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METHOD FOR CALCULATION OF UPPER LIMIT INTERNAL ALPHA DOSE RATES TO AQUATIC ORGANISMS WITH APPLICATION TO PLUTONIUM-239 IN PLANKTON

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ABSTRACT. A method for calculation of upper limit internal alpha dose rates to aquatic organisms is presented. The mean alpha energies per disintegration of radionuclides of interest are listed to be used in standard methodologies to calculate dose to aquatic biota. As an application, the upper limits for the alpha dose rates from ^{239}Pu to the total body of plankton are estimated based on data available in open literature.

RESUMO. Um método é apresentado para o cálculo dos limites superiores das taxas de doses internas de partículas alfa para organismos aquáticos. As energias médias das partículas alfa por desintegração são arroladas para radionuclídeos de interesse para serem usadas para cálculo de dose para a biota aquática em metodologias convencionais. Como uma aplicação, estimou-se os limites superiores para as taxas de dose para o corpo inteiro de plancton, devido a desintegração alfa de ^{239}Pu, baseando-se em dados disponíveis na literatura.

INTRODUCTION

Recent report on the effects of ionizing radiation on aquatic organisms and ecosystems (IA76) recognizes that the dosimetric models to aquatic biota need improvement. However, the existing models allow to estimate non refined dose rates to organisms of distinct species for purposes of comparison. An estimation is presented here on the upper limits for the alpha dose rates from ^{239}Pu to the total body of plankton based on data available in the open literature. The mean alpha energies per disintegration of radionuclides of interest are listed here to be used in standard methodologies to evaluate dose to aquatic biota.

STANDARD METHODOLOGIES

Most aquatic organisms are in contact with water and sediments as well, and can concentrate radionuclides directly from water or through their food chains. It is well recognized that the radiation dose to aquatic biota should be evaluated for external exposure to potentially contaminated water and sediments and also for internal contaminants.

The methodology to evaluate external and internal dose from beta and gamma radiation is well established (Wo73, So73, Le75). However, estimations of dose to aquatic biota have been also made by other investigators at various levels of detail (Fo57, Ch64, Sh72, Fo73, Wr73, Pa76, He76). An examination of the various approaches used thus far in dosimetric estimations to aquatic biota has shown that, despite some points in common, there is not yet an universally adopted way to estimate internal dose to aquatic organisms. Besides, the alpha contribution to the internal dose may be misestimated if the suggested standard methodologies (So73, Le75) are used, because faulty computational parameters are listed by So73 (file CRITEN to be used in the CRITS Program) for the mean alpha energies available for deposition per disintegration. Those computational parameters would be approximately correct if used to estimate dose equivalent, in rem, to humans, since the values for effective alpha energy per disintegration listed by So73 are about ten times higher than the alpha energy values accepted currently and compiled in Nuclear Data Sheets and He69. However, when estimating absorbed dose, in rad, the use of such parameters will lead to misestimations in the alpha doses. Since the variations in relative biological effectiviness (RBE) between alpha-radiation and beta- and gamma-radiation are not well established yet for most aquatic organisms, the dose estimations for those organisms are usually expressed in rad rather than in rem.

The contribution from manmade alpha emitters, like $^{239,240}Pu$ for example, is not expected to be generally dominant to the overall radiation dose to aquatic biota. However, the internal alpha dose should be considered in standard computational methodologies used to estimate radiation dose to aquatic biota when alpha emitters are involved in routine or accidental releases of radionuclides to the aquatic environment. Independent estimations made by several investigators (Wo73, Ch64, Sh72, Fo73, Pa76) have shown that the natural alpha internal dose to some species of aquatic biota might not be necessarily negligible in dosimetric estimations. Wo73 suggests also that the alpha dose from ^{239}Pu to plankton may become more significant in the long term.

MEAN ALPHA ENERGIES FOR DOSIMETRIC CALCULATIONS

The mean alpha energies, E_α, listed in Table 1 provide the basis for calculating the upper limit internal alpha dose rates from radionuclides which may be present, in minute amounts though, in aquatic organisms. Those radionuclides may be incorporated into aquatic organisms either due to bioaccumulation of natural occuring radionuclides or by internal contamination with manmade radionuclides.

