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PLUTONIUM - ITS BEHAVIOR IN NATURAL-WATER SYSTEMS
AND ASSIMILATION BY MAN

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ABSTRACT

There are a number of factors which must be considered in establishing whether or not the inadvertent intrusion of a sizable amount of plutonium-bearing material into a natural-water system may have a significant impact on the health of those individuals who use that system as a drinking-water resource. These factors include the chemical form(s) and solubility of plutonium in natural waters, its behavior in relation to natural processes (geochemical and biological), its fate in water-treatment systems, and its uptake by man from drinking water. From the results obtained in our investigations of the behavior in natural-water systems, it appears that (1) the chemical forms of plutonium dissolved in natural waters are Pu(IV) and Pu(V), (2) the soluble plutonium in many waters is bound to the organic constituents which probably enhances plutonium solubility, (3) the natural process responsible for the removal of plutonium from water is adsorption onto sediments, and (4) in water-treatment systems, soluble plutonium is oxidized to the VI state and this form is not removed. From our

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investigations of gastrointestinal absorption, it appears that the value for f_1 , the fraction transferred from the gut to blood, is surely greater than 1×10^{-3} and may be as high as 2×10^{-1} .

Consideration of these and other factors indicates that the concentration of plutonium could, in certain small natural-water systems, if the intrusion was of a particular nature, approach, and perhaps even exceed the MPC for plutonium. However, the overall effect that this would have on the affected population would be small in comparison with that due to the consumption by this population of radium-bearing well-water at its MPC. In most systems the rates of those natural processes that result in the removal of soluble plutonium from the water are such that the period when the plutonium would be above its MPC would be short.

INTRODUCTION

Support for research on the behavior of radium and the actinide elements in man is, to a major degree, the consequence of (1) the formation of very significant amounts of plutonium and the other transuranics in nuclear reactors, (2) the conclusion, based on radium toxicity in man and plutonium toxicity in animals, that they are hazardous once incorporated, even at very low levels, and (3) the possibility of their release to the environment and transport to man via the food-chain. There is a sharp contrast between the degree of understanding we have about the toxicity of these elements and their behavior once incorporated and that which we have of their behavior in natural systems and the probabilities of their being incorporated. Our understanding of the former is probably superior to that for any other toxicant while our understanding of the latter has, until quite recently, been very sketchy. Investigations to establish the behavior of plutonium in natural systems have been underway for a number of years but the factors which are controlling have only recently been identified and their quantification has just begun; the value of the factor which is most important in environment to man the transport of plutonium from the environment to man, the fraction transferred from the gut to blood, has not, in our opinion, been established.

In this paper we deal with a number of these factors as well as how they relate to the question: Is there a way that plutonium could become incorporated in man in an amount that is in excess of the established limit through consumption of drinking water? In answering this question we will present a summary of our views about the interrelated factors

that control the behavior of plutonium in natural water systems, and consequently the concentration of plutonium in the water. We will also present evidence which suggests that the present value for the gastrointestinal absorption factor of plutonium in man, f_1 , may be three orders of magnitude too low and hence the maximum permissible concentration too high. And finally on the basis of our understanding of plutonium behavior in natural water systems and our speculative value for f_1 we will show that for Lake Michigan, the drinking water resource of millions of individuals and the source of cooling water for a number of nuclear reactors, there is virtually no way in which a serious long-term plutonium contamination problem can arise due to a reactor accident.

PLUTONIUM BEHAVIOR

In the Environment

Present knowledge of the behavior of plutonium in freshwater systems is based, to a major degree, on studies of the behavior of fallout plutonium. Between 1950 and 1962 approximately 6 metric tons were dispersed in the stratosphere (Ha80), and within a very few years it was deposited, rather uniformly, on the surface of the geosphere, mainly in the northern hemisphere.

