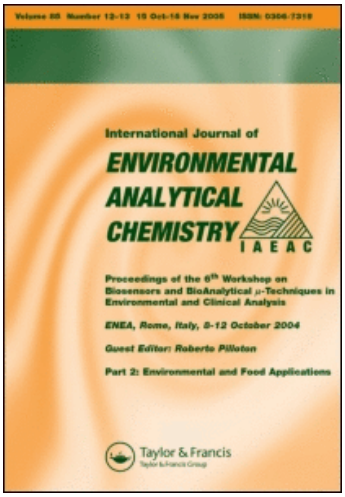

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MARINE AND LACUSTRINE RADIOECOLOGICAL RESEARCHES IN ANTARCTICA. 1992–1994

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Researches on environmental radioactivity in the Ross Sea – Terra Nova Bay area have been carried out with the aim of evaluating the role of different abiotic and biotic matrices in the cycling of radionuclides in the marine and lacustrine ecosystems. The transfer of Sr-90, Cs-137, Pu-238 and Pu-239(240) from the water to the benthic organisms and the sediment layer has been investigated. Contamination levels in both ecosystems and biogeochemical cycles of the main fission and transuranic products are therefore presented and discussed. A comparison of radioecological data from different areas of the Antarctic Ocean and the Mediterranean Sea is given. In the framework of the data obtained, concentrations of the natural radionuclides K-40, Th-232 and U-238 in the sediment samples, are also presented.

Keywords: Antarctica; artificial radionuclides; natural radionuclides; radioecology; bioindicators; Ross Sea

INTRODUCTION

In the first part of this reaseach a complete radioecological survey of contamination levels detected in several matrixes of the marine, lacustrine and terrestrial ecosystems was carried out. The data referred to the period 1987 – 1991 and was published elsewhere[1,2,3]. The previously reported results referred mostly to the anthropogenic Cs-137 with some preliminary data about Sr-90, Pu-238 and Pu-239, together with natural radioactivity determinations of K-40, Th-232 and U-238 in soil and sediments[4].

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The results demonstrated that Antarctica and the surrounding ocean had erroneously longly been considered as unpolluted areas. In spite of the low contamination levels of Sr-90, Cs-137 and plutonium isotopes in most specimens, some interesting concentrations were detected especially in species of the marine and lacustrine environments[5] that can well be considered as bioindicators.

In this paper we report the second phase (1992 – 1994) of our study which deals with investigations on marine and lacustrine processes that influence the cycling of the previously investigated radionuclides in the Terra Nova Bay environments. Researches refer to the transfer of the above cited fission and transuranic products from the water to the benthic biomass and the sediment layer as a final point for pollution deposit.

Analyses have been carried out on samples of surface seawater, surface coastal sediments and benthic organisms *(Adamussium colbecki, Pagothenia bernacchii* and *Chionodraco hamatus)* collected during the 1993 – 94 Italian National Program for Antarctic Research (PNRA) Scientific Expedition.

A comparison with the fresh-water system of Carezza and Tarn Flat Lakes of the Terra Nova Bay inlet has been conducted by means of analyses on samples of surface water, surface sediments and unclassified algae collected both at the lake center and side.

Concentrations of the natural radionuclides K-40, Th-232 and U-238 are also reported only for the sediment samples.

The data obtained for artificial radioactivity are compared to concentrations detected in samples collected in the same period in the Mediterranean Sea.

MATERIALS AND METHODS

Analyses were performed on seawater, fish, clams and sediment samples collected in the same sites during the 1993 – 94 scientific expedition of the PNRA. During the same expedition samples of lacustrine water, sediment and unclassified algae were also collected. A complete list of the samples analyzed and collection sites is reported in Table I. In Figure 1 an overall view of the study area is also reported.

The radioanalytical methodologies carried out are summarized in Figure 2.

