STUDIES ON ENVIRONMENTAL RADIOACTIVITY IN BRAZIL - | RADIOPROTECTION

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ABSTRACT

This work presents the results of the continuous environmental survey programmes carried out by the Radioecological Laboratory of the Coast (LARELI) of the Agency for Coordination of Special Projects of the Sao Paulo State (COPESP). Artificial radionuclides in marine samples have been routinely monitored in the Laboratory's research monitoring programme since 1990. Some examples of analyses carried out in our laboratory are discussed mainly as large sample volumes are required. Marine samples (seawater, fish) are monthly collected from the Brazilian coast since Rio Grande do Sul State (latitude 32° 11'S) to Para State (latitude 00° 26'S) and the artificial radionuclide concentration was evaluated. Data are used to describe the radioecological situation along the Brazilian coast, to obtain radionuclide reference levels as well as to obtain the data for estimating the exposure of our population to fall-out radionuclides. This knowledge is necessary as a baseline for the detection of any future contamination. The main source of the radionuclides detected is due to the testing of nuclear devices in the atmosphere or accidents (like Chernobyl) occured in the northern hemisphere.

INTRODUCTION

The study of environmental contamination caused by radioactive nuclides is of great Scientific interest. Human and living organisms can be affected by both natural and artificial radiation. Radioactive substances have always been present in the environment.

There are more than 60 natural radionuclides, they include radionuclides of elements from Periodic Table like K-40, Rb-87, U-235, U-238, Th-232, the decay products of long lived Uranium and Thorium isotopes (such as U-234, Th-230, Ra-226, Pb-210, Po-210, Rn-220, Ru-222) and cosmogenic radionuclides formed through the interaction of cosmic rays with the atmosphere or with the Earth's crust (like H-3, C-14).

Radioactive material of technogenic origin, mainly fission products of uranium and plutonium were found in the atmosphere since 1945 after nuclear tests. Artificial radionuclides have been created in cyclotrons and reactors, as well as produced from nuclear weapons tests and from the operation of research and power reactors.

According to date from the National Council of Radiation Protection and Measurements (USA), the radon emission, formed through the decay of natural uranium, is responsible for about 55% of the annual mean dose of radiation received by the general population. Medical sources contribute 15%, internal irradiation from natural radionuclides (K-40 and U and Th radionuclides) 11%, cosmic rays 8% and the radioactivity of rocks and minerals 8%. The remaining 3% of this annual irradiation dose comes from professional irradiation and from global radioactive fallout as a result of the activities of the nuclear power in industry. [1]

ARTIFICIAL RADIONUCLIDES

Artificial Radionuclides Resulting from Nuclear Weapons Tests:

Between 1945 and 1980, atmospheric nuclear weapons tests carried out by different countries totaled 423. Most of them was conducted between 1955 and 1958 and 1961 and 1962. There has no testing since 1980. Artificial radionuclides produced by tests include C-14, Cs-137, H-3, Sr-90 as well as plutonium and other transuranic elements.

The radioactive debris generated by the tests has been delivered to the oceans from the troposphere and the stratosphere in both northern and southern hemisphere.

The estimates of input were 6.1×10^2 (Cs-137), 8.2×10^2 (Pu-239+240) and 3.7×10^2 Pbq for Sr-90. For the southern hemisphere the values have been half of those in the northern hemisphere. [2]

ARTIFICIAL RADIONUCLIDES RESULTING FROM NUCLEAR POWER FACILITIES

Sources of artificial Radionuclides produced in nuclear power facilities include the reactor, fuel reprocessing plants, the radioactive waste storage and operational discharges of effluents. There are anthorized routine releases of low level radioactive wastes to the environmet from the facilities mentioned above.

The discharges of effluents reach the oceans through the atmosphere, rivers and by direct releases to the marine environment. Discharges of radioactive effluents have been made into European waters since 1952 and according to the United Nations Committee on the Effects of Atomic Radiation (UNSCEAR Report) discharges from Sellafield, U.K., were of the 0.38 PBq in 1984-85, from Cap de La Hague, France, the Cs-137 releases were 0.027 PBq in 1983-85.

