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**Soil Cleanup at Los Alamos National Laboratory:  
Sediment Contamination in the South Fork of Acid Canyon**

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# **Soil Cleanup at Los Alamos National Laboratory**

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As always, the author remains solely responsible for the contents of this report, its conclusions and recommendations, and any omissions or errors.

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## Section One: Introduction

Between 1944 and 1964, multiple liquid radioactive waste streams were released into the South Fork of Acid Canyon from Los Alamos National Laboratory. From 1944 to 1951, “untreated radioactive effluent from former Technical Area (TA) 1 was discharged into the head of the South Fork of Acid Canyon” and from 1951 to 1964 a “radioactive liquid waste treatment plant at former TA-45” discharged its effluent into the canyon. Today, this area is located within 1,000 feet of a residential neighborhood and less than a mile from a local high-school.<sup>1</sup> We chose to examine the remediation of Acid Canyon because; (1) it is a site that is already accessible to the general public, (2) it has already had remediation efforts undertaken based, in part, on analyses conducted by DOE for site-specific exposure scenarios, and (3) it illustrates some of the general concerns that will arise at Los Alamos and other sites which have actinide contamination (uranium, plutonium, neptunium, americium, etc.) as the main driver of risk.

In the South Fork of Acid Canyon the following radionuclides were identified by DOE as being of potential concern:

Tritium (H-3), Strontium-90, Cesium-137, Uranium-234, Uranium-235, Uranium-238, Plutonium-238, Plutonium-239, and Americium-241.<sup>2</sup>

Given the lack of edible plants in the canyon and that fact that no hunting or fishing is allowed, the authors of the *Interim Report on Sediment Contamination in the South Fork of Acid Canyon* (hereafter the *Interim Report*) considered only the external gamma, soil ingestion, and soil inhalation pathways in conducting their analysis. In light of the proximity of residential areas to the canyon, it was assumed that the canyon could be used by children as an extension of their backyards and that adults could use the hiking/jogging trails in the canyons which cross and pass near contaminated areas.<sup>3</sup> Except for tritium, which is not a major contaminant of concern in Acid Canyon, the extended backyard scenario is the most restrictive scenario evaluated by DOE, and therefore will be the focus of our current review. This is because the present case study is focused on a review of Los Alamos’s calculations of the consequences of its remediation strategy.

Of the nine radionuclides considered in the *Interim Report*, plutonium-239 was by far the primary driver of risk with americium-241 and cesium-137 a distant second and third respectively as can be seen in Table 1.<sup>4</sup> This is mostly because the residual concentrations of plutonium-239 in the canyon soil are far higher than the other radionuclides.

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<sup>1</sup> DOE 2000 p. 2 and Figure 1

<sup>2</sup> DOE 2000 p. 7

<sup>3</sup> DOE 2000 p. 6-7

<sup>4</sup> DOE 2000 p. 13 and 16

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Table 1: Single radionuclide soil guidelines (SRSGs) for extended backyard scenario and the area averaged surface soil contamination in Acid Canyon as reported by the Department of Energy.<sup>5</sup>

Radionuclide	Extended backyard scenario SRSG (pCi/gm)	Maximum detected value before cleanup (pCi/gm)	Canyon average concentration before cleanup (pCi/gm)	Canyon average concentration after cleanup (pCi/gm)
Tritium (H-3)	38,000	1.86	0.53	0.2
Strontium-90+D	5,500	80	6.86	1.9
Cesium-137+D	210	148	7.50	3.5
Uranium-234	3,000	21.5	2.92	3.6
Uranium-235+D	710	2	0.25	0.2
Uranium-238+D	2,000	16.6	1.92	1.9
Plutonium-238	310	37.3	0.97	0.6
<b>Plutonium-239,240</b>	<b>280</b>	<b>7,780</b>	<b>211</b>	<b>112</b>
Americium-241	270	278	13.8	5.4

While the *Interim Report* was “not intended to be a final assessment of the potential risk from contaminants in Acid Canyon, but instead to be an interim report to address specific concerns raised by stakeholders in Fall 1999 and to evaluate the need for immediate remedial action,” we chose to examine its analysis in some detail because it was used, along with ALARA (as-low-as-reasonably-achievable) guidelines, to set cleanup goals for remediation efforts that occurred in the summer and fall of 2001.<sup>6</sup>

With respect to the extended backyard scenario used by DOE to set the preliminary remediation guidelines summarized in Table 1, we have found that:

1. Despite the focus of the scenario on protecting children, the authors of the *Interim Report* did not make use of the age-specific dose conversion factors which were available from the International Commission on Radiological Protection and chose instead to incorrectly use the older dose conversion factors derived for the 154 pound adult male worker.
2. The assumption made regarding the length of time children may be exposed to the contamination in Acid Canyon (200 hours per year) is not adequately conservative for a screening calculation.
3. The ingestion of plutonium contaminated soil dominates the risk for the extended backyard scenario. As such, the *Interim Report* fails to adequately take into account the potential for children to intentionally consume large quantities of soil, a behavior known as geophagia or soil pica.
4. The *Interim Report* does not consider the potential for children to track contaminated soil into their homes creating additional routes of exposure for themselves and for the other people in their family.