Gamma emitters

The surface unfiltered seawater samples have been treated by the ammonium-phosphomolybdate (AMP) tecnique[6] for the cesium isotopes separation. Each sample (200 liters volume) was acidified with HCl to pH 1, 60 mg of Cs carrier

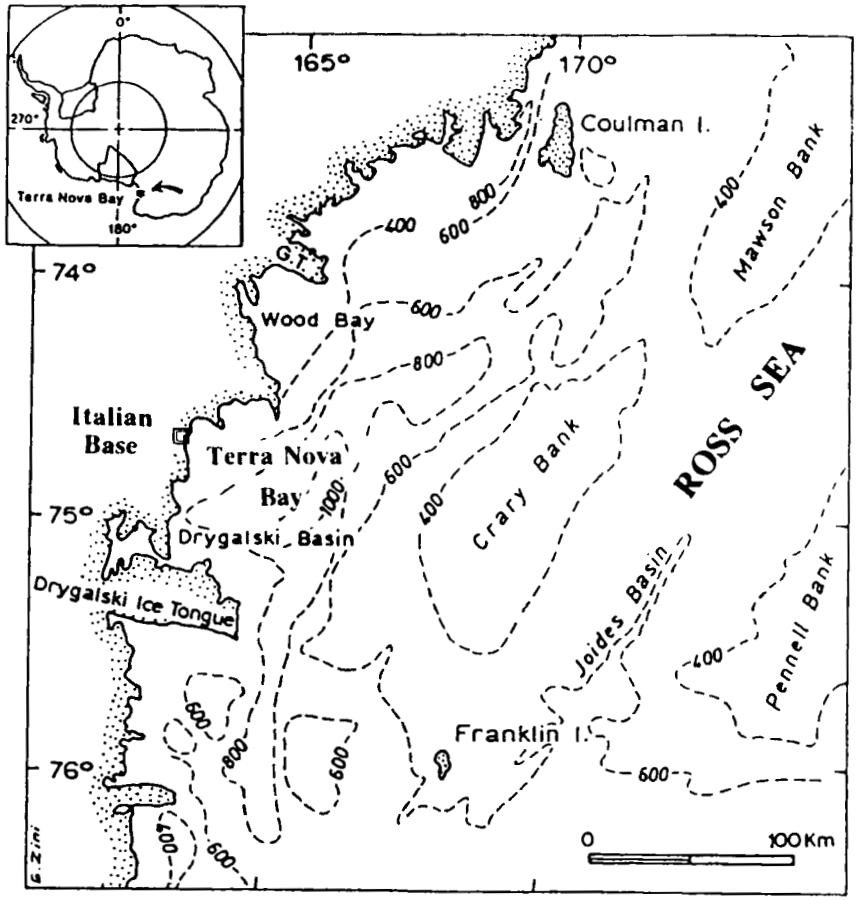

FIGURE 1 Terra Nova Bay and Ross Sea sampling area

and 10 Bq of Cs-134 spike were added to evaluate the radiochemical separation yield. After a 15–20 minute stirring to reach an isotopic equilibrium, 60 g of AMP - $(NH_4)_3PO_4 \cdot 12MoO_3 \cdot 3H_2O$ – powder (specific Cs isotopes reactant) were introduced into the solution to obtain caesium isolation. After a 3 hour slow stirring and a 48 hour settling the clarified surnatant liquid was extracted and eliminated. The AMP fraction was then centrifuged and the solid residue obtained was oven-dried at 105°C and stored into standard plexiglass source containers to be counted for gamma spectrometry. Separation yields ranged between 68% and 88%.

