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THE RADIOLOGICAL HAZARDS OF ALPHA - CONTAMINATED WASTE

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ABSTRACT

The radiological hazards of alpha-contaminated wastes is discussed in this overview in terms of two components of hazard: radiobiological hazard, and radioecological hazard. Radiobiological hazard refers to human uptake of alpha-emitters by inhalation and ingestion, and the resultant dose to critical organs of the body. Radioecological hazard refers to the processes of release from buried wastes, transport in the environment, and translocation to man through the food chain. Besides detailing the sources and magnitude of hazards, this brief review identifies the uncertainties in their estimation, and implications for the regulatory process.

RADIOLOGICAL HAZARDS OF ALPHA-CONTAMINATED WASTE

I. Introduction.

The purpose of this review is to briefly assess the state of our understanding of the radiological hazards from alpha-contaminated wastes for the purpose of discussion in the Alpha-Contaminated Waste Workshop.

At the outset, a terminological distinction should be noted between "hazard" and "risk". The term "hazard" implies the existence of some threat to human health or well-being, whereas the term "risk" implies knowing both the exsistence of a threat and its potential for occurrence. The point of emphasizing this distinction is the reminder that the exsistence of a hazard says nothing about the level of risk relative to it. For there to be a risk, and a threat to become meaningful, potential pathways for exposure to man must exist. This potential is expressed as a probability. Failure to make this distiction has added much heat and little light to the nuclear debate. The press protrayal of plutonium as being "fiendishly toxic", for example, invites confusion on this point. The studies of Slovic and his colleagues (1) of the public perception of risks have found that the qualitative aspects of dread, and of catastrophic potential strongly influence a person s risk perceptions. So being told that plutonium is exceedingly toxic (say, on a gram basis), evokes images of disaster attendant on its use or disposal. When lay people are asked to evaluate risks, they seldom

have statistical evidence on which to base judgement. Instead, they rely on inferences based on what they remember hearing or observing about the risk in question. These judgemental rules, known technically as heuristics, are used to reduce difficult mental tasks to simpler ones. While valid in some applications, they can lead to large biases in others, particularly in the case of the "availability" heuristic, which has been studied by Slovic in depth (1). If instances of an event or problem are easily imagined or recalled, then people are apt to judge the frequency or likelihood of it as high. The problem with the availability heuristic is that it is influenced not only by actual recurrence, but also by factors unrelated to frequency of occurence such as vivid images of disaster. Many people expect nuclear power and nuclear waste management to lead to disasters of immense proportions due to such availability effects. Don't be mislead by this apparently unwarrented expectation, however. As Slovic's group point out (1), the psychological reality is that public fears are not based on irrationality. Instead, they reflect normal ways of thinking that, when applied to low-probability, high consequence events, lead to heightened and persistant fears. Besides, experts rely on heuristic thinking too, and are often susceptible to more subtle traps associated with their use. One such is overconfidence in matters of technical judgement. An example is the Rasmussen Reactor Safety Study (2), which has been criticized for using procedures which greatly overestimate the precision with which the probability of core melt could be assessed (3). It may be impossible in principle

to support probability estimates on the order of one in a billion per reactor year with convincing theoreatical arguments. Unwarrented certainty sometimes arise from a techinical person's insensitivity to the weakness of assumptions and lack of hard information. Some examples noted by Slovic (1) of common ways in which experts may lead to overconfidence include: a) failure to anticipate human response to safety measures, b) insensitivity to how a technological system functions as a whole, c) over optimism about the state of scientific knowledge, and d) failure to anticipate human response to safety measures. We in the waste management area would do well to be alert for these pitfalls. There is a further element of overconfidence which is particularly relevant to this review of hazards associated with management of alpha waste. That is, every technology involves risks and benefits, and its attractiveness depends on the probability and size of possible gains and losses. People have difficulty resolving risk/benefit conflicts in even simple gambles. One way to reduce discomfort when uncertainty is introduced in the decision-making process, is to deny that uncertainty. Denial of uncertainty represents as additional powerful source of overconfidence. One form that denial can take, for example, under the pressure of conflict between immediate needs for results by regulatory agencies and the limited state-of-the-art evaluation capability (models and data), is the resort to "conservative" assumptions and computer models. The asumptions of conservatism can be the weakest link in the entire regulatory chain, as was pointed out by the Lewis Committee in the case of the Reactor