<sup>5</sup> DOE 2000 p. 12-13 and 16 and DOE 2002 p. 17

<sup>6</sup> DOE 2000 p. 2 and DOE 2002 p. 1

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5. Finally, the soil guidelines derived by Los Alamos for this scenario are about right due to the approximate canceling of over and underestimates in the *Interim Report*.

Overall, IEER's principal finding is that significant additional remediation of the South Fork of Acid Canyon will likely be required when the assessment of surface water impacts is made by DOE. We have found that the area averaged plutonium concentrations in the canyon soil are significantly larger than the values which could lead to surface water concentrations above 0.15 pCi per liter if they were present in the stream bed. The level of 0.15 pCi per liter is the current statewide surface water limit for Colorado and has been recommended by IEER and other groups for adoption by the Environmental Protection Agency as the federal drinking water limit. While we have not made specific recommendations for the final remediation guidelines for Acid Canyon in this report, we have concluded that the present level of residual contamination is likely too high by at least a factor of ten. IEER's previous recommendations for the cleanup goal at Rocky Flats (1 to 10 pCi per gram of plutonium in the soil, with the lower end of the range corresponding to the protection of drinking water onsite) is consistent with this conclusion.<sup>7</sup>

### Section Two: Generally Protective Assumptions of the Interim Report

To begin we will briefly review the decisions that were made by the authors of the *Interim Report* that we agree are generally protective of public health and have a sound basis for use in quantitative risk assessment. First and most important is their adoption of a 15 millirem per year dose limit as the standard against which compliance was judged. The authors explained their choice as follows:

The radiation dose limit of 15 mrem/yr follows proposed Environmental Protection Agency (EPA) guidelines, and is more protective of possible human health effects than the dose limit of 25 mrem/yr proposed by the US Nuclear Regulatory Commission for unrestricted use of a site (10 CFR 20, *Standards for Protection Against Radiation*) and the limit of 100 mrem/yr in US Department of Energy (DOE) Order 5400.5, *Radiation Protection of the Public and the Environment*. The dose limit of 15 mrem/year is also consistent with developing guidance from DOE/Albuquerque Operations Office.<sup>8</sup>

The use of a 15 millirem per year dose limit rather than 25 or 100 millirem per year is a good practice for radiation protection standards which will apply to the general public and is consistent with the typical level of "acceptable risk" used in regulating other carcinogens. The use of this lower dose limit is also supported by the 2005 report from the BEIR VII Committee of the U.S. National Academy of Sciences that concluded that exposure to low-dose radiation carries even higher risks of causing cancer than was thought to be the case in 1999 when the EPA published its recommendations on radiation risks in Federal Guidance Report 13.<sup>9</sup> The fact that children are the focus of the extended backyard scenario and are also at significantly higher risk from

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<sup>7</sup> Makhijani and Gopal 2001 p. 7-10 and 43-44

<sup>8</sup> DOE 2000 p. 5

<sup>9</sup> NAS/NRC 2005 p. 28 and EPA 1999 p. 182

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radiation exposure compared to adults also supports the use of the more protective 15 millirem per year dose limit.

In addition to the 15 millirem per year dose limit from all pathways, however, it is important that all cleanup standards also include a separate sub-limit of 4 millirem per year to the maximally exposed organ from the drinking water pathway. While the drinking water pathway was not evaluated in the *Interim Report* (see section four), such a sub-limit should be generally included in all cleanup goals in order to help ensure that the most restrictive criteria in each particular case will be used to guide the overall remediation efforts. In the case of Acid Canyon, it appears very likely that meeting the surface water standard of 0.15 pCi per liter proposed by IEER will be the controlling factor behind setting the final cleanup levels and that further remediation of the canyon will be required.

Second, the authors of the *Interim Report* chose to use the EPA's recommended "upper-bound values" for the exposure factors considered in the scenarios.<sup>10</sup> The use of upper-bound values is an appropriate choice for this type of screening analysis. While we do not believe that appropriate upper-bound values were used for the amount of time children may spend playing in Acid Canyon or for the amount of soil that they may ingest, other pathways, such as inhalation, did make use of appropriately conservative assumptions. In assessments where plutonium is the major contributor to the risk, the inhalation pathway must be carefully considered due to the higher dose received from plutonium inhaled into the lung compared to the same amount of plutonium ingested (see Table 2 below). For the extended backyard scenario, the authors of the *Interim Report* made adequately conservative assumptions for the typical level of dust loading, which accounts for how much contaminated soil will be resuspended into the air, as well as for the average inhalation rate of the children playing in the canyon.<sup>11</sup> The one important exception to this conclusion regarding the inhalation pathway may be for children engaging in the intentional consumption of soil (see section three). For these children, particular care should be taken to estimate the amount of soil that is inhaled through the nose and mouth during the close contact that will accompany the ingestion of large amounts of dirt.

