Sources of radioactivity in the marine environment and their relative contributions to overall dose assessment from marine radioactivity (MARDOS)

Final report of a co-ordinated research programme



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FOREWORD

The International Atomic Energy Agency's Marine Environment Laboratory has carried out a five years Coordinated Research Programme (CRP) on Sources of Radioactivity in the Marine Environment and their Relative Contributions to Overall Dose Assessment from Marine Radioactivity (MARDOS).

The objectives of the CRP were to summarize available data and provide new results on ¹³⁷Cs and ²¹⁰Po measurements in seawater and biota, to provide radiological assessment of doses to the world population from ¹³⁷Cs and ²¹⁰Po in marine food and to support and encourage marine radioactivity investigations in Member States. The results obtained represent the most complete data set available to Member States on radioactivity levels in the marine environment and on doses to the world population from marine radioactivity through ingestion of marine foods.

Three Research Coordination Meetings have been organized (IAEA-MEL Monaco, 1989; Risø National Laboratory, Roskilde, Denmark, 1991; IAEA-MEL, Monaco, 1993), where the objectives of the CRP, the classification, organization, compilation and synthesis of data and the assessment of radiological doses based on ¹³⁷Cs and ²¹⁰Po in fish and shellfish were discussed. The presentations and discussions covered both aspects of the CRP - radioactivity levels of ¹³⁷Cs and ²¹⁰Po in water and biota of the oceans and seas of the world and the assessment of doses to world population. A. Aarkrog, of Denmark, worked as chairman of all three meetings. The scientific secretaries of the meetings were A. Sanchez (1989, 1991) and P.P. Povinec (1993). The topics were discussed in three Working Groups:

Working Group 1:	¹³⁷ Cs and ²¹⁰ Po concentrations in water (Chairman: H. D. Livingston, USA),
Working Group 2:	¹³⁷ Cs and ²¹⁰ Po concentrations in biota (Chairman: A. de Bettencourt, Portugal),
Working Group 3:	Dose assessment (Chairman: E. Holm, Sweden).

The Chief Scientific Investigators were: A. Aarkrog of Denmark, A.O. Bettencourt of Portugal, R. Bojanowski of Poland, A. Bologa of Romania, S. Charmasson of France, I. Cunha of Brazil, R. Delfanti of Italy, E. Duran of the Philippines, E. Holm of Sweden, R. Jeffree of Australia, H.D. Livingston of the USA, S. Mahapanyawong of Thailand, H. Nies of Germany, Li Pingyu of China, J.N. Smith of Canada and D. Swift of the United Kingdom. The IAEA-MEL staff involved in the CRP were: M.S. Baxter, I. Osvath, P.P. Povinec and A. Sanchez.

The success of the CRP was due to the full collaboration of the participating institutions and Chief Scientific Investigators. The IAEA would like to express its gratitude for the information provided and for a most fruitful collaboration.

The work was coordinated in the Radiometrics Section of the IAEA's Marine Environment Laboratory in Monaco and the Responsible Officer was P.P. Povinec.

EDITORIAL NOTE

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SUMMARY

The document provides comprehensive information on radionuclide levels in the marine environment and estimates doses from marine radioactivity through ingestion of marine food. Two radionuclides - natural ²¹⁰Po and anthropogenic ¹³⁷Cs - are studied, as they are radiologically the most important representatives of each class of marine radioactivity on a global scale. Radioactivity levels of ²¹⁰Po and ¹³⁷Cs in sea water and biota (fish and shellfish) have been estimated for the FAO fishing areas on the basis of measurements which have been carried out in recent years. 1990 has been chosen as the reference year. Collective doses are calculated for each FAO area using radioactivity data for water and biota. A good agreement has been found between the results calculated by these two methods, with the exception of the doses from ²¹⁰Po by consumption of shellfish. The collective effective dose commitment for ¹³⁷Cs in marine food in 1990 is 160 man Sv with an estimated uncertainty of 50 %. The corresponding dose from ²¹⁰Po is 30 000 man Sv with an estimated uncertainty within a factor of 5.

The results confirm that the dominant contribution to doses comes from natural ²¹⁰Po in fish and shellfish and that the contribution of anthropogenic ¹³⁷Cs (mostly coming from nuclear weapons tests) is negligible (100 to 1000 times lower).

The results obtained in the framework of the MARDOS CRP provide the most complete data set available to Member States on radionuclide levels in the marine environment and on doses to the world population from marine radioactivity through ingestion of marine foods. The results will be used as the international reference source on the average radionuclide levels in the marine environment and corresponding collective committed effective doses from fish and shellfish consumption in each FAO fishing area.

1. INTRODUCTION

The International Atomic Energy Agency's Marine Environment Laboratory has carried out a five year Coordinated Research Programme (CRP) on "Sources of Radioactivity in the Marine Environment and their Relative Contributions to Overall Dose Assessment from Marine Radioactivity (MARDOS)". The objectives of the CRP were:

i) To summarize available data and provide new results on ¹³⁷Cs and ²¹⁰Po measurements in sea water and biota, characterizing FAO fishing regions

ii) To provide an assessment of doses to the world population from anthropogenic (^{137}Cs) and natural (^{210}Po) sources of radioactivity in marine food

iii) To support and encourage marine radioactivity investigations in Member States, especially in those which need methodological assistance.

Two radionuclides - anthropogenic ¹³⁷Cs and natural ²¹⁰Po - have been chosen, as they are from the radiological point of view the most important representatives of each class of marine radioactivity. The former is the most abundant anthropogenic radionuclide present in the marine environment and the latter is known to lead to the highest doses amongst natural radionuclides.

However, two different approaches were followed for these two radionuclides. Actually, their distribution in the marine environment is governed by different factors. The concentrations of 137 Cs vary from region to region, according to the different sources of contamination. The main global source of 137 Cs in the marine environment is fallout from nuclear tests performed in the atmosphere. In some regions, like the Irish Sea, the Baltic Sea and the Black Sea, the concentration of 137 Cs in the marine environment depends on the input due to discharges from the reprocessing facilities and from the Chernobyl accident, and in these regions the evolution of its concentration is quite dynamic. On the other hand, the changes in concentrations seem to be less dependent on the region, but they might vary by orders of magnitude, according to the species under consideration. In each species they range as widely from one tissue to another. Furthermore, as most of the 210 Po in marine species is relatively in excess of its radioactive parent 210 Pb, it decays when food is preserved for some time before consumption.

Laboratories representing 16 IAEA Member States participated in the work of the CRP, which started in 1989. The CRP participants collected huge amounts of data from their respective regions of interest as well as from other regions, consideration being given to the entire coverage of the oceans of the world. In addition, specific tasks were assigned to collect information available from countries not participating in the CRP so that these data could also be included in the CRP database. The data bank for the dose assessment included radionuclide concentrations in water and fish, statistics of the fisheries' catches in the various oceans as well as information on consumption habits.

The collected information is of high scientific value and represents the reference source on average radionuclide levels in the marine environment for each FAO fishing area. Several CRP participants have benefitted from the methodological help provided by the Agency. The present report summarizes the results obtained in the framework of the CRP. Radioactivity levels of 210 Po and 137 Cs in sea water and biota (fish and shellfish) have been estimated for the FAO fishing areas on the basis of measurements carried out in the framework of the CRP, other data provided by the CRP participants and data taken from the literature. Collective committed effective doses and mean individual doses from 210 Po and 137 Cs by consumption of fish and shellfish have been calculated separately using the water and biota data.

2. ¹³⁷Cs AND ²¹⁰Po CONCENTRATIONS IN WATER

 137 Cs and 210 Po activities of sea water normalized to 1990 in each FAO fishing region have been evaluated. Since these data would be used in global dose assessment models, they should be representative of the marine environment associated with the major commercial sources of fish and shellfish.

2.1. ¹³⁷Cs Compilation

Sea water data have been compiled both from the literature and from unpublished results provided by CRP participants. GEOSECS data [1] which were first decay-corrected to 1990 and then reduced by an additional 10% to account for surface sea water decreases owing to physical mixing, were used to corroborate other data sets or were used as the primary data set in the absence of other results from a given region.

For regions in which ¹³⁷Cs distributions were reasonably homogenous, a mean of the existing data set was determined. In cases in which there was a stronger latitudinal gradient in ¹³⁷Cs distributions, and most of the recent data were confined to one area of the FAO region, the representative ¹³⁷Cs activity was inferred from the GEOSECS data set. In cases in which the existing data set exhibited large variations which appeared to represent analytical uncertainties, decisions were occasionally made with regard to the reliability of each data set and these were weighted accordingly. For regions exhibiting great variability in ¹³⁷Cs distributions owing to oceanographic/geographic variability, a representative ¹³⁷Cs activity was determined by weighting the data according to the principal geographic focus of fishing activity. Data from regions in which the ¹³⁷Cs source function exhibits negligible variability were simply decay-corrected to 1990. These data being sufficiently recent, they did not require the mixing correction applied to the GEOSECS data set. For regions in which the ¹³⁷Cs source function was undergoing changes in 1990, the 1990 results were given primary consideration. If these data were not available, the existing historical data set for that region was extrapolated to derive a 1990 value.

