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COMPARATIVE FOOD-CHAIN BEHAVIOR AND DISTRIBUTION OF ACTINIDE ELEMENTS IN AND AROUND A CONTAMINATED FRESH-WATER POND

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COMPARATIVE FOOD-CHAIN BEHAVIOR AND DISTRIBUTION OF ACTINIDE ELEMENTS IN AND AROUND A CONTAMINATED FRESH-WATER POND*

ABSTRACT

The bioaccumulation of $^{233,234}U$, ^{238}U, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm in both native and introduced biota was studied at Pond 3513, a former low-level radioactive waste settling basin (0.4-ha surface area) at Oak Ridge National Laboratory. This system, which was decommissioned in 1976 after more than 30 years use, contains approximately 5 Ci of $^{239,240}Pu$; inventories of other actinide isotopes are considerably less. Significantly higher concentrations of actinides in fish that were allowed access to sediments indicated that sedimentary particulates may be the primary source of transuranics to biota in shallow fresh-water ecosystems. Our study determined habitat, in particular the degree of association of an organism with the sediment-water interface, to be the primary factor in controlling transuranic concentrations in aquatic biota. In most of the biological samples analyzed, excluding samples suspected of being contaminated by sediment, $^{241}Am/^{239}Pu$, $^{244}Cm/^{239}Pu$, and $^{238}U/^{239}Pu$ ratios were greater than the respective ratio in sediment while $^{233,234}U/^{238}U$, and $^{239,240}Pu/^{238}Pu$ (aquatic biota only) ratios were not different from the respective ratios in sediment. The relative uptake of actinides from contaminated sediment by aquatic and terrestrial biota at this site was U > Cm ≥ Am > Pu. The relative extractability of actinides from shoreline sediment (using 1 M HNO_3 and 0.01 M HCl) was U > Cm ∿ Am > Pu; we also observed the same relative ranking for sediment-water exchange *in situ*. Concentrations of transuranics in water, terrestrial vegetation, and vertebrate carcasses were less than 10% of the recommended public exposure maximum permissible concentration (MPC) of the ICRP, even in this highly contaminated system which has radioactive inventories atypical of any other fresh-water site now in use or planned for future use in association with nuclear power technologies.

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1. INTRODUCTION

Long-lived synthetic actinides (isotopes of U, Pu, Am, and Cm) are a significant part of radioactive waste from nuclear fuel cycles [1]. In the event of actinide releases to the environment, field studies are necessary to supply information to use in forecasting the long-term biogeochemistry of these elements, many synthetic. Studies of both naturally occurring and synthetic actinides are needed in order to assess environmental effects and human health risks from these elements in food chains. Much ecological research on actinides has dealt with plutonium, as evidenced by its coverage in review papers [2,3,4,5,6]. Other actinides, like U, Am, and Cm, have not been extensively studied from an ecological perspective. Even fewer studies [7,8,9,10] have attempted to make comparisons of the relative bioaccumulation potential of various actinides under field conditions. The research objectives at Oak Ridge on long-lived actinides have been to characterize and compare the chemical and biological behavior of different actinides in the environment and thereby gain understanding that will enable us to predict the long-term fate of actinides released to the environment. These objectives have covered a variety of research efforts on environmental chemistry and bioaccumulation by aquatic and terrestrial biota.

This paper summarizes the environmental chemistry and comparative uptake of isotopes of U, Pu, Am, and Cm by aquatic and terrestrial biota in and around the environs of a former low-level radioactive waste settling pond at Oak Ridge National Laboratory, Tennessee, USA. We believe that the comparative approach taken in these field studies will be useful to forecasting relative similarities and differences in the bioaccumulation of various actinide elements. Of particular interest is the comparative bioaccumulation of actinides by fish and small mammals and the importance of sediment as a source of contamination to biota. Technical details of the methods and results will be presented elsewhere [11,12,13]. Here we discuss the major findings, general trends, and conclusions from these studies which lasted over several years.