Our observations of the behavior of this plutonium in freshwater lakes, rivers and the oceans indicates that it is now dispersed in these systems as individual ions, either dissolved in the water or adsorbed onto the surface of sediments, and that it has arrived at a thermodynamic steady-state. The bioavailability of this plutonium has

been maximized; in terms of the ICRP model for plutonium metabolism, it is in every sense a class D compound. The If plutonium, entered Lake Michigan as a result of a nuclear accident, its behavior would, in time, be identical to that of the fallout plutonium. The only phenomena that could occur which would result in a change, other than a proportional increase in concentration in the water and on the sediments, are: the formation of a solid phase, i.e., the solubility of the plutonium bearing compound that is least soluble in the water was exceeded, and/or the adsorption capacity of sediments was exceeded. The probability of either of these occurring appears to be highly unlikely.

Other evidence of plutonium behavior in water systems has been obtained from studies of its fate subsequent to deliberate or accidental discharges of wastes from nuclear processing plants. An example of the former is the $^{239,240}\text{Pu}$ in wastes discharged from the U.K. Windscale Processing Plant to the Irish Sea (Mi70); an example of the latter is the ^{238}Pu , which, as the result of an accident at the Mound Laboratory in 1968, entered an abandoned section of the Miami-Erie Canal (Ro74). In both of these situations it appears that the plutonium is ionic in nature and is in a steady state.

From the results of our investigations (Ne80; Ne78; Ed79), we conclude that the plutonium atoms which enter the water interact rapidly with the natural constituents of the system to establish three competitive equilibria: (1) a complexation reaction with Pu(IV) in which the complexants are primarily dissolved organic materials (presumably humic acids); (2) an oxidation-reduction reaction involving Pu(IV) and Pu(V) in which the oxidant is elemental oxygen; and (3) an

adsorption-desorption process between the Pu(IV) and the suspended and unconsolidated bottom sediments. The plutonium in Lake Michigan is, in comparison with that in most other lakes, bound less strongly to the organics (the concentration of dissolved organic carbon is low) and as a consequence, the fraction of Pu(IV) oxidized to Pu(V) is greater than in most other lakes (the ratio of Pu(V) to Pu(IV) is about 4 while in some eutrophic lakes it is less than 0.1) and the adsorption of Pu(IV) onto sediments is particularly high. Because the concentration factor (concentration in sediments/concentration in water) is 3×10^5 L/kg for total plutonium (2×10^6 for Pu(IV)), only 3% is dissolved in the water. There has been no measurable decrease in plutonium concentration in the water (5×10^{-16} Ci/L) since the first measurements were made (1971). The rate of sediment burial in Lake Michigan is very slow, (the rate of sedimentation is <10 mg/cm²/yr), and the flushing time is about 100 years.

In Water Treatment

In the Chicago and other large metropolitan water treatment plants, Lake Michigan water is chlorinated immediately after coarse filtration and hence the plutonium that is in solution is oxidized to the VI state (La78). Since a low concentration of chlorine is maintained in the water during subsequent treatment and in the distribution system, the behavior of plutonium would be expected to be like that of uranium and hence not be removed. Our analysis of Chicago drinking water has shown this. As there is no known treatment method for removing uranium from drinking water inexpensively, it would appear that the modification of a water treatment plant to remove plutonium would require major changes in

both the treatment method and the plant.

In the Food Chain

It does not appear that the consumption of foodstuff from natural waters such as Lake Michigan would lead to the incorporation of plutonium in man. A number of species of freshwater fish taken from Lake Michigan have been analyzed for plutonium in our laboratory (Ma75) and the results show that the assimilation of plutonium by these fish is insignificant. In no case was the concentration factor (the concentration in the fish divided by the concentration in the water) greater than 30. As these values were for whole fish, the concentration factors for edible flesh are certainly much lower.

When Ingested

The values we have obtained for the absorption of plutonium in drinking water (La81) are much higher than those obtained by earlier investigators. In fasted mice and rats that were administered solutions of either Pu(VI) or Pu(IV) at a concentration of about 1×10^{-10} M (to simulate chlorinated and unchlorinated drinking water, respectively, at the present MPC for ^{239}Pu), the fractional absorptions were 2×10^{-3} and 3×10^{-3} , respectively. In the fasted dog the value was 7×10^{-4} (To82). In fed mice we have shown recently that the value is about 2×10^{-4} (La82). Sullivan et al. (Su79) obtained a value of 4×10^{-3} for Pu(VI) in the fasted rat. The results obtained by Stather et al. (St81) in the hamster show, as do our data, that plutonium absorption is not affected by oxidation state and that there is a factor of 10 difference in absorption between fasted and fed animals. Their values for plutonium absorption in the hamster are, however, an order of magnitude

