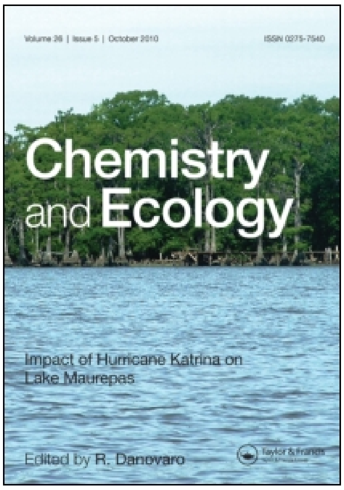

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RADIOACTIVE CONTAMINATION IN THE ITALIAN SEAS: AN OVERVIEW ALSO IN THE LIGHT OF THE EUROPEAN MARINA-MED PROJECT

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Following a similar project performed for the Northern European Seas in the years 1985–1990, a study on the radioactive contamination of the Mediterranean Sea has recently been conducted by a group of experts of the European Union (MARINA-MED Project). Its aim was to assess the radiological exposure of the population of the European Union, due both to natural and man-made radionuclides in the Mediterranean Sea. A review of the conclusions of this study is presented, with special attention to the environmental radioactive data. The Italian radioactive monitoring organisation is described. Starting from this experience, some proposals for new research projects are presented.

KEY WORDS: Radioactivity, natural/man made, Mediterranean Sea, Italy.

INTRODUCTION

In the years 1992–1993, a group of experts was established by the Commission of the European Union to prepare a report on the radioactive contamination in the Mediterranean Sea, in order to assess the doses to European population due to both natural and man-made radionuclides in the seas and marine matrices.

A similar project, called "MARINA", was conducted for the Northern European Seas in the years 1985–1990. For this project, the group of experts appointed by Member States was asked to elaborate a report on four main items: 1) the disposal of solid and liquid radioactive waste in Northern European marine waters; 2) existing marine dispersion models, with a view to agreeing on a valid comprehensive model; 3) quantities, distribution and end-uses of marine produce; 4) exposure of the public. The project results were published in a European Report (C.E.C., 1990).

For the Mediterranean Sea project, the same procedure was followed: the experts nominated by Member States were asked to analyse the total radiological exposure of the population in the European Member States, due to natural and man-made radionuclides in the Mediterranean Sea. The project (named MARINA-MED) was conducted in the years 1992–1994, considering data from 1980 to 1990, and, whenever possible, adding data from the years 1991–1993.

In order to achieve this objective, the following working groups were set up: 1) sources of radioactivity in the Mediterranean Sea; 2) radioactivity in the Mediterranean waters; 3) survey of the quantities and utilisations of associated marine products; 4) radiological impact on E.C. Member States of routine discharges into the Mediterranean Sea. These are also the section titles of the final report, which was published in 1993 (E.C., 1993).

The main sources of artificial radioactivity in the Mediterranean Sea are routine discharges from nuclear power and research plants, and the radioactive fallout due to atmospheric nuclear weapon tests as well as that from the Chernobyl accident. The nuclear plants are placed in Spain, France and Italy, but the plants discharging in the Black Sea also contribute. As, after the Chernobyl accident, the Black Sea also contributed greatly to radioactive contamination where in 1990, ^{137}Cs concentrations are by an order of magnitude higher than those in the remaining part of the Mediterranean. This was considered as a real source of radioactivity for the Mediterranean Sea. In the Mediterranean, waste dumping operations have never been carried out, but the only officially-confirmed accident with losses of radioactive substances occurred in this sea, when in January 1966 a US B-52 bomber carrying four thermonuclear weapons crashed near the Almanzora river between Palomares and Cuevas de Almanzora (Spain). As a consequence, 2.26 km^2 of territory were contaminated with ^{239}Pu and ^{240}Pu (the total Pu content in the bombs is still unknown, being covered by US military secret). Among the sources, the 1964 accident to the US Transit 5BN-3 satellite is also quoted. It was carrying 1 kg of metallic ^{238}Pu, and re-entering the atmosphere, it released 629 TBq of ^{238}Pu in the environment, due to vaporisation and dispersion of nuclear fuel into the stratosphere.

As regards natural radionuclides, they are ubiquitous and they have always been present on the Earth or originate from outer space. The most frequent in the sea are ^{40}K and ^{87}Rb, the nuclides of uranium and thorium families (above all ^{226}Ra, ^{210}Pb and ^{210}Po) and the cosmogenic nuclides ^{14}C and ^{3}H. ^{210}Po gives the most relevant contribution (about 80%) to the collective dose from natural radioactivity in the sea, due to its high concentration factor in shellfish (see the following discussion).

