The Proposed Ward Valley Radioactive Waste Facility: Papers Submitted to the National Academy of Sciences

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Panel on Ward Valley Board on Radioactive Waste Management National Academy of Sciences/ National Research Council 2001 Wisconsin Avenue, N.W. Washington, D.C. 20007

Dear Member of the NAS/NRC Panel on Ward Valley:

I believe that the enclosed material should prove useful in the deliberations of your committee. It merits your careful scrutiny.

Sincerely, int Como q-

Robert Cornog, Ph.D. [co-discoverer of tritium, 1939]

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EMPIRICAL MEASUREMENT OF RADIONUCLIDE MIGRATION AT LLRW DISPOSAL SITES IN ARID LOCATIONS

Abstract: Comprehensive radiation monitoring data for the US Ecology LLRW site at Beatty, Nevada, published in the last few days, provide a unique opportunity to evaluate the validity of optimistic transport models that have been used to predict travel times to groundwater in the tens of millennia. The newly available data show gross alpha readings in groundwater in excess of action levels in eight different years, gross beta in violation of action levels seven years, and tritium in excess of action levels four years, with significantly elevated tritium (>1,000 pCi/L) but below action levels an additional four years. The data provide clear evidence that radioactive materials have migrated from the disposal trenches to groundwater, 300 feet beneath the surface, in a few decades. The presence of elevated gross alpha, gross beta, and Cobalt-60 in the groundwater, in addition to substantial tritium, rule out vapor-phase migration. These empirical observations of rapid radionuclide migration contrast sharply with predictions by Prudic (1994) for Beatty and Ward Valley using Chloride Mass Balance calculations.

Introduction

Proponents of the Ward Valley LLRW project have attempted through various theoretical models and assumption-driven calculations to demonstrate that radioactivity buried in an arid location such as the existing US Ecology facility at Beatty, Nevada, or the proposed site at Ward Valley will assertedly take tens of thousands of years to migrate through the vadose zone. These models and calculations are dependent upon a long list of controversial assumptions: that there is an upward gradient at the sites, that matric potential and other soil parameters needed as inputs for the calculations have been accurately measured, that heterogeneities in the soil profiles can be effectively ignored, that all water movement in the unsaturated zones in question can be accurately described with a simple piston-flow displacement model and that there aren't potential bypass modes such as preferred pathways or mobile/immobile phases of soil water, that (in the case of the chloride mass balance approach) chloride deposition rates over the last 50,000 years are well known and can be estimated from measurements taken in the current period far from the location in question, and so on.

The problem, of course, is that models may or may not accurately represent what occurs in the field. Furthermore, they are only as good as the input assumptions upon which they are based, and the input assumptions at work here are untested and heavily disputed. Leaving aside for the moment the question of the serious problems with the measurements (e.g., thermocouple psychrometer readings) upon which US Ecology attempts to rely, at base what exists is an almost theological dispute. By that we mean that advocates of the proposition that there is essentially no deep percolation in arid zones and that what does infiltrate migrates only over tens of millennia

are being strongly challenged by skeptics, but the former belief is based to a significant degree on faith (i.e., theory). True, there are serious arguments that can be advanced to support the theory, as there are to challenge it, but theory it remains. The evolution of science, however, makes clear that theories change or are abandoned over time as more data are obtained, or as intellectual fashions in a particular field shift.

Hazardous/Radioactive Waste Disposal History One of Optimistic Initial Models Subsequently Abandoned After Facilities Fail

Indeed, this pattern of the promulgation of optimistic theory and the necessity subsequently to abandon the theory when events in the real world tragically disproved it, has been at the root of the troubled history of toxic low-level radioactive waste disposal in this country. The State of California approved the Stringfellow Acid Pits based on the fact that annual evaporation exceeded annual precipitation; 15 years later groundwater contamination was extensive and spreading, and now a court has saddled the state with a cleanup bill approaching \$800 million due to inadequate review when it first approved the project (Environment Week, 1992). During much of the 1950s and 1960s, radioactive waste was dumped in 55 gallon oil drums off the east and west coasts of the United States, on the assumption that the drums would contain the radioactive materials until they had decayed and that should any be released, it would be dispersed. In the mid-1970s, however, subsequent studies found that many of the barrels were already corroding and breached, releasing radioactivity; and that the radioactivity adsorbed onto bottom sediments where they were ingested by bottom-dwelling organisms which in turn were subsequently consumed by higher species, concentrating radionuclides up the food chain (cf. Davis, 1982). Monitoring was so inefficient that responsible agencies lost track of even the locations of something on the order of half of the ocean dump sites (Hirsch, 1981).

Initial predictions for US Ecology's LLRW facility at Sheffield, Illinois, were that it would safely contain the radioactive waste for millennia; within 15 years of opening, the facility had failed and had to be closed after extensive radionuclide migration and contamination resulting from the failure to adequately characterize the site beforehand (failure to identify sand lenses and their capability of acting as fast-track migration pathways) and reliance on models that eventually were found to have substantially underestimated travel times (U.S. Congress, 1976). US Ecology's LLRW facility at Maxey Flats similarly failed, when radionuclides such as tritium and plutonium were found to have migrated offsite in as little as a decade despite initial predictions such migration would take many thousands of years (Shrader-Frechette, 1992); the failure to consider the effects of complexing agents on increased mobility and decreased soil

retention were among the causes identified (Cleveland and Rees, 1981; Weiss and Czyscinski, 1981; Fowler and Polzer, 1988).

The history of the heavily contaminated Department of Energy nuclear complex strongly reinforces this same theme – initial optimistic models predicting extremely slow travel times proved by experience to be tragically wrong, as radioactive wastes have contaminated vast areas, with cleanup costs estimated on the order of \$155 billion (U.S. Congress, 1991; U.S. Department of Energy, 1991). As the National Research Council (1989, p. 37) noted in its evaluation, "Virtually every facility in the weapons complex has some amount of environmental contamination within its boundaries while many also have some contamination outside the boundaries."

Models are transient, changing, readily abandoned. Radioactive contamination is, in human terms at least, permanent, and abandonment of aquifers or land are considerably more costly than subsequent abandonment of a model that turned out to be a mistake. Mistaken models can thus be extremely costly to the human enterprise and the environment in general.

Best Evidence: Has Radioactivity Migrated at Arid Sites?

It should be remembered that in the case at hand, the models and theory-based calculations are all designed to answer only one question. Can the radioactive materials proposed to be buried in unlined trenches at Ward Valley reach groundwater or the surface prior to having decayed away? The best evidence to answer that question is not the theoretical models advanced by facility proponents. Theories are, after all, merely hypothesis. Science is not the promulgation of theory – that is merely the first step. The core of science is the testing of hypothesis against hard evidence obtained from controlled experimentation.

The best evidence is thus, by definition, not theoretical models but actual data. The best data for the question at hand are those that indicate whether there has been, at arid LLRW sites, radioactive migration faster than predicted by the optimistic models of the Ward Valley project proponents. US Ecology (1990) has conceded that it has had troubles at its now-closed eastern LLRW sites involving radioactive contamination but asserts its facilities located in arid western sites (Richland, Washington, and Beatty, Nevada) have been free of such difficulties. The problems its facilities have experienced, US Ecology asserts, are due neither to its reliance on a design involving no containment (i.e., unlined trenches) nor its own track record, but rather location in humid climates (Kentucky and Illinois).

Furthermore, US Ecology has said that its Beatty site can be relied upon as an analog for Ward Valley; indeed, the company has used infiltration estimates from Beatty for its computer model for Ward Valley (License Application, Appendix, p. A-11 - A-12). Prudic (1994) has based his conclusion that it would take more than 50,000 years for radioactive material to migrate to a depth of 30 meters beneath the Ward Valley site on chloride mass balance calculations he has applied identically to both the Beatty and Ward Valley sites. The best evidence of whether he may be right would be to examine whether and how far radioactive material from the Beatty waste trenches has indeed migrated and compare that to his assertion, based on his chloride mass balance (CMB) calculations, that no movement of moisture has occurred beneath 10 meters for 15,000 years and that before that time the migration rate was only 0.2 centimeter per year (Prudic, 1994, p. 18). Evaluating actual radioactive migration at Beatty would be a good test of his even more dramatic assertions regarding the Ward Valley site, of migration rates of a mere 2-3 centimeters (cm) per 1,000 years (Prudic, 1994, p. 18).

The chloride mass balance technique utilized by Prudic is handicapped by the fundamental fact of chemistry that stable chloride is by definition *stable* (i.e., it does not decay, so its age cannot be determined by measuring how many half-lives have elapsed). A radioactive isotope, in contrast, in essence carries with it a clock by which its age can be directly measured.

Techniques based on stable chloride are further handicapped, because stable chlorine has been in existence since the earliest history of the earth, as opposed to artificial radionuclides whose existence or abundance is due to recent human activity, for example, nuclear weapons testing or wastes from nuclear reactors. Such artificial tracers carry an additional "clock" with them, as their time of origin is more or less known (e.g., 1945 or thereafter for A-bomb fallout, 1952 or thereafter for H-bomb fallout, 1962 or thereafter for wastes migrating from commercial LLRW sites). Reliance on such artificially-produced tracers makes direct estimates of travel times possible without resort to theoretical calculations highly dependent on input assumptions and the unproven validity of the model itself.¹

¹ It is for this reason that the tritium found beneath Ward Valley is of such significance. That tritium carries with it its own clock. Given the 12.3 year half-life of tritium, and pre-bomb tritium concentrations in precipitation that should be less than the 7 TUs currently measured there in atmosphere moisture, it must have taken something on the order of three half-lives, or ~35 years, to travel that distance. If the tritium is from bomb fallout, the same time period is at work, as the first thermonuclear bombs were detonated in the early 1950s, about 35 years before the tritium measurements were made at Ward Valley.

The simplest, and most important test of the assertions of project proponents thus would be to examine US Ecology's other arid sites as to whether there is any evidence of radionuclide migration at those facilities. Of course, those sites have been operating for only a relatively short time (~30 years) and the migration times of concern are far longer, given the longevity of many of the radionuclides buried. Thus, the absence of evidence of migration might not be determinative of the larger question, but its presence would be.

Evidence of Radionuclide Migration at Beatty

Depth to groundwater at US Ecology's Beatty site is 85 to 115 meters and mean annual precipitation is approximately the same at Beatty as at Ward Valley, 12.8 cm for the former and 11.7 cm for the latter (Prudic, 1994, p. 2). Prudic estimates that there has been no movement of water beneath 9 meters of the surface in 20,000 years and a downward percolation rate of 0.2 centimeters per year below that depth, although he asserts that that rate was probably only applicable about 20,000 years ago and current percolation rates would be even lower. Taking that rate as the current rate, however, it would take 35,000 years to travel 70 meters, the minimum depth to groundwater from the deepest Beatty waste trench, 15 meters deep [Conference of Radiation Control Program Directors (CRCPD), 1994, p. 4-8]. Clearly, if Prudic's CMB assumptions and calculations are correct, there should be absolutely no radioactive material from the Beatty waste trenches in groundwater at the site, at least not for another 35,000 years.

During his presentation before the NAS-NRC Ward Valley panel in July 1994, Prudic did not volunteer that there were groundwater contamination data from Beatty that called into question the model and calculations he was presenting. Upon repeated questioning by the panel, he eventually conceded that samples taken from a well drilled by USGS downgradient of the LLRW site were positive for tritium.

Well MR-3 was drilled in 1987 and sampled for the first and only time in August 1989, showing levels of 12.2 ± 1.9 and 6.4 ± 1.9 pCi/L (Prudic, 1993a; 1993b). This was part of a United States Geological Survey (USGS) program collecting data on groundwater quality near the Beatty LLRW site; six wells were sampled in 1989 (including the one that tested positive for tritium, well MR-3) and four separate wells were sampled in 1992 (Prudic, 1993b, p. 1).

Prudic speculated (1993a, Table 4 - 1989) that the positive finding for tritium, which was confirmed by replicate analysis, might have been caused by remnant drilling fluid. This is

unlikely, as he himself noted that more than 2,000 gallons of water had been pumped from the well immediately prior to sampling, and the sampling itself occurred two years after drilling. In addition, it is unclear why this problem would assertedly manifest itself only in this well, when there were other wells also monitored within 2 years of drilling (wells 600, 604, and W001).

The more reasonable interpretation that the tritium finding is valid is supported both by the fact that it was confirmed by replicate analysis and by noting the location of the well in which the tritium was found compared to those in which it was not found. Of the wells monitored by Prudic in his review, only MR-3, the one in which tritium was found, was directly downgradient of the radioactive waste facility. Eight were upgradient of the LLRW facility, and one (W001) was off to the side. Only MR-3 was downgradient of the LLRW site. (See map from Prudic, 1993a, on p. 7 of this report.)² If one were going to find radioactive contamination, it would be in MR-3, which is indeed precisely where it was found. The other wells, upgradient, serve as controls, demonstrating that the tritium found in MR-3 appears tied to the waste facility.

Upon repeated prodding by the NAS-NRC panel, Prudic conceded there was other evidence of radioactive contamination having reached groundwater at Beatty, in addition to his own measurements discussed above. After many years in which there were no monitoring wells, either on the LLRW site or downgradient from it, in 1982 US Ecology drilled two wells on the LLRW site itself, one (301) just inside the upgradient boundary and one (302) just inside the downgradient boundary (see map on p.7). From the time of the opening of the LLRW facility in 1962 until wells 301 and 302 were added, only the site supply well was monitored³ (CRCPD, 1994, p. 4-15). The site well is in the buffer zone outside the LLRW facility boundary and upgradient of it (see map).

The very first sample taken from the new downgradient well (302) found extremely elevated tritium levels – $410,000 \pm 10,000$ pCi/L (Administrative Record, 1993, p. 123-00190 - 123-00191). This is more than 20 times the U.S. Environmental Protection Agency (EPA) Safe Drinking Water standard for tritium⁴ (EPA, 1976, p. 155) and 200 times the Beatty facility's

² Prudic monitored the site well, and wells MW 313, 314, 315, 316, 600, 604, 311, W001, as well as MR-3. (Prudic, 1993a). It is difficult to understand the purpose of sampling wells almost all of which are upgradient of the facility one is attempt to monitor.

³ Again, it is difficult to comprehend how a LLRW facility could be effectively monitored for such a long period via only well, one that was not even on the LLRW site and which was upgradient of it.

⁴ A higher standard employed by the U.S. Nuclear Regulatory Commission for tritium in effluents from nuclear facilities (10 CF 20 Appendix B) is sometimes cited for comparison purposes, but the more restrictive EPA standard is controlling for concentrations in water supplies.



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New wells deilied during 1990, and intersayies collected in December 1112.

Source: Prudic, 1993a (handwritten note is from Prudic) action level (CRCPD, 1994, p. 4-10). Tritium was also found in upgradient well 301, but in levels generally considerably lower than those found in the downgradient well. The discovery of tritium contamination via the opening of the new wells led to increased frequency of monitoring. Tritium continued to be found in the wells, at significant levels, month after month, although the concentration appeared to decline over the next two years. The data are reproduced in Table 1.

Prudic (1993b, p. 2) says that well 302 "reportedly" went dry in August 1983 and has been dry since that time, however it is clear from the data that measurements of groundwater continued through the end of January 1985, so if the well went dry, it would appear to have occurred after January 1985. Fischer (1992, p. 7) reports well 302 as dry in December 1988. In any case, the available data indicate elevated tritium in the downgradient well (302) for virtually all of the more than 2-year period, 1982 through 1985, for which data are available, as well as lower but still significant tritium levels in upgradient well 301 on half a dozen occasions during the 1 1/2 years for which measurements are available for it.