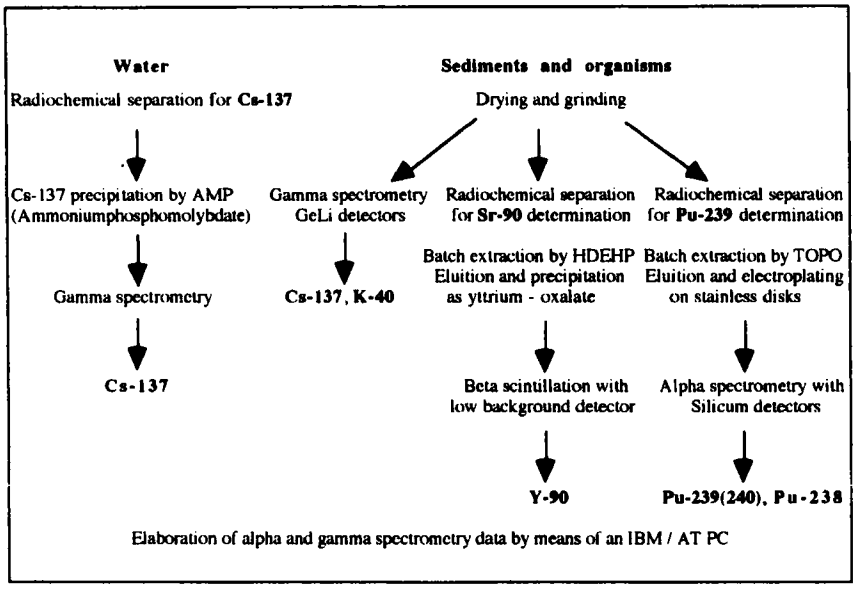

FIGURE 2 Radioanalytical methodologies tested and used

TABLE I List of samples collected during the 1993–94 PNRA Scientific Expedition

Matrix	*Sample code*	*Date*	*Depth (meters)*	*Latitude (South)*	*Longitude (East)*	*Study area*
Marine Ecosystem						
Water	SW C	28.12.93	0.5	74°42′	164°09′	Icaro Camp
	SW E	09.01.94	0.5	74°22′	165°26′	Wood Bay
Coastal	MS C	28.12.93	238	74°42′	164°09′	Icaro Camp
Sediment	MS E	09.01.94	346	74°22′	165°26′	Wood Bay
Adamussium c.	AC C	25.01.94	238	74°42′	164°09′	Icaro Camp
Pagothenia bernacchii	PB C	15.01.94	238	74°42′	164°09′	Icaro Camp
	PB C	20.01.94	238	74°42′	164°09′	Icaro Camp
Chionodraco hamatus	CH C	24.01.94	238	74°42′	164°09′	Icaro Camp
	CH C	29.01.94	238	74°42′	164°09′	Icaro Camp
Lacustrine Ecosystem						
Water	LW 17 c	04.01.94	0.5	74°43′	164°01′	Carezza Lake
	LW 17s	15.01.94	0.5	74°43′	164°01′	Carezza Lake
Sediment	LS 17	15.01.94	1	74°43′	164°01′	Carezza Lake
Algae	AG 17 s	15.01.94	0.5	74°43′	164°01′	Carezza Lake
	AG 17 c	07.02.94	1.6	74°43′	164°01′	Carezza Lake

c: center s: side

Coastal sea sediments collected by grab and box corer were oven-dried at 105°C, ground and introduced into suitable containers. Dry/humid percentages and organic matter contents were also determined[5].

Samples of soft bottom macrofauna *(Adamussium colbecki,* Mollusca Bivalvia) and ichthyofauna *(Pagothenia bernacchii* and *Chionodraco hamatus,* Osteichtyes Perciformes) were collected by dredge and nets in coastal stations of Terra Nova Bay. After biometric analyses, the samples were dissected, dried (105°C), ground and stored into source containers for gamma spectrometry.

All sources were then counted by gamma spectrometry[5] by means of high resolution (FWHM 1.8 keV at 1.33 MeV) GeLi and Ge high purity detectors (PGT Silena). Within the energy range 0.08 – 1.5 MeV, K-40 and Cs-137 were determined by direct spectrometry while Th-232 and U-238 were detected through the equilibrium descendants Bi-212 for Th, Pb-214 and Bi-214 for U. Measurement times ranged between 50 and 100 hours, error (1 sigma) was due to the counting statistics. The data were elaborated by means of a IBM Personal Computer.

Alpha emitters

Determinations of plutonium isotopes were carried out after a specific radiochemical separation that is herein briefly discussed[7,8].

A sample pretreatment was executed in relation to the matrix composition. After the preliminary treatment, the ashed matter were spiked with Pu-242 (3 dpm) as internal tracer and submitted to HNO_3 attack. Different treatments concerning batch extraction with Microthene-TOPO (tri-n-octylphosphine oxyde) and eluition with HI in HCl on chromatography columns were carried out. A classical electrodeposition on steel plates was then performed. Electroplated disks were afterwards counted for alpha emissions for two days by means of two EG&G-Ortec silicon chambers for the identification of Pu-238 (5.5 MeV), Pu-239(240) (5.15 MeV) and Pu-242 (4.9 MeV). Separation yields ranged between 20% and 35%.

Beta emitters

Strontium was determined by starting in two different ways. Either by processing the eluate from the plutonium extraction or in case this solution was not available, by doing a sample pretreatment (leaching) similar to the one used for plutonium. After the sample pretreatment, a specific separation technique[8] was carried out.

