundance of the parent (^{235}U) is very low in the earth's crust, less than 1 percent of either uranium or thorium (U.S. EPA, 2000). Of the other radionuclides in drinking water, thorium is extremely insoluble and not subject to mobilization in most groundwater environments (U.S. EPA, 2000). Radon is a noble gas that rapidly volatilizes from water when there is an air-water interface. High radon-222 concentrations have been associated with uranium-enriched phosphate strata that contain sulfuric acid water (USGS, 1998).

The remaining radionuclides include uranium-234 and -238; radium-224, -226 and -228; lead-212; and polonium-210. The occurrences of uranium and radium-226 and -228 are described in separate PHG documents (to be developed), and their levels in drinking water are regulated by separate MCLs. There is little or no occurrence data on lead-212 (U.S. EPA, 2000). However, a recent survey performed by the USGS and the U.S. EPA measured radium-224 and polonium-210 in 103 targeted wells from 27 states (USGS, 1998). These results are summarized in Table 3.

Table 3. Summary of Ground Water Analysis Results of the USGS (1998) Survey

Statistic	**^{224}U**	**^{210}Po**
Number of samples	104	95
Arithmetic mean	3.2 pCi/L	0.1 pCi/L
Median	0.3 pCi/L	0.01 pCi/L
Standard deviation	10.1 pCi/L	0.5 pCi/L
Maximum	73.6 pCi/L	4.9 pCi/L

The concentrations of radium-224 found ranged from less than 1 to 73.6 pCi/L (0.04 – 2.73 Bq). The maximum concentration was found in a coastal plain aquifer in Maryland. Most samples contained radium-224 in concentrations less than 1 pCi/L, with half of the concentrations being less than 0.3 pCi/L. Only 9 percent of the samples exceeded 7 pCi/L of radium-224. Concentrations of radium-224 and radium-228 were highly correlated because both are derived from the thorium-232 decay series. The few samples with high concentrations of radium-224 were from water with very low pH (< 4).

Polonium-210 was detected above the minimum detectable concentration, 1 pCi/L, in only two wells. The highest concentration (4.9 pCi/L, 0.18 Bq) was detected in a private well located in the coastal plain of Virginia.

The municipal water systems in the State of California have reported results from 1994 to 2001 on about 20,000 public drinking water supply wells for various alpha emissions, including radium-226, uranium, and gross alpha particle activity. The gross alpha particle MCL was exceeded about 100 times each year (DHS, 2002). The regulation requires that if 5 pCi/L is exceeded (5 pCi/L is the combined Ra-226, -228 MCL), specific analyses for uranium and radium must be conducted. No other alpha particle emitters are evaluated. Po-210 and other alpha-particle-emitting-radionuclides are not measured, and therefore not reported.

In order to determine a suitable screening level employing a health/toxicity criterion, the type and amount of the contaminants of concern that are expected to be measured by this technique should be considered. The cancer risk can be estimated from alpha-particle-emitting radionuclides other than uranium and radium-226 in two relevant ways. The first is to calculate the risk from measured values of radium-224 and polonium-210. The second is to assume that the entire risk from the gross alpha particle MCL (15 pCi/L) is from radium-226 at its MCL (5 pCi/L), and the remaining 10 pCi/L from other non-uranium radionuclides.

To calculate cancer risk, the cancer morbidity risk coefficients from the Federal Guidance Report No. 13 were used. Table 4 below lists the cancer risk coefficients for the water ingestion exposure route. In addition, the table lists the unit lifetime risk concentrations for drinking water (in risk per pCi/L), assuming a drinking water rate of two liters/day for a 70-year lifetime.

Table 4. Cancer Risk Coefficients and Unit Risk Values for Drinking Water

Radionuclide	Morbidity Risk Coefficients (per Bq)	Risk per pCi/L in drinking water
^{212}Pb	6.76×10^{-10}	1.26×10^{-6}
^{210}Po	4.9×10^{-8}	9.40×10^{-5}
^{224}Ra	4.50×10^{-9}	8.5×10^{-6}
^{226}Ra	1.04×10^{-8}	1.97×10^{-5}
^{228}Ra *	2.81×10^{-8}	5.32×10^{-5}
^{227}Th	1.28×10^{-9}	2.42×10^{-6}
^{228}Th	2.90×10^{-9}	5.48×10^{-6}
^{230}Th	2.46×10^{-9}	4.65×10^{-6}
^{232}Th	2.73×10^{-9}	5.16×10^{-6}
^{231}Pa	4.67×10^{-9}	8.83×10^{-6}

*beta emitter

CANCER RISK AT THE GROSS ALPHA MCL

Using the unit risks for drinking water in Table 4 above, the cancer morbidity risk at the gross alpha activity MCL of 15 pCi/L can be estimated. Assuming that radium-226 (and/or radium-228) was at its MCL value of 5 pCi/L, then the remainder of the activity (10 pCi/L) would come from each naturally occurring alpha emitting radionuclide that could be included in the gross measurement. The results are listed in Table 5. The table shows that except for polonium-210, the combined cancer risk at the MCL ranges from 1.0 to 1.9 x 10^{-4}. With polonium-210, the combined cancer risk at the MCL is 10^{-3}.