2. HISTORY AND SITE CHARACTERISTICS OF POND 3513

Pond 3513 is a former low-level radioactive liquid waste settling basin at Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA. In early 1976 the routine release of radioactive liquid effluent to the pond stopped after over 30 years use. Our studies were conducted from late 1976 through 1978. The pond sediments, mostly greater than 20 cm

deep, are a mixture of natural materials (clays, calcium carbonate) used or produced in the lime-soda-clay waste treatment of liquid radioactive waste [14,15,16]. The clay component of the treatment was predominantly illite with some kaolinite. Organic matter in surface sediments (approximately 7% of dry wt.) is derived primarily from aquatic plants. The pond is shallow (approximately 1 m deep), square in outline, and has a surface area of about 0.4 ha (63 m x 63 m).

The pond is a freshwater system which is chemically and biologically stabilized because chronic radionuclide inputs to the pond are absent. Yet this system contains high concentrations of numerous actinide elements derived from nuclear fuel cycle sources. Before our studies were begun in 1976, the water in Pond 3513 underwent rapid turnover by rainwater runoff. A weir at the pond outlet has slowed the water turnover time to more than 2 years, still entirely due to runoff from rainfall. Effects on the pond biota from nonradiological, toxic pollutants are minimal because the low level radioactive liquid effluent to the pond had a similar chemical composition to that of laboratory tap water prior to lime-soda-clay treatment [16]. The high diversity of aquatic macroinvertebrates observed at the start of our studies support this. The source of actinides to the pond was primarily soluble low-level wastes. Based on radioactivity in sediment cores the inventory of ^{239}Pu in Pond 3513 is approximately 5 Ci [17]. We use ^{239}Pu throughout this paper to refer to both ^{239}Pu and ^{240}Pu. We also use the designation ^{233}U to refer to both ^{233}U and ^{234}U.

Pond 3513 is eutrophic with a mixed species composition of phytoplankton [18]. Benthic filamentous algae and submerged macrophytes are seasonally abundant. Goldfish (Carassius auratus), channel catfish (Ictalurus punctatus), and bluegill (Lepomis macrochirus) were introduced into the pond in May 1977 to supplement an existing vertebrate fauna consisting of frogs (Rana catesbeiana and R. palustris) and possibly some turtles. The pond shoreline is densely vegetated by an approximately 1-m-wide strip of emergent aquatic macrophytes. The gently sloped and grassy banks around the pond are 2 to 3 m wide from the shoreline to top of the pond banks. Cotton rats (Sigmodon hispidus) frequent the densely vegetated pond banks.

3. APPROACH TO COMPARISONS OF ACTINIDE TRANSPORT

To impart only the general tendencies in actinide bioaccumulation, we have rounded the results to one significant digit in this paper. For details about variance within individual sets of data the reader is referred to other papers [11,12,13].

Aside from direct comparisons of dry weight concentrations of actinides in various biotic and abiotic items, we compared actinide dynamics in water, sediment, and biota using isotopic activity ratios and discrimination factors. Each sample of water, sediment, or biota taken from the pond was analyzed simultaneously for concentrations of several different actinide elements [11,12,13]. We calculated each isotopic ratio as the ratio of the untransformed concentration of one actinide to the untransformed concentration of a different actinide in the same sample (for example, dpm per g ^{238}Pu in fish/dpm per g ^{239}Pu in fish). Plutonium concentration data and isotopic ratio data were log transformed prior to analysis. Frequency distributions of radionuclide concentrations in environmental samples tend to be positively skewed and in that respect resemble a lognormal distribution [19,20,21]. Concentration data and isotopic ratio data sometimes contained extreme values that biased the arithmetic mean; therefore, we chose the geometric mean (i.e., exponent of the mean of log transformed data) as the best measure of central tendency.