The California Department of Health Services (DHS) has dismissed the tritium findings, claiming they were "greater up-gradient than down-gradient," that the readings allegedly occurred only in 1983, and that "there was no recurrence" (DHS, 1993, p. 27). In particular, DHS has stated, "There has been no tritium detected in Beatty wells in 1991 or the recent past." As we have shown above, and will show in more detail below, each of these assertions is incorrect. The tritium findings were repeated samples, essentially monthly, from 1982 into the end of 1984. There is no evidence of subsequent measurements showing no tritium from 1985. on; in fact, Prudic and Fischer claim the downgradient well went dry, and when USGS put in a new well downgradient, quite near well 302, the only measurements from it, in 1989, were also positive for tritium. Furthermore, as shall be shown below, in addition to Prudic's measurement in well MR3 in 1989, other measurements at Beatty found elevated readings 100 times higher than Prudic's in 1989 and, in 1991 as well. Lastly, the well (302) with the very high tritium readings from 1982 through 1984 was downgradient, not upgradient as DHS asserted (see map, above), precisely where one would most expect elevated levels. The well downgradient from the LLRW trenches was an order of magnitude higher than the well on the upgradient portion of the facility, a clear indication that the contamination was indeed coming from the trenches. DHS does say that investigations by US Ecology and the State of Nevada were "not able to ascertain the specific cause" of the contamination (DHS, 1993, p. 27).

Well	Date Sampled	Tritium, pCi/L
301	* 6-28-82	0.0 ± 1,000
	* 9-09-82	24,000 ± 1,000
	10-26-82	< 1,000
	* 2-03-83	200 ± 70
	* 3-03-83	< 2,000
	3-30-83	< 200
	3-30-83	< 200
	4-07-83	< 200
	7-11-83	< 200
	7-11-83	< 220
	7-11-83	< 200
	8-02-83	0 + 200
	8-22-83	500
	9-02-83	500
	9-30-83	500
	10-27-83	300 + 200
	11-30-83	200 1 200
	12-21-83	0 + 200
	1-26-84	0 ± 200
	1 20 04	0 1 200
302	* 10-26-82	410,000 ± 10,000
	* 2-03-83	$48,900 \pm 3,000$
	* 3-03-83	65,200 ± 6,520
	3-11-83	50,100
	3-30-83	30,700
	3-30-83	31,000
	4-07-83	30,000
	5-03-83	46.700
	5-03-83	47.000 ± 2.000
	6-03-83	$13,000 \pm 600$
	6-29-83	11.000 ± 600
	7-11-83	10,000
	7-11-83	18,200 ± 900
	8-02-83	$13,200 \pm 700$
	8-22-83	< 500
	9-30-83	2,800 ± 750
	10-27-83	5,800 + 300
	11-30-83	4.200 + 200
	12-21-83	3,000 + 300
	1-26-84	3,600 + 200
	3-02-84	2 900 + 100
	4-14-84	2 100 + 200
	5-31-84	1 600 + 200
	6-20-84	1,000 ± 200
	7-21-04	1,000 ± 200
	7-31-84	1,000 ± 200
	0-07-04	1,400 ± 200
	9-27-84	800 ± 200
	10-30-84	500 ± 200
	11-30-82	300 ± 200

Table 1. Beatty, Nevada, Well Water Sampling Results

Analyzer

Eberline Eberline Teledyne Teledyne Teledyne EAL USGS USGS USGS EPA EAL EAL CEP CEP CEP EAL EAL EAL EAL

Eberline Teledyne Teledyne EPA USGS USGS USGS ЕРА EAL EAL EAL EAL EAL EAL CEP CEP EAL EAL

EAL.

EAL

 Samples collected and analyzed by US Ecology contractor. Rest of samples collected by State and analyzed by different agencies or contractors.

Source: Ward Valley Administrative Record, 123-00190 to 123-00191.

12-28-84

1-31-85

500 ± 200

0 ± 200

There have been suggestions that the contamination found in the groundwater during the 1982-1984 period may have been due to sabotage by a disgruntled employee. While this is not a particularly comforting an explanation – as sabotage is a mechanism for radionuclide transfer to and contamination of groundwater not considered in any environmental impact or safety review for either the Beatty or Ward Valley projects – the evidence makes such an explanation highly unlikely. First of all, as will be shown below, similar contamination was found in other wells before (in 1979 and 1980) and after (1985, 1989, and 1991) (CRCPD, 1994, p. 4-16; Prudic 1993a). Second, contamination was found in both the upgradient and downgradient wells, and far greater in the downgradient one. Third, the suggestion that well 302 was a dry hole and water strangely appeared in it for a time (Prudic, 1993b, p. 2), water that turned out to be contaminated, before becoming dry again, cannot explain the dropping *concentrations* of tritium in the well. The amount of water in the well could perhaps drop, if it was originally a dry hole and someone had poured contaminated water in it, but the *concentration* of tritium in that water should remain essentially constant over the two years of measurements. Prudic (1993b, p. 2-3) agrees that the drop in concentration eliminates the sabotage possibility as a reasonable explanation.

The 12.3 year half-life of tritium cannot explain the drop in concentration in Well 302 from 410,000 pCi/L on October 26, 1982, to 47,000 pCi/L in May 1983, not to 1,400 pCi/L in August 1984. Tritium leaking from particular degrading waste packages from the waste trenches, migrating down to groundwater, and then traveling downgradient in the aquifer *can* readily explain the data, and is in fact the only reasonable explanation. The data present a picture of a contamination front passing the monitoring well, with tailing concentrations traveling behind. Subsequent releases from the trenches can have the same effect, and the measurements of elevated tritium before and after the 1982-1984 findings paint the same picture – a leaking facility, with leachate reaching and contaminating the groundwater beneath it.

Indeed, despite US Ecology's efforts at the NAS-NRC Ward Valley panel meeting in June to explain the tritium findings away as potential sabotage, its monitoring reports for Beatty conclude that the most probable cause for the presence of tritium in ground water is migration down to the ground water from the disposed waste (CRCPD, p. 4-15 and 4-34, citing US Ecology's annual monitoring reports).

It may be argued by Ward Valley/Beatty defenders that the tritium somehow reached the groundwater 300 feet below the surface via gas phase migration and is not indicative of potential solute travel. It is essentially impossible to show a mechanism for gas phase migration that would result in tritium concentrations in groundwater 300 feet below, 20 years after the facility

opened, resulting in levels of over 400,000 pCi/L. Furthermore, as will be discussed below, elevated gross alpha, gross beta, and cobalt-60 levels in groundwater demonstrate that the contamination is not just by tritium but by other radionuclides that cannot travel as a gas and must travel as a solute.

On November 21, 1984, the Nevada Department of Human Resources (State of Nevada, 1984) cited US Ecology for violating its license, in particular the requirement "that when the concentrations of radioactive material in water samples are found to be above action levels (30 picocuries per liter for alpha and 90 picocuries per liter for beta) the Division will be notified." The citation stated:

Contrary to the above requirement, during this inspection it was learned that the company had been notified on October 18, 1984 of analyses of two water samples that had radioactive materials in concentrations above the action levels and the Division had not been notified.

A 1987 inspection report by the State of Nevada noted that wells 301 and 302 "have shown elevated levels of gross alpha and beta, and tritium in the past" (State of Nevada, 1987). The gross alpha and beta contamination indicate solute contamination. Tritium alone could be arguably vapor phase, but not elevated gross alpha and gross beta.⁵

Additional Monitoring Data, Showing Repeated Contamination at Beatty – Alpha, Beta, Cobalt-60 – in Groundwater, Unsaturated Zone Soil, and Offsite Vegetation

It is our understanding that the NAS-NRC Ward Valley panel, interested in learning whether there is any further evidence of migration of wastes from the US Ecology facility at Beatty, as a test of whether proffered models asserting no such migration could occur in arid zones, has requested from US Ecology and various regulatory bodies all such monitoring data. We understand little if any such data have as yet been received. Indeed, US Ecology asserted at the August meetings of the panel that all other monitoring data have shown no indication of any radioactivity and that they would provide all the data.

We have just received new data, dated October of this year, summarizing 30 years of monitoring at Beatty (CRCPD, 1994). The data compilation was issued by the Conference of

⁵ Because of the very weak beta given off by tritium, and the analytic methods employed for gross beta scans, tritium is not included in gross beta readings and is measured and reported separately.

Radiation Control Program Directors, Inc., and prepared for it by the EG&G contractors at Idaho Falls, Idaho, who run the National Low Level Radioactive Waste Management Program for the Department of Energy.⁶ CRCPD is the national organization of chiefs of state radiation protection programs. The new data demonstrate far more clearly than previous information presented to the NAS panel that radioactivity has migrated from the waste trenches at Beatty – into groundwater beneath the site (vertical migration), soil in dry wells downgradient from the site (lateral migration), and in vegetation (upward migration). The contaminants include tritium, cobalt-60, and gross alpha and gross beta contamination, demonstrating that liquid-phase solute transport is involved. The new data present a long and consistent pattern – not an isolated allegedly anomalous reading or two, as asserted by Prudic and US Ecology at the meetings of the NAS panel.

Table 2, summarizing groundwater monitoring data for gross alpha, gross beta, and tritium since Beatty opened, is taken from the CRCPD report, p. 4-16. For eight separate years, beginning as early as eight years after the facility first opened and continuing into the 1990s, gross alpha activity in groundwater beneath the site exceeded the Action Levels set by US Ecology and the State of Nevada, at times by more than a factor of 20. For seven separate years, gross beta in groundwater exceeded Action Levels, at times by an order of magnitude. Measurable tritium (in excess of 500 pCi/L) was found 8 out of the 13 years for which there are data, ranging from 1,000 to 49,000 pCi/L.⁷ By contrast, current tritium levels in rainfall are about 20-60 pCi/L. With tritium's 12.3 year half-life, no measurable tritium whatsoever should be showing up in groundwater, let alone at these high concentrations, whether from rainfall or leachate, if the Ward Valley proponents' were right that migration rates are on the order of thousands or tens of thousands of years. Whereas US Ecology and Prudic at the NAS meetings appeared to suggest there were just a couple of anomalous readings, the elevated tritium is showing up most years for which there are data. Despite suggestions of sabotage being the cause for the 1982-1984 readings, this report (p. 4-15 and 4-34), says US Ecology's own monitoring reports have attributed the tritium in groundwater to "migration from the disposed waste."

⁶ The report was prepared by the "E-5 Committee" of the Conference of Radiation Control Program Directors, Inc. The "E-5 Committee" is the Waste Management Oversight Committee.

⁷ The authors of the CRCPD study (p. 4-15) reported the high tritium level found in 1982, 410,000 pCi/L \pm 10,000 pCi/L, in the text of their report but did not include it in their table on p. 4-16. Similarly, the highest 1983 reading reported in the Administrative Record table on p. 123-00190 is 65,200 \pm 6520, higher than the value reported in the CRCPD report and with a far smaller error bar. The error margin given in the CRCPD report for the figure 49,000 \pm 29,000 appears to be an error; the Administrative Record table for 1984 are much smaller, e.g., 3,600 \pm 300.

	Gross alphaª	Gross beta ^a	Tritium	
 Year	pCi/L	pCi/L	pCi/L	
1962	3 ± 2	54 ± 4	no data	
1963	no data	no data	no data	
1964	8 ± 3	50 ± 26	no data	
1965	20 ± 5	60 ± 31	no data	
1966	10 ± 5	60 ± 34	no data	
1967	10 ± 4	40 ± 28	no data	
1968	14 ± 5	52 ± 3	no data	
1969	6 ± 3	41 ± 41	no data	
1970	\rightarrow 39 ± 7	\rightarrow 94 ± 30	no data	
1971	no data	no data	no data	
1972	10 ± 4	9 ± 4	no data	
1973	\rightarrow 46 \pm 7	\rightarrow 549 ± 47	no data	
1974	16 ± 10	\rightarrow 132 ± 77	no data	
1975	\rightarrow 47 ± 9	\rightarrow 173 ± 55	no data	
1976	12 ± 5	40 ± 32	no data	
1977	< 3	<30	no data	
1978	3 ± 2	<20	no data	
1979	10 ± 5	<20	\rightarrow 3,800 ± 1,100	
1980	< 5	10 ± 4	•• 1,700 ± 900	
1981	21 ± 7	31 ± 4	0	
1982	\rightarrow 710 ± 183	\rightarrow 340 ± 49	$ ightarrow$ 24,000 \pm 1,000	
1983	\rightarrow 140 \pm 98	\rightarrow 930 ± 150	\rightarrow 49,000 ± 29,000	
1984	$\rightarrow 63 \pm 29$	\rightarrow 140 ± 24	\rightarrow 5,000 ± 4,000	
1985	25 ± 18	26 ± 10	•• 1,100 ± 600	
1986	15 ± 9	14 ± 5	< 500	
1987	no data	no data	no data	
1988	$\rightarrow 31 \pm 11$	10 ± 3	< 500	
1989	20 ± 14	30 ± 23	•• 1,548 ± 508	
1990	\rightarrow 78 ± 24	63 ± 11	< 500	
1991	10 ± 6	11 ± 5	•• 1,079 ± 551	
1992	7 ± 3	13 ± 3	< 500	

Table 2. Gross alpha, gross beta, and tritium activity in groundwater, Beatty LLRW facility, 1962-1992.

Action Levels: gross alpha = 30.0 pCi/L; gross beta = 90.0 pCi/L; tritium = 2,000 pCi/L

a. Indicates highest value for each year. \rightarrow Indicates Action Level exceeded. •• Indicates < Action Level but >1000 pCi/L H-3

Source: Conference of Radiation Control Program Directors, Inc., 1994, Environmental Monitoring Report for Commercial Low-Level Radioactive Waste Disposal Sites: Frankfort, KY, Conference of Radiation Control Program Directors, Inc., p. 4-16.

Vapor-phase migration is contradicted, both by the very high concentrations of tritium found in groundwater and by the evidence of migration of soluble radionuclides as shown by the repeated elevation of gross alpha and gross beta above action levels in groundwater. In addition, cobalt-60 was found in sediment in groundwater taken from one of the monitoring wells.⁸ Cobalt-60 is an artificial isotope with a five-year half-life (in 50 to 100 years it decays to non-radioactive levels) and found in large amounts in low level radioactive waste, its presence is likewise indicative of migration of leachate to groundwater since the facility opened in 1962. Cobalt-60 has been found to migrate rapidly at other radioactive sites, particularly when in chelated form (Means and others, 1978, Killey and others, 1984).

In Table 3, we have reprinted the soil sample data, which, until 1984, were taken primarily from dry wells dug downgradient to monitor for possible lateral migration from the trenches (CRCPD, 1994, p. 4-21 - 3). These dry wells were normally located downgradient of the completed trenches and extended at least 10 feet below the established bottom of the trench. Two additional dry wells were located downgradient of the site itself. Note that for four years, gross alpha in the soil samples exceeded action levels. For six years, gross beta was in excess of action levels. This suggests lateral subsurface flow in the unsaturated zone, a matter raised by the Wilshire group regarding Ward Valley (Wilshire and others, 1993, p. 3-7).

In September 1984, the State of Nevada eliminated the requirement for soil sampling of the dry wells, in part because most of the soil had been removed from the dry wells during the years of sampling, leaving behind mainly rocks (CRCPD, 1994, p. 4-22). The fact that subsequent to that time, action levels for soil have not been violated would appear to be resulting primarily from the elimination of the requirement to continue sampling the dry wells where the previous violations of action levels had been detected.

In Table 4, the table for vegetation sampling is reproduced (CRCPD, 1994, p. 4-27). Gross beta limits were exceeded in vegetation in six different years.

⁸ The CRCPD report indicates that during some time after the very elevated tritium readings were found, at least during part of 1985 and perhaps parts of 1983 and/or 1984 (it is not clear from the text), gamma spectroscopy and fluoroscopy were performed on the suspended fraction (as opposed to dissolved fraction) taken from the water samples to attempt to ascertain contribution to gross alpha and gross beta levels resulting from naturally occurring radionuclides. Action levels were not exceeded in 1985. Furthermore, it is not clear that the artificial radionuclides of concern would be in the suspended fraction rather than the dissolved fraction. If rapid migration were resulting from chelation, the complexed radionuclides would be primarily in the dissolved fraction and thus missed. Additionally, gross alpha or gross beta levels would not fluctuate, year to year, from 10% of the Action Level to 10 times the Action Level, if the sole source of activity were naturally occurring radionuclides.