Table 5. Cancer Risk of Drinking Water at the Gross Alpha MCL

Radionuclide	Risk per pCi/L	Risk at 10 pCi/L	Total risk including Ra-226 at 5 pCi/L
^{212}Pb	1.26 x 10^{-6}	1.26 x 10^{-5}	1.1 x 10^{-4}
^{210}Po	9.40 x 10^{-5}	9.40 x 10^{-4}	1.0 x 10^{-3}
^{224}Ra	8.51 x 10^{-6}	8.51 x 10^{-5}	1.8 x 10^{-4}
^{227}Th	2.42 x 10^{-6}	2.42 x 10^{-5}	1.2 x 10^{-4}
^{228}Th	5.48 x 10^{-6}	5.48 x 10^{-5}	1.5 x 10^{-4}
^{230}Th	4.65 x 10^{-6}	4.65 x 10^{-5}	1.4 x 10^{-4}
^{232}Th	5.16 x 10^{-6}	5.16 x 10^{-5}	1.5 x 10^{-4}
^{231}Pa	8.83 x 10^{-6}	8.83 x 10^{-5}	1.9 x 10^{-4}

CANCER RISKS FROM ALPHA-EMITTING RADIONUCLIDES IN DRINKING WATER OTHER THAN URANIUM AND RADIUM-226

The cancer risk from alpha-particle-emitting radionuclides other than uranium and radium-226 and -228 was calculated using the cancer morbidity risk coefficients from the Federal Guidance Report No. 13. Table 6 below lists the cancer risk coefficients for the water ingestion exposure route. In addition, the table lists the concentration (activity) at the one in a million, *de minimis* risk level for drinking water (in risk per pCi/L) assuming a 2 L/day drinking water rate for a 70-year lifetime.

Table 6. Cancer Risk Coefficients and Unit Risk Values for Drinking Water

Radionuclide	Morbidity Risk Coefficients (per Bq)	Concentration in pCi/L at the 10^{-6} risk level
^{212}Pb	6.76×10^{-10}	0.8
^{210}Po	4.97×10^{-8}	0.01
^{224}Ra	4.50×10^{-9}	0.1
^{226}Ra	1.04×10^{-8}	0.05
^{228}Ra	2.81×10^{-8}	0.02
^{227}Th	1.28×10^{-9}	0.4
^{228}Th	2.90×10^{-9}	0.2
^{230}Th	2.46×10^{-9}	0.2
^{232}Th	2.73×10^{-9}	0.2
^{231}Pa	4.67×10^{-9}	0.1

USE OF THE GROSS ALPHA PARTICLE MEASUREMENT SCHEME FOR SCREENING

A gross alpha particle PHG is not needed, principally because the predominant contributors to naturally-occurring alpha radioactivity in drinking water are addressed in the uranium PHG and the radium-226 and -226 PHGs (to be developed). The other natural radionuclides that could contribute to the gross alpha activity are most likely radium-224 and polonium-210; these are not measured specifically by the gross alpha particle method. To measure radium-224, which has a shorter half-life, samples may need to be assessed more quickly. Radium-224 might be estimated from measurements of radium-228, since they are part of the same decay series. Measurements from a national survey strongly suggest that these two radionuclides either do not occur very often in drinking water or occur at relatively low levels. The national survey by the USGS (1998) detected polonium-210 in only two out of 95 wells; the highest measurement was 4.9 pCi/L (corresponding to a 4.9×10^{-4} lifetime risk level). Radium-224 was detected more frequently than polonium-210, and 91 percent of the measurements were below 7 pCi/L (a 7×10^{-5} lifetime risk level). For Ra-224, 30 percent of the measurements were above 1 pCi/L (a 1×10^{-5} risk level).

While it is true that the radium-226 PHG (to be developed) and the uranium PHG may address the large majority of the alpha particle radioactivity in drinking water, other gross alpha particle radionuclides would be a potential health concern. The risk comparison above shows that Po-210 is very radiotoxic compared to the other naturally occurring alpha-emitting radionuclides, exceeding their risk by an order of magnitude or more (Table 6). Polonium-210 is even two times more potent than radium-226 with regard to cancer morbidity. However, polonium-210 is not expected to be present in drinking water supplies at levels above its analytical detection level. Radium-224 occurs more commonly than

polonium-210 but is not included in the present evaluation. In the future, if or when measurements of radionuclides other than radium-226, -228 and uranium are conducted, the health risks from other alpha-particle-emitting-radionuclides in drinking water could be addressed by the values given in the tables above.

Another consideration could be to include more natural beta emitters to be assessed under the gross alpha particle measurement scheme. Perhaps it would be reasonable to abandon the "gross alpha particle" title and simply address the whole category as "natural radionuclides," as is done in the California regulations (Title 22, Division 4, Chapter 15, Article 5). For example, radon-222 is a gas that is included in the gross alpha measurement scheme. Its daughter product, lead-210, could be measured as a beta particle emitter under the gross beta particle measurement or under the gross alpha scheme. The gross beta particle/photon measurement is required to be assessed only for certain communities that utilize surface water supplies. Supplies that include groundwater, a major source of natural radioactivity, are not necessarily screened for the natural beta emitters like lead-210. Therefore, natural beta emitters would be more appropriately measured under the conditions specified for the gross alpha particle screening scheme.

The foregoing discussion presents the argument that the current MCL for gross alpha particle radioactivity may not be protective of humans from exposure to key members of this class of radionuclides at the *de minimis* cancer risk level. If screening levels were to be based on providing health risk information to the public for risks above the *de minimis* level (regarded by OEHHA as 10^{-6}), the initial screening level of 5 pCi/L discussed above might logically be lowered by 10-fold or more. The actual risk level depends on the isotope or the mixture of isotopes present. Also, because other radionuclides which could pose a risk to human health are not being measured at all, it may be useful to consider revision of the measurement scheme to include the related natural beta/photon emitters. These isotopes would be more appropriately monitored under the regulatory conditions specified for the gross alpha particle category, where measurements would include ground water sources rather than just surface water sources. Thus, revising both the screening level and the screening procedures would provide more relevant information on activities above the *de minimis* risk level for these regulated radionuclides in drinking water.

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