The mean alpha energy, $\bar{E}_{\alpha i}$, available for deposition per desintegration of a radionuclide i is calculated by the following expression:

$$\bar{E}_{\alpha i} = \sum_j f_{ji} E_{\alpha ji}$$

where f_{ji} = the frequency per disintegration of the emission of the j^{th} alpha particle; and

$E_{\alpha ji}$ = the energy of the j^{th} alpha particle (in MeV) emitted by the radionuclide i.

The upper limit to the average internal alpha dose rate to total body of an organisms can be easily calculated under the following assumptions:

1. Any alpha emitting radionuclide i is uniformly distributed throughout the organism body; and
2. The mean alpha energy, $\bar{E}_{\alpha i}$, available for deposition per disintegration is totally absorbed within the organism body. The values of $\bar{E}_{\alpha i}$ are presented in Table 1 based on data taken from reference He69 and Nuclear Data Sheets.

The average internal alpha dose rate, D (in μrad/hr), is usually calculated (Wo73, So73, Le75, Pa76, He76) by the following expression:

$$D = 2.13 \sum_i \bar{E}_{\alpha i} C_i$$

where: $2.13 \frac{\text{dis}}{\text{pCi/gm}} \cdot \frac{\mu\text{rad/hr}}{\text{MeV}} = 3.7\times10^{-2} \frac{\text{dis}}{\text{sec}\cdot\text{pCi}} \; 1.6\times10^{-6} \frac{\text{ergs}}{\text{MeV}}$

$$\times\; 10^{4} \frac{\mu\text{rad}}{\text{ergs/gm}} \; 3.6\times10^{3} \frac{\text{sec}}{\text{hr}} \;;$$

$\bar{E}_{\alpha i}$ = mean alpha energy per disintegration (see Table 1) (in MeV);

C_i = concentration of radionuclide i in the organism (in pCi/gm).

UPPER LIMITS TO ALPHA DOSE FROM Pu-239 TO TOTAL BODY OF PLANKTON

Plutonium-239, an alpha emitter with 2.4×10^{4} years half-life, has already found its way to the aquatic environment in measurable amounts through fallout from nuclear weapons, accidents with military airplanes carrying nuclear weapons, and releases from the nuclear industry. Data on the plankton content of ^{239}Pu are available from the literature. However, to the best of our knowledge, the only reported attempt to estimate the internal alpha radiation dose rate to plankton from ^{239}Pu thus far was made by Wo73 (reproduced in IA76), based upon data from Pi64 and No71.

Table 2 presents estimations for the upper limits of the internal alpha radiation dose rate received by plankton due to the bio-accumulation of ^{239}Pu. The assumptions made to calculate the dose rates presented in Table 2 do not take into account the internal distribution of the alpha emitters $^{239,240}Pu$. However, keeping in the proper perspective the variations due to internal distributions of ^{239}Pu in distinct species, one can still make the observation that some phytoplankton from North Atlantic collected in 1966 (No71) and zooplankton from Thule (Åa71) might have received higher total internal alpha dose than the plankton collected in the Bikini Atoll in 1964 (We69). Assuming plankton life spans higher than one year and concentration of ^{239}Pu constant in time, the upper limits for annual internal alpha dose to total body of zooplankton from ^{239}Pu would remain under 1 rad with the sole exception of the dose to zooplankton from a specific site in Greenland where an accident with a military airplane in the winter before the collection time has been reported (Åa71). However, more recently Åakrog has observed that the Plutonium concentrations in aquatic organisms from Thule, Greenland have been decreasing with time (Aa77). The internal annual alpha dose to phytoplankton from the North Atlantic (No71) might also be over 1 rad.

DISCUSSION AND CONCLUSIONS

The mean alpha energies available for deposition per disintegration listed in Table 1 should be used in standard methodologies to calculate dose to aquatic biota, whenever the concentrations of those radionuclides in the organisms are known.