Discrimination factors (DF) were calculated as the ratio of the geometric mean isotopic ratio in a receptor compartment to the geometric mean isotopic ratio in a donor compartment. Differential transport of actinides from a donor to receptor compartment is indicated when DF values are greater or less than unity. Similar isotopic activity ratios in donor and receptor indicate no differential uptake of the actinides (DF = 1). The discrimination factor approach has been used in previous radioecological studies to compare the food chain transport of Ca and ^{90}Sr as well as K and ^{137}Cs [22].

4. PHYSICOCHEMICAL BEHAVIOR OF ACTINIDES IN SEDIMENT AND WATER

Field measurements revealed a mean pH of surface water in Pond 3513 of 9.1 (range 7.9 - 9.6) with a dissolved oxygen content generally at or above saturation. Pond water is alkaline due principally to high primary productivity and the presence of sedimentary carbonates from the lime-soda-clay treatment process. Actinides in pond water have been determined in both the dissolved (0.22 μm filtrate) and particulate (seston) fractions (Table I). Due to the higher solubility of trivalent ^{241}Am and ^{244}Cm and hexavalent ^{238}U relative to ^{239}Pu, ^{241}Am/^{239}Pu, ^{244}Cm/^{239}Pu, and ^{238}U/^{239}Pu, activity ratios are higher in filtered water than in seston or sediment.

The water soluble Pu in Pond 3513 has been shown to be primarily in the (V) oxidation state ($\geq$ 75%) under normal conditions (i.e., in the absence of massive algal blooms or

other factors which produce reducing conditions in surface sediments) [23]. This finding of the predominance of pentavalent Pu is significant because similar observations of Pu(V) oxidation-state dominance in solution have now been made in Lake Michigan [24], the Marshall Islands [25], and in rainwater [26]. The small fraction of reduced Pu(IV), and the Am(III) and Cm(III) in pond water were found to be associated primarily with naturally produced organics having molecular weights $\geq$ 6000 by means of gel filtration chromatography. Ultrafiltration experiments indicated that $\geq$ 90% of the U passed through a 500 mol. wt filter, probably as a U(VI) carbonate complex [27]. During our studies on Pond 3513, the fraction of Pu(V,VI) in filtered water varied from $\leq$ 10 to > 80% as a result of biological and chemical cycles in the pond. Investigators may expect significant temporal variability in valence behavior. Nevertheless, in four extremely dissimilar aquatic systems, the Marshall Islands [25], the Irish Sea [24], Lake Michigan [24], and Pond 3513, oxidized Pu predominates the soluble form of Pu; dissolved concentrations range from 10^{-13} to 10^{-18} M.

Isotopic activity ratios of transuranics in submerged offshore and shoreline (at water's edge) sediments are shown in Table I. The high actinide concentrations in sediment coupled with the large mass of sediment makes pond sediments the primary standing pool for actinides in the system. We used weak-acid extraction (0.01 M HCl) as an estimate of relative biological availability of actinides from sediment. Extraction of the actinides from shoreline sediment revealed that 44 to 77% of the ^{233}U and ^{238}U, 3 to 5% of the trivalent ^{241}Am and ^{244}Cm, and 0.2% of the ^{239}Pu could be removed with 0.01 M HCl. Based on this acid extraction the rank order availability of actinides from sediment was U > Cm = Am > Pu. Pu in pond sediment is apparently reduced (IV) and not oxidized Pu (V or VI) because oxidized Pu would likely exhibit extractability comparable to U(VI).

4. FATE OF TRANSURANICS IN AQUATIC BIOTA

Concentrations of ^{239}Pu and isotopic activity ratios for the transuranics in aquatic biota are presented in Table II. Animals were usually kept in clean water for more than two days to permit the GI tract to empty. The degree of an organism's association with sediment was important in determining its plutonium concentration. Dragonfly nymphs that crawl through sediments had higher actinide concentrations than invertebrates like water bugs or snails that swim or climb on plants. The high transuranic concentrations in algae were probably due to their high surface to volume ratio. Submerged macrophytic plants and

small shoreline macrophytes, that are occasionally submerged, had higher transuranic concentrations than large emergent macrophytes, which are never submerged, as observed earlier in laboratory microcosm studies [28]. Vertebrate carcasses (whole body minus GI tract and gills) after a 28-week exposure period had among the lowest actinide concentrations found in pond biota.

