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ASSESSMENT OF THE STATE OF POLLUTION

IN THE MEDITERRANEAN SEA BY RADIOACTIVE SUBSTANCES

In cooperation with

IAEA

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TABLE OF CONTENTS

BACKGROUND

The protocol for the Protection of the Mediterranean Sea against pollution from Land-based Sources (LBS Protocol) was adopted by the Conference of Plenipotentiaries of the Coastal States of the Mediterranean Region on 17 May 1980 in Athens, Greece. The protocol has so far been ratified by 17 Contracting Parties to the Barcelona Convention and entered into force on 17 June 1983.

Article 5 of the LBS Protocol stipulates that:

- The parties undertake to eliminate pollution of the Protocol Area from land-based sources by the substances listed in Annex 1 to this protocol.

- To this end they shall elaborate and implement, jointly or individually, as appropriate, the necessary programmes and measures.

- These programmes and measures shall include, in particular, common emission standards and standards for use.

- The standards and the time-tables for the implementation of the programmes and measures aimed at eliminating pollution from land-based sources shall be fixed by the Parties and periodically reviewed, if necessary every two years, for each of the substances listed in Annex I in accordance with the provisions of article 15 of this protocol.

Annex I of the protocol lists radioactive substances, including their wastes.

At the Fourth Ordinary meeting (Genoa, September 1985), the Contracting Parties to the Convention for the Protection of the Mediterranean Sea against Pollution and its related Protocols agreed that, with regard to the technical implementation of the Protocol for the Protection of the Mediterranean Sea against Pollution from Land-based Sources, the Secretariat would propose an order of priority and a realistic time-table for the development of programmes and measures for at least two substances annually, including common emission standards and standards for use, required for the implementation of the Protocol, and that in preparing such a proposal, substances listed in the Annex I to the Protocol should be accorded priority.

A meeting of experts on the technical implementation of the LBS Protocol (December 1985) recommended that an "assessment document" on the state of pollution in the Mediterranean Sea should be prepared for each of the substances listed in Annexes I and II of the LBS protocol.

The Contracting Parties which approved the recommendation of the above meeting gave the guidelines for preparing an "assessment document". The guidelines state that such assessment documents should include inter alia chapters on:

- sources, point of entries and amounts of pollution from industrial, municipal and other discharges to the Mediterranean Sea

- levels of pollution

- effects of pollution

- present legal, administrative and technical measures at national and international levels.

In compliance with the above the present document on the assessment of the state of pollution of the Mediterranean Sea by artificial radionuclides was prepared by the Secretariat with the help of consultant and in close cooperation with international organizations, especially with IAEA. This document does not deal with all radionuclides, but concentrates on dealing with the artificial ones, since possible environmental events considered as "pollution" are only attributable to some of these radionuclides.

1. Introduction

The acute public concern over the effects on human well-being of ionizing radiation resulting from the existence of radioactive substances in the environment has only arisen in the recent years, despite the fact that natural radioactive substances had already existed at the beginning of the universe and, since then, have always been present in the environment. This concern is certainly owing to recent recognition by the public of increasing anthropogenic releases of artificial radionuclides into the environment by developing nuclear industries, by large-scale nuclear weapon testing and, especially, by spectacular nuclear accidents, such as those of Three Mile Island, Chernobyl, etc. For the scientific assessment of the effects of environmental pollution with artificial radionuclides, however, one should always bear in mind that life occurring on the earth has been consistently and constantly under the influence of ionizing radiation from naturally occurring radionuclides. The radiological effects of ionizing radiation on life are identical whether they come from natural or artificial radionuclides. Although the introduction of artificial radionuclides into the environment has been a recent event, its scientific significance is entirely different from that of some types of toxic organic pollutants, which had never been present in the environment before their introduction.

The present document intends to outline the actual state of pollution of the Mediterranean Sea by artificial radionuclides, to provide a scientific rationale for establishing control measures for preventing radioactive pollution to be adopted by the Contracting Parties.

In order to facilitate general understanding of the problems, a brief scientific outline on characteristics of radionuclides and comparisons between natural and artificial radionuclides are given prior to the description of the situation of contamination by artificial radionuclides in the Mediterranean Sea.

Chapter I, which deals with the assessment of the state of pollution in the Mediterranean Sea, provides information on the sources and inputs of artificial radionuclides, describes biogeochemical processes by which various released radionuclides are redistributed into different marine environmental compartments and presents a summary of available data on levels of artificial radionuclides in sea water, sediments and biota in the pre-Chernobyl and post-Chernobyl periods. The chapter also provides information on the effects of radiation exposure on marine organisms as well as man, assessing risks of such exposure.

Chapter II, which deals with control measures for preventing pollution effects, reviews available information on existing national and international control measures and outlines the scientific rationale for establishing these control measures.

2. General Characteristics of Radionuclides

Since radionuclides (radioisotopes) are chemically identical with stable elements, their biogeochemical behaviour in the marine environment is similar to that of stable elements and depends on their chemical properties. They undergo various environmental processes taking place in situ, such as dissolution, precipitation, sorption, complexation, biological ingestion and excretion, etc. in the manner similar to those of stable elements. This is especially true for natural radionuclides coexisting with their stable counterparts in the environment. For example, radioactive potassium-40 (K-40) behaves in situ exactly like stable potassium-39 (K-39, natural isotopic abundance, . 93%) and potassium-41 (K-41, natural isotopic abundance, . 7%), although its isotopic abundance amounts to only 0.01%. Similarly, artificial radionuclides, such as cobalt-60 (Co-60), cesium-137 (Cs-137), etc. which have been introduced into the marine environment as the

result of human activities, behave similarly to their natural occurring stable counterparts, cobalt-59 (Co-59, natural isotopic abundance, 100%), cesium-133 (Cs-133, natural isotopic abundance, 100%), etc., provided that both the chemical and physical forms of the artificial radionuclides introduced are identical with those of their stable counterparts.

Atoms of radionuclides are, however, characterized by the instability of their nuclei. This instability causes a radioactive atom to decay to another atom, either radioactive or stable, with a probability specific to the initial atom. In other words, the probability of decay of a radionuclide within a given time depends on the radionuclide involved. The half-time of the decay (which follows the exponential law) is a constant for that specific radionuclide regardless of the amount of radionuclides present. This half-time of decay, traditionally known as a "half-life", is considered a primary physical property characterizing each radionuclide. This specific time-dependency of the decay of radionuclides could be used not only for ensuring the identification of certain radionuclides during their measurements, but also for applying them as unique tools for determining rate constants of various environmental events, such as, the time-scale of geochemical processes. The latter use of natural as well as artificial radionuclides has been developed for studying various environmental processes, proving the usefulness of some radionuclides as "geochemical tracers".

In addition to the time-specificity, each radionuclide decays in its own specific scheme, emitting spontaneous particles and/or radiation with characteristic energies. This is another important characteristic of radionuclides which facilitates their identification in environmental materials with certainty. Due to the decay-scheme and energy specificity, aided sometimes by the time specificity of the decay described above, most radionuclides can be identified and quantitatively determined at very low levels often found in complex environmental matrices. The accuracy of radiological measurements suffer, in general, much less from reagent blanks and procedural contamination inherent in all trace measurements but especially critical for stable trace element measurements. In addition, high sensitivity and accuracy in radiological measurements can be attained by relatively modest laboratory facilities and instrumentation. For example, the combination of radiochemical separation procedures and successive á-spectrometry achieves the measurement of concentrations of transuranic elements as low as 10^{-20}-10^{-22} gram of the elements per 1 gram of environmental matrices with associated errors within ± 20% or less (Fukai et al., 1976; Holm et al., 1980). It is also possible, with the recent developments in computerized gamma-spectrometry, to conduct continuous measurements and control of gamma-emitting radionuclides, for example, in radioactive waste effluents being released from nuclear installations.

The sensitivity, certainty and ease of radionuclide measurements, which are based on the physical characteristics of radionuclides described above, are important aspects to be taken into account in considering environmental and effluent monitoring as well as control measures for preventing radioactive pollution in the environment.

3. Natural and Artificial Radionuclides

3.1. Natural radionuclides

Although the general scope of the present document is to deal with the state of pollution of the Mediterranean Sea derived from the introduction of artificial radionuclides by human activities, it is not possible to completely ignore the presence of and environmental role played by naturally occurring radionuclides in the sea since, as it has already been pointed out, the life existing on earth has always been exposed to their radiation.

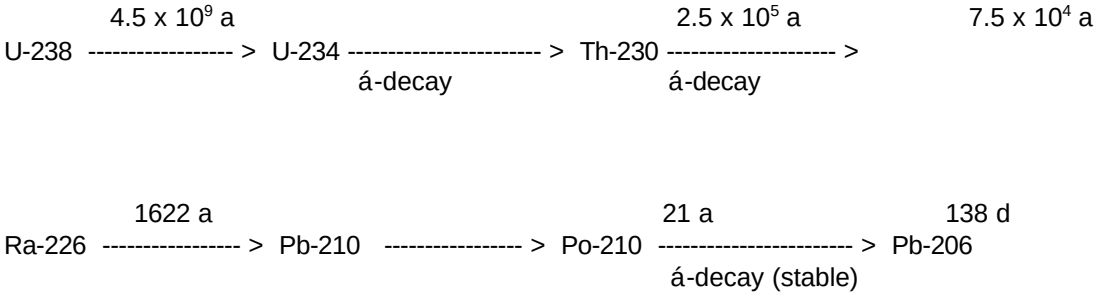
More than 60 radionuclides are known to occur naturally in the environment. These are classified into two groups according to their origin: terrigenous and cosmogenic.

3.1.1. Terrigenous radionuclides: The terrigenous radionuclides are believed to have been present already in the rocks and minerals of the earth's crust when the earth was formed and include long-lived primordial nuclides co-existing with their stable element counterparts as well as the three primordial actinide parent nuclides, thorium-232 (Th-232), uranium-235 (U-235) and uranium-238 (U-238) and their descendant products.

At least 14 radionuclides are known to occur in nature at present as primordial nuclides coexisting with stable element counterparts. The common characteristic of these radionuclides is their very long half-lives, which range from 10^7 years to 10^{15} years. Although their chemical properties vary widely, all of these radionuclides are considered to be well exchanged isotopically with their stable isotopes in the environment due to primordial occurrence at the formation of the earth. Thus the environmental behaviour of these radionuclides is similar to that of their stable counterparts.

The principal members in the above category are potassium-40 (K-40) and rubidium-87 (Rb-87). Both radionuclides belong to the alkali metal group, so that they are dispersed widely over the whole environmental spheres, especially in the hydrosphere, due to their susceptibility to weathering. The radioactivity of K-40 thus represents over 90% of the total radioactivity of sea water, while that of Rb-87 corresponds with approximately 1%.

As previously stated, another group of the primordial terrigenous radionuclides includes three actinide parents and their daughter nuclides which consist of about 35 radioisotopes of Pb, Bi, Po, Rn, Ra, Ac, Th, Pa, and U. Depending on their parent's mass numbers, these isotopes form three independent decay series. Among these decay series, the uranium decay series, characterized by mass numbers of 4n+2 (where n represents an arbitrary integral number), includes a series of more familiar radionuclides encountered in the marine environment. Principal members of the uranium decay series (half-lives longer than 100 days) are schematically presented below:



where solid arrows represent the direct decay from a parent to its daughter; broken arrows, indirect decay from a parent through short-lived intermediate nuclides to its descendant; numbers above the arrows, half-lives of parent nuclides, and signs below the arrows, types of decay.

In a closed system of sufficient age in the environment a descendant radionuclide in a given decay series exists in a steady state equilibrium, since the radioactive decay law dictates that the rate of production of the descendant from its immediate parent is equal to the rate of decay of the descendant itself to its daughter. In the dynamic environment such as the hydrosphere, however, the closed system is often disrupted by the separation of a daughter nuclide from its immediate parent, as the daughter may be quite different chemically from its parent. This causes radiological disequilibrium between the parent nuclides and its daughter. Since the restoration of the steady state in the original system, if it is reclosed, takes approximately six half-lives of the daughter, the degree of the disequilibrium observed for the original system indicates a measure of the length of time lapsed between the reclosure of the system and the observation. Depending on the circumstances, several

environmental processes are known to cause isotopic disequilibria between parent and daughter nuclides in the actinide decay series. Thus, the observations on the degree of the isotopic disequilibria in the actinide decay series in some environmental systems may bring useful information on time-scales with which various biogeochemical processes are taking place in situ.

In connection with the isotopic disequilibria for the uranium decay series encountered in the marine environment, it is important to note that some marine organisms are receiving much higher natural radiation doses than organisms living on land, due to the extensive accumulation of polonium-210 (Po-210), the descendant nuclide of U-238, in some organs of their bodies (Cherry & Shannon, 1974; Cherry & Heyraud, 1981). In fact, it has been observed that radiation doses due to Po-210 received by several marine organisms are much higher than those due to artificial radionuclides in significantly contaminated areas around some nuclear installations (Pentreath et al., 1980).

3.1.2. Cosmogenic radionuclides: At least 14 radionuclides are known to be continually produced in the earth's atmosphere by nuclear reactions between gaseous atoms of the earth's envelope (hydrogen, oxygen, nitrogen, argon, etc.) and cosmic-ray particles, such as high-energy protons originating from the outer space. The fluctuations in the rate of production of these radionuclides have been estimated from the measurements on meteorites, etc. and are found to be within a factor of about two, if the average values over periods of their half-lives are considered.

These radionuclides are brought down to the earth's surface by precipitation and dry fallout or they enter directly into geochemical processes taking place on and/or above the earth's surface in a gaseous phase. Although stable isotopes of all known cosmogenic radionuclides exist in nature, the degree of the isotopic exchange between cosmogenic radionuclides and their corresponding stable isotopes depends on the forms in which they are introduced into the geochemical processes.

The cosmogenic radionuclides include hydrogen-3 (tritium, H-3) and carbon-14 (C-14) as their principal members. The naturally occurring global inventories of H-3 and C-14 are estimated to be respectively 1.3×10^{18} Bq and 11×10^{18} Bq (Lal & Peters, 1967). In addition to these inventories, excess amounts of these two radionuclides have been produced artificially by a number of explosion tests conducted mainly during the 1960's and introduced into the earth's environment. These amounts are estimated to be 1.7×10^{20} Bq for H-3 (Schell, 1974; Michel, 1976) and 0.2×10^{18} Bq for C-14. Thus, it has become necessary to take into account the presence of the artificially injected H-3 and C-14 in the environment, when they are to be used as tracers for various environmental processes, such as water mixing, sedimentation, etc.

3.1.3. Environmental abundance of natural radionuclides: In order to obtain a general picture of the relative abundance of natural radionuclides in the marine environment, the average mass and radioactivity concentrations of principal members of natural radionuclides in sea water and marine sediments are compared with those in the earth's crust in Table I. The table is based on the compilation made by Fukai & Yokoyama (1982). In the compilation the heterogeneity existing within a given geochemical sphere has been disregarded by presenting average concentrations for the geochemical sphere. Therefore, it should be borne in mind, when one refers to the numbers in Table I, that the variability of the concentrations may be quite large within a given geochemical sphere, especially for the heterogenous lithosphere. Nevertheless, the table gives useful information on the levels of natural radionuclides in the marine environment in relation to the concentrations of artificial radionuclides released into the sea as the result of human activities.

Table I

Average mass and radioactivity concentrations of natural radionuclides in the marine environment and the earth's crust

Radionuclide	Half-life (years)	Marine environment				Continental Earth's Crust	
		Sea water		Sediment			
		($\mu g l^{-1}$)	($mBq l^{-1}$)	($\mu g kg^{-1}$)	($mBq kg^{-1}$)	($\mu g kg^{-1}$)	($mBq kg^{-1}$)
<u>Terrigenous Radionuclides</u>							
Potassium-40	1.25×10^{9}	48	1.26×10^{4}	3000	7×10^{5}	2400	6.3×10^{5}
Rubidium-87	4.7×10^{10}	34	110	-	-	2.4×10^{4}	8.1×10^{4}
<u>Uranium Decay Series</u>							
Lead-210	21	6×10^{-10}	1.9	5×10^{-5}	1.5×10^{5}	1×10^{-5}	3.3×10^{4}
Polonium-210	0.38	7×10^{-12}	1.1	9×10^{-7}	"	2×10^{-7}	"
Radium-226	1662	1×10^{-7}	3.7	0.004	"	0.001	"
Thorium-230	7.52×10^{4}	2×10^{-8}	0.015	0.2	"	0.04	"
Uranium-234	2.48×10^{5}	2×10^{-4}	44	0.08	1.9×10^{4}	0.15	"
Uranium-238	4.5×10^{9}	3	37	1000	1.1×10^{4}	110	"
<u>Cosmogenic Radionuclides</u>							
Hydrogen-3	12.3	1.7×10^{-12}	0.6	-	-	-	-
Carbon-14	5730	2×10^{-8}	3.7	$(0.1\text{-}1) \times 10^{-4}$	0.0015-0.015	-	-

3.2. Artificial radionuclides

Several hundreds of artificial radionuclides are produced intentionally or unintentionally as the result of human activities, such as the applications of nuclear reactors and particle accelerators to various purposes, testing of nuclear weapons and nuclear accidents. Many of these radionuclides are short-lived and decay quickly after their production. Some of the artificial radionuclides are, however, longer-lived and released into the environment under controlled or uncontrolled conditions. Since radioactive waste effluents from nuclear fuel reprocessing and power production are treated in order to reduce the levels of radionuclides contained after certain periods of storage, the quantities of artificial radionuclides in the effluents released into the environment are low and the numbers of the radionuclides contained are limited. As there is no control of environmental releases of artificial radionuclides during weapon testing and accidents, the quantities and number of artificial radionuclides released into the environment are much larger than those in the cases of nuclear industries. Even within these uncontrolled releases many short-lived radionuclides decay in the environment before giving any significant radiation effects on marine organisms and man. Thus, only a limited number of artificial radionuclides are often encountered in the marine environment.

The artificial radionuclides which are often found in the marine environment are traditionally classified into the following categories depending mainly on their mode of production: light nuclides, fission products, activation products and transuranic nuclides. Principal members of the radionuclides in each category are listed in Table II with their half-lives, modes of radioactive decay and major sources for environmental release.

3.2.1. Light nuclides: Although tritium (hydrogen-3, H-3) and carbon-14 (C-14) are natural cosmogenic radionuclides, as has been described previously, they have also been produced massively during the nuclear weapon testing carried out extensively during the 1960's and distributed in the earth's environment, especially in the marine environment. In addition, H-3 is known to be released from both nuclear power and reprocessing plants. Although the global inventory of bomb-produced H-3 largely surpasses either that of natural H-3 or that from the nuclear industry at present, it has been estimated that the amounts originating from the nuclear industry may affect the total inventory after the year 2000, if the projected developments of the nuclear industry for the early 21st century are pursued (Preston, 1971).

3.2.2. Fission products: Since the operations of nuclear reactors are, in any case, based on controlled fission reactions of fissionable radionuclides, such as uranium-235, plutonium-239, etc., the utilization of any type of a nuclear reactor results in the production of fission products, including strontium-90, ruthenium-106 and cesium-137. As the aim of the nuclear fuel reprocessing is to remove fission products produced within fuel materials in order to recover reusable fissionable nuclides, the wastes resulting from the fuel reprocessing contain a whole series of fission products. On the other hand, as the use of nuclear weapons is based on uncontrolled fission reactions which attain the state of explosion, the nuclear weapon testing also produces a whole series of fission products with activation products simultaneously formed by neutron activation of structural materials of the weapon. Thus, atmospheric explosion tests of nuclear weapons have introduced a variety of fission as well as activation products into the marine environment through the atmosphere.

