DETERMINANTS OF RADIOLOGICAL DOSE FROM SR AND CS ACCUMULATED BY TROPICAL FRESHWATER FISH

J. R. Twining, S. J. Markich and J. M. Ferris
ANSTO, Australia

ABSTRACT

Recent studies of the bioaccumulation of strontium and caesium by tropical freshwater fish were in some ways consistent with temperate data. For example, biodistribution of radionuclides and the influence of chemical analogues Ca and K were in general agreement with earlier data.

However, in other matters there were systematic inconsistencies when compared with the standard IAEA models. These differences gave rise to transfer (concentration) factors for both Cs and Sr that were approximately an order of magnitude below the expected values based on temperate data (e.g. 13 and 0.7 compared with 440 and 4.7 respectively). Compatible results were found for a range of tropical species studied by different investigators.

In this presentation, some of the factors believed to influence bioaccumulation, and hence radiological dose, in tropical environments are discussed in relation to the above findings. These factors include: water physico-chemistry, temperature, fish size and physiology, whether or not the data were acquired from field or laboratory studies, and how the studies were carried out.

Critical group exposure pathways in tropical environments and the benefits and shortcomings of applying default temperate models of Cs and Sr accumulation by freshwater fish are also discussed.

INTRODUCTION

Nuclear power will have a role in the rapid industrial development occurring within the tropics. Hence, an understanding of the kinetics and degree of radionuclide bioaccumulation in these regions is required for realistic dose assessment. Models under development to determine the consequences of a nuclear release in south east Asia have perceived that specific transfer factors (TFs) for these environments are sparse or non-existent (Harris *et al.*, 1997).

In recognition of this need, the International Atomic Energy Agency (IAEA) and the United Nations Food and Agriculture Organisation (FAO) established a Co-operative Research Program (CRP) to assess the 'Transfer of radionuclides from air, soil and fresh water to the food chain of man in tropical and sub-tropical environments'. The CRP developed standardised experimental protocols that specified factors such as water quality, exposure conditions, etc., for all participants to follow (IAEA, in prep). The resultant data were tabulated and will be included in updated manuals of radionuclide TF data (e.g. IAEA, 1994).

The primary aim of this paper is to compare the bioaccumulation of Cs and Sr by tropical fish with the previously acquired temperate data and to assess factors that may contribute to any apparent differences. In addition, the implications of the recent findings to critical group dose assessment in tropical areas will be addressed.

DATA ACQUISITION AND PRELIMINARY COMPARISON

The tropical fresh weight TF and biodistribution data used in this paper were derived primarily from studies undertaken within the IAEA/FAO CRP (IAEA, in prep; Mollah *et al.*, 1994, '95, '97; Ngo & Binh, 1997; Sinakhom *et al.*, 1997; Twining *et al.*, 1997). Some additional information was acquired from Srivastava *et al.* (1990, 94). The TFs and biological half-lives ($T_{½}$s) for Cs and Sr in tropical fish, derived within the CRP, were determined by fitting a single compartment exponential uptake and loss model to experimental uptake and elimination phase kinetic data (IAEA in prep). Srivastava *et al.* (1990, 94) derived their values graphically. A synopsis of these results and other relevant parameters is given in Table 1.

Measured TFs for Cs in the flesh of tropical species were analysed for nine different species and a variety of different water qualities. They consisted of values some two orders of magnitude less than the conservative factor of 2000 expected by the IAEA (1994) for Cs accumulation by freshwater fish from temperate waters. Indeed, for all tropical studies except one, the measured TFs for Cs were below the lower end of the relatively broad range (30 - 2000) reported by the IAEA (1994).

The measured TFs for Sr ranged from 0.1 to 19 across five species of fish studied by three separate investigators. These values were consistently less than the TF of 60 expected by the IAEA (1994) based on data for temperate freshwater fish. The lowest values were beyond the lower end of the reported range (1 - 1000).