DUMPING OF LOW-LEVEL PACKAGED WASTES

Packaged radioactive waste has been disposed of into European waters since 1948.

Dumping operation was interrupted in 1982 as a result of the London Dumping Convention. The total amount of material dumped at sea was of about 60 PBq.

NUCLEAR ACCIDENTS

Four major nuclear accidents with radionuclide releases to the environment occured in Sellafield (Windscale, U.K, 1957), Chelyabinsk (USSR, 1957), Three Mile Island (USA,1979) and Chernobyl (USSR, 1986). A few accidents with nuclear powered satellites and nuclear submarines have also occurred.

In the Windscale accident, the principal route for contamination was radiodine in milk while at Chelyabinsk exposure resulted from Sr-90 contaminated milk. At Three Mile Island most of the realease consisted of Xe-133. At the Chernobyl accident the cesium-137 deposition in the oceans was of 4.7 PBq out of the total of 70 PBq released.

ATOMIC POWER GENERATING UNITS

At the present, nuclear power plants make an essential contribution to the global production of electricity. According to data from the IAEA, in 1987, 417 atomic power generating units were operating in 26 countries. Today, France is the second world producer of nuclear generated electricity (56 nuclear reactors) after the United States and ahead of the USSR, Japan and Germany. In 1991, generated nuclear electricity represented nearly 73% of French electricity production. France has a leading position, ahead of Belgium (59%), Sweden (52%), South Korea (47,5%), Hungary (46%), Swit-Zerland (42%), Taiwan and Spain (39%), Bulgaria (35,5%), Germany (32%) and Finland (29%).

PATHWAYS LEADING TO HUMAN EXPOSURE

Radionuclides in the sea may contribute to

exposure of organisms, including man, through several pathways. The main pathway is the consumption of various types of seafoods, mainly the fish consumption. For marine organisms interest is primarily in population effects such as survival, growth and reproductive performance.

In regard to internal exposure from ingestion of food and water and to the contamination of environmental materials, the most important radionuclides to be assessed are Cs-137, I-131, Sr-90, Pu-239+240 and Am-241.

According to the Gesamp Report, internationally recommented approaches to authorized radionuclides disposal to the sea from man's activities have efficiently restricted marine contamination to limited areas and to low concentrations.

Individual doses via ocean pathways from radionuclide releases are low compared to the dose limit 1 m SVy^{-1} defined by ICRP for members of the public.

Radiation doses arising from reprocessing plant releases are about one tenth of the annual average dose from natural background. However, in some case as Sellafield the values are higher.

Some releases such as from the Sellafield or accidents like Chernobyl have been studied, as the radionuclides released offer an opportunity to trace water and particulate movements.

ARTIFICIAL RADIONUCLIDE CONCENTRATION IN MARINE ENVIRONMENT

According to the Gesamp Report, the Cs-137 contribution originating from nuclear weapons tests was only a few $Bq.m^{-3}$ in the Atlantic surface water. In the Mediterranean, the values vary from about zero near the botton to up to 5 to 10 $Bq.m^{-3}$ at the surface. In some closed shallow brackkish-water sea areas, sush as the Baltic Sea, the concentrations of Sr-90 and Cs-137 reached 20-40 Bq.m , and those of plutonium 0.002-0.004 $Bq.m^{-3}$.

In the North Atlantic and Artic Oceans the concentration of Pu was around 0.013 $Bq.m^{-3}$ (1980). Levels of plutonium up to 0.026 $Bq.m^{-3}$ were obseved in waters near south of Norway, due to Sellafield. In 1970 in the Surface water of the Pacific Ocean, concentrations of plutonium ranged from 27 to 37 $mBq.m^{-3}$.

Significant quantities of Pu have been accumulated in ocean sediment. In the North East Atlantic Sediments were found 0.015 to 1.3 $mBq.q^{-1}$ of Pu. In Sediments of the Baltic Sea 100 $mBq.g^{-1}$ for cesium-137 and 5 $mBq.g^{-1}$ for Plutonium have been measured.

