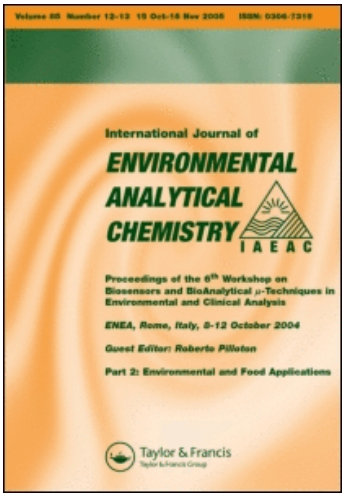

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Evolution of persistent Anthropogenic radioactivity in Antarctic ecosystems

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EVOLUTION OF PERSISTENT ANTHROPOGENIC RADIOACTIVITY IN ANTARCTIC ECOSYSTEMS

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The primary objective of this study was to observe the evolution of anthropogenic radioactivity contamination in the Antarctic continent throughout the period 1997–1999. Moreover, results have been compared with those obtained for previous expeditions, starting from 1987. As far as ^{137}Cs is concerned, interesting considerations could be made due to the great amount of available data. On the whole, radioactive contamination seems to be higher in continental than in marine environments. For lake algae, contamination seems to decrease gradually in the order: Tarn Flat, Edmondson Point, Carezza Lake. Focusing on ^{137}Cs activity data, a clear temporal decreasing trend was observed in all samples: for sea water, values decreased from mean values of 0.9 Bq/m^3 in 1987 to 0.5 Bq/m^3 in 1999, a 56% decrease (20% of the total is due to natural decay of ^{137}Cs). For lake waters and lake algae, the decreases are higher (80 and 30%, respectively) and the same can be assessed for sediments and soils, even if the resulting distributions are more complicated. The highest values for all radionuclides analysed were detected in terrestrial organisms (mosses, lake algae, and lichens). As a consequence, these matrices appear to be good bioindicators of radioactive contamination. Finally, although the Antarctic continent is affected by some degree by anthropogenic radioactive pollution, our results for ^{137}Cs show that we are facing a progressive decrease. Moreover, contamination in other parts of the world is much higher: from 6–10 times in the Mediterranean Sea and 20–50 times in the North Sea and Black Sea.

Keywords: Radioecology; Pollution; ^{137}Cs; Plutonium; ^{241}Am

INTRODUCTION

The Antarctic continent, despite what is widely thought, is not free from pollution which heavily characterizes the northern hemisphere and the industrialized Australian territories [1–3]. The low population density is not sufficient to ensure an uncontaminated environment, even as far as anthropogenic radiocontamination is involved [4–8], mainly because of global atmospheric and oceanic circulation

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of air and water masses, which transport materials and polluted substances in a north–south direction.

In previous decades, as a part of the Italian National Research Programme in Antarctica (PNRA), the Laboratory of Radioecology of the University of Parma has been strongly involved in the measurement of artificial and natural radionuclides in samples from continental and marine environments (sediments, waters, and organisms). First, attention was focused on ^{137}Cs (the most abundant) and several natural radionuclides (^{40}K, ^{232}Th, and ^{238}U), and only recently have highly toxic and artificial radionuclides with very long decay times (^{238}Pu, $^{239+240}Pu$, ^{241}Am, and ^{90}Sr) been analysed.

In this paper, we present the results from 1997–1999 PNRA scientific expeditions and discuss the evolution of anthropogenic radioactivity contamination in the Antarctic continent throughout the period 1987–1999.

EXPERIMENTAL

All samples were collected during two different scientific expeditions (1997–1998 and 1998–1999). Figure 1 shows the continental and marine sample sites.

Soils were collected by hand, and grab hooks were used for marine and lacustrine sediments. *Pagothenia bernacchii, Adamussium colbecki*, *Chionodraco hamatus*, *Euphausia superba*, and *Iridea cordata* were the marine organisms considered in this study. The various species of lichens were *Usnea antractica, Candelaria* sp., *Lepraria* sp., and *Umbilicaria* sp. Mosses and lacustrine algae were not classified.

