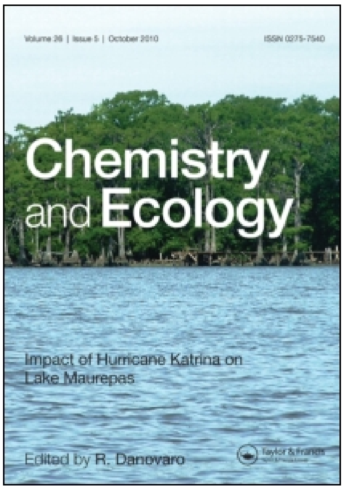

Chemistry and Ecology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713455114

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Francesco Nonnis Marzano[a]; Cesare Triulzi[a]; Marina Vaghi[a]
[a] Department of Evolutive and Functional Biology, University of Parma, Viale delle Scienze, Parma, Italy

To cite this Article Marzano, Francesco Nonnis , Triulzi, Cesare and Vaghi, Marina(1996) 'Evolution of Radiocontamination of the Northern and Middle Adriatic Sea in the Period 1979-1990', Chemistry and Ecology, 12: 4, 239 — 246

To link to this Article: DOI: 10.1080/02757549608039089

URL: http://dx.doi.org/10.1080/02757549608039089

Chemistry and Ecology, 1996, Vol. 12, pp. 239–246
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EVOLUTION OF RADIOCONTAMINATION OF THE NORTHERN AND MIDDLE ADRIATIC SEA IN THE PERIOD 1979–1990

FRANCESCO NONNIS MARZANO, CESARE TRIULZI and MARINA VAGHI

Department of Evolutive and Functional Biology, University of Parma, Viale delle Scienze, Parma, Italy

(Received 2 February 1996)

Several investigations were carried out on biotic and abiotic samples to determine the difference between Chernobyl and preChernobyl environmental radiocontamination of the Adriatic Sea. Special emphasis was put on the determination of ^{137}Cs which resulted in the most abundant radionuclide in a wide variety of samples (sea water, sediment, pelagic and benthic species). Attention was also given to the neutron activation products ^{110m}Ag and ^{134}Cs that were first discovered in the Adriatic ecosystem after the Chernobyl event. In particular, although the ^{110}Ag fallout deposition over the area was neglected in comparison to that of ^{137}Cs, its bioaccumulation in macroalgae and molluscs was much higher suggesting some particular physiological accumulation mechanism.

KEY WORDS: Adriatic sea, artificial radioisotopes, bioaccumulation, caesium, silver.

INTRODUCTION

A ten-year radioecological investigation (1979–1990) was carried out in the Adriatic Sea to assess the wide fluctuations of anthropogenic radionuclides introduced by both direct fallout deposition and river inputs.

The first part of the research related to the preChernobyl period (1979–1985) and during that time ^{137}Cs was largely investigated in connection with intense atmospheric nuclear testing which generated in the sixties and seventies a source of global fallout (Triulzi *et al.*, 1983). A complete mapping of the ^{137}Cs concentrations in the study area together with the distribution between water, biomass and sediment, was obtained.

During the following years (1986–1990) a full assessment of the Chernobyl–derived radioactivity was performed. In fact, the accident at the Chernobyl nuclear reactor during April–May 1986 injected a considerable amount of fission and activation products into the atmosphere. Its delivery over the Mediterranean area through fallout depositions followed soon the event. Since the Adriatic was one of the seas significantly contaminated by the accident, studying the amounts of radionuclides from the atmospheric source to reach the aquatic environment was of special interest.

Although special emphasis was put on the determination of ^{137}Cs which became the most abundant radionuclide in a wide variety of samples (sea water, sediment, pelagic and benthic species), attention was also given to the neutron activation products ^{110m}Ag and ^{134}Cs that were first discovered in the Adriatic ecosystem after the Chernobyl event. In particular, although the ^{110m}Ag fallout deposition over the area was neglected in comparison to that of ^{137}Cs, its bioaccumulation in macroalgae and molluscs was much higher suggesting some particular physiological accumulation mechanism.

Some interesting considerations about biogeochemical cycles and ecological processes were pointed out. Other data concerning ^{90}Sr and plutonium isotopes is reported elsewhere (Desideri *et al.*, 1996).