lower than our values for absorption in the mouse and the rat. In the most definitive and comprehensive of the earlier investigations, Weeks et al. (We56) obtained a value of 3×10^{-5} when 0.01 M nitric acid solutions of Pu(IV) were administered to fed rats, and it would appear that the present ICRP value for f_1 was, to a major degree, based on their values. [Prior to 1978 the value of f_1 was 3×10^{-5} (ICRP72); the most recent value is 1×10^{-4} (ICRP79).] As the value of Weeks et al. for fed rats is a factor of 10 lower than our values for both fed mice and fed rats and a factor of 100 lower than our value for fasted animals, it would appear that a re-evaluation of f_1 is warranted. Particular consideration should be given to man's habits with respect to the consumption of drinking water. Since it is frequently not concurrent with his consumption of food, the values obtained with fasted animals, rather than fed ones, should probably be the basis for setting the value of f_1 .

There is circumstantial evidence to support the view that the value of f_1 for plutonium may be as high as 2×10^{-1} , two orders of magnitude higher than the values obtained for the fractional absorption in fasted mice and rats. The rationale for this view is based on the knowledge that (1) the value of f_1 for uranium is 2×10^{-1} (ICRP79), (2) in the mouse and the rat the values for the fractional absorption of uranium and plutonium are essentially the same (our value for U(VI) is 3×10^{-3} (La79)), and (3) plutonium and uranium are close chemical analogs when their oxidation states are the same. Coupled with this knowledge is the possibility that Pu(VI) and U(VI), the forms present in chlorinated drinking water (La78), are reduced to Pu(IV) and U(IV) by the contents

of the small intestine. (The possibility of either plutonium or uranium being reduced to the III state in the G.I. tract does not appear to be warranted since these reductions in neutral media require a very strong reductant, e.g., elemental zinc.) Soluble iron in the small intestine is surely in the II state, and it is well known that Pu(VI) is rapidly reduced to Pu(IV) by Fe(II). That U(VI) is reduced to U(IV) by Fe(II) in the small intestine is suggested by the results of an in vitro experiment we conducted. When a stoichiometric amount of ferrous chloride was added to a solution that was 0.01 M in U(VI), and 0.1 M in sodium bicarbonate, (pale yellow), reduction to U(IV) (deep emerald green) was immediate. The gastrointestinal absorption data we have obtained is consistent with the view that both Pu(VI) and U(VI) are reduced in the small intestine. Of course, it may be fortuitous that the absorptions of plutonium and uranium in the mouse and in the rat are essentially the same in spite of a difference in their oxidation states in the small intestine.

The consequence of an increase in the value of f_1 for plutonium, from 3×10^{-5} to 2×10^{-1} , would of course result in a corresponding decrease in the MPC, from 5×10^{-9} Ci/L to 7×10^{-13} Ci/L, and a marked increase in the probability of plutonium becoming a significant drinking water contaminant. This topic is addressed in the following section.

REACTOR ACCIDENTS AND NATURAL WATER SYSTEMS

Nuclear Fuel Dissolution Processes

In a discussion of the possibilities of plutonium in a nuclear power reactor entering the environment, and, through a series of events

becoming a constituent of a body of water that is used as a resource for public drinking water, one must have some knowledge of the nature of the fuel itself, the material used to contain it, and the chemical interactions that would have to take place to bring these materials into solution. In the present generation of nuclear plants being operated in the U.S., the fuel is high-fired uranium dioxide in the form of cylindrical pellets (about 1 cm in length and 1 cm in diameter). They are contained (clad) in thin-walled tubes (about 3.5 m in length) of a zirconium alloy (zircalloy). These tubes are in contact with the primary coolant. The primary coolant is separated from the secondary coolant, lake water, by the steam cycle. The discharge of heat is accomplished by either evaporating secondary coolant water or discharging it to the lake. The amount of uranium in each of these fuel elements is about 4 kg and there are between 20,000 and 30,000 of these elements in the reactor. The total amount of uranium is about 100,000 kg. The amount of plutonium in the fuel after protracted reactor operation is about 1,000 kg, or about 2×10^5 Ci. (The specific activity of this plutonium is significantly higher than that of ^{239}Pu due to its ^{238}Pu and ^{240}Pu content.)