DISCUSSION

Factors affecting bioaccumulation of Cs and Sr by freshwater fish

Water chemistry can systematically modify the TFs of Cs and Sr in freshwater fish. The concentrations of K, Ca and suspended solids (SS) have been shown to correlate with the TFs of both radionuclides (IAEA, 1994; Rowan & Rasmussen, 1994). The IAEA (1994) has published empirical models which incorporate these variables, thus enabling calculation of more specific, 'modified' TFs for Cs and Sr. These models are based largely on field data for temperate freshwater fish (Rowan & Rasmussen, 1994). The model predicting the modified TF for Cs includes the concentrations of both K and SS in water (increased SS and K^+ reduces TF), with a categorical variable for the trophic preference of the fish (*i.e.* piscivorous (1) or non-piscivorous (0); declared as zero because fish were not fed radioactively labelled food in the tropical studies). The model predicting the modified TF for Sr incorporates the Ca concentration in the water (increased Ca reduces TF) (IAEA 1994). When the mean values of these variables for the tropical studies were used in the temperate models, the modified TFs predicted for Cs and Sr (Table 1) were 2 - 60 times less than the recommended values.

Despite an approximate order of magnitude reduction from the IAEA recommendation, the TFs measured for Cs in tropical fish were still 15 - 65 times less than these modified values (Table 1). Similarly, apart from one value derived using exceptionally high Ca water concentrations, the TFs measured for Sr were 2 - 20 times less than the modified values.

The modification of TFs by water chemistry also contributes to explaining the differences in measured TFs between the tropical studies. Allowance for the chemical differences resulted in the adjusted tropical TFs for Cs being generally brought closer together with the exception of a slightly increased divergence by the value of Srivastava *et al.* (1990). Similarly for Sr, except for the outlier identified earlier, the adjusted tropical TF values differed only by a factor of ~8. The results from the tropical laboratory studies were thus closely corroborative and they consistently estimated lower TFs for Cs and Sr than were predicted on the basis of existing empirical models.

Temperature is perhaps the most obvious and biologically important physical factor differentiating temperate and tropical freshwater environments. Rowan & Rasmussen (1994) comprehensively reviewed field-collected bioaccumulation data for radionuclides of Cs in fish from temperate, freshwater and marine systems of latitudes ranging from approximately 32 - 69°N. These authors showed that TFs for Cs were positively related to mean annual air temperature and to the thermal zone inhabited by the fish (*i.e.* epilimnetic > hypolimnetic), and hypothesised that TFs for fish would increase in response to increasing habitat temperature. The findings of the tropical studies did not generically support this hypothesis. In addition, Srivastava *et al.* (1994) tested one species of fish for the effect of increasing temperature and observed a directly opposite response.

A single compartment exponential uptake and elimination model is the simplest approach to describe the kinetics of accumulation. Using this approach, TF values are determined at steady state and represent the ratio between the uptake and loss rate coefficients (Whicker & Shultz, 1982). Hypotheses implying that higher temperatures lead to higher uptake rates, and hence higher TF values, ignore the importance of this ratio. Should the animals be endemic to tropical climates, and hence, have evolved metabolisms adapted to those environments, then there is no reason to suppose that the ratio should be biased towards greater uptake. Ugedal *et al.* (1992) and Cocchio *et al.* (1995) reported that the $T_{½}$ for Cs administered orally to trout decreased with increasing temperature to an extent similar to that predicted by the fishes' increasing metabolic rate. This finding is consistent with the reasoning that the relatively smaller TFs observed in the tropical studies result from an evolved metabolic shift towards increased elimination relative to uptake rate. However, most of the tropical studies reported $T_{½}$s for Cs that are comparable with those found for temperate systems (Table 1). This suggests, *prima facie*, that the rate of uptake should decrease in tropical conditions to yield lower TFs whilst maintaining similar $T_{½}$s.

A possible explanation for these findings is that a one-compartment model may not adequately describe the loss kinetics of metals from fish. Indeed, several studies (Adam *et al.*, 1997; Willis & Jones, 1977; Carbonell & Tarazona 1994; and references therein) have shown that the loss of metals from a range of fish follows at least a two compartment model (*i.e.* shorter and longer-lived sub-compartments).