MATERIALS AND METHODS

Samples of sea water, zooplankton, icthyofauna, benthos and sediments (different strata) were collected between November 1979 and June 1990. The study area covered an extended sea surface going from the Gulf of Trieste to the Gargano promontory. The sampling stations were located along seven different transects stretching between the borders of the Italian and Croatian territorial waters and facing main localities of both sides (Fig. 1).

Further, fallout samples were collected monthly in Parma in the framework of the Italian national radioprotection monitoring programme (Morani *et al.*, 1989).

All different methods and radioanalytical techniques to obtain gamma spectrometry sources have beeen reported elsewhere (Nonnis Marzano and Triulzi, 1994). The ^{110m}Ag, ^{134}Cs and ^{137}Cs concentrations were determined through direct gamma spectrometry by means of GeLi and GeHP (High Purity) (Ortec and PGT Silena) detectors. The data were then elaborated by an IBM PC equipped with an EG&G Ortec software programme.

All data were decay-corrected to sampling time and reported as $Bq\,m^{-3}$ for sea water, $Bq\,kg^{-1}$ dry weight for the sediment, $Bq\,kg^{-1}$ wet weight for biotic samples and $Bq\,m^{-2}$ for the fallout. Errors considered as standard deviation (1 sigma) were referred only to the counting statistics.

RESULTS AND DISCUSSION

In the Mediterranean Sea fallout has been the main source of environmental radiocontamination especially during the last decade. In particular, two different periods of global fallout contamination were observed.

Atmospheric dry and wet depositions over the basin were influenced by the intense nuclear testing which generated in the sixties and seventies a source of global fallout. After the interruption of any nuclear atmospheric test (last explosion was the Chinese test which happened in 1980) (Whicker and Schultz, 1982), a fast decrease of all artificial gamma emitters except that of ^{137}Cs, was observed. In spite of the ^{137}Cs continuing presence, its concentrations were generally low between 1981 and April 1986. During that period Morani *et al.* (1989) reported concentrations in the range 0.6–0.10 $Bq\,m^{-2}$ as ground deposition in the northern part of the country.