In both cases strontium and yttrium carriers were added and brought to pH 0.5 with citric acid. Y-90 (Sr-90 daughter) was then extracted with HDEHP, di-(2-ethylhexyl)-phosphate, eluted with 6M HNO_3 and precipitated with oxalic acid at pH 1.5.

Y-90 sources were therefore counted by means of an "ASPN" beta counter for 4 hours. Concentrations of Sr-90 were directly obtained by the Y-90 data after calculation of the correct decayment. Separation yields ranged between 55% and 80%[8].

RESULTS AND DISCUSSION

Concentrations of Cs-137 were generally well determinable, although often at very low activity levels. On the contrary, concentrations of Sr-90 and Pu isotopes were below the detection limit in most matrices. Quantified data were therefore determined only in one sample of sea sediment and the algae. Sr-90, Cs-137, Pu-238 and Pu-239 concentrations detected in samples of the marine and lacustrine ecosystems are reported in Table II.

TABLE II Anthropogenic radionuclides concentrations (Bq/kg dw, Bq/m^3*) detected in different Antarctic environmental samples

Matrix	*Sample code*	*Date*	*Cs-137*	*Sr-90*	*Pu-238*	*Pu-239(240)*
Marine Ecosystem						
Water*	SW C	28.12.93	0.44±0.21	–	<0.26	<0.026
	SW E	09.01.94	0.83±0.37	–	<0.15	<0.19
Sediment	MS C	28.12.93	0.26±0.05	–	<0.006	0.025±0.004
	MS E	09.01.94	0.14±0.07	0.36±0.02	<0.083	<4.28
A. colbecki	AC C	25.01.94	0.20±0.09	<0.63	<0.07	<0.10
P. bernacchii	PB C	15.01.94	0.19–1.28	–	–	–
	PB C	20.01.94	0.18–1.00	–	–	–
C. hamatus	CH C	24.01.94	0.18–0.98	–	–	–
	CH C	29.01.94	0.19–0.70	–	–	–
Lacustrine Ecosystem						
Water*	LW 17 c	04.01.94	0.38±0.21	–	–	–
	LW 17s	15.01.94	0.48±0.31	–	<3.03	<6.50
Sediment	LS 17	15.01.94	0.11±0.09	<3.21	<4.28	<5.67
Algae	AG 17s	15.01.94	15.2±0.46	5.7±0.26	<0.10	0.66±0.07
	AG 17 c	07.02.94	10.8±0.30	2.5±0.10	<0.030	0.28±0.04

Ranges of concentrations determined in fish samples to the viscera and skeleton.

Concentrations of Cs-137 in coastal seawater reflected the high cesium persistency as soluble cationic element in the pelagic environment. The higher concentrations in the water compared to the sediment seemed to confirm such statement, although we have often reported in previous papers[5,9] a possible role of particulate matter in the transport of cesium from the water to the sediment layer.

Changes in the physical form of the radionuclide could happen during proceeding months of the summer season together with increasing availability of phytoplankton. In fact, in the past we have assessed very high Cs-137 Concentration Factors in antarctic organisms at high trophic activity[9], in connection with organic matter availability.

Pu-239(240) was detected in only one sample of surface marine sediment. This value was in good agreement with plutonium determinations of previous expeditions[10]. Pu-238 was below the detection limit in all samples.

A comparison between the marine and lacustrine sediment layers showed higher Cs-137 radiocontamination in coastal sediments of Terra Nova Bay (0.14–0.26 Bq/kg dw) rather than in the Carezza Lake (0.11 Bq/kg dw). This seemed very controversial. In fact, we usually found high activity levels in lake sediments during previous campaigns and especially at Carezza Lake[2,4]. However, the LS17 sample of the 1993–94 expedition was made mostly by coarse sand and stones (0.5 – 1 cm) which have very low affinity for cesium. After sieving the sample with a 1 mm – mesh sieve, the Cs-137 concentration raised to 0.26±0.05 Bq/kg dw.

The unclassified algae certainly were the most interesting samples with very high Cs-137 concentration factors (CF: 3×10^4) and major concentrations also for strontium and plutonium isotopes. For the other environmental matrices, Sr-90 and plutonium isotopes were generally hardly detectable with values often below the detection limits.