Safety Study (4). They observed that it does not follow that because an assumption is more conservative than another, the result is conservatism in an absolute sense. An example from the nuclear waste management area can be found in the Nuclear Regulatory Commission (NRC) environmental impact statement (EIS) on Licensing Requirements for Land Disposal of Radioactive Waste (5). In this EIS, four hypothetical regional disposal facilities are described in which site-specific conditions potentially influencing adverse environmental impacts vary from site to site. But only one "generic" site was selected for analysis and regulation development. The decision rests mainly on the grounds that key parameters such as water percolation, which dominate in groundwater release and transport pathways, are most "conservatively" estimated at one site: the humid southeastern site (5). However, the final waste classification concentration limits, declared by NRC to be the culmination of the Part 61 rulemaking effort (5), are derived from intruder exposure limits. These are dominated (especially in the case of long-lived actinides) by exposures in resuspension pathways, not groundwater pathways, as will be seen in the following review. Hence the apparent conservatism in site parameter selection for modeling groundwater migration leads to overconfidence in having bounded the uncertainties in the rulemaking process, and indirectly to a form of denial of the relative unknowns about the resuspension-inhalation pathways. There is no simple answer to the question of how to avoid this type of overconfidence in the face of incertainties. We will return to this issue in the concluding remarks.

Consider now the main elements of this review, which will be an overview of radiobiological hazards associated with transuranuim elements taken into the body, and radioecological hazards associated with various transport pathways in the environment. Both are parts of the overall radiological hazard of alpha-contaminated waste disposal by earth burial.

II. Radiobiological Hazards of Alpha-emitters

The two routes of entry of radioactive materials into the human body which are of most significance to evaluating the health hazards from transuranics released in the environment from buried waste are inhalation and ingestion. Let us begin with inhalation hazards.

A reasonable approximation to our understanding of the kinetics of inhaled radionuclides is contained in the ICRP Task Group Lung Model (6). In this model, inhaled small particles of actinides such as plutonium and thorium from environmental sources are most often retained in the lung with clearance half-times on the order of 500 days, while others such as uranium, americium, and curium particulates may have large fractions more rapidly cleared with 50 day half-times. Plutonium and other transurancis absorbed from the lung into the bloodstream are deposited principally in the liver and skeleton. Both human and animal data show great variability in relative absorption and depostion in these organs. ICRP has recommended values of 0.45 for the fraction of plutonium deposited from blood to

both bone and liver (6). The human biological half-times in these organs are presumed to be on the order of 100 years for retention in bone, 40 years in liver.

How well have the ICRP model predictions matched human autopsy plutonium data? McInroy's data on plutonium burdens in the general population from fallout sources, shown in Table 1, provide a good source for comparison with Task Group Lung Model predictions (7). The burden in the lung is evidently well predicted by the model; however, redistribution to other organs is not. Note that there is about 35% greater observed liver deposition, and about a factor of 2 less bone depostion than was predicted. But given the many uncertainties in the multitude of parameters in the model, agreement within a factor of two is still good confirmation of the general assumptions of the model.

As a rule, ingestion is a relativily minor route of entry into the body for transuranics, compared with inhalation. The same is not true for other alpha-emitting actinides, such as uranium and neptunium. The variation in reported mammalian gut uptake fractions is quite large, spanning at least two orders of magnitude or more (6). There are several sources of this variation, the most important being: age at exposure, actinide species, mass of actinide ingested daily, and chemical form of the ingested actinide. In young rats, for example, assimilation is about two orders of magnitude greater than adults. While the percentage uptake of ^{239}Pu-nitrate in adult rats on the order of 0.004%, in the neonatal

animal it is on the order of 0.41% (14). By way of reference, the ICRP 30 recommended value for most plutonium compounds is 0.01% (6). Similar, but somewhat smaller increases are reported for uptake of other actinides in the juvenile mammal.

The effects of actinide species on gut uptake is not as pronounced perhaps as with age, but is still significant. The largest percentage uptake of nitrate forms of actinides in the rat for example, is exhibited by neptunium (>1.2%), followed by uranium, americium and curium (0.06%), and last, plutonium and thorium (<0.001%) (13).