Table II

List of artificial radionuclides found often in the marine environment with their half-lives, modes of decay and major sources for environmental release

Radionuclide	Half-life	Mode of decay	Major Sources for Environmental Release
Light Nuclides			
Hydrogen-3	12.3a	B^-	Natural cosmogenic production Nuclear weapons testing
Carbon-14	5730a	"	H-3: Nuclear fuel reprocessing & power reproduction
Fission products			Nuclear fuel reprocessing Nuclear weapons testing
Cesium-137	30a	B^-	
Cesium-144	285d	"	
Ruthenium-106	1a	"	
Strontium-90	29a	"	
Technetium-99	$2.1x10^5$a	"	
(Other potentially important fission products include: ^{155}Eu, ^{131}I, ^{85}Nb-Zr, ^{103}Ru, ^{125}Sb, ^{89}Sr & ^{91}Y		"	
Activation products			Nuclear power production Nuclear weapons testing
Cesium-134	2.2a	B^-	
Chromium-51	27.7d	EC	
Cobalt-60	5.3a	B^-	
Manganese-54	291d	EC	
Silver-110m	253d	IT, B^-	
Zinc-65	245d	B^+, EC	
Transuranic Nuclides			Nuclear fuel reprocessing Nuclear weapons testing
Americium-241	433a	á	
Neptunium-237	2.1×10^6a	"	
Plutonium-238	87.7a	"	
Plutonium-239	2.4×10^4a	"	
Plutonium-240	6.56×10^3a	"	
Plutonium-241	14.4a	B^-	

Plutonium-239 cannot be differentiated from plutonium-240 in the present-day alpha-spectrometrical measurements. In order to avoid repeated tedious expression, the term and sign, plutomium-239 and Pu-239 are used below instead of the more precise term and sign, plutonium-(239+240) and Pu-(239-240).

As all of the fission products listed in Table II have their natural occurring stable isotopes in the marine environment, except for technetium-99, their biogeochemical behaviour after environmental releases follow, in principle, that of their stable counterparts. The similarity of the behaviour between the radioactive and corresponding stable isotopes depends on the degree of, and time required for, the isotopic exchange between themselves in the marine environment. It has been observed in some cases that fission products such as ruthenium-106, cerium-144, etc. in effluents released into the marine environment after waste treatments are in entirely different chemical forms from those of their stable counterparts occurring in sea water. Therefore, the radionuclides released behave quite differently from their stable counterparts (IAEA, 1975; Guegueriat, 1975).

3.2.3. Activation products: This category includes mainly radioisotopes of transition metals which are produced principally by neutron activation of stable metal isotopes, which are frequently used for piping, casing, etc. of nuclear reactors as well as nuclear weapons. As for nuclear power reactors, the metal piping is an essential structure for cooling water circulation and the circulating water may contain traces of piping material which could be activated while it circulates through reactor cores, or of radioactive metal corrosion products coming from piping material. As the primary heat exchange in nuclear power reactors is normally made in closed circuit systems, the activation products produced in the circulating water are not released directly into the environment. However, these activation products are removed from time to time from the primary circulating water and, after waste treatments, are discharged at low levels into the environment. Thus, some activation products, such as cobalt-60, zinc-65, etc. are often found in the effluents released from nuclear power plants. As previously mentioned, activation products are also introduced into the environment as the result of atmospheric nuclear weapon testing as well as nuclear fuel reprocessing, although quantities of these activation products produced, relative to those of fission products, are considerably lower.

3.2.4. Transuranic nuclides: One of the members, plutonium-239 (Pu-239), is especially important in this category because of its fissionable character suitable as nuclear fuel and bomb material, as well as its long half-live (2.4×10^4 years). As Pu-239 is produced through neutron capture by uranium-238 and successive beta-decay, both power reactor operations and nuclear explosions produce this radionuclide. Since one of the main purposes of nuclear fuel-reprocessing is to recover as much as possible the produced Pu-239 from the spent fuel materials, only a small fraction of Pu-239 escapes during the reprocessing operation into the waste fraction with fission products. Nevertheless, small amounts of Pu-239 are always contained in waste effluents released from nuclear fuel reprocessing plants and find their way to the environment with much greater amounts of fission products. The release histories of other transuranic nuclides from the fuel reprocessing are more or less similar, although they are only by-products of nuclear power production. As can be understood from their mode of production, the transuranic nuclides listed in Table II have been also produced during the past nuclear weapon testing and introduced into the marine environment through the atmosphere. Plutonium-238 and Pu-239 have also been known to be introduced into the marine environment through the disintegration of the SNAP satellite carrying Pu-238 in 1964, the loss of a nuclear weapon above Spain in 1966 and the crash of an aircraft carrying nuclear weapons at Greenland in 1968.

These transuranic nuclides listed are characterized by their long half-lives and most of them decay emitting á-particles. Since there are no stable counterparts existing in nature for these nuclides, their biogeochemical behaviour in the marine environment depends solely on their chemical properties, which are different from one to another.

I. ASSESSMENT

4. Sources and Inputs of Artificial Radionuclides into the Mediterranean

4.1. Nuclear installations as discharge sources

In considering radioactive pollution sources for the Mediterranean Sea, it is necessary to differentiate well-defined point sources of radioactive effluent discharges such as nuclear power plants, fuel reprocessing plants, etc. from wide-spread or combined sources like atmospheric fallout, river run-off, etc. Since many nuclear installations in the Mediterranean Region are located along major rivers, artificial radionuclides discharged from these installations enter into the Mediterranean Sea through rivers. While travelling along the rivers, these discharged radionuclides are subjected to various geochemical processes, such as hydrolysis, precipitation, sorption, ion-exchange, etc., so that the concentrations of the radionuclides entering the marine environment tend to be reduced in the rivers as compared with the original levels at which they were released. The situation is quite different, for example, for the nuclear fuel reprocessing plants located at sea coast such as at Sellafield (UK), or at La Hague (France), the major radioactive effluent discharge points in Europe, where effluents are directly released into the Irish Sea or the English Channel. Thus, in considering the sources for radioactive pollution of the Mediterranean Sea the individual point sources of radioactive effluent discharge become less important than the combined sources like contaminated rivers, which introduce combined amounts of radionuclides from several point sources. Nevertheless, possible discharges from various nuclear installations in the Mediterranean Region are reviewed below in order to obtain ideas on quantitative contribution of discharges from these installations to radioactive pollution of the Mediterranean Sea as a whole. The distribution of various nuclear installations in the Mediterranean Region is illustrated in Fig. 1 based on the compilation made by Calmet & Granby (1988).

4.4.1. Nuclear fuel reprocessing plants: As point sources for radionuclide discharge into the environment nuclear fuel reprocessing plants are known to be the largest contributors among various types of installations in the nuclear industry. Although some fuel reprocessing activities are being conducted in the Mediterranean basin, i.e., in France (Marcoule) and Italy (Sallugia) as shown in Fig. 1, the sizes of these reprocessing facilities are much smaller than those of industrial fuel reprocessing plants installed at Sellafield (UK), or La Hague (France). In addition, these facilities are located along the major rivers in the Mediterranean basin, Rhone and Po, and discharge their waste effluents into these river systems. As described in Section 4.1, the concentrations of radionuclides released into these rivers tend to be reduced by various biogeochemical processes during the water transport along the rivers, while some amounts of radionuclides originated from waste effluents of other nuclear installations may be added. As far as the Mediterranean marine environment is concerned, radioactive contamination is influenced only by the quantities of radionuclides in river run-off into the Mediterranean Sea, regardless of those in original releases into the rivers. Although it should not be overlooked that the discharges of radionuclides from these fuel reprocessing facilities pose radioprotection problems (such as levels of radionuclides in drinking and irrigation waters, those in local food products, etc.) in the vicinity of the points of radioactive releases, these problems should be dealt with in the scope of terrestrial pollution, but not of marine pollution.

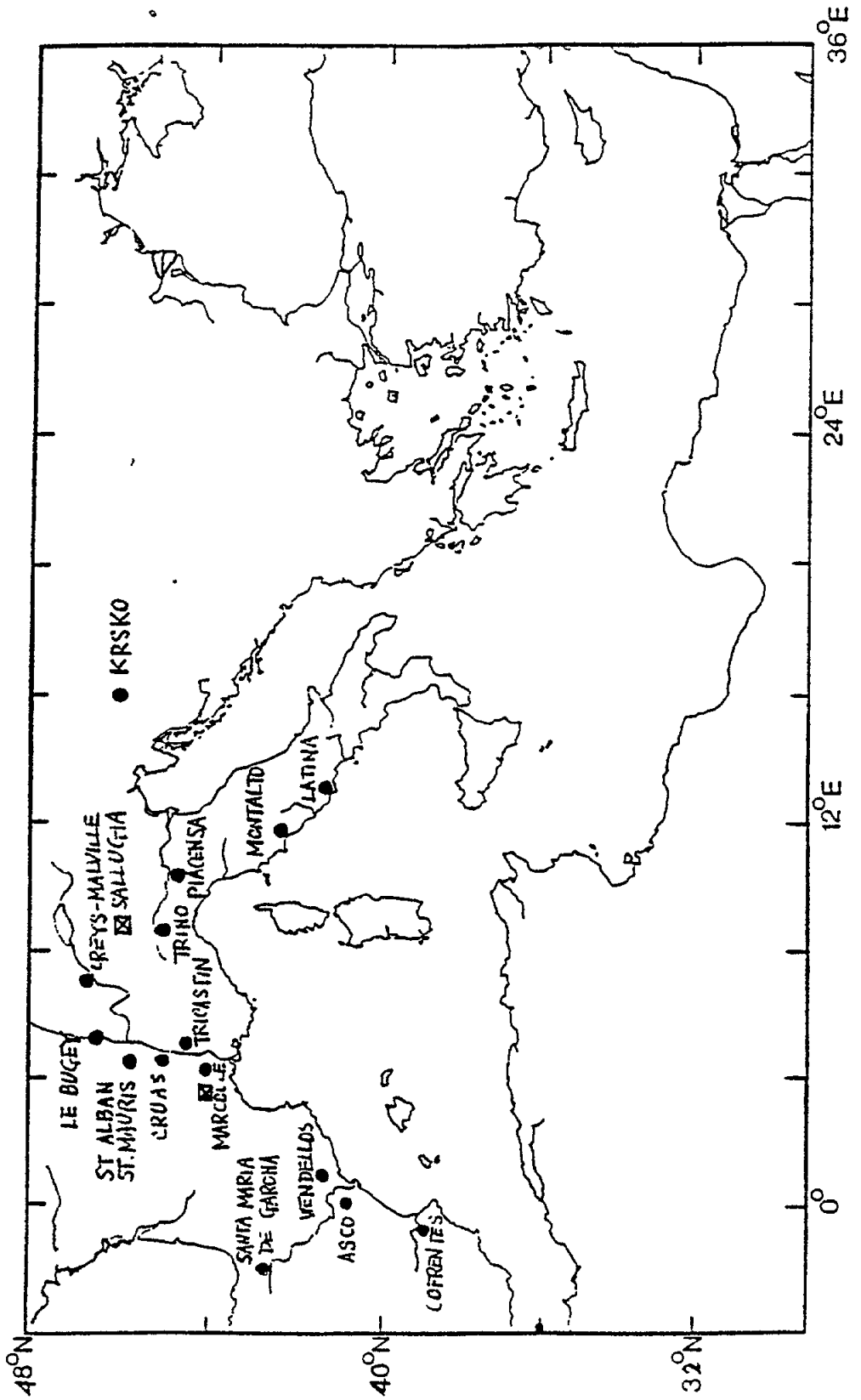


Figure 1: Distribution of nuclear installations in the Mediterranean Region
(• power station; ⊠ fuel reprocessing station).

As to the quantities of radionuclides released into the rivers, detailed data have not been published, although it is quite certain that these quantities have been monitored and controlled by competent national authorities. On the other hand, detailed data on annual discharge rates of various radionuclides into the marine environment have been published for the industrial fuel reprocessing plants at Sellafield, UK (Pentreath, 1985) and at La Hague, France (Calmet & Guegueniat, 1985). In order to obtain general ideas on quantities of selected radionuclides discharged from large-scale fuel reprocessing operations, the ranges of annual discharge rates of these radionuclides during the 1970s from the above plants are given in Table III. The data presented in Table III demonstrate that the annual discharge rates of major fission products are close to or in the order of magnitude of PBq a^{-1} (10^{15} Bq a^{-1}), although these rates fluctuate considerably, depending on many factors, such as size of the plant, schedule of reprocessing operation, type of irradiated fuel to be processed, methodology of waste treatment adopted, time for waste storage, etc. The quantities of radioactive wastes discharged from Sellafield Works have been considered as the largest intentional discharge so far made by the nuclear industry into the marine environment and some radionuclides released from the Sellafield plant into the sea could be traced several thousands of kilometers in the oceanic marine environment from their point of discharge. Judging from the data available for the quantities of river run-offs of some radionuclides into the Mediterranean Sea, the radionuclide discharges from the Mediterranean nuclear fuel reprocessing facilities into the environment are estimated to be at least three orders of magnitude or, more likely, from four to five orders of magnitude lower than those made from the Sellafield plant.

4.1.2. Nuclear power stations: Recent estimates published by IAEA (1988) demonstrate that nuclear power stations are operational in only four Mediterranean countries, France, Italy, Spain and Yugoslavia. The estimates for the numbers of power reactor units and net combined capacities of these reactors in operation and under construction in the above-mentioned four countries are given in Table IV (IAEA, 1988c). Since the numbers given for France in the upper line in the table cover the whole of France, those applicable only to the Mediterranean part of France have been estimated on the basis of the information given by Calmet & Granby (1988) and presented in the lower line of the table. These numbers show that only one-third of the total nuclear power production in France has been allocated to the Mediterranean basin. Since many power reactors in operation in the Mediterranean Region are located along rivers, as shown in Fig. 1, the fate of radionuclides in waste effluents discharged from these power stations into the river environment would be similar to that in the cases for the fuel reprocessing facilities, as described in Section 4.1.1. The situation may be different for a few power stations, where the waste effluents are discharged more directly into the marine environment.

In order to obtain a general idea on the quantities of radionuclides discharged from nuclear power plants, examples of annual discharge rates of selected radionuclides from different types of power reactors are given in Table V (Fukai & Murray, 1973). The data in the table show that annual discharge rates of different radionuclides vary widely depending on size and types of power reactors, conditions of reactor operations, procedures for waste treatment, etc. Therefore, it is difficult to estimate the quantities of radionuclides discharged from individual power stations into the environment, without basing on systematic monitoring and control data for each power station present in the region concerned. Such data have certainly been made available to competent national authorities, but not to the general public. On the basis of the data presented in Table V, however, it may be reasonable to assume that a range of radionuclide discharge rates from power reactor stations would be between 0.1 and 1 TBq a^{-1} (10^{12} Bq a^{-1}) in terms of the discharge of Cs-137 for 1000 MWe power reactor operation. If the range assumed is acceptable, it follows that the total annual discharge rate from the nuclear power stations operating in the Mediterranean Region would be approximately 5 TBq a^{-1} in terms of the Cs-137 discharge, allowing a factor of around 5.

Table III

Ranges of annual discharge rates for selected radionuclides
from industrial nuclear fuel reprocessing plants at Sellafield and La Hague
during 1970s (PBq a^{-1} or 10^{15} Bq a^{-1})

Radionuclide	Sellafield, UK	La Hague, France
Hydrogen-3	0.7 - 1.5	0.1 - 0.7
Strontium-90	0.2 - 0.5	0.003 - 0.2
Ruthenium-106	0.3 - 1.5	0.02 - 0.8
Cesium-134	0.2 - 1.3	0.03 - 0.05
Cesium-137	0.9 - 5.2	0.02 - 0.3
Cesium-144	0.07 - 0.7	0.001 - 0.04
Plutonium-239 (*)	0.02 - 0.07	(0.2-6) x 10^{-4}

(*) Including other alpha-emitting plutonium isotopes.

Table IV

Nuclear power reactors in operation and under construction in countries
in the Mediterranean Region (preliminary data in Dec. 1987)

Country	In Operation		Under Construction	
	Number of Units	Net Capacity (MWe)	Number of Units	Net Capacity (MWe)
France				
Whole country	53	49398	10	13124
Mediterranean Region	17	16273	-	-
Italy	2	1120	3	1999
Spain	9	6529	1	990
Yugoslavia	1	632	-	-
Total (Mediterranean)	29	24554	4	2989

Table V

Estimated annual discharge rates for selected radionuclides from nuclear power stations
(TBq a^{-1} or 10^{12} Bq a^{-1})

Power Station	Type of reactor	Capacity (MWe)	Estimated discharge rate		
			H-3	Co-60	Cs-137
Tarapur (India)	BW [1]	400	0.2	0.004	0.01
Carigliano (Italy)	"	150	0.4	< 0.2	0.07
Dresden I (USA)	"	210	0.2	0.4	0.07
Trino (Italy)	PW [2]	252	22	0.06	0.04
Indian Point I (USA)	"	265	41	0.2	0.1
Lanina (Italy)	GG [3]	200	1.5	0.002	0.3
Bradwell (UK)	"	300	5	0.02	1

[1] BW = Boiling water type

[2] PW = pressurized water type

[3] GG = Graphite-moderated and gas-cooled type

4.1.3. Research reactors and other point sources: The presence of research reactors in the Mediterranean countries is wider-spread than that of nuclear power reactors. The numbers of research reactors in operation in the Mediterranean Region are given by country in Table VI (IAEA, 1988c). As the total number presented in the table includes research reactors in operation in the non-Mediterranean area of France, the actual total number for the whole Mediterranean Region would be around 25. Since the normal power capacity of a research reactor is about three orders of magnitude lower than that of a power reactor unit often used at present and being around a few megawatt, the production of radioactive wastes by a research reactor would accordingly be lower although the quantities of radionuclides discharged from reactor sites are not proportional to power capacities of nuclear reactors concerned, but depend on various other factors involved in the waste effluent discharges. Nevertheless, it may be reasonable to estimate that the reduction factors for quantities of radionuclides released from independent research reactor sites into the environment are higher than those from nuclear power plants since, in addition to waste treatments in the sites, the waste effluents usually go through municipal sewage treatment systems with further reduction of radionuclide quantities. Thus, the total discharge rate of radionuclides from research reactors in operation in the Mediterranean Region would be less than 1 GBq a^{-1} (10^{-9} Bq a^{-1}) in terms of the Cs-137 discharge.

As a whole, the discharge of radionuclides into the environment at the above rate is considered negligible as compared with that from the nuclear industry. One can argue about the possibility of high local accumulation of certain radionuclides resulting from the discharge by research reactors in some environmental matrices under unusual circumstances, which might cause significant radiological effects. Such argument is, however, entirely theoretical and, especially in the marine environment, coincidental synchronization of

various favorable conditions for the unusual accumulation is quite unlikely to occur and can practically be disregarded.

Hospitals and scientific laboratories utilizing radioisotopes for medical and scientific purposes may be considered as possible point sources for radionuclide discharges into the marine environment. However, the possibilities of any significant discharges of radionuclides from these establishments are small, due to the fact that the quantities of radionuclides handled in these establishments are much smaller than those handled in the other types of sources already mentioned. Also the handling of radionuclides are performed under strict control in order to avoid accidental contamination of personnel and facilities, and radioactive wastes at significant levels are usually treated before discharging into municipal sewage systems. In addition, only short-lived radionuclides such as technetium-99m (half life: 6h), iodine-131 (half-life: 8d), gold-198 (half-life: 2.7d), etc. are used for medical purposes in hospitals. Although there are always risks of accidental contamination of personnel and facilities, these are not problems of marine pollution.