### **Section Three: Some Assumptions of the Interim Report not Adequately Protective of Public Health**

#### Doses to Children

First, despite the very specific focus of the extended backyard scenario on children, the authors of the *Interim Report* chose to use the dose conversion factors developed for a 154 pound adult male workers.<sup>12</sup> The authors justified this choice "[b]ecause dose conversion factors for

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<sup>10</sup> DOE 2000 p. 6

<sup>11</sup> DOE 2000 p. 8, NCRP 77 p. 42, Till et al. 2000 p. 6-2, and EPA 1997 p. 5-24

<sup>12</sup> The Reference Man model used to develop the dose conversion factors for adult worker was described by the ICRP as follows:

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populations other than adult workers have not been published by DOE.”<sup>13</sup> In discussing the potential impact of this choice, however, the author’s noted that

**There are no data to estimate the dose conversion factors for children so this uncertainty must remain qualitative.** However, because of their higher metabolism it can be surmised that children are more sensitive to the carcinogenic effects of ionizing radiation than are adults. Therefore, action may be warranted at lower dose environmental concentrations of radionuclides for children than for adults.<sup>14</sup>

When the *Interim Action Completion Report* was published in September 2002 following the remediation efforts in the canyon, the dose conversion factors for the adult male worker were still being used to evaluate doses to children in the extended backyard scenario.<sup>15</sup>

While it is true that the Department of Energy had not published its own collection of age-specific dose conversion factors, by the time the *Interim Report* was published in April 2000, the International Commission on Radiological Protection had published dose models for five different age groups that had been widely accepted by international radiation protection schemes. The ICRP efforts date back to the aftermath of the Chernobyl disaster which raised awareness within the radiation protection community of the need to accurately calculate doses to people of various ages as a result of internally deposited radionuclides. In March 1987, the Task Group on Age-dependent Dosimetry was created within the ICRP. This Task Group (later renamed the Task Group on Internal Dosimetry), along with the Task Group on Dose Calculations published a series of five ICRP reports between 1989 and 1996 that provide dose conversion factors for a number of radionuclides.<sup>16</sup> The specific age groups that were considered by the ICRP are:

- 3 month old (0 to 1 years old),
- 1 year old (1 to 2 years),
- 5 year old (2 to 7 years old),
- 10 year old (7 to 12 years old),
- 15 year old (12 to 17 years old), and
- Adult (over 17 years old).<sup>17</sup>

Since 2001, the ICRP has also published dose conversion factors for the embryo/fetus and for the breast fed infant.<sup>18</sup> Plutonium, the main contaminant of concern in Acid Canyon, was discussed in four of the five ICRP reports issued prior to the *Interim Report*.<sup>19</sup> These newer age-specific

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“Reference man is defined as being between 20-30 years of age, weighing 70 kg [154 pounds], is 170 cm [5 feet 7 inches] in height, and lives in a climate with an average temperature of from 10° to 20° C. He is a Caucasian and is Western European or North American in habitat and custom.” [ICRP 23 p. 4]

<sup>13</sup> DOE 2000 p. 6

<sup>14</sup> DOE 2000 p. 24 (emphasis added)

<sup>15</sup> DOE 2002 p. 17

<sup>16</sup> ICRP 56, ICRP 67, ICRP 69, ICRP 71, ICRP 72 and ICRP 2005b p. A-1

<sup>17</sup> ICRP 72 p. 11

<sup>18</sup> ICRP 88 and ICRP 95

<sup>19</sup> ICRP 72 p. v



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dose models were rapidly accepted by the international radiation protection community. By 1996, the ICRP models had already been incorporated into the European Union’s European Basic Safety Standards and the International Atomic Energy Agency’s International Basic Safety Standards.<sup>20</sup> The EPA issued its own collection of age specific dose and risk factors in a 2002 CD supplement to its Federal Guidance Report 13. The dose conversion factors in this EPA database are generally the same as those of the ICRP.<sup>21</sup>

It seems hard to justify the claims made by the authors of the *Interim Report* in 2000 that there was “no data to estimate the dose conversion factors for children” and that the values for the adult worker had to be used in the DOE analysis. It stretches credulity to believe that the authors were unaware of the ICRP’s efforts, and if they were it would reflect very poorly on their competence to carry out these types of dose calculations.

In the particular case of plutonium, the author’s presumption that using age-specific dose conversion factors would tend to increase the dose relative to that estimated for the adult worker turns out, in fact, to be incorrect. Table 2 summarizes the ingestion and inhalation dose conversion factors for plutonium-239 as estimated by the EPA and the ICRP.

