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ANTHROPOGENIC RADIONUCLIDES IN PLANTS, ANIMALS AND THEIR ENVIRONMENTS IN ANTARCTICA

© S. Giuliani, C. Triulzi, M. Vaghi

Laboratory of Radioecology, Department of Evolution and Functional Biology, University of Parma, Italy

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In the framework of the Italian National Research Program in Antarctica (PNRA), the Laboratory of Radioecology of the University of Parma developed from 1987 several researches on terrestrial and marine samples. At first the principal aim was the study of ^{90}Sr, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ and ^{241}Am concentrations in samples of soil, ice, seawater and biota collected near the Italian Antarctic Base (Ross Sea, Terra Nova Bay).

Key words: Antarctica, pollution, ^{137}Cs, $^{239+240}Pu$, ^{238}Pu and ^{241}Am

Начиная с 1987 г., Лаборатория радиоэкологии университета г. Парма выполнила ряд исследований в рамках Итальянской Национальной Антарктической программы. Основной задачей было изучение содержания антропогенных радионуклидов ^{90}Sr, ^{137}Cs, ^{238}Pu, $^{239+240}Pu$ и ^{241}Am в пробах грунта, льда, морской воды, а также в гидробионтах, собранных в районе Итальянской Антарктической станции (море Росса, бухта Терра-Нова).

Ключевые слова: Антарктика, загрязнение, ^{137}Cs, $^{239+240}Pu$, ^{238}Pu, ^{241}Am

As is widely known, the progresses of the research in the nuclear field and the use of this kind of energy for civil and military purposes caused, in the last decades, the worldwide diffusion of anthropogenic radionuclides.

Up to 1945 radionuclides in the environment were solely of natural origin (see Fig.1, left). Cosmogenic radionuclides (^{3}H, ^{7}Be and ^{14}C) are produced by the interaction between the atmosphere and cosmic rays, whereas primordial ones are present on earth's rocks and, by diffusion in the atmosphere, they get in touch with all the other components of the biosphere. Major elements of this kind are ^{40}K, ^{87}Rb and $^{147,148,149}Sm$. The natural radioactive series of ^{232}Th, ^{235}U and ^{238}U together with their descendants (i.e. radon and toron gases) are also very important.

Anthropogenic radionuclides contamination was principally caused by radioactive fallout generated by nuclear and thermonuclear essay explosions in the atmosphere (see Fig.1, right). At the beginning, in 1945 (Tab.1), USA tested some weapons in the Nevada Test Site, contaminating the part of territory.