Uranium mines may be also considered as possible sources for radionuclide discharge since they mobilize some natural radionuclides belonging in the uranium decay series. Except for radioprotection problems against inhalation of powdered ores containing radium-226 by workers of the mines, environmental impacts of the mining activities are, however, considered negligible especially for pollution of the marine environment.

Table VI

Number of research reactors in operation in countries
in the Mediterranean Region (Dec. 1987)

Country	Number of research reactors in operation
Egypt	1
France	21 [(1)]
Greece	2
Israel	1
Italy	6
Libyan Arab Jamahiriya	1
Spain	1
Turkey	2
Yugoslavia	3
Total	38

[(1)] For the whole of France

4.2. Inputs through different routes

As has been pointed out in the previous section, the most important factor for controlling the radioactive pollution status of the Mediterranean Sea is the actual amount of artificial radionuclides introduced into the Mediterranean marine environment through different routes rather than their amounts released by various point sources into the atmosphere and the fresh-water environment, since the latter tends to be considerably modified before entering into the sea. Thus, in the present section attempts are made to quantify the amounts of artificial radionuclides brought into the Mediterranean through atmospheric fallout, river run-offs and water exchange via straits. It should be borne in mind, however, that there has been a major change of the radioactive pollution situation in the Mediterranean Region before and after the Chernobyl accident in 1986. The discussions given below regarding the quantitative inputs of radionuclides into the Mediterranean Sea, therefore, deal with those in the pre-Chernobyl period separately than with those in the post-Chernobyl period. Considering the abundance of existing data necessary for quantitative estimations to be conducted, the following discussions are focused in cesium-137 and plutonium-239 as the representatives of artificial radionuclides introduced into the Mediterranean marine environment.

4.2.1. Atmospheric input in the pre-Chernobyl period: Radioactive fallout resulting from large-scale explosion tests of nuclear weapons conducted during the 1960s, followed by similar tests in smaller-scale by the Chinese in the 1970s and afterwards, had represented the predominating route for the introduction of artificial radionuclides into the Mediterranean marine environment until the Chernobyl accident took place. The radionuclides produced by these tests were advected to the stratosphere, well-mixed and then transported gradually to the earth's surface in the form of dry and wet fallout. Global atmospheric fallout distributions and inventories show a strong latitudinal dependance (Hardy et al., 1972; Volchok & Toonkel, 1974; Kupferman et al., 1979).

Annual fallout delivery rates of Cs-137 and Pu-239 have been measured in the Mediterranean Region at Monaco during 1978-79 and reported to be 13.0 ± 0.2 Bq m^{-2} a^{-1} and 0.300 ± 0.004 Bq m^{-2} a^{-1}, respectively (Thein et al., 1980). These reported data are in good agreement with those obtained at Tokyo (Miyake et al., 1975) and at New York (Bennett, 1976) for the period of 1972-75. They are, however, very much lower than the rates observed during the period of early 1960s, when large fallout peaks took place. Applying the annual delivery rates cited above and the value for the surface area of the Mediterranean Sea (2.97×10^6 km^2), the annual delivery of Cs-137 and Pu-239 during the period of 1975-1985 are estimated to be respectively 39 TBq a^{-1} and 0.9 TBq a^{-1}. Although systematic measurements of the delivery rates of radionuclides by atmospheric fallout during the peak period of fallout and the successive period have not been performed in the Mediterranean Region, the information on the integrated fallout delivery covering the whole period of radioactive fallout can be obtained on the basis of the data of radionuclide measurements on soil samples. Based on the soil measurement data obtained at Monaco and elsewhere, the integrated delivery of Cs-137 and Pu-239 through atmospheric fallout up to 1986 have been estimated to be 3.3 ± 0.6 kBq m^{-2} and 76 ± 6 Bq m^{-2}, respectively (Ballestra et al., 1987; Holm et al., 1988b). Applying again the surface area of the Mediterranean Sea to these values, the estimates of 10 ± 2 PBq for Cs-137 and 0.23 ± 0.02 PBq for Pu-239 are computed as the total fallout delivery of these radionuclides into the Mediterranean Sea up to 1986.

4.2.2. River input in the pre-Chernobyl period: Since many nuclear installations existing in the Mediterranean Region are located along major rivers, river run-offs of artificial radionuclides into the sea represent major routes for the introduction of radionuclides released from these installations. The quantities

of major radionuclides released are systematically monitored at the sites, although little of this monitoring data has been published. On the other hand, on the basis of field measurements, an attempt was made to estimate the quantities of Cs-137, Pu-239 and other transuranic nuclides brought into the Mediterranean marine environment through the route of river run-offs (Fukai et al., 1981). On the basis of data obtained from radionuclide measurements performed seasonally in 1977 on river water samples collected at stations set up at the mouths of the Var and the Rhone, the annual average concentrations of Cs-137 and Pu-239 were computed for each of these rivers, taking into account seasonal variations of the river water discharges into the sea. Assuming that the annual average concentrations of Cs-137 and Pu-239 in the Var and the Rhone respectively represent those in uncontaminated and contaminated Mediterranean rivers with the radioactive releases from nuclear installations and applying the annual water discharge rates for uncontaminated rivers to be 3.3×10^8 t a^{-1} and that for contaminated rivers (the Ebro, Po and Rhone) to be 1.2×10^8 t a^{-1}, the total annual input rates of Cs-137 and Pu-239 through rivers into the Mediterranean Sea are estimated to be 1.2 TBq a^{-1} for Cs-137 and 17 GBq a^{1} for Pu-239 (Fukai et al., 1981). Since the annual average concentrations of these radionuclides in the Var and the Rhone are not necessarily representative respectively for other uncontaminated and contaminated rivers, the estimated input rates are admittedly crude. The data indicate, however, that more than 65% of Cs-137 and 90% of Pu-239 are brought into the sea in a form of suspended matter, which deposits to the bottom without allowing much time for interaction with sea water. Thus, the major fractions of these radionuclides carried down by river water into the sea do not significantly affect the distribution in Mediterranean sea water beyond the vicinity of estuarine areas.

The annual river-inputs of Cs-137 and Pu-239 into the Mediterranean Sea estimated above for 1977 are approximately 3% and 2%, respectively, of the annual fallout delivery rates of these radionuclides in 1978-79 (Thien et al., 1980). It has not been known whether these percentages had been constant or not through the whole period under consideration. If the constant percentages could be assumed for the period from early 1960s to 1985, then the total quantities of Cs-137 and Pu-239 introduced by the river input into the Mediterranean sea up to 1986 would be respectively 0.4 PBq and 5 TBq, considering the corresponding fallout delivery of 10 PBq for Cs-137 and 0.23 PBq for Pu-239. The estimated values for river inputs are considered to be valid only for indicating the order of magnitude of the quantities concerned due to the uncertainty of the assumption adopted. In any case, however, the river inputs represent the minor addition of these nuclides within the errors associated to the fallout delivery values (Holm et al., 1988b).

4.2.3. Strait inflow in the pre-Chernobyl period: It has been well-known that there is a net in-flow of sea water from the Black Sea and the Atlantic Ocean into the Mediterranean Sea (Sverdrup et al., 1942; Fairbridge, 1966). In both cases the incoming waters into the Mediterranean flow in the upper layers (4.0×10^2 km^3 a^{-1} from the Black Sea and 5.5×10^4 km^3 a^{-1} from the Atlantic), which are more or less compensated by the outflowing waters in the lower layers (1.9×10^2 km^3 a^{-1} to the Black Sea and 5.3×10^4 km^3 a^{-1} to the Atlantic). If concentration gradients of radionuclides exist between the upper and lower layers, there would be increased net inflow or outflow of radionuclides depending on the slope of the concentration gradients.

If we take the Cs-137 concentrations 16 mBq l^{-1} and 5 mBq l^{-1} respectively for the Black Sea surface water and the outgoing Mediterranean water (Vakulovskii et al., 1980), then a net annual Cs-137 input of 5.4 TBq a^{-1} into the Mediterranean is computed. Since the levels of Cs-137 in the Black Sea surface water have been known to be much higher in the past, this could lead to an input of 0.1 PBq during the period of 25 years up to 1986 after the decay correction. Similarly, the annual Atlantic in-flow of Cs-137 may be estimated to be 0.12 PBq a^{-1}, considering the Cs-137 concentration of 5 mBq l^{-1} for the inflowing Atlantic water and that of 3

mBq l^{-1} for the outflowing Mediterranean water. This corresponds to the input of about 1.5 PBq during the period of 25 years up to 1986 (Holm et al., 1988b).

Taking the Pu-239 concentration of 50 mBq l^{-1} for the inflowing Black Sea water as well as for the outflowing Mediterranean water, of 10 mBq l^{-1} for the inflowing Atlantic water and of 40 mBq l^{-1} for the outflowing Mediterranean water, similar computations estimate the annual inflow rates of 10 GBq a^{-1} and -1.5 TBq a^{-1} respectively from the Black Sea and the Atlantic as well as corresponding Pu-239 inputs of 0.25 TBq and -38 TBq during the period of 25 years up to 1986. While the net input of Pu-239 from the Black Sea may be higher than the estimated value due to possible higher levels of Pu-239 in the Black Sea surface water in the past, there seems to be a net loss of Pu-239 through the Strait of Gibraltar into the Atlantic. No consideration was given to the effects of water flow from the eastern to the western Mediterranean in these calculations.

4.2.4. Total input in the pre-Chernobyl period: In order to summarize the various estimations made as to the quantities of Cs-137 and Pu-239 introduced into the Mediterranean marine environment through various routes, the values representing total amounts of input of these radionuclides up to 1986 are presented in Table VII. Although the errors involved in these estimations are large, the values in the table indicate that the inputs of the radionuclides through atmospheric fallout predominate those through the other routes. While the river inputs represent only a small additional delivery of both radionuclides to that brought down by fallout, the exchange of water through the Strait of Gibraltar might significantly influence the total quantities of radionuclides, especially those of Cs-137 occurring in the entire Mediterranean Sea.

4.2.5. Chernobyl fallout: In April 1986 the operational accident of the reactor No. 4 at the Chernobyl Nuclear Power Station in USSR caused severe radioactive fallout deposition from highly radioactive air plumes spread and transported mainly over Europe beyond national boundaries. Due to the location of the accident site and meteorological conditions at the time of the accident, the Mediterranean Region had been subjected to the relatively strong influence of radioactive contamination resulting from the Chernobyl fallout. Envelopes of trajectories followed by the radioactive air plumes over Europe after the Chernobyl accident are presented in Fig. 2 (Calmet & Granby, 1988). As can be seen from the figure, these trajectories were rather complicated so that the fallout deposition at different localities in the Mediterranean Region varied considerably depending on the geographical, topographical and meteorological factors at the individual locations concerned. To demonstrate the heterogeneity of the fallout deposition in the Mediterranean Region, examples of the results of measurements on the amounts of the Cs-137 deposition from the Chernobyl fallout observed at Mediterranean stations and elsewhere are presented in Table VIII. Differences of a few orders of magnitudes exist in some cases between the results obtained within the Mediterranean Region. Under the circumstances, it is rather difficult to estimate the change caused by the Chernobyl fallout in addition to the quantities already delivered by fallout as a result of the nuclear detonation tests up to 1986. It may be deduced, however that the Chernobyl deposition was significantly higher in the northern parts of the Mediterranean. Approximations may lead to a range of estimates between 25 and 40% increase of the Cs-137 deposition for the Mediterranean Region, compared with that already existing in the region up to 1986 (Holm et al., 1988a).

Table VII

Inputs of cesium-137 and plutonium-239 into the Mediterranean marine environment up to 1986 through different routes

Route	Quantities introduced	
	Cs-137 (PBq) [1]	Cs-239 (PBq) [1]
Atmospheric fallout	10 ± 2	0.23 ± 0.02
River run-off	0.04 ± 0.1	0.05 ± 0.002
Exchange via Strait with the Black Sea	0.1	0.003 >
Exchange via Strait with the Atlantic Ocean	1.5	- 0.04
Total	12 ± 2	0.19 ± 0.02

[1] PBq = 10^{15} Bq.

Table VIII

Deposition of Cs-137 by the Chernobyl fallout in May 1986 at various locations in the Mediterranean Region

Location	Cs-137 deposition (k Bq m^{-2})	Literature
Chernobyl, USSR	**1 x 10^4**	**USSR State Comittee (1986)**
Ljubljana, Yugoslavia	26	Juznic & Fedina (1986)
C. & N. Greece	45*	Papanicolaou & Kritidis (1988)
S. & W. Greece	5*	"
N. Italy	15	ENEA (1986)
Ispra, Italy	19	Pharabod et al. (1986)
Milano, Italy	10	"
La Spezia, Italy	1.5	Delfanti & Papucci (1988)
C. Italy	3.7	ENEA (1986)
S. Italy	0.7	"
Monaco	3.1	Ballestra et al. (1987)
Cadarache, France	3.6	Pharabod et al. (1986)

* Values not corrected for Cs-134 and Cs-137 due to earlier fallout.

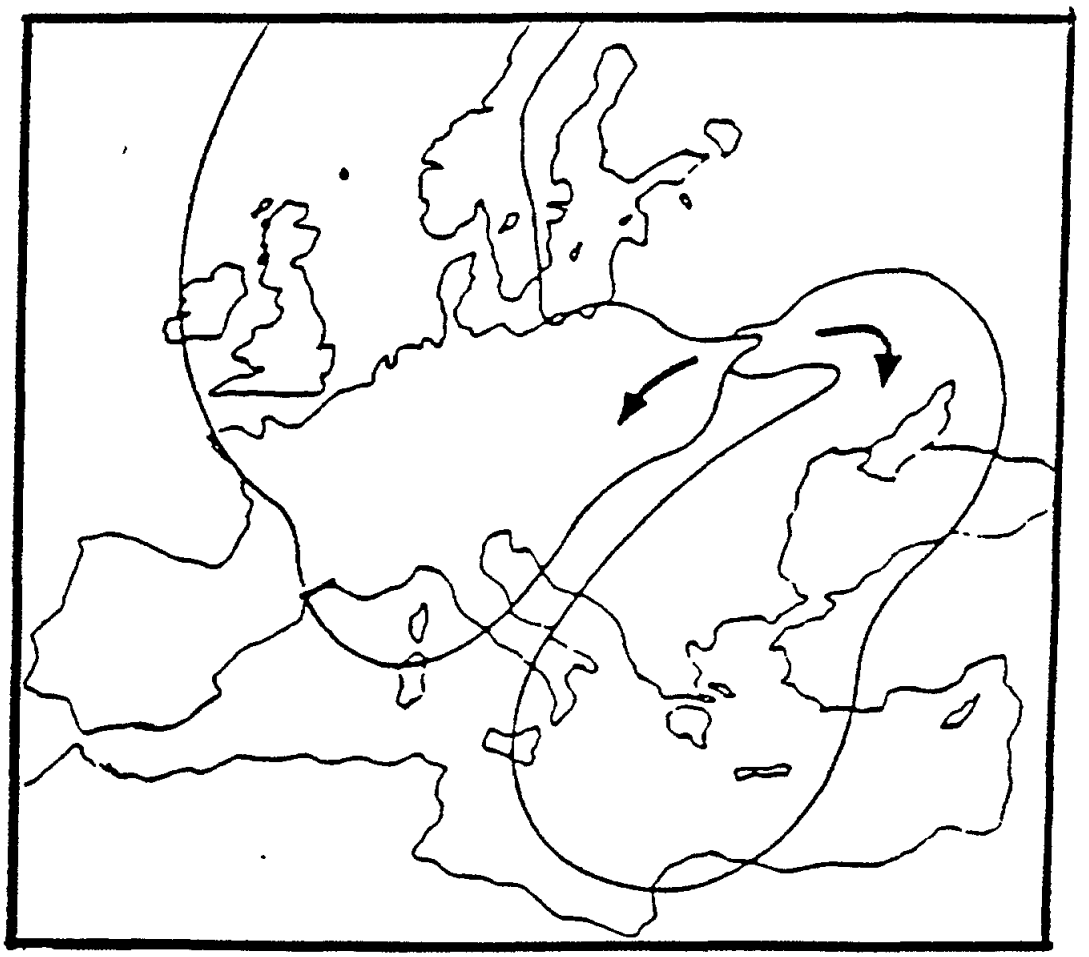


Figure 2: Trajectories of the highly radioactive plumes after the Chernobyl accident over the Mediterranean Region.

The deposition of Pu-239 and other transuranic nuclides from the Chernobyl fallout in the Mediterranean Region was much smaller than that of Cs-137. The activity ratio Pu-239/Cs-137 was in the order of 5 x 10^{-5} for Chernobyl fallout compared with that of 1-2 x 10^{-2} for fallout from nuclear weapon testing. The deposition of Pu-239 following the Chernobyl accident was about 10 mBq m^{-2} or only 0.1% of previous integrated deposition (Holm et al., 1988a).

5. The Biogeochemical Behaviour of Radionuclides in the Mediterranean

All radionuclides introduced into the marine environment undergo various physical, chemical and biological processes taking place in the sea. These processes may be simple physical dispersion, but in many cases may involve complicated chemical and biological interactions of the radionuclides introduced with other environmental matrices such as inorganic and organic suspended matter, variety of living organisms, bottom sediments, etc. In order to interpret or predict the levels of radionuclides in various marine environmental compartments, it is essential to understand, step by step, the operating mechanisms of biogeochemical processes in situ. The behaviour of radionuclides in the sea depends primarily on their chemical properties, but it may also be influenced significantly by properties of interacting matrices and other environmental factors. In the present section general descriptions are given, at first, on characteristics of the Mediterranean marine environment as well as on the important parameters such as sediment and biological concentration factors required for pollution prediction and assessment. These descriptions are followed by brief reviews of characteristic behaviour of selected radionuclides encountered often in the marine environment.

5.1. Characteristics of the Mediterranean marine environment

The Mediterranean Sea is a fairly deep water body surrounded by land and is characterized generally by low precipitation, high evaporation, high salinity, low tidal action, low nutrient content, low suspended load, low biological productivity, etc. While these characteristic features of the Mediterranean Sea result in hydrographical conditions quite different from those in other seas and oceans, the fundamental biogeochemical processes taking place in coastal boundaries as well as in water columns are considered not much different from those in other seas. For example, in shallower enclosed seas like the Baltic Sea some radionuclides introduced through atmospheric fallout are brought down quickly to the sea bottom by high suspended load in sea water, while in the much deeper Mediterranean Sea the same radionuclides stay in solution for a fairly long time from several years to tens of years. This is due to the lower suspended load and greater depths of the Mediterranean than those of the Baltic. The above example demonstrates that even though the sorption of radionuclides onto the surface of suspended matter operates similarly in both seas, the resultant levels of radionuclides in sea water are quite different between the Mediterranean and the Baltic.

The longer length of time for which the introduced radionuclides stay in solution is considered important in determining their behaviour thereafter in the Mediterranean, since this allows the radionuclides more time to equilibrate with their stable counterparts occurring naturally in sea water. As a result, the behaviour of these radionuclides should be exclusively controlled by that of their stable counterparts as soon as the isotopic exchange between them has been completed. The process described is considered to have significant influence on the levels of radionuclides, such as Sr-90, Cs-137, etc. in the Mediterranean water column since the major route for their introduction in the Mediterranean Sea has been identified as atmospheric fallout, and the whole surface of the Mediterranean is represented as its receptor.