From the trends in Table II it can be concluded that, as suggested earlier [29], benthic detritivores will be most likely to accumulate high concentrations of actinides because of their habit of ingesting organic sediments. An experiment where some fish were released into Pond 3513 and allowed free access to sediment and some fish were confined in enclosures that prohibited access either to sediment (nets) or to both sediment and contaminated food ("Living Streams") confirms the previous idea [11,29]. Transuranic concentrations in goldfish carcasses and GI tracts were less in fish from the enclosures than in free roaming fish (Table III). This further identifies sediment associations as being important to actinide uptake by fish.

Discrimination factors for aquatic invertebrates, vertebrates, and plants for ^{238}Pu and ^{239}Pu, ^{241}Am and ^{239}Pu, and ^{244}Cm and ^{239}Pu, assuming either pond water or sediment as potential donors to biota, are shown in Figure 1. Using either sediment or water as possible sources there was no appreciable difference in the uptake of ^{238}Pu and ^{239}Pu by pond biota (DF values all approximate unity). Americium-241 was slightly (2-3 times) more available than ^{239}Pu to pond biota when sediment is considered the source of contamination. The opposite pattern is observed if filtered pond water is used as the donor to biota. The uptake of ^{244}Cm by pond biota relative to ^{239}Pu is similar to ^{241}Am since DF values indicate an enrichment of 2 to 5 times more Cm than Pu. Again the opposite pattern is observed if filtered water is used as the donor compartment. It has been concluded that DF values based on pond water are artifacts because direct uptake of transuranics from pond water does not appear to be a significant pathway into biota [11]. Because of the affinity of transuranics for sedimentary particulates, suspended or bed sediments that adhere to invertebrate and plant surfaces and are ingested by vertebrates are the most important source of actinides to pond biota. In many cases only a few mg of sediment sorbed or associated with plants or invertebrates is sufficient to account for their entire actinide concentration. A sediment source would be most important in shallow zones of lakes and in ponds. It is in such areas that maximal uptake by organisms will occur and this will result in maximum potential doses to people from ingesting contaminated aquatic foods. Thus, estimates of biological uptake based on our

data should be conservative (maximal) with respect to values obtained in other, deeper fresh-water systems.

Despite the concentrations of actinides in Pond 3513 sediment, Pu concentrations in fish are among the lowest observed in pond biota. Dry weight concentrations of transuranic elements in fish carcasses are less than 10% of the recommended public exposure maximum permissible concentration of the ICRP [11,30]. Moreover, transuranic concentrations in fish muscle tissue are at least ten-fold less than in fish carcasses, which further lessens the potential dose to humans from ingestion of fish from contaminated aquatic environments. Therefore we conclude that, since concentrations in Pond 3513 biota should be maximized by the nature of the exposure environment, the aquatic food chain transport of transuranics in other environments will not likely result in potentially hazardous radiation doses to people as defined by regulations for the nuclear fuel cycle in the U.S.A. [11,30].