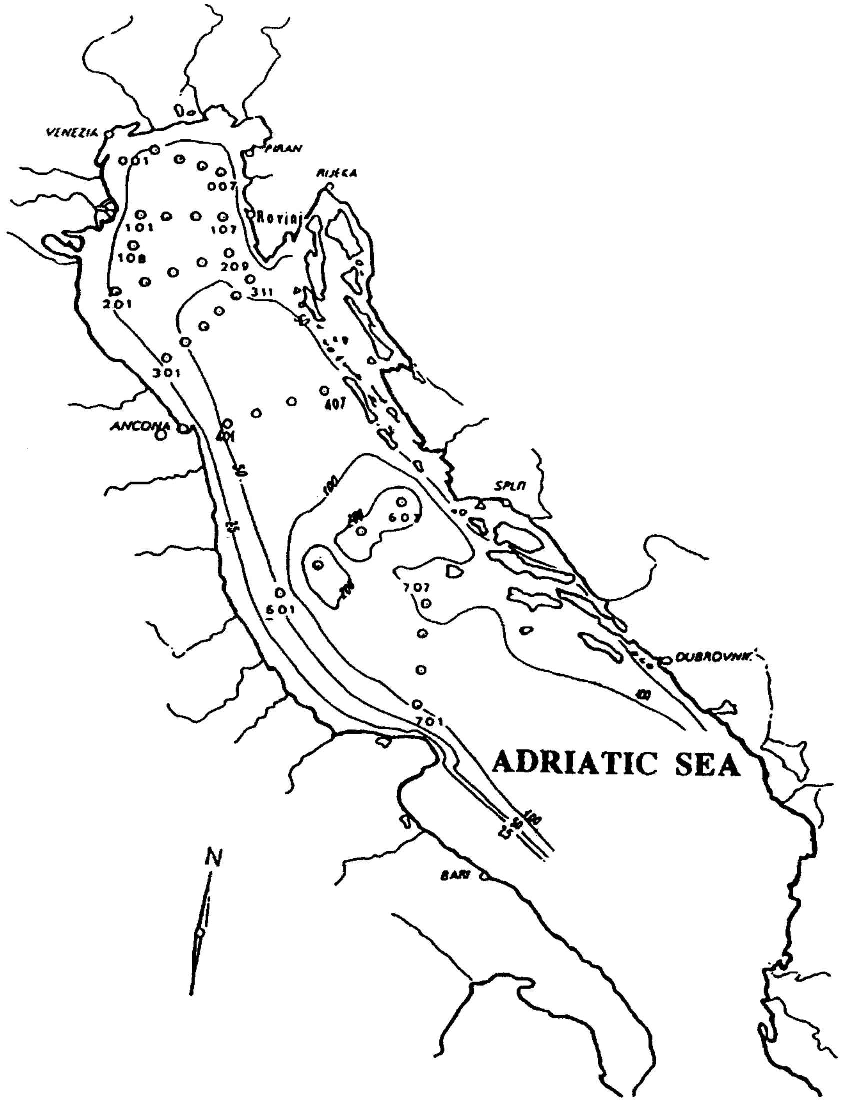

Figure 1 Study area in the Northern and Middle Adriatic Sea.

After the Chernobyl accident (April 26, 1986) (ENEA, 1986) a rapid increase of ^{137}Cs, together with other short half-lived radionuclides, was measured. Radiocaesium concentrations as high as 3500 Bq m^{-2} for ^{137}Cs and 1740 Bq m^{-2} for ^{134}Cs were detected in fallout samples of the Po river plain during May 1986. In Figure 2 a trend of the ^{137}Cs fallout concentrations between 1986 and 1990 is reported.

In general, the fallout deposition over Italy was higher in the north rather than in the central and southern part of the country. Furthermore, the distribution of the concentrations showed an east-west gradient with lower values in the Western

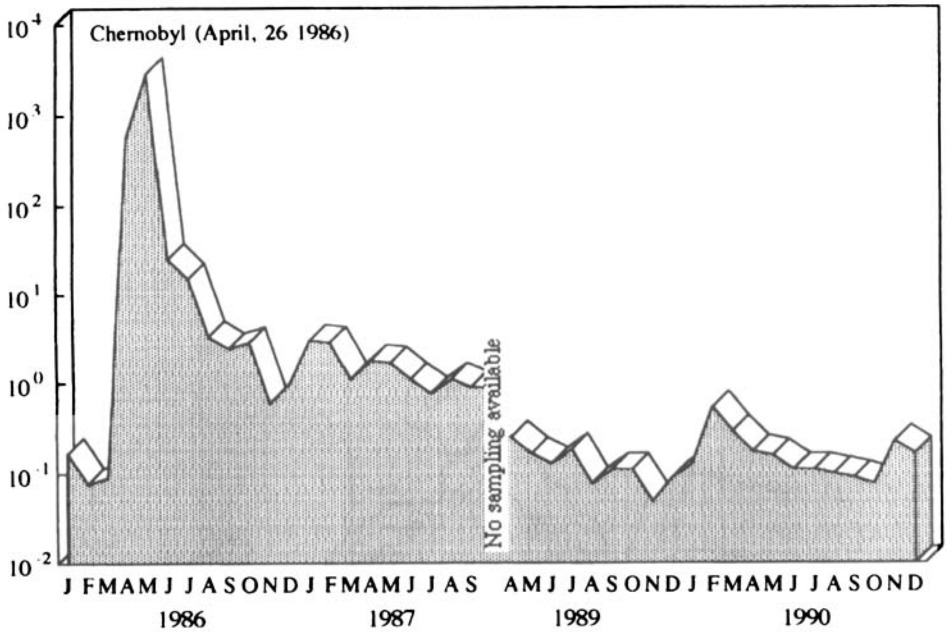

Figure 2 Monthly depositions of ^{137}Cs ($Bq\,m^{-2}$) detected in the Po River plain (1985–1990).

Mediterranean and higher ones in the Eastern basin. In fact, Ballestra *et al.* (1987) reported values of 1400 and 700 $Bq\,m^{-2}$ reespectively for ^{137}Cs and ^{134}Cs in fallout samples of the Gulf of Lion.

The ^{137}Cs concentrations had a fast decrease in fallout samples collected after June 1986 with just slight increases during months of heavy rainfall. The data finally reached preChernobyl values during the middle of 1989 and have steadily remained around 0.10–0.20 $Bq\,m^{-2}$ since that time. On the other hand, ^{134}Cs fell below the detection limits during December 1989.