In the case of river run-offs of radionuclides in the Mediterranean Sea, the conditions associated with lower tidal action are considered to accelerate settling of suspended matter carried by rivers and, at the same time to limit the interaction of radionuclides with bottom sediments (vigorous tidal currents would induce active resuspension of sedimented materials into the water column). Generally speaking, the specific environmental characteristics of the Mediterranean Sea seem to act towards the direction of moderating interactions between radionuclides introduced and various environmental matrices. This is done in such a way as to stabilize the levels of these radionuclides in different environmental compartments, except for cases where special local conditions disturb the general tendency.

5.2. Sediment concentration factor

Although the major route for the input of radionuclides into the Mediterranean Sea has been attributed to atmospheric fallout, a major part of radionuclides discharged from nuclear installations located in the Mediterranean Region reach the sea through river run-offs, as discussed in Sections 4.1 and 4.2. Soluble fractions of the radionuclides transported to the sea by river run-offs are then subjected to interactions with suspended matter and bottom sediment particles prevailing in estuarine waters, although major fractions of the radionuclides (more than 60% for Cs-137 and more than 90% for Pu-239) enter into the sea in a sorbed state onto riverine suspended matter. In estuaries, the interactions stated above are enhanced by current disturbances which bring bottom materials into the water phase and the settling of river-borne particles also takes place. This is the first marine geochemical process encountered by the riverine radionuclides introduced into the sea. Thereafter, similar interactions are continued to take place even in the off-shore marine environment - the extent and senario of which depend on various local conditions and the time-scale to be considered.

In order to describe the affinity of various radionuclides in solution with sediment materials, settled or suspended, a term "sediment/water distribution coefficient (K_d)" or "sediment concentration factor" has been traditionally utilized. The term is defined as the ratio of concentration of an element or a radionuclide, which is exchangable with that in solution, in the sediment phase to concentration of the same element or radionuclide in the solution phase, which is in contact with the sediment phase. Since the term "distribution coefficient" has been derived based on a model involving riverside reactions governed by the law of mass action under steady state equilibrium conditions, it is not certain whether the term is appropriate for expressing the partition of radionuclides between solution and solid phases in complicated environmental processes prevailing in situ, where mechanisms of the partitioning may not be clearly understood. Therefore, the use of a looser term "sediment concentration factor" may be preferable when complicated environmental processes are involved. This is especially true for coastal areas, where steady state conditions are unlikely to be fulfilled.

If a sediment concentration factor for a radionuclide could be uniquely determined as to various types of sediment materials, then it would become possible to predict the level of the radionuclide to be attained in sediments on the basis of its concentration in ambient sea water by utilizing the determined value of the sediment concentration factor. This implies practical importance for radioprotection practices of the environment, since the control of radioactive contamination of sediments may be possible by controlling the sea water concentration of the radionuclides concerned. In view of this practical importance, much effort has been devoted for determining sediment concentration factors of various radionuclides for different types of sediment materials by conducting laboratory experiments as well as by investigating radiologically

contaminated fields. However, as can be immediately understood from the difficulties in defining the term "sediment concentration factor" based on precise mechanisms involved in the partitioning of radionuclides, a wide range of values for sediment concentration factors have been experimentally obtained for each radionuclide depending on types of sediments dealt with, conditions of the experiments as well as field observations, methodologies of measurements, etc. Thus, the situation had become confused as to the values of the sediment concentration factors to be utilized for practical prediction of sediment contamination. In view of the practical importance of a good choice of reasonable values of sediment concentration factors for control and prediction of radioactive contamination of sediments, the IAEA took the initiative to clarify this confused situation by critically reviewing various values reported for different types of sediments under different conditions and compiled the recommended mean values of sediment concentration factors with minimum and maximum values (IAEA, 1985). In the compilation, distinction was made between "sediment/water distribution coefficients" to be used for pelagic ocean conditions and "sediment concentration factors" to be used for coastal conditions. The former is considered important for a situation like deep-sea dumping of radioactive wastes, while the latter applies to coastal discharge of radioactive waste effluents. Taking into account the present situation of radioactive contamination in the Mediterranean Sea, the mean recommended values for coastal sediment concentration factors of selected radionuclides often encountered in the marine environment are given in Table IX. The values presented in the table vary from one order of magnitude to 10^6 with the minimum/maximum ranges by a factor from 5 to 10 towards both sides. These values represent relative degrees of partitioning of the radionuclides listed between sediments and ambient water phase and are useful, with careful examination of local conditions of areas to be considered, for the prediction and control of radioactive contamination of sediment materials.

Table IX

Recommended mean values of coastal sediment concentration factors
for selected radionuclides

Radionuclide	Mean Coastal Sediment Concentration Factor
Hydrogen-3	1
Cobalt-60	2×10^5
Strontium-90	1×10^3
Ruthenium-106	3×10^2
Cesium-137	3×10^3
Plutonium-239	1×10^5

5.3. Biological concentration factor

In contact with a variety of living organisms occurring in the marine environment, the radionuclides introduced into the sea undergo various biological processes and enter into complicated marine food-webs. The major route of radiation exposure of man to artificial radionuclides occurring in the marine environment should be through ingestion of radiologically contaminated marine organisms. It will be discussed below that the levels of radioactive contamination of edible marine organisms represent the key factor for radioprotection of the marine environment.

On the analogy to the cases for marine sediments, biological concentration factors are defined as the ratio of concentration, usually on wet weight basis, of an element or a radionuclide in biological materials to concentration of the same element or radionuclide in ambient sea water. Although the term has been defined by assuming steady state conditions, these conditions are rarely fulfilled in biological processes taken place in situ. In addition, since the partition of radionuclides between biological materials and the ambient sea water is considered to be achieved through complicated combinations of biological and biochemical processes, such as assimilation, excretion, grazing, specific absorption, ion-exchange, etc., it is practically impossible in many cases to understand precise operating mechanisms of the partitioning. This is why the biological concentration factors do not make any reference to the partitioning mechanisms, but represent simply concentration ratios of an element or a radionuclide between biological materials and their ambient water.

If a biological concentration factor of a radionuclide could be uniquely determined for each of the major groups of marine organisms, the level of the radionuclide in the organisms would be predicted and controlled based on concentration of the radionuclide in ambient water. A great number of experimental and field studies for determining biological concentration factors of many radionuclides for a variety of marine organisms have been carried out under different conditions, resulting in values of biological concentration factors varying in wide ranges. Similarly to what has been done in the case of the sediment concentration factors, a number of values reported as biological concentration factors of various radionuclides for different groups of marine organisms has been critically reviewed and compilations of the recommendable mean values with the minimum/maximum ranges were made for major groups of marine organisms (IAEA, 1985). On the basis of the above compilation, the values of mean biological concentration factors of selected radionuclides for major groups of edible parts of marine organisms are given in Table X. The values presented in the table vary in a range of 1 - 10^4 and represent specific tendencies of the radionuclides listed in the assimilation by different types of marine organisms.

Table X

Recommended mean values of biological concentration factor for selected radionuclides as to different groups of marine organisms

Radionuclide	Mean Biological Concentration Factor		
	Molluscs (Except Cephalopods)	Crustaceans	Fish
Hydrogen-3	1	1	1
Cobalt-60	5×10^3	5×10^3	1×10^3
Strontium-90	1	2	2
Ruthenium-106	2×10^3	100	2
Cesium-137	30	30	100
Plutonium-239	3×10^3	300	40

5.4. Specific behaviour of individual radionuclides

5.4.1. Hydrogen-3 (H-3, tritium): While the major global inventory of H-3 at present has resulted from the nuclear weapon testing, it is also introduced into the marine environment by waste discharges from nuclear fuel reprocessing and power plants. Since practically all H-3 atoms produced in the different processes enter into the marine environment in the form of a tritiated water molecule, the behaviour of H-3 is considered to be identical with that of the normal water molecule. Strictly speaking, however, the behaviour of the tritiated water molecule is slightly different in physical processes such as evaporation and condensation from that of the normal water molecule due to their mass difference (isotopic effect), but such an effect is too small to have any practical significance. In this sense, H-3 is an ideal tracer for studying water movements in the marine environment. In fact, H-3 has been utilized extensively for tracing mixing and movement of water masses in the oceanic areas. The behaviour of H-3 in biological processes has been also experimentally studied, showing that tritiated water could be incorporated into the organic fractions of marine organisms such as algae, mussels, shrimps, etc. to an extent close to the level of the normal water molecule, but at slightly lower levels (Kirchman et al., 1979; Bonotto et al., 1981). This means that the values of the coastal sediment concentration factor and the biological concentration factors of H-3 do not exceed one, being slightly lower than one in usual cases, as indicated in Tables IX and X.

5.4.2. Cobalt-60 (Co-60): This radionuclide is a member of the activation products, which include other radionuclides of transition metals such as manganese-54, iron-59, zinc-65, etc. (section 3.2.3), and has been introduced into the marine environment by waste discharges from both nuclear power and fuel reprocessing plants as well as by atmospheric fallout (Table II). Since trace amounts of stable isotopes of these metals occur naturally in sea water at a level between a few ngl^{-1} and several μgl^{-1}, the behaviour of these radionuclides including Co-60 is identical, in principal, to that of their stable isotopes after the isotopic equilibration between stable and radioactive nuclides has been completed. These metals tend, however, to be subjected to inorganic and organic complexing and hydrolysis in seawater and to interact with sediment materials by ion-exchange, coprecipitation and other adsorption reactions. They often exhibit more than one oxidation state and oxidation/reduction reactions may occur under changes of environmental conditions. These chemical characteristics specific to these transition metal nuclides tend to complicate their behaviour in the environment. In addition, transition metal radionuclides discharged by nuclear industries tend to be subjected to complicated waste treatment procedures utilizing complexing agents, so that they are often in complexed forms before they are released. This may retard their quick isotopic equilibration with stable counterparts when they are introduced into the sea. In general terms, nevertheless, the transition metal radionuclides have high sediment concentration factors ($>10^3$). Due to the great interest in recent years directed towards heavy metal pollution of marine edible organisms, the uptake and loss of many transition metals by a variety of marine organisms have been extensively studied under various laboratory and field conditions, resulting in accumulation of abundant data on ingestion/excretion dynamics of these transition metals by the marine organisms. Although the data obtained have not always been consistent or comparable, the transition metal radionuclides including Co-60 have, in general, high values of the biological concentration factors around the order of 10^3 for different groups of marine organisms (Table X).

5.4.3. Strontium-90 (Sr-90): Stable and radioactive strontium belongs chemically to the alkali earth elements and the radionuclide is considered to be brought into the marine environment in the divalent ionic form specific to the alkali earth elements, regardless of whether it comes through

atmospheric fallout or it is released from nuclear installations. Sea water usually contains around 8 mg of stable strontium per litre, depending on its salinity. Thus, as soon as Sr-90 in the ionic form enters into the marine environment the

isotopic exchange of Sr-90 with its stable counterpart is quickly achieved and thereafter the behaviour of Sr-90 follows exactly that of its stable counterpart. This is important for assessing the extent of radiological impact of Sr-90 on radiation exposure of man, since strontium tends to be incorporated in human bones and gives radiation to human bone marrow. The complete isotope exchange of Sr-90 with stable strontium in the marine environment described above reduces the radiation dose received by the human bone marrow when it is incorporated into the bone. The isotope dilution effect of Sr-90 by its stable strontium in the marine environment is amplified by the presence of a large amount of calcium (about 400 mg Ca l^{-1}), a chemical homologue of strontium, as one of the major dissolved constituents in sea water. In many geochemical and biological processes calcium acts very similar to strontium, so that the presence of calcium in sea water has the effect of further isotopic dilution. Both calcium and strontium are not very reactive in sea water and tend to remain in solution without association with terrigeneous particulate matter. Even in the assimilation of calcium as well as strontium by marine organisms as materials for their calcareous tissues, the levels of Sr-90 taken up are suppressed by the isotopic dilution effect described above. This is why the values of biological concentration factors of Sr-90 for various marine organisms are generally lower compared to those of other radionuclides (Table X).

5.4.4. Ruthenium-106 (Ru-106): Although Ru-106 has also been introduced into the marine environment through atmospheric fallout, this fission product constitutes one of the major members of radionuclides in waste effluents from nuclear fuel reprocessing plants such as those at Sellafield, U.K. and La Hague, France. Because the oxidation state of ruthenium changes easily between 0 and 8 depending on ambient conditions in solution, Ru-106 forms various complexes with different ligands in acidic media. Thus, Ru-106 in the acid solution of the mixed fission product tends to escape various steps of waste effluent treatments at reprocessing plants. After the dissolution of spent nuclear fuel with nitric acid at fuel reprocessing plants, Ru-106 is present in the form of various complexes of nitrosyl ruthenium, such as nitrato nitrosyl complexes and nitro nitrosyl complexes having varying electric charges. The chemical speciation of Ru-106 is further complicated by the polymerization of nitro-complexes in diluted acid or neutral solution. Thus, depending on the condition and history of the treatments of waste effluent, different degrees of complexation and polymerization of Ru-106 occur when the effluents become in contact with sea water. The polymerized fraction of Ru-106 complexes is expected to serve as nuclei for the formation of colloids and particles which may be charged electrically negative or positive. While positively charged colloids and particles tend to be sorbed onto the surface of seaweeds and other marine organisms, those negatively charged remain in the medium for several weeks (Gueguoniat, 1975; IAEA, 1975). This is why the behaviour of Ru-106 in the marine environment is rather independent from that of stable ruthenium in sea water, although trace amounts of stable ruthenium occur naturally in sea water (0.7 ng l^{-1}).

Since the Ru-106 released from the Sellafield fuel reprocessing plant was known to be taken up extensively by one particular species of seaweed, Porphyra and the seaweed is used as food by some local populations around the Irish Sea, the transfer of Ru-106 through the route- (waste effluent - sea water - seaweed - man) - had constituted in the past the critical pathway of radionuclides leading to human radiation exposure. Thus, extensive studies on the uptake of Ru-106 not only by the Porphyra but also by other marine organisms have been conducted in laboratories as well as in contaminated areas. While biological concentration factors of Ru-106 for Porphyra as well as some species of molluscs have been found to be high, those for other marine organisms such as crustaceans and fish were lower (Table X), suggesting that the major mechanism of the Ru-106 accumulation may be adsorption of this radionuclide onto the body surface of marine organisms.

5.4.5. Cesium-137 (Cs-137): Cesium belongs to the alkali metal elements which exhibit always positive monovalent ionic form in aqueous solution and Cs-137 in the sea water medium is not an exception. Although stable cesium occurs naturally in sea water at a level around 3 µg l^{-1}, the isotopic dilution effect similar to that described for Sr-90 by stable strontium in sea water is not as pronounced since the amounts of stable cesium present in sea water are much smaller than those of stable strontium (8 mg l^{-1} in sea water). In addition, the chemical similarity between cesium and potassium, a chemical homologue of cesium, is not as close as that between strontium and calcium. Therefore, the extent of the amplification of the isotopic dilution effect by the presence of a chemical homologue of the diluting stable element, so described in the case of stontium and calcium, is very limited for cesium and potassium. Thus, the behaviour of Cs-137 in the marine environment is rather independent from that of its chemical homologue, potassium, although similar tendencies in their behaviour, in general terms, exist between these chemical homologues.

Since considerable amounts of Cs-137 have been introduced into the marine environment through atmospheric fallout as well as through the discharges from nuclear installations and Cs-137 tends to be distributed widely from its point of the introduction staying in solution, a great number of measurements of this radionuclide have been carried out so far on various marine environmental matrices. This is probably the most widely measured radionuclide in the marine environment. Due to its simple ionic state Cs-137 undergoes adsorption onto naturally occurring suspended solids, especially onto clays, by ion-exchange reactions, so that its coastal sediment concentration factor is moderately elevated, being at a level of around 1000 (Table IX). Since Cs-137 is known to be incorporated into muscle tissues of various marine organisms, which may be eaten by man, biological uptake of this radionuclide by marine organisms is important from the radioprotection viewpoint. On the basis of results obtained in a number of studies on the uptake and loss of Cs-137 by various types of marine organisms, biological concentration factors of around 100 and biological half-lives of around 100 days have been deduced for general use in radioprotection practices.

5.4.6. Plutonium-239 (Pu-239): In view of its high radiotoxicity and long half-life, Pu-239 is considered as one of the most hazardous radionuclides found in the marine environment. Since no stable element exists for Pu-239, the behaviour of Pu-239 in the environment is governed solely by its chemical characteristics. Plutonium is known to occur in sea water in different oxidation states; Pu(III), Pu(IV), Pu(V) and Pu(VI). Plutonium-239 in higher oxidation states, Pu(V) and Pu(VI), is considered to be more soluble in sea water than in lower oxidation states, Pu(III) and Pu(IV). In fact, Lovett & Nelson (1981) found that the plutonium in sea water in the Irish Sea consisted primarily of Pu (V+VI) and that the distribution coefficient values for this pair are less than 10^4, whereas those for the Pu (III+IV) pair are about 10^6. Fukai et al. (1987) have also demonstrated that Pu-239 in atmospheric fallout dissolved in rain is in higher oxidation states, Pu(V) and Pu(VI), at Monaco and brought down to the Mediterranean surface most probably in the major form of Pu(V). The changing oxidation state of Pu-239 complicates its behaviour in the marine environment, since the rate and degree of interactions of Pu-239 with sedimentary materials and organisms vary depending on its oxidation state. Thus, the oxidation-reduction reactions taking place in the marine environment become important for deciding the direction of the behaviour of Pu-239. Relatively high sediment concentration factors of Pu-239 observed for some types of sedimentary materials are considered to suggest prevailing reducing conditions at the water-sediment interfaces, where the non-detrital hydrous oxides of iron and manganese may be playing an important role (Aston & Stanners, 1981). The behaviour of plutonium in biological processes have been extensively studied in laboratories by

applying a gamma-emitting isotope of Pu, Pu-237. Studies on the uptake of Pu-237 by invertebrates have shown that specific organs of some marine organisms, such as branchial heart of octopus and hepatopancreas of crabs, accumulate Pu at high levels (Guary & Fowler, 1982; Guary & Negrel, 1981). The biological concentration factors of Pu-239 are comparatively high, in general, but vary considerably for different groups of marine organisms (Table X).

6. Levels of Artificial Radionuclides in the Mediterranean

Artificial radionuclides released from various sources are introduced into the Mediterranean Sea through different routes and then distributed in sea water, marine sediments and marine biota after having been subjected to a variety of environmental processes, such as sorption, biological assimilation and excretion, food-web transfer, horizontal and vertical transports by physical and biological mechanisms, etc. Therefore, the levels at which these radionuclides are found in various marine environmental compartments in the Mediterranean Sea should not be considered to be static, but to be dynamic in nature. There is no reason to believe at present that the steady state equilibria have been attained between inputs and physical decay or outputs of these radionuclides in the Mediterranean marine environment. On the contrary, even apart from the unexpected inputs due to the Chernobyl accident, the rates of inputs of artificial radionuclides into the Mediterranean Sea are expected to increase in coming years, if the projected developments of nuclear industries are accepted, resulting in the elevation of levels of some long-lived radionuclides in Mediterranean environmental compartments. The most important environmental problem is, therefore, to identify the increase of the input rates by periodical reviews of the levels of longer-lived radionuclides in different environmental compartments at appropriate intervals. In the present section the data so far obtained on the levels of selected radionuclides in the Mediterranean marine environment are reviewed for sea water, marine sediments and marine biota in order to deduce the trends, if any, for changing levels. Since the quality of the data dealt with has prerequisite importance on the conclusions to be drawn, the reliability of the data must be examined beforehand. As significant changes in the levels of some radionuclides have occured before and after the Chernobyl accident, the reviews are made respectively for the pre-Chernobyl period and the post-Chernobyl period.