The GEOSECS tritium data set was used as a check on the 137 Cs information for the Indian Ocean. Thus the 137 Cs/ 3 H ratios in the Atlantic Ocean were used to derive 137 Cs values at equivalent latitudes in the Indian Ocean and these agreed with the few available direct observations.

Two regions (FAO 27 - the NE Atlantic, and 37 - the Mediterranean and the Black Seas) were sufficiently heterogeneous in terms of their ¹³⁷Cs distributions and fish catches to require special treatment. These regions were first divided into sub-regions for which a representative ¹³⁷Cs activity was determined and then a representative ¹³⁷Cs activity was

FAO Area	Data and (Bq m	source 1 ⁻³)	Recommended value (Bq m ⁻³)
Pacific Ocean			
61 NW	5.4	PHI	4.0
	4.2	USA	
	3.7	CPR	
	2.8	JPN (NIRS)	
	3.3	Geosecs	
67 NE	3.7	Geosecs	3.9
	4.0	USA	
71 W Central	l 2.1	JPN	2.4
	4.8	PHI	
	3.6	THA	
	2.4	Geosecs	
77 E Central	2.6	FRA	2.7
	5.0	USA	
	2.7	Geosecs	
81 SW	2.2	JPN (MRI)	1.3
	1.3	NZ	
	1.0	Geosecs	
	1.4	AUS	
87 SE	2.6	JPN (MRI)	1.9
	1.2	Geosecs	
88 Antarctic	1.1	JPN (MRI)	0.3
	0.3	Geosecs	
Indian Ocean			
51 W	2.9	JPN (MRI)	2.9
57 E	2.8	JPN (MRI)	2.8
58 Antarctic	0.6	JPN (MRI)	0.5
	0.6	Geosecs	

TABLE I.SURFACE137Cs CONCENTRATIONS (on 01-01-1990) IN VARIOUS
OCEAN AREAS

FAO Area		Data and s (Bq m ⁻¹	ource] ³)	Recommended value (Bq m ⁻³)
1	8 Arctic	7.6	DEN (EGC/RISO)	7.6
2	21 NW	2.9	CAN	2.9
		2.6	USA	
		2.9	Geosecs	
3	1 W Central	2.0	USA	2.4
		2.7	Geosecs	
3	4 E Central	3.0	USA	2.4
		2.4	SWE	
		2.7	Geosecs	
		1.8	FRA	
4	1 SW	1.4	BRA	1.4
		1.4	SWE	
		1.4	Geosecs	
4	7 SE	1.3	GER	1.4
		1.4	Geosecs	
4	8 Antarctic	0.4	SWE	0.5
		0.5	GER	
		0.6	Geosecs	
Special area	IS			
3	7 Mediterranean	5.0	ITA (Coastal)	5.4
		3.7	ITA (W. Med.; oper	n)
		3.5	IAEA (W. Med.)	
		4.9	RUS (E. Med.; oper	n)
		11.8	GRE (Aegean)	
	Black Sea	48	ROM	52
		58	USA	

Note: Area 37 mean value = 13 (Based on weighted average Black Sea/Mediterranean Sea fish catch data.

FAO Area	Data an (Bq 1	d source n ⁻³)	Recommended value (Bq m ⁻³)		
27 NE Atlantic		<u> </u>	······································		
Baltic	125	GER	125		
Danish Straits	73	DEN	73		
North Sea	12	GER (1992)	12		
Faroese Waters	3.1	DEN	3.1		
Nor/Greenland Sea	6.8	DEN	6.8		
Iceland	2.8	DEN	2.8		
Irish Sea	55	GER/UK	55		
NOAMP area	3.0 3.2	GER SWE	3.1		
Barents Sea	10	UK	10		

Note: Area 27 mean value = 21 (Based on weighted average fish catch data).

<u>Data from CRP members</u>: (AUS = Australia; BRA = Brazil; CAN = Canada; CPR = China; DEN = Denmark; FRA = France; GER = Germany; GRE = Greece; IND = India; ITA = Italy, NOR = Norway; PHI= Philippines, POL = Poland; ROM = Romania; RUS = Russia, SWE = Sweden; THA = Thailand, UK = United Kingdom; USA = United States of America).

Other data: JPN (NIRS) = Japan/National Institute of Radiological Sciences, JPN (MRI) = Japan/Meteorological Research Institute, Papers in Meteorology and Physics, 39, 95-113 (1988); Geosecs = Derived from Geosecs surface sections, after correction for decay and mixing; EGC/RISO = East Greenland Current/Riso Nat. Lab., Denmark; RUS = Russia, Stepanets et al., Analyst, 117, 813-6, 1992.



FIG. 1. Concentrations of ¹³⁷Cs in water for FAO fishing areas.

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chosen for each entire region with some weighting for the magnitude of the fish catch in each sub-region.

Recommended values of 137 Cs in seawater arranged according to FAO fishing areas are listed in TABLE I and shown in Fig. 1.

2.2. Removal of Radiocaesium from the Upper-Mixed Layer of the Oceans

The fate and distribution of 137 Cs during 16 years in the surface waters of the North and South Atlantic can be assessed by using data from the GEOSECS expedition in 1972-73 [1], the Polish expedition in 1977-78 [2], and the Swedish Antarctic Expedition in 1988-89 [3].

The results for 137 Cs from the three scientific expeditions are displayed in Fig. 2. In order to evaluate removal processes other than radioactive decay of 137 Cs (dilution, sedimentation, biological removal) from the Atlantic surface waters during 1972 to 1990, all results were corrected for physical decay to 1990.

The results from the three expeditions show, after correction for physical decay, a very good agreement in the activity concentrations from 30°N to 70°S. The conclusion is that radiocaesium from nuclear test fallout has behaved conservatively in open Atlantic surface water. We must, however, consider other input sources since 1973.



FIG. 2. ¹³⁷Cs activity concentrations ($Bq m^3$) in surface waters from the North and South Atlantic in 1972/1973 (GEOSECS expedition [1]), 1977/1978 (Polish expedition [2]) and 1988/1989 (SWEDARP expedition [3]). All data are corrected for physical decay to 1990.

The estimated fission yields of atmospheric nuclear tests since 1973, mainly Chinese and French tests, are about 6 Mt, compared to a total of 217 Mt since 1945 [4]. The mean residence time of particulate debris injected into the stratosphere is of the order of one to a few years [4] and the remaining activity from older tests might give some contribution. The total deposited activity from 1973 to 1986 can be estimated as 6% of the total cumulative deposit in the northern hemisphere and 7% in the southern hemisphere. The cumulative deposit was measured over land and not at sea and probably includes some aeolian redistribution. The aeolian redistribution of radioactive material from land to sea is interesting and might be an important input to surface waters in certain areas, but few studies have been done.

The run-off from rivers is important in marginal seas such as the Baltic Sea, the Black Sea and eventually the Arctic Seas. This is due to the relatively large water input from rivers, large catchment and drainage areas contaminated by, for example, the Chernobyl accident.

In taking radioactive decay into account, the deposition of 137 Cs since 1973 can be estimated to be about 10% or less of the total integrated delivery up until 1990. We can derive from this that the half-life of 137 Cs in the surface waters of the North and South Atlantic is in the order of 100 years, corrected for physical decay. The effective half-life of 137 Cs will be about 25 years.

2.3. ²¹⁰Po Compilation

Sea water data have been compiled from the literature and from the unpublished results provided by CRP participants. The existing data sets were inspected to determine variability in each FAO region. There were two primary sources of variation related to input and removal of ²¹⁰Po. Some regions, receiving higher inputs of ²¹⁰Pb to the surface ocean owing to higher rates of atmospheric ²¹⁰Pb inputs, have higher levels of ²¹⁰Po in surface sea water.

Uptake of 210 Po onto particle surfaces (fractionated towards organic phases) and into phyto- and zooplankton results in removal of 210 Po from the more productive, shallower, marine regions with consequent diminished sea water activities. Scavenging of 210 Pb by particles (biased towards inorganic phases) has a smaller, but measurable, effect on reducing 210 Po activities in shelf regions. Despite the spatially and temporally heterogeneous distributions of 210 Po in the surface ocean, the existing data set indicates that there are only minor latitudinal or temporal gradients and that an average value of 1 Bq m⁻³ is acceptable with an uncertainty of 0.5 Bq m⁻³.