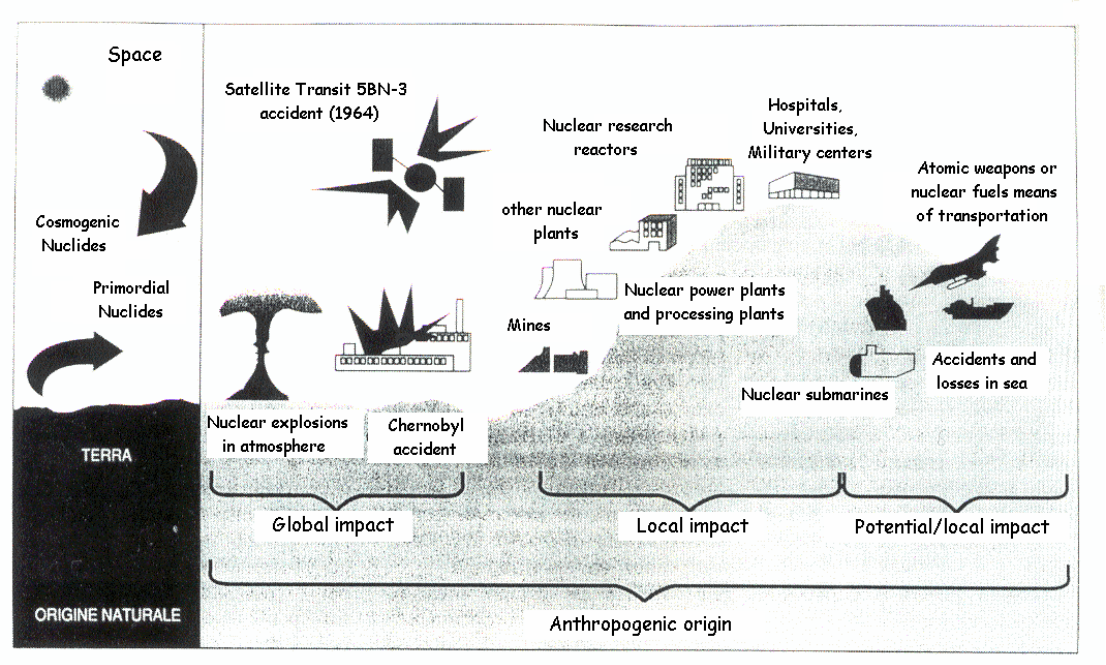


Fig.1. Origin and sources of natural and anthropogenic radionuclides
Рис. 1. Происхождение и источники природных и антропогенных радионуклидов

From that moment, US nuclear experiments are continued in the Marshall Islands and in other Pacific equatorial and sub-equatorial sites. UK did the same in Christmas Islands and Bikini Islands, whereas USSR testing site was Semipalatinsk, at the border between Mongolia and Siberia. Chinese weapons were tested at Lop Nor Lake, near the border with USSR and also France made some nuclear experiments: in the early 60's in the Sahara Desert and then at Mururoa in Polinesia. The radioactive red rain caused by explosions in the desert was detected even by our laboratory in Parma (Italy).

The nuclear agreement between USA, UK and USSR in 1962 stopped nuclear explosions in the atmosphere but permitted to continue the experiments in caves, not contaminating the environment. China did not sign the agreement and continued the tests in the atmosphere until 1980. In recent years, other countries (i.e. India and Pakistan) produced nuclear bombs and tested them in caves. The well-known "Chirac Bombs" in Mururoa have been the last test explosions. They did not contaminate the environment because the energy was low and they were performed 1000 m below sea level.

On the whole, 423 nuclear explosions were realized from 1945 to 1980 in the Northern and Southern Hemisphere and in the equatorial belt. 25 - 26 % of the radioactive fallout generated by these explosions reached the Southern Hemisphere and part of it contaminated Antarctica.

Other possible radioactive contamination sources can be both satellites that fall down to earth carrying ^{238}Pu batteries and nuclear power plants accidents like the one that happened in Chernobyl (26.04.1986). As far as this accident is concerned, the impact was really large in extension but was restricted to the lower part of the atmosphere: the toxic cloud did not reach the stratosphere but has risen on some kilometers in

height, involving most of Central Europe, Russia and Baltic regions territories. The winds that generally blow eastward to Moscow, in those days pushed the contamination in the contrary direction, reaching Greece, Italy, Central Europe and Sweden. The latter was the first country to realize that something strange had happened, even before the soviet authorities had informed the international community. Emergency alarms in some Swedish nuclear plants rang in the outside, whereas the situation inside was under control. That was the way they discovered that the problem was not due to their own plants but was coming from far away, and gave the alarm. Chernobyl was a particular nuclear power station, because it had not a second container. The remaining 220 nuclear stations in Europe and 200 ones in the rest world have such container.

Other local contamination sources can derive from Uranium and Thorium production cycles, from mines to processing plants (see Fig.1, right). Radioactive wastes must be safely collected and isolated by law, in order not to pollute the environment.

Nuclear energy can be also used for pacific purposes in many civil fields, i. e. sanitary, agriculture and research fields, but they also can produce a local and limited impact. Finally, in the sea, we can find other contamination sources, most of all regarding ice-breaker navies and nuclear submarines, like the Kursk (see Fig.1).

As a consequence, the variations of radioactivity levels measured in the environment since the beginning of the nuclear era, led to the development of many scientific studies. At first, the principal aim was human health protection and only in a second time biogeochemical cycles analyses and large-scale effects on the environment were studied, leading to the birth of a new scientific branch: Modern Radioecology.

As to Antarctica, different studies [3, 8, 16], some of them also regarding anthropogenic radioactive contamination [9, 10, 13, 14, 15], contradicted the general belief that the continent was free from pollution of any kind. For this reason, radioecological studies are really important also to evaluate transfer processes and anthropogenic contaminants distribution in all environmental matrices and to define temporal trends and evolutions of