Only seawater samples needed to be pre-treated before any radiochemical analyses, concentrating large volumes of water using different techniques: the ammonium

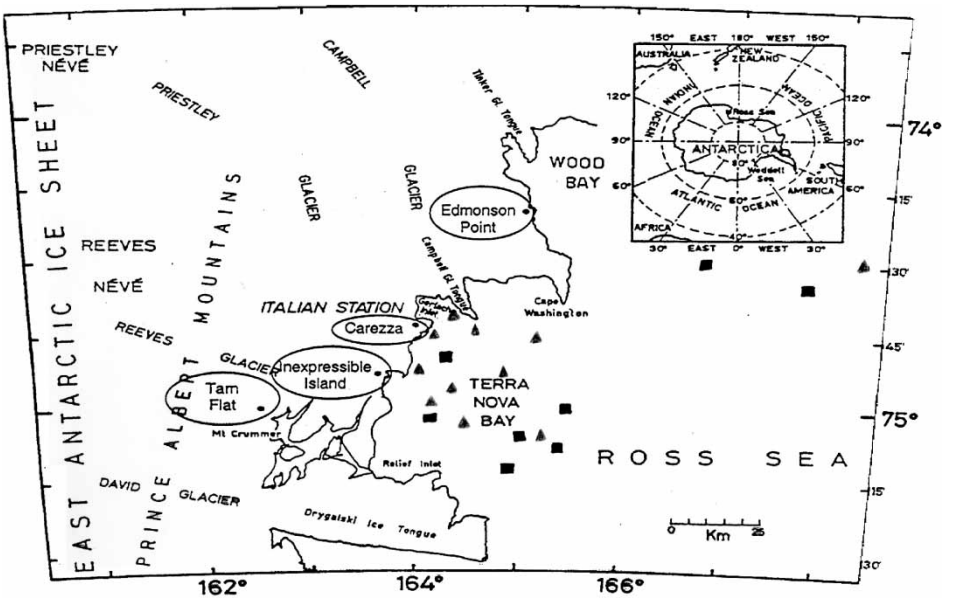

FIGURE 1 Sample sites. Circles represent lake areas where terrestrial and lacustrine samples have been collected. Triangles and squares denote superficial water and sediments sample sites, respectively.

phosphomolibdate method for ^{137}Cs, and the iron hydroxide (III) method for $^{239+240}$Pu, ^{238}Pu, and ^{241}Am [9]. Subsequent procedures were the same for all matrices, starting with oven drying at 105°C for 24 h.

For γ-emitters (^{137}Cs and all natural nuclides), activity in all sample types was determined directly using gamma spectrometry with high-resolution (FWHM 1.8 keV at 1.33 MeV), Ge(Li)- and Ge high-purity detectors (PGT Silena). ^{232}Th activities were determined through the equilibrium descendant ^{212}Bi. Counting times were generally set in 2–3 days. The detection limit was 0.01 Bq/kg, and the detection efficiency was 50–80%.

For the analyses of ^{90}Sr (beta emitters), activity was determined after a complex radiochemical separation: batch extraction with di-(2-ethylhexyl)orthophosphoric acid (HDEHP), precipitation with yttrium oxalate, and measurement of ^{90}Y (descendant of ^{90}Sr) activity through low-level beta scintillation.

Finally, isotopes of Pu and ^{241}Am, both α-emitters, were determined by a specific method, consisting of a previous ashing at 600°C, then acid digestion, passage through Microtene-tri-normal-octylamine (TNOA) and/or HDEHP columns, and electrodeposition on steel plates. Deposition conditions (voltage and time) are different between Pu and Am. Alpha counting was carried out by means of two EG&G Ortec silicon chambers for 4–5 days. The detection limit was 0.0003 Bq/kg, and the detection efficiency spanned 30–60%. For specific details, see [10,11].