These results are complicated by another effect, the effect of daily mass ingested on uptake. A comparison of uptake of nearly equivalent activity, but different masses of plutonium isotopes increasing in the order: ^{237}Pu, ^{238}Pu ^{241}Pu, and ^{239}Pu, indicates that there is a corresponding <u>decrease</u> in uptake with increasing mass ingested. The percentage uptake of nitrate forms in the rate are: 0.12% (^{237}Pu), 0.03% ($^{238,241}Pu$), and 0.04% (^{239}Pu) (13). This effect is particularly significant in the case of small quantities of actinides moving in environmental food chains of man.

The last of these effects, the effect of chemical form due to mineral cycling in plant communities on uptake, is also quite significant. Uranium perhaps most dramatically illustrates the point, since we have human data from both industrial and environmental contexts, and the differences in behavior are large. On the basis of dietary studies, the human gut uptake from food ranges from 2% to

32% (10). These estimates are one or two orders of magnitude greater than both the measured assimilation of nitrate forms, and the occupational exposure limiting value used by ICRP for uptake of U_3O_8 (6, 10). The data on uptake of plutonium shows a dependency on chemical forms as well. The highest range of uptake fractions measured in experimental animals are those associated with plutonium added or incorporated in feed (as opposed to pure chemical forms) (11). In contrast, the expectation that the fraction of "insoluble plutonium" (i.e., high-fired oxide forms) transferred from the GI tract to blood may be less than for "soluble plutonium" by an order of magnitude (15), has not been supported by recent data (13). Particularly when the oxide particles are in the more likely-to-be encountered polydispersed form. In fact the polydispersed "insoluble oxide" of ^{241}Am and ^{244}Cm as well as $^{238,239}Pu$, are all reportedly absorbed about as well as their more soluble nitrate forms in the rat (13). These findings suggest that the value of GI-tract uptake for actinides may have as much as three orders of magnitude of uncertainty associated with it due to the effects of varying chemical form. Furthermore, the use of uptake parameters derived for industrial exposures (or derived maxmium allowed intakes or MPC's based on them) must be used with considerable caution when assessing environmental sources.

The next level of expression of radiobiological hazard of the actinides is in terms of dose delivered to critical organs following inhalation or ingestion.

Fractional uptake, fractional deposition, retention half-times, and radiological decay properties, are all critical parameters which combine in the estimation of dose. A useful tool for making comparisons of hazards among mode of entry (inhalation or ingestion), actinide species, and critical organs is computed rem per microcurie utilizing dosimetric models. A current tabulation of internal dose conversion factors based on inhalation modeling using the Task Group Lung Model, on ingestion modeling using a four-segment catenary model, and on retention in other organs modeled by linear combinations of exponential functions (16), was used to compile the factors shown in Table 2. These computed factors clearly demonstrate that inhalation is by far the more hazardous intake mode from the standpoint of dose per unit intake. But also note that some of the actinides (Am in particular) are more hazardous than plutonium, particularly from the ingestion pathway. This difference is accentuated if the higher gut uptake factors (not taken into account in Table 2 for Np, Am, and Cm) are folded into the assessment. Also the previously demonstrated effects of higher uptake resulting from exposure to small quantities in food sources, and most important, exposure of the young, would tend to drive these simulated dose factors even higher.

To conclude this sequence of considerations relevant to assessing the radiobiological component of the hazards of alpha-emitters, the question of how the many uncertainties discussed might be combined to obtain an estimate of the overall uncertainty in the estimate of health hazard should be addressed. To simply combine

the range of values for separate components would tend to place probably misleading emphasis on extreme values. But, surveying these various elements, particularly fractional gut uptake from environmental sources, and age at exposure, it seems an overall uncertainty in health hazard estimates of at least 2 or 3 orders of magnitude is a reasonable expectation. Others have come to similar conclusions (17).

Note if disposal processes provide effective isolation of waste from the biosphere, and of their transport through various pathways in the ecosystem to man are highly inefficient, then the human biological component of hazard is offset, and conversely, amplified if they are not. So one must carefully examine the effectiveness of the major pathways for translocation of radionuclides from burial waste to man. The measure of pathway effectivness will here be called the radioecological hazard of alpha-emitters.