*Table 2: Dose conversion factors for plutonium-239 as reported by the EPA in Federal Guidance Report 11 for the adult male worker and the age-specific dose conversion factors reported by the EPA in the CD Supplement to Federal Guidance Report 13.<sup>22</sup>*

Age Group	Ingestion Dose Conversion Factor (Sv/Bq)	Inhalation Dose Conversion Factor for Class M (Sv/Bq)	Inhalation Dose Conversion Factor for Class S (Sv/Bq)
3 month old	$4.19 \times 10^{-6}$	$8.00 \times 10^{-5}$	$4.27 \times 10^{-5}$
1 year old	$4.22 \times 10^{-7}$	$7.73 \times 10^{-5}$	$3.85 \times 10^{-5}$
5 year old	$3.33 \times 10^{-7}$	$6.04 \times 10^{-5}$	$2.66 \times 10^{-5}$
10 year old	$2.71 \times 10^{-7}$	$4.81 \times 10^{-5}$	$1.86 \times 10^{-5}$
15 year old	$2.46 \times 10^{-7}$	$4.72 \times 10^{-5}$	$1.68 \times 10^{-5}$
adult (25 year old)	$2.51 \times 10^{-7}$	$5.01 \times 10^{-5}$	$1.60 \times 10^{-5}$
FGR 11 (adult male)	$9.56 \times 10^{-7}$	$1.16 \times 10^{-4}$	$8.33 \times 10^{-5}$

It is true that young children will receive a higher dose than a 25 year old adult within the newer dose models. However, due to changes in tissue weighting factors, different assumptions made about the behavior of plutonium in the body, and refinements in the model used to represent the respiratory system, the dose received by inhaling or ingesting plutonium has gone down from the older estimates used in the *Interim Report*. For ingestion, which is the most important pathway in the extended backyard scenario, the dose conversion factor for a 2 to 12 year old child is about three to three and a half times less than the one used in the *Interim Report*.

<sup>20</sup> ICRP 2005b p. A-1

<sup>21</sup> EPA 2002

<sup>22</sup> EPA 2002 and EPA 1988 p. 151 and 177

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While this means that the doses estimated by the DOE analysis for plutonium were, in fact, conservative in this regard they were not based on the latest available scientific information. Of potential significance in other remediation situations is the fact that for many other radionuclides, children can receive higher doses than would be estimated using the older adult male model. For example, the EPA and ICRP age-specific dose conversion factors for ingesting strontium-90 are 22 to 88 percent bigger for a 2 to 12 year old than those for the adult worker used in the *Interim Report*.<sup>23</sup> In all future assessments, the DOE should make use of the latest available dose conversion factors that have been published by the International Commission on Radiological Protection or the Environmental Protection Agency.

Finally, for radionuclides (such as cesium-137) for which external radiation is an important exposure pathway, the estimated gamma dose should be modified to account for the smaller size of children and the fact that they often spend a greater amount of time near the ground and are thus in closer proximity to the contamination. While the effect these factors will have depends on the energy of the gamma rays, and thus on the particular radionuclide involved, the National Council on Radiological Protection has recommended scaling the external gamma dose estimated for adults by  $1.3 \pm 0.1$  to get the dose for children up to at least 12 years of age.<sup>24</sup> This type of guidance has been followed by the NRC Staff in the context of evaluating reactor decommissioning plans for the Haddam Neck plant.<sup>25</sup>

### Exposure Duration

Second, as discussed above, the authors of the *Interim Report* stated that they sought to use “upper-bound values” for the exposure factors in their analyses. One of the areas in which IEER does not believe that the choice made in the *Interim Report* is adequately conservative was in the length of time children were taken to play in the canyon. The authors of the *Interim Report* assumed that a child would spend 200 hours per year playing in the canyon, which would amount to approximately one hour per day for seven months of the year. They note that this assumption is “based on professional judgement, incorporating input from NMED.”<sup>26</sup> However, the 95<sup>th</sup> percentile value reported by the EPA was that a child age 1 to 11 years old would spend as much as eight or nine hours in outdoor activity per day. Even focusing on just the average values, the study cited by the EPA recommendations estimated that children between two and eleven spend 2.2 hours outdoors at home and an additional 1.9 hours outdoors at parks, etc.<sup>27</sup>

The dose received by a child in the extended backyard scenario is directly proportional to the amount of time the child spends in the canyon. Thus, for a screening calculation which is meant to provide a conservative basis upon which to base the need for or adequacy of cleanup efforts, it is important to make consistent use of “upper-bound values” for all parameters, including exposure duration. The choice of an adequately conservative estimate for the length of time

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<sup>23</sup> EPA 2002 and EPA 1988 p. 160

<sup>24</sup> NCRP 129 p. 56-57

<sup>25</sup> NRC 2003 p. 39

<sup>26</sup> DOE 2000 p. 7

<sup>27</sup> DOE 2000 p. 8, EPA 1997b p. 15-187, and EPA 2002b p. 9-48 and 9-59

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children may play in the contaminated areas should be made with input from the local residential population. Based on the studies underlying the EPA recommendations, it would be likely that an exposure time of 300 to 400 hours per year would be a more appropriate screening level for the case of Acid Canyon.