Typical values for Cs-137 in deep sea fish muscle are about of 0.1 to 0.5 $Bq.kq^{-1}$ and for Pu are 10 to 10 $Bq.Kq^{-1}$. In the Baltic Sea these values vary from 1 to 4 $Bq.Kg^{-1}$.

THE RESEARCH PROGRAMME OF THE LARELI

Considering the environmental pollution problems and their impact on man, the LARELI has developed a research programme on environmental radioactivity studies. [3,4]

The objetives of the continuous environmental survey programmes carried out by the LARELI are to determine the quantity of radioactive substances accumulated in the environment as well as to estimate the exposure dose. So, proper sampling procedures and radiochemical methods were developed for radionuclide analysis in aerosol, sediment, seawater and marine products (Such as Seaweeds, fish, phytoplancton). Total gama activity, total beta activity, Cs-137, Sr-90, Po-210 and Pu-239+240 can be measured.

Data are used to describe the radiological situation along the Brazilian Coast, to obtain radionuclide reference levels as well as to obtain the necessary data for estimating the exposure of our population to fall-out radionuclides.

The LARELI includes laboratory rooms, storage

rooms, counting equipments and all the facilities necessary to provide a broad range of radionuclide analysis support services.

The monitoring programme relative to collecting samples is already implemented and montly we have received marine samples from differents points of the Brazilian Coast, from Rio Grande do Sul State (latitude 00° 26'S) to Para State (latitude 32° 11'S), one total of about 7000 Km being monitored.

SEAWATER SAMPLE ANALYSIS

Surface Seawater samples are being monthly collected at the fixed stations (See Figure 1) and cesium was preconcentrated from 100 liters of sample using ammonium phosphomolybdate (AMP), previously syntethized in our laboratory. Cesium-137 was assayed by gamma spectrometry.

The strontium was preconcentrated using sodium carbonate, purified by both iron hydroxide scavenger and sulphate precipitation and stored for 14 days to reach the radioactive equilibrium. Yttrium-90 was separated as hydroxide from strontium-90, converted to oxalate and counted in a low background Geiger Muller detector.

FISH ANALYSIS

For fish analysis, the edible part (2Kg) was weighed and dried at 110° C for one week and later on the temperature was gradually increased up to 450 C until ashes were obtained. Ashes were counted in polyethylene pots in a hyperpure Ge detector to determine Cs-137 activity. Further, in order to determine Sr-90 they were submitted to wet digestion and to the same procedure employed for Sr-90 analysis in seawater.

The main fish consumed by our population, such as sardine, ballistes, weak fish, dog fish, mullet, saw fish, red, mackerel scad, brazilian croakar and white

grunt were analysed. Fish samples are purchased fromfishermen's cooperative associations, that know the exact points of collecting.

ESTIMATING DOSE RATES

In order to estimate dose rate from cesium-137, it was necessary to obtain data relative to catch of fish, fish consumption by local population and the average cesium concentration for fish caught. The area chosen was the southern coast of the Sao Paulo State (23° 52'S LAT., 45° 50'W LONG.), with a population of 18 million inhabitants. This rate of population includes regions, from Santos until Rio de Janeiro. Table 1 gives the estimations on sea foodhabits in the studied area. The average consumption of sea fish in the area is 7.8 Kg per year, based on statistics of the total population.

The quantity of sardine caught in the Santos region are of 87,000 ton, being that 80% are consumed as fresh fish.In the case of ballistes sp, the total fish caught are of 35,000 ton and 65% are consumed. The other part is employed for the preparaton of animal diets.

The total catch of weak fish is of 33,000 ton and 85% are consumed. The quantity of corvine and dog fish caught are of 17,000 ton (70% of consumption) and 9,000 ton (90% of consumption) respectively.