The Gulf of Lion underwent a separate analysis due to its peculiarities. The nuclear plants along the Rhône river discharge their liquid wastes in the Gulf and, for this reason, almost 50% of the collective dose from marine artificial nuclides received by European Mediterranean inhabitants (from France, Greece, Italy and Spain) following the discharge of liquid radioactive effluents from the E.U. nuclear installations comes from this sea area alone. The Marcoule reprocessing plant (France) is the main contributor (95%) to the total collective dose of about 2 per man Sv over the period 1980–1991: its release represents 76.7% of the total beta-gamma and more than 99.9% total alpha discharges. The main contributor to total beta-gamma discharges is ^{3}H (92.26%). Leaving aside ^{3}H, the most important contributor is $^{106}Ru + ^{106}Rh$ (79.69%), followed by the decreasing contribution by $^{90}Sr + ^{90}Y$ (7%), ^{137}Cs, ^{134}Cs, ^{58}Co and ^{54}Mn. The main contributors for total alpha discharges are ^{241}Am (31.4%), $^{239+240}Pu$ (27.87%) and U_{nat} (24.79%). The highest contribution to the total collective dose is given by ^{106}Ru (39%, of which 95% is due to ingestion of molluscs).

However, in the years 1980–1991, the radioactivity from European nuclear civil plants released in the Mediterranean was more than two orders of magnitude lower than that in the Northern Europe waters (E.C., 1993).

In order to analyse the radioactive contamination of sea water and marine matrices, the working group (2) collected available data from each country regarding the activity concentration of ^{137}Cs and ^{210}Po in the years 1981–1990, integrating them, whenever possible, for the years 1991–1993. Matrices such as surface waters, fish, shellfish and surface sediments were taken into account. The Mediterranean Sea was divided in ten basins, in many cases surrounded by more than one country, in order to compare and average the data. A small special report was prepared for each country by its representative. Summaries of these reports can be found as annexes to chapter 2 of the European Report (E.C., 1993).

In Italy this analysis was made only for ^{137}Cs, due to the absence of available systematic data for ^{210}Po. Even in other countries, data for this radionuclide were scattered but as a first approximation, despite their lacking of statistical representativeness, they were used to calculate the mean values and to assess the doses.

As regards ^{137}Cs, in Italy a lot of data were available. Since the mid-fifties, in this country, two types of environmental radioactivity networks have been operating. The *local network* was set up near nuclear power or research plant sites, in order to monitor the environment in the neighbourhood of the plants. The *national network* was aimed to evaluating the exposure of the population from all radioactive sources throughout the national territory. They have been upgraded through these years, particularly in the aftermath of the Chernobyl accident: Regional Radioactivity Control Laboratories were set up, constituting the *regional network*. As a result, new laboratories began operating on the national territory and this enhanced the development of new experimental projects. These networks carry out periodic measurements on environmental radioactivity contamination and, for this reason, they represent the most reliable source of systematic data in the field. Additional data come from papers published by some research groups, especially from Universities, that have concentrated their experimental work on radioactivity in marine matrices.

For the network procedures, it should be stressed that in general the sampling sites are not homogeneously distributed along the coasts or in the Italian seas. Due to the main aims of radioprotection, samplings are more frequent near possible contamination sources, that is near the outlet of big rivers, along the coast near nuclear plants, in harbours also used by nuclear fuel-propelled naval vessels or submarines, etc. This is a possible cause for overestimation.

This analysis on radioactivity in the Mediterranean Sea, based on experimental measurements, leads to some interesting conclusions (E.C., 1993). First, the ^{137}Cs concentrations in sea water, fish, shellfish and sediments are similar in all the Mediterranean basins, with the exception of some small local variations: in 1990, e.g., mean values for the different basins were some $Bq\,m^{-3}$ for water, some tenths of $Bq\,kg^{-1}$ fw for fish, the same for shellfish, and varying from a few $Bq\,kg^{-1}$ dw to 15 ± 13 $Bq\,kg^{-1}$ dw (in the Adriatic Sea) for surface sediments. Secondly, the contribution of radioactive contamination from the Chernobyl accident is clearly evident in water and fish (^{137}Cs concentrations are by an order of magnitude higher), less evident in shellfish and in surface sediments, due probably to considerable inhomogeneity of the samples used to calculate the mean value. In general, in 1990 activity concentrations decreased to the pre-Chernobyl levels, apart from those in the Black Sea and the northern part of the Aegean, which still show increased levels. For doses of annual ingestion from ^{210}Po contaminated seafood are shown to be almost by an order of magnitude higher than those from ^{137}Cs. That is the Mediterranean mean individual effective dose received from Chernobyl ^{137}Cs through marine food showed to be approximately 0.5 μSv, whereas the annual dose from ^{210}Po is 33 μSv. Moreover, the doses to the critical population groups from the 1990 intake resulted in 0.5 μSv from ^{137}Cs, while 0.54 mSv arises from ^{210}Po. However, the very few Mediterranean ^{210}Po data available may make dose estimates for ^{210}Po less reliable. As for the total collective dose to the E.U. population derived from direct measurements of ^{137}Cs in fish and shellfish in 1990 (1 year contribution) this resulted to be 1.7 man Sv, that is 1.4 from fish, 0.25 for molluscs and 0.072 from crustacean ingestion. Collective dose rates for ^{137}Cs calculated using as a source term the discharges from E.U. nuclear installations are more than two orders of magnitude lower (about $1 \cdot 10^{-2}$ man Sv y^{-1}), therefore the ^{137}Cs activity concentrations in the Mediterranean Sea and in the Black Sea are mostly due to fallout from experimental nuclear weapons and releases from the Chernobyl nuclear plant. Concentrations factors (CF) of seafood to sea water ($Bq \cdot kg^{-1}$ fresh water/$Bq \cdot l^{-1}$ sea water) were calculated. The ^{137}Cs mean concentration factor for fish (80) was in good agreement with the recommended value by IAEA (1985), i.e. 100. However, the value found for shellfish (200) was higher than the IAEA value of 30. It could be explained primarily by some high levels in shellfish from Tyrrhenian and Ionian Seas, which received Chernobyl fallout in a very patchy way. As for ^{210}Po, the CF resulted in $6 \pm 4 \cdot 10^3$ for fish and $19 \pm 5 \cdot 10^3$ for shellfish, not incompatible with the IAEA-recommended values ($2 \cdot 10^3$ and $1\text{–}5 \cdot 10^4$, respectively).