III. Radioecological Hazards of Alpha-Waste

The factors which are of interest in discussing transport through various pathways to man include corrosion and release from the waste matrix, sorption or retention during groundwater migration, and uptake and accumlation in the food chain of man. But in keeping with the intent here to dicuss hazard, and not risk, the discussion will not cover all the elements of a full-blown pathway analysis. The approach to discussing radioecological hazard will be to consider for each of the major pathways the sequence: release from buried waste, transport mechanisms in the environment, and

finally human exposure.

Consider first the aquatic pathway: release by erosion, followed by sediment transport to surface water, then human exposure via surface water uses. Erosion rates are highly variable across the continent, depending on climate, soils, and man activities. The maximum rate of erosion is experienced in regions of limited rainfall, with decreased rates in more humid as well as more arid climates (18). Sediment yield is a response not only to amount of precipitation, but also to vegetative cover and rainfall frequency. Erosion hazard ranges over almost an order of magnitude.

Transuranics released by this mechanism would be expected to become rapidly attached to small particles, principally either by hydrolysis and precipitation, or by cation exchange reactions with particles or surfaces. These reactions are vital determinants of the ultimate fate of plutonium in the environment. Some nuclides may enter the environment as relatively stable organs-complexes and be transported in soluble form in surface water. Initial chemical reactions and tendencies to remain soluble depend on the initial chemical form of the transuranium element. However, the original source characteristics become less important as time goes on and weathering and aging proceed. These tendencies lead to the generalization noted in a recent review of the research literature (19), that under aerobic conditions, the ultimate behavior of transuranics in the environment, regardless of chemical form, will be determined by processes involving hydrolysis and sorption on particulates or

surfaces, and the formation of soluble complexes with organic and inorganic ligands. In time, the influences of the source diminish.

Next consider release phenomena related to infiltration of precipitation into burial pits. This is the groundwater pathway: leaching of the waste matrix, and hydraulic transport to an aquifer. The process typically begins with the infiltration of water into buried waste. This depends on the characteristics of the cover materials. In some soils there is very little percolation, even with 50 cm to 75 cm of rain, while in other soils, almost all of the precipitation infiltrates to depth. In typical shallow land waste burial conditions, waste are emplaced in a trench in host soil and covered with a cap of material which may be more permeable than the host soil, or less permeable if special precautions are taken. In any case, there is competition in the annual water budget between runoff, infiltration, evaporation, evapotranspiration, and seepage. If infiltration is large, and is not compensated by evaporation and evapotranspiration, this can lead to one of two problems. One is related to conditions involving seepage into waste under conditions of high precipitation and low host soil permeability. If severe enough, this can lead to saturating the trench contents with leachate, and is called the "bathtub effect". When it occurs, there can be a release of leachate at the ground surface. The other problem, more likely related to conditions of high host soil permeability and high precipitation, can lead to leachate migration downward to groundwater.

In addition to site hydrology, the processes affecting the leaching of radionuclides from waste materials are prime factors in the groundwater pathway. Among the processes accelerating leaching are the creation of corrosive, oxidizing substances from the decomposition of organics, and, the action of natural or man-made chelates which tend to bind radionuclides in soluble form. In this case, the characteristics of the wastes themselves, and the conditions of the site favoring microbial decomposition contribute most to the uncertainty in release magnitude. On the other extreme, some solidified waste forms, such as wastes encapsulated in concrete, exhibit releases which are essentially diffusion limited. That is, the radioactive atoms behave much as a cloud of gas slowly expanding away from a concentrated source in a porous medium. Hence the rate of release is governed as much by the diffusion path length as anything else. Effective diffusivity is thus proportional to surface to volume ratio (19). So if and when monoliths of encapsulated waste physically crumble, release rates are greatly accelerated. In this case, waste form stability may be the primary determinant of uncertainty in cumulative fraction of alpha-waste release.

Once transuranics are released to groundwater, transport is subject to convective, diffusive and sorption-disorption processes in the host rock or soil. Thus, for example, uncertainty in groundwater migration hazard is dependent on the ionic species of the radionuclide, which affects geochemical retardation [e.g., uranium (VI) is more mobile than uranium (IV)]. The potentially many

unknowns concerning the physical and chemical nature of the sorptive surfaces along the transport path from disposal site to receptor location may introduce large uncertainties in hazard estimation as well. Of course, groundwater velocity variations (influencing convective transport rate), and groundwater chemistry factors (which affect sorption) introduce uncertainties of their own.