### Soil Ingestion

Third, soil ingestion is by far the most important exposure pathway in the extended backyard scenario accounting for more than 90 percent of the DOE's estimated total dose from all radionuclides present. Given its dominant role in governing the cleanup goals for Acid Canyon, it is particularly important that the soil ingestion pathway be addressed completely. The authors of the *Interim Report* start with the EPA's recommended 95<sup>th</sup> percentile soil ingestion figure of 400 milligrams per day and assume that this ingestion of soil occurs uniformly over the entire time the child spends outdoors (5.6 hours per day). The authors then calculate what the total amount of soil ingestion would be during the 200 hours the child is assumed to spend playing in the canyon. As noted above, the dose from soil ingestion will thus be directly proportional to the length of time the child is assumed to play in the canyon. From this, the authors of the *Interim Report* estimate that a child in the extended backyard scenario will consume 14.3 grams of contaminated soil over the course of a year.<sup>28</sup> However, despite the existence of a number of studies examining soil ingestion, there remain significant uncertainties both about the actual long-term rate of ingestion and about the variability between individuals and groups.<sup>29</sup> A review of studies on soil ingestion, published in *Health Physics* following the publication of the EPA's *Exposures Factor Handbook*, recommended using a 95<sup>th</sup> percentile value for soil ingestion for a suburban lifestyle which was more than four times higher than the 95<sup>th</sup> percentile value recommended by the EPA.<sup>30</sup>

More important than the uncertainties in the estimated amount of routine soil ingestion, is the issue of how the critical group in the *Interim Report* is defined. In conducting risk assessments, once the exposure scenarios are generally defined, the next step is to identify a group of individuals that are expected to receive the highest doses and that is also "small enough to be relatively homogenous with respect to age, diet and those aspects of behaviour that affect the doses received."<sup>31</sup> As noted by the International Commission on Radiological Protection in its draft 2005 recommendations

Such a group is chosen to be representative of the most highly exposed individuals as a result of the source. Its characteristics should be derived from the mean of a homogeneous and sustainable group. Additionally, it is important that the habits used in calculating the dose to the individuals are the average habits in the critical group and not the habits of a single extreme individual. The critical group may, however include some

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<sup>28</sup> DOE 2000 p. 8

<sup>29</sup> EPA 1997 p. 4-20, Simon 1998 p. 659, and Royal Society 2002 Annexe C p. 2-3

<sup>30</sup> Simon 1998 p. 661-663

<sup>31</sup> ICRP 26 p. 17

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individuals with extreme or unusual habits and should be selected such that all relevant habits are taken into account.<sup>32</sup>

In the draft foundation document on dose calculations supporting these recommendations, the ICRP reiterated that, when conducting probabilistic assessments, “[c]are must be used to include all hypothetical individuals whose dose could possibly be representative of persons receiving the highest dose, including extremes.”<sup>33</sup> The ICRP went on to conclude that

Close attention must be paid to suggestions from members of the public of existing or likely exposure situations that might reflect extremes in the population. . . . If it can be shown that such a pathway, in combination with other exposure, is likely to affect a few tens of persons and elevate their doses above the dose constraint, then a revision of the analysis must be undertaken. If such a homogeneous group is found to exist above the dose constraint, then the mean dose to this group becomes the basis for compliance.<sup>34</sup>

In the case of soils contaminated with actinides such as plutonium-239 and where children are likely to play or otherwise come into close contact with the soil, particular care must be taken to ensure that the critical group includes the potential for the intentional ingestion of large quantities of soil.<sup>35</sup> If a significant number of children are ultimately determined to be expected to exhibit this kind of behavior, than it must be included in the underlying definition of the critical group against which compliance should be judged.

Geophagia, the intentional ingestion of large quantities of soil, has been documented for centuries and is commonly viewed as a particular manifestation of a behavior known as pica which is the intentional ingestion of all non-food stuffs such as paint, string, and soil. It has been found to occur across “geographic, ethnic and cultural boundaries” and has “been noted not to be a rare event.”<sup>36</sup> In its 1985 Superfund Guidance, the EPA acknowledged that short term soil ingestion well above the typical 95<sup>th</sup> percentile are possible and recommended that risk assessments consider potential exposures of 5 grams per day.<sup>37</sup> In studies of lead poisoning in children, the intentional ingestion of soil and paint chips is commonly viewed as playing a significant role.<sup>38</sup> In their 1997 *Exposure Factors Handbook*, the EPA concluded that “it can be assumed that the incidence rate of deliberate soil ingestion behavior in the general population is low.” However, the EPA went on to note that “the prevalence of pica behavior is not known” and that due to the short time period over which children have so far been studied, “[i]t is plausible that many children may exhibit some pica behavior if studied for longer periods of time.”<sup>39</sup> As summarized by Calabrese et al.

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<sup>32</sup> ICRP 2005 p. 44

<sup>33</sup> ICRP 2005b p. 17

<sup>34</sup> ICRP 2005b p. 18

<sup>35</sup> Simon 1998 p. 656

<sup>36</sup> Simon 1998 p. 649 and 659

<sup>37</sup> Calabrese et al. 1997

<sup>38</sup> Mielke and Reagan 1998

<sup>39</sup> EPA 1997 p. 4-18 and 4-20

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Realistic estimates of soil pica are problematic. Estimating the frequency, magnitude, variability, and duration of soil pica has not been the object of extensive research. In the course of three soil ingestion studies, we have observed unambiguous soil pica in two children.... These data suggest that soil pica may vary considerably both between and within individuals and are consistent with observations that generalized pica behavior is common in normal children, but may be more prevalent and of longer duration in mentally retarded children.

