

SOURCES OF RADIOACTIVITY IN THE MARINE ENVIRONMENT AND THEIR RELATIVE CONTRIBUTIONS TO OVERALL DOSE ASSESSMENT FROM MARINE RADIOACTIVITY - AN IAEA COORDINATED RESEARCH PROGRAMME

COLEÇÃO PTC

DEVOLVER AO BALCÃO DE EMPRESAS

MARDOS Collaboration

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ABSTRACT

A five year Coordinated Research Programme of the IAEA has been completed. It provides comprehensive information to Member States on levels of radioactivity in the marine environment and it estimates doses from marine radioactivity through ingestion of marine foodstuffs. Two radionuclides - natural ^{210}Po and anthropogenic ^{137}Cs - have been studied, as they are radiologically the most important representatives of each class of marine radioactivity. 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 which have been carried out in recent years. Collective doses and individual dose rates have been 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 except for the doses from ^{210}Po by consumption of shellfish. The collective effective dose commitment for ^{137}Cs in marine foodstuff in 1990 has been found to be 160 man Sv with an estimated uncertainty of 50 %. The corresponding dose from ^{210}Po is 30 000 man Sv with an estimated uncertainty of a factor of 5.

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 ^{137}Cs and ^{210}Po measurements in sea water and biota, characterizing FAO fishing regions.
- ii) To provide radiological 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 - natural ^{210}Po and anthropogenic ^{137}Cs - have been chosen, as they are from the radiological point of view the most important representatives of each class of marine radioactivity. Radioactivity levels of ^{210}Po and ^{137}Cs in sea water (Working Group I) and biota (fish and shell-fish) (Working group II) 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 literature data. Collective committed effective doses and mean individual doses from ^{210}Po and ^{137}Cs by consumption of fish and shell-fish have been calculated separately using the water and biota data (Working Group III).

The present report summarizes the results obtained in the framework of the CRP. A full report will be available soon in the form of an IAEA TECDOC report.

2 ^{137}Cs and ^{210}Po Concentrations in Water

The objective of this exercise was to determine sea water ^{137}Cs and ^{210}Po activities to which marine organisms were exposed in 1990 in each FAO fishing region. 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 shell-fish.

2.1 ^{137}Cs 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 ^{137}Cs distributions were reasonably homogenous, a mean of the existing data set was determined. In cases in which there was a stronger latitudinal gradient in ^{137}Cs distributions, and most of the recent data were confined to one area of the FAO region, the representative ^{137}Cs activity was inferred from the GEOSECS data set [1]. 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 ^{137}Cs distributions owing to oceanographic/geographic variability, a representative ^{137}Cs activity was determined by weighting the data according to the principal geographic focus of fishing activity. Data from regions in which the ^{137}Cs source function exhibits negligible variability were simply decay-corrected to 1990. These data were sufficiently recent so that they did not require the mixing correction applied to the GEOSECS data set. For regions in which the ^{137}Cs 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 /tritium ratios in the Atlantic Ocean were used to derive ^{137}Cs values at equivalent latitudes in the Indian Ocean and these were in good agreement with the few available direct observations.

Two regions (FAO 27 and 37) were sufficiently heterogeneous in terms of their ^{137}Cs distributions and fish catches to require special treatment. These regions were first divided into sub-regions for which a representative ^{137}Cs activity was determined and then a representative ^{137}Cs activity was chosen for each entire region with some weighting for the magnitude of fish catch in each sub-region [2].

An uncertainty of 25 % was recommended for the average ^{137}Cs concentrations in sea water.

2.2 ^{210}Po compilation

Sea water data have been compiled from the literature and using 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 ^{210}Po . Some marine regions, receiving higher inputs of ^{210}Pb to the surface ocean owing to higher rates of atmospheric ^{210}Pb inputs, have higher levels of ^{210}Po 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}

3 ^{137}Cs and ^{210}Po Concentrations in Biota

3.1 ^{137}Cs Compilation

The data reported by the CRP participants were reviewed and tabulated according to the FAO regions [2]. However, the ^{137}Cs data existing for the NE Atlantic and adjacent seas (27) were subdivided into Baltic, Irish Sea, North Sea, Barents Sea and general NE Atlantic. Also the data from area 37 were subdivided into Mediterranean and Black Sea. 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.

The data were classified into fish, mollusc and crustacean concentrations. A value for shell-fish was finally chosen for each region, as the differences for ^{137}Cs in molluscs and crustacea were not significant.