Soil sorptive properties are measured by the distribution coefficient of the radionuclide being transported. This coefficient, K_d, is the equilibrium ratio of the nuclide concentration in the soild phase to its concentration in the liquid phase. As such, it is a lumped parameter, roughly reflecting the combined equilibrium effects of geochemical and physical processes on contaminants in a solid-solution system. It is difficult to generalize groundwater transport hazard across the great variety of rock, soil and groundwater conditions that could be encountered in the field. One would assume that site selection would provide an initial boundary condition on the hazard, which could be limited to some extent by sound site selection. But the fact that we are dealing with very long-lived actinides means that uncertainties arising from transport over great distances and long times must be contended with. With site heterogeneity in mind, it is evident from a comparison among actinides that uranium and neptunium, for example, have a tendency to exhibit low K_d in a variety of settings (Table 3). And moreover, they are susceptible to change in mobility with change in geochemistry (20). Although plutonium often has a higher K_d than uranium or neptunium,

its behavior is hard to predict. It has the potential for switching to forms having very different K_d's. So it belongs with U and Np in a category suggested by Bondietti (20), of elements having chemical behavior such that there is the largest potential for migration assessment error due to unknowns in site conditions. In contrast to these, at least thorium, americium, and curium tend not only to have relatively high K_d, but also to be relatively stable under changing conditions of groundwater chemistry and rock type. Thus, they are classed together as having the least potential for causing errors in migration assessment due to unknowns in geochemistry at a given site.

A third category of release phenomena can be roughly grouped together as biological intrusion. The release process may involve plant root invasion, burrowing animals, or the consequences of human habitation and land use following loss of institutional control. The relative severity of these intrusive processes can, of course, be considerably amplified if the burial trench cover has been partially or entirely eroded away before intrusion occurs. The half-life of many of the actinides is large enough that this possibility could be realized even at relatively low erosion rates.

Once released to the surface terrestrial environment by any of these mechanisms, actinides are subject to transport mainly by the forces of wind and water, but also by biotic activity and mechanical disturbances of man. Contaminants will be entrained in the mineral cycles of the ecosystem. For example, activity released to the soil environment of a site ecosystem can cycle into the atmosphere, plant,

and subsoil compartments which in turn, return it through deposition, plant mortality, and root uptake, respectively.

Of the processes affecting transfer of actinides from soil to plants and animals, physical transport via contaminated dusts dominate over plant root uptake, and uptake from the gut. In man, this is seen in the greater hazard of inhalation compared with ingestion. In the case of plants, this predominance can be seen in a comparison of actinide concentration ratios (i.e., concentration in plant divided by concentration in soil the plant is grown in) in plants grown in the field or in greenhouse conditions.

Concentration ratios in pot cultures of plants are several orders of magnitude smaller than these plants grown in field environments (21). This can also be seen in the observation of one or two orders of magnitude higher concentration ratios in exposed parts of plants (leaves, stems) compared with protected parts (grain) (21). Vegetative components closely associated with the soil (roots, litter, and herbaceous species), exhibit the highest concentration ratios (22).

The migration hazard from vegetable portions of the diet strongly depend, therefore, on whether the edible portion is root, leafy (thus exposed to dusts), or is protected such as a seed. However, harvesting can increase contamination in protected parts such as wheat grains (23), so the distinction has to be used with care in assessing ingestion hazards. It should be noted that bioaccumulation in plants

depends on actinide species as well. Concentration ratios from uptake of neptunium are in the range 10^{-2} to 10^{-1}; for ^{214}Am, 10^{-4} to 10^{1}; for ^{239}Pu, 1^{-7} to 10^{-3} (21). So assessing the hazards of the biologic pathway cannot be conservatively done based just on plutonium behavior in the environment.

A discussion of biological intrusion related hazard would not be complete without mentioning human intrusion. The translation of concentration ratios to concentration depends entirely on activity levels in soil. But whether these are large or small depends decisively on the nature and scope of human intrusion into the waste burial site following loss of institutional controls. This controversial issue cannot be fully explored here; suffice it to say that the effects institutional controls, disposal technology, waste form modification, and so forth, might have considerable effect on the consequences of the potential human intrusion.