Increasing radiocontamination in the Adriatic environment was observed just a few days after the Chernobyl event. The distribution of ^{110m}Ag, ^{134}Cs and ^{137}Cs between sea water, biomass and sediment was therefore evaluated.

The different temporal trends of ^{137}Cs detected in sea water, zooplankton, *Sardina pilchardus* and coastal sediments are reported in Figure 3, 4 and 5. Such data were determined between 1980 and 1990 along both the Italian and Croatian coastline.

As it can be noticed, the radiocaesium concentrations in water, plankton and *Sardina Pilchardus* had very similar trends. These data were indicating a fast transfer of the radiocontaminant from the water to the pelagic biocenosis. Furthermore, the fast uptake and transfer from plankton to a planktivorous fish is well demonstrated in Figure 4. In fact, in our samples the transfer of ^{137}Cs within the pelagic trophic web resulted faster for planktivorous fish such as *Sardina pilchardus* and much slower for bigger predators such as *Merluccius merluccius* (Nonnis and Triulzi, 1994). Moreover, a fast decrease was observed in the zooplankton due to the rapid turnover and short life cycle of the organisms. This was noticed especially in the November 1986 sample.

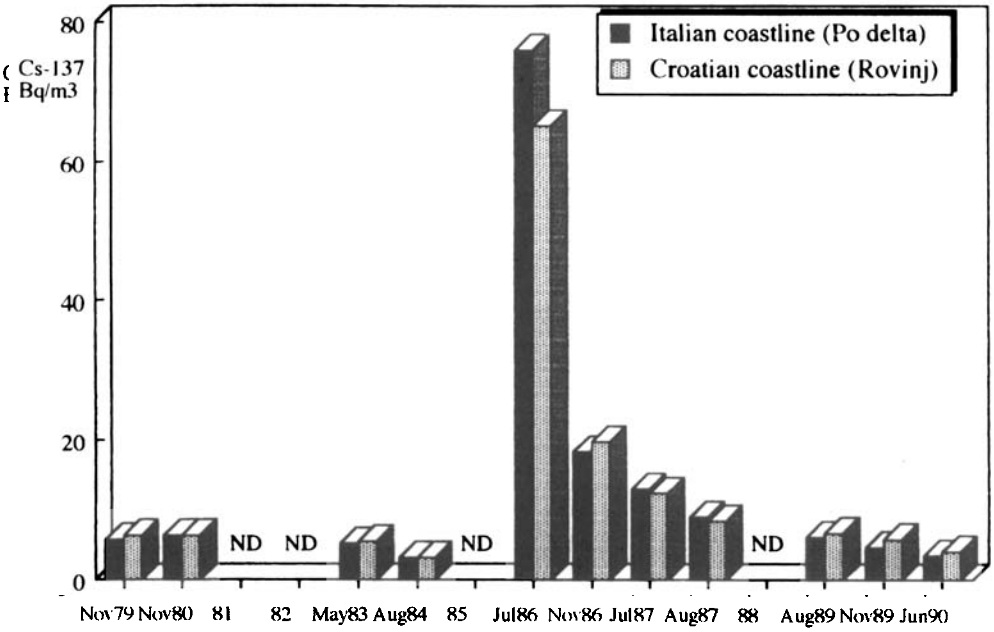

Figure 3 Temporal trends of ^{137}Cs ($Bq\,m^{-3}$) detected in sea water samples from 1979 to 1990 (ND: Sample not available).