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	Gross alpha ^a	Gross beta ^a	
Year	pCi/gm	pCi/gm	
1962	no data	no data	
1963	no data	no data	
1964	no data	no data	
1965	1.9 ± 0.63	72 ± 4.4	
1966	2.7 ± 1.2	73 ± 5.3	
1967	1.7 ± 0.64	3.5 ± 0.34	
1968	2.94 ± 0.41	5.03 ± 0.57	
1969	9.5 ± 3.7	37 ± 4.1	
1970	no data	no data	
1971	8.9 ± 3.8	80 ± 4.9	
1972	13 ± 5	\rightarrow 108 ± 32	
1973	6 ± 3	\rightarrow 110 ± 40	
1974	18.2 ± 8.2	\rightarrow 253.6 ± 111	
1975	$\rightarrow 64 \pm 15$	$\rightarrow 614 \pm 60$	
1976	\rightarrow 42 ± 7.7	$\rightarrow 257 \pm 28$	
1977	20 ± 6.1	60 ± 24	
1978	18 ± 6	60 ± 24	
1979	\rightarrow 31 ± 13	80 ± 3.1	
1980	23 ± 6.1	$\rightarrow 90 \pm 16$	
1981	\rightarrow 32 ± 9.8	60 ± 15	
1982	25 ± 6	66 ± 18	
1983	24 ± 7	79 ± 4	
1984	25 ± 7	52 ± 1.5	
1985	16 ± 3	40 ± 17	
1986	10.2 ± 1.6	9.6 ± 1.0	
1987	1.3 ± 0.3	7.0 ± 0.8	
1988	5.7 ± 1.1	6.6 ± 0.9	
1989	9.3 ± 2.5 (wet)	21.5 ± 1.3 (wet)	
1990	12.8 ± 3 (dry)	51.5 ± 7 (dry)	
1991	3.5 ± 1.0 (dry)	22.0 ± 1.3 (dry)	
1992	5.4 ± 2.7 (dry)	28.4 ± 2.3 (dry)	
		(-))	

Table 3. Soil sample analysis - Beatty LLW Site.

Action Levels: gross alpha = 30.0 pCi/gm; gross beta = 90.0 pCi/gm

a. Indicates highest value for each year.

 \rightarrow Indicates Action Level exceeded.

Source: CRCPD, 1994, p. 4-23

	Gross alpha ^a	Gross beta ^a
Year	pCi/gm	pCi/gm
1962	0.73 ± 0.32	\rightarrow 126 ± 3.1
1963	no data	no data
1964	no data	no data
1965	0.13 ± 0.04	21 ± 0.5
1966	0.9 ± 0.45	\rightarrow 110 ± 5.4
1967	0.39 ± 0.22	8.0 ± 0.4
1968	0.16 ± 0.04	13.3 ± 0.2
1969	0.17 ± 0.12	31.3 ± 0.27
1970	no data	no data
1971	0.19 ± 0.16	2.8 ± 0.3
1972	1.4 ± 1.0	\rightarrow 722 ± 35
1973	0.36 ± 0.32	27.2 ± 3
1974	3.8 ± 4.1	\rightarrow 420 ± 110
1975	3.49 ± 2.2	\rightarrow 146 ± 30
1976	9 ± 3	\rightarrow 220 ± 20
1977	0.3 ± 0.006	39.6 ± 14.5
1978	0.7 ± 0.03	36.9 ± 9
1979	0.7 ± 0.6	29.3 ± 4.2
1980	2.4 ± 1	50 ± 5.1
1981	9 ± 4	17.6 ± 1.4
1982	2.4 ± 2	30 ± 4.9
1983	· 6±3	55.7 ± 4.9
1984	6.3 ± 1.8	15.5 ± 2.3
1985	7.2 ± 1.3	16 ± 1
1986	0.8 ± 0.2	5.8 ± 0.2
1987	5.3 ± 2.7	77.6 ± 2.5
1988	3.2 ± 0.4	10 ± 0.3
1989	0.6 ± 0.2 (dry)	65.5 ± 8.1 (dry)
1990	3.1 ± 2.4 (dry)	16.3 ± 3.6 (dry)
1991	0.5 ± 0.2 (dry)	5.9 ± 0.3 (dry)
1992	11.4 ± 2.3 (dry)	48.9 ± 2.8 (dry)

rable 4. Vegetation sample analysis - Deatty LLW Site	Table 4.	Vegetation	sample analysis	- Beatty	/ LLW Site
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Action Levels: gross alpha = 30.0 pCi/gm; gross beta = 90.0 pCi/gm

a. Indicates highest value for each year. \rightarrow Indicates Action Level exceeded.

Source: Conference of Radiation Control Program Directors, Inc., 1994, Environmental Monitoring Report for Commercial Low-Level Radioactive Waste Disposal Sites: Frankfort, KY: Conference of Radiation Control Program Directors, Inc., p. 4-27

Table 5 reproduces tritium readings for vegetation for one time period, March 1982, taken from outside the facility boundary (CRCPD, 1994, page 4-30). The readings are extraordinary – up to 1000 pCi/ml. (These readings are questioned in the report as "not readily explained." Split samples with the State of Nevada resulted in widely divergent readings, with US Ecology saying they may possibly be related to chemiluminescence.)

Location	Analysis	Concentration
200 ft. southeast of south fence in dry wash	H-3	34 ± 2 pCi/ml
	Cs-137	1 ± 0.4 pCi/gm
240 ft. southeast of south fence in dry wash	H-3	1,000 ± 100 pCi/ml
	Cs-137	1.5 ± 0.6 pCi/gm
300 ft. southeast of south fence in dry wash	H-3	630 ± 10 pCi/ml
	Cs-137	1.4 ± 0.5 pCi/gm
400 ft. southeast of south fence in dry wash	H-3	340 ± 10 pCi/ml
	Cs-137	0.9 ± 0.5 pCi/gm
320 ft. south of south fence, NE	H-3	60 pCi/ml
	Cs-137	0.8 ± 0.3 pCi/gm
320 ft. south of south fence, SE	H-3	170±10 pCi/ml
	Cs-137	1.5 ± 0.4 pCi/gm
320 ft. south of south fence, SW	H-3	15 ± 1 pCi/ml
	Cs-137	0.5 ± 0.3 pCi/gm
320 ft. south of south fence, NW	H-3	ll±lpCi/ml
	Cs-137	1.3 ± 0.4 pCi/gm
a. Data for other years are not available.		

Source: Conference of Radiation Control Program Directors, Inc., 1994, Environmental Monitoring Report for Commercial Low-Level Radioactive Waste Disposal Sites: Frankfort, KY: Conference of Radiation Control Program Directors, Inc.

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Lastly, even the direct gamma exposures measured by thermoluminescent dosimeters (TLDs) above the surface at the site fenceline are remarkable, measuring up to 1,140 mrem per quarter, or about forty times background (CRCPD, 1994, p. 4-31).

Discussion

When Prudic gave his presentation to the NAS-NRC Ward Valley panel about Beatty and his models supposedly demonstrating that no migration of radionuclides was possible at the site – and by implication, any similarly arid site – it was unfortunate that he did not volunteer the best possible data for assessing his assertions, actual measurements of leachate reaching groundwater. Similarly, it is unfortunate that his recent paper on the subject (Prudic, 1994) is likewise silent on the subject and that he hasn't published his own findings of tritium in groundwater at Beatty. Even when the NAS-NRC panel tried diplomatically to extract from him information about such data, he was reluctant to disclose it. Unfortunately, these data now appear to be but the tip of the iceberg.

It now appears that there is ample evidence of radionuclide migration from the US Ecology trenches at Beatty to groundwater 300 feet below, migration that must be in the liquid phase. This contradicts the claims of Ward Valley proponents, including Prudic, that water basically does not move in the vadose zone in arid locations, and raises very serious questions about his theoretical chloride mass balance calculations, based on idealized model assumptions (e.g., pure piston flow, uniform and well-known chloride deposition rates over long times). Prudic's chloride paper (1994) asserts moisture movement at Beatty of only a millimeter per year and even less than that at Ward Valley, purportedly taking tens of thousands of years to travel 10 meters – yet radioactive waste has reached groundwater at Beatty, 100 meters below the surface, within ten years of the facility opening.

Theoretical models have their place, but it is data that matters. Models are merely to help us assess whether radioactive material can migrate at appreciable rates in conditions of arid climates. The best possible answer to that question would be not models, but actual measurements of whether radioactive materials *have* reached groundwater. The Beatty radiological monitoring data make clear not only that it *can* happen, but that it already *has* happened.⁹ In science, Theory must defer to Fact.

⁹ A discussion of new evidence about contamination at US Ecology's Richland LLRW site is found in Appendix A.

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APPENDIX A

NEW EVIDENCE OF CONTAMINATION OF THE VADOSE ZONE AT THE US ECOLOGY LLRW SITE IN RICHLAND, WASHINGTON

We have recently obtained data showing elevated tritium levels (~400,000 pCi/L) in soil pore water from the vadose zone beneath US Ecology's Richland LLRW facility, strongly suggesting rapid migration in that arid site as well. Claims have repeatedly been made about Richland, similar to the claims regarding Beatty and Ward Valley, that because average annual pan evaporation potential so exceeds average annual precipitation (~6 inches at Richland), there is essentially no infiltration and recharge at the site. Those claims are in similar jeopardy as a result of vadose zone monitoring just completed.

In November 1991, US Ecology installed three wells into the unsaturated zone at its LLRW facility at Richland, located in the semi-arid region of eastern Washington State, as part of a two-year research project conducted at the request of the Washington State Department of Health. Vadose Well (VW) #101 was installed in the vicinity of waste trenches 4 and 5, and VW-102 was placed near trenches 10 and 11A. A background well (VW-100) was placed away from the disposal areas in the northwest corner of the site. A map identifying the locations of the two monitoring vadose wells and of the background control vadose well follows, as well as a completion log for VW-100 (US Ecology, 1994a). The wells extended approximately 85 feet below the surface. Silica gel packs were placed in a perforated cylinder into the bottom of the well to absorb water vapor present in the vadose zone; the vadose well was then sealed off at about the 45 foot depth (US Ecology, 1994a, p. 3; US Ecology 1994b, p. 5-141). (Potential contamination from atmospheric moisture would not be a concern, as the monitored tritium levels in the vadose wells were on the order of 400,000 pCi/L, about four orders of magnitude higher than tritium in atmospheric moisture.) Silica gel packs were replaced quarterly and the accumulated soil moisture was then monitored for tritium. Results for 1993 are reproduced in Table A-1 from US Ecology (1994b).

The reader will readily see that tritium concentrations in the vadose zone beneath the waste trenches range as high as $4.5 \times 10^{-4} \mu \text{Ci/cc}$ (450,000 pCi/L), over 20 times the U.S. Environmental Protection Agency (EPA) Safe Drinking Water Level. The concentrations found in the vadose zone near the burial locations average three orders of magnitude higher than at the control/background location (see Figures A-1 and A-2). The control location, located about

A-1

600 feet farther from the burial trenches than the locations where the elevated tritium was found, is nonetheless still in the middle of the U.S. Department of Energy (DOE) Hanford Reservation. Therefore, the contamination found in the vadose zone near the US Ecology burial locations cannot be coming from other operations at Hanford but must be coming from the US Ecology waste trenches.

The vadose zone monitoring program was initiated at the request of the Washington Department of Health in part to verify markedly elevated tritium readings in vegetation at the US Ecology site, in particular in vegetation growing on the trench caps. The State of Washington concluded, after reviewing the vadose zone data and the vegetation data, that "there was a correlation between the two" (Washington, 1993, p. 46). Thus, there is strong evidence that tritium is migrating in substantial quantities both upwards to the surface and downward in the vadose zone fairly deep below the waste trenches.

TABLE A-1. 1993 Vadose Zone Monitoring Results From US Ecology Richland, Washington, LLRW Site.

		(1	Units of µCi/cc)		
Location	1st Quarter	2nd Quarter	<u>3rd</u> Quarter	4th Quarter	Average
VW#100	1.44 ± 0.13 E-6	-2.23 ± 9.51 E-8	9.19±9.39 E-8	3.77 ± 9.58 E-8	3.87 ± 2.10 E-7
VW#101	2.03 ± 0.01 E-4	2.14 ± 0.01 E-4	2.16 ± 0.01 E-4	$2.39 \pm 0.01 \text{ E}-4$	2.18±0.01 E-4
VW#102	4.50 ± 0.02 E-4	4.22 ± 0.02 E-4	4.22 ± 0.01 E-4	4.05 ± 0.01 E-4	$4.25 \pm 0.02 \text{ E-4}$

Vadose Zone Tritium Measurements

Source: Palmer, A.J., and Ledoux, M.R., 1994, Annual Environmental Monitoring Report for Calendar Year 1993: US Ecology Richland, Washington Low-Level Radioactive Waste Disposal Facility: Richland, WA, US Ecology.



Figure A-1. Map of US Ecology Richland, Washington, LLRW facility.

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Figure A-2. Vadose zone monitoring well log.

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THE PROPORTION OF WATER AS VAPOR AND AS LIQUID IN WARD VALLEY SOIL

Introduction

The objective of these calculations is to estimate the relative amounts of water that exist as vapor and as liquid in the unsaturated zone beneath the proposed Ward Valley low-level radioactive waste disposal site.

<u>Analysis</u>

Data from the licensing documents are: volumetric moisture content is approximately $7\%^1$; the soil temperature is approximately 23° C (296 K)²; the soil porosity is $30\%^3$; the vapor pressure of water is 21.07 torr.⁴ We use the common gas law, PV = nRT. One atmosphere of pressure is 760 torr. The molecular weight of water is 18.016 g/mol. The gas constant R is 0.0821 L-atm/mol-K.

First we consider 1 L of saturated air in the soil, and calculate its water content.

$$n = \frac{PV}{RT} = \frac{21.07 \text{ torr} \left(\frac{1 \text{ atm}}{760 \text{ torr}}\right) \times 1 \text{ L}}{0.0821 \frac{L \times \text{ atm}}{\text{mol} \times \text{K}} \times 296 \text{ K}} = 1.14 \times 10^{-3} \text{ mol } \text{H}_2\text{O}$$

$$1.14 \times 10^{-3} \text{ mol } H_2O\left(\frac{18.016 \text{ g}}{1 \text{ mol}}\right) = 0.026 \text{ g} H_2O \text{ vapor in } 1 \text{ L air}$$

This is the amount of water vapor in 1 liter of air under these conditions.

¹ Dames & Moore, Inc., 1994, Wilshire Report Analysis, p. 5: Santa Ana, CA, Dames & Moore, Inc.

² Seracuse, J.M., Lappala, E.G., and Ohland, G.L., 1990, Supplemental Unsaturated Zone Data Report, Vol. I, California Low-Level Radioactive Waste Disposal Project, Ward Valley, California, Figure 25 - Temperature and Water Potential Versus Depth, Seasonal Plot (Administrative Record, p. 128-00278): Tustin, CA, Harding Lawson Associates.

³ *Ibid.*, Table 11 - Soil Characteristics of WV-1SB-1 Soil Samples (Administrative Record, p. 128-00066); License Application, Table 2420.B-7 and Table 6120.A-8.

⁴ The vapor pressure (torr or other units) depends on temperature. See, e.g., Handbook of Chemistry and Physics table on "Vapor Pressure of Water."

Now we apply this result to 1 cubic meter (1,000 L) of soil, using the porosity and fraction by volume of the soil which is air:

30% void (porosity) = 1,000 L × 0.30 = 300 L
7% liquid H₂O (by volume) = 1,000 L × 0.07 = 70 L
Air volume = 300 L - 70 L = 230 L
230 L
$$\left(\frac{0.0206 \text{ g H}_2\text{O}}{1 \text{ L}}\right)$$
 = 4.74 g H₂O as vapor in 1 m³ of soil at 23°C

This is the amount of water present as vapor in 1 cubic meter of soil.

The ratio of the amount of water as liquid to the amount as vapor is:

Ratio =
$$\frac{70 \text{ L liquid H}_2 O\left(\frac{1,000 \text{ g}}{1 \text{ L}}\right)}{4.74 \text{ g H}_2 O \text{ as vapor}} \approx \frac{14,800 \text{ units of liquid}}{1 \text{ unit of vapor}}$$

Conclusion

There are about 15,000 times more water molecules in the liquid phase than residing as vapor in the unsaturated zone beneath the Ward Valley site. While the rate of migration may be faster as a gas than as a liquid, the flux of water in the vapor is very much smaller owing to the great density difference between the phases. The molecules of water vapor may move relatively quickly, but that cannot compensate for the fact that for every molecule in the gas phase there are 15,000 molecules of liquid. Measurable tritium concentrations in soil moisture at 100 feet beneath the surface seem very unlikely to have resulted from vapor migration, in part because of the 1:15,000 ratio of vapor to liquid in the soil.