IV. Conclusions

It is difficult to put all of the foregoing components of radiological hazards of alpha-wastes together in one collective statement. This is especially so since much has been evaluated in a relative sense, and there are such large uncertainties involved. One possibility for combining the several components is suggested by the approach Healy used to derive a generalized limit for plutonium in the environment (24). The radiobiological hazard of plutonium (dose to critical organs per unit quantity inhaled or ingested), is combined with its radioecological in terms of potential quantities moved

through various pathways to man, by finding limiting concentrations of plutonium in surface soils anywhere such that no more than 500 mrem/yr would be delivered to the critical organ of an exposed individual under normal conditions of land use. Clearly mechanical resuspension of dusts, and growing food crops are exposure modes defining limiting pathways. This will be true whether the surface soil was contaminated by erosion processes, plant invasion, irrigation with contaminated water, or direct human intrusion into the wastes.

The discussion moves toward the grey area between assessing hazard and assessing risk in this process, but perhaps that is unavoidable. Continuing on that line, the relative hazards in terms of dose potential depending on actinide species can be folded in by lowering soil limits by factors of 2.5 to 35 relative to plutonium. Similarly considering the effects of ecosystem transport potential, corresponding limits for other actinides might be pushed down by factors of 2 to 5. The reasoning behind this is as follows: Assuming intake via all pathways from a source in surface soil, one can look at expected concentration ratios of actinides in a critical organ in man such as bone, for a clue as the cumulative effect. Some appropriate autopsy data exists for this. Bondietti (8) has estimated the lifetime accumulation of the actinides in human bone to result in concentration ratios on the order of 0.002 for Th and Pu, 0.005 for Am and Cm, and 0.01 for uranium. Which is to say, about 2.5 times as much Am and Cm may accumulate in bone than Pu, physical and chemical factors assumed constant. Uranium may show a 5-fold greater accumula-

tion. Use of concentration ratios helps integrate many of the uncertainties mentioned earlier. Taken together, these consideration suggest that some actnide species have the potential for both greater accumulation in bone and greater dose per unit intake than plutonium, and thus should have a corresponding lower soil limit.

Naturally, if long-t[illegible] control, deep burial, and waste form modification to control leac[illegible]ng are considered, hazard for some species could be offset by radioactive decay, sorption, dilution, and so forth, and limits go up. Thus, for example, ^{244}Cm with a half-life of 18 years may have a hazard limited principally by the accumulation of daughter products (^{240}Pu).

Finally, then, when a full-blown <u>risk</u> assessment is done, leading to recommended disposal concentration limits in wastes, the final value should reflect judgements on these sometimes conflicting, sometimes additive tendencies with respect to hazard, and their respective uncertainties. All with the aim of ensuring that the manifestation of any of them is unlikely to be realized.

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TABLES

CONCENTRATION OF FALLOUT Pu IN MAN 1974

(McINROY 1981)

ORGAN	Pu CONC. (pCi/kg) AUTOPSY	TGLM
BONE	0.09	0.20
LIVER	0.73	0.54
LUNG	0.14	0.12
LYMPH NODE	2.0	26.5
KIDNEY	0.01	0.02

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ACTINIDE DOSE PER UNIT INTAKE

(REM/ MICROCURIE)

	INTAKE MODE•			
	INHALATION		INGESTION	
NUCLIDE	BONE	LUNG	BONE	LIVER
Th-232	4530.	454	18.	0.02
U-238	2.9	480	11.	0.05
Np-237	4240.	59	87.	16.0
Pu-238	3270.	608	2.1	0.44
Pu-239	4160.	580	2.6	0.49
Am-241	9730.	64	80.	17.0
Cm-244	4560.	67	38.	0.44

• THE UPTAKE FRACTION FOR Np. Pu. Am. AND Cm WERE ASSUMED TO BE EQUAL 10^{-3}

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COMPARISONS OF ACTINIDE SORPTION ON ROCK

K_d, ml/g

SOLID	U(VI)	Np(V)	Pu(IV)	Am(III)
QUARTZ	0.3	0.5	225	1323
BASALT	2.8	0.5	60	1033
SHALE	6.7	49	145	962
GRANITE	6.3	63	320	$3X\,10^5$
BENTONITE/QUARTZ	16	13	160	500

ACTINIDES RELATIVELY STABLE TO CHANGES IN GEOCHEMISTRY: Th, Am Cm

ACTINIDES SENSITIVE TO CHANGES IN REDOX, WATER CHEMISTRY, ROCKTYPE: Pu, Np, U

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