5. ACTINIDE BIOACCUMULATION FROM THE POND SHORELINE

There is a narrow zone of seasonally exposed shoreline sediment around Pond 3513 that is heavily vegetated by emergent aquatic macrophytes. Cotton rats that frequent the densely vegetated pond banks were trapped in 1977 and 1978 along transects parallel to and 3 m distant from the pond shoreline. The turnover rate for the cotton rat population around the pond was about 16 weeks in 1977. High radiocesium concentrations in rats captured along the pond shoreline as well as the presence of runways through shoreline plants during summer indicated that rats had access to contaminated vegetation and sediment [31]. Plutonium concentrations and isotopic activity ratios in rat carcasses (whole body minus pelt and GI tract) and rat GI tracts are presented in Table IV. Rat carcasses from around the pond had ^{239}Pu concentrations approximately 100 times background fallout levels of 0.05 fCi/g dry weight (.0001 dpm/g) in cotton rats from noncontaminated sites in South Carolina [32]. Actinide concentrations in rat GI tracts were greater than or equal to concentrations in both grass collected 3 m away from the shoreline and in plants collected at the pond shoreline (Table II). The high actinide concentrations in rat GI tracts were strong evidence that rats were foraging in the contaminated zone around the pond shoreline.

Actinide concentrations in soil decrease rapidly with distance from the pond shoreline. Plutonium concentrations in surface soil (0-2 cm deep) taken 3 m away from the pond

shoreline were < 2% of the ^{239}Pu concentrations in shoreline sediment. Isotopic ratios for $^{241}Am/^{239}Pu$, $^{244}Cm/^{239}Pu$, and $^{233}U/^{238}U$ were not different between shoreline sediment and soil collected 3 m away from the shoreline. Therefore, even if rats forage away from the pond shoreline our comparisons of actinide uptake are not seriously compromised. Nevertheless, soil and grass in areas away from the shoreline were not considered an important source of actinides for cotton rats because of the low concentrations in grass relative to GI-tract contents. Instead, shoreline sediment is considered a likely source of actinides to cotton rats because cotton rats and other small mammals accidentally or intentionally ingest soil [33,34]. Typically, for this system, less than 10 mg of ingested shoreline sediment per dry g of GI tract contents would contribute the entire actinide concentration to the rat GI tract.

Discrimination factors for plants and rats from the pond shoreline are presented in Figure 2. Since rats may derive only a portion of their diet from the shoreline, DF values for rats were based on both shoreline sediment and soil collected 3 m from the pond shoreline. For plants not subject to submersion in pond water (grass and large macrophytes) and rat carcasses, ^{241}Am was bioaccumulated 3 to 9 times more than ^{239}Pu from shoreline sediment or soil. The pattern for the bioaccumulation of ^{244}Cm relative to ^{239}Pu was similar to that for ^{241}Am except that the enrichment of Cm was more than for Am. Uptake of ^{238}U by plants and rats was 3 to 40 times greater than the uptake of ^{239}Pu, but using either soil or sediment as a possible source of actinides there was no appreciable difference in the bioaccumulation of ^{233}U and ^{238}U (DF values were approximately one).

6. CONCLUSIONS

1. Oxidized Pu (V) is the dominant form of water soluble (0.22 μm filtrate) Pu in Pond 3513. Reduced Pu (IV) and Cm (III) appear to be associated with organics having molecular weights $\leq$ 6000. The dominant valence state of sediment associated Pu appears to be (IV).

2. The degree of contact between an organism and the pond sediment or resuspended sedimentary particulates is extremely important in determining the concentration of actinides in or associated with biota. Fish and rodent GI tracts, that probably contain sediments, contain higher concentrations than internal tissues. Limiting access to sediment significantly reduces the actinide concentrations in fish.

3. Transuranic concentrations in fish carcasses from this highly contaminated freshwater pond, containing an estimated 5 Ci of sedimentary ^{239}Pu, were less than 10% of the recommended maximum permissible concentration of the ICRP for public exposure. The favorable conditions in Pond 3513 that maximize actinide bioaccumulation indicate that, in environments receiving discharges of actinides from nuclear fuel cycle facilities, there will be little aquatic food chain transport of transuranics.

4. In and around this pond, there is no difference in the relative bioaccumulation of ^{238}Pu and ^{239}Pu nor in the uptake of ^{233}U and ^{238}U by biota from contaminated substrates (sediment or soil). This is the expected result when uniform source term exposure exists.