QUESTION: WHAT IS THE PROBABILITY THAT THE FINDING OF TRITIUM IN THE UNSATURATED ZONE BENEATH WARD VALLEY IS SPURIOUS?

Background

US Ecology found measurable tritium in samples of soil pore water taken at different depths in different locations beneath the proposed Ward Valley radioactive waste disposal site. Of the 14 measurements taken, only one measured zero within an error probability of one sigma. Four measurements were taken at approximately 100' beneath ground surface. The one finding of zero tritium was discovered at this depth, the rest being positive for tritium. The finding of measurable tritium at depth, in concentrations one-seventh that at the surface, given the 12.3-year half-life of tritium, indicated that tritiated water had migrated at least 100' downward in a few decades. This in turn called into question the transport model of project proponents, which predicted migration rates on the order of thousands of years (e.g., see US Ecology Interrogatory Response No. 0600A6151.1.7).

If the transport model employed by US Ecology were right, or if the assumptions made by Prudic (1994) of very low (mm/yr) migration rates were right, there should be no measurable tritium at any of these locations, all well below the root zone. Gas phase migration is extremely unlikely to be able to get sufficient flux to these depths (see accompanying analysis, "The Proportion of Water as Vapor and Liquid in Ward Valley Soil"). Thus the discovery of tritium beneath the site of the proposed Ward Valley facility is of substantial importance.

During the discussion of these findings at the National Academy of Science Ward Valley Panel meeting in Needles, California, one of the panel members asked whether the reported error rate represented one or two sigma. When informed that the values were one-sigma, a question arose about the statistical reliability of the finding of tritium beneath the site. That issue is readily resolved via the following treatment.

<u>Questions</u>

Question One: What is the likelihood that all of the positive tritium measurements are spurious? Question Two: What is the likelihood that the three positive tritium measurements at 100' beneath ground surface are spurious?

Date	Air Piezometer	Sample Depth (feet)	Tritium Value* (Tritium Units)
6/26/89	GB-1	21.5	1.39 +/- 0.57
6/26/89	GB-1	35.0	1.72 +/- 0.51
6/7/89	GB-1	60.0	0.74 + - 0.41
6/6/89	GB-1	99.5	-0.01 +/- 0.58
5/6/89	GB-4	16.5	5.60 +/- 0.37
5/7/89	GB-4	58.0	1.37 +/- 0.49
5/7/89	GB-4	99.7	1.02 +/- 0.33
6/2/89	GB-4	16.5	6.00 +/- 0.72
6/3/89	GB-4	58.0	1.15 +/- 0.66
6/4/89	GB-4	99.7	1.18 +/- 0.54
6/24/89	GB-6	18.5	3.94 +/- 0.73
6/23/89	GB-6	33.0	2.07 +/- 0.89
6/23/89	GB-6	59.0	1.38 +/- 0.62
6/16/89	GB-6	99.7	1.66 +/- 0.39
		Air Moisture Sam	ple 6.91 +/- 1.03

Analyses

These questions can be answered through binomial probability analyses (see, e.g., Johnson, 1980, pp. 188-197). The first step in answering Question One is to calculate the binomial coefficient, which is used to calculate the number of ways that exactly x "successes" can occur in a set of n trials. This is done using the formula:

$$\binom{n}{x} = \frac{n!}{x!(n-x)!}$$

where n = the number of trials, and x = the number of "correct" or "successful" occurrences. (Note that for this analysis, since we are testing to see if the assumption that there is no tritium
beneath Ward Valley is correct, a "correct" or "successful" occurrence would be when the measured tritium was within one sigma of zero.) Thus,

$$\binom{n}{x} = \binom{14}{1} = \frac{14!}{1!(14-1)!} = 14$$

The next step is to determine the probability of only one measurement within one sigma of zero tritium beneath Ward Valley if, in reality, there is no tritium beneath Ward Valley. This determination is made using the binomial probability function:

If, for a binomial experiment, p is the probability of success and q is the probability of a failure on a single trial, then the probability P(x) that there will be exactly x successes in n trials is

$$P(x) = \binom{n}{x} \cdot p^{x} \cdot q^{n-x} \text{ for } x = 0, 1, 2, \dots, n$$

For both questions, since the associated error was identified as one sigma, the probability of a correct measurement is 68.26%. Accordingly, the probability of an incorrect measurement is 1 - p, or 31.74%. Thus, for Question One,

$$P(1) = {\binom{14}{1}} \cdot (0.6826)^{i} \cdot (0.3174)^{14-i}$$
$$= 14 \cdot 0.6826 \cdot 3.32 \times 10^{-7}$$
$$= 3.17 \times 10^{-6}$$

Thus, there is less than a four-in-a-million chance that only 1 of the 14 tritium measurements came out to be zero (i.e., within one sigma of zero) if all of the tritium measurements should have been zero.

To answer Question Two, a similar analysis can be performed:

$$\binom{n}{x} = \binom{4}{1} = \frac{4!}{1!(4-1)!} = 4$$
$$P(1) = \binom{4}{1} \cdot (0.6826)^{1} \cdot (0.3174)^{4-1}$$

 $= 4 \cdot 0.6826 \cdot 0.031975752024$ = 0.0873

Thus, there is less than a 9% chance that the three positive tritium readings at 100' beneath ground surface at Ward Valley were spurious.

This analysis can be expanded to assess the probability of spurious tritium findings with an error rate of two sigma (i.e., when p = 0.9544 and q = 0.456). Only 3 of the 14 tritium measurements are within two sigma of zero. The probability of 11 spurious readings with a two sigma error would be:

$$\binom{n}{x} = \binom{14}{3} = \frac{14!}{3!(14-3)!} = 364$$
$$P(3) = \binom{14}{3} \cdot (0.9544)^3 \cdot (0.0456)^{14-3}$$
$$= 364 \cdot 0.8693 \cdot 1.77 \times 10^{-15}$$
$$= 5.61 \times 10^{-13}$$

Thus, for a two sigma analysis, there is less than **one chance in a trillion** that all of the positive tritium readings beneath Ward Valley were spurious.

Conclusion

The probability, with 13 of 14 samples positive for tritium at one-sigma, that there is in fact no measurable tritium at any of those locations is less than four in a million. Even if one ignores the readings at depths 16.5 to 60 feet and focuses only on the measurements at 100 feet, with three of the four measurements at 100' positive for tritium at one-sigma, the probability that all four of those samples are in fact devoid of detectable tritium is less than 9%. The probability, with 11 of the total 14 measurements positive for tritium at two-sigma, that there is in fact no measurable tritium is less than one in a trillion.

The possibility of measurement error always remains. The logical way of addressing that question is to replicate the measurements, preferably using far more sensitive techniques. Lawrence Livermore National Laboratory has developed a technique for using mass spectrometry that has sensitivities several orders of magnitude lower than traditional techniques for measuring tritium. If there is any question about possible measurement error or cross-

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contamination, one should resolve the issue by replicating the measurements, using more sensitive techniques.

References

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Prudic, D.E., 1994, Estimates of Percolation Rates and Ages of Water in Unsaturated Sediments at Two Mojave Desert Sites, California - Nevada: U.S. Geological Survey Water-Resources Investigations Report 94-4160, Carson City, Nevada, 10 p.

CHELATION AND K_d VALUES: THE EFFECT ON RADIONUCLIDE MIGRATION

[T]he presence in the waste of complexing agents such as organic chelates used in decontamination operations and natural organic acids from the soil promotes the formation of strong complexes with certain radionuclides that reduce the adsorption capacity of the . . . soil for the radionuclide.

(Means and others, 1978, p. 1477)

[L]aboratory-derived K_ds did not reflect the effects of long-term leaching of soiladsorbed radionuclides by natural or manmade organic complexing substances to which the in situ soils were exposed. The in situ K_ds more accurately express the sorption capacities of the mobile forms of these radionuclides, which migrate at much faster rates than predicted from laboratory-derived K_d values.

(Fruchter and others, 1985, p. 8, emphasis added)

"[T]he effect of complexants on toxic element sorption should be examined with soils from specific proposed commercial low-level waste disposal sites."

(Swanson, 1984, p. 2)

Introduction

It has long been recognized that the presence of naturally occurring organic complexing agents, either in soil or the waste itself, or stronger chelating compounds such as EDTA used in nuclear operations for decontamination of piping and the like, causes the formation of highly stable metal complexes that dramatically increase mobility of radionuclides in soil. These chelated radionuclides have a radically reduced distribution coefficient or soil retention factor (K_d) and will migrate very much faster in soil than in their non-chelated state. Far more rapid migration of such radionuclides than would otherwise be expected has been observed at numerous radioactive waste facilities (e.g., Maxey Flats, Kentucky; West Valley, New York; Oak Ridge, Tennessee; Chalk River, Canada) and attributed to the presence of complexing agents (Means and others, 1978, Cleveland and Rees, 1981, Akers and others, 1994). A brief

introduction to the chemistry of chelates and the related matter of colloidal-enhanced radionuclide migration is attached hereto as Appendix A.

Wilshire and others (1993, p. 38 and Attachment 8) have pointed out that the potential for chelating agents to contribute to the mobility of radioactive waste at Ward Valley had not been addressed in the License Application and urged serious review of the question.

The distribution coefficient or soil retention factor, K_d , is an essential input in analyzing potential risks of transfer of radioactive materials from the Ward Valley disposal trenches to the surface above, the aquifer below, or, once in the aquifer, to discharge locations at Danby Lake or the Colorado River. The use of inappropriate K_d factors can lead to substantial underestimation of travel times and environmental risk. It must be made clear at the outset that there is no single, universal K_d value for a particular radionuclide. K_d depends upon the chemical composition and other properties of the site soils, the trench leachate, and the chemical form of the radionuclide (e.g., whether it is chelated, and if so, the nature of the chelating agent). To calculate risk, one cannot just pull a K_d value out of a book, or use one for a different site, or use one for a different chemical form (e.g., unchelated) than may be present in the situation of concern. US Ecology, however, did just that.

To estimate K_ds appropriately, one must test the specific radionuclides of importance in experiments on soils from the site in question; one needs to use radionuclides that are complexed using different chelating materials; and test the materials over different concentrations, pH, and associated conditions.

US Ecology used K_ds obtained from other sites (e.g., the U.S. Department of Energy Hanford Reservation) rather than use K_ds based on tests on Ward Valley soils. It ignored the K_ds it did measure using Ward Valley soils. Furthermore, the K_ds it used in its performance analyses were for unchelated materials. All of these actions resulted in the use of K_ds that were grossly too high, resulting in migration rate estimates and consequence conclusions dramatically too low.

In an effort to determine K_ds for use in its performance assessment, US Ecology undertook batch sorption testing on Ward Valley site soils for five radionuclides [cesium-137 (Cs-137), cobalt-60 (Co-60), radium-226 (Ra-226), natural uranium, and nickel (as a proxy for nickel-59)], which were chosen "because they are typical low-level radioactive waste (LLRW) constituents" (License Application, p. 2600-58 and Table 2600.A-10).

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With the exception of uranium, however, these were *not* the radionuclides US Ecology chose to analyze in its performance assessment. The radionuclides that were in the end analyzed were plutonium-239 (Pu-239), iodine-129 (I-129), carbon-14 (C-14), and uranium-238 (U-238) (License Application, p. 6151-12 - 6151-14).¹ Numerous isotopes of importance were ignored. Of greatest importance is the fact that no tests whatsoever on Ward Valley soil were conducted on plutonium, chelated or not.

Neither the batch sorption tests conducted on Ward Valley soil, nor the literature values for other soils upon which US Ecology relied for the K_d values it ended up employing, were based on *chelated* radionuclides. Chelation causes K_d values to drop dramatically. By failing to use K_d s obtained for Ward Valley site soils, and failing to assess the full range of potential radionuclides and chemical forms of concern, the potential transfer to the general environment may have been severely underestimated.

Discussion

The profound effect that chelating agents can have on K_d values is exemplified by the measured effect founds by Means and others (1978, p. 1477):

The following experimentally measured distribution coefficients (K_d) illustrate the pronounced effects that organic ligands have on the adsorption capacity of sediment for trace metals. We determined that the K_d values for Co⁶⁰ in weathered Conasauga shale at *p*H 6.7 and 12.0 were approximately 7.0 x 10⁴ and 0.12 x 10⁴ respectively. In the presence of 10⁻⁵M ethylenediaminetetra-acetic acid (EDTA) the K_d values were reduced to 2.9 and 0.8.

Thus, in the setting analyzed above, chelation reduced the K_d for Co-60 by approximately four orders of magnitude.

The results of such a reduction in K_d on radionuclide transport time can be quite dramatic. Means and others (1978), citing Marsily and others (1977), give the following example. Pu-239 deposited at the bottom of a 500 meter thick geologic formation, in a setting with high sorptive capacity characterized by a K_d of 2 x 10³, could take more than a million years to migrate

¹ When the controversy arose over the tritium finding at depth, an analysis for tritium was performed as well, although, as we have discussed in detail elsewhere (see Cornog and others, 1992, p. 33-47), the model was unable to match the measured data.

500 meters if the geological formation were of low permeability. Given its half-life, the plutonium would have decayed completely by the time it reached the environment. If, however, chelating compounds were present, resulting in a K_d approaching zero, the plutonium could reach the environment in as little as six years. In formations of low to moderate permeability, the plutonium would travel the 500 meters in only tens to several hundreds of years, four to five orders of magnitude faster than if the plutonium were unchelated.

Description of US Ecology's Analysis

Waste Stream Identification and Selection of Radionuclides for Transport Analysis

US Ecology's first step was to attempt to identify the Ward Valley waste stream. US Ecology's waste stream projection is reprinted in Table 1.²

Although the table is titled "Major Nuclides Considered for Analysis," it does not actually represent the nuclides that *were* analyzed in the performance assessment. Apparently, as a careful reading of the title suggests, it represents only those nuclides "considered for analysis," not those that were actually analyzed. In the end, no analysis was performed on 80% of these radionuclides (which, indeed, themselves represent only a small fraction of the scores of radionuclides which will be present in the Ward Valley trenches).

US Ecology proceeded to narrow the scope of its analysis further to just one of the five trenches projected for Ward Valley, the BC30 trench. This is problematic because (1) it ignores a substantial portion of the waste, (2) the wastes for the Class A trenches are not required to be stabilized (FEIR/S, p. 2.1-14), so subsidence of the trench cap, resulting in ponding and cracking and increased infiltration is likely, and (3) the Class A trench caps are just earth fill, with a mere 100 year design life, and without the "intruder barrier and capillary break" built into the BC30 trench cap design (as questionable as may be the effectiveness of the latter cap design) (FEIR/S, Appendix - Response to Comments on Draft EIR/S, p. 96). The more primitive trench cap and lack of waste form stabilization to prevent subsidence, cracking and ponding produce substantially increased potential for downward percolation through the waste, but those wastes

² US Ecology initially presented its waste stream estimates originally in License Application Table 6110-9 and several other tables. Prompted by a DHS interrogatory asking why there were discrepancies between Table 6110-9 and the other tables in the License Application, particularly Table 6151-1 for the BC30 Trench, US Ecology notified DHS that Table 6110-9 was in error and replaced it with Table 566B.1-1, which is presented here (Interrogatory Response Nos. 061B6151.1.7 and 0566B6110.B).