6.1. Reliability of environmental radionuclide measurements

Any measurements on trace amounts of pollutants in environmental matrices are always a delicate affair and the data produced in these measurements should be critically examined before utilizing any scientific purposes. Although standard practices for performing analytical data quality control of specific measurements are available and much effort has been devoted to improving the reliability and comparability of the data produced, the data quality of trace pollutant measurements are not always satisfactory. The situation for environmental radionuclide measurements is not an exception, but generally speaking, it seems that they suffer much less from their inherent sources of errors than do other types of trace measurements, such as trace heavy metal measurements, trace organic pollutant measurements, etc. This is considered due to the following aspects specific to environmental radionuclide measurements:

(i) Sensitivity and specificity for identifying and quantifying a radionuclide are usually higher than those for many other trace measurements;

(ii) Chances for contaminating the samples to be analysed during sampling and analytical

manipulations are much smaller;

(iii) Reagent blanks for the measurements are practically negligible;

(iv) Only a limited number of qualified and well-equipped laboratories in the region are engaged in the measurements; and

(v) Well-organized intercalibration exercises and standard reference materials exist for analytical data quality control.

In spite of the fact that the aspects stated above may give assurance on the quality of data on the levels of radionuclides in the Mediterranean marine environment, the limited abundance of the available data with respect to space and time does not allow to make accurate deductions as to the changing trends of these levels.

6.2. Levels in the pre-Chernobyl period

In the pre-Chernobyl period up to 1986 the radionuclide data covering significant areas of the Mediterranean Sea have been available for only two long-lived radionuclides, Cs-137 and Pu-239, since other radionuclides introduced into the Mediterranean Sea through atmospheric fallout resulted from the extensive nuclear weapon testing in the early 1960s had already decayed to very low levels in the late 1970s, except for Sr-90. As to Sr-90, extensive measurements of this radionuclide has never been conducted in the Mediterranean marine environment, because of the technical difficulties involved in the Sr-90 measurement as well as the fact that a constant ratio has been known to exist between radioactivity concentrations of Sr-90 and Cs-137 in their fallout delivery. For these reasons the levels of only Cs-137 and Pu-239 in sea water, marine sediments and marine organisms are primarily dealt with in the following reviews.

6.2.1. Levels in sea water: The radioactivity concentration levels of Cs-137 measured for surface waters of the Mediterranean Sea by various authors up to 1986 are given in Table XI. In Fig. 3, the locations of sampling stations are given for the data in Table XI obtained by Fukai et al. (1980). The data in the table include those of systematic long-term measurements of Cs-137 in surface sea water performed at La Spezia, Italy covering a period from early 1960s to 1986 (Delfanti et al., 1988). These data show clearly the decrease of the Cs-137 level from early 1960s to late 1960s and its leveling off in the 1970s and afterwards. These changes in the levels correspond to the decrease of amounts of radioactive fallout due to the nuclear weapon testing at the time. Excluding a value obtained in the Rhone Estuary and the data obtained before 1970, the values presented in Table XI for both coastal and off-shore areas are in a relatively narrow range of 2.6-5.6 mBq Cs-137 l^{-1}. Considering the half-life of Cs-137 (30 years) it seems reasonable to conclude that the base-line level of Cs-137 in Mediterranean surface water in 1985 had been around 4 mBq l^{-1} or slightly lower.

The vertical distributions of Cs-137 in Mediterranean water columns measured at the four profile stations shown in Fig. 3 are illustrated in Fig. 4 (Fukai et al., 1980). As shown in Fig. 4, the vertical profiles of Cs-137 distributions are characterized by its concentration maxima between 50m and 250m, although the maximum concentration of Cs-137 is different for each profile. The formation of the Cs-137 maxima in the vertical profiles was considered to be caused by vertical water movement rather than by sinking of particulate matter. Different slopes of decrease in Cs-137 concentrations below the maxima found between the western and eastern basins of the Mediterranean were attributed to the differences in rate of vertical water mixing for two basins. Basing on these profile data and comparing these with the amounts of Cs-137 brought down into the Mediterranean by fallout it was concluded that at least 70%, probably much more, of the total amounts of the Cs-137 delivery should be still in the Mediterranean water column (Fukai et al. 1980).

Table XI

Levels of cesium-137 in surface waters of the Mediterranean Sea
during the pre-Chernobyl period

Location	Area No	Time	No. of data	Cs-137 Level mBq l^{-1}	Literature
Coastal Zone					
French Coast	II	Apr.'86	8	3.1	Calmet & Granby (1988)
Rhone Estuary	"	"	1	15.5	"
La Spezia, Italy	IV	'62-'65	8	32	Delfanti et al. (1988)
		'66-'70	10	11	"
		'71-'75	10	2.6	"
		'76-'80	10	3.2	"
		'81-'85	10	4.1	"
Rovinj, Yugoslavia	V	'78-'85	8	4.4	Bauman (1990)
Rijeka, Yugoslavia	V	" "	8	4.6	"
Split, Yugoslavia	V	" "	8	4.1	"
Dubrovnik, Yugosl.	V	" "	8	4.2	"
Off-Shore Area					
W. Med.	II-III	'70		8.1	Kautsky (1977)
"	II-III	'74	4	5.6	"
N.W. Med.	II	'75-'78	7	5.0	Fukai et al. (1980)
S.W. Med.	III	'75	3	4.4	"
Tyrrhenian	IV	'75-'77	6	4.4	"
Adriatic	V	'77	3	5.0	"
Ionian	VI	'77	6	5.0	"
S. Levantine	X	'77	6	5.0	"
Whole Med.			33	4.6	"

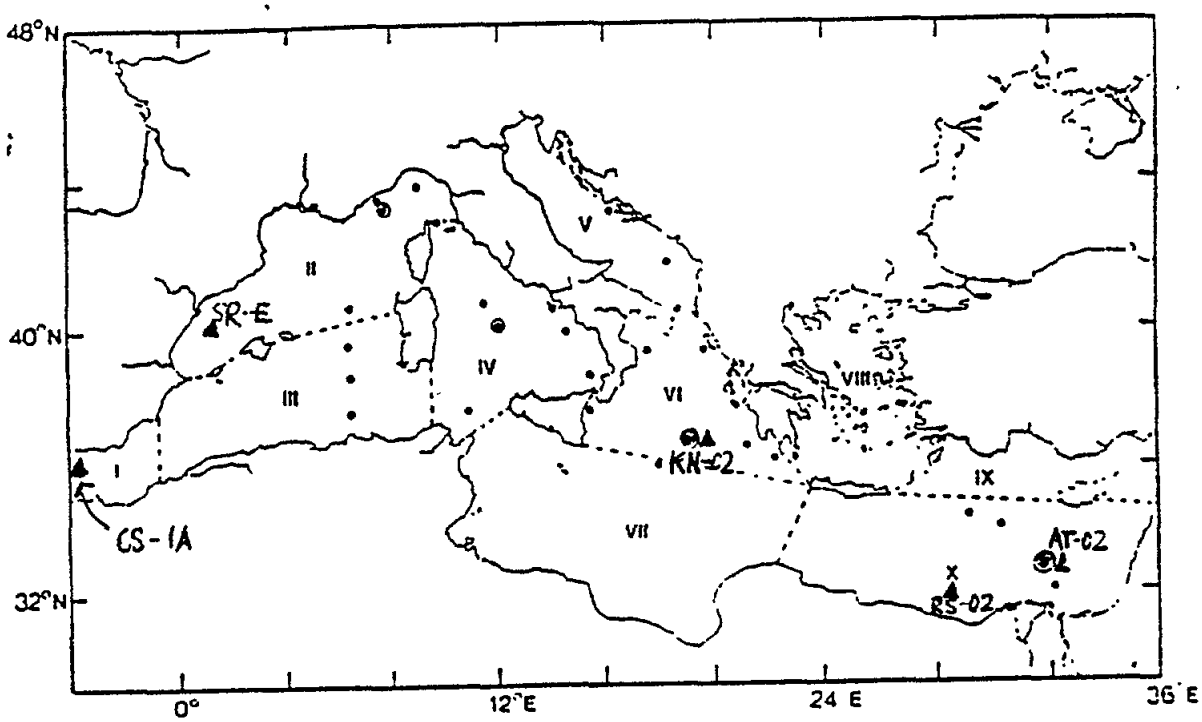


Figure 3: Sampling stations for cesium-137 and plutonium-239 measurements in the Mediterranean Sea (• surface station for Cs-137; ⊙ vertical profile station for Cs-137; ▲ vertical profile station for Pu-239 and Am-141).

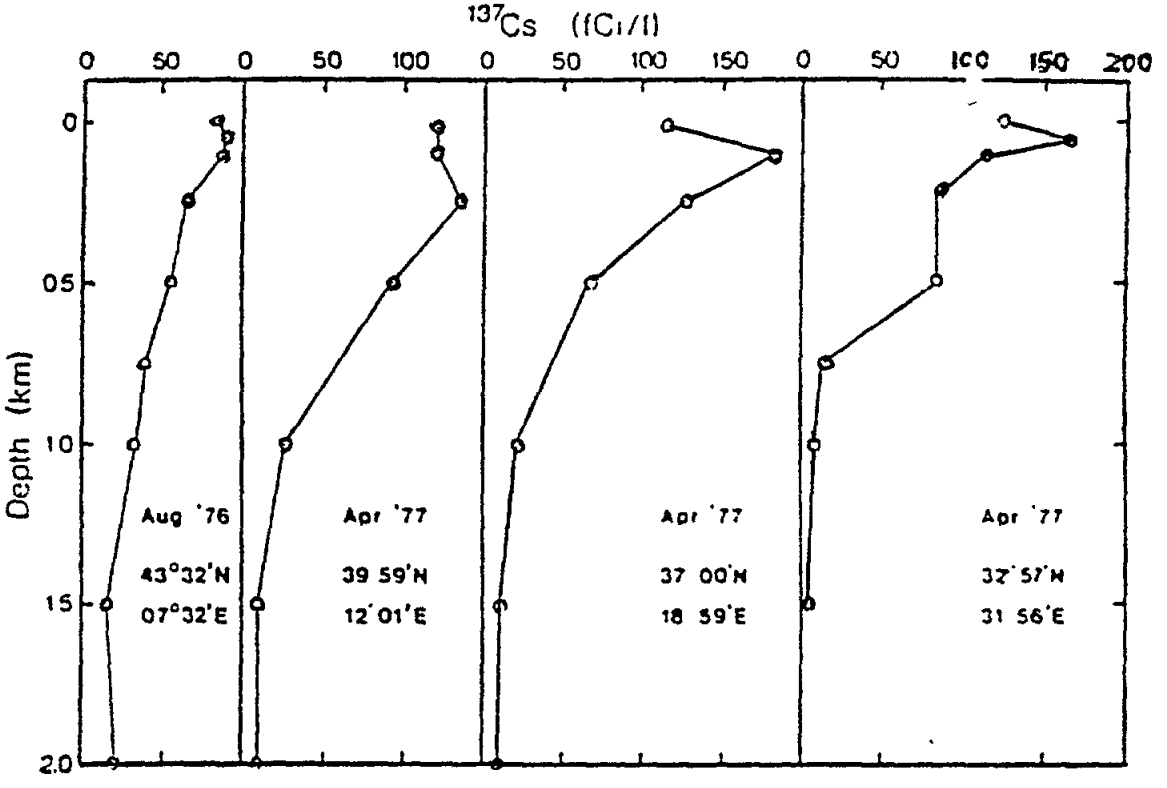


Figure 4: Vertical distributions of cesium-137 in the Mediterranean Sea (1 fCi = 37 μ Bq = 0.037 mBq).

The number of data available for the Pu-239 measurements on Mediterranean sea water is much smaller than that for Cs-137. The vertical distributions of Pu-239 and Am-241 (americium-241) measured at various locations in the Mediterranean Sea are illustrated in Fig. 5. The locations of these profiles are indicated in Fig. 3. As shown in Fig. 5, concentrations of Pu-239 and Am-241 in the Mediterranean surface waters are respectively around 0.4 mBq l^{-1} and less than 4 µBq l^{-1}. All profiles presented in Fig. 5 show sub-surface concentration maxima for both Pu-239 and Am-241, the depths of which are around 250m for Pu-239 and below 500m for Am-241. The cause of the formation of these concentration maxima was attributed to sinking of particulate matter, contrary to that for Cs-137, to which vertical water movements were considered to be responsible. The differences of the depths of the concentration maxima found between Pu-239 and Am-241 were believed to be related to the difference in their chemical characteristics, which differentiate them in particle formation processes in situ. On the basis of its vertical distribution patterns it was concluded that the most part of Pu-239 introduced into the Mediterranean up to 1986 by fallout remained in the water column (Fukai et al., 1982).

6.2.2. Levels in marine sediments: The radioactivity concentration levels of Cs-137 measured for surface sediments of the Mediterranean Sea by various authors up to 1986 are given in Table XII. Excluding a series of values obtained in the Rhone Estuary, the values presented in Table XII for both coastal and off-shore areas are in a relative narrow range of 2-10 Bq Cs-137 kg^{-1} dry, with an average value of around 6 Bq kg^{-1} dry. Although observations demonstrated that higher levels of Cs-137 tend to be found in areas under the influence of rivers where bathymetric belts (depths 50-100m) characterized by maximum Cs-137 concentrations exist (Delfanti et al., 1988), it seems reasonable to consider 6 Bq Cs-137 kg^{-1} dry as a base-line level for Mediterranean surface sediments in 1985, with an allowance factor of 2 in either side. The higher values found in the Rhone Estuary are considered to represent the discharge from the Rhone, which was redistributed under the influence of near-bottom currents.

The number of data available for the Pu-239 measurements on Mediterranean sediments is much smaller than that for Cs-137, although a series of observations have been conducted in the coastal zones of Spain and Italy. The data obtained on core samples collected from the Gulf of Vera, Spain, demonstrated that the Pu-239 concentrations in surface sediments are around 3 Bq kg^{-1} with an average integrated concentration for the cores of 280 Bq m^{-2} (Gasco Leonarte et al., 1988). The other series of measurements were made on core samples collected from the bay of La Spezia and the Gaeta Area along the Italian coast. The surface and integrated concentrations of Pu-239 for these cores were respectively 0.8 Bq kg^{-1} dry and 700 Bq m^{-2} for those from Bay of La Spezia and 1.5 Bq kg^{-1} dry and 160 Bq m^{-2} for those from the Gaeta Area (Delfanti et al., 1988). Although it has been observed that the concentrations of Pu-239 in surface sediments vary rather widely depending on the influences of river run-offs, near-shore bottom currents, etc., a range of 1-3 Bq Pu-239 kg^{-1} dry is considered reasonable as the base-line range of the Pu-239 concentration for surface sediments in the coastal Mediterranean Sea. The data on core samples taken from off-shore stations in the Mediterranean Sea demonstrated that an average integrated concentration of Pu-239 in deep-sea Mediterranean sediments is around 7 Bq m^{-2} with a range of 2-15 Bq m^{-2} (Livingston et al., 1977; Livingston et al., 1978).

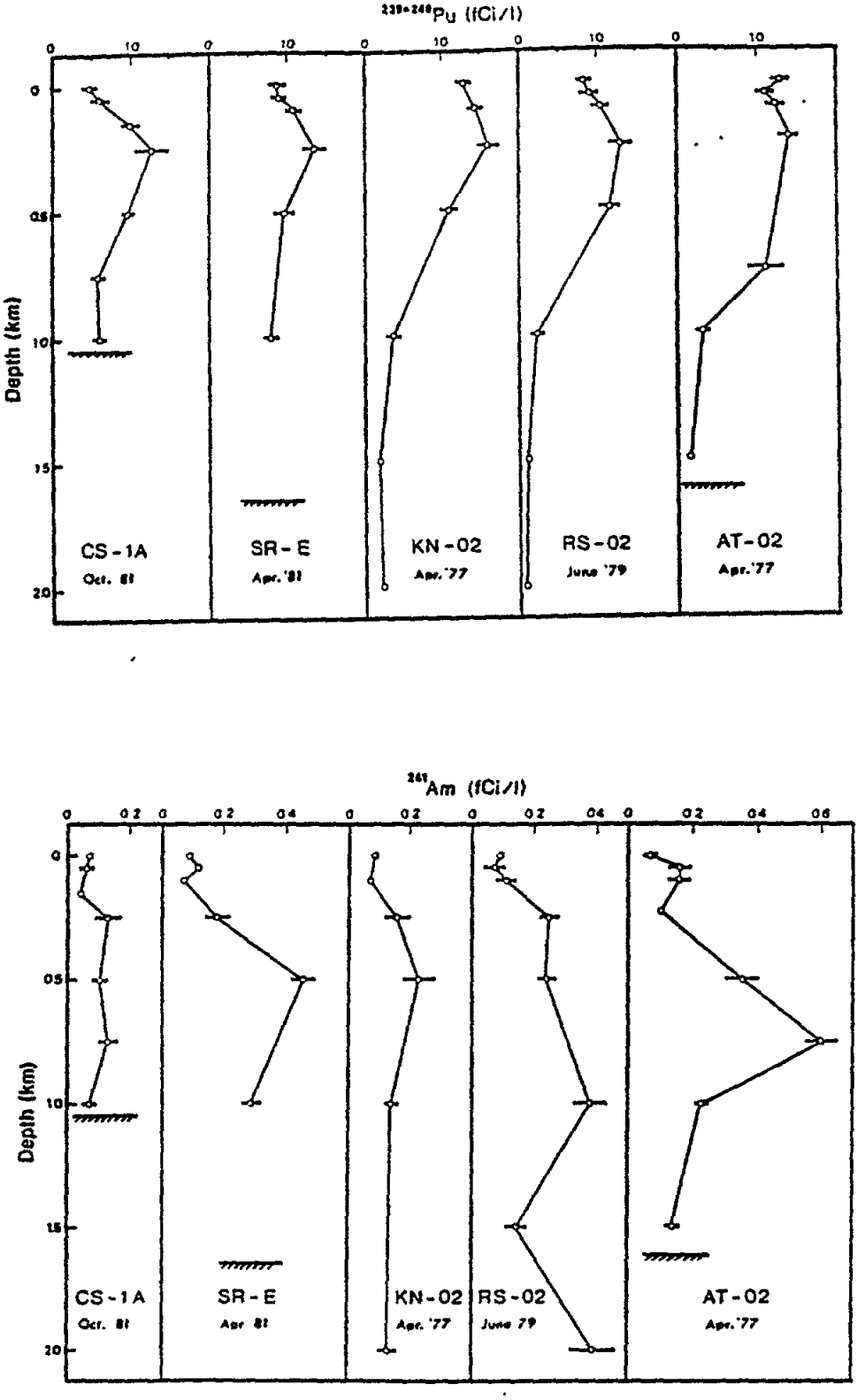


Figure 5: Vertical distributions of plutonium-239 and americium-241 in the Mediterranean Sea (1 fCi = 37 μ Bq = 0.037 mBq).