...The findings also support the hypothesis that there is considerable interindividual variation with respect to soil pica frequency and magnitude. Thus, for the majority of children, soil pica may occur only on a few days of the year, but much more frequently for others. If soil pica is seen as an expected, although highly variable, activity in a normal population of young children, rather than an unusual activity in a small subset of the population, its implications for risk assessment become more significant.<sup>40</sup>

Estimates for the amount of soil that a pica child might intentionally ingest carry even greater uncertainties than estimates of routine ingestion. Accurately estimating the amount of soil ingestion requires "extensive knowledge of the living conditions and cultural attitudes of the population of interest."<sup>41</sup> Generally, however, the assumptions that have been made are that a child experiencing pica will consume between 5 and 10 grams per day. This has been the assumption adopted by risk assessments and recommendations of the Environmental Protection Agency, the Centers for Disease Control, and the Agency for Toxic Substances and Disease Registry.<sup>42</sup> In 1997, the EPA officially recommended the use of 10 grams per day as the ingestion rate for a pica child.<sup>43</sup> However, smaller estimates (one to five grams per day) and larger estimates (26 to 85 grams per day) have been considered by other sources.<sup>44</sup> For the purposes of screening calculations in which soil ingestion is a major pathway, an acute exposure from the consumption of at least 30 to 40 grams of soil per year, occurring on a small number of days, should be considered in addition to the chronic exposure from routine, inadvertent soil ingestion.

Finally, given that intentional soil ingestion events are most likely to be short-term, acute exposures, the inhomogeneous distribution of the radionuclide contamination should be considered in estimating the potential impact of pica events. This is particularly true for transuranic elements which are known to result in highly inhomogeneous contamination patterns from studies of fallout around Chernobyl and the Marshall Islands.<sup>45</sup> In the case of Acid Canyon, for example, there were hot-spots with a combined area of 50 m<sup>2</sup> (4.5 percent of the contaminated land in the canyon) which had an average plutonium-239 concentration of 2,740

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<sup>40</sup> Calabrese et al. 1997

<sup>41</sup> Simon 1998 p. 659

<sup>42</sup> EPA 1997 p. 4-20 and Simon 1998 p. 661

<sup>43</sup> EPA 1997 p. 4-20 and 4-25

<sup>44</sup> Simon 1998 p. 661-663 and Royal Society 2002 Annexe C p. 4

<sup>45</sup> Simon 1998 p. 666

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pCi per gram.<sup>46</sup> A single pica event in which a child consumed 10 grams of soil from these hot-spots would have alone resulted in a dose greater than 25 millirem. Although no mention was made of the potential for such acute doses from pica, these two areas of contamination were subsequently removed during the summer and fall of 2001 as part of attempts to maintain doses as low as reasonably achievable.<sup>47</sup> While both the probability and the consequences of acute exposures need to be considered in risk assessments, the potential for pica children consuming large amounts of the most heavily contaminated soils should be addressed in the process of setting the final remediation guidelines. In the case of Acid Canyon, the application of other criteria lead to a cleanup level that does not pose a radiologically significant threat from acute soil ingestion of plutonium, but this may not always be the case for other sites or for other radionuclides.<sup>48</sup>

### Transported Soil as a Potential Exposure Potential Pathway

Our fourth concern with the extended backyard scenario relates to the fact that it does not consider what may be a potential pathway of exposure, namely the fact that children may track contaminated soil into their homes. This type of pathway has been noted by the EPA and ATSDR in some cases for exposures to lead, mercury, arsenic, polychlorinated biphenyls (PCBs), and other toxic chemicals.<sup>49</sup> In addition to increasing the exposure of the older children who tracked in the soil, this pathway creates the possibility that infants at home could be exposed despite never traveling to the canyon if they have older siblings who play there. This potential exposure pathway may be important to consider because the ingestion dose conversion factor for infants is more than four times larger than the dose conversion factor used in the *Interim Report* (see Table 2) and infants have heightened hand-to-mouth behavior and spend much of their time in contact with furniture or the floor which can bring them into increased contact with contaminated dust.