Table 1. US Ecology Ward Valley	Waste Stream F	Projections.
MAJOR NUCLIDES	CONSIDERED	FOR ANALYSIS
** All act:	ivities in c	curies **
NUCLIDE	ACTIVITY	PERCENT
AM-241 C-14 CM-243 CM-244 CO-60	1.391E+1 7.559E+2 7.271E-1 6.714E+0 1.482E+5	<0.1 <0.1 <0.1 <0.1 2.8
CS-137 FE-55 H-3 I-129 NB-94	1.894E+4 1.690E+4 4.868E+6 2.134E+1 3.054E+0	0.4 0.3 92.3 <0.1 <0.1
NI-59 NI-63 NP-237 PU-238 PU-239	1.305E+2 1.191E+4 4.393E-3 2.612E+3 3.448E+3	<0.1 0.2 <0.1 <0.1 <0.1
PU-240 PU-241 RA-226 RN-222 SR-90	8.963E-1 6.925E+4 3.816E+1 3.816E+1 5.746E+2	<0.1 1.3 <0.1 <0.1 <0.1
TC-99 TH-231 TH-234 U-235 U-238	6.105E+0 1.781E+1 4.034E+2 1.781E+1 <u>4.034E+2</u> 5.272E+6	<0.1 <0.1 <0.1 <0.1 <u><0.1</u> 99.2

Source: US Ecology Table 566B.1-1

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were excluded from the groundwater transfer analysis (License Application, Section 6151, and Appendix 6151.A). Wastes of the same longevity are permitted in the Class A trenches as the BC30 trench (License, Appendix A). The 10-fold difference in concentration limits is such that, when coupled with the increased volume of Class A wastes, significant amounts of long-lived materials can be present in the Class A trenches, , where deep infiltration may be more likely because of increased probability of subsidence, ponding, and cracking of the trench cap. Yet these wastes were excluded from analysis.

As seen in **Table 2**, a reprint of Table 6151-1 from US Ecology's License Application section detailing its Analysis of the Transfer Mechanism to Groundwater, the number of radionuclides "considered" in the groundwater transfer analysis shrank by 40% from US Ecology's Table 566B.1-1, the "major radionuclides considered for analysis." Important radionuclides such as plutonium-241 and its major decay product, americium-241, were not included, nor was consideration of the buildup of their inventory resulting from decay of parent nuclides.³

In the end, US Ecology conducted transport analyses on only *four* radionuclides: I-129, C-14, U-238, and Pu-239. (A fifth nuclide, tritium, was later considered when the tritium finding at depth was disclosed and the adequacy of US Ecology's transport model was challenged by USEPA and the California Regional Water Quality Control Board.) The nearly two hundred radioisotopes identified in the waste stream inventory of the License Application (Appendix 6110.D) were reduced to 25 in Table 566B.1-1, then reduced to 15 in Table 6151-1, and finally reduced to the four radioisotopes used in actual transport analysis. In the process, important radionuclides (e.g., more toxic than C-14 or U-238, or mobile than the mobility assumed by US Ecology for unchelated plutonium) were ignored.

Determination of K_d Values

US Ecology, in order to conduct the transport analyses for the groundwater performance assessment, needed to determine appropriate K_d values for radionuclides that would be present at the Ward Valley facility, if opened. After describing its decision to analyze only for C-14,

³ For example, the nearly 70,000 curies of plutonium-241 (Pu-241) destined for Ward Valley will decay into approximately 2,000 curies of americium-241. Unlike Pu-241, which is a beta-emitter with a relatively short half-life of 14.4 years, Am-241 is a highly toxic alpha-emitter with a half-life of 433 years. By ignoring the buildup of Am-241, US Ecology severely underestimated the amount of risk involved with the proposed Ward Valley project.

Isotope	Half-Life (Years)	Activity (curies)	Specific Activity (curies/g)	Kd (ml/g)
Ac-227ª	2.18 x 10 ¹	2.8922	7.23 x 10 ¹	40
C - 14	5.73 x 10 ³	272.04	4.46	0
I - 129	1.57 x 10 ⁷	9.9466	1.77 x 10-4	0
Nb - 94	2.03 x 10 ⁴	2.9577	1.87×10^{-1}	350
Ni - 59	7.50 x 10 ⁴	106.62	8.10 x 10 ⁻²	101
Pa - 231ª	3.28 x 10 ⁴	2.8922	4.72 x 10 ⁻²	40
Pa - 234m ^b	7.65 x 10 ⁻⁴	173.63	2.00×10^{6}	40
Pu - 239	2.41×10^4	3507.80	6.20 x 10 ⁻²	73
Pu - 242	3.76 x 10 ⁵	7.6722	3.93 x 10 ⁻³	73
Ra - 226	$1.60 \ge 10^3$	15.994	9.89 x 10 ⁻¹	200
TC - 99	2.13 x 10 ⁵	2.7589	1.70 x 10 ⁻²	0
Th - 231ª	2.91 x 10 ⁻³	2.8922	5.32 x 10 ⁵	40
Th - 234 ^b	6.60 x 10 ⁻²	173.63	2.32 x 10 ⁴	40
U - 235	7.04 x 10 ⁹	2.8922	1.92 x 10 ⁻⁶	0
U - 238	4.47 x 10 ⁹	173.63	3.35 x 10 ⁻⁷	1

Table 2. Properties of Radionuclides in the BC30 Trench (US Ecology Table 6151-1)

Information on table provided by US Ecology (1989a) with the exception of Kd values for U-235 and U-238 and specific activity data. Kd values for U-235 and U-238 obtained from Langmuir (1981). Specific activity values obtained from Wang (1969).

^a daughter products of U-235 that reach secular equilibrium

^b daughter products of U-238 that reach secular equilibrium

Source: US Ecology, License Application, Table 6151-1.

U-238, I-129, and Pu-239, US Ecology stated that K_{ds} for these radionuclides are shown on Table 6151-1 and were "provided by US Ecology on the basis of adsorption tests conducted on these radionuclides" (License Application p. 6151-14). Despite the extensive discussion of adsorption tests performed by US Ecology using Ward Valley site soils (License Application, p. 6151-7 - 6151-8; p. 2600-56 - 2600-61; License Application Appendix, p. 2600.1A-17 -2600.1A-28), the K_d values used in the transport analysis for carbon, uranium, and plutonium in fact were *not* obtained from adsorption tests by US Ecology using those nuclides and Ward Valley soils.

US Ecology did perform batch sorption tests on selected radionuclides using Ward Valley soils, but *none* of those tests were used in establishing the K_d values US Ecology employed in its groundwater transfer analysis. Indeed, with the exception of uranium, none of the batch sorption tests performed on Ward Valley soils were even for radionuclides considered by US Ecology in its performance assessment.

In License Application Table 2600-9, US Ecology identifies the radionuclides for which sorption tests were performed using Ward Valley soils. They are cobalt-60, cesium-137, nickel, radium-226, and uranium. Yet US Ecology did *no* analysis for transport to groundwater for any of these but uranium. The most important radionuclide for which US Ecology did do a transport analysis, Pu-239, was subjected to no testing whatsoever using Ward Valley site soils.

US Ecology took the plutonium K_ds from published values for an entirely different site, the U.S. Department of Energy (DOE) Hanford Reservation (Grant, 1994, p. 2). US Ecology used a K_d for plutonium of 73 (see **Table 2**; note that Grant, 1994, p. 2, mistakenly says the value used was 72). Grant (1994) reports that plutonium K_ds had been measured in laboratory column and batch tests using Hanford site soils over 15 years ago and that based on those tests, DOE used a K_d value of 71 in a 1987 radionuclide transport analysis for Hanford (as discussed below, this characterization is not correct). Grant (p. 2) asserts that "the values at the two sites should be comparable, since the soils and the environment are *similar*" (emphasis added). The applicability of K_d values based on measurements from soils a thousand miles apart is questionable at best, and exacerbated by the wide disparity that exists between laboratory and *in situ* measurements. For example, Coles and Ramspott (1982) reported measuring extremely rapid ruthenium migration (as fast as tritium) at the Nevada Test Site despite laboratory batch sorption tests on Hanford soil produced K_ds of 40 to 752, yet *in situ* observations found significant mobility. Coles and Ramspott (p. 1236) concluded that K_d values obtained from batch sorption tests "should be used with caution and verified with field radionuclide migration studies or more relevant laboratory studies." They pointed out that an understanding of the chemical speciation of the radionuclides in question, particularly selected actinides, "is needed in order to predict their behavior in a ground-water environment" (p. 1236). This conclusion is especially important in light of US Ecology having used a K_d from another location rather than conducting sorption tests on site soils for the isotopes in question, as well as having failed to use K_ds for plutonium in chelated form.

The significance of this is readily observable from its own table of K_d values (Grant 1994, p. 6 and 8). Asserting that the appropriate K_d value for plutonium at Ward Valley is approximately that used for Hanford soils because they are allegedly "similar," US Ecology then chose a K_d for uncomplexed plutonium at Hanford ($K_d = 71$) based on laboratory tests rather than the K_d of 1.4 measured at Hanford for plutonium migration in the presence of suspected organic contaminants (see Table 3, comparing Grant, 1994, p. 6 and p. 8). Because one does not know the characteristics of those suspected organic contaminants, the Kd for plutonium that is purposely complexed with a powerful chelating agent such as EDTA (e.g., reactor primary coolant decontamination wastes that would go to Ward Valley) is likely to be even lower than 1.4 for Hanford soils. US Ecology chose a Kd of 73 for unchelated plutonium rather than a measured value for those soils in the range of 1 for suspected chelated plutonium. As Grant (1994, p. 1-2) notes, radionuclides with Kds orders of magnitude greater than 1 are "essentially immobile," yet US Ecology nonetheless chose a K_d yielding essentially immobility in the face of measured values for the soils and environment it claimed were similar to Ward Valley that showed very much lower Kds (close to no retardation at all) and very much higher mobility for actual plutonium moving in those actual soils.

As to US Ecology's claim that the K_d it used for plutonium, 73, was the figure used by DOE for calculating radionuclide transport at Hanford in its safety analysis, the document they cite (DOE, 1987, p. S.17-19) directly contradicts that assertion. That study indicated that the K_d for Hanford soils would extend over a range from an upper limit of 71 for dilute uncomplexed plutonium to 0.63 for concentrated complexed plutonium. DOE created a probability density function (pdf) demonstrating that over the range of conditions examined for their assessment, the K_d would be much closer to 0 than to 71, and indeed used a median K_d of 6.0 for their analysis of the no action alternative. US Ecology thus took a figure *higher* than the *maximum* K_d value cited by DOE for dilute uncomplexed plutonium and applied it to wastes for Ward Valley that would contain substantial amounts of powerful chelating agents for which the K_d should be vastly lower. If the DOE data from Hanford were to be used at all-as opposed to measuring actual

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1 a b c c c c c b b a b b c c c c c c c c	Table 3.	Comparison of Kd	Values for Plutonium at the	DOE Hanford Reservatio
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Source	Kd Value (ml/g)	Environment	Comments	Grant's Reference
Grant (1994, p. 6)	71	Unsaturated zone at Hanford Reservation 300 Area.	Kd value said (by Grant) to be used by DOE for Hanford plutonium transport analyses.	DOE (1987).
Grant (1994, p. 8)	1.4	Hanford site wastes.	Addition of suspected organic contaminants.	Knoll (1985, 1969) and Hajek and Knoll (1966) reported in EPA (1978).

 K_{ds} for heavily chelated plutonium using Ward Valley soils-a K_{d} in the range of the 0.63 cited in the DOE report for chelated plutonium would have been the most appropriate estimate.

Table 4 is a reprint of the relevant table from the DOE Environmental Impact Statement (EIS) for Hanford on which US Ecology claims it based both its decision to use a K_d of 73 for plutonium for Ward Valley soils and its defense of that figure for *chelated* plutonium.

Table 4. Kd Values Used in Hanford Plutonium Transport Analyses.

Environmental Impact Assessment	Median	Low (Concentrated Complexed)	High (Dilute Noncomplexed)
Plutonium K _d for No Action Alternative	6.0	0.63	71

Source: DOE, 1987, p. S.17 - S.19.

One readily sees that even were it appropriate to use K_{ds} determined for Hanford soils and apply them to the Ward Valley situation, there is no basis for a K_d of 73 for plutonium if it is combined with a complexing agent. Since the transuranic wastes to go to Ward Valley would be complexed with the strongest and most persistent chelates possible (e.g., EDTA), in order to strip the "crud" from the inside of reactor piping during reactor primary coolant decontamination operations, an appropriate K_d for these heavily chelated wastes headed for Ward Valley would be in the range of the 0.63 cited in the Hanford study, not the 73 US Ecology claims the study recommends.

US Ecology has claimed that the K_{ds} it used are appropriate for chelated radionuclides such as plutonium because the values are assertedly at the lower end of reported values (for unchelated nuclides). Let us examine for a moment the data that do exist regarding sorptive capacity of Ward Valley soil, and whether it is at the low or high end of measured values reported in the literature for other locations and soil types. As indicated above, US Ecology did batch sorption testing using Ward Valley soils for five *unchelated* radionuclides or their surrogates: Co-60, Cs-137, nickel (as a surrogate for Ni-59), radium-226, and uranium. The mean, minimum, and maximum K_{ds} measured are listed in **Table 5** (License Application, Table 2600-9). Also see **Table 6** (License Application, Table 2600-10), which includes a compilation of literature values for K_{ds} for various soils for the same radionuclides, prepared by US Ecology.

When compared to literature values from a range of soils, Ward Valley soils have relatively little sorptive capacity—even for uncomplexed radionuclides. The measured mean value for Co-60 at Ward Valley was 64.88; US Ecology's literature search showed a range of 24 to 800. The mean at Ward Valley for Cs-137 was 166.85, the literature range is from 30 to 3165. Uranium was 6.92 at Ward Valley, and ranges from 0 to 270 in US Ecology's literature compilation.

The most interesting results were for radium-226 and nickel. The mean K_d measured value for nickel at Ward Valley was a surprising 0.20-essentially as mobile as water. This is *below* the range of 5 to 102 US Ecology reports from its literature search. For radium-226, the mean measured value at Ward Valley is 1.98, while the low end of the range US Ecology reports from the literature *begins* two orders of magnitude higher, 200 to 500.

Thus, actual K_{ds} for Ward Valley soils are not only at the very low end of reported literature values, but in two very important cases, nickel and radium, they are far below any literature value reported. When Grant (1994) asserts that K_{ds} for plutonium go far higher than the 73 used in the US Ecology assessment, that is no argument for use of a high K_{d} for Ward Valley. Ward Valley soils, by US Ecology's measurements, are at the very low end of sorptive capacity-even without considering chelating compounds.

Table 5. Mean, Minimum, and Maximum Distribution Coefficients for Ward Valley Soils (License Application, Table 2600-9).

<u>Radionuclide</u>	Mean	Minimum	Maximum
Cobalt-60	64.88	46.57	96.11
Cesium-137	166.85	66.05	266.27
Nickel	0.20	0.06	0.28
Radium-226	1.98	0.14	3.36
Uranium	6.92	3.86	17.56

Table 6. Literature Compilation of Distribution Coefficients (Kd) for Soils (License Application, Table 2600-10).

	Radionuclide	<u>Kd Range</u>	Reference	
	Cobalt-60	24 - 800	7	
	Cesium-137	30 - 3165	1, 2, 3, 4	
	Nickel	5 - 102	6	
	Radium-226	200 - 500	6	
	Uranium	0 - 270	5	
Reference	28:			
1 - Nork e	et al. (1971)	5 - Rancon (1973)		

2 - Parsons (1962) 3 - Isherwood (1981) 7 - Ames and Rai (1978)

4 - Dosch and Lynch (1978)

6 - US Ecology (1989)

License Application, Tables 2600-9 and 2600-10. In Table 2600-9 (shown here as Table 5), US Ecology reported the results of its batch sorption tests, listing the mean and range of Kd values measured for five radionuclides in Ward Valley soils. In Table 2600-10 (shown here as Table 6), US Ecology reported the range of Kd values found for these radionuclides in its literature search. Measured mean Kd values from Ward Valley soils for cobalt-60, cesium-137, and uranium correspond to low-end Kd values in the ranges reported in the literature. The measured mean Ward Valley soil Kd values for nickel (0.20) and radium-226 (1.98) fall below the range values reported in the literature (nickel = 5, radium-226 = 200). Thus, when compared to measurements at other locations, Ward Valley soils appear to have very little sorptive capacity. In its assessment of radionuclide transport to groundwater, US Ecology analyzed the impact of plutonium-239, iodine-129, carbon-14, uranium-238, and tritium relying upon Kd values from the literature. (Source: License Application, p. 2600-60.)