The upper limits for internal alpha dose rates presented in Table 2 and also those estimations made by Wo73 should be considered only as preliminary efforts towards valuable dose comparisons. The upper limit for the annual internal alpha dose for plankton from ^{239}Pu has been estimated, under bounding assumptions, to be about 1 rad based on data taken from the literature. Such a dose is usually regarded as insignificant as far as radiobiological effects are concerned. However, there are discrepant, view-points on the radioresistance of marine organisms (Po66, Bo71, IA76).

The total alpha doses from ^{239}Pu incorporated into the organisms to the total body of zooplankton of several origins and distinct species are in general in the same order of magnitude of the natural alpha dose, 27 mrad/yr, to the total body of the zooplankton Gammarus from the Hudson River (Pa76). However, in cases of accidental releases the alpha dose from ^{239}Pu to total body of plankton may be considerably higher.

The internal distribution of alpha emitters inside zooplankton, for example, may result in dose rates to a particul^r organ greater than tenfold the dose rate estimated to the total body of the organism, as Pa75 has shown to occur, under laboratory conditions, with the internal alpha dose from ^{226}Ra to Gammarus. Besides that, the outside-to-inside ratios for ^{210}Po and ^{239}Pu have been reported to be 1000 and 200 respectively for the giant brown algae, Pelagophycus porra (Wo72). Recently, Hi77 reported that the concentration of $^{239,240}Pu$ in euphausiid, Meganyctiphanus norvegica faecal pellets is two orders of magnitude higher than the concentrations in the exoskeleton and wholebody.

Seabed has been receiving serious consideration as a valid option for the ultimate disposal of high level nuclear wastes (Ho77). The U.S. Environmental Protection Agency has shown recently (EP77) that the alpha emitting wastes from phosphate and uranium mining present also a disposal problem. The lack of knowledge to make a decision at present time whether seabed can be the site for ultimate disposal of high radioactive waste has been prudently recognized (He77). However, in the long range alpha emitters can find their way to the aquatic environment by leaking, leaching and running off irrespective seabed will or will not be chosen as the site for ultimate disposal of radioactive wastes. Therefore, an effort should be made to improve the experimental and calculational methodologies to estimate the dose per generation of aquatic organisms of lower trophic levels taking into account the internal distribution of alpha emitters.

REFERENCES

Åa71 Åakrog A., 1971, "Radioecological Investigations of Plutonium in an Artic Marine Environment", Health Phys. 20, 31.

Åa77 Åakrog A., 1977, "Environmental Behaviour of Plutonium Accidentally Released at Thule, Greenland", Health Phys. 32, 271.

Bo71 Bowen V. T., Olsen J. S., Osterberg C. L. and Rivera J., 1971, "Ecological Interaction of Marine Radioactivity", In Radioactivity in the Marine Environment. National Academy of Sciences. National Research Council 200.

Ch64 Cherry R.D., 1964, "Alpha Radioactivity of Plankton", Nature 203, 139.

EP77 Environmental Protection Agency (E.P.A.), 1977, "Issues and Objectives Statement", In A Workshop of Issues Pertinent to the Development of Environmental Protection Criteria for Radioactive Wastes, Reston, Virginia, Feb. 3-5.

Fo57 Folsom T. and Harley J., 1957, "Comparison of Some Natural Radiation Received by Selected Organisms", In The Effects of Atomic Radiation on Oceanography and Fisheries, Publication 551, National Academy of Sciences. National Research Council, 28.

Fo73 Folsom T. R. and Beasley T. M., 1973, "Contributions from Alpha Emitter, Polonium-210, to the Natural Radiation Environment of the Marine Organisms", In Radioactive Contamination of the Marine Environment, International Atomic Energy Agency (I.A.E.A.), Vienna, 625.

He77 Heath G. R., 1977, "Barriers to Radioactive Waste Migration", Oceanus 20, 26.

He69 Heath R. L., 1969, "Table of Isotopes", In Handbook of Chemistry and Physics (Robert C. Weast, editor). The Chemical Rubber Co., Cleveland, B-265.