2.4. Evaluation of Errors in the ¹³⁷Cs and ²¹⁰Po Compilation

2.4.1. 137Cs Data

Generally, the analytical errors associated with the measurement of concentration levels of this radionuclide in the surface ocean are rather small, typically in the range of 3-6% and probably not greater than 10% in areas where levels are lowest, like Antarctic waters.

The real source of uncertainty comes from the fact that in some FAO regions there is a considerable gradient in concentrations which characterize the area. In some cases sampling has been biased to parts of these areas and in others few recent measurements have been made and it has been necessary to extrapolate average values from historical data using the methods described above.

Therefore, it is not realistic to provide meaningful estimates of uncertainties on a region by region basis. Rather it seems more appropriate to select an average estimated value to use for all the regions - and to add the caveat that the **actual** uncertainty may be more or less in individual areas. This will not be a major source of uncertainty in the overall calculation of doses therefore, this empirical approach may be justified.

An uncertainty of $\pm 25\%$ is to be associated with the average values shown in TABLE I and Fig. 1 for ¹³⁷Cs concentrations.

2.4.2. 210 Po Data

It was found impossible to derive meaningful data for surface 210 Po values for the FAO ocean areas because of the regional and seasonal variability which characterize these data. An average value of 1 Bq m⁻³ and a range of 0.5-1.5 Bq m⁻³ was used for each area.

3. ¹³⁷Cs AND ²¹⁰Po CONCENTRATIONS IN BIOTA

3.1. ¹³⁷Cs Compilation

The data reported by the CRP participants were reviewed and tabulated according to the FAO regions (TABLE II) [7]. However, the 137 Cs data existing for the NE Atlantic and adjacent seas (area 27) were subdivided into the Baltic, Irish, North, and Barents Seas and general NE Atlantic. Also the data from area 37 were subdivided into the Mediterranean and Black Seas. A global value for 137 Cs concentrations in areas 27 and 37 was then calculated, computing the average concentrations for those sub-regions against the seafood catch in each of them (TABLE III).

The data were classified into fish, mollusc and crustacea concentrations. A value for shellfish was finally chosen for each region, as the differences for ^{137}Cs in molluscs and crustacea were not significant (TABLE IV). Concentrations were expressed in terms of Bq kg⁻¹ wet weight of the edible fraction, whenever this distinction was available. Values reported as Bq kg⁻¹ dry weight were converted using a general ratio of 0.2 for dry weight/fresh weight.

Data reported by the CRP participants were used as much as possible for the dose calculations. However, in some regions, very few data were available and some did not seem to be sufficiently representative for the area. Therefore, these data were completed by literature survey, whenever necessary.

All data were then critically reviewed to analyse whether they could be considered representative of the significant seafood in each region. In the first phase, concentration factors were calculated for each region (and some sub-regions) using the available values for

TABLE II CON	/IPILATION OF 137C	Cs CONCENTRATIONS	S IN FISH, MOLLUS	CS AND CRUSTACE	A FOR THE DIFFERENT	Γ REGIONS
OF THE WORI	.D'S OCEANS (Bk/k	(g w w)				

Record	Reg	Fish	NI	81	Molluses	N2	S 2	Crustacea	N3	S 3	Year	Ref	Observations
1	21	0 34	1		0 21	3	0 13	0 11	2	0 07		CAN90	
2	21	0 28	1					0 08	1			CAN90	
4	21				0 03						90	USA93	
5	27	0 52	6	0 25							79-84	РТ93	Corrected for 1990
6	27	0 47	7	0 21							85-89	РТ93	Corrected for 1990
7	27	14 4	17	3 56							90	POL	Baltic Sea
8	27	14 8	22	5 79							91	POL	Baltic Sea
9	27	10 5	3	10								DEN90	Baltic Sea
10	27	1 06	18	0 34				06	4		90	UK93	North Sea (Crust-1990)
11	27	0 56	5	0 33								UK93	General North Atlantic
12	27	14	35	0 55	0 48	6	0 22	08	6	0 44	90	GER93	North Sea
13	27	2	6	07	0 52	6	0 24	0 95	5	0 49	85-87	DEN-MT	M North Sea S
14	27	12 2	33	59							91	GER93	Baltic Sea
15	27	119	81	15	10 2	53	89				90	UK93	Irish Sea - Shellfish
16	27	0 44	3	0 16								DEN90	Faroes
21	27	0 51	5	0 08							90	POL93	# Mean 27 Barents Sea
22	31	0 14	6	0 14							85-87	FRA93	Corrected for 1990
23	31	0 14	3	0 02							88	FRA93	Corrected for 1990
25	34	0 12	6	0 04							85-87	FRA93	Corrected for 1990
26	34	0 08	3	0 07							88	FRA93	Corrected for 1990
27	34	0 06	1								86	ROM	
28	34	0 58	1								88	ROM	
29	34	1 43	2	0 97							89	ROM	
30	34	0 49	14	0 34							79-84	РТ93	Corrected for 1990
31	34	04	11	02							85-89	РТ93	Corrected for 1990
33	37	44	22	19	1 56	8	0 94				87	ROM93	Black Sea
34	37	3 10	15	09	1 37	29	0 71				88	ROM93	Black Sea
35	37	3 5	18	07	1 39	30	0 53				89	ROM93	Black Sea
36	37	29	5	08	1 1 1	14	0 39				90	ROM93	Black Sea
37	37	0 35	3		0 3 1	1						MC90	
38	37	0 46	12	0 33	0 25	4	0 13					ITA90	
41	41	0 1 1	10	0 03								BRA93	
42	41	0 07	8	0 02	0 03	11	0 03				90	USA	
43	41	0 07	1		0 05	1					91	USA	
45	41	0 07	10	0 02	0 03	14	0 02				90-91	POL93	

Record#	Reg	Fish	NI	S 1	Molluses	N2	\$2	Crustacea	N3	\$3	Year	Ref	Observations
46	47	0.48	2	0.02							89	ROM	# Mean 47
47	48	0.33	6	0.36				0.34	1		89	SWE93	
48	48							0.01	3	0.01	91-93	POL93	
49	51	0.09	12	0.05							85-87	FRA93	Corrected for 1990
50	51	0.09	12	0.05							88	FRA93	Corrected for 1990
52	57										90	AUS93	Values below DL
84	57	0.14	8	0.09	0.04	2	0.01	0.09	3	0.04	90-91	THA93	# Mean 57
53	61	0.24	232	0.12	0.07	63	0.05				80-87	JPN	
54	61	0.19	57	0.08	0.05	17	0.01				88-89	JPN	
55	61	0.36	33	0.1							90	USA	
56	61	0.38	8	0.08							91	USA	
57	61	0.11	26		0.14	26		0.18	28		90	CPR93	Converted FR DW
58	61	0.13	1		0.05	7	0.01	0.05	3	0.01		PHI90	
59	61	0.21	36	0.08	0.09	26						PHI90	
60	61	0.24	102								85-87	PHI90	
62	61	0.34	53	0.07							90-91	POL93	
63	67	0.7	1								88	USA	67 and 77
64	67	0.3	1								89	USA	67 and 77
65	67	0.56	5	0.21							90	USA	67 and 77
67	67	0.5	10	0.2							88-91	POL93	
68	71	0.58	1					0.06	1			AUS	
69	71	0.41	7	0.51							85-87	FRA93	Corrected for 1990
70	71	0.13	3	0.02							88	FRA93	Corrected for 1990
72	71	0.53	48	0.47	0.24	13	0.2	0.23	4	0.12	88-93	PHI93	
71	71	0.11	12	0.06	0.04	3	0.01	0.03	8	0.01	89-91	THA93	
74	77	0.18	89	0.19	0.09	46	0.11	0.09	36	0.05		FRA90	
75	77	0.29	120	0.22								FRA90	
77	81	0.04	4		0.03	2		0.02	2			AUS	
79	87	0.07	8	0.03							85-87	FRA93	Corrected for 1993
80	87	0.06	4	0.01							88	FRA93	Corrected for 1993
81		0.06	4									FRA-MT	
82			0		0.15	4	0.13					CPR90	
Reg:	Region			Fish:		Concentra	tion in Fish		N	Molluscs:	Concentration	in Molluscs	
Crustacea:	Concentratio	on in Crustace	a	N:	1	Number of	f Samples (or	r pool samples)	1	S:	Standard Devi	ation	
Year:	Year of the r	measurements		Ref:]	Reference							

•

TABLE II. (continued)