Table XII

Levels of cesium-137 in surface sediments of the Mediterranean Sea
during the pre-Chernobyl period

Location	Area No.	Time	No. of Data	Cs-137 Level Bq kg^{-1} dry	Literature
Coastal Zone					
French Coast	II	Feb'86	2	5	Calmet & Granby (1988)
Rhone Estuary	"	"	4	40-170	"
Corsican Coast	"	'85	6	12	"
Montalto di Castro, Italy	IV	'83		2-10	Toccafondi et al. (1988)
La Spezia, Italy	"	'82-'85	21	8	Delfanti et al. (1988)
Sardinian Coast	"	"	16	4	"
Latina Area, Italy	"	"	14	5	"
Gaeta Area, Italy	"	"	23	10	"
Apulia Region, Italy	V-VI	"	36	6	"
Off-Shore Area					
Gibraltar	I	'81-'82	5	7	Calmet & Granby (1988)
N.W. Med.	II	"	3	3	"
S.W. Med.	III	"	6	10	"

6.2.3. Levels in marine biota: Due to very low levels of Cs-137 and Pu-239 in marine biota found in the region the measurements of these radionuclides on biological matrices have not been carried out extensively in the Mediterranean Region. Nevertheless, the measurements of these radionuclides on a few species of seaweed (Codium tomentosum, Corallina mediterranea, Dictyota dichotoma, etc.) as well as mussels (Mytilus sp.) were conducted repeatedly at Monaco. While the concentration levels of Cs-137 measured were 0.5-1 Bq kg^{-1} dry and around 1 Bq kg^{-1} dry respectively for the seaweeds and mussels (Whitehead et al., 1987; Holm et al., 1988a), the concentration levels of 0.1 - 0.8 Bq kg^{-1} dry and 0.02 -0.1 Bq kg^{-1} dry were found for Pu-239 respectively in the above two

groups of marine organisms (Ballestra, 1980; Holm et al., 1988b).

In order to facilitate the comparisons of concentration levels for these radionuclides between sea water, marine sediments and selected groups of marine organisms approximate concentration ranges of Cs-137 and Pu-239 expected to be found in these environmental compartments in the Mediterranean are given in Fig. 6. The concentration ranges for sea water and marine sediments presented in the figure were derived on the basis of the data presented previously, while the concentration ranges for different groups of organisms were estimated from the concentration levels of these radionuclides in sea water by utilizing the recommended biological concentration factors (IAEA, 1985).

6.2.4. Total Mediterranean inventories up to 1986: If one could know the concentration distributions of a radionuclide in sea water, marine sediments and marine biota for a specific region of the sea, it would be possible, in principle, to calculate the total inventory of the radionuclide in the sea region concerned by applying the volume or mass of the environmental compartments containing the radionuclide. The precision of this sort of calculation depends on the availability of detailed data representative for the concentration distributions of the radionuclide in each environmental compartment. Although the data at hand had not been sufficient for precise inventory computations, an attempt was made to estimate the total inventories of Cs-137 and Pu-239 in the Mediterranean marine environment based on existing data (Holm et al., 1988b).

The average vertical distributions of Cs-137 and Pu-239 in the Mediterranean water column estimated on the basis of the data so far obtained (cf. Section 6.2.1) are given in Fig. 7. Based on these average distributions, the integrated concentrations of Cs-137 and Pu-239 for a Mediterranean water column were estimated to be 3.8 ± 1.2 kBq m^{-2} for Cs-137 and 55 ± 6 Bq m^{-2} for Pu-239 in 1985. Taking into account varying depths of individual water columns the above integrated concentrations lead to the total water column inventory of 10.2 ± 1.2 PBq for Cs-137 and 0.16 ± 0.02 PBq for Pu-239 covering the entire Mediterranean Sea. Similarly, based on the average integrated concentrations for sediment cores respectively of 162 Bq m^{-2} for Cs-137 and of 7 Bq m^{-2} for Pu-239, the total sediment inventory was estimated to be 0.5 ± 0.2 PBq for Cs-137 and 0.02 ± 0.01 PBq for Pu-239 covering the entire Mediterranean Sea. Due to the lack of total biomass data for the whole Mediterranean as well as measurement data on various marine organisms, the inventory for the Mediterranean biomass could not be estimated. The results of these estimations are summarized in Table XIII.

Table XIII

Inventories of cesium-137 and plutonium-239
in the Mediterranean marine environment (1985)

Inventory	Cs-137 PBq *	Pu-239 PBq *
Water column Sediment Biomass	10.2 ± 1.2 0.5 ± 0.2 ?	0.16 ± 0.02 0.02 ± 0.01 ?
Total	11 ± 1	0.18 ± 0.02

* PBq = 10^{15} Bq.

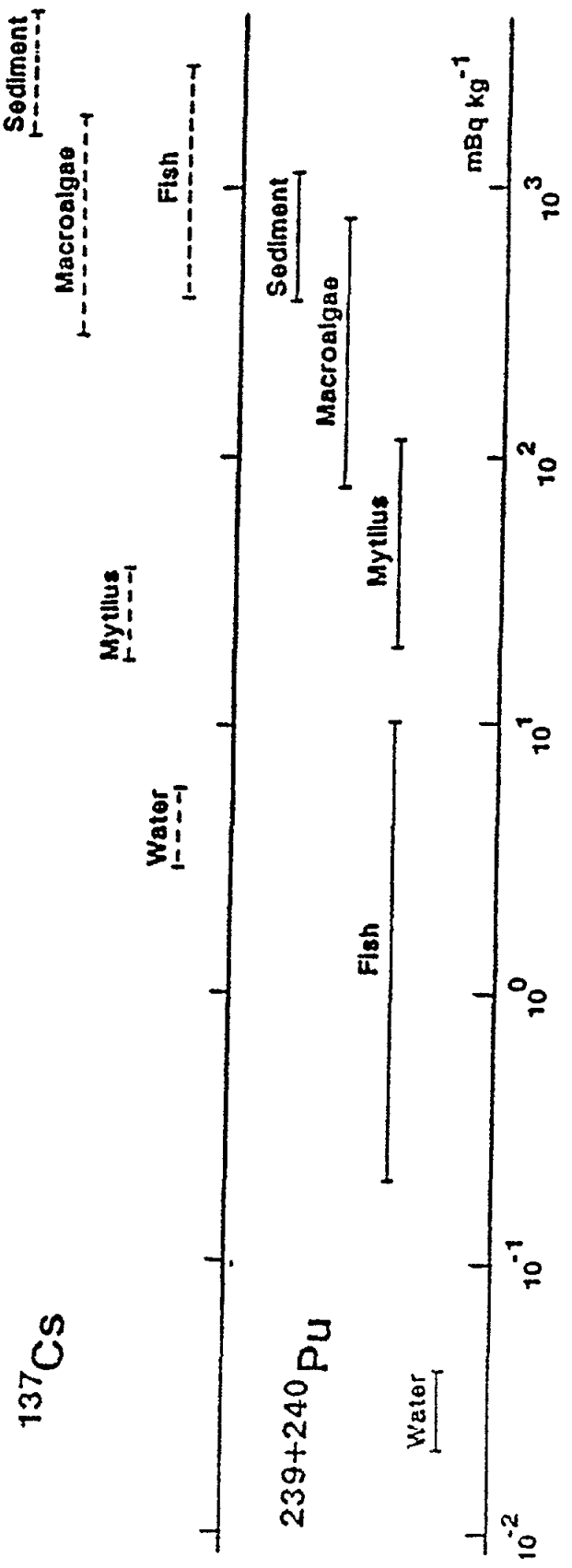


Figure 6: Ranges of activity concentrations for cesium-137 and plutonium-239 in various Mediterranean environmental compartments.

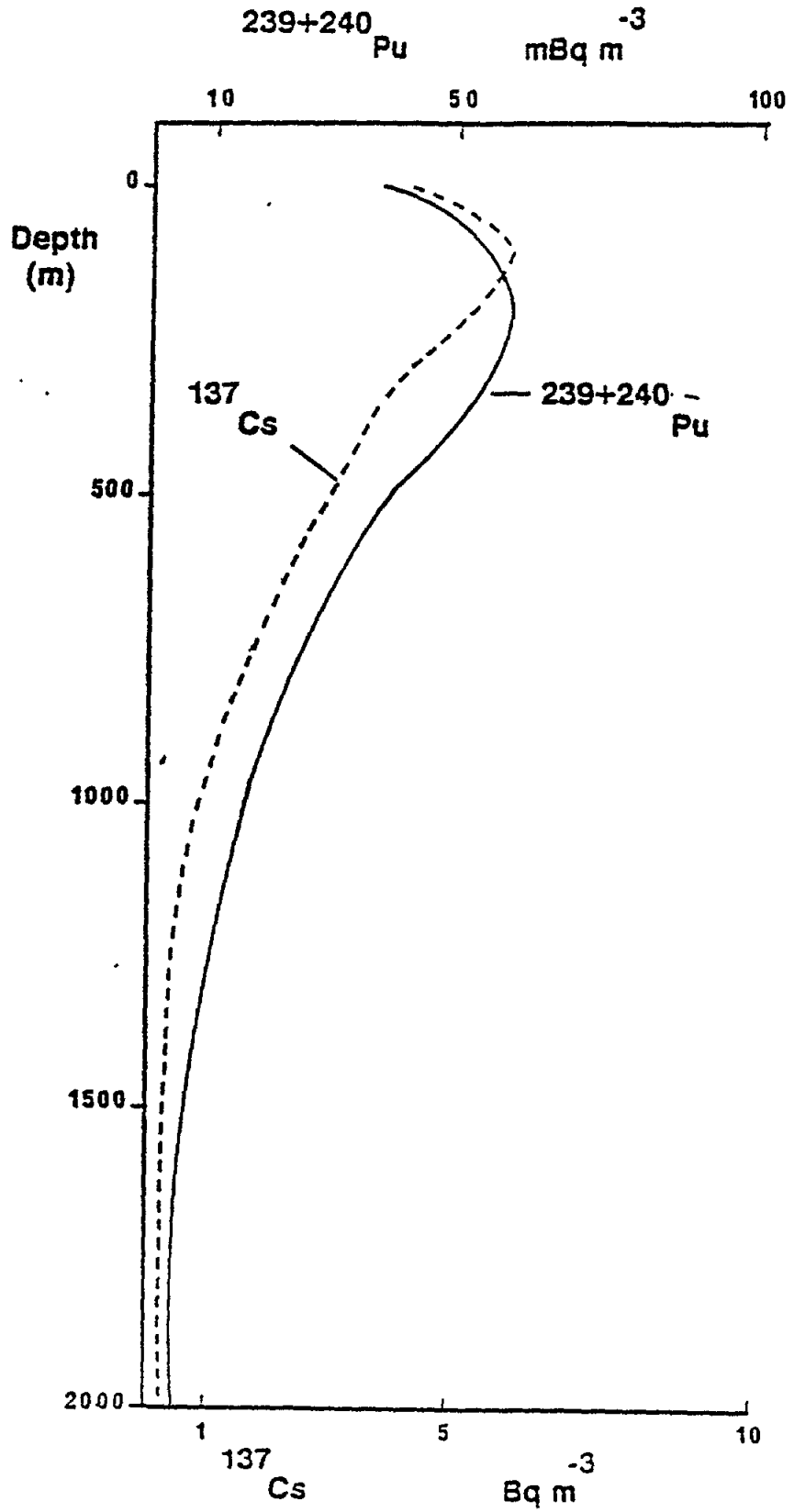


Figure 7: The idealized average vertical distributions of cesium-137 and plutonium-239 in the Mediterranean water column (based on pre-1982 data).

Comparisons of the total inventories of the radionuclides given in Table XIII with the values of total inputs of these radionuclides into the Mediterranean Sea (Table VII) indicate that the agreements between the inputs and the existing inventories are surprisingly good. Considering the rather large errors associated with these estimates, however, excellent numerical agreements of these values are likely to be coincidental. Nevertheless, the agreements even in the order of magnitude suggest that the general approach adopted for these estimations are not largely in error. The accuracy of these estimations will be substantially improved when more data will be made available for a better coverage of the entire Mediterranean marine environment.

6.3. Levels in the post-Chernobyl period

Although more than 30 radionuclides were identified in Monaco by gamma-spectrometry of the Chernobyl fallout immediately after the accident in early May 1986 (Ballestra et al., 1987), most of them decayed quickly thereafter and a few months later only longer-lived radionuclides, such as ruthenium-106 (half-life: 372d), silver-110m (250d), tellurium-129m (109d) cesium-134 (2.06a), cesium-137 (30.2a), etc., were measurable in the Mediterranean marine environment. Taking into account its long half-life and its radioactivity concentration levels found in the fallout, Cs-137 is considered to be the most important radionuclide delivered by the Chernobyl fallout for the assessment of marine pollution by artificial radionuclides. For this reason the measurements of Cs-137 in various environmental compartments have been conducted on a world-wide scale. Although these measurements in the Mediterranean Region have been limited in number and are still being continued, the data published so far are reviewed and significant findings are summarized below.

Levels of Cs-137 measured on surface sea waters of the Mediterranean Sea by various authors after the Chernobyl accident are summarized in Table XIV. Comparing the values in Table XIV with those in Table XI which gives the Cs-137 concentrations in the pre-Chernobyl period, one can observe that the levels of Cs-137 in Mediterranean coastal waters increased by one or two orders of magnitude immediately after the Chernobyl accident, but they decreased again to lower levels within a relatively short time probably due to dispersion by coastal water movements. While the levels of Cs-137 in near-shore waters along the French coast, except for those in the Rhone Estuary, decreased to only a few times of the base-line value 4 mBq l^{-1} towards the end of 1986, the levels along the Italian coast remained at higher levels, approximately 5 times of the base-line value, in the same period. Since the Cs-137 levels in near-shore surface waters are considered to vary constantly within rather wide ranges under the influence of precipitation, land run-offs and coastal currents, not too much significance should be attached to the quantitative differences found between the measured Cs-137 levels. It may be roughly estimated, however, that the higher Cs-137 concentrations found in the Mediterranean near-shore waters after the Chernobyl accident should have decreased to levels approximately 50% higher than the base-line concentration towards the end of 1989. The level to be found in a specific location is considered to be very dependent on local conditions, especially on run-offs of accumulated fallout radionuclides in soil after rains. The result of measurements in time-series of vertical distribution of Cs-137 conducted in the western off-shore Mediterranean are given in Fig. 8 (Calmet & Granby, 1988). The figure demonstrates the evolution with time of penetration of Cs-137 delivered by the Chernobyl fallout from the surface into the deeper layers of the water column.

The levels of pollution in water, air, soil and nutrients taken by the relevant authorities in Syria at the time of the Chernobyl disaster were found to be "very" low and did not expose the population to dosages exceeding the accepted levels.

Levels of Cs-137 measured on surface sediments of the Mediterranean Sea by various authors after the Chernobyl accident are summarized in Table XV. Comparing the values in Table

Table XIV

Levels of cesium-137 in surface waters of the Mediterranean Sea
during the post-Chernobyl period

Location	Area No.	Time	No. of data	Cs-137 level mBq l^{-1}	Literature
French Coast	II	Nov'86	10	5.7	Calmet & Granby (1988)
		Sep'87	11	3.6	"
Off Rhone Estuary	II	Sep'86	13	10-25	"
Golfe du Lion	II	Sep'86	22	2-12	"
		Dec'86	18	2-8	"
Monaco	II	May'86	3	65-510	Holm et al. (1988a)
		June'86	2	25	"
La Spezia, Italy	IV	May'86	3	405	Delfanti & Papucci (1988)
		June'86	1	58	"
		Oct'86	2	14	"
Naples, Italy	IV	Sep'86	1	24	"
		Oct'87	1	22	"
Trieste, Italy	V	June'86	1	99	"
		Sep'86	1	65	"
Venice, Italy	V	Oct'86	1	31	"
Adriatic	V	Summer'86	3	51	Korun et al. (1988)
Rovinj, Yugoslavia	V	May'86	1	100	Bauman (1990)
		Apr.'87	1	63	"
		May'88	1	<15	"
		Apr.'89	1	<5.5	"
Rijeka, Yugoslavia		May'86	1	74	"
		Apr.'87	1	200	"
		May'88	1	17	"
		Apr.'89	1	<5.5	"
Split, Yugoslavia		May'86	1	490	"
		Apr.'87	1	240	"
		May'88	1	24	"
		Apr.'89	1	<6.4	"
Dubrovnik, Yugoslav.		May'86	1	150	"
		Apr.'87	1	350	"
		May'88	1	<15	"
		Apr.'89	1	<6.4	"

Table XV

Levels of cesium-137 in surface sediments of the Mediterranean Sea
during the post-Chernobyl period

Location	Area No.	Time	No. of data	Cs-137 level Bq kg^{-1} dry	Literature
French Coast	II	Nov.'86	6	20	Calmet & Granby (1988)
Rhone Estuary	"	"	1	390	"
Corsican Coast	"	Sep'86	4	28	"
		July'87	4	22	
La Spezia, Italy	IV	June'86	1	5*	Delfanti & Papucci (1988)
		Oct.'86	2	7*	"
Naples, Italy	"	Sep.'86	1	4*	"
		Oct.'86	1	5*	"
Venice, Italy	V	June'86	1	7*	"
		Oct.'86	1	14*	"
Taranto, Italy	VI	Sep'86	1	1*	"
Adriatic	V	Summer'86	6	8	Korun et al. (1988)

* Levels are expressed in Bq kg^{-1} wet

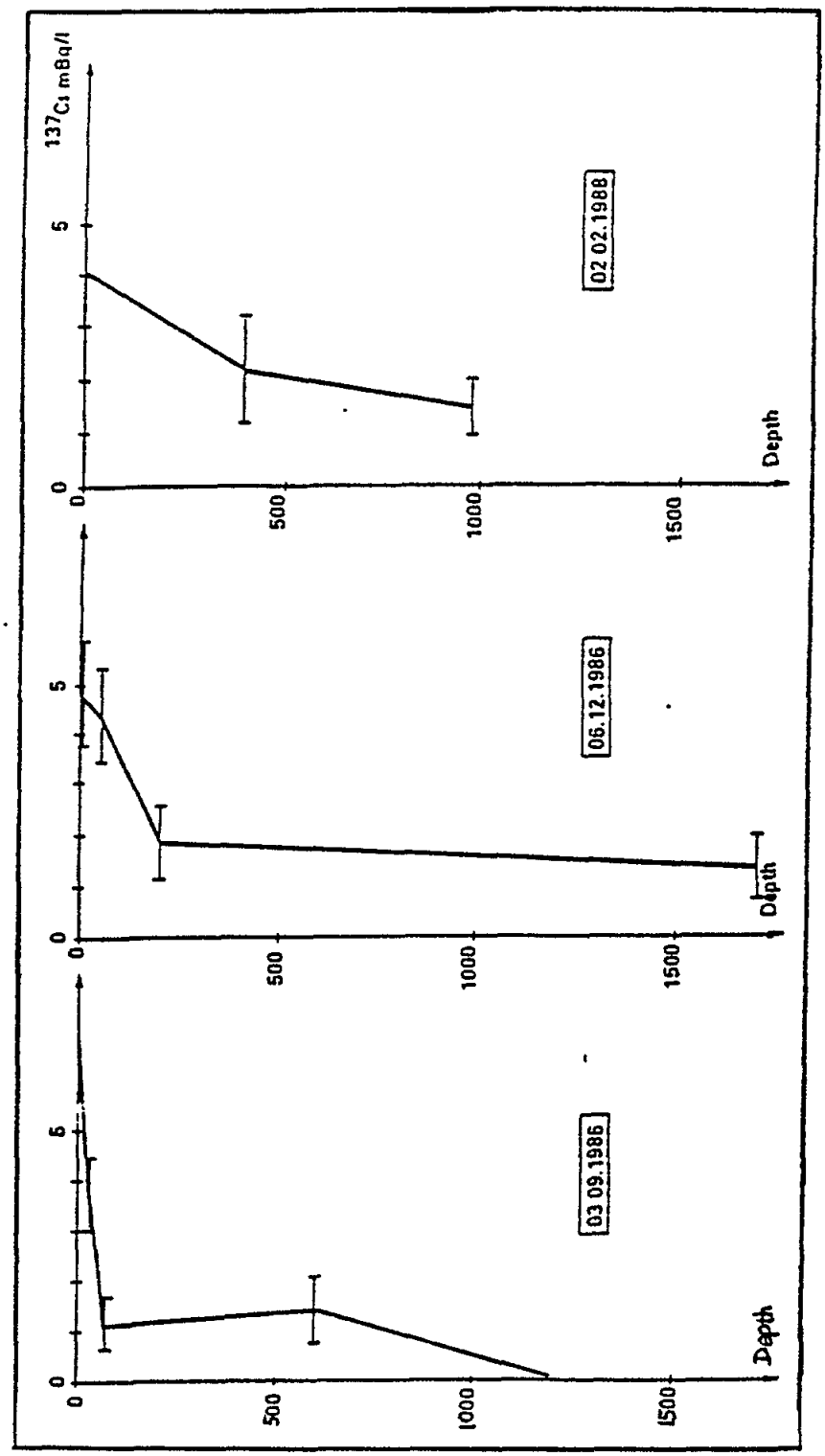


Figure 8: Evolution with time of the vertical distribution of cesium-137 after the Chernobyl accident at a station in the Gulf of Lion (42° 19'N; 04° 59.5' E).