He76 Hetherington J. A., Jefferies D. F., Mitchell N. T., Pentreath R.J. and Woodhead D. S., 1976, "Environmental and Public Health Consequences of the Controlled Disposal of Transuranic Elements to the Marine Environment", In Transuranium Nuclides in the Environment, Proceedings of a Symposium, San Francisco, Nov 17-21, 1975, I.A.E.A., Vienna, 139.

Hi77 Higgo J. J. W., Cherry R. D., Heyraud M., Fowler S.W., 1977, "Rapid Removal of Plutonium from the Oceanic Surface Layer by Zooplankton Faecal Pellets", Nature 266, 623.

Ho77 Hollister C.D., 1977, "The Seabed Option", Oceanus 20, 18.

IA76 International Atomic Energy Agency (I.A.E.A.), 1976, "Effects of Ionizing Radiation on Aquatic Organisms and Ecosystems", Vienna, Technical Reports Series No. 172.

Le75 LeClare P., Hairr L., Schmirl L., Philbin T., Stoecker R., and Eisenbud M., 1975, "Standard Methodology for Calculating Radiation Dose to Lower Form Biota", Atomic Industrial Forum, Inc. AIF/NESP-006.

Mi75 Miettinen J. K., Jaakkola T. V. A., and Järvinen M.,1975, "Plutonium Isotopes in Aquatic Foodchains in the Baltic Sea", In Impacts of Nuclear Releases into the Aquatic Environment, Proceedings of a Symposium, Otaniemi, 30 June-4 July,I.A.E.A., Vienna, 147.

ND70 Nuclear Data Sheets, 1970, B4, 543.

ND71a Nuclear Data Sheets, 1971, B5, 561.

ND71b Nuclear Data Sheets, 1971, B6, 209.

ND71c Nuclear Data Sheets, 1971, B6, 539.

ND73 Nuclear Data Sheets, 1973, 10, 555.

ND76a Nuclear Data Sheets, 1976, 17, 329.

ND76b Nuclear Data Sheets, 1976, 19, 1.

No72 Noshkin V. E., 1972, "Ecological Aspects of Plutonium Dissemination in Aquatic Environments", Health Phys. 22, 537.

No71 Noshkin V. E., Bowen V. T., Wong K. M., and Burke J. C., 1971, "Plutonium in North Atlantic Ocean Organisms: Ecological Relationships", In Proceedings of the Third National Symposium on Radioecology (Nelson D. J., editor), Oak Ridge, Tennessee, May 10-12, 681.

Pa75 Paschoa A. S., 1975, Ph.D. thesis, New York University.

Pa76 Paschoa A. S., Wrenn M. E., and Eisenbud M., 1976, "Natural Radiation Dose to Gammarus from Hudson River", In Proceedings of the Fourth Symposium on Hudson River Ecology, Bear Mountain, New York, March 28-30, 10.

Pi64 Pillai K. C., Smith R. C., and Folsom T. R., 1964, "Plutonium in the Marine Environment", Nature, 203, 568.

Po66 Polikarpov G. G., 1966, "Radioecology of Aquatic Organisms", Reinhold, New York.

Sh72 Shannon L. V., 1972, "Marine Alpha-Radioactivity off Southern Africa 1 - Gross Alpha Activity, Radiation Dose, Alpha-Spectrum and Variation in Alpha Activity of Marine Life", Division of Sea Fisheries, Investigational Report no. 98, South Africa.

So73 Soldat J. K., Shipler D. B., Baker D. A., Denham D. H., and Robinson N. M., 1973, "Computational Model for Calculating Doses from Radionuclides in the Environment", In AEC Final Environmental Statement - ALAP-LWR Effluents, Volume 2, NTIS Report WASH-1258.

Wa75 Wahlgren M. A. and Marshall J. S., 1975, "The Behaviour of Plutonium and Other Long-Lived Radionuclides in Lake Michigan. I Biological Transport, seasonal cycling and residence times in the water column", In Impacts of Nuclear Releases Into the Aquatic Environment, Proceedings of a Symposium, Otaniemi, 30 June-4 July, I.A.E.A., Vienna, 227.