5. The aquatic and terrestrial (shoreline) studies at Pond 3513 consistently show that the trivalent transuranics, Am and Cm, are bioaccumulated slightly more than tetravalent Pu. Terrestrial shoreline studies show that hexavalent U is bioaccumulated more than the transuranics. This pattern of uptake by biota agrees with the relative extractability of actinides from shoreline sediment with weak acid (0.01 M HCl). The overall comparative uptake of actinides by biota at this site appears to be U > Cm $\geq$ Am > Pu.

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Figure 1. Discrimination factors, based on water (0.22 μm filtrate) or sediment, for various pairs of actinide elements in aquatic plants, invertebrates, and vertebrates (fish, frogs) from Pond 3513 (a low-level radioactive waste settling basin) in Tennessee.

Figure 2. Discrimination factors, based on soil 3 m from the pond or based on shoreline sediment, for various pairs of actinide elements in plants and cotton rats collected along the shoreline and banks of Pond 3513 (a low-level radioactive waste settling basin) in Tennessee.

Table I. Geometric mean dry weight concentration of ^{239}Pu and isotopic activity ratios in seston, filtered water, submerged sediment (0-5 cm deep), and exposed shoreline sediment (0-2 cm deep) at Pond 3513, Oak Ridge, Tennessee.

Sample	dpm ^{239}Pu/g	Activity ratios			
		^{238}Pu: ^{239}Pu	^{241}Am: ^{239}Pu	^{244}Cm: ^{239}Pu	^{238}U: ^{239}Pu
Water					
Seston[a]	0.01[e]	0.07	0.3	0.4	-
0.22-μm filtrate[b]	0.003[e]	0.1	1	20	10
Sediment					
submerged[c]	1400	0.09	0.3	0.5	0.05
shoreline[d]	530	0.05	0.2	0.1	0.04

[a]Sample size (N) = 10, [b]N = 31, [c]N = 25, [d]N = 8, [e]dpm/mL water filtered.

Table II. Geometric mean dry weight concentration of ^{239}Pu and isotopic activity ratios in representative aquatic biota from Pond 3513, Oak Ridge, Tennessee.

Sample	dpm ^{239}Pu/g	Activity ratios		
		^{238}Pu: ^{239}Pu	^{241}Am: ^{239}Pu	^{244}Cm: ^{239}Pu
Aquatic plants				
algae[a]	20	0.09	0.4	1
submerged macrophytes[b]	5	0.2	0.9	2
small emergent macrophytes[c]	0.7	0.05	0.3	0.3
large emergent macrophytes[d]	0.04	0.2	2	2
Invertebrates				
dragonfly nymphs[e]	30	0.1	0.7	2
giant water bug[f]	2	0.1	0.8	2
snail[g]	2	0.1	0.7	2
Vertebrate Carcasses				
goldfish[h]	0.1	0.1	0.4	0.8
catfish/bluegill[i]	0.05	0.2	0.5	1
tadpoles[j]	0.2	-	2	1

[a] *Hydrodictyon* (N = 8), [b] *Potamogeton* (N = 1), [c] *Eleocharis* (N = 18), [d] *Typha* and *Juncus* (N = 7), [e] *Plathemis* (N = 15), [f] *Belostoma* (N = 7), [g] *Physa* (N = 30), [h] *Carassius* (N = 7) - 28 week exposure time, [i] *Ictalurus* and *Lepomis* (N = 13) - 28 week exposure time, [j] *Rana* (N = 5) - 2 years old.

Table III. Geometric mean dry weight concentration of ^{239}Pu, ^{238}Pu, ^{241}Am and ^{244}Cm in goldfish after 28 weeks in enclosures or free roaming in Pond 3513, Oak Ridge, Tennessee.