FAO region	Fish	Number of samples	Molluscs	Number of samples	Crustacea	Number of samples	Observations
21	0.31	2	0.2.6	4	0.10	3	
27	1.50	59	0.50	6	0.78	15	North Sea
27	13.0	750					Baltic Sea
27	11.90	81	10.2	53			Irish Sea
27	0.54	21					remaining areas
27	0.51	5					Barents Sea
31	0.14	9					
34	0.45	38					
37	3.48	60	1.36	81			Black Sea
37	0.40	8	0.28	5			Mediterranean Sea
41	0.18	29	0.03	29			
47	0.48	2					
51	0.09	24					
57	0.14	8	0.14	2	0.09	3	
61	0.24	548	0.08	139	0.12	31	
67	0.52	17					
71	0.30	71	0.14	16	0.11	13	
77	0.23	209	0.09	46	0.09	36	
81	0.04	4	0.03	2	0.02	2	
87	0.06	16	0.115	4			

TABLE III. ARITHMETIC MEANS OF ¹³⁷ Cs CONCENTRATIONS IN FISH, MOLLUSCS AND CRUSTACEA, CALCULATED FOR FAO REGIONS (Bq kg ⁻¹ w.w.)

the concentrations of 137 Cs in water and seafood. The following ranges of concentration factors (CF) were found:

23 to 144 for fish, excluding three high values6 to 40 for molluscs, excluding one value5 to 52 for crustacea, excluding two values.

These values are in reasonable agreement with the concentration factors of 100 for fish and 30 for molluscs and crustacea, previously recommended by the IAEA [5]. Exceedingly high CF values should therefore not be used except if they are duly confirmed.

The data reported were not presented in a totally uniform manner, in particular with regard to the detection limits, which ranged over almost two orders of magnitude, and the average calculation which was sometimes an arithmetic mean and at others a geometric mean. For ¹³⁷Cs, arithmetic means were used, as it was not possible to treat all the available data in another way.

When there were few values below detection limits, these were not considered. However, some sets of values consisted of a large number of results below detection limits with only a few detectable ones, which occasionally were inconsistently high. In such cases, the average was critically reviewed to take account of the inconsistencies.

TABLE IV. ¹³⁷Cs CONCENTRATIONS IN FISH AND SHELLFISH FOR FAO REGIONS RECOMMENDED TO BE USED IN DOSE ASSESSMENT (Bq kg⁻¹ w.w.)

FAO		
Region	Fish	Shellfish
21	0.3	0.1
27	2.4 *	(1)
31	0.5	(0.1)
34	0.4	(0.1)
37	1.0 *	0.5 *
41	0.07	0.03
47	(0.1)	(0.04)
48		(0.02)
51	0.2	(0.08)
57	(0.2)	0.06
61	0.3	0.1
67	0.5	(0.15)
71	0.3	0.09
77	0.3	0.09
81	0.04	0.03
87	0.07	0.03

* Weighted for catch

() Estimated values

For three regions (47, 48 and 57) either there were no data available or there were too few values which were not reliable enough or did not seem representative for the region. For regions 47 and 57, values were suggested taking into account the general distribution of 137 Cs in the oceans and the values observed in the neighbouring regions. For region 48, it was decided that no value should be given.

It would have been possible to convert radionuclide concentration in sea water to that in fish using the recommended CF. However, this has been avoided in order to keep both calculations as independent as possible.

In one or two cases, the available values were not consistent with those observed in the neighbouring regions, and the values were corrected accordingly. In many regions there were no values at all for molluscs and especially for crustacea. As the values were not too different, it was decided to use only one value for shellfish in each region. In those areas where values were still not available, the concentration in shellfish was deducted from that in the fish from the same region.

The average concentrations for each group of seafood and for each FAO region are given in TABLE II. From these values, a mean concentration was calculated for each region (or sub-region of regions 27 and 37), (TABLE III).



FIG. 3. Concentrations of ^{137}Cs in fish for FAO fishing areas.

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FIG. 4. Concentrations of ¹³⁷Cs in shellfish for FAO fishing areas.

Record #	Reg	Fish	Number of samples	Standard deviation	Molluscs	Number of samples	Standard deviation	Crustacea	Number of samples	Standard deviation	Year	Ref	Observations
1	27	3.1	9	2.1	77	58	89	50	4	3	85-89	PT90	
2	27	0.7	16	0.6							1990	POL90	
3	27	0.8	20	0.5							1991	POL90	
4	27	7.5	4	4.4							1990	POL90	
5	27	0.6	2	0.3							1991	POL90	
6	27	0.5	1								1991	POL90	
8	34	1.2	14	1.1	35	1		100	2	85	85-89	PT90	
9	37	2.3	6	0.8							1991	ROM93	Black Sea
10	41	0.1	7	0.03	0.8	10	0.1				1990	USA	
11	41	0.2	1		1.1	1					1991	USA	
13	57	0.8	4	0.3			7.1	2	0.3			AUS	
14	61	0.2	41	0.08							1990	USA	
15	61	0.3	8	0.1							1991	USA	
16	61	2	11		2.4	5		1.6	8		90-91	CPR93	
17	61	0.2	58	0.1							90-91	POL93	
19	67	0.9	5	0.6							1990	USA	67 & 77
20	71	3.1	37	2.2	1.8	6	1.4	1.4	1		89-93	PH193	
21	71	1.2	10	0.9	4	2	0.2	5.6	6	7	90-91	THA93	
23	81	2.5	10	2	48	1		4.8	1			AUS	
24	27	6.9	14	8.9	42	51	52	59	10	39	82-90	UK93	
25	27	0.1	2	1.6	9.4	4	4.5				1990	ROM93	
26	27										92-93	FRA93	
27	27	21	5	25	101	8	99	43	6	20	90-92	FC93	Mussels & Squids
28	37	6.5	2	3.5	4.8	3	3.2	17	3	27		FC93	Mussels & Squids
29	41	0.1	9	0.07	0.4	13	0.1				89-91	POL93	
30	48	1.6	6	1.2								SWE93	
31	48							0.4	3	0.2	91-93	POL93	
32	57	3	8	2.4	11	2	13	9	4	8	90-91	THA93	
33	67	0.8	11	0.6							90-91	POL93	
35	77	0.9	5	0.6							1991	USA90	Mean of 77, 67,77

TABLE V. CONCENTRATIONS OF ²¹⁰ Po IN FISH, MOLLUSCS, AND CRUSTACEA FOR FAO REGIONS (Bq kg ⁻¹ w.	'.w.)
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FAO region	Fish	Number of samples	Molluscs	Number of samples	Crustacea	Number of samples	Observations
27	5.8	73	57	121	38	21	
34	1.2	14	35	1	100	2	
37	4.4	8	4.8	3	17	3	
41	0.1	17	0.8	24			
48	1.6	6			0.4	3	
57	1.9	12	11	2	8.1	6	
61	0.7	118	2.4	5	1.6	8	
67	0.9	16					
71	2.1	47	2.9	8	3.5	7	
77	0.92	5					Mean of 77, 67 & 77
81	2.5	10	48	1	4.8	1	, ,

TABLE VI. AVERAGE ²¹⁰Po CONCENTRATIONS OF FISH, MOLLUSCS AND CRUSTACEA, CALCULATED FOR FAO REGIONS (Bq/kg w.w.)

These data were critically reviewed and final ¹³⁷Cs concentrations in fish and shellfish for the different regions, as recommended values to be used in dose assessment, are given in TABLE IV. These recommended concentrations are shown in maps, for fish and for shellfish, in Figs. 3 and 4, respectively. Values within brackets were estimated as mentioned above.

3.2. ²¹⁰Po Compilation

While for ¹³⁷Cs the data were compiled for each FAO region, a similar treatment for ²¹⁰Po showed first that, as expected, there are very few or no values available for some regions, and secondly that there seem to be no significant differences in concentration from one ocean to another. The regional differences, if any, are below the fluctuations observed from one species to another or even from one species to another.

For the above reasons, the data for each group of seafood, from all regions of the world's oceans, were combined and analyzed together. Geometric means for the concentrations of ²¹⁰Po in each of the seafood groups were calculated both for the data reported by the CRP participants and from the literature survey.

The compilation results are shown in TABLE V. The averaged concentrations are presented in TABLE VI and Figs. 5, 6 and 7. It can be observed that in several regions there are no values available and, in others, the average is calculated over very few values.



FIG. 5. Concentrations of ²¹⁰Po in fish for FAO fishing areas.

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FIG. 6. Concentrations of ²¹⁰Po in molluscs for FAO fishing areas.



FIG. 7. Concentrations of ²¹⁰Po in crustacea for FAO fishing areas.