Wa76 Wahlgren M. A., Alberts J. J., Nelson D. M., and Orlandi, K.A., 1976, "Study of the Behaviour of Transuranics and Possible Chemical Homologues in Lake Michigan Water and Biota", In Transuranium Nuclides in the Environment, Proceedings of a Symposium, San Francisco, Nov. 17-21, 1975, I.A.E.A., Vienna, 9.

We69 Welander A. D., 1969, "Distribution of Radionuclides in the Eniwetok and Bikini Atolls: August 1964", In Proceedings of the Second National Symposium on Radioecology, Ann Arbor, Michigan (Nelson D. J. and Evans F. C., editors) May 15-17, 346.

Wo71 Wong K. M., 1971, "Radiochemical Determination of Plutonium in Sea Water, Sediments and Marine Organisms", Anal. Chim. Acta, 56, 355.

Wo70 Wong K. M., Burke J. C., and Bowen V. T., 1970, "Plutonium Concentration in Organisms of the Atlantic Ocean", In Proceedings of the Fifth Annual Health Physics Aspects of Nuclear Facility Siting, Idaho Falls, Idaho, Nov. 3-6, 529.

Won71 Wong K. M., Hodge V. F., and Folsom T. R., 1971, "Concentrations of Plutonium, Cobalt and Silver Radionuclides in Selected Pacific Seaweeds", In Proceedings of Environmental Plutonium Symposium, Los Alamos Scientific Laboratory (LASL) LA-4756, Aug. 4-5, 93.

Wo72 Wong K. M., Hodge V. F., and Folsom T. R., 1972, "Plutonium and Polonium inside Giant Brown Algae", Nature 237, 460.

Wo73 Woodhead D. S., 1973, "Levels of Radioactivity in the Marine Environment and the Dose Commitment to Marine Organisms", In Radioactive Contamination of the Marine Environment, I.A.E.A., Vienna, 499.

Wr73 Wrenn M. E., Jinks S. M., Cohen N., and Hairr L. M., 1973, "Cs-137 and Cs-134 Distribution in Sediment, Water, and Biota of the Lower Hudson River and their Dosimetric Implications for Man", In Proceedings of the Third International Congress of the International Radiation Protection Association, Sep. 9-14, Washington D.C., 279.

Table 1 - Alpha emitting radionuclide mean alpha energies $\bar{E}_{\alpha}$ per disintegration for deposition, and radiological decay constants.

Nuclide	E_{α_i} (MeV)*	λ_r (day)$^{-1}$	Reference
Bi-212	2.2 (= 6.0×0.36)**	1.6E+01†	He69
Po-210	5.3	5.0E-03	ND71a
Po-212	5.6 (= 8.8×0.64)**	2.0E+11	He69
Po-214	7.7	3.7E+08	He69
Po-218	6.0	3.3E+02	He69
Rn-220	6.3	1.1E+03	ND76a
Rn-222	5.5	1.8E-01	He69
Ra-223	5.8	6.1E-02	He69
Ra-224	5.7	1.9E-01	ND76a
Ra-226	4.8	1.2E-06	He69
Ac-225	5.7	6.9E-02	ND73
Ac-227	~0.1	8.8E-05	He69
Th-227	5.9	3.7E-02	He69
Th-228	5.4	9.9E-04	ND76a
Th-229	4.9	2.6E-07	ND71b
Th-230	4.7	2.5E-08	ND70
Th-232	4.0	1.4E-13	ND70
Pa-231	4.9	5.8E-08	ND71b
U-232	5.3	2.6E-05	ND70
U-233	4.8	1.2E-08	ND71b
U-234	4.8	7.7E-09	He69
U-235	4.2	2.7E-12	ND71b
U-236	4.5	7.9E-11	ND70
U-238	4.2	4.2E-13	ND70
Np-237	4.8	8.9E-10	ND71c
Pu-238	5.5	2.2E-05	ND70
Pu-239	5.1	7.8E-08	ND71c
Pu-240	5.2	2.9E-07	ND70
Pu-242	4.9	4.9E-09	ND70
Pu-244	4.6	2.3E-11	ND76a
Am-241	5.5	4.4E-06	ND71c
Am-243	5.3	2.6E-07	ND76b
Cm-242	6.1	4.3E-03	ND70
Cm-243	5.8	6.7E-05	ND76b
Cm-244	5.8	1.0E-04	ND76a
Cm-245	5.4	2.2E-07	ND76b
Cm-246	5.4	4.0E-07	ND76a
Cm-248	4.7	5.6E-09	ND76a
Bk-247	5.6	1.4E-06	ND76b
Cf-252	5.9	7.2E-04	ND76a