Typical household dust is made up of a mixture of soil from outdoors, paint, plaster, biological material such as dead skin, and other materials. What fraction of household dust is dirt from outside is highly variable and depends on a variety of site specific factors. For example, three different studies estimated the fraction of soil in household dust to be 14 to 15 percent, 30 to 40 percent, and 75 to 100 percent respectively.<sup>50</sup> Significant variations have been found from one contaminant to another and from one house to the next. For example, one of the most heavily studied contaminants with respect to soil ingestion is lead. Estimates for the amount of lead in household dust that is attributable to soil from outside range from 20 to 95 percent. Some studies found the level of indoor lead to be associated with the level of lead outdoors while other studies found no such correlation. In light of these uncertainties, the EPA's Integrated Exposure Uptake

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<sup>46</sup> DOE 2000 p. 16

<sup>47</sup> DOE 2002 p. 1 and 17

<sup>48</sup> For example, the consideration of a child ingesting 30 to 40 grams of soil would lower the single radionuclide soil guidelines for uranium-234, uranium-238, and strontium-90 reported in the *Interim Report* for the extended backyard scenario. Even these modified cleanup guidelines, however, would be far above the levels of contamination reported as measured in the canyon. [DOE 2000 p. 16 and DOE 2002 p. 17]

<sup>49</sup> EPA 1999c, EPA 2005, ATSDR 1994, ATSDR 1998, and ATSDR 2002

<sup>50</sup> Wong et al. 2000 p. 443 and Royal Society 2002 Annexe C p. 1

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Biokinetic Model for Lead in Children makes the default assumption that 70 percent of dust is made up of dirt from outside.<sup>51</sup> In its assessment of the health risks from depleted uranium munitions the UK Royal Society made a similar assumption and concluded that “a value of 75% [of household dust being soil from outside] would seem appropriate, even though this is almost certainly cautious in many cases.”<sup>52</sup>

In addition, when dealing with the transfer of contamination from outside to inside, there are a number of factors that may act to enhance the concentration of contaminants in dust. These factors include the fact that there are fewer ways for contaminants on household dust to degrade or be transported away compared to outdoors, the fact that carpets can act to store dust over long times, and the fact that some dust is derived from biological material such as molds and fungi that can act to bioaccumulate certain contaminants.<sup>53</sup> Studies of these effects, however, have shown significant variability. For example one study found no significant enhancement of lead indoors but did find an enhancement of copper on household dust.<sup>54</sup> Another study, however, found significantly higher concentration of “lead, cadmium, antimony and mercury” in household dust compared to either street dust or garden soil, but found the opposite trend for “aluminum, barium and thallium.”<sup>55</sup> A third study found the levels of arsenic and lead to be higher indoors than outdoors for residences on or near fruit orchards which had used lead arsenate insecticide, and concluded that this enhancement was associated with soil having been tracked in from outside.<sup>56</sup> The lack of any generally applicable rules regarding the possible correlation between indoor and outdoor contaminant levels makes it difficult to make any generic assumptions which can be used in risk assessments. In order to determine if this potential pathway is of importance in such cases as Acid Canyon, measurements in and around local residences will be required.

In performing these measurements, one additional complication that must be dealt with is the issue of pets. Studies have found that elevated levels of lead in children correlate with elevated levels of lead in their pets. While it is not fully understood if the pets are a route of exposure or not, it has been noted that “houses that had dogs and cats appeared to have a higher level of metals” and that “[t]his may be due in part to the fact that pets usually bring in dust from outdoors” given that “[t]hey stay close to the ground... and spend most of their time playing with dirt or dust.”<sup>57</sup> In addition, pets may increase a child’s access to soil for either ingestion or inhalation “by digging or by accumulating soil and dust in their fur.”<sup>58</sup> The choice of sampling locations should take note of the existence of pets and whether they can go outdoors in order to establish any potential impact they may have on the importance of this exposure pathway.

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<sup>51</sup> Wong et al. 2000 p. 443 and Rasmussen, Subramanian, and Jessiman 2001 p. 126 and 136

<sup>52</sup> Royal Society 2002 Annexe C p. 1

<sup>53</sup> Tong 1998 p. 130, Wong et al. 2000 p. 443-444, and Rasmussen, Subramanian, and Jessiman 2001 p. 137

<sup>54</sup> Tong 1998 p. 123

<sup>55</sup> Rasmussen, Subramanian, and Jessiman 2001 p. 130

<sup>56</sup> Wolz et al. 2003 p. 293, 296-297

<sup>57</sup> Wong et al. 2000 p. 447 and 449 and Tong 1998 p. 128-129

<sup>58</sup> Wong et al. 2000 p. 444

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### Section Four: Surface Water Assessment

The scope of the *Interim Report* explicitly excluded an analysis of “[w]ater-related exposure pathways” due to “the lack of surface water data from Acid Canyon.”<sup>59</sup> The authors went on to note that the assessments of “risk from contamination in surface water are pending further data analysis and interpretation.”<sup>60</sup> When this assessment of potential surface-water impacts is carried out it will be important that it consider the most up to date science on plutonium health risks. As detailed in the IEER report *Bad to the Bone: Analysis of the Federal Maximum Contaminant Levels for Plutonium-239 and Other Alpha-Emitting Transuranic Radionuclides in Drinking Water*, the science underlying the current drinking water limit for gross alpha-activity (which would include plutonium activity) is more than four decades old and is no longer a satisfactory basis for the protection of public health. IEER has recommended reducing the concentration limit for plutonium and other long-lived alpha emitting transuranic elements from its current value of 15 pCi per liter to 0.15 pCi per liter. IEER and other groups have requested that the EPA take this information into account as part of their 2006 review of the Safe Drinking Water Act standards, and the EPA has agreed to consider the findings of the IEER report.<sup>61</sup> Of particular significance in the present case, we note that the State of Colorado has already adopted a 0.15 pCi per liter state-wide surface water standard for plutonium.<sup>62</sup> We also note that New Mexico governor Bill Richardson has written to the EPA and encouraged them to lower the allowable limit for plutonium in drinking water along the lines recommended by IEER.<sup>63</sup>

To illustrate the potential significance of the surface water impacts from the known contamination in Acid Canyon, we considered the typical levels of plutonium in stream sediment that would lead to an equilibrium concentration of 0.15 pCi per liter in the surface water. Table 3 summarizes our results using typical values of the partition coefficient for plutonium for various soil types.