The dissolution of this plutonium in natural water would require first the dissolution of the zircalloy cladding, a material that is essentially impervious to water at 300°C in a high irradiation field, and then the dissolution of the uranium dioxide, a material that would dissolve at a very slow rate in most waters (the oxic ones). The dissolution of uranium dioxide in natural water would take place by a process in which dissolved oxygen converts U(IV) to U(VI) and the U(VI)

is complexed by bicarbonate and carbonate ions. The dissolution rate of the plutonium would be the same as that of the uranium since the plutonium that is formed is interstitial in the uranium oxide. The amount of plutonium that would be in solution would depend, of course, on the number of fuel elements involved. It should be noted that in the Three Mile Island accident a significant fraction of the cladding was lost (as the result of a high temperature metal-water reaction) but there was essentially no dissolution of the uranium dioxide in the anoxic cooling water (Nu80).

In summary, it appears highly unlikely that a significant fraction of the fuel in a nuclear reactor would, even in the event of an accident, enter Lake Michigan, but if it did the dissolution rate of the fuel, and hence the introduction of plutonium, would be extremely slow.

Total Dissolution of Nuclear Fuel

In the unlikely event of a nuclear accident in which reactor fuel entered Lake Michigan, the concentration of plutonium in metropolitan drinking waters that would in time result, can be calculate. We know the total amount in the reactor (2×10^5 Ci), the disposition of soluble plutonium in the lake (3% in the water, 97% in the sediments), the fate of plutonium in the treatment plant (no removal), and the volume of the lake (5×10^{15} L). If all the plutonium went into solution, the concentration would be 1.2×10^{-12} Ci/L. This concentration is more than three orders of magnitude lower than the present MPC for plutonium in drinking water, 5×10^{-9} Ci/L ($f_1 = 3 \times 10^{-5}$) and a factor of only 2 higher than 7×10^{-13} Ci/L, the MPC based on a f_1 value of 2×10^{-1} , (the speculative value arrived at in a preceeding section). This simple

calculation neglects two rate factors, that of fuel dissolution in the water and that of water-mixing subsequent to a point source input. Since the rate of dissolution of highly irradiated, clad fuel has been shown to be about 8%/year, even when the fuel has been out of the reactor for some time (Ka80), it is apparent that despite the fairly rapid mixing that occurs, ($t_{1/2} < \text{one year}$) (All82) there would be localized regions of the lake in which the plutonium would exceed the speculative MPC, but not the present value.

Arriving at the conclusion that the plutonium in an existent nuclear reactor does not constitute a threat to drinking water from Lake Michigan obviously does not resolve more general questions such as: What is the consequence of (1) the plutonium in the largest reactor source, a breeder, entering Lake Michigan, (2) the plutonium in an existent reactor entering some other body of water, or (3) some other radionuclide in a reactor entering Lake Michigan? The amount of plutonium in a breeder reactor would be an order of magnitude higher than that present in one of the existent reactors; bodies of water that are used as sources of both reactor coolant and drinking water are generally much smaller than Lake Michigan; the other actinides and the fission products may or may not behave the same as plutonium.

Plutonium Removal by Sediment Adsorption

Subsequent to the introduction of plutonium into a body of water and the establishment of steady-state conditions, the concentration in the water is a function of the amount introduced and of two factors that describe the degree of sediment adsorption: the amount of unconsolidated sediments and the distribution ratio of plutonium (curies

per gram of sediment/curies per gram of water). The concentration is related to size, area of a lake, and/or the volume of water in it, in only a very general way: large lakes and rivers usually have more total unconsolidated sediments than small ones. Although the ratio of the volume of water to the weight of unconsolidated sediments is very high in large bodies of water (in Lake Michigan it is about 1×10^4), the fact that the distribution ratio of plutonium is 1 to 3×10^5 results in most of the plutonium being in the sediments. (In Lake Michigan it is 97%.) Thus, if the volume of water in such lakes were to decrease by a factor of two and the amount of sediments were to remain the same, the concentration of plutonium in the water would not change appreciably. Half the plutonium now in the water would have been adsorbed onto the sediments but this would not have resulted in an appreciable increase in the concentration in the sediments. The amount of plutonium in the water would only increase by a factor of two if the concentration in the sediments increased by a factor of two.