* Mean alpha energy available for deposition per disintegration of radionuclide i.

** Corresponding to (Po-212, Tl-208) branching ratio of Bi-212.

† 1.6E+01 = 1.6×10^{1}

Table 2 - Plutonium-239 content and upper limits for alpha dose rates* to total body of plankton.

Year of Collection	Origin	Pu-239 Content Phyto- (pCi/gm wet)	Pu-239 Content Zoo- (pCi/gm wet)	Alpha Dose Rates Phyto- (μrad/hr)	Alpha Dose Rates Zoo- (μrad/hr)	Reference
1961	Atlantic	-	[a]2.0E+00**	-	2.2E+01	No70,No72
1964	California Coast	4.3E-04(4)[b]	1.1E-03	4.7E-03	1.2E-02	Pi64
1964	Bikini Atoll[c]	2.5E-03	[d]1.4E-01**	2.7E-02	1.5E+00	We69
1964	Bikini Atoll[e]	-	[d]2.2E-00**	-	2.4E+01	We69
1965	Arabian Sea[f]	[g]2.3E-04(3)	-	2.5E-03	-	Pi76
1966	Atlantic	8.4E+01	-	9.1E+02	-	No71
1968	Thule, Greenland	-	9.0E-01 to 3.5E+01	-	9.8+00 to 3.8E+02	Åa71
1970	Thule, Greenland	-	1.1E-03[h]	-	1.2E-02	Åa77
1970	Atlantic	-	1.0E+00	-	1.1E+01	Wo71
1970-1971	Atlantic	[g]6.0E-01(16)	-	6.5E+01	-	Won71
1971	Arabian Sea[f]	[g]1.9E-03(3)	-	2.1E-02	-	Pi76
1971	Atlantic	9.0E-02	-	9.8E-01	-	No71
1972	Lake Michigan	6.0E-03[h]	-	6.6E-02	-	Wa76
1973	Lake Michigan	6.5E-04[h]	2.0E-04[h]	7.2E-03	2.2E-03	Wa76
1973	Arabian Sea[f]	[g]3.8E-04(2)	-	4.1E-03	-	Pi76
1973	Arabian Sea[i]	[g]4.3E-04(2)	-	4.6E-03	-	Pi76
1974	Lake Michigan	4.3E-03(22)[h]	2.6E-04[h]	4.8E-02	2.9E-03	Wa75
1974	Baltic Sea	-	[j]7.0E-04[h]	-	7.8E-03	Hi75
1974	Thule, Greenland	-	4.0E-04[h]	-	4.5E-03	Åa77
1975	Lake Michigan	6.0E-03(2)[h]	1.1E-04(2)[h]	6.6E-02	1.2E-03	Wa76
1976	not reported	-	8.5E-05	-	9.4E-04	Hi77

*Upper limits - assuming uniform distribution of alpha emitter throughout the body, constant concentration, and complete absorption of the mean alpha energy

**not specified whether phyto- or zoo- plankton

a 2.0E+00 read 2.0×10^{0}

b Parentheses indicate number of samples to calculate the average

c Off Enyu Island

d Assuming 0.20 gm dry/gm wet

e Off Romurikku Island

f Tarapur

g Seaweed

h Pu-239,240: $\bar{E}_{\alpha}$ = 5.2 Mev/dis

i Mangalore

j Benthic Isopoda (Mesidotea entomon)