RESULTS AND DISCUSSION

Tables I and II summarize the results for ^{137}Cs and natural radionuclides of the samples collected during the PNRA scientific expeditions 1997–1998 and 1998–1999. Data for $^{239+240}$Pu, ^{241}Am, ^{90}Sr, and ^{137}Cs from 1987 to 1996 [9] are listed in Table III. Our discussion will also include previous ^{137}Cs results obtained by the same research group of the Laboratory of Radioecology [4–8].

Contamination seems to be higher in continental environments (see Tables I–III). Marine environments are less polluted, probably because of the presence of a strong oceanic circulation, which helps the dilution and autodepuration mechanisms. Concerning the lacustrine environment, Tarn Flat algae are more contaminated than those of Edmondson Point, and Carezza Lake algae are the least polluted of all [4–9]. For clarity, Fig. 2 shows a histogram with the mean concentrations of ^{137}Cs measured in matrices both from the continent and from the sea.

The following paragraphs detail the results obtained for each matrix.

TABLE I Natural and anthropogenic radionuclide concentrations in terrestrial and lacustrine samples[a]

Sample	*Date of collection*	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
LW13	18/02/98	n.d.	n.d.	n.d.	0.19 ± 0.11
LV14	30/12/98	4.23 ± 0.7	24.07 ± 0.9	261 ± 16	7.08 ± 0.52
LS13	18/02/98	47.9 ± 1.7	70.4 ± 2.0	1028 ± 52	1.80 ± 0.2
LS14 (fine fraction)	30/12/98	23.0 ± 0.9	54.1 ± 1.6	1464 ± 73	< 0.5
LS14 (gross fraction)	30/12/98	22.7 ± 0.8	35.0 ± 1.0	1323 ± 65	< 0.3
TV13 (soil)	18/02/98	46.7 ± 1.2	71.4 ± 2.1	955 ± 49	13.9 ± 0.8
TV13 (mosses)	18/02/98	36.1 ± 1.4	61.3 ± 2.0	829 ± 44	12.6 ± 0.8

[a]Data refer to scientific expeditions 1997–1998 and 1998–1999. Results are in Bq/kg dry weight, with the exception of lake waters (Bq/m^3). LW, lake waters; LV, lake algae; LS, lake sediments; TV, terrestrial vegetal. n.d., not determined.

TABLE II Natural and anthropogenic radionuclide concentrations in marine samples[a]

Sample	*Date of collection*	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
SWY1 (sup.)	07/12/97	n.d.	n.d.	n.d.	< 0.57
SW13 (2 m)	24/11/97	n.d.	n.d.	n.d.	0.53 ± 0.38
SW13 (sup.)	07/02/98	n.d.	n.d.	n.d.	0.50 ± 0.44
MV13	08/02/98	1.3 ± 0.3	1.8 ± 0.2	506 ± 35	< 0.4
MA13-TB	12/02/98	< 1.5	< 2.6	332 ± 14	0.7 ± 0.2
MA13-AC (shells)	04/02/98	< 0.1	1.1 ± 0.2	< 0.6	< 0.4
MA13-AC (flesh)	04/02/98	< 2.1	< 2.9	363 ± 26	< 0.9
MA13-CH	17/11/97	< 1.5	< 2.3	302 ± 17	< 0.6
MA-PHN Krill	23/12/97	n.d.	n.d.	422 ± 20	0.17 ± 0.09
MS13 (430 m)	03/02/98	24.5 ± 1.1	48.2 ± 1.6	614 ± 37	< 0.4
MS13 (420 m)	22/11/97	35.3 ± 1.3	63.6 ± 1.8	647 ± 34	< 0.5
MS13 (220 m)	17/11/97	16.5 ± 0.6	31.9 ± 1.7	792 ± 40	0.32 ± 0.10
MSY1 (1002 m)	08/12/97	19.6 ± 0.5	44.3 ± 1.3	772 ± 28	< 0.3

[a]Data refer to scientific expeditions 1997–1998 and 1998–1999. Results are in Bq/kg dry weight, with the exception of sea waters (Bq/m^3). SW, sea waters; MV, marine algae; MA, marine animals; MS, marine sediments. n.d., not determined.