There are, however, several other aspects of these tropical laboratory studies that may have further reduced the measured TFs, particularly for Cs, relative to those determined from field data. Patel (1975) compared laboratory- and field-derived data for marine fish and found that the TFs for Cs were generally 2 - 4 times less in laboratory studies than in field studies. Similarly, Rowan & Rasmussen (1994) reported that laboratory studies overestimate elimination by a factor of 1.4. Although the CRP protocols required simulation of natural environments as closely as possible, it is inevitable that the experimental conditions did not achieve this ideal.

In the case of Cs, the reduced TFs may be due, in part, to the administration of activity via the water column and not via the diet. The dietary uptake pathway has been shown to dominate Cs accumulation in a number of studies. The animals in the tropical laboratory studies were fed unlabelled food. In contrast to Cs, Patel (1975) reported that the TFs for Sr were potentially greater, by up to an order of magnitude, in laboratory studies than in field studies. This finding would suggest that the tropical TFs derived in laboratories should be greater than the field-derived, temperate values, not less as observed.

Twining *et al.* (1997) found that stressed fish accumulated less Sr. Also, Srivastava *et al.* (1990) observed an increased loss of ^{137}Cs from a fish just prior to its death. Hence, there is some indication that experimental stress may have contributed to lowering the TFs. This possibility is supported by Pottinger and Calder (1995) who measured increased stress enzyme levels in fish with higher experimental disturbance and a concurrent effect on toxicological endpoints.

The results of Ngo & Binh (1997) and Twining *et al.* (1997) both showed a negative correlation between fish size and TF. These findings accord with some, though not all (see Rowan & Rasmussen, 1994), previous reports that size is inversely related to the TFs for Cs (Koulikov & Ryabov, 1992) and Sr (Pally & Foulqier, 1979). The selection of larger, edible fish in the CRP studies may therefore have favoured lower TFs assuming that the field sampling was not similarly biased towards larger fish.

In some tropical studies whole fish were used as the sample rather than edible flesh which was used to determine the IAEA (1994) values. For Cs this may have some consequence as Cs is concentrated in flesh compared to other tissues. However, the observed results within the CRP were consistently low irrespective of sample type (Table 1). For Sr, most of the activity was found in the bones and the scales (Twining *et al.*, 1997). Hence, whole animal values would be expected to indicate higher TFs, not lower as noted. These effects can be observed to some degree in the findings reported by Sinakhom *et al.* (1997) and Mollah *et al.* (1995, 1997) (Table 1).

One other factor that may have contributed to the lower observed TF values is the possibility of reduced uptake due to saturation of the uptake mechanism by the higher radioactivity levels during the experimental exposures. That is, the TF may be non-linear with respect to water activity level. This hypothesis is not supported if it assumed that the Cs and Sr are accumulated as analogues of the metals K and Ca. In this case the variation in activity is trivial in terms of the overall elemental concentrations that are not approaching saturation. There is strong support for the metal analogue hypothesis both within the tropical studies and in the previous temperate literature to refute the possibility of non-linear TFs.

Radiological dose assessment

Based on the recent studies, Cs is concentrated above environmental levels by about an order of magnitude in the flesh of tropical freshwater fish species. Sr was accumulated to slightly lower levels (Table 1). As mentioned above, the TFs for both radionuclides are lower than those expected for temperate freshwater fish (IAEA 1994) and, as such, imply that use of their default temperate TFs for radiological dose assessment may result in an overestimation of dose when applied to tropical freshwater fish. The average $T_{½}$ derived from the tropical studies implies equilibrium in the flesh of adult fish under tropical conditions in just over a year for Cs and within approximately 7 months for Sr (based on 5 $T_{½}$s or 97% equilibrium).