Based on the the cesium-137 radioactivity levels and by taking into consideration the dose conversion factor recomended by ICRP, we can estimate the annual effective dose equivalent as being of the 0.013 uSv.a^{-1}. This value shows that the dose via ocean patways from cesium-137 is very low compared to the dose limit (1 mSv.a^{-1}) recommended by ICRP to protect members of the public. The above estimation gives a rough idea of the present exposure levels in the coastal region of the Sao Paulo State.

RESULTS

Cesium-137 levels in seawater and fish samples found at each local are shown in the Tables 2 and 3. This radioactivity present in marine samples correspond to typical values due to fallout deposition in the southern hemisphere.

Cesium-137 levels in seawater range from 0.5 to 2.2 Bq.m^{-3}, and in fish vary from 0.1 to 0.4 Bq.Kg^{-1}. Cesium-137 has been monitored by most member states of the European Communities and the Gesamp Report [2] gathered data relative to the levels of this radionuclide. Cesium-137 measurements in seawater and fish over the period 1980 to 1985 were performed. The cesium-137 levels in Portuguese waters are low (<20 Bq.m^{-3}) and include a significant contribution due to the fallout from nuclear weapons tests. Areas like northwest coast of Scotland and northern Ireland are influenced by input from Sellafield (cesium-137 levels 100-200 Bq.m^{-3}) and the Irish Sea contains the highest concentrations determined (>200 Bq.m^{-3}).

Cesium-137 concentrations in fish range from about 0.3 to 3.0 Bq.Kq^{-1} (wet) with the higher levels being found in the Baltic Sea.

Bettencourt et al [5] performed cesium-137 measurements in fish collected in Portuguese oceanic waters. From the artificial radionuclides analysed, Cs-137 is the most significant contributor for the dose to man through fish consumption. The cesium-137 concentrations in fish muscle range from 0.1 to 1.7 Bq.Kg^{-1}.

In this work, cesium-137 levels obtained for marine samples are in agreement to the published values in the literature and they can be considered reference levels to our country. Any increase in these levels could be attributed to some possible future contamination.

Today, nuclear safety is a necessary condition to make possible the pratical applications of nuclear technology in the industry, medicine and the power generation.

The nuclear accidents at Three Mile Island and the Chernobyl highlighted the needs for nuclear safety and for environmental consideration. Having in mind this ecological concern, the LARELI has infrastructure and capability to offer to the community laboratory services in order to carry out a monitoring programme, to evaluate the influence of unexpected releases of radioactive materials and to verify that the public dose equivalent of environmental artificial radiations is lower than the limit of annual dose equivalent.

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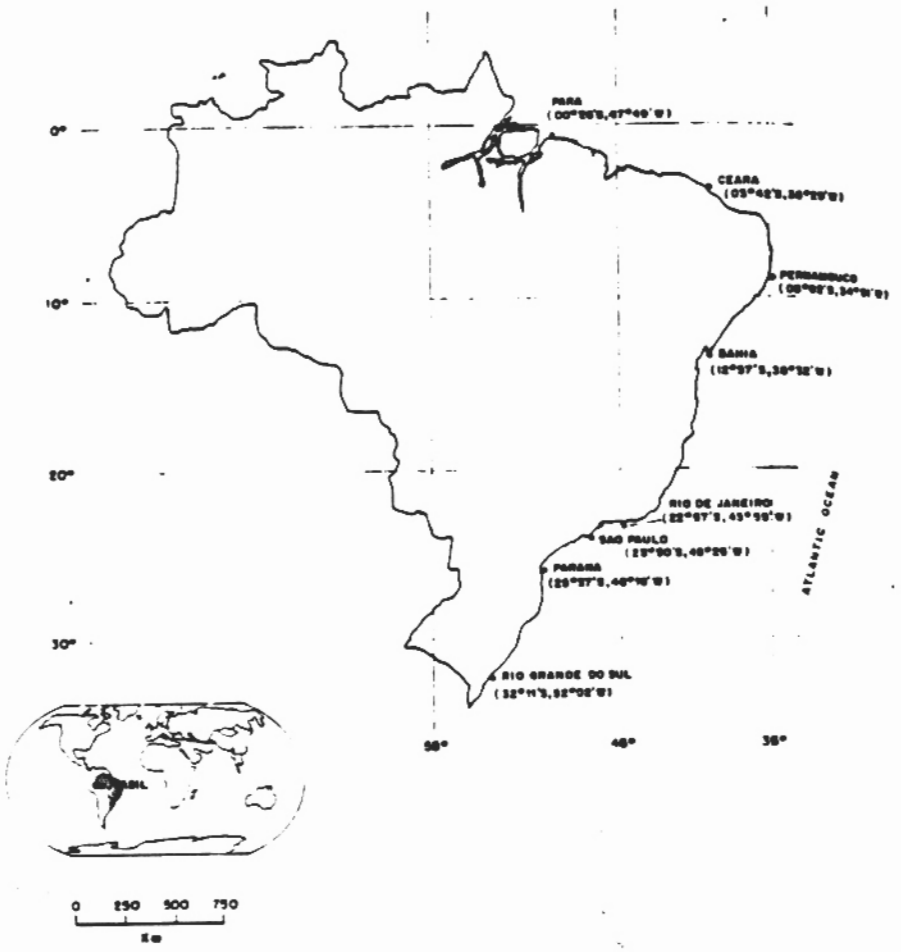