TABLE III $^{239+240}$Pu, ^{241}Am, ^{90}Sr, and ^{137}Cs concentrations in marine and terrestrial samples[a]

Sample	*Date of collection*	$^{239+240}Pu$	^{241}Am	^{90}Sr	^{137}Cs
SW1	15/01/96	8.1 ± 1.0	2.5 ± 0.5	n.d.	0.39 ± 0.25
SW2	23/01/96	0.64 ± 0.07	0.29 ± 0.07	n.d.	0.09 ± 0.02
SW3	03/02/96	0.71 ± 0.09	0.53 ± 0.12	n.d.	0.06 ± 0.03
A12	15/01/91	0.658 ± 0.026	0.305 ± 0.023	3.30 ± 0.34	37.2 ± 2.6
A17	15/01/91	0.201 ± 0.009	0.040 ± 0.014	n.d.	10.4 ± 0.41
A15	15/01/96	0.417 ± 0.019	0.149 ± 0.013	9.31 ± 0.90	17.6 ± 2.2
Fish (flesh)	15/01/89	0.0455 ± 0.0054	0.0087 ± 0.0026	0.11 ± 0.08	0.97 ± 0.30
Fish (bones)	15/01/90	0.0010 ± 0.0005	0.0006 ± 0.0004	0.08 ± 0.03	0.30 ± 0.15
MS22	29/12/90	0.0056 ± 0.0017	0.0066 ± 0.0014	0.21 ± 0.08	0.96 ± 0.08
MS38	07/01/91	0.0169 ± 0.0022	0.0074 ± 0.0014	0.16 ± 0.05	0.36 ± 0.04
MS77	02/02/95	< 0.0003	0.0024 ± 0.0013	< 0.10	< 0.10
MSC	25/01/95	0.0806 ± 0.0051	0.0440 ± 0.0028	< 0.18	0.40 ± 0.04
MS104	10/02/95	< 0.0003	0.0008 ± 0.0005	< 0.10	< 0.17
LS8	15/01/91	0.0969 ± 0.0068	0.0298 ± 0.0030	0.56 ± 0.09	2.87 ± 0.36
LS17	15/01/91	0.0051 ± 0.0010	0.0036 ± 0.0008	0.31 ± 0.04	0.27 ± 0.19
LS12	15/01/92	0.0057 ± 0.0009	0.0023 ± 0.0009	0.41 ± 0.04	0.14 ± 0.09
LS15a	94–95	0.0327 ± 0.0029	0.0144 ± 0.0020	0.18 ± 0.05	1.16 ± 0.07
LS15a	94–95	0.0801 ± 0.0065	0.0412 ± 0.0047	0.76 ± 0.09	3.93 ± 0.35
TS8-1	91–92	0.0190 ± 0.0020	0.0130 ± 0.0016	0.20 ± 0.04	1.40 ± 0.08
TS8-2	91–92	0.0010 ± 0.0004	0.0010 ± 0.0005	0.08 ± 0.04	< 0.12
M1	15/01/89	0.908 ± 0.096	0.614 ± 0.084	16.2 ± 1.9	41.4 ± 2.5
M2	15/01/89	0.788 ± 0.096	0.216 ± 0.049	14.5 ± 2.2	29.2 ± 9.4
M3	15/01/89	0.435 ± 0.061	0.210 ± 0.042	24.4 ± 3.2	49.9 ± 3.7
M15	15/01/89	0.637 ± 0.028	0.276 ± 0.012	18.8 ± 1.3	20.7 ± 2.0
M14	15/01/91	0.328 ± 0.007	0.178 ± 0.013	9.2 ± 0.5	11.3 ± 1.7
L1	15/01/89	4.64 ± 0.38	1.94 ± 0.14	n.d.	162 ± 4

[a]Data refer to scientific expeditions from 1988 to 1996. Results are in Bq/kg dry, with the exception of sea waters (Bq/m^3 for ^{137}Cs and mBq/m^3 for $^{239+240}$Pu and ^{241}Am). SW, sea waters (SW2 and SW3 are filtered samples). A, marine algae; MS, marine sediments; LS, lake sediments; TS, terrestrial soils; M, mosses; L, lichens; n.d., not determined.