Very high concentrations of fission products such as Sr-90 and Cs-137 had already been discussed in the past at Carezza Lake and Edmonson Point[2,5]. The well detectable concentrations of Sr, Cs and Pu and their high CF in the algae make this lacustrine matrix, together with moss and lichens[5] a good bioindicator of environmental radiocontamination of the lacustrine ecosystem.

Algae certainly play a major role in the cycling of radionuclides in the lake environments and further investigations will be necessary to assess the main biological and environmental processes that influence such uptake by the algae. The role of marine macroalgae at Terra Nova Bay has not yet been investigated.

For what concerns primordial radioactivity, concentrations of the main natural radionuclides are shown in Table III. The uranium and thorium data have been within ranges of concentrations detected during the past expeditions[1,2,4,5]. The observed concentrations of Th-232 (42 – 57 Bq/kg dry) and U-238 (17 – 59 Bq/kg dry) were calculated assuming secular equilibrium among parents and descendants of the two natural radioactive families.

TABLE III Concentrations of primordial radionuclides (Bq/kg dw) detected in samples of marine and lacustrine surface sediments

Matrix	*Sample code*	*Date*	*% Organic substance*	*Th-232*	*U-238*	*K-40*
Marine Ecosystem						
Sediment	MS C	28.12.93	2.2	47.52±1.80	24.03±0.46	945.66±6.95
	MS E	09.01.94	3.4	57.04±1.10	58.88±1.10	941.30±6.88
Lacustrine Ecosystem						
Sediment	LS 17	15.01.94	1.8	41.87±1.18	17.59±0.35	1247.1±9.01

The K-40 value in the lacustrine sample (Carezza Lake) was quite high (1247 Bq/kg dw). The same situation was observed in the algae sample of the same lake with K-40 concentrations as high as 720 Bq/kg dw. This is a further stimulus for future investigations on the role of macroalgae in the accumulation of radionuclides in the Antarctic lacustrine ecosystem. It has to be remarked that the term "lake" for the water bodies herein considered is still controversial even among the geologists who would preferably define them as "ponds", because of their small size. Their existence is limited to the period of thaw, about two months a year, due to the seasonal release of meltwater from the ice and snowfields[11].

Anthropogenic radioactivity data of marine samples collected during this expedition has been compared with results detected in similar matrixes of the Mediterranean Sea during the period 1992 – 94. The different radiocontamination levels of the two environments can be observed in Table IV.

TABLE IV Comparison between anthropogenic radiocontamination (Bq/kg dw., Bq/m^3) in the Antarctic Ocean and the Mediterranean Sea

Matrix	*Cs-137*	*Sr-90*	*Pu-239*
Antarctica (1993–94)			
Water	0.44 – 0.83	–	<0.026
Sediment	0.14 – 0.26	0.36	0.025
Organisms	0.18 – 1.28	–	<0. 10
Mediterranean (Adriatic Sea 1990–91)			
Water	3.36 – 5.18	–	–
Sediment	1.5 – 18.5	1.5 – 15	0.08 – 1.47
Organisms	1.0 – 2.57	<0.5 – 1.7	<0.01 – 0.09
Mediterranean (Aeolian Is. 1994–95)			
Water	2.92 – 3.2	–	–
Sediment	0.32 – 7.8	0.5 – 1.5	0.23
Organisms	–	–	–

In general the radiocontamination of the Mediterranean Sea is 5 – 10 times higher than Terra Nova Bay. We remind that just after the Chernobyl accident the difference was even as high as 100 – 1000 times[10].

Within the Antarctic marine environment our results on Cs-137 were in good agreement with data reported by Hashimoto et al.[12] for the same study area (Ross Sea) but were lower than the ones detected by Holm[13] and Roos et al.[14] at the Antarctic Peninsula (Weddel Sea). In spite of the different contamination levels between the two Antarctic areas, high Cs-137 concentration levels and Concentration Factors were also reported for several organisms of the Antarctic Peninsula[13,14]. Such observation has confirmed the results we have obtained during several expeditions on the opposite side of Antarctica. These remarkable Cs-137 levels in the biota will stimulate future investigations directed to the evolution of environmental processes and ecological mechanisms that influence the radionuclide accumulation in some Antarctic organisms.

Acknowledgements

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