US Ecology chose not to use its own actual measurements of Kds in Ward Valley soil when assembling the table of K_d values to be used in its performance analyses (License Application Table 6151-1). Despite the clear evidence that nickel would be essentially nonsorbed in Ward Valley soils, and a measured K_d value below the minimum it says was reported in the literature, US Ecology used a K_d of 101 (License Application, Table 6151-1), at the very top of the literature range for other soils and 500 times higher than it had measured on Ward Valley soils. The measured very low K_d for nickel at Ward Valley-and failure to use that K_d or even perform a transport analysis at all for nickel-is significant given the very long life of Ni-59 (75,000 years half-life) and the relatively large amount estimated to be present in the Ward Valley waste (130 curies, according to US Ecology's Table 566B.1-1, 293 curies in our estimate (CBG, 1994, Ward Valley waste stream hazards index Table 1), enough to contaminate on the order of half a million acre-feet of water to levels in excess of EPA drinking water standards (CBG, 1994, hazards index Table 2). With that level of toxicity and environmental persistence, a measured K_d in Ward Valley soil of very nearly zero (0.2), even when unchelated, is cause for serious concern. The failure to perform any transport analysis whatsoever on Ni-59 and the reliance, in deciding not to perform such an analysis, on a K_d one hundred times higher than that measured at Ward Valley, are difficult to explain,

For radium-226, US Ecology measured a mean K_d in Ward Valley soils of 1.98, with a minimum measured value of 0.14 and a maximum of 3.36 (License Application, Table 2600-9). Yet it used a K_d of 200 (License Application, Table 6151-1), one hundred times higher than its own measured value. Since US Ecology cites the same source⁴ for all the K_d values it did employ in Table 6151-1, with the exception of uranium, it is reasonable to assume that the same over-estimation of K_d values occurred for the radionuclides for which sorption tests were not performed at Ward Valley (e.g., plutonium). It is clear that Ward Valley soils were measured and found to have very low sorptive capacity for the nuclides tested, and yet far higher K_ds , taken from other soils, were apparently used.

[US Ecology obtained K_d values for uranium-235 and -238 from a source different from the one it used for all others isotopes. For those two isotopes, US Ecology presents a K_d of 0 and 1 respectively. K_ds should be the same for isotopes of the same element, so the use of two different K_ds is clearly in error. The DHS reviewer pointed this out in Interrogatory No.

⁴ US Ecology's cited source of Kd's is, in fact, not a published study but rather an April 20, 1989, letter from one of its own employees, J. Glenn, to D. Hochmuth, an employee of US Ecology's contractor, Harding Lawson Associates (see License Application, p. 6151-13 and 6151-28). It is unfortunate that the document US Ecology cites as its sole source for all the Kds it employed except for uranium is neither available in the published literature nor even part of the public Administrative Record, frustrating efforts to confirm US Ecology's Kd assumptions.

0601B6151.1.7, but US Ecology declined to use a consistent K_d for uranium. The groundwater transport assessment was done only for U-238, using a K_d of 1. Since US Ecology reports a K_d of 0 for U-235, and K_ds should be identical for both isotopes, had that K_d been used in the U-238 transport calculation, 5 to 11 times less soil retention (Grant, 1994, p. 1), and equivalently faster travel times, would have resulted. Note that measurements for uranium K_d in Ward Valley soil based on unchelated uranium produced a low value, so a K_d of 0 for chelated uranium is not unreasonable.]

Additional Concerns About Chelation

Despite assertions to the contrary by Grant (1994), many of the chelating agents expected to be present in Ward Valley wastes-for example, EDTA, widely used in reactor coolant loop decontamination operations-are extremely stable and will not appreciably biodegrade, even over very long periods of time (see Appendix A).

In addition to dramatically increasing mobility of radionuclides in soil, chelating agents also enhance migration out of waste packages. For example, the migration of radioactive materials out of cement is increased by one to two orders of magnitude when chelating agents are present (McIsaac and others, 1991, p. 108). Furthermore, chelating agents also markedly increase uptake and retention of radionuclides ingested. For example, DTPA has been found to increase the amount of plutonium absorbed approximately 700-fold (Baxter and Sullivan, 1972). Although chelates also increase excretion, the net effect is an overall increase in plutonium retained in the body, a 2.7-fold increase in retention for DTPA and a 80-fold increase for citrate. The dose to a person from the same amount of plutonium, for example, would be 3 to 80 times higher if the radionuclide were chelated with either of these complexing agents.

Conclusions

1. US Ecology performed batch sorption tests using Ward Valley soils for only five radionuclides, but then discarded the values it had measured. The measured values were among the lowest reported anywhere in the literature, indicating very low sorptive capacity for Ward Valley soils.

2. US Ecology then used K_{ds} from the literature, values much higher than it had measured for Ward Valley soils, indeed, as much as two orders of magnitude higher.

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3. In the end, US Ecology's performance assessment was, in all but one case, based on different isotopes, for which no Ward Valley measurements of K_d had been made. Only five radionuclides were analyzed; many extremely important ones were not.

4 The K_{ds} US Ecology ended up using, particularly for plutonium, were based not on Ward Valley but Hanford soils, a thousand miles away.

5. Reliance on literature values for Hanford soils, rather than measured values for Ward Valley, is improper. Even so, US Ecology chose the wrong K_ds from the Hanford data-very much higher than DOE recommended be used.

6. None of the K_d values reported in the License Application were for chelated radionuclides, and thus were all too high, greatly overestimating (by orders of magnitude) travel times and retardation factors.

7. Environmental risks were therefore severely under-assessed.

Recommendations

1. K_d measurements should be made on actual Ward Valley soils using a range of such soils from different depths, given the heterogeneity present; using a significant sampling of radionuclides, not just five, but certainly including plutonium; using a range of complexing agents, including strong chelates used in reactor decontamination operations; and measured over a range of concentrations and pH.

2. In the absence of such measurements, transport calculations should be redone done with $K_d = 0$ for uranium-238, and between 0 and 1 for plutonium.

3. Conclusions based on the US Ecology calculations in the License Application, premised as they are on erroneous K_{ds} and the absence of chelation, should be discarded.

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APPENDIX A

A BRIEF DESCRIPTION OF CHELATE COMPLEXES AND COLLOIDS

Historical. – Nearly 250 years ago in Europe a primitive technology was developed to produce pigments for artists' paint and for inks. It consisted of (1) leaching ashes, filtering the solution, and evaporating to dryness (the residue was mostly potassium carbonate); (2) mixing this material with scrap iron and animal byproducts, such as leather, dried blood, and hooves, and then heating strongly; (3) after cooling, extracting the mass with water, filtering, and evaporating to dryness. This yielded crystals known as "yellow prussate of potash," from which pigments such as Prussian blue and Berlin green were made. About a century later, chemists identified "yellow prussate of potash" as a double cyanide of potassium and iron, formulated as $4\text{KCN Fe}(\text{CN})_2$. Brilliant research in the 1890s by the Swiss chemist Werner demonstrated that in these "complex" ion is octahedral, as sketched. Thus the formula was written as K_4 Fe(CN)₆. Hundreds of thousands of complex compounds are known today. The atoms or molecules bonded to the central atom are known as ligands. Aside from octahedral structures, others are linear, triangular, tetrahedral, square, or other geometrical forms. Many biologically active substances, such as hemoglobin and chlorophyll, are complex compounds.



Chelates. – Many ligands are bonded to the central atom by two or more atoms. An example is the oxalate complex of iron, sketched. The oxalate ion is:



Since the iron atom, in the center of an octahedron, has its bonds directed to the six corners, it can accommodate six oxygen atoms, and thus three oxalate ions. When this type of structure was first realized, the oxalate ion, seizing the iron atom in pincers, was likened to the claw of a crab. The name chelate was adopted for such complexes, from the Greek word for claw, *chele*.



Chelates in general tend to be more stable than simple complexes with the same type of atoms. Stability is quantified by the equilibrium constant representing its formation. Thus for the copper ion (Cu^{2+}) and ammonia (NH_3) , the complex is $Cu(NH_3)_4^{2+}$. Its formation constant is 10^{12} . But if four ammonia molecules are bound together into a linear molecule (by replacing some of the hydrogen atoms with organic groups), the ligand becomes:

NH₂CH₂CH₂NHCH₂CH₂NHCH₂CH₂NH₂

On reacting with copper ion, the long molecule wraps around the metal atom and all four nitrogen atoms link to the metal, forming a strongly-bonded chelate. Its formation constant is 10²⁰, a value one hundred million times that of the simple ammonia complex.

Chelates in Reactor Waste. – It is this enhanced stability, among other factors, which enable certain chelating agents to remove the scale of oxides and related materials from the cooling piping of nuclear reactors. The most important chelating agents used are:

EDTA, ethylenediaminetetraacetic acid, $[HOOC-CH_2]_2NCH_2CH_2N[CH_2-COOH]_2$ NTA, nitrilotriacetic acid, N(CH₂COOH)₃ Picolinic acid, alpha-pyridinecarboxylic acid, C₅H₄NCOOH Oxalic acid, HOOC-COOH Citric acid, HOOC-CH₂COH(COOH)-CH₂COOH

In the case of EDTA, loss of the four hydrogen atoms from the -COOH groups permits all four oxygen atoms, as well as the two nitrogen atoms, to bond to a metal. Thus this single molecule is able to form exceedingly stable chelates. The structure of the iron chelate is shown here:



EDTA is resistant to degradation by radiation and to microbial decomposition. It is frequently employed in reactor cleaning cycles. Both oxalic and citric acids degrade by microbial action.

Sorption of Ions by Soil. – Many components of soil are capable of sorbing ions of metals. These include clays such as kaolinite and montmorillonite, zeolites, hydrated iron oxides, hydrated silica and titanium dioxide, and organic matter. The process is quantified by an approximate equilibrium constant (the distribution constant, K_d), which has dimensions of volume per mass unit of metal ion. When this constant is zero, it means no adsorption takes place. The higher the value, the more strongly the metal is adsorbed. The values are affected by the acidity level (pH) and other factors, all of which must be specified.

There is frequently a considerable difference in the degree of adsorption of a metal ion (positively charged) and the same metal in chelated form (generally negatively charged). For example, on kaolinite at pH 6, unchelated cadmium ion (Cd^{2+}) has a K_d value of 15, meaning it is sorbed significantly. In chelated form (CdEDTA²⁻) the value is 4, meaning it is not sorbed nearly as much. In general terms, this behavior can be attributed to the charge at sorption sites on the kaolinite. If these sites bear a negative charge, as is often the case with clays, they attract the positively-charged cadmium ion in unchelated form, but repel the negatively-charged chelate ion. In addition, there is a size effect: The smaller cadmium ion can closely approach a small site, while the larger chelate cannot. These arguments are not always the case, however, and laboratory tests using samples of the soil in question should be conducted. No simple rule applies to all cases. Owing to the porous nature of soil components, equilibrium is generally reached slowly.

Any radioactive waste which contains EDTA or similar chelating agent is more likely to leak dangerous radionuclides than waste without the complexants. This mobilization of radioactive elements is a serious consideration to be taken into account in repositories.

Colloids. – Colloids are aggregates of molecule or ion assemblies which have radii of 50 to 100 nanometers. Ions range up to 1 nanometer in radius. Colloidal suspensions look turbid owing to their property of scattering light. Suspended in water, colloids pass through ordinary filter paper, and require an ultra centrifuge to throw out of suspension. Most often, colloids bear either a positive or negative charge because of adsorbed cations (positive) or anions (negative). Positively charged colloidal particles, on close approach to each other, mutually repel; negative colloids behave the same way. This property leads to stabilization of colloidal suspension. If the charge on a colloid is neutralized by sorption of a suitable ion of opposite charge, the charge on the particles disappears and coagulation takes place; in fact that is how deltas are formed when colloidal silt runs into the ocean, and suspended material precipitates.

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Clearly colloidal particles have large surfaces. If they sorb radionuclides, they are capable of transporting the material great distances. This possibility is another which one must cope with in designing radioactive waste repositories. Test should be made for the presence of colloidal matter in groundwater, and a study of its carrying capacity made.

TRITIUM, RADIONUCLIDE MIGRATION, AND POTENTIAL GROUNDWATER CONTAMINATION¹

Tritium in trace quantities has already been discovered 100 feet below ground surface at Ward Valley. This finding raises serious questions about the adequacy of the environmental analysis conducted by the California Department of Health Services (DHS), the United States Bureau of Land Management (BLM), and US Ecology. In this part of our study, we discuss these issues and question the adequacy of the environmental impact review and licensing process.

US Ecology Failure to Consider Potential Tritium Migration in Its License Application

In a disturbing yet telling omission, US Ecology failed to consider potential tritium migration when analyzing various contamination scenarios in its License Application. Although, by its own estimates, tritium represents the largest amount (in curies) of any radionuclide to be sent to Ward Valley (License Application, p. 6110-16), and almost all of it will be disposed of in the BC-30 trench (Interrogatory Response No. 0536B5120.2), US Ecology failed to include tritium in a list of radionuclides destined for the BC-30 trench (License Application, p. 6151-13; Table 6151-1, reprinted here as **Table 1**). In its License Application, US Ecology presented analyses for four radionuclides, iodine-129, uranium-238, plutonium-239, and carbon-14, but neglected to perform any analysis of tritium. Most stunning, US Ecology misrepresented iodine-129 as "the most mobile radionuclide" and plutonium-239 as "present in the waste at the highest total activity" even though tritium represents the most mobile radionuclide and the highest total activity of the Ward Valley waste stream (License Application, p. 6151-14).

A DHS reviewer noted this omission and asked US Ecology to explain its failure to include tritium in its analyses of contamination scenarios. US Ecology's reply exposed the dangerous assumptions and cavalier manner with which it treated the threat of tritium contamination:

QUESTION (DHS/BLM): Table 6151.1 does not include tritium. Explain the reason for not including tritium in the evaluation releases from the BC-30 trench.

¹ This material is excerpted from the "Report of the Ward Valley Technical Review Panel" (April, 1992). The panel included: Dr. Robert Cornog, Prof. James C. Warf, Daniel O. Hirsch, Dr. Earl Budin, Dr. Joseph Lyou, Dr. Bennett Ramberg, and Thomas Schmidt. The reader is referred to the full report for a more detailed analysis.

Isotope	Half-Life <u>(Years)</u>	Activity (curies)	Specific Activity (curies/g)	Kd <u>(ml/g)</u>
Ac-227ª	2.18 x 10 ¹	2.8922	7.23 x 10 ¹	40
C - 14	5.73 x 10 ³	272.04	4.46	0
I - 129	1.57 x 10 ⁷	9.9466	1.77 x 10 ⁻⁴	0
Nb - 94	2.03 x 10 ⁴	2.9577	1.87 x 10 ⁻¹	350
Ni - 59	7.50 x 10 ⁴	106.62	8.10 x 10 ⁻²	101
Pa - 231ª	3.28 x 10 ⁴	2.8922	4.72 x 10 ⁻²	40
Pa - 234m ^b	7.65 x 10 ⁻⁴	173.63	2.00 x 10 ⁶	40
Pu - 239	2.41 x 10 ⁴	3,507.80	6.20 x 10 ⁻²	73
Pu - 242	3.76 x 10 ⁵	7.6722	3.93 x 10 ⁻³	73
Ra - 226	1.60 x 10 ³	15.994	9.89 x 10 ⁻¹	200
Tc - 99	2.130 x 10 ⁵	2.7589	1.70 x 10 ⁻²	0
Th - 231ª	2.91 x 10 ⁻³	2.8922	5.32 x 10 ⁵	40
Th - 234 ^b	6.60 x 10 ⁻²	173.63	2.32 x 10 ⁴	40
U - 235	7.04 x 10 ⁹	2.8922	1.92 x 10 ⁻⁶	0
U - 238	4.47 x 10 ⁹	173.63	3.35 x 10 ⁻⁷	1

Table 1. Properties of Radionuclides in the BC30 Trench (US Ecology Table 6151-1).

Information on table provided by US Ecology (1989a) with the exception of Kd values for U-235 and U-238 and specific activity data. Kd values for U-235 and U-238 obtained from Langmuir (1981). Specific activity values obtained from Wang (1969).