Sample	dpm/g			
	^{239}Pu	^{238}Pu	^{241}Am	^{244}Cm
From Pond (free)				
goldfish GI tract[a]	500	40	200	300
goldfish carcass[b]	0.1	0.01	0.04	0.08
From enclosure (nets)				
goldfish GI tract[a]	300	20	100	200
goldfish carcass[a]	0.05	0.007	0.02	0.04
From enclosure ("Living Streams")				
goldfish GI tract[c]	50	4	20	40
goldfish carcass[a]	0.02	0.002	0.01	0.02

[a]Sample size (N) = 6, [b]N = 7, [c]N = 5.

Table IV. Geometric mean dry weight concentration of ^{239}Pu and isotopic activity ratios in soil, grass (Festuca sp.), and rats (Sigmodon hispidus) collected 3 m away from the Pond 3513 shoreline

Sample	dpm ^{239}Pu/g	Activity ratios			
		^{241}Am: ^{239}Pu	^{244}Cm: ^{239}Pu	^{238}U: ^{239}Pu	^{233}U: ^{238}U
Soil[a]	7	0.2	0.2	0.3	3
Grass[a]	0.009	0.5	1	3	2
Cotton Rats					
GI tract[b]	4	0.3	0.6	0.04	5
Carcass[b]	0.02	0.9	2	2	2

[a]Sample size (N) = 8, [b]N = 15.

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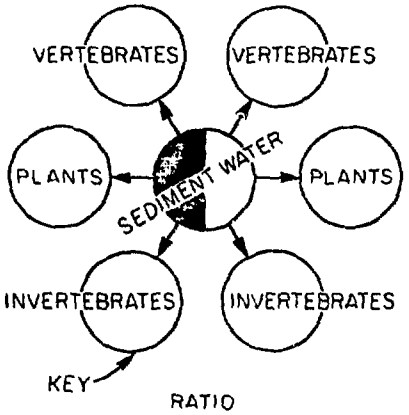
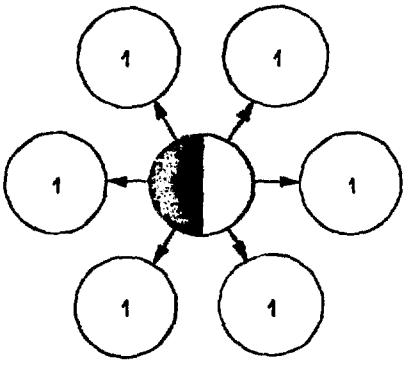
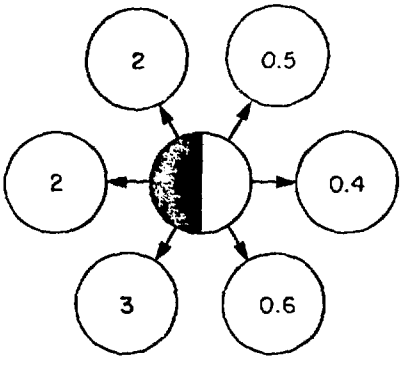
$$\text{DISCRIMINATION FACTOR} = \frac{\text{ISOTOPIC RATIO IN BIOTA}}{\text{ISOTOPIC RATIO IN SEDIMENT OR WATER}}$$

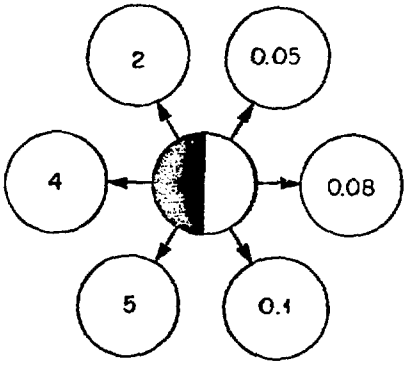




$^{238}Pu : {}^{239}Pu$



$^{241}Am : {}^{239}Pu$



$^{244}Cm : {}^{239}Pu$

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DISCRIMINATION FACTOR = $\frac{\text{ISOTOPIC RATIO IN BIOTA}}{\text{ISOTOPIC RATIO IN SEDIMENT OR SOIL}}$