This can also be seen by deriving the equation that expresses the concentration of plutonium in the water in terms that are known, have been measured, or can be reasonably estimated and then substituting actual values into this equation:

$$\text{Water concentration} = \text{curies input} / (\text{lake volume} + \text{sediment weight} \cdot K_d)$$

The fallout plutonium input to Lake Michigan was about 100 Ci, the volume is 5×10^{15} L, the estimated amount of unconsolidated sediments is 5×10^{11} kg, and the measured distribution ratio K_d is 3×10^5

$L \cdot kg^{-1}$. It can be seen from the calculation of water concentration that its value is very insensitive to changes in lake volume, the value of the product of sediment weight and distribution ratio being a factor of about 30 higher than the volume. That the amount of sediments, and not the volume of water is the controlling factor is demonstrated by the higher concentration of fallout plutonium in Lake Michigan than that in Lake Erie (Alb81). The input per unit area to these lakes was the same, and although the volume of water in Lake Erie is a factor of 10 lower than that in Lake Michigan, the concentration of plutonium in the water of Lake Erie is less than that in Lake Michigan. One of the reasons for this is that the amount of sediments in Lake Erie, i.e., those that interact with plutonium in the water, is larger than that in Lake Michigan. The distribution ratios in these two lakes are very nearly the same.

It would appear, that the largest impact on water quality due to an influx of soluble plutonium would occur in rivers. In the Mississippi River, which has a discharge rate of 5×10^{14} L/yr, the concentration of plutonium might exceed the speculative MPC of 7×10^{-13} Ci/L, but would very probably not exceed the present MPC of 5×10^{-9} Ci/L. The rate and degree of plutonium removal from the water by interaction with sediments would surely be higher than they are in Lake Michigan: there is greater mixing of sediments and water, the ratio of amount of sediments to amount of water is much larger, and sediments are continuously transported out of the system (3×10^{14} g/yr). It is obvious that the problems that would arise in smaller rivers would be more severe than those in the Mississippi. Of course, these situations

in rivers are based on the same highly improbable premise that was used above for lakes, namely, all or a very significant fraction of the irradiated fuel from a reactor entering the water.

It must be pointed out that the behavior of other radionuclides, and hence the consequences of their intrusion, may be quite different from that of plutonium. An example is ^{90}Sr . Once the fuel cladding was breached, the strontium would very probably dissolve rapidly (Ka80), since fission-product strontium is not interstitial in the fuel. If all the ^{90}Sr in an operating reactor, about 4×10^6 Ci, were to dissolve in Lake Michigan the concentration would be 8×10^{-10} Ci/L, a factor of 10 higher than its MPC. Unlike plutonium, strontium is not strongly adsorbed onto sediments, but like plutonium, it is not removed in most conventional water treatment systems.

The particular point to be made is this: when considering the impact of plutonium or any other radionuclide on any natural water system, it must be recognized that the degree of the impact will depend on the nature of the system, and that there is in many of these systems a factor (a combination of factors) that may prevent the concentrations from reaching an acceptable level.

CONCLUSION

The probability of all the plutonium in a nuclear reactor entering and dissolving in a body of water such as Lake Michigan is extremely low, but if such an event were to occur, the level in the water would, in relation to an MPC which is three orders of magnitude lower than its present value, be comparable to that for ^{226}Ra in the drinking waters

that a million or so of the people in the U.S. now consume. Concerns about radioactive materials in drinking water, should be directed primarily toward those situations where "high" levels of radioactivity in drinking waters already exist. If there is little or no public concern for the radium problem, a major concern about plutonium in drinking water is surely not justifiable.

In comparing present knowledge of the behavior of plutonium in aquatic systems with present knowledge of the assimilation of plutonium by man, it is our view that much more is known about the former than the latter.

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