The CRP studies observed biodistribution of radioactivity between tissues with a general ranking of flesh > skin/scales > bone for Cs, and bone ≥ skin/scales >> flesh for Sr. These results are consistent with several other published findings (Stanek *et al.*, 1990; Hewett & Jeffries, 1978; Guimarães, 1992). For the skin/scales tissue sections, the greatest fraction of Sr was shown to be in the scales, where specific activities were ~15 times those in the skin itself (Twining *et al.* 1997). The data of Sinakhom *et al.* (1997) show very high values for Sr accumulation in the head of catfish, comprising up to 80% of the total accumulated body burden.

The bone and, therefore, probably the scales, represent long-lived compartments with slow exchange. Strontium in fish scales could contribute to radiological dose if the scales were consumed directly, as in very small fish (*e.g.* whitebait and anchovy), in stews, soups or other preparations using whole fish. The IAEA (1994) report negligible losses of Sr from the boiling of mammalian bones. Given the chemical similarities between the bones of mammals and the bones and scales of fish, it is reasonable to assume that fish bones and scales would be similarly retentive of Sr when boiled. In considering the consumption of whole marine fish, the IAEA (1985) estimated that relatively few people consumed whole fish such as anchovy. Understandably, the same bias towards temperate-derived data that gave rise to this study appears to apply to the consideration of food preparation techniques and consumption patterns. The diverse cultures occupying the tropics may be expected to have both different techniques for preparing fish and also a different proportion of fish in their diets than is evident in European cultures. There is a need to include the contribution of scales and bones to any estimation of dose from whole body loads of Sr radionuclides in teleost fish.

CONCLUSIONS

The equilibrium TFs for radionuclides of Cs and Sr measured in tropical freshwater fish were consistent across a range of species as measured in eight studies. However, the mean TFs for Sr and Cs were markedly lower than those expected for temperate freshwater fish based on the existing IAEA database. The apparent differences between the measured TFs in the flesh of tropical fish and the recommended TFs for temperate freshwater fish were greatly reduced by incorporating specific water chemistry variables into predictive accumulation models (*i.e.* the concentrations of K and suspended solids for Cs accumulation and the Ca concentration for Sr accumulation). Other factors, notably fish size and the experimental nature of the studies may have contributed to the low values. The effect of temperature was contrary to previous expectations. Although further data are required on the accumulation of Cs and Sr in tropical freshwater fish, the current results do not support the hypothesis of increased TFs for Cs in tropical freshwater fish. On the contrary, the results indicate that the use of recommended TFs for radiological dose assessment models, based on temperate freshwater fish, may provide overestimates of dose when applied to tropical fish.

Strontium was found to overwhelmingly reside in the scales and bones. Although these tissues may not be frequently consumed directly, they may contribute to radiological dose through leaching into soups, stews and other preparations using whole fish. It should be more broadly recognised that whole body Sr loads include a considerable fraction in scales as well as in bone. Any critical group analysis performed within tropical regions should take particular note of meals that include fish bones or skin in their preparation.

REFERENCES

Adam C., J. Garnier-Laplace and J.P. Baudin. Uptake of water, release and tissue distribution of ^{54}Mn in the rainbow trout (*Oncorhynchus mykiss* Walbaum). *Environ. Poll.* 97 (1 - 2), 29-38, 1997.

Carbonell G. and J.V. Tarazona. Toxicokinetics of copper in rainbow trout (*Oncorhynchus mykiss*). *Aquat. Toxicol.* 29, 213-221. 1994.

Cocchio L.A., D.W. Rodgers and F.W.H. Beamish. Effects of water chemistry and temperature on radiocesium dynamics in rainbow trout, *Oncorhynchus mykiss*. *Can. J. Fish. Aquat. Sci.* 52, 607-613. 1995.

Guimarães J.R.D. Bioaccumulation of ^{137}Cs and ^{60}Co by a tropical marine teleost *Epinephelus sp. Sci. Total Environ.* 120, 205-212. 1992.