^a daughter products of U-235 that reach secular equilibrium

^b daughter products of U-238 that reach secular equilibrium

Table 1. Properties of Radionuclides in the BC30 Trench. Although tritium represents the most mobile radionuclide in the Ward Valley waste stream, and despite the fact that according to US Ecology's estimates, in terms of radioactivity, there will be far more tritium than any other isotope, US Ecology failed to include tritium in its initial accident analyses. Tritium, which should be included here as "H-3," isn't even mentioned in the list of radionuclides to be disposed of in the BC30 trench. (Source: License Application, p. 6151-13.)

RESPONSE (U.S. ECOLOGY): Tritium was not considered because of its short half-life (12.3 years) relative to the time of travel of water in the unsaturated zone (*thousands of years*). (Response to Interrogatory No. 0600A6151.1.7, emphasis added).

DHS and BLM Failure to Include Tritium Finding in Draft EIR/S

The failure to analyze for potential tritium migration at Ward Valley is unacceptable in light of the history of tritium leakage at other facilities. As one of the members of the DHS blue ribbon hydrology and monitoring task force wrote:

> Tritium migration has been found in *all* low level waste sites in the U.S. where investigated. The proposed sampling system [designed to monitor tritium migration] will merely verify the well established fact that tritium indeed *always* does migrate.² (emphasis in original)

(Given this well-established threat of tritium migration should the Ward Valley facility be approved as currently planned, we are concerned that the on-site monitoring plan adopted by US Ecology contains essentially no plan of action in case the monitoring reveals that radioactive contamination has exceeded any of the so-called "action levels.")³

While basing almost its entire claim of safety for the Ward Valley site on assertions of radionuclide migration rates of thousands of years, US Ecology possessed hard data from actual measurements at the site that indicated migration rates of decades rather than millennia. These data, and their significance, may never have seen the light of day in the environmental impact review process were it not for the diligence of two government agencies other than those

² Letter from Robert K. Schulz, Ph.D., Department of Soil Sciences, University of California, Berkeley (and member of the Special Committee on Vadose Zone Monitoring) to Mr. Reuben Junkert, P.E., State of California, Department of Health Services, dated 18 October 1990. Released to members of the Special Committee on Vadose Zone Monitoring. See section labeled, "DHS Vadose Zone Interrogatory." See also Schulz, R. K., and others, 1991, August, Tritium Migration Studies at the Nevada Test Site, DOE/NV-345, UC-721, p. 1: Washington, DC, U.S. Department of Energy. In referring to the "high mobility" of tritium, Schulz et al. state, "It is no surprise then, that tritium migration has been found at all commercial Low-Level Radioactive Waste (LLRW) disposal sites studied."

³ The section of the monitoring plan that deals with "action level responses" is only about one page long and contains no specified responses. See US Ecology, 1991, Environmental Monitoring Report for the Proposed Ward Valley California Low Level Radioactive Waste (LLRW) Facility, p. IV-12: Auburn, CA, US Ecology.

conducting the environmental review. DHS and BLM did not mention the tritium finding at all in their Draft EIR/S, which was issued in June 1990. DHS and BLM repeated instead US Ecology's claim of migration times of millennia. These agencies did, however, belatedly disclose the finding in their FEIR/S (see **Table 2**), but only in an appendix⁴ after receiving letters of concern and inquiry from the U.S. Environmental Protection Agency (EPA) (Pardieck, 1990) and the California Regional Water Quality Control Board (RWQCB) (Gruenberg, 1990),⁵ insisting that the matter be included in the EIR/S and resolved:

> One additional concern, somewhat outside the original request for review, is the problem posed by the presence of tritium at depth beneath the Ward Valley site. This elevated background will complicate interpretation of the monitoring data and *raises questions about the validity of the radionuclide transport modeling in the license application*. (Pardieck, 1990; emphasis added)

EPA's Pardieck went on to say:

The most obvious source for this material is tritium from atmospheric nuclear tests in the 1950s and early 1960s carried to depth by the percolation of rainwater. If this is the case, the apparent rapid infiltration of this material raises questions about the validity of US Ecology's vadose zone transport modeling.

⁴ The appendix includes no independent analysis by BLM or DHS, merely reproduction without comment submissions by US Ecology's contractor, Harding Lawson Associates, attempting to explain away the findings.

⁵ Gruenberg wrote, "The DEIR/S does not mention detection of tritium in the vadose zone. The License Application (LA) briefly mentions that tritium was detection from 0-100+ feet in the vadose zone. The extent of penetration of the tritium and the mechanism of transport were not described in the LA in any detail. Discussion of tritium detection should be included in the final EIR/S along with discussion of the relationship between tritium migration and the infiltration rate model presented in the license application." In a letter dated 9 November 1990, from Robert Perdue of RWQCB to Junkert at DHS, questions were again raised about the tritium findings: "The tritium detected at the site must be fully characterized to enable the VZM [Vadose Zone Monitoring] System to be calibrated to background tritium levels. *Sources of tritium migration pathways, and rates of migration should be delineated and compared to infiltration modeling as described in the US Ecology license application. . . . [W]e are extremely interested in whether the computer modeling program used in the license application either forecasted these results or incorporated this information during calibration... (emphasis added).*

ble 2. Tritium Results for Unsaturated Zone Soil Vapor				
Date	Air Piezometer	Sample Depth (feet)	Tritium Value* (Tritium Units)	
6/26/89	GB-1	21.5	1 39 +/- 0.57	
6/26/89	GB-1	35.0	1.72 +/- 0.51	
6/7/89	GB-1	60.0	0.74 +/- ().41	
6/6/89	GB-1	99.5	-0.01 +/- 0.58	
5/6/89	GB-4	16.5	5.60 +/- 0.37	
5/7/89	GB-4	58.0	1.37 +/- 0.49	
5/7/89	GB-4	99.7	1.02 +/- 0.33	
6/2/89	GB-4	16.5	6.00 +/- 0.72	
6/3/89	GB-4	58.0	1.15 +/~ 0.66	
6/4/89	GB-4	99.7	1.18 +/- 0.54	
6/24/89	GB-6	18.5	3.94 +/- 0.73	
6/23/89	GB-6	33.0	2.07 +/-0.89	
6/23/89	GB-6	59.0	1.38 +/- 0.62	
6/16/89	GB-6	99.7	1.66 +/- 0.39	

Air Moisture Sample 6.91 +/- 1.03

*Note: +\-values indicate the amount of accuracy associated with the tritium value.

Source: US Ecology License Application, Table 2420, B-10,

Note: Tritium Units (TUs) are a measure of tritium concentration in water. One TU is equal to 3.2 picocuries per liter of liquid (as opposed to gaseous) water.

Had the EPA and RWQCB neglected to comment on the tritium finding, DHS and BLM may have let the issue drop without further investigation or debate. There are only two explanations of the failure of DHS and BLM to include the tritium finding in the DEIR/S: DHS and BLM either did not recognize the importance of the finding, or they purposefully suppressed the results of the site characterization study in an effort to avoid crincism of the assumptions made in their analysis of radionuclide migration at Ward Valley. Both conclusious give cause for concern. On the one hand, if DHS and BLM did not recognize the importance of the tritinm finding-that the finding basically invalidated the assumptions made by US Ecology in developing its radionuclide migration models-then the ability of DHS and BLM to regulate the Ward Valley contractor must be questioned. On the other hand, if DHS and BLM suppressed the results to avoid criticism, then their ability and willingness to exercise objectivity in licensing and regulating the proposed project must be questioned. Both explanations suggest that the environmental review process undertaken to date has guaranteed neither public safety nor environmental protection at Ward Valley.

What are these measurements?, and why is the failure to address them adequately so disturbing in terms of the potential environmental impact of the proposed project?

Water vapor in soil samples show the presence of tritium as far down as measurements were taken of soil pore water-100 feet below the proposed Ward Valley facility site (Final EIR/S, Appendix - Responses to Comments on Draft EIR/S, Appendix 9 - Evaluation of Tritium Transport).⁶ This finding suggests that radioactive contamination at Ward Valley could represent a much greater threat to public health and the environment than DHS, BLM and US Ecology first assumed. US Ecology discovered the tritium while cunducting a site characterization study in preparation of its License Application. At best, the discovery indicates thut tritium migrated to depths of at least 100 feet in fewer than a few decades. This finding severely undermines the assumption made by US Ecology that tritium would take "thousands of years" to reach the Ward Valley groundwater basin, which is 650 to 700 feet below the surface and about 600 feet below the waste trenches (US Ecology Response to Interrogatory No. 0600A6151.1.7).

Tritium Migration Beneath Ward Valley

When faced with the implications of the data, US Ecology has variously attempted to explain them away while, on occasion, admitting that it does appear tritium can migrate at least 100 feet in 35-40 years rather than the thousand-year tange they had initially asserted:

"Analytic gas diffusion calculations performed by HLA (Harding Lawson Associates, a US Ecology contractor) indicate

⁶ US Ecology took measurements of groundwater from the aquifer about 700 feet beneath the surface, but did not take any measurements between 100 feet below ground surface and the aquifer. The results of the groundwater analyses for tritium reported by US Ecology (License Application, Table 2600.A-5) appear to contradict in part the results reported to US Ecology by its contractors (Amold, 1989; Top, 1989). Three of the 15 samples reported by US Ecology measured above zero within an error rate of one signts. Even assuming that these three positive readings were erroneous, since the detection limit for the technique used for the vadose zone measurements was 0.5 TUs, about half the average level found at 100 feet, "non-detect" findings for the groundwater samples would merely indicate a reduction in tritum concentrations by a minimum factor of two over the 600 foot distance—that is, a minimum of one half-life, or about 12 years travel time, assuming no dilution of tritiated water with non-trituated water. For this reason, "non-detect" readings in the aquifer have little value, which is why we propose additional measurements be made beneath the 100 foot level using more sensitive measurement techniques.

that it is possible that gas diffusion can transport tritium to depths of 100 feet within 35 to 40 years." (License Application Appendix 2420.B, p. 37.)

The migration rate of radionnelides is a key variable in establishing the threat to groundwater at Ward Valley. To determine the rate of tritium migration, it is necessary to consider tritium concentrations in rainwater (i.e., the identified source of the tritium) and the decay rate of tritium. Small quantities of tritium are formed naturally by cosmic ray interaction with elements in the atmosphere. In addition, from about 1954 to 1963, large scale above-ground testing of hydrogen bombs led to elevated levels of tritium in the atmosphere. Since then, atmospheric tritium levels have waned, but they remain somewhat higher than before open-air hydrogen bomb testing began. As a hydrogen isotope, utifium combines with oxygen or exchange with stable hydrogen in already existing water molecules to form tritiated water. DHS, BLM, and US Ecology assume that the tritium found beneath the proposed facility originated in the atmosphere and was then deposited onto the soil as rainwater.

Hydrogen bomb testing of weapons using substantial amounts of tritium-the source of tritium fallout-began in 1954;⁷ the Ward Valley measurements were mode in 1989, 35 years later. Therefore, if the tritium found beneath Ward Valley came from rainwater that contained tritium from fallout from nuclear weapons testing, that tritium must have migrated there in fewer than 35 years.

US Ecology has at times attempted to claim the tritium could not have come from fallout, because the tritium concentration in raitwater during the periods of heavy nuclear weapons testing was considerably higher than that found now betteath the Ward Valley site. Tritiated water from fallout-contaminated rain would, however, as it migrated betteath the site, mix with older water already there, in which utilitin would have decayed substantially.⁸ The new, higher concentration tritiated water from fallout rainwater would dilute with older, subsurface,

⁷ The first U.S. hydrogen bomb test was in November 1952, but it employed liquid denterium as its thermonuclear fuel. In 1954, the U.S. began testing H-bombs using lithium deuteride as the primary thermonuclear fuel; neutron bombardment of the lithium in the midst of the explosion produces large amounts of tritum. The first Soviet H-bomb test was in 1955; Britain, France, and China followed suit in the years thereafter.

⁸ Older wher should have less tritium, because of time for decay. Water that has been separated from the carth's surface for more than about 250 years (20 half-lives of minium) should be essentially "dead water," that is, without measurable tritium. Mixing of new and old water should produce tritium concentrations lower than that for new water alone.

low-concentration soil water. Taking into account this dilution factor, concentrations of tritium in soil pore water at Ward Valley would be *expected* to be lower than that found in rainwater in the midst of nuclear testing.

Unfortunately, in terms of site safety and suitability, assuming the tritium came from pre-hydrogen bomb testing rainfall does little to elongate the possible migration time. Because the concentration of tritium in atmospheric moisture measured at the surface of the Ward Valley site was only about seven times higher than that found 100 feet below, and similar concentrations to that found now at the surface are reported for pre-testing rainfall, migration times of fewer than three half-lives (three periods of 12 years, or about 35 years total) would be indicated, even if one did not assume any of the material came from fallout. Assuming that some of the reduction in concentration resulted not from travel time, but from dilution with older water at lower levels, the travel time could be measured in years.

Tritium content in atmospheric moisture at the surface of Ward Valley has been ineasured at 6.9 TUs.⁹ The four measurements made at 100 feet beneath the surface averaged about 1 TU.¹⁰ If that difference were because of decay of the tritium alone that decay would require about three half-lives. (In one half-life, or about 12 years, tritium would have decayed in half, to about 3.5 TUs; after 24 years, or two half-lives, it would be about 1.7 TUs; after 36 years, it would be about 0.85 TUs, less than the average tritium measurement actually found.)

While estimating the longest period for tritium to have migrated 100 feet below Ward Valley is a straightforward process, estimating the shortest possible time between surface contamination and the tritium discovery is much more difficult. The problem is that tritiated

⁹ The California Health and Welfare Agency has claimed that atmospheric tritium has been measured at the Ward Valley surface at 5 to 20 TUs (Gould, 1992). In fact, the measurement was 6.9 TUs (US Ecology License Application, Table 242(1,B-10).

¹⁰ In his lener to Congressman Miller, former Health and Welfare Secretary Russell Gould (1992) misleadingly states, "The records further indicate that the maximum measured tritium concentrations in three bore holes at 100 feet were about 1 TU at two locations and were below detection limits at the third location." By noting the number of borehole sites instead of the number of measurements made, Gould gives the impression that there were only three measurements made at 100 feet. In fact, two samples were taken from one of the boreholes. Thus, there were four samples taken at 100 feet, three of which were above 1 TU. Specifically, outside the one measurement of z4 ro, the actual findings were $1.02 \pm 1/-0.33$, $1.18\pm 1/-0.54$, and $1.66\pm 1/-0.39$. Even assuming the maximum error, and that all the errors were in one direction, the measurements are above the detection limit of 0.5 TUs. We have independently checked the reputation of the laboratory that performed the tritium measurements, and found it to be highly respected. We were told that if the lab says it can detect 0.5 TUs, and reports readings above that, one should have confidence in the readings. (This does not mean, however, that contamination of samples or other problems could not have occurred before the samples reached the tab. It is possible that the measurements are erroreous, but the only way to test these findings is to take additional, more sensitive measurements, which is what we recommend.)

rainwater is likely to be diluted with older, non-tritiated water found in the soil pores. The extent of old-water dilution of tritiated water below Ward Valley is unknown. Given sufficient dilution, it is possible that tritium migrated to 100 feet below Ward Valley in as little as a few years. In other words, the factor of seven reduction from surface to 100 feet may not have been a consequence of time-that is, three half-lives or so of radioactive decay-but the result of mixing with pre-existing, older soil pore water that has a lower tritium content. Unfortunately, without knowing the amount of dilution taking place in the soil, it is impossible to establish a fully reliable worst-case assessment of tritium migration rates in the soil beneath the proposed facility.¹¹ In addition, one doesn't know how much further below 100 feet ritium is at about 1 TU. The concentration was fairly consistent from about 60 feet on down. If tritium is found below 100 feet beneath ground surface, the travel time could even be faster.

Tritium Migration: Observed Data vs. Simulation Models

After being prodded by EPA and the Water Quality Control Board, DHS and BLM did add an appendix reprinting an analysis by US Ecology's contractor attempting to explain away the tritium findings that contradict the earlier assumption of extremely long migration times. No independent assessment by DHS or BLM is found in the Final EIR/S.