These conclusions give rise to some considerations and proposals.

First of all, this experience should stimulate the onset of a new study project on the radioactive contamination of the Mediterranean Sea, foreseeing an international scientific collaboration among all European and non-European countries surrounding the Mediterranean Sea. It should be noted that the MARINA-MED project was limited to the E.U. Member States, even if some published data from Croatia (for the Adriatic Sea) and from Ukraine and Rumania (for the Black Sea) were used. For example, the participation of other countries, such as some countries of Northern Africa facing the Mediterranean Sea, would have improved the knowledge of the southern Mediterranean area. This project should not fix radiation-protection but only research objectives and be organised with different criteria. Sampling sites should be chosen in order to be statistically representative and additional measurements should be foreseen in those areas, where very few data are now available. Moreover, this project should be extended to some natural radionuclides, such as ^{210}Po and ^{210}Pb, which are not covered presently by routine measurements.

From the comments on concentration factors, other research projects can originate. In some papers the calculated CFs seem to be significantly different from those recommended by IAEA. For example, McDonald *et al.* (1992) quote this discrepancy

Table I Half-lives, adult ingestion dose coefficients and concentration factors for fish, crustaceans and molluscs for some natural and artificial nuclides frequent in the marine environment.

Nuclide	$T_{1/2}$	*Ingestion dose conversion coefficients** $Sv\,Bq^{-1}$	CF^{**}_{fish}	$CF^{**}_{crustaceans}$	$CF^{**}_{molluscs}$
Cs-137	30.174 y	$1.4 \cdot 10^{-8}$	100	30	30
Cs-134	753 d	$1.9 \cdot 10^{-8}$			
Ru-103	39.35 d	$7.3 \cdot 10^{-10}$	2	100	2000
Ru-106	368.2 d	$7.0 \cdot 10^{-9}$			
Po-210	138.4 d	$1.2 \cdot 10^{-6}$	2000	50000	10000
Pb-210	21 y	$7.0 \cdot 10^{-7}$	200	1000	1000
Ra-226	1602 y	$2.8 \cdot 10^{-7}$	500	100	1000

* ICRP, 1994
** IAEA, 1985

for ^{210}Po and ^{210}Pb, stressing that this is due partly to the few data used by IAEA for the calculation. The needs for further study is thus clearly pointed out.

Finally, it would be interesting to devote special attention to the content of natural radionuclides, particularly ^{210}Po and ^{210}Pb, in the Italian waters. Table I clearly shows why these two radionuclides have a great impact on population doses: both the conversion coefficients from ingested radioactivity to the dose and the concentration factors from sea water to fish, crustaceans and molluscs are very high. They are 1 to 3 orders of magnitude higher than those of caesium isotopes. As a matter of fact, IAEA- recommended concentration factors would be better expressed by a range, because they are affected by a high uncertainty, but this fact does not change the previous comment.

High levels of natural radioactivity in marine matrices have been detected along the coasts of England, Ireland, the Netherlands and France (see, e.g. McDonald *et al*, 1992; Oosterhuis, 1992; Germain *et al.*, 1992; McCartney *et al.*, 1992). They seem to be caused mainly by discharges from phosphate ore processing plants, but in some cases also from sea dumping of coal spoil and from the Sellafield reprocessing plant. McDonald *et al.* (1992) have calculated an individual dose up to 3.2 mSv per year, mainly for ^{210}Po, for people eating mussels from an area where the Whitehaven (UK) phosphate plant discharges.

Also in Italy, this problem should be given more attention, due to the fact that about 2 Mton of phosphatic materials, mainly from Northern Africa, are processed yearly. It has been estimated that such materials have a mean uranium concentration of about 100–200 ppm, as well as a frequently similar concentration of thorium (Cappadona *et al.*, 1983).

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