Harris F., R. Domel and J. Crawford. The relative importance of radioecological parameters in tropical and subtropical regions - The use of a radiological consequence model to focus research. *Proc. International Meeting on Influence of Climatic Characteristics upon Behaviours of Radioactive Elements*. Inst. Environ. Sci. Rokkasho Village, Aomori, Japan. Oct 14-16, 1997.

Hewett C.J. and D.F. Jeffries. The accumulation of radioactive caesium from food by the plaice (*Pleuronectes platessa*) and the brown trout (*Salmo trutta*). *J. Fish Biol.* 13, 143-153. 1978.

IAEA. *Sediment Kds and Concentration Factors for Radionuclides in the Marine Environment*. Tech. Rep. Ser. No. 247. International Atomic Energy Agency, Vienna, 1985. p. 71.

IAEA. *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments.* Tech. Rep. Ser. No. 364. International Atomic Energy Agency, Vienna. 1994.

IAEA, in prep. *Transfer of radionuclides from air, soil and freshwater to the foodchain of man in tropical and subtropical environments*. IAEA TECDOC. Vienna.

Koulikov A.O. and I.N. Ryabov. Specific cesium activity in freshwater fish and the size effect. *Sci. Total Environ.* 112, 125-142. 1992.

Mollah A. S., A. Begum, S. Akther, M.A. Malek, S.M. Ullah and Z.H. Khan. Studies on radionuclide transfer from soil and freshwater to the foodchain of man in tropical environment of Bangladesh. *Report to the IAEA/FAO CRP*. 12-16 December 1994. Damascus.

Mollah A. S., A. Begum, S.M. Ullah and Z.H. Khan. Studies on radionuclide transfer from soil and freshwater to the foodchain of man in tropical environment of Bangladesh. *Report to the IAEA/FAO CRP*. 6-10 November 1995. Istanbul.

Mollah A. S., A. Begum, S.M. Ullah and Z.H. Khan. Studies on radionuclide transfer from soil and freshwater to the foodchain of man in tropical environment of Bangladesh. *Report to the IAEA/FAO CRP*. Final meeting, 16-20 June 1997. Vienna.

Ngo N. T. and N.T. Binh. Factors of radionuclide transfer from air, soil and fresh water to the food chain of man in monsoon tropical condition of Vietnam. *Report to the IAEA/FAO CRP*. Final meeting, 16-20 June 1997. Vienna.

Pally M. and L. Foulqier. Étude bibliographique sur la capacité et les modalites de la fixation du radiostrontium par les poissons. *CEA-BIB-232 (part 2).* Commissariat á L'Energie Atomique, Gif-sur-Yvette, p. 350. 1979.

Patel B. 1975. Field and laboratory comparability of radioecological studies, in *Design of Radiotracer Experiments in Marine Biological Systems.* IAEA Tech. Rep. Ser. No. 167. International Atomic Energy Agency, Vienna, pp. 211-239.

Pottinger T.G. and G.M. Calder. Physiological stress in fish during toxicological procedures: a potentially confounding factor. *Environ. Toxicol. Water Quality.* 10, 135-146. 1995.

Rowan D.J. and J.B. Rasmussen. Bioaccumulation of radiocesium by fish: The influence of physicochemical factors and trophic structure. *Can. J. Fish. Aquat. Sci.* 51, 2388-2410. 1994.

Sinakhom F., P. Supaokit, M. Punnachiya, N. Chantaraprachoom, N. Prasertchewchan, and P. Yamkate. Transfer factors of ^{137}Cs and ^{85}Sr by fresh-water fish in tropical environments. *Report to the IAEA/FAO CRP*. Final meeting, 16-20 June 1997. Vienna.

Srivastava A., H.O. Denschalag, O. Kelber and K. Urich. Accumulation and discharge behaviour of Cs-137 by zebra fish (*Brachydanio rerio*) in different aquatic environments. *J. Radioanal. Nucl. Chem.* 138, 165-170. 1990.