		Literature limits				Participants limits				All limits			
	G.M.	(S.D.)	(S.E.)	N	G.M.	(S.D.)	(S.E.)	Ν	G.M.	(S.D.)	(S.E.)	N	
Fish	2.1	13	2.7	66	2 1	13	4		24	12.7	2.8		
	2.1	0.35	1.7	00	3.1	0.76	2.4	33	2.4	0.45	2.0	99	
		61	22			73	18			68	18		
Molluscs	18			35	12			24	15			59	
		5.2	14			2.1	8.5			3.3	12		
		24	9.3			21	8.1			23	7.9		
Crustacea	6.5			14	5.2			10	6			24	
		1.7	4.5			1.3	3.3			1.6	4.5		

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TABLE VII. GEOMETRIC MEANS OF ²¹⁰Po CONCENTRATIONS IN FISH, MOLLUSCS AND CRUSTACEA (Bq/kg w.w.)

Reviewing the kind of species which were analyzed, it was concluded that the differences were related to species rather than to regions. An attempt was made to find typical values for species within the larger groups of fish, molluscs and crustacea. However, there are too few values to obtain reliable mean concentrations.

The frequency distribution of ²¹⁰Po concentrations in the three groups of seafood was therefore analyzed, for the data reported by the participants, for the literature survey, and for both. The results of this analysis are presented in TABLE VII, with the geometric mean and respective upper and lower limits, taking into account the standard deviation of the values and the standard deviation of the mean. It should be noticed that the number of observations corresponds to the number of references used and not to the number of samples analyzed.

A global concentration of 2.4 Bq kg⁻¹ w.w. in fish, 15 Bq kg⁻¹ w.w. in molluscs $6 Bq kg^{-1} w.w.$ in crustacea was then calculated for ²¹⁰Po and used in the dose assessment.

3.3. Expected Reliability of the Methods

3.3.1. 137Cs Data

Taking into account the large number of data existing on ¹³⁷Cs concentrations in the marine environment and the amount of research work performed on this radionuclide, it may be expected that the results obtained for this radionuclide are reliable. The uncertainties increase in the areas where the concentrations are lower and fewer studies have been made.

In the areas receiving ¹³⁷Cs discharges, where the higher doses are expected to arise, the uncertainty can be expected to be lower. However, in these regions, mainly the North East Atlantic and the Mediterranean, the distribution of concentrations is far from uniform. Therefore the populations will be exposed to quite different dose domains, according to the fishing zones.

Considering the methods used in the treatment of the data, the uncertainty can be expected to be not more than 50% in those areas where values were estimated. Regions 3, 47, 57, 77 and 81, where the uncertainties seem to be higher, only account for about 5% of the dose. This means that an uncertainty of 50% in the data from these regions would lead to an uncertainty in the dose of less than 3%.

The uncertainty will be higher for shellfish than for fish, but the impact in the final exposure will be much less because the consumption of shellfish only accounts for about 10% of the dose caused by the intake of 137 Cs through seafood consumption. An uncertainty of 50% in 137 Cs concentrations would lead to a final uncertainty of around 5%.

The highest contributions to dose come from areas 27 and 61. Concerning area 27, the main error would come from the uneven distribution of 137Cs in this region and from its continuous evolution. It is difficult to define a level of uncertainty for this region, but it is reasonable to expect that it will not be greater than 30%.

3.3.2. 210 Po Data

The reliability of 210 Po results is much lower than for 137 Cs. The main reasons for the uncertainties in 210 Po are the following:

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a) The number of studies on 210 Po in the marine environment is considerably less than those on 137 Cs. In many regions very little data are available on 210 Po concentrations in fish and especially shellfish.

b) The methods of analysis are more complex than those used for 137 Cs and fewer international intercalibration exercises have been performed.

It can be observed that the standard deviations are quite high, reflecting the wide range of reported concentrations. In the case of molluscs, this does sometimes reflect the large differences in concentrations between squids and mussels, the latter displaying significantly higher concentrations than the former.

c) There is a very important variability of 210 Po concentrations, which may be related to the trophic level of the species. Furthermore, large differences are observed in the concentrations in different tissues, and it is difficult to take due account of them, as the food habits also change from place to place and from species to species. For example, small planktivorous fish, which show the highest 210 Po concentrations, are eaten whole, whilst in other fish, only the flesh is eaten.

d) An average delay between fishing and consumption has to be assumed to account for the radioactive decay of 210 Po.

As mentioned above, the uncertainty in 210 Po concentrations is much higher than for 137 Cs. An envelope of values might reasonably be given by the standard deviation of the mean.

3.4. Future Studies on Radioactivity in Biota

Concerning biota, a good data set exists for 137 Cs in fish, enabling a quite reliable dose assessment. Data for shellfish are, however, less abundant and values had to be estimated for several regions. Missing data can, however, be deduced from concentrations in sea water or from concentrations in fish, when these exist.

The CRP contributed significantly to increasing the amount of existing data on 210 Po, particularly in regions where such data were totally non-existent. The data obtained here on this radionuclide have enabled the assessment of the dose to the world population, and emphasise the importance of 210 Po as a major contributor to the global dose.

There is need to establish a uniform presentation of results from all the laboratories of the world. Some recommendations might be useful for future treatment of data, for 137 Cs as well as for other radionuclides:

a) Concentrations in seafood should always be reported in units of Bq kg⁻¹ wet weight.

b) Values should always refer to the fraction of the fish or shellfish analyzed (whole body, flesh only, which edible part, etc.).

c) Concentrations should be accompanied by the sampling date and the date of measurement if these differ.

d) The detection limit should be indicated whenever values below detection limit are reported; also the confidence interval of such detection limits should be indicated.

4. DOSE ASSESSMENT

4.1. Concentration Factors

The dose from consumption of marine food is calculated by two different methods, i.e. using the estimated activity concentrations of 137 Cs and 210 Po in water (for 1990) for different fishing areas and applying recommended concentration factors (Method 1), and also using estimated concentrations in the marine products (for 1990), fish and shellfish (Method 2). The concentration factors used were based on IAEA recommended values [5]. The recommended concentration factor for 210 Po is lower for molluscs (10 000) than crustacea (50 000), but the MARDOS CRP expert group felt that it should be the opposite. A value of 30 000 was chosen for shellfish including both molluscs and crustacea in agreement with the MARINA project [6] (Table VIII).

The fish catch for different major fishing areas was calculated using FAO statistics for 1990 [7]. The factors used for the committed effective dose calculations for adults from intake of radionuclides are those used in the MARINA project [6], i.e. $1.2 \cdot 10^{-8}$ Sv Bq⁻¹ for 137Cs and $4.3 \cdot 10^{-7}$ Sv Bq⁻¹ for 210Po. Using an effective half-life of 70 days for 137Cs in the human body, the first year dose from a one-time oral intake is 97 % of the committed effective dose.

Matrix	Caesium	Polonium
Fish	100	2 000
Shellfish	30	30 000

TABLE VIII. CONCENTRATION FACTORS FOR POLONIUM AND CAESIUM IN MARINE PRODUCTS

4.2. Calculation of Doses

Method 1. The doses were calculated using the following formulae:

$$D_{Cs}(fish) = C_{w} \cdot 100 F_{c}F_{h}F_{e} \cdot 1.2 \cdot 10^{-8} = 4.2 \cdot 10^{-7}C_{w}F_{c} \quad [man \ Sv],$$
(1)

and

$$D_{Cs}(\text{shellfish}) = C_w \cdot 30 F_c F_h F_e \cdot 1.2 \cdot 10^{-8} = 1.8 \cdot 10^{-7} C_w F_c \quad [\text{man Sv}],$$
 (2)

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where

- D_{cs} is the collective committed effective dose from ¹³⁷Cs by consumption of fish and shellfish respectively from intake during 1990,
- C_w is the activity of sea water (Bq l⁻¹),
- F_c is the catch calculated from FAO statistics (kg per year),
- F_h is the fraction of the catch which goes for human consumption and is assumed to be
- 0.7 for fish and 1.0 for shellfish, and
- F_e is the fraction actually eaten and is assumed to be 0.5.

A delay factor (D_f) between catch and consumption has to be considered for polonium since the physical half-life of ²¹⁰Po is 138 days. Statistics show that 30% of seafood is eaten fresh, 30% frozen, 20% smoked and 20% canned. The delay in time between the different products is 0.1, 2 and 12 months respectively, giving a weighted mean of 93 days, i.e. slightly less than one physical half-life of ²¹⁰Po, but one half-life is applied in the calculations. The doses for polonium can accordingly be calculated as:

$$D_{Po}(fish) = C_w 2000 F_c F_h F_e D_f 4.3 \cdot 10^{-7} = 1.5 \cdot 10^{-4} C_w F_c \quad [man \; Sv],$$
(3)

and

$$D_{Po}(shellfish) = C_w 3.10^4 F_c F_h F_e D_f 4.3.10^{-7} = 3.2.10^{-3} C_w F_c \text{ [man Sv]}.$$
(4)

Numerical values for concentration factors, as given in TABLE VIII, were introduced in the above formulae.