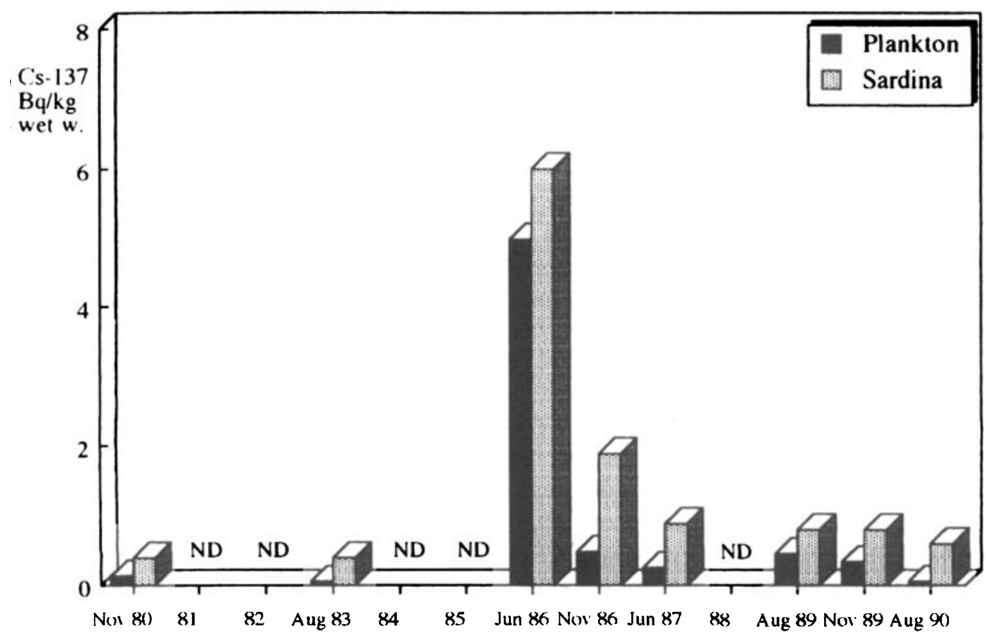

Figure 4 Temporal trends of ^{137}Cs ($Bq\,kg^{-1}$ wet weight) detected in plankton and *Sardina pilchardus* samples collected in front of the Po delta (ND: Sample not available).

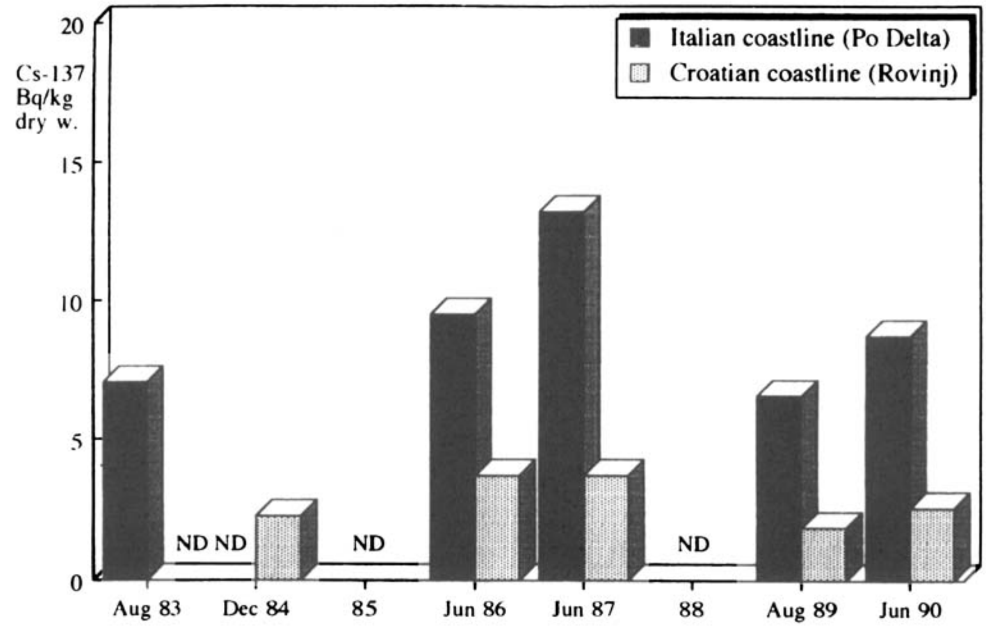

Figure 5 Temporal trends of ^{137}Cs (Bq kg^{-1} dry weight) detected in sediment samples (ND: Sample not available).

The zooplankton seemed to be the main vector of radioactivity from the pelagic to the benthic compartment through packaging of radionuclides into large faecal pellets which rapidly sank to the bottom. It was also noteworthy that ^{137}Cs trends in mixed plankton samples were correlated with their biomass. Higher concentrations were consequently detected in samples collected during periods of high production.

The sediment compartment which represents the final point of pollution accumulation, had major ^{137}Cs contamination more than one year after the accident (July 1987). Such a delay represented the time needed for transport of radiocontaminants from the pelagic to the benthic compartment, even considering the Po river discharges (very intense during autumn 1986 and spring 1987).