US Ecology has attempted to explain the measured tritium as the result of either liquid phase or gas phase migration, preferring the latter explanation.¹² Whatever the cause of the migration, it appears as though tritium can move at this site very much faster than US Ecology or DHS and BLM had previously assumed.

¹² We have some skepticism about the gas phase migration claim. At best, it is not yet proven. US Ecology would obviously prefer the migration mechanism to be gas phase rather than liquid phase, because if the rapid migration a result of relatively rapid liquid migration downward at the site, then radionuclides other than tritium, particularly those in water soluble forms, could also migrate quickly. Whatever form of migration is occurring, however, does little to provide assurance with regard to the migration of the large amounts of tritium planned to be disposed of al Ward Valley. No matter what mechanism is causing the tritium migration, it appears from the data to be migrating rapidly; because millions of curies of tritium are projected to be disposed of there, rapid migration, in either a liquid or gas phase, is problematic. These problems are magnified to the extent that other radionuclides (e.g., iodine-129 and carbon-14) are also subject to gas phase migration. Note also that, even if US Ecology were right in its claim about gas phase migration, which it says is gradient-driven, large quantities of tritium disposed of at the sure could create a markedly stronger gradient and driving force producing even more rapid migration

¹¹ If the travel time were the maximum indicated by these measurements-35 years for 100 teet-and if the travel time were linear with distance, it could take a couple of hundred years for tritium to reach groundwater. This would be sufficient time for tritium to decay to insignificant levels (with a half life of 12.3 years, 20 half lives would be about 250 years, at which time only a millionth of the tritium would remain). For longer-lived materials such as strontium and cesium, however, it would take about 600 years or more to have the same reduction in amount through decay. If the travel time were shorter than 35 years for 10) feet-because, for example, the reduction in concentration observed resulted not just from radioactive decay but also from dilution with older water, tritium could potentially reach groundwater sufficiently quickly that a substantial portion would remain radioactive.
US Ecology's consultant, Harding Lawson Associates (HLA), attempted to downplay the contradictory findings by presenting a new "worst-case" scenario for possible tritium contamination of the groundwater below Ward Valley. This new "worst-case" scenario is based on HLA's "calibrating" of their previous model with the new data from the actual tritium measurements. In this scenario, however, the threar of contamination is underestimated through reliance on a migration model that conflicts with data obtained from the site characterization study. The failure of their migration model to predict and conform to the observed tritium findings can be seen in the report submitted by HLA on behalf of US Ecology (FEIR/S, Appendix - Response to Comments on Draft EIR/S, Appendix 9 - Evaluation of Tritium Transport). The key figure from that report is reproduced here as Figure 1.

We include here as Figure 2 the tritium measurements in the location in the holes from which the measurements were made. In Figure 3, we have plotted the data from the actual tritium measurements. Tritium concentrations drop from about 7 TUs at the surface to on the order of 1-2 TUs at 35 feet and remain fairly constant as far below surface as soil pore water measurements were made in the unsaturated zone-to the 100 foot below ground surface level.

Now examine the graph prepared by HLA of the same data, included here as Figure 1. (The broken curve we have added and will discuss shortly.) Upon careful review of Figure 1, one finds that the unbroken curve purported by HLA to represent simulated tritium concentration levels fails to match the observed data. For example, at the 100 foot level, where the observed data average just under 1.0 TU, the simulated level is virtually zero. At the surface, where the measured level is 6.9 TUs,¹³ the HLA curve starts not with the measured value but with much higher values that do not even appear as data points on the graph. At the 100 foot level, four measurements were made. One was essentially zero, the other three were all above 1 TU. HLA ignores the three positive readings and projects its curve near zero at 100 feet. Whereas the actual data indicate little if any change in concentration with depth below 35 feet, and measurable readings at the lowest depth studied, 100 feet, HLA asserts that tritium barely penetrates beneath 100 feet. We find this practice disturbing: HLA, under contract with US Ecology, generated a curve that does not fit the data, and then relied upon the model-derived curve for its worst-case environmental estimates rather than relying upon the data at hand-then

¹³ Pre-bomb test concentrations would be expected to be even lower; values as low as 0.5 TUs have been reported (see, Evans, 1966).



Figure 1. Simulated and Observed Tritium vs. Depth Profile, Gas Phase Simulations, With "Best-Fit" Prediction Curve. Harding Lawson Associates (HLA), under contract with US Ecology, "calibrated" its trutum migration computer simulation model to analyze potential migration scenarios in response to unexpected unium findings beneath the proposed Ward Valley waste site. Even after calibration, HLA's model failed to correspond with existing data. HLA assumed that surface tritium concentration levels of 20 TUs when actual data show a surface concentration level of 6.9 TUs. HLA assumed that mitium concentration levels approach 0 TUs at depths of 100 feet beneath ground surface while actual data show an average of about 1.0 TU at 100 feet. A "bestfit" prediction curve based on the observed data is represented here as a dashed line. The "best-fit" curve shows the difference between the assumptions of HLA's tritium migration model and actual tritium findings at Ward Valley. (Adapted from: FEIR/S, Appendix - Responses to Comments on Draft EIR/S, Appendix 9 - Evaluation of Tritium -Transport).

Tritium Findings at Ward Valley (in Tritium Units)

SAMPLE SITE AND DATE



6.9 TUs · Air Moisture Sample

Figure 2. Tritium Findings at Ward Valley. Soil vapor samples for tritium were taken at depths of 100 feet in the unsaturated zone beneath the proposed Ward Valley radioactive waste facility. The findings suggest tritium migration rates of 100 feet in fewer than 35 years, calling into question US Ecology's initial assumptions of tritium migration rates to groundwater of "thousands of years."



Figure 3. Observed Tritium vs. Depth Profile, Data Points Only.

DHS and BLM accepted this fabrication as absolute assurance that radionuclide migration will not threaten the Ward Valley environment.

Using curvilinear regression analyses, we have generated a "best-fit" curve based on the actual data. In Figure 1, we have superimposed that "best-fit" curve, using a broken line, on the HLA graph. One can see that the existing data in no way suggest zero concentrations of tritium beneath 100 feet, as the HLA model simulation purports to demonstrate.

HLA compounds its failure to model accurately the actual below-ground tritium findings by assuming that the ground-level tritium concentration is an abnormally high 20 TUs. This assumption serves two purposes: First, it allows US Ecology to claim that it took tritium as long as 60 years to migrate 100 feet below Ward Valley. If they had to assume more conservative initial tritium levels, such as 5 TUs, then they would have to admit that, given dilution of open air hydrogen bomb testing era levels, tritium could have migrated to the 100 foot level in only a few years. Second, the assumption of an initial concentration level of 20 TUs allows HLA to project a "steeper" concentration reduction gradient, which lends credence to the assumption that actual tritium levels approach zero at the 100 feet below ground surface. The observed data, however, suggest that the tritium concentration gradient levels off just past depths of 30 feet and remains fairly constant all the way down to depths of 100 feet or more.

The 20 TUs ground-level concentration assumption is called into question by the 6.9 TUs air moisture sample collected by HLA at Ward Valley, and by pre-1954 atmospheric tritium concentration measurements and estimates reported in other studies. The authors cited by HLA gave the highest estimate of pre-1954 atmospheric tritium concentrations found in our literature review. We found estimates of 0.5 to 5 TUs (Evans, 1966), approximately 6 TUs (Feinendegen, 1967), and 8 to 10 TUs (Bradbury, 1991), but nothing as high as the 20 TUs cited in Freeze and Cherry (1979) by HLA. In fact, Freeze and Cherry (1979) contradicted their own citation of 5-20 TUs by including a reference to data showing pre-1953 tritium concentration in precipitation in Ottawa, Canada, as less than 10 TUs. Furthermore, Freeze and Cherry relied upon Payne (1972) for the 5-20 TUs figure. Payne, citing Lal and Peters (1962) stated that global average precipitation levels from non-bomb tritium would be "between 1 and 2 tritium units" (p. 100). Payne added that lower concentrations would be expected over oceans and higher values over the continents, and that seasonal variations may occur as well. He stated, "Unfortunately, few measurements of cosmic ray-produced tritium exist" (p. 100), and went on to say that, in the absence of reliable data, "roughly one can expect" pre-bomb tritium in precipitation "to be in the range 5-20 TUs depending upon location and season of the year"

(p. 100; emphasis added). Thus, HLA's reliance on Freeze and Cherry is not based on actual measurements; Freeze and Cherry relied on Payne, who bemoaned the lack of measurements, and the 5-20 TUs range that was Payne's "rough expectation" depending upon location and season does not imply an expectation that a place like Ward Valley would have 20 TUs on average, even if its location put it at the high end of the range, because 20 TUs was the estimate for the location with the highest pre-hydrogen bomb tritium levels at the time of year when the concentrations were the highest.

It is unfortunate that HLA used the upper end of such a "rough estimate," ignoring much lower actual measurements that are reported in the literature. This is particularly disturbing given that actual measurement taken at Ward Valley of tritium in atmospheric moisture was one third of the value used by HLA for its worst-case analysis assumptions, and that those measurements were taken while tritium from fallout was present (i.e., pre-hydrogen bomb tritium levels at Ward Valley must be assumed to be below the 6.9 TUs measured there now, assuming that the 6.9 TUs measurement was reliable.)

From a standpoint of conservative safety analysis, it is inappropriate to use the least conservative figure (i.e., 20 TUs). Using the actual measurements of atmospheric moisture tritium content at the site itself, the maximum migration period would be approximately 35 years. With dilution of tritiated and older water, or a starting figure based on lower pre-bomb surface concentrations than are found today, tritium may well migrate faster than 100 feet in 35 years.

US Ecology, DHS, and BLM violated a fundamental scientific principle: Given inconsistencies between theory and data, the theory must be revised or abandoned. Here, prior to taking actual measurements, US Ecology assumed that tritium would take "thousands of years" to migrate 650 feet. After the discovery of tritium 100 feet below the proposed facility, US Ecology formulated a "calibrated" model of the rate of tritium migration at Ward Valley:

The effective diffusion coefficient for tritium was *assumed* to be 1.0 ft² / day as *estimated* from the *calibration* of the ATLAS-SOLUTE model to measured tritium concentrations in the unsaturated zone at the Ward Valley site.¹⁴ (emphasis added)

¹⁴ FEIR/S Appendix - Responses to Comments on Draft EIR/S, Appendix 9 - Evaluation of Tritium Transport, Attachment D, Gas Phase Diffusion Modeling Bounding Case Scenario, p. 3.

The calibrated model does not, however, accurately reflect the observed data. Since the model does not fit the data, we conclude that the model is flawed and that we cannot rely upon the assurances offered by US Ecology, DHS, and BLM that radionuclide migration at Ward Valley will not endanger public health or the environment.

Indeed, US Ecology claims, "Measurements of background tritium in water vapor in the vadose zone showed that the concentration of tritium *decreases about one order of magnitude for every 50 feet below ground surface.*"¹⁵ As is readily apparent from the data, however, the concentration of tritium decreases by a factor of about 3-5 in 35 feet, and then remains fairly constant for the next 65 feet. Indeed, there is only a seven-fold reduction in concentration in 100 feet, not the 100-fold reduction US Ecology claims about its own data. Most important, there is no evidence to suggest what happens below 100 feet below ground surface, the greatest depth at which measurements were taken.

The Absence of Tritium Measurements in the Unsaturated Zone Below 100 Feet

US Ecology complicated their already problematic model assumptions by limiting tritium sampling to only the top 100 feet of the unsaturated zone. The failure to measure for tritium in the unsaturated zone at distances deeper than 100 feet below the surface represents perhaps the most troublesome aspect of the site characterization analysis. Without such measurements, there is no way to tell how far or how rapidly tritium has migrated at Ward Valley. This failure leaves open questions about the possibility that tritium migrated rapidly to depths well below 100 feet.

The Need to Use the Most Sensitive Measurement Technique in Future Tritium Sampling

Both the tritium findings and the failure to sample below 100 feet beneath the surface suggest a need for further study of site characteristics at Ward Valley. Should additional tritium sampling take place, an alternative technique would need to be adopted to assure accurate measurement of the extremely small tritium levels that may be found beneath the 100 foot level. There are at least three ways to measure tritium concentrations in water. The first method is to measure tritium levels directly. This method is the easiest, least expensive, and least sensitive. The second method, used by US Ecology to measure tritium in the unsaturated zone, involves

¹⁵ US Ecology, 1991, December 12, Final Environmental Monitoring Report, Version 2, Page III-93, (emphasis added).

enriching a sample by some set amount of tritium, thus boosting the tritium level to one in which relatively sensitive measures can be taken. This method allows researchers to detect tritium levels as low as 0.1 TUs (University of Miami, 1987).

There is an even more sensitive measurement technique that could be used to detect extremely low levels of tritium in soil samples at Ward Valley. This technique is currently being used by scientists at the Sandia National Laboratory branch in Livermore, California, as well as the Lawrence Livermore National Laboratory, to measure amounts of tritium well below 1 TU. The process involves the removal of all helium from a sample. The sample is then left to sit for a time while tritium naturally decays to helium-3. Helium-3 is then measured with a mass spectrometer and, having taken into account the 12.3 year half-life of tritium and the time between initial helium extraction and the time of measurement, one can calculate the amount of tritium present in a given soil sample with high precision and at very low concentrations.

Using a similar technique, Professor Harmon Craig at the University of California, San Diego, says tritium can be measured in concentrations as low as 0.005 TUs.¹⁶ Note that these measurements are *100 times more sensitive* than those DHS and BLM cite as the lower limit of detectability.

Sensitive measurements, well controlled and independently verified, should be made of soil pore water at Ward Valley all the way down to groundwater. One cannot confidently estimate migration times to groundwater without such measurements; the measurements made to date, however, do make it clear one cannot at this time rely on the migration model employed by US Ecology and endorsed by DHS and BLM.¹⁷

¹⁶ Personal communication, 1 April 1992. We confirmed this with Dr. Robert Poreda, University of Rochester, who says there are about eight places in the U.S. that can perform such measurements at approximately those levels. His own laboratory can measure tritium at concentrations as low as 0.01 TUs.

¹⁷ US Ecology, DHS, and BLM have repeatedly claimed that water only migrates upward at the Ward Valley site. They justify this claim in part by citing the annualized average pan evaporation rate at the site (how much water would evaporate from a pan of water kept outdoors all year) compared to the annual rainfall at the site. This homogenization of the data misses the central point that most of the rain occurs during the winter, when the evaporation rate is the lowest, and that substantial storms do occur that result in ponding and downward migration of water (see Figure 4). Note also that the model is based on the rainfall currently at Ward Valley, not a situation in which there has been climate change. Given the multi-thousand year period for which some of these waste remain dangerous, it is not prudent to assume that weather and geological conditions will remain the same for as long as the wastes represent a risk to the environment and public health. Furthermore, the US Ecology model only takes into account rainfall, not the additional water being added to the site by planned routine spraying of water at the site by the company for dust abatement.

The "blue ribbon" panel established by DHS to deal with hydrological and monitoring issues associated with Ward Valley was very concerned about the tritium findings and what these findings implied about the validity of US Ecology's optimistic claims with regard to radionuclide migration rates.¹⁸ Yet, rather than permit the panel to attempt to resolve the matter, DHS "steered" the panel away from the issue (see, e.g., Bianchi, undated). In a letter to DHS, Steve Pardieck (1991), Chief of EPA's Drinking Water and Ground Water Protection Branch (Region IX) stated that the finding of tritium beneath the site is "an issue which has not yet been resolved to our satisfaction." Nor to ours.

¹⁸ Dr. William Bianchi, who served on the DHS panel and was concerned about these matters, is also a member of the Ward Valley Technical Review Panel.



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Figure 4. Climatologic Data for Ward Valley Vicinity. Ward Valley proponents argue that the risk of downward migration of radionuclides at Ward Valley is minimal because the pan evaporation rate exceeds the average precipitation level. Those who use this argument ignore data showing that rainfall in the desert is more likely when temperatures are cool, and evaporation potentials low. As might be expected, pan evaporation rates are higher in the summer months when, on average, precipitation tends to occur during the winter. (Source: US Ecology License Application, Figure 6120-6.)

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