Method 2. With notations similar to those used for Method 1, the following equations were applied to calculate doses:

$$D_{Cs}(fish) = C_b \cdot F_c F_h F_e \cdot 1.2 \cdot 10^{-8} = 4.2 \cdot 10^{-9} C_b F_c \quad [man \; Sv],$$
(5)

$$D_{Cs}(\text{shellfish}) = C_b \cdot F_c F_h F_e \cdot 1.2 \cdot 10^{-8} = 6.0 \cdot 10^{-9} C_b F_c \quad [\text{man Sv}], \tag{6}$$

$$D_{Po}(fish) = C_b F_c F_h F_e D_f 4.3 \cdot 10^{-7} = 7.6 \cdot 10^{-8} C_b F_c \quad [man \; Sv],$$
(7)

and

$$D_{Po}(\text{shellfish}) = C_b F_c F_h F_e D_f 4.3 \cdot 10^{-7} = 1.1 \cdot 10^{-7} C_b F_c \text{ [man Sv]},$$
 (8)

where

 C_b is the radionuclide concentration in the edible part of marine biota (Bq kg⁻¹ w.w.).

The collective effective dose commitment for fish and shellfish caught during 1990 calculated for FAO areas using Methods 1 and 2 are shown in TABLES IX-XII and Figs. 8-11.

The resulting collective effective dose commitment from fish and shellfish caught during 1990 calculated using the two different methods are summarized in Table XIII. The mean individual doses for a world population of $5.3 \cdot 10^9$ are shown within parentheses.

		¹³⁷ Cs concentration in		¹³⁷ Cs concentration in fis	h
Area	Fish-catch (t)	water (Bq/m ³)	CEDC (man Sv)	(Bq/kg)	CEDC (man Sv)
21	2171721	2.9	2.65	0.3	2.74
27	7872569	21	69.44	2.4	79.36
31	1225398	2.4	1.24	0.5	2.57
34	3813212	2.4	3.84	0.4	6.41
37	1131267	13	6.18	1	4,75
41	1381933	1.4	0.81	0.07	0,41
47	1507349	1.4	0.89	0.1	0.63
48	43076	0.5	0.00		
51	3040221	2.9	3.70	0.2	2,55
57	2296242	2.8	2.70	0.2	1,93
58	4657	0.5	0.00		
61	1994942	4	33.52	0.3	25,14
67	2723513	3.9	1.46	0.5	5,72
71	6259975	2.4	6.31	0.3	7.89
7 7	1294078	2.7	1.47	0.3	1,63
81	883004	1.3	0.48	0.04	0.15
87	1360863	1.9	10.86	0.07	4.00
			Total 148.5 (28.0 nSv/person)		Total 145.9 (27.5 nSv/nersc

Table IX. COLLECTIVE EFFECTIVE DOSE COMMITMENT (CEDC) FOR DIFFERENT FAO AREAS FROM ¹³⁷Cs BY FISH CONSUMPTION (1990), BASED ON ESTIMATED CONCENTRATIONS IN FISH AND WATER

Area	Fish Catch (t)	¹³⁷ Cs concentration in water (Bq/m ³)	CEDC (man Sv)	²¹⁰ Po concentration in fish (Bq/kg)	CEDC (man Sv)
21	2171721	1	327.93	2.4	393.52
2.7	7872569	1	1188.76	2.4	1426 51
31	1225398	ī	185.04	2.4	222.04
34	3813212	1	575.80	2.4	690.95
37	1131267	1	170.82	2.4	204.99
41	1381933	1	208.67	2.4	250.41
47	1507349	1	227.61	2.4	273.13
48	43076	1	6.50	2.4	7.81
51	3040221	1	459.07	2.4	550.89
57	2296242	1	346.73	2.4	416.08
58	4657	1	0,70	2.4	0.84
61	1994942	1	3012.33	2.4	3614.84
67	2723513	1	411.25	2.4	493.50
71	6259975	1	945.26	2.4	1134.31
77	1294078	1	195.41	2.4	234.49
81	883004	1	133.33	2.4	160.00
87	1360863	1	2054,90	2.4	2465.89
			Total 10450 (1970 nSv/person)		Total 12540 (2370 nSv/person)

TABLE X. COLLECTIVE EFFECTIVE DOSE COMMITMENT (CEDC) FOR DIFFERENT FAO AREAS FROM ²¹⁰Po BY FISH CONSUMPTION (1990), BASED ON ESTIMATED CONCENTRATIONS IN FISH AND WATER

	Crustacea	Molluscs	Crustacea and	¹³⁷ Cs concentration		¹³⁷ Cs concentration in	
Area	catch (t)	catch (t)	molluscs catch (t)	in shellfish (Bq/kg)	CEDC (man Sv)	water (Bq/m ³⁾	CEDC (man Sv)
21	276552	744208	1020760	0.1	0.61	2.9	0.53
27	238418	792335	1030753	1	6.18	21	3.90
31	251059	210966	462025	0.1	0.28	2.4	0.20
34	62901	207701	270602	0.1	0.16	2.4	0.12
37	55758	287336	343094	0.5	1.03	13	0.80
41	85746	561229	646975	0.03	0.12	1.4	0.16
47	12384	12489	24873	0.04	0.00	1.4	0.01
48	350213		350213	0.02	0.04	0.5	0.03
51	279174	46610	325784	0.08	0.16	2.9	0.17
57	248819	131627	380446	0.06	0.14	2.8	0.19
58	30343		30343	0.5	0.09	0.5	0.003
61	1407298	3648803	5056101	0.1	3.03	4	3.64
67	156090	103810	259900	0.15	0.23	3.9	0.18
71	494226	477765	971991	0.09	0.52	2.4	0.42
77	77424	129636	207060	0.09	0.11	2.7	0.10
81	6282	137411	143693	0.03	0.03	1.3	0.03
87	125970	162657	288627	0.03	0.05	1.9	0.10
88	658		658	0.5	0.00	0.5	0.000
					Total 12.8		Total 10.6
					(2.4 n Sv/person))		(2.0 nSv/person)

TABLE XI. COLLECTIVE EFFECTIVE DOSE COMMITMENT (CEDC) FOR DIFFERENT FAO AREAS FROM ¹³⁷Cs BY SHELLFISH CONSUMPTION (1990), BASED ON ESTIMATED CONCENTRATIONS IN BIOTA AND WATER

TABLE XII. COLLECTIVE EFFECTIVE DOSE COMMITMENT (CEDC) FOR DIFFERENT FAO AREAS FROM ²¹⁰Po BY CONSUMPTION OF SHELLFISH (1990), BASED ON ESTIMATED CONCENTRATIONS IN BIOTA AND WATER

Area	Crustacea catch (t)	²¹⁰ Po concentration in crustacea (Bq/kg)	CEDC (man Sv)	Molluscs catch (t)	²¹⁰ Po conc. in molluscs (Bq/kg)	CEDC (man Sv)	Crustacea and molluscs catch (t)	²¹⁰ Po concentration in water (Bq/m ³)	CEDC (man Sv)
21	276552	6	178.38	744208	15	1200.04	1020760	<u> </u>	3297.05
27	238418	6	153,78	792335	15	1277.64	1030753	1	3329.33
31	251059	6	161.93	210966	15	340.18	462025	1	1492.34
34	62901	6	40.57	207701	15	334.92	270602	1	874.04
37	55758	6	35,96	287336	15	463.33	343094	1	1108.19
41	85746	6	55.31	561229	15	904.98	646975	1	2089.73
47	12384	6	7.99	12489	15	20.14	24873	1	80.34
51	279174	6	180,07	46610	15	75.16	325784	1	1052.28
57	248819	6	160,49	131627	15	212.25	380446	1	1228.84
61	1407298	6	907.71	3648803	15	5883.69	5056101	1	16331.21
67	156090	6	100.68	103810	15	167.39	259900	1	839.48
71	494226	6	318,78	477765	15	770.40	971991	1	3139.53
77	77424	6	49.94	129636	15	209.04	207060	1	668.80
81	6282	6	4.05	137411	15	221.58	143693	1	464.13
87	125970	6	81.25	162657	15	262.28	288627	1	932.27
48	350213	6	225.89				350213	1	1131.19
58	30343	6	19.57				30343	1	98.01
88	658	6	0.42				658	1	2.13
			Total 2683 (506 nSv/person)	-		Tota1 12340 (2330 nSv/person)			Total 38160 (7200 nSv/person)



FIG. 8. Contribution from individual FAO fishing areas to the collective effective dose commitment from ¹³⁷Cs in fish caught in 1990 (water/biota data).