Anomalous $^{137}Cs/^{134}Cs$ ratios of 1.4–1.7 were determined in June 1987 in sediment samples of the Fossa di Pomo (Middle Adriatic). The Fossa di Pomo is a trench and the only northern-middle Adriatic area that reached steep depths (about 250 m). Considering the two different decay-times of ^{137}Cs and ^{134}Cs an average value of 3.5–4 was expected. The difference between theoretical and measured values suggested a major contribution of ^{134}Cs than expected. A high ^{134}Cs deposition probably took over as a consequence of the very inhomogeneus and scattered Chernobyl fallout which determined wide local differences.

As already said before, the different composition of the Chernobyl fallout compared to the previous bomb-fallout (Delfanti and Papucci, 1988), introduced in the Adriatic ecosystem new radionuclides which had never been discovered there before.

An evaluation of the uptake times and bioaccumulation mechanisms was therefore investigated especially for ^{110m}Ag and the ^{134}Cs.

In particular, for what concerned ^{110m}Ag, although its ground deposition was much lower than the one of caesium and hardly detectable in fallout and sea water samples, its accumulation by benthic organisms resulted in higher and well measurable values. Molluscs such as *Patella coerulea* provided sensitive indication of silver pollution levels. In Figure 6 a comparison between ^{137}Cs and ^{110m}Ag concentrations in samples of *Patella coerulea* collected in the Gulf of Trieste, is illustrated.

The persistence of ^{110m}Ag in cephalopods and gasteropod molluscans suggested that biological half life is unusually long in these taxa and, particularly in *Patella*, the grazing on algae in succeeding months seemed to replace the radioactive decay and biological excretion of the radionuclide. A very low ^{110m}Ag value was detected in June 1987. Such data were difficult to explain because of sampling not being available during the previous months. Some ecological or physiological aspect was probably responsible for such a sudden radiosilver decrease.

In general it can be summarized that different biogeochemical behaviours for the investigated radionuclides were highlighted. It was observed as ^{134}Cs and ^{137}Cs were present mostly in solution and therefore easily available for the pelagic communities. On the contrary, ^{110m}Ag and its particle reactive form (Coughtrey and Thorne, 1983) was easily driven to the benthic compartment and available for filter-feeders and macrograzers.

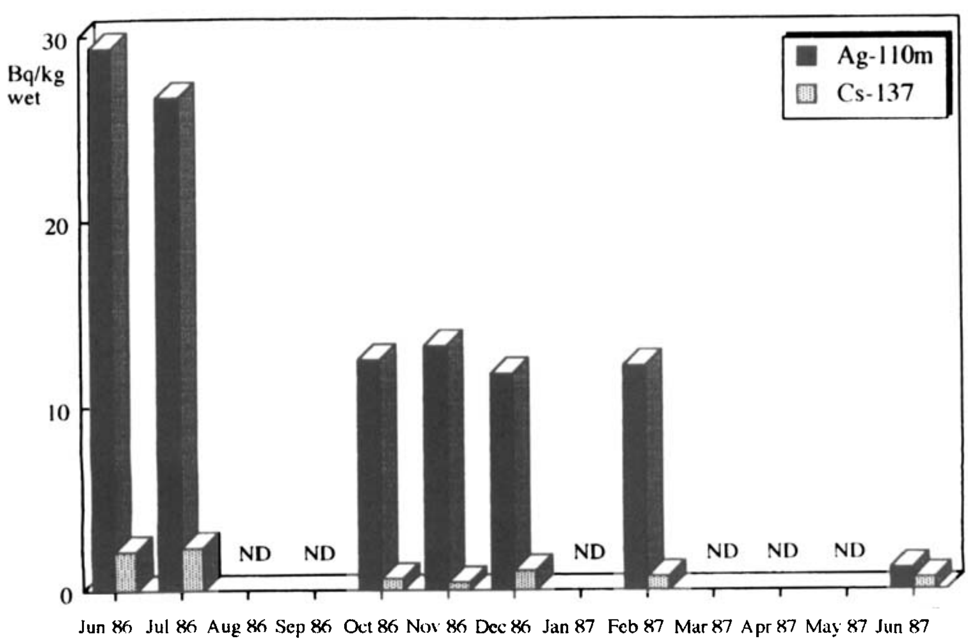

Figure 6 Temporal trends of ^{110m}Ag and ^{137}Cs (Bq kg^{-1} wet weight) detected in samples of *Patella coerulea* of the Gulf of Trieste (ND: Sample not available).

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