Srivastava A., S.J. Reddy, K. Urich and H.O. Denschalag. Uptake and release kinetics of ^{134}Cs by goldfish (*Carassius auratus*) and ^{137}Cs by zebra fish (*Brachydanio rerio*) in controlled aquatic environment. *J. Radioanal. Nucl. Chem.* 182, 63-69. 1994.

Stanek Z., M. Penáz and E. Wohlgemuth. Accumulation and kinetics of ^{90}Sr in fishes and other components of an artificial aquatic system, Folia Zool. 39, 375-383. 1990.

Twining J.R., J.M. Ferris and S.J. Markich. Measurement of transfer factors for radionuclides, including ^{137}Cs and ^{85}Sr, in freshwater fish and plant crops from tropical and sub-tropical Australia: Research summary. *Report to the IAEA/FAO CRP*. Final meeting, 16-20 June 1997. Vienna.

Ugedal O., B. Jonsson, O. Njåstad and R. Naeumann. Effects of temperature and body size on radiocaesium retention in brown trout, *Salmo trutta*. *Freshwater Biol.* 28, 165-171. 1992.

Whicker F. W. and V. Schultz. *Radioecology. Nuclear Energy and the Environment, Vol. 1.* CRC Press, Boca Raton. 1982.

Willis J. N. and N. Y. Jones. The use of uniform labelling with zinc-65 to measure stable zinc turnover in the mosquito fish, *Gambusia affinis* - I. Retention. *Health Phys.* 32, 381-387. 1977.

Table 1.

Results of experiments to determine Cs and Sr transfer factors and biological half-times in tropical freshwater fish and parameters measured to assist interpretation. Also included are the expected values and the results of applying predictive models from the temperate data (IAEA 1994). Blanks indicate that the data were not reported in the reference. FW - fresh weight.

		Cs-137			Sr-85/90								
Species	size (SL or mass)	TF modelled	TF measured L/kg (FW)	T½ days	TF modelled	TF measured L/kg (FW)	T½ days	Temp. C	K^+ mg/L	Ca^{2+} mg/L	Suspended solids mg/L	Notes	Reference
zebrafish *Brachydanio rerio*	0.24 g	1015	16	51				26	2.7		nil	whole fish	Srivastava 90
goldfish *Carassius auratus*	2-6 g	1015	4	19				28	2.7		nil	”	Srivastava 94
tilapia *Tilapia sp.*	30-40 g[a]		26	12								“	Ngo & Binh 97
carp *Cyprinus carpio*	10-30 g		40	31								“	
catfish *Clarias sp.*	160-270 g	59	4	29				27	29	67	100	flesh & skin	Sinakom et al. 97
		106	2	36	1	5	39	26.5	13	42	“	whole fish	
		72	3	41	2	0.1	4	29.6	22	64	“	flesh	
singhi *Heteropneustus fossiles*	22-24 cm		6	93		10	62	28	6.9	3.7	110	flesh	Mollah et al. 94
magur *Clarias batrachus*	20-25 cm	175	7	94	37	11	54	“	“	“	“	“	
climbing perch *Anabas testudines*	16-19 cm		6	81		14	80	“	“	“	“	“	
singhi	20-23 cm		6	110		15	73	27	7.2	4.8	118	whole fish	Mollah et al. 95
magur	18-21 cm	174	9	104	26	19	65	“	“	“	“	“	
climbing perch	14-17 cm		9	88		13	78	“	“	“	“	“	
singhi	≥ 20 cm		16	160		3	7	“	“	“	“	flesh	Mollah et al. 97
magur	“	174	19	182	26	4	8	“	“	“	“	“	
climbing perch	“		16	119		4	7	“	“	“	“	“	
silver perch *Bidyanus bidyanus*	18-25 cm	440	13	19	5	0.7	5	25	2.0	20	92	flesh	Twining et al. 97
Expected TF (range) from temperate data			2000 (30-3000)			60 (1-1000)							IAEA 94

a. 20 g fish (juveniles) were excluded from this analysis on the basis of failing normality and heteroscedasticity tests.