XV with those in Table XII, which gives the Cs-137 concentrations in the pre-Chernobyl period, one can remark that the levels increased significantly by a factor of 2 to 4, along the French and Corsican coasts after the Chernobyl accident, while the changes in the levels were not evident along the Italian coast. Even considering the fact that the Italian data were expressed in a wet weight basis, local variability masks the changes which might have happened between the two periods, although measurable Cs-134 (originated from the Chernobyl fallout) was present in almost all sediments measured. Since the stabilization of Cs-137 levels in coastal sediments is considered to take some time, it may be too early to draw any general conclusions at present as to the levels of Cs-137 in these sediments.

Levels of Cs-137 in different seaweed species measured at Monaco increased from 0.5-1 Bq kg^{-1} dry to 10-50 Bq kg^{-1} dry after the Chernobyl accident with biological half-lives of 70-350 days (Holm et al., 1988a). The measurements at Toulon, France on a seaweed species (Ulva sp.) showed the level of 96 Bq kg^{-1} dry with a biological half-life of approximately 30 days (Calmet & Granby, 1988). In mussels an increase of the Cs-137 level from 1 Bq kg^{-1} dry to 30 Bq kg^{-1} dry was observed at Monaco with a biological half-life of about 20 days (Whitehead et al., 1987). Similar observations were made on mussels at La Spezia, Italy, reporting a value of 7.5 Bq Cs-137 kg^{-1} wet with two components of biological half-life of 2 days and 63 days (Delfanti & Papucci, 1988). These biological half-lives measured are much shorter than the physical half-life of Cs-137, 30.2 years and, thus, the concentrations of Cs-137 in marine organisms living in the Mediterranean marine environment are considered to have decreased already to levels close to those in the pre-Chernobyl period towards the end of 1989.

On the basis of the amounts of deposition of various radionuclides, especially those of Cs-137, resulting from the Chernobyl fallout at several locations in the Mediterranean region, it was estimated that the Chernobyl fallout increased the Cs-137 deposition approximately 25-40% in addition to the amounts existing in the region up to 1986 (Section 4.2.5.). As has already been stated, the deposition of radionuclides through the Chernobyl fallout was very heterogeneous depending on the trajectories of high radioactivity plumes, although the radioactivity levels were, generally speaking, much higher in the northern Mediterranean region than those in the southern part. This heterogeneity of the deposition makes it necessary to collect much more data covering different parts of the Mediterranean Sea for reasonably estimating the additional Mediterranean inventory of radionuclides resulting from the Chernobyl fallout. Considering the coverage at present of the data on the levels of radionuclides in various environmental compartments for the entire Mediterranean Sea, it seems that such inventory estimations are not yet warranted.

7. Risk Assessment

7.1. Effects on marine organisms

Radiological effects on living organisms caused by the introduction of radionuclides into the marine environment are primarily a problem of public health. The potential effects of increased radiation exposure on marine organisms, however, should also be assessed with respect to environmental consequences of such exposure on populations of marine living resources which are being utilized as human food. In the field of radiobiology the effects of ionizing radiation on various organisms have been and still are one of the major topics with practical significances and extensive experimental and model studies on this topic have been conducted for several decades by using experimental organisms belonging to widely varying phyla from bacteria to mammals, including various species of marine organisms. Although the results of many of these studies in the early period did not have any relevance to the situations found in environmental contamination

with radionuclides, general tendencies were observed that the degree of response to irradiation appeared to be inversely correlated with organism complexity despite the wide variations within phyla. Thus, the greatest sensitivity was found in actively proliferating cell systems such as blood cells, the gonads, skin, the gut lining of fish, etc. while the early embryonic stages were also identified as being particularly radiosensitive.

As to specifically aquatic organisms and ecosystems, an expert panel was convened by the IAEA in order to consider the effects of ionizing radiation (IAEA, 1976). Detailed assessments conducted by this panel on the basis of data obtained from laboratory experiments and field studies, have shown that there are only isolated situations where radiation dose rates from artificial radionuclides introduced into the marine environment as the result of fallout and waste disposal significantly exceed the natural background. Taking into account the applicability of the data obtained in these studies to realistic environmental situations, special attention in the assessments was drawn to studies employing a chronic low dose exposure regime and to those dealing with the effects of radionuclides in the ambient water on developing fish eggs. Earlier indications of an extreme radiosensitivity of developing fish eggs were definitively discounted by these assessments and it was concluded that very minor effects might be noted at dose rates approximately 5 times the maximum predicted to occur in one of the specific contaminated marine environments. The lowest dose rate cited was approximately 0.4m Sv h^{-1}. Since in natural aquatic systems, the population rather than the individual is more important for maintaining balanced ecosystems, an attempt was made to assess the consequences for populations of the radiation effects on individuals. At the dose rates prevailing in contaminated environment it was concluded that it would not be possible to detect any radiation-induced effect separate from that generated by other environmental factors, although it was considered that the minimum dose rates causing significant effects on populations are at least one order of magnitude higher than those causing the minor effects on individuals.

It has been widely recognized at present that the most heavily contaminated marine environment with artificial radionuclides in the world is to be found in the vicinity of the waste effluent discharge point of the nuclear fuel reprocessing plant at Sellafield, U.K., in the Irish Sea. Because of the elevated levels of artificial radionuclides in the marine environment around this area extensive measurements of radiation doses received by various marine organisms were carried out in this connection and their effects on these organisms were studied in detail. These studies demonstrated that the beta and gamma irradiation dose rates to developing fish embryos in sea water containing levels of artificial radionuclides similar to those in sea water around Sellafield, are less than those from natural backgrounds due to potassium-40 (Woodhead, 1970) and that the alpha dose rates from the presence of Pu-239 were also negligible due to the short hatching time of the eggs (. 2 weeks) (Hetherington et al., 1976). The irradiation dose rates to the gonads of adult plaice as well as other marine organisms such as molluscs, crustaceans, etc. were also estimated, showing that internal and external dose rates of these organisms are in the order of 10^{-2} µrad h^{-1}, although they vary considerably depending on biological and environmental conditions (Woodhead, 1973; Woodhead, 1974). On the whole, it was concluded that the radiation dose rates received by marine organisms living in sea water around Sellafield are at least one order of magnitude, more probably two to three orders of magnitude below those which would be expected to elicit any effect.

Levels of artificial radionuclides found in the Mediterranean marine environment are at least three orders of magnitude, normally four orders of magnitude lower than those found in the marine environment around Sellafield. Even considering possibilities of unusual local accumulation of artificial radionuclides under special circumstances, the radiation effects of artificial radionuclides introduced in the Mediterranean environment on living marine organisms could be completely disregarded at present.

7.2. Human exposure

Significant quantitative assessments of increased risks with the increased radiation dose received by man as the result of introduction of artificial radionuclides into the Mediterranean marine environment are so complicated that, it seems fair to state, such assessments are hardly possible on the basis of the present knowledge at hand. Nevertheless, some ideas on the degree of risk increases due to the introduction of artificial radionuclides into the Mediterranean Sea may be obtained by comparing the levels of artificial radionuclides in the Mediterranean marine environment with those in specific contaminated areas where risk assessments have been deemed necessary.

Since the biological consequences of exposure to ionizing radiation depends on the type of radiation and the organ of the body that is irradiated, it is necessary to take these factors into consideration when one tries to relate irradiation doses to effects of man. Thus, the effective dose equivalent expressed in Sieverts (Sv) has been defined along this sense. Extensive data were reviewed by the United Nations Scientific Committee on the Effects of Atomic Radiation and these data showed that the harm directly attributable to radiation results at an effective dose equivalent above 0.5 Sv received within a short time (UNSCEAR, 1982). For doses much less than this level, the harm has a low probability, but it is assumed that there is a risk of harm that increases in direct proportion to the dose received. It is possible that at very low doses there is no risk at all, i.e., there is a threshold below which the body is able to repair any damage to cells from ionizing radiation. However, it is generally thought that, even if such a threshold exists, the natural background radiation to which man is exposed, typically 0.1 Sv per lifetime or 2×10^{-3} Sv per year, already exceeds this threshold and therefore, in practice, there is a risk of harm from non-natural sources of radiation that is directly proportional to the incremental dose (Brewers & Garrett, 1987).

In order to illustrate the contents of human radiation exposure relative contributions of natural and man-made radiation sources to human radiation doses are given in Fig. 9 (IAEA, 1988b). As shown in the figure approximately three quarters of a human radiation dose comes from natural background, while doses resulted from nuclear weapon testing and nuclear power production contribute only fractions of the percentage to the total dose received by an average man. The radiation risk is generally thought to correlate with the effective dose equivalent in such a way that one unit increase of the effective dose equivalent corresponds to a chance of 10^{-2} of developing fatal cancer and a chance of 6×10^{3} of genetic damage that would affect offspring (UNSCEAR, 1982) thus, the aggregate risk may be rounded off to about 2×10^{-2} Sv^{1}. On this basis, each individual faces a lifetime risk of severe harm of about one in 500 due to naturally occurring background radiation. It should be emphasized, however, that this harm is wholly probabilistic and will not be correlated with any specific cause. The risk factor of about 2×10^{-2} Sv^{1} is not known to any great certainty and could be wrong by a factor of about three in either direction (Brewer & Garrett, 1987).

If one accepts the above dose-effect relationship, it follows that one case in 10^{5} of severe harm may have been caused by weapons testing (taking a ratio of the contribution of the total human dose by weapon testing to that by the natural background). Since the major part of human radiation exposure is considered to be normally due to terrestrial origin, the part resulted from radioactive contamination of the marine environment is not likely to exceed 10% of the weapon testing contribution. Thus, the increased radiation exposure risk due to the introduction of artificial radionuclides into the Mediterranean marine environment by weapon testing may correspond approximately to the increase of one case of severe harm in 10^{6} during the pre-Chernobyl period. As has been discussed in detail in Section 4, the introduction of artificial radionuclides through the other routes into the Mediterranean Sea contributes much less to the human radiation exposure.

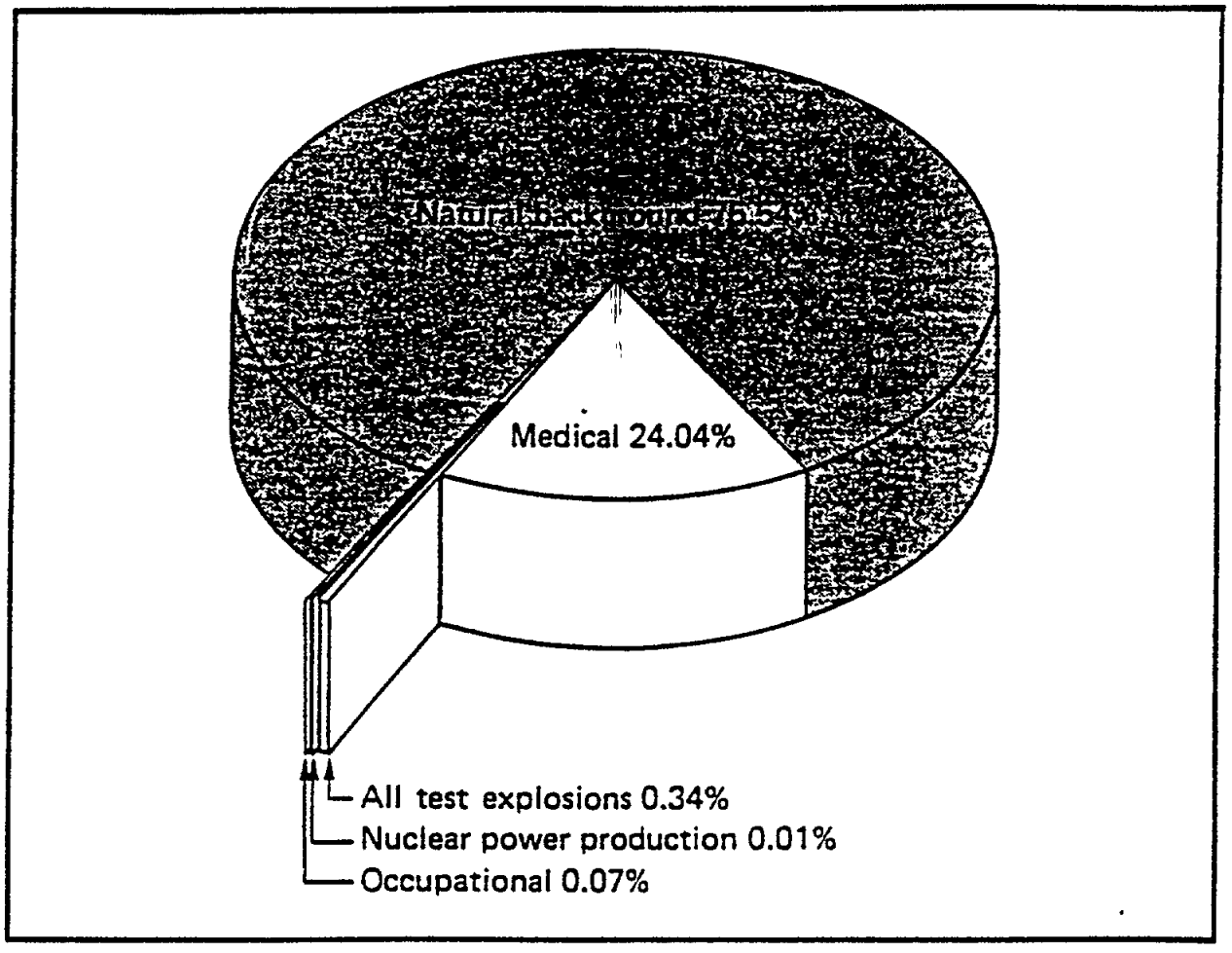


Figure 9: Relative contributions of natural and artificial sources of radiation to human radiation doses.

On the other hand, it was observed that the effective dose equivalents received by sea-food consumers in local fishing communities around Sellafield, U.K. were an order of magnitude less than the dose limit of 5m Sv a^{-1}. Although concentration of radionuclides in the environment cannot be directly correlated to the effective dose equivalent, at least three orders of magnitude levels lower of artificial radionuclides in the Mediterranean marine environment than those in the vicinity of Sellafield, suggest that effective dose equivalents received by the Mediterranean inhabitants due to sea-food consumption may be roughly three orders of magnitude lower. The risk increase was thus estimated to be approximately 5 cases of severe harm in 10^7. These estimated risk increases are admittedly crude and should be regarded only as an index of their magnitude.

In Fig. 10 the ranges of the effective dose equivalent commitments of different countries from the Chernobyl accident, based on preliminary data from UNSCEAR, are illustrated (IAEA, 1988a). The figure demonstrates that ranges vary considerably from less than 1 mSv to more than 10 mSv depending on geographical locations of the countries concerned. Among the Mediterranean countries the values are highest for Yugoslavia, followed by Greece, Italy, France, Turkey and Spain, in that order. These differences found are considered to be due to the complicated trajectories of the high radioactive plume over the Mediterranean region immediately after the accident (Fig. 2). Comparing these values of the effective dose equivalent commitments with annual effective dose equivalents from natural background 2.2 mSv a^{-1} and applying the risk factor 2×10^{-2} Sv^{-1}, it may be possible to obtain a general indication of increased risk from the Chernobyl accident for each Mediterranean country.

8. Conclusions on the Assessment

The assessments made on sources, inputs, biogeochemical behaviour and levels of artificial radionuclides occurring in the Mediterranean marine environment as well as their effects on marine organisms and man have led to the following conclusions:

(1) Judging from the data available for river run-offs of some radionuclides, radionuclide discharges from the Mediterranean nuclear fuel reprocessing facilities into the aquatic environment are estimated to be at least three or, more probably, four to five orders of magnitude lower than those made from the Sellafield plant, U.K., while the environmental discharge rates from nuclear power stations and research reactors operating in the Mediterranean Region are estimated to be about 5 TBq a^{-1} and less than 1 GBq a^{-1} respectively in terms of the Cs-137 discharge. Discharges from other types of point sources are considered negligible.

(2) Atmospheric, river and strait-exchange inputs of Cs-137 into the Mediterranean Sea by 1985 are estimated to be respectively 10 ± 2 PBq, 0.4 ± 0.1 PBq and 1.6 PBq, which add up to the total input of 12 ± 2 PBq in 1985 for the entire Mediterranean Sea (Table VII).

(3) Base-line levels of Cs-137 in Mediterranean surface sea water and surface coastal sediments are estimated to be respectively 3-4 mBq l^{-1} and around 6 Bq kg^{-1} dry in 1985. The total Mediterranean inventory of Cs-137 in 1985, 11 ± 1 PBq, estimated on the basis of data available, agrees well with the total input value up to 1986. Although the agreement may be rather coincidental, it is considered to indicate that the general approach adopted for estimating the input and inventory is not grossly in error.