Concentrations in biota 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 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 the concentrations of ^{137}Cs in water and sea food. The following ranges of concentration factors (CFs) were found:

- 23 to 144 for fish, excluding three high values,
- 6 to 40 for molluscs, excluding one value,
- 5 to 52 for crustacea, excluding two values.

These values are quite in agreement with the concentration factors of 100 for fish and 30 for molluscs and crustacea, previously recommended by IAEA [3]. Values leading to exceedingly high CFs 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 ^{137}Cs , 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 in 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.

3.2 ^{210}Po Compilation

While for ^{137}Cs the data were compiled for each FAO region [2], a similar treatment for ^{210}Po 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 the other. The regional differences, if any, are below the fluctuations observed from one species to another or even from one specimen to another of the same species.

For the above reasons, the data for each group of seafood, from all regions of the world oceans, were combined and analysed together. Geometric means for the concentrations of ^{210}Po in each of the seafood groups were calculated both for the data reported by the CRP participants and from the literature survey. A global concentration for ^{210}Po in fish, molluscs and crustaceans was then calculated and used in the dose assessment.

4 Dose Assessment

4.1 Concentration Factors

The dose from consumption of marine food was 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 shell-fish (method 2). The concentration factors used were based on IAEA [3]. A value of 30 000 was chosen for shell-fish including both mollusks and crustaceans in agreement with the MARINA-MED project (Table 1).

The fish catch for different major fishing areas was calculated using FAO statistics for 1990 [2]. The factors used for the committed effective dose calculations for adults from intake of radionuclides were extracted from [5] and data used in the MARINA project [4], i.e. $1.2 \cdot 10^{-8} \text{ Sv Bq}^{-1}$ for ^{137}Cs and $4.3 \cdot 10^{-7} \text{ Sv Bq}^{-1}$ for ^{210}Po . Using an effective half-life of 70 days for ^{137}Cs in the human body, the first year dose from a one-time oral intake is 97 % of the committed effective dose.

4.2 Calculation of Doses

The doses were calculated using the following formulae;

$$D_{\text{Cs}}(\text{Fish}) = C_w 100 F_c \cdot 1.2 \cdot 10^{-8} F_h F_e = C_w F_c 4.2 \cdot 10^{-7} \text{ Sv Bq}^{-1} \quad (1)$$

and

$$D_{\text{Cs}}(\text{Shellfish}) = C_w 30 F_c \cdot 1.2 \cdot 10^{-8} F_h F_e = C_w F_c 1.8 \cdot 10^{-7} \text{ Sv Bq}^{-1}, \quad (2)$$

where D_{Cs} is the collective committed effective dose from ^{137}Cs by consumption of fish and shell-fish 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 shell-fish 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 ^{210}Po is 138 days. Statistics show that 30% is eaten fresh, 30% frozen, 20% smoked and 20% canned. The delay 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 ^{210}Po , but one half-life is applied in the calculations. The doses for polonium can accordingly be calculated as:

$$D_{\text{Po}}(\text{fish}) = C_w 2000 F_c 4.3 \cdot 10^{-7} F_h F_e D_f = C_w F_c 1.51 \cdot 10^{-4} \text{ Sv Bq}^{-1} \quad (3)$$

and

$$D_{\text{Po}}(\text{shell-fish}) = C_w 3 \cdot 10^4 F_c 4.3 \cdot 10^{-7} F_h F_e D_f = C_w F_c 3.23 \cdot 10^{-3} \text{ Sv Bq}^{-1} \quad (4)$$

The resulting collective effective dose commitment from fish and shell-fish caught during 1990 calculated using the two different methods are given in Table 2. The mean individual doses for a world population of $5.3 \cdot 10^9$ are shown within parentheses.

The two different methods give almost identical results except for the doses from ^{210}Po by consumption of shell-fish. Considering the possible errors in the different estimations, even this difference, by a factor 2.5, is acceptable. If the lower concentration factor, such as 10 000, had been used for crustacea, the dose from shell-fish would decrease from 38 000 to 30 000 man Sv.

The collective effective dose commitment from fish caught during 1990 calculated for FAO areas using the method 2 are shown in Fig. 1. The contribution of ^{137}Cs to the collective effective dose commitment from fish and shell-fish consumption is negligible, only 0.4% of that for ^{210}Po .