Water (Sea and Lake)

Concentrations for all nuclides are higher in unfiltered seawater samples (SWY1, SW13 in Table II, SW1 in Table III) than in the others, thus showing the very high affinity of these elements for the particulate matter in the water column: with these particles,

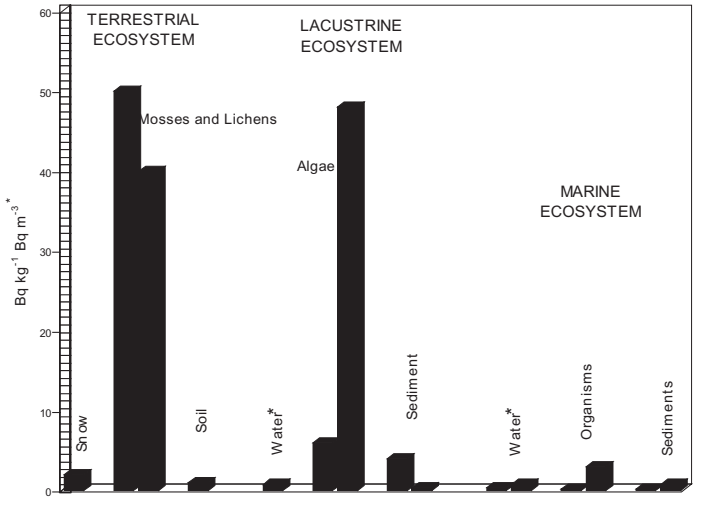

FIGURE 2 Maximum and minimum ^{137}Cs concentrations measured in terrestrial, lacustrine, and marine samples.

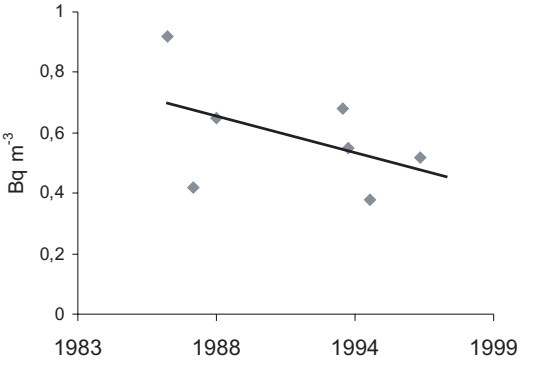

FIGURE 3 Evolution of ^{137}Cs concentration in sea waters (1988–1998). Data are mean annual values.

adsorption and sinking to the bottom are the principal phenomena which remove radionuclides from the water and are commonly known as scavenging processes.

Anthropogenic radioactive contamination is present in this environment, but in the last 12 years (from 1987 to 1999), we have registered a progressive decreasing in seawater (from a mean value of 0.9 to 0.5 Bq/m^3; see Fig. 3) equal to 56% (20% of the total is due to natural decay of ^{137}Cs). For lake water, the decrease is much more evident (80%; see Fig. 4).

Sediments and Soils

As mentioned earlier, values are generally higher in the lacustrine sediments than in the marine sediments and soils. This is evidence of a more contaminated terrestrial-lacustrine environment. Soil collected under mosses and lichens (TV13; Table I) shows a considerably high activity, for ^{137}Cs above all.

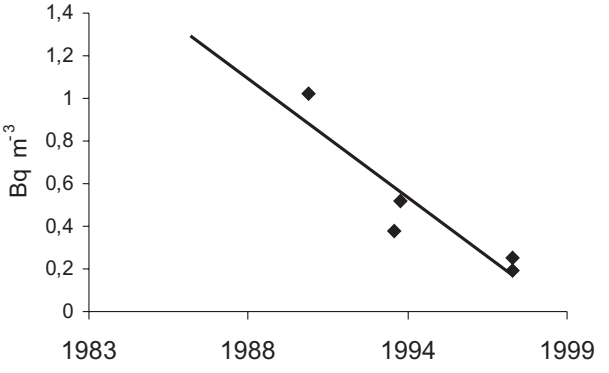

FIGURE 4 Evolution of ^{137}Cs concentration in lake waters (1991–1998). Data are mean annual values of all lakes.