Fig.1 - Collecting Points of Marine Samples.

TABLE 1. THE ESTIMATION ON SEA FOOD HABITS

Fish	Level of cesium 137 (Bq.Kg^{-1} edible part)	Quantity of fish caught(t)	Porcentage of fish consumed	Consumption of fish per year (Kg. man^{-1} a^{-1})
Sardine (Sardineella brasiliences)	0.1	87.000	80%	3.87
Ballistes sp	0.1	35.000	65%	1.26
Weak-Fish (Cynoscion Jamaicensis)	0.1	33.000	85%	1.56
Corvine (Micropogonia furnieri)	0.3	17.000	78%	0.66
Dog Fish	0.3	9.000	90%	0.45

TABLE 2 - CESIUM-137 LEVELS IN SEAWATER (Bq.m^{-3})

STATE	LAT	LONG	4ºBim/91	1ºBim/92	2ºBim/92	3ºBim/92	4ºBim/92
Rio G. Sul	32°11'S	52°02'W	1.5	---	1.2	1.2	0.7
Parana	25°37'S	48°16'W	1.5	1.7	1.0	1.3	0.9
Sao Paulo	23°00'S	44,5° W	1.7	1.5	1.5	0.9	1.2
Rio de Janeiro	22°57'S	43°55'W	1.5	1.2	---	---	0.7
Bahia	12°57'S	38°32'W	1.6	1.0	1.5	1.6	0.7
Pernambuco	08°02'S	34°51'W	1.8	1.2	1.1	1.6	0.7
Ceara	03°42'S	38°29'W	2.1	2.2	1.1	1.7	1.3
Para	00°26'S	47°49'W	0.7	0.6	0.5	0.8	1.3

TABLE 3 - CESIUM-137 LEVELS IN FISH

STATE	(Bq.Kg^{-1} Edible Part)		
Rio G. Sul	Mackerel Scad (cavalinha) 0.11	Weak Fish (pescada) 0.04-0.19	Brazilian Croakar (curvina) 0.15
Parana	Weak Fish 0.11	Sardine 0.18	Mullet (pescada) 0.10
Sao Paulo	Sardine 0.10	Ballistes 0.10	Weak Fish 0.10
	Brazilian Croakar 0.30	Dog Fish 0.30	
Rio de Janeiro	Mullet (Tainha) 0.07-0.15	Sardine 0.15	
Bahia	RED (Vermelho) 0.12	Mackerel Scad (Xixarro) 0.12	
Pernambuco	Mackerel Scad 0.11-0.22	Saw Fish (Serra) 0.28	Garajuba 0.39
Ceara	White Grunt (Biquara) 0.10		
Para	Mullet 0.11-0.16	Brazilian Croakar 0.21	White Mullet (Pratiqueira) 0.01