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

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

where $D(\infty)$ is the integrated dose from 1990 to infinity and D_i is the collective committed effective dose from consumption during 1990. The dose becomes 5 300 man Sv (or a mean individual dose commitment of $1 \mu\text{Sv}$ for a world population of $5.3 \cdot 10^9$) from consumption of fish and 450 man Sv (or a mean individual dose commitment of $0.08 \mu\text{Sv}$) from consumption of shell-fish. These figures can be compared with the doses of 10 000 man Sv received in one year by consumption of ^{210}Po in fish and 15 000 man Sv (using the biota value which is perhaps more precise) by consumption of shell-fish (or annual mean individual doses of 2 and $3 \mu\text{Sv}$, respectively).

The dose can also be calculated for a critical group (Area 27, NE Atlantic) tentatively consuming 100 kg of fish and 10 kg of shell-fish per year (1990). The annual dose from ^{137}Cs will be 3 μSv by consumption of fish and 0.1 μSv by consumption of shell-fish. The corresponding figures for ^{210}Po are 100 μSv for fish and 55 μSv for shell-fish.

5. Discussion

5.1 Reliability of the Assessment

The good agreement for ^{137}Cs between the two methods suggests that the ^{137}Cs concentrations in the marine environment are fairly well known. The major uncertainty of 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 ^{210}Po , the radioactivity measurements are less reliable than those for ^{137}Cs , particularly for biota. Furthermore, inhomogeneity in the internal distribution of ^{210}Po 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 [6,7], 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 fishermen's 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. 3], 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 % to the collective dose from ^{137}Cs and ^{210}Po in the marine environment.

The world ocean also contains other radiologically significant radionuclides besides ^{137}Cs and ^{210}Po . For example, from nuclear weapons testing in the atmosphere, ^{90}Sr , $^{239,240}\text{Pu}$, ^{241}Am , ^3H , 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:

$$^{90}\text{Sr}/^{137}\text{Cs} = 0.66 \text{ [ref. 8]}$$

$$^{239,240}\text{Pu}/^{137}\text{Cs} = 2\text{-}3 \cdot 10^{-3} \text{ [ref. 9]}$$

$$^{241}\text{Am}/^{137}\text{Cs} = \sim 1 \cdot 10^{-3} \text{ [ref. 8,9].}$$

Tritium and ^{14}C from global fallout will not be dealt with as their present marine dose contributions are relatively low. In order to estimate the doses from ^{90}Sr and transuranic elements in the marine food-chain in 1990, we may use the calculations for ^{137}Cs (equations (1) and (2)) and correct them by weighting for the appropriate water concentrations, dose factors and concentration factors for fish and shell-fish. The estimated collective doses from human intake of ^{90}Sr in fish and shell-fish in 1990 become 5 and 0.7 man Sv, respectively, for $^{239,240}\text{Pu}$, 11 and 22 man Sv and for ^{241}Am , 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 ^{90}Sr and transuranic elements relative to ^{137}Cs 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 ^{137}Cs .

Pentreath [10] has estimated that the dose from marine pathways by naturally occurring radionuclides other than ^{210}Po is one third of the dose from ^{210}Po , the main contributors to this dose being ^{210}Pb , ^{40}K , ^{87}Rb , $^{226,228}\text{Ra}$, $^{235,238}\text{U}$ and ^{14}C .

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) [11]. 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 ($0.03 \mu\text{Sv}$) corresponds to 7 minutes' effective dose from all natural sources (2.4 mSv per year) [8]. The corresponding dose from ^{210}Po in seafood ($5 \mu\text{Sv}$) corresponds to about 1 day's effective dose from all natural sources. In 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 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 ocean. 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 South Atlantic, the mean residence time seems long for ^{137}Cs and somewhat shorter for plutonium [9]. In the North East Atlantic, the mean residence times for ^{90}Sr and ^{137}Cs are relatively short [4]. Future studies of marine radioactivity should follow the time trends of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in the mixed layer of the different parts of the world ocean 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 of 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 doses from ^{137}Cs show a significant geographical variation. The highest doses were received by the populations eating fish 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 concentrations in the waters of the NE Atlantic, and thus also of the biota produced there, were 5 times higher than 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, initially deposited in 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. 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.

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 radioactive sources, the doses from anthropogenic radionuclides in the marine environment are insignificant.

7. References

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Table 1. Concentration factors for polonium and cesium to marine products.

Matrix	Cesium	Polonium
Fish	100	2 000
Shell-fish	30	30 000

Table 2. Collective effective dose commitment from fish and shell-fish caught in 1990. Average individual doses (μSv), within parentheses.