FIG. 9. Contribution from individual FAO fishing areas to the collective effective dose commitment from ²¹⁰Po in fish caught in 1990 (water/biota data).



FIG. 10. Contribution from individual FAO fishing areas to the collective effective dose commitment from ¹³⁷Cs in shellfish caught in 1990 (water/biota data).



FIG. 11. Contribution from individual FAO fishing areas to the collective effective dose commitment from ²¹⁰Po in shellfish caught in 1990 (water/biota data).

TABLE XIII. COLLECTIVE EFFECTIVE DOSE COMMITMENT FROM FISH AND SHELLFISH CAUGHT IN 1990. AVERAGE INDIVIDUAL DOSES (μ Sv), WITHIN PARENTHESES.

	13	⁷ Cs	210p ₀ [man Sv]			
	[ma	in Sv]				
Matrix	Method 1	Method 2	Method 1	Method 2		
Fish	150 (0.03)	146 (0.03)	10 000 (1.9)	12 000 (2.3)		
Shellfish	11 (0.002)	13 (0.002)	38 000 (7.2)	15 000 (2.8)		

The two different methods give almost identical results except for the doses from 210 Po by consumption of shellfish. Considering the possible errors in the different estimations, even this difference, by a factor 2.5, is acceptable. If, for instance, a lower concentration factor, such as 10 000, had been used for crustacea, the dose from shellfish would decrease from 38 000 to 30 000 man Sv. The concentration factors used in Method 1 apply to the whole organisms (or whole soft parts) whereas results in Method 2 were obtained on edible parts only.

The contribution of 137 Cs to the collective effective dose commitment from fish and shellfish consumption is negligible, below 1% of that for 210 Po (Fig. 12).

Assuming that there will be no additional sources or changes in predicted input of these nuclides to the oceans and that the effective residence time for radiocaesium is 25 years, the integral dose to the population from 1990 onwards can be calculated according to the formula

$$\mathbf{D}(\infty) = \int_0^\infty D_i e^{-\frac{\ln 2}{25}t} dt,$$

where

 $D(\infty)$ is the integrated dose from 1990 to infinity, and D_i is the collective effective dose commitment from consumption during 1990.

The dose becomes 5 300 man Sv (or a mean individual dose commitment of 1 μ Sv for a world population of 5.3·10⁹) from consumption of fish and 450 man Sv (or a mean individual dose commitment of 0.08 μ Sv) from consumption of shellfish. These figures can be compared with the doses of 10 000 man Sv received in one year by consumption of ²¹⁰Po in fish and 20,000 man Sv (an average value) by consumption of shellfish (or annual mean individual doses of 2 and 4 μ Sv, respectively).



5 FIG. 12. Contribution from individual FAO fishing areas to the collective effective dose commitment from ¹³⁷Cs and ²¹⁰Po in fish caught in 1990 (biota data).

The dose can also be calculated for a critical group (Area 27, NE Atlantic) tentatively consuming 100 kg of fish and 10 kg of shellfish per year (1990). The annual dose from ¹³⁷Cs will be 3 μ Sv by consumption of fish and 0.1 μ Sv by consumption of shellfish. The corresponding figures for ²¹⁰Po are 100 μ Sv for fish and 60 μ Sv for shellfish.

4.3. Uncertainties in Doses

The concentration factors for caesium in fish depend on size and species, but, because commercial fishing mainly takes place in fish-rich-regions and is selective as far as the size and species of the fish are concerned, the overall error in the concentration factor for caesium is estimated at 10%. Fewer data are available for polonium and an error of 30% is estimated. According to the IAEA [5], the concentration factors of radiocaesium in molluscs and crustacea are similar, but differ in the case of polonium. The MARDOS expert group considered that molluscs should have a higher CF than crustacea, as opposed to the values recommended by the IAEA. In the present calculations, a general value of 30 000 was used for 210 Po in shellfish. Large variations are found in the literature and therefore the overall error must be assessed at 50%.

The factors for the collective effective dose commitment from intake of radionuclides are assumed to be correct within 10%. The fish catch, F_c , may be slightly underestimated because of unreported catches and year-to-year variations. An uncertainty in the FAO values of 10% for both fish and shellfish is considered. The fraction going to human consumption, F_h , is the major fraction in all cases and an uncertainty of 10% is estimated. The fraction eaten, F_e , varies according to species and food habits. The factor of 0.5 may be an underestimation for certain regions. On the other hand, food is often discarded after cooking. The overall error for F_e is estimated at 10%.

If we assume that the uncertainties are independent of each other, the total of the estimated errors in the calculations can be worked out to 30 and 50% for estimates of 137 Cs doses due to the consumption of fish and shellfish using Method 1 (water data) and 50 and 70%, respectively, using Method 2 (biota data for both fish and shellfish.).

For polonium, the delay in time between catches and consumption is very important and is estimated at 90 ± 20 days which gives a correction factor of 0.6 ± 0.1 The estimated uncertainties in calculated doses for polonium for the consumption of fish and shellfish become 60 and 70%, respectively, using Method 1. Method 2 gives estimations of doses from ²¹⁰Po in fish and shellfish within a factor of about 5.

5. DISCUSSION

5.1. Reliability of the Assessment

The good agreement between the two methods for ^{137}Cs suggests that the ^{137}Cs concentrations in the marine environment are fairly well known. The major uncertainty in the dose assessment comes primarily from the fact that the actual intake of ^{137}Cs with marine foods strongly depends on the reliability of catch statistics, on knowledge of the fraction of marine products actually eaten and on possible losses of ^{137}Cs during cooking. It is believed that these uncertainties in general contribute to an overestimation here of the doses from ^{137}Cs .

In the case of ²¹⁰Po, the radioactivity measurements are less reliable than those for ¹³⁷Cs, particularly for biota. Furthermore, inhomogeneity in the internal distribution of ²¹⁰Po within an organism makes it additionally difficult to estimate the actual intake of this nuclide.

The global 137 Cs dose assessment is estimated to be correct within 50%, but the 210 Po assessment probably has an uncertainty factor of about 5.

5.2. Importance of other Marine Pathways and Radionuclides

In the present study, only the fish/shellfish-man pathway has been considered. From studies, particularly in the UK around Sellafield [8,9], it is, however, well known that other marine pathways may also be of interest, particularly for radionuclides other than those dealt with here. Critical pathways have involved 106 Ru in edible seaweed, transuranics in molluscs and external dose to fishermens' hands and to boat-dwellers. On a global scale, although the consumption of seaweed is generally low, this pathway is of some regional importance, e.g., especially, in the Far East (Japan). In the case of 210 Po, the food-chain is the only important marine pathway. As the concentration factor for 210 Po in macro algae is 10^3 [ref. 5], there may be a contribution from this source. In a global context, the contribution from pathways other than that for fish/shellfish ingestion is believed to be less than 10 % of the collective dose from 137 Cs and 210 Po in the marine environment.

The world's oceans also contain other radiologically significant radionuclides besides 137 Cs and 210 Po. For example, from nuclear weapons testing in the atmosphere, 90 Sr, 239,240 Pu, 241 Am, 3 H, and 14 C are still present in measurable quantities. In surface ocean waters contaminated only by global fallout, the concentrations relative to 137 Cs are as follows:

 ${}^{90}\text{Sr}/{}^{137}\text{Cs} \approx 0.66 \text{ [ref. 10]}$ ${}^{239,240}\text{Pu}/{}^{137}\text{Cs} \approx 2 \cdot 3 \cdot 10^{-3} \text{ [ref. 3]}$ ${}^{241}\text{Am}/{}^{137}\text{Cs} \approx 1 \cdot 10^{-3} \text{ [ref. 10, 3]}.$

Tritium and ¹⁴C from global fallout will not be dealt with as their present marine dose contributions are relatively low. Doses from ⁹⁰Sr and transuranic elements in seafood in 1990, are estimated in a similar way to those from ¹³⁷Cs (equations (1) and (2)). The committed collective doses from human intake of ⁹⁰Sr in fish and shellfish in 1990 are 5 and 0.7 man Sv, respectively, for ^{239,240}Pu, 11 and 22 man Sv and for ²⁴¹Am, 6 and 22 man Sv. These estimates are biased towards the high side because it has not been taken into account that the liquid discharges from Sellafield and the Chernobyl accident, which are the main sources responsible for the enhanced levels in the NE Atlantic (FAO area 27), had lower contents of ⁹⁰Sr and transuranic elements relative to ¹³⁷Cs than were observed in global fallout. If this factor is corrected for the above, dose estimates are reduced by approximately a factor of two. Hence the contribution from other anthropogenic radionuclides to the collective dose from marine food-chains in 1990 was in the order of 25% of the dose from ¹³⁷Cs.