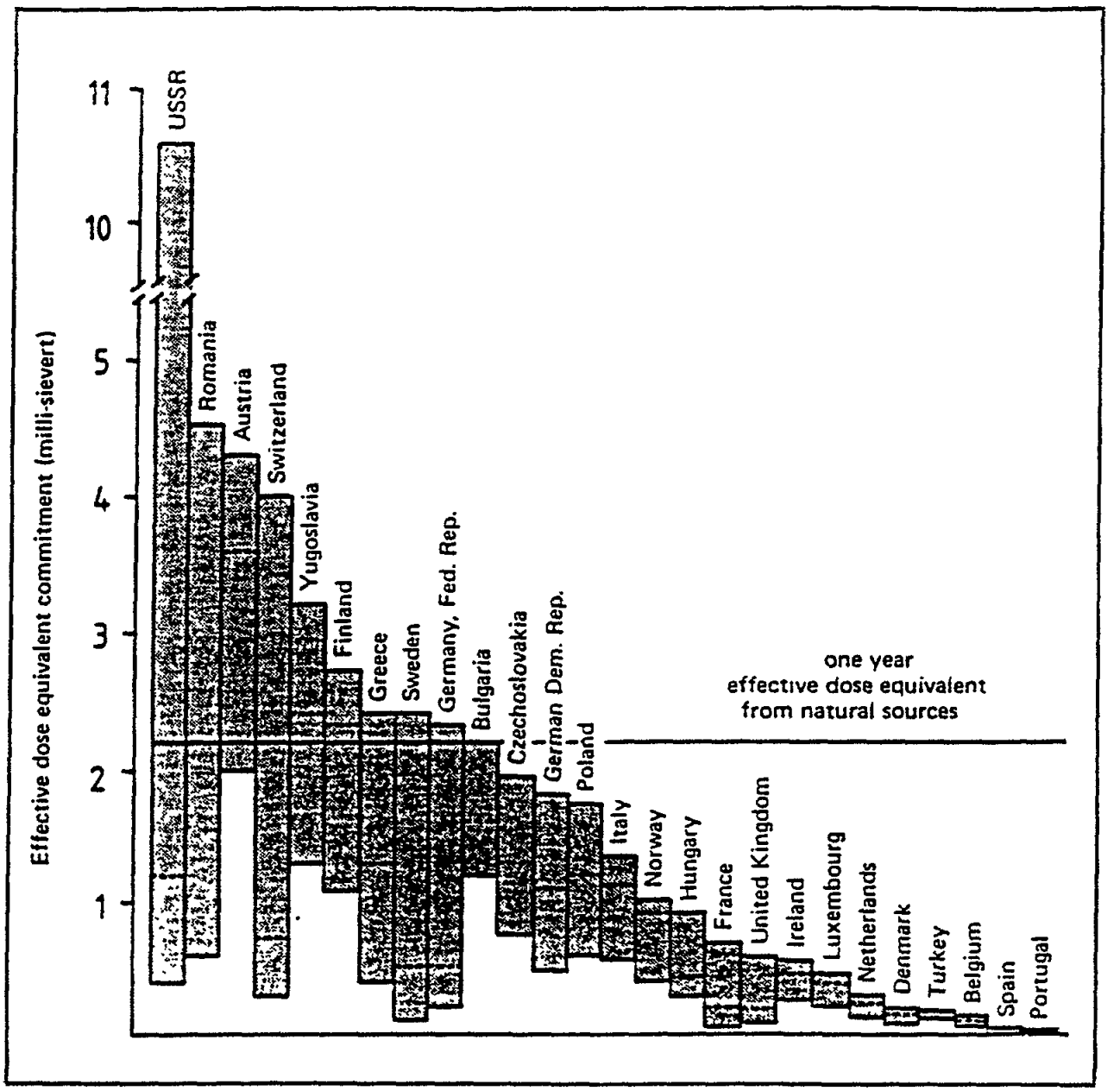


Figure 10: Ranges of effective dose equivalent commitments from the Chernobyl accident in different European countries.

(4) The recommended average values for sediment and biological concentration factors of selected radionuclides vary between 1 and 10^5 depending on particular radionuclide and are thought to be useful in considering radioprotection measures in the Mediterranean Region.

(5) On the basis of the amounts of deposition of various radionuclides, especially those of Cs-137, resulting from the Chernobyl fallout at several locations in the Mediterranean region, it was estimated that the Chernobyl fallout increased the Cs-137 deposition approximately 25-40% in addition to the amounts existing in the region up to 1986. The deposition of radionuclides through the Chernobyl fallout was very heterogeneous depending on the trajectories of high radioactivity plumes, although the radioactivity levels were, generally speaking, much higher in the northern Mediterranean region than those in the southern part.

(6) Significant increases in the Cs-137 levels by a factor of 2 to 4 were observed in surface coastal sediments along the French coast in 1986-87. The high Cs-137 levels found in various marine organisms after the Chernobyl accident are considered to have decreased to levels close to those in the pre-Chernobyl period towards the end of 1989.

(7) While the effects of the presence of artificial radionuclides in the Mediterranean Sea on living marine organisms are considered negligible, the increased radiation risk for man may correspond to one case of severe harm in 10^6, mainly due to artificial radionuclides introduced by fallout from nuclear weapon testing.

(8) Although concentration of radionuclides in the environment cannot be directly correlated to the effective dose equivalent, at least three orders of magnitude levels lower of artificial radionuclides in the Mediterranean marine environment than those in the vicinity of Sellafield, suggest that effective dose equivalents received by the Mediterranean inhabitants due to sea-food consumption may be roughly three orders of magnitude lower. The risk increase was thus estimated to be approximately 5 cases of severe harm in 10^7. These estimated risk increases are admittedly crude and should be regarded only as an index of their magnitude.

II. CONTROL MEASURES

9. Existing International and National Controls and Measures

9.1. International control measures

A special situation of the protection of man against hazards rising from radiation exposure, compared with the situations found for other environmental pollutants, may be that an international consensus already exists on the radiation dose limits which ensure man's safety. The recommended radiation dose limits are the results of extensive and elaborated research conducted by the International Commission on Radiological Protection (ICRP). The ICRP is a non-governmental international scientific body created in the early 20th century in order to deal with all scientific aspects of the protection of man against hazards caused by ionizing radiation and consists of representative scientific and medical experts in the field of radiological protection. It makes recommendations on all aspects of radioprotection including basic safety standards; based on hard scientific facts without political considerations, although social and economical implications of these recommendations are not completely ignored. Thus, the ICRP recommendations have been widely accepted among international communities regardless of their political systems and the scientific competence of the ICRP has rarely been contested. On the other hand, the IAEA as a governmental international organization consists of its member states, represented by their governments, and its recommendations take into account their political implications. Therefore, when the ICRP recommendations are accepted by the IAEA, it means that these recommendations are not only scientifically sound, but also acceptable to the governments of the IAEA member states including their implications in political aspects. This is the case of the ICRP recommendations on the basic safety standards for radiation protection laid down in its publication No. 26 (ICRP, 1977), which form essential parts of the IAEA basic safety standards (IAEA, 1982). These are recommendations which constitute the basis for controlling radioactive pollution of the environment due to the introduction of artificial radionuclides.

Although the above ICRP recommendations include those specifying detailed conditions for their applications under various circumstances, the essence of the recommendations may be condensed to the following three principles:

(1) No practice shall be adopted unless its introduction produces a positive net benefit (justification);

(2) All radiation exposures shall be kept as low as reasonably achievable, economic and social factors being taken into account (optimization);

(3) The radiation dose to individuals shall not exceed the annual effective dose equivalent limit of 5 mSv for members of the public or that of 50 mSv for the individual organs and tissues of members of the public (dose limitation).

The original ICRP recommendations have not been written in the form of regulations. Taking into account difficulties in their applications to regulatory purposes for the radiation protection of members of public by competent national authorities, the recommendations have been rewritten into the form of regulations (IAEA, 1982). The national authorities may refer to the rewritten text and make national regulations more specifically adapted to their particular needs and conditions.

Since the recommendations on the basic safety standards for radiation protection concern mainly with the effective dose equivalent limits for man, it cannot be applied directly for limiting

releases of radioactive substances into the marine environment, but the dose limits have to be translated into the terms of emission standards for the releases in order to serve as regulatory tools. This translation is a rather complicated procedure, involving a stepwise approach where various aspects have to be taken into account. Guidelines for controlling radioactive waste discharges into the marine environment on the basis of the ICRP recommendations were elaborated by several expert panel meetings and published by the IAEA (IAEA, 1978; IAEA, 1983). The approach recommended in these publications are briefly outlined below (Section 10). It has been generally recognized that, since environmental conditions of coastal areas receiving radionuclide releases may be so different from one area to the other depending on where these areas are located, it is not scientifically sound nor practical to adopt a series of common emission standards of artificial radionuclides for different discharge sites even within one country.

Although international regulations do exist and are the basis for controlling radionuclide releases into the environment, which have been elaborated by the Commission of the European Communities (CEC, 1981; CEC, 1984), they will be dealt with in the following section.

9.2. National control measures

Efforts have been made recently to collect information on the national regulations for controlling radionuclide releases into the marine environment existing already in the Mediterranean countries. Table XVI lists the national legal provisions available as the basis for controlling radioactive pollution of the marine environment, including those of the Commission of the European Communities. These legal documents are very difficult, however, to be compared directly with each other, since they have been prepared in different contexts, taking into account prevailing circumstances in each country. Nevertheless it has been remarked that there are two categories of the Mediterranean countries towards the control of radionuclide discharges into the marine environment. The first category includes countries where ICRP principles for radiation protection were fully accepted as the basis and the regulations for control of radionuclide discharges into the environment were set up more or less following the guidelines drawn by the IAEA. The second category includes countries where practically no legal provisions have been prepared, since there has been no intentional release of radionuclides into the environment. Although the situation is not clear, it is likely that, even in the second-category countries, legal provisions for the basic safety standards for radiation protection based on the ICRP recommendations may exist, since medical applications of radionuclides are rather widely exercised also in these countries.

In the first-category countries where nuclear industries have already been developed the legal permits for environmental releases of radionuclides are normally accompanied by imposed monitoring of effluents released as well as environmental monitoring of the areas receiving discharges. The monitoring operations are normally conducted by operators of nuclear installations or competent national authorities in order to ensure the safety of the release operations. Therefore, a number of monitoring data have been and will be available to competent national authorities, although these data have not always been published. In order to follow the long-term trend of radioactive contamination of the Mediterranean Sea within the framework of the implementation of the Mediterranean Action Plan it is considered important to assemble these monitoring data, especially the data on the amounts of radionuclides introduced into the Mediterranean marine environment from each country, at a centralized competent body dealing with environmental pollution of the Mediterranean Sea.

Table XVI

Existing national legislations for controlling radioactive marine pollution in the Mediterranean countries

Country	Legal provisions available for marine pollution control	Acceptance of ICRP principles as the basis for these regulations
Commission of the European Communities	! Directive of the Commission on the basic standards for radioprotection of the general public and radiation workers (1980) ! Directive of the Commission on the fundamental measures for radioprotection in medical examinations and treatments (1984) ! Directive of the Commission for modifying the basic standards for radioprotection of radiation workers (1984)	Yes
France	! Law for combatting pollution of water systems (1964) ! Regulatory order on discharge limits of radioactive effluents from nuclear centres and measures for environmental monitoring (1976)	Yes
Israel	! The prevention of sea pollution law (1983) ! The prevention of sea pollution from Land-based sources law (1988)	?
Libya	! Regulation on ionized radiation use and protection against its hazards (1981) ! Maximum allowed limits for radioactive materials in foodstuff	?
Morocco	! Law on maritime fisheries (1973) ! Draft order on the protection against ionizing radiation and radioactive substances	?
Spain	! Spanish nuclear legislation (1964) ! Directive for regulating nuclear installation and radioactivity (1972) ! Directive on regulations for health protection against ionizing radiation (1982)	Yes
Syria	! Series of basic safety criteria for the radioactive protection ! Maximum levels of radioactivity tolerated in food	?

10. Scientific Rationale for Establishing Restriction and Control

As has been stated, since the radiation dose limits for man have to be translated to the emission standards for radionuclide releases into the marine environment in order to serve as regulatory tools, it is necessary to develop rational procedures relating to the quantities of potential releases of radionuclides to the resultant dose. Although several methods for obtaining this relationship were tried, the past experiences have shown that at present only two approaches, the system analysis approach and the concentration factor approach, are recognized to serve reasonably well for achieving the goal (IAEA, 1983). These two approaches are briefly presented below. Taking the latter approach, which has been used more frequently, as an example, stepwise procedures for relating the radiation dose limits to the release rates of radionuclides from nuclear installations are outlined.

Since the ICRP principles for radiation dose limitations include those for the justification and optimization, it is important to demonstrate beforehand to national decision-making authorities that specific release operations of radionuclides into the environment are justified and optimized compared to other options. These procedures are not simple, since they normally involve the analyses of balances between the effects and benefits, costs and benefits, health and social convenience, etc. Quantitative evaluations of these factors are always hard to make, as the evaluations depend very much on the basis of which they are made. When the decisions for the release operations are taken at national levels, however, it means that national radiation dose limits to be tolerated are accepted in relation to the releases of radionuclides into the environment. These national radiation dose limits may be either at the levels equivalent to those recommended by the ICRP or fractions of the recommended dose limits.

10.1. System analysis approach

The system analysis approach intends to model the dynamic behaviour of the radionuclides released in the marine environment by applying complicated and sophisticated mathematics. It is particularly applicable to situations where radioactive releases are pulsed, or where environmental situations are such that steady-state conditions are not likely to exist. In conducting this approach it is necessary to solve a series of first-order differential equations and the biggest difficulty is generally the availability of values for the transfer coefficients involved, which, in reality, vary with time and space. As the expressions become more realistic, unique analytical solutions of these differential equations do not generally exist and numerical solutions have to be adopted to simulate temporal behaviour of radionuclides released. The procedures for obtaining the numerical solutions are complex and require extensive calculations by computer and associated specific softwares for executing this work.

10.2. Concentration factor approach

The concentration factor approach includes, as its special case, the so-called critical pathway approach and is applicable where release rates are uniform over relatively long periods of time and where steady-state conditions can be assumed to exist in the environment receiving releases of radionuclides, although, strictly speaking, such conditions are rarely fulfilled in real situations. When the radiation dose limits to be tolerated by the general public (or a fraction of the general public) are decided at a national level in relation to the releases of radionuclides from nuclear installation sites into the marine environment, the concentration factor approach starts with the dose limits and goes step by step backwards through various environmental compartments in order to arrive at release limits of radionuclides at discharge points. The outline of the approach is presented below step by step.

10.2.1. Selection of possible pathways: When radioactive waste effluents are discharged from nuclear installation sites into the marine environment, radionuclides contained in the wastes may cause radiation exposure to man through various routes. They may be accumulated on beaches causing the external exposure of bathers or taken up by edible marine organisms causing the internal exposure of persons who ingest these organisms as seafood. In this way there are potentially a number of routes, so-called "pathways", by which man may be exposed to radiation resulting from the radionuclides released. In practice, however, for any given site, only one pathway or at most a small number of pathways, may prove to be limited. Therefore, if the radiation exposure of the public along these "critical pathways" is kept well within the national dose limits, the exposure through other pathways will not cause these limits to be exceeded. Similarly, although a large number of radionuclides may be discharged, only a small number of "critical radionuclides" will account for the major fraction of the exposure. When the "critical pathways" can be well identified, then the procedures involved in the concentration factor approach become simpler and the approach is specifically called the "critical pathway approach". Thus, in order to avoid unnecessary complexity involved in evaluating numerous pathways, it is important at first to select out significant pathways on the basis of the general information available for the radionuclide composition of effluents to be discharged, the degree of accumulation of certain radionuclides in some types of marine organisms, amounts of consumption of the marine organisms as seafood, etc. In view of the present situations in the Mediterranean Region, probable critical pathways are most likely to be through the ingestion of seafood.

10.2.2. Annual limits on intakes (ALIs): The radiation dose limits set up can be translated to the annual limits on intakes of individual radionuclides, if the relationships between the amounts as well as distributions of these radionuclides in certain human organs or the whole body, and effective dose equivalents received by them are known. The translation work has been completed by the ICRP on the basis of the data obtained from extensive experimental radiation dose measurements and chemical analyses of human organs, producing several publications (ICRP; 1979; ICRP 1980; ICRP, 1981; IAEA, 1982). These publications include extensive tables listing annual limits on intakes of each radionuclide for occupational exposure (expressed in Bq). The values for the members of the general public should be one tenth of the values listed in these tables. The values found in the tables can be applied directly to the identified possible critical radionuclides.

10.2.3. Demographic survey: In order to estimate the dose to the most exposed individuals within the dose limits it is essential to obtain the information on the working, eating and recreational habits of the local population being expected to receive higher radiation doses and, in certain cases, of populations at some distance from the discharge sites. For example, these surveys may include estimates of the types and amounts of ingested marine organisms harvested from specific areas, the number of hours spent on the beach, at work or for recreation, the number of hours spent handling fishing gear on the beach and at sea, etc. Since it is considered most unlikely that the external radiation exposure at beaches become limiting in the present situations encountered in the Mediterranean Region, the surveys should be concentrated in the feeding habit of the local populations around the release sites. On the basis of the results of the demographic surveys conducted on the local populations it may be possible to identify probable critical groups of people within the local population, who may have the highest probability for the radiation exposure caused by the releases of radionuclides from the specific discharge sites. It should be emphasized that, since feeding habits of the local populations are different from one place to another, the resultant doses received by local populations from radionuclide releases are site-specific even though concentrations of radionuclides in consumed seafood are similar for different localities. It should be also noted that the ingestion of the seafood is not only the cause of radiation doses for the populations but the ingestion of terrestrial food constitutes normally the major cause of the radiation doses especially in the Mediterranean Region.

10.2.4. Limits for environmental levels: Knowing the values of the ALIs for possible critical radionuclides contained in the waste effluent and the feeding habit of the local populations around the release site of the radionuclides into the sea, one can determine concentration limits of specific radionuclides in marine organisms being consumed as major seafood by the local population. Thus, the identification of the critical groups within the local population may be possible at this stage. By utilizing the recommended values of biological concentration factors of the critical radionuclides for the marine organisms being consumed as major seafood, the concentration limits of the radionuclides in the ambient sea water can be determined in such a way that by maintaining the concentration limits in sea water the resultant radiation doses received by the members of the critical group would not exceed the national dose limits set out at the beginning of the concentration factor approach. For the preliminary estimations of the concentration limits of the possible critical radionuclides in the ambient sea water it may be sufficient to apply the recommended mean biological concentration factors for the specific groups of marine organisms (IAEA, 1985). However, it may be more prudent to utilize specific concentration factors of the critical radionuclides for local species involved in the critical pathways, which have to be determined experimentally at the site, in order to obtain increased accuracy of the concentration limits in the final stage of the concentration factor approach.

10.2.5. Release limits at discharge points: When the effluents containing radionuclides are released into the coastal marine environment they are immediately mixed and diluted with coastal sea water. The degree of the dilution depends on prevailing local hydrographical conditions, such as coastal currents, eddy diffusion, prevailing wind, bottom topography, etc.. These hydrographical conditions around the discharge points are important in deciding the dilution factor of the effluents, and to be investigated extensively beforehand. Taking into account the local hydrographical conditions as well as chemical forms of critical radionuclides in the effluents and their expected behaviour in the coastal marine environment, the release rates of the radionuclides from the discharge points can be determined so as to maintain the concentration limits of the radionuclides in the ambient coastal sea water. As the coastal hydrographical conditions are variable within a relatively short time, it is often necessary to apply averaged hydrographical parameters in describing prevailing coastal conditions.

10.3. Pre-operational survey and monitoring

As can be seen from the preceding descriptions on the approaches to be made for determining the release limits of radionuclides into the marine environment from discharge points, several environmental and demographic parameters are needed to proceed with these approaches. These parameters have to be estimated on the basis of either the results of pre-operational local surveys or reasonable assumptions. Thus, the accuracy of the results of the approaches depends on how accurately the individual parameters are able to describe real processes involved in the models adopted. Uncertainties involved in selecting appropriate parameters influence considerably the results obtained. Therefore, it is often necessary to verify the results of the pre-operational approaches by trials of low-level release operations. When the results of the pre-operational approaches are proven to be satisfactory, the authorization for full release operations may be granted. In order to ensure that the dose to the most exposed individuals remain within the national authorized dose limits after the full operation of radionuclide releases, it is essential to monitor regularly the release operations at the release points and in the marine environment expected to be influenced by these release operations. Although competent national authorities and/or management of nuclear installations concerned are considered to be responsible for implementing adequate monitoring operations of the effluents released and the marine environment under consideration, it may be appropriate to harmonize internationally the mode of the monitoring in the framework of the Mediterranean Action Plan.

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