Matrix	^{137}Cs [man Sv]		^{210}Po [man Sv]	
	Method 1	Method 2	Method 1	Method 2
Fish	150 (0.03)	145 (0.03)	10 000 (2.0)	12 000 (2.3)
Shell-fish	11 (0.002)	14 (0.003)	38 000 (7.2)	15 000 (2.8)

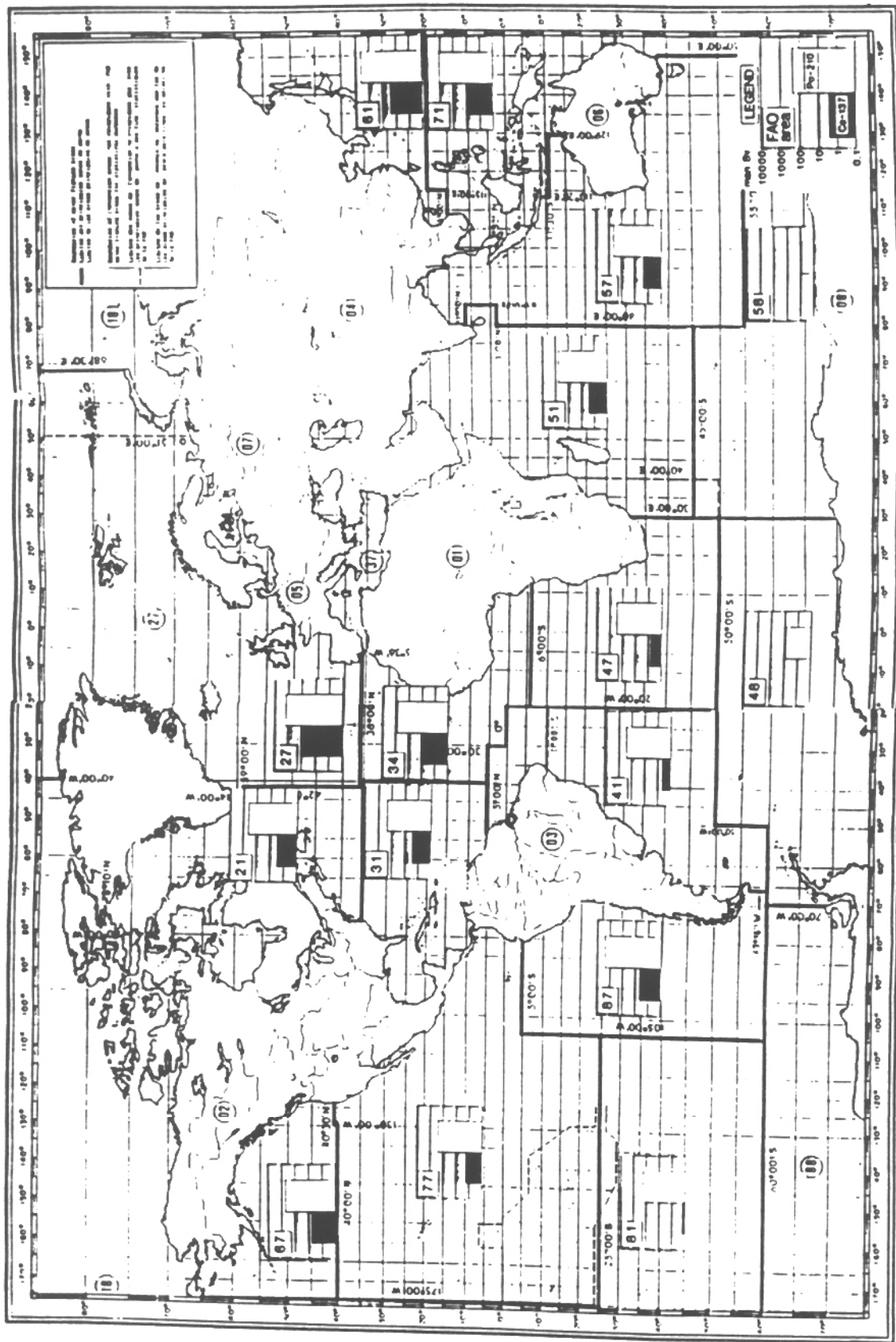


Figure 1 Collective effective dose commitment from fish caught in 1990 calculated for FAO areas using radioactivity data for biota.