Pentreath [11] has estimated that the dose from marine pathways by naturally occurring radionuclides other than ²¹⁰Po is one third of the dose from ²¹⁰Po, the main contributors to this dose being ²¹⁰Pb, ⁴⁰K, ⁸⁷Rb, ^{226,228}Ra, ^{235,238}U and ¹⁴C.

5.3. Comparison with Doses from the Terrestrial Environment

The doses received from 137 Cs via marine foods are in general lower than those received from terrestrial foods. This is particularly true if global fallout 137 Cs is considered. In 1964, when global 137 Cs levels in human diet peaked, less than 1% of the 137 Cs in total foods in Western Europe (Denmark) was derived from the marine environment (fish) [12]. In periods with a low input of fresh atmospheric fallout, the relative contribution of 137 Cs from the marine food-chains increases. If the terrestrial and the marine environments received the same deposition of 137 Cs per unit area, the dose commitment received by man from the marine food-chain will typically be 2 orders of magnitude less than that received from the terrestrial food-chain.

The global mean individual dose from 137 Cs in seafood in 1990 (0.03 µSv) corresponds to 7 minutes' effective dose from all natural sources (2.4 mSv per year) [10]. The corresponding dose from 210 Po in seafood (6 µSv) corresponds to about 1 day's effective dose from all natural sources. Regarding the NE Atlantic (FAO area 27), which has received most of the 137 Cs from Sellafield and Chernobyl, the individual mean dose received from 137 Cs in seafood from 1990 was one order of magnitude higher than the global mean dose, i.e. corresponding to about 1 hour's effective dose from natural sources.

5.4. Comparison with Doses from Global Fallout

The global fallout 137 Cs concentrations have been decreasing from 1966 to 1990 in the northern and southern hemispheres with an effective half-life of about 10 and 15 years, respectively. From 1961 to 1966 the concentrations of 137 Cs in surface seawater increased by a factor of 2 every 4 years [13]. It is assumed that this trend had persisted since 1952, when the first thermonuclear weapons tests began. The integrated water concentrations of 137 Cs from 1952 to 1966 in the northern hemisphere can be estimated as:

$$\int_{0}^{14} 17e^{-\frac{\ln 2}{4}t} dt = 89 \text{ Bq m}^{-3} \text{ year}$$

and in the southern hemisphere :

$$\frac{5}{17}$$
89 = 26 Bq m⁻³ year,

where surface seawater concentrations of respectively 17 Bq m^{-3} and 5 Bq m^{-3} were taken as 1966 representative values for the northern and southern hemispheres.

From 1966 to 1990 the integrated ¹³⁷Cs water concentrations in the northern hemisphere became:

$$\int_{0}^{24} 17e^{-\frac{\ln 2}{10}t} dt = 199 \text{ Bq m}^{-3} \text{ year}$$

and in the southern hemisphere:

$$\int_{0}^{24} 5e^{-\frac{\ln 2}{15}t} dt = 72 \text{ Bq m}^{-3} \text{ year.}$$

The annual fish catches in 1990 were $47 \cdot 10^9$ kg in the northern hemisphere and $22 \cdot 10^9$ kg in the southern hemisphere. The catches were lower in the periods prior to 1990. However, we have used the 1990 figures and have arbitrarily reduced the estimated dose for

1952 to 1990 by ~20% i.e. to ~6000 man Sv. The dose commitment from 1990 and onwards has been calculated to be 5750 man Sv (see §4.2), about half this dose being due to Sellafield and Chernobyl, i.e. the global fallout contribution becomes ~3000 man Sv. Hence the total commitment from global fallout becomes 9000 man Sv. According to UNSCEAR [14], this dose is about 1.3% of the total collective dose commitment from global fallout 137 Cs in human diet.

5.5. Future Studies

The actual global mean doses from anthropogenic radionuclides (e.g. ^{137}Cs) are very low and are not presenting any significant health hazard. The dose from ^{210}Po in marine foods is presently 2 to 3 orders of magnitude higher than that from ^{137}Cs . As discussed above, there is still a need for a better estimate of this dose to man and further studies of ^{210}Po in marine biota are therefore encouraged.

A number of lost nuclear submarines and probably also other nuclear devices (satellites, isotope batteries) reside on the sea-bed in the world's oceans. A better understanding of the long-term behaviour of, in particular, very long-lived radionuclides (e.g. transuranics) in the deep-ocean might be desirable in this context. Useful information may be obtained by studying the dumped nuclear reactors in the shallow waters of the Kara Sea east of Novaya Zemlya, radioactive wastes dumped in the Sea of Japan and the sunken nuclear submarine Komsomolets in the Norwegian Sea.

The calculation of future doses to man from marine food-chains depends on knowledge of the mean residence times of the radionuclides in the mixed layer of the ocean. In the North and South Atlantic, the half-life seems long for 137 Cs (100 years or longer) and somewhat shorter for plutonium (7-8 years) [3]. Future studies of marine radioactivity should follow the time trends of 90 Sr, 137 Cs and 239,240 Pu concentrations in the mixed layer of the different parts of the world's oceans in order to improve knowledge on mean residence times of these radionuclides.

6. CONCLUSIONS

The aim of the MARDOS project has been to assess the doses to the world population due to 137 Cs and 210 Po in marine food products in 1990. The collective dose commitment for 137 Cs is found to be 160 man Sv with an estimated overall uncertainty of 50%. The corresponding dose from 210 Po is 30 000 man Sv with an estimated uncertainty within a factor of 5. While the individual doses from 210 Po are assumed to be evenly distributed globally depending only on the amounts of marine products consumed, the individual doses from 137 Cs show a significant geographical variation. The highest doses were received by the populations eating marine food from the NE Atlantic Ocean, the FAO area No. 27. Approximately half of the global collective dose from 137 Cs in marine foods from 1990 was received from fish and shellfish produced in this area. The 137 Cs from Sellafield in the mean concentrations in the other ocean regions. Discharges of 137 Cs from Sellafield in the late seventies and early eighties and the deposition of 137 Cs from the Chernobyl accident in 1986 were the main reasons for the enhanced levels in the NE Atlantic. Higher concentrations were also observed in the Mediterranean (FAO area No. 37), these being primarily due to Chernobyl debris, coming mainly from the Black Sea.

The global collective dose commitments from 137 Cs in marine foods contaminated by liquid discharges from W. European civil nuclear sites until 1984 can be estimated to be approx. 3 000 man Sv and the corresponding dose commitment from the Chernobyl accident to be 2 000 man Sv [6]. The total collective dose commitment from 137 Cs in marine foods due to all nuclear weapons tests in the atmosphere can be estimated to be 9 000 man Sv. Hence the total dose commitment from marine-derived 137 Cs from these 3 sources is $1.4 \cdot 10^4$ man Sv, which corresponds to half of the dose received in one year from 210 Po in marine foods.

Data concerning ¹³⁷Cs in the oceans seem to be quite reliable. The behaviour of this radionuclide in the marine environment is well known, its concentration factors are well established, and predictions can easily be done about its fate in the case of accidental releases.

Data referring to 210 Po are much less abundant and concentration factors from the literature do not seem to be supported enough by field studies, at least in some cases. A wide range of 210 Po concentrations in the marine environment can be observed and the reason for such ranges of values is not yet sufficiently understood.

It should also be noticed that there are some other natural radionuclides, like 210 Pb and 226 Ra, which will also contribute to the population exposure. Although leading to doses significantly lower than 210 Po, they can still result in exposures more important than those due to 137 Cs. As referred to above, 210 Po was selected as representative of the natural radionuclides.

The doses to man from anthropogenic radionuclides in the marine environment are generally 1 to 2 orders of magnitude less than the doses from such radionuclides in the terrestrial environment. Compared with doses from natural radionuclides, the doses from anthropogenic radionuclides in the marine environment are insignificant.

Therefore, efforts should concentrate in getting a better knowledge of the behaviour of natural radionuclides in the marine environment, in particular ²¹⁰Po, as well as on metabolic models improving the reliability of the dose factors.

The results obtained in the framework of the MARDOS CRP provide the most complete data available to Member States on radionuclide levels in the marine environment and on doses to world population from marine radioactivity through ingestion of marine foods. The results will be used as the international reference source on the average radionuclide levels in the marine environment and corresponding collective committed effective doses from fish and shellfish.

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