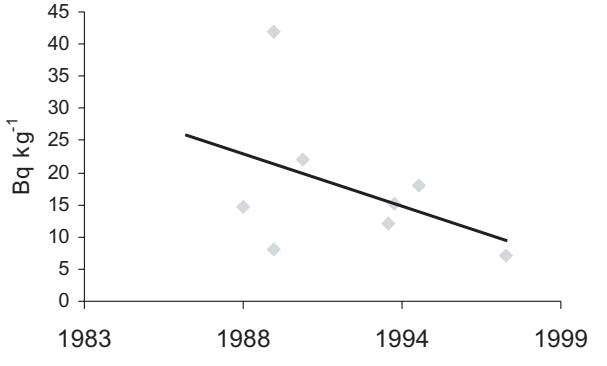

FIGURE 5 Evolution of ^{137}Cs concentration in lake algae (1991–1998). Data are mean annual values of all lakes.

Even if the distribution is much more complicated than that observed for waters, for superficial marine samples we have also noticed a progressive decrease in Cs-137 contamination levels, from 0.4–1.5 Bq/kg dry weight in 1988 to 0.3–0.4 in 1998.

Mosses, Lichens, and Algae

Values obtained for these matrices are the highest of all samples analysed in this research, decreasing in the order: lichens, mosses, and algae. As a consequence, all these organic matrices can be considered good bioindicators of radioactive pollution, principally because they accumulate these elements from the environment during growth. Therefore, high activity values are due to the specific behavioural and physiological characteristics of these organisms: high surface/volume rate, low growth rate, and long life.

For lake algae, the decrease in ^{137}Cs concentration in time is fairly high (30%), as illustrated in Fig. 5.

TABLE IV Concentrations levels of anthropogenic radionuclides (Bq/m^3, Bq/kg dry weight) in marine matrices from Antarctica and several Mediterranean Seas

Sample	^{137}Cs	^{90}Sr	$^{239+240}Pu$	^{238}Pu	^{241}Am
Antarctica					
B.T.N.-Ross Sea 1990–1996					
Water (surface)	0.44–0.89		0.0058–0.0067	0.00013	0.0005–0.0025
Sediment (surface)	0.14–1.50	< 0.10–0.36	0.0056–0.0908	0.0014–0.0044	0.0024–0.0440
Animals	0.18–1.28	0.08–0.11	< 0.0022–0.0455	< 0.0022–0.013	0.0006–0.0089
Mediterranean Sea					
Adriatic Sea 1990–1996					
Water (surface)	3.36–5.18		0.013–0.025	0.0009–0.0074	
Sediment (surface)	1.50–18.5	< 0.16–0.51	0.080–0.683	0.004–0.022	0.036–0.188
Animals	1.00–2.57		< 0.01–0.034	0.0020	
Po Delta 1990–1996					
Water (surface)	2.8–4.3				
Sediment (surface)	9.90–31.3	< 0.16–5.80	0.68–1.23	0.02–0.03	0.037–0.042
Animals	1.00–9.00		0.034	0.002	
Eolie Islands 1994–1995					
Water (surface)	2.92–3.20		0.012–0.020		
Sediment (surface)	0.32–7.8	0.5–1.5	0.11–0.23	0.0066–0.0089	0.029–0.161

Marine Organisms

Very briefly, it seems that autodepuration processes of the marine ecosystem work well, and only small ^{137}Cs activities could be detected for *Pagothenia bernacchii* and *Euphausia superba*. In the marine macroalga *Iridea cordata,* ^{137}Cs concentrations were found to be very low.

Finally, our results show that even the Antarctic continent suffers from radioactive pollution, even though ^{137}Cs concentrations show a progressive decrease. Moreover, contamination in other places of the world is higher: 6–10 times higher in the Mediterranean Sea (see Table IV) and 20–50 times higher in the North Sea and Black Sea [12].

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