*Table 3: Concentrations allowable in the stream sediment for different soil types in order to maintain the equilibrium concentration of the surface water below the 0.15 pCi per liter limit recommended by IEER.*<sup>64</sup>

Partition Coefficient ( $K_d$ )	Plutonium concentration in water (pCi/L)	Plutonium Concentration in sediment (pCi/gm)
550 (geometric mean value for sand)	0.15	0.083
2,000 (ResRad default value)	0.15	0.30
5,100 (geometric mean value for clay)	0.15	0.77

<sup>59</sup> DOE 2000 p. 6

<sup>60</sup> DOE 2000 p. 25

<sup>61</sup> Makhijani 2005 p. 6-9 and Blette 2005

<sup>62</sup> Makhijani 2005 p. 21 and Colorado Reg. 31, 2005/08/08 p. 25 and 64

<sup>63</sup> Richardson 2005

<sup>64</sup> EPA 1999b p. 2.16 and Yu et al. 2001 p. E-9 to E-13



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Given that the average concentration of plutonium-239 in the canyon's soil as reported by the DOE, even after the remediation that took place in 2001, was 112 pCi per gram, the potential for this contamination to adversely affect the surface water is clear. The issue of the impact of residual plutonium in the soil on surface and ground water at the Los Alamos site must be carefully addressed by the DOE in all aspects of waste management and cleanup activities. No remediation guideline should be accepted that would not maintain the concentration of all long-lived alpha emitting transuranic elements in both surface and ground water below the limit of 0.15 pCi per liter. In the specific case of Acid Canyon, the requirement to protect the surface water will almost certainly be a more restrictive criterion than the extended backyard scenario, and will thus likely determine the final remediation goals for this location.

### Section Five: Conclusion

In summarizing the results of the *Interim Report*, the authors concluded that

Although we did not perform a quantitative uncertainty analysis on these parameters it is highly likely that actual doses would be less than those calculated in this evaluation because upper end exposure assumptions were made for key parameters (like exposure time).<sup>65</sup>

However, we have found that both for the question exposure duration and for the issue of intentional soil ingestion, the *Interim Report* is not adequately conservative. While the appropriate upper-bound estimate for the length of time children may spend playing in the canyon should ultimately be guided by input from local residents as well as expert judgment, values from 50 to 100 percent longer than those used in the *Interim Report* seem reasonable based on existing EPA guidance. In addition, the value of 14.3 grams per year used for the total amount of contaminated soil that a child might consume may be less than the amount of soil consumed by a pica child in a single event in some instances.<sup>66</sup> In cases such as Acid Canyon, where soil ingestion dominates the risks, the critical group should consist of children who will display pica behavior in addition to the inadvertent soil ingestion considered by the authors of the *Interim Report*. Using current EPA recommendations, a reasonable value for these acute exposures would likely be on the order of 30 to 40 grams for exposures lasting a few days per year. Finally, a voluntary measurement program should be conducted in the local communities to gauge the potential radiological significance, if any, of children tracking contaminated soil into their homes.

While not addressed by the *Interim Report*, we have found that considerations of plutonium's impact on the surface water in Acid Canyon is likely to lead to a far more restrictive cleanup criteria than the extended backyard scenario used to guide the 2001 remediation efforts. Information on the level of plutonium contamination that may impact the surface water is not available in the *Interim Report*, however, the levels of contamination remaining the soil of Acid

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<sup>65</sup> DOE 2000 p. 24

<sup>66</sup> Calabrese et al. 1997 and Royal Society 2002 Annexe C

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Canyon are likely to be too high by at least a factor of ten if the amount of plutonium and other transuranic elements in the surface water is to be maintained below 0.15 pCi per liter over the long term. This concentration is the current statewide surface water limit for Colorado, and has been recommended by IEER and other groups for adoption by the EPA as the federal drinking water limit. While we have not proposed specific remediation guidelines for Acid Canyon pending further assessment by the Department of Energy, we note that IEER has previously recommended setting a cleanup goal at Rocky Flats of between 1 and 10 pCi per gram of transuranic elements in the soil with the lower end of the range corresponding to the protection of drinking water onsite.<sup>67</sup> This recommendation is consistent with our expectations for the level of residual contamination that will ultimately be set for Acid Canyon.

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<sup>67</sup> Makhijani and Gopal 2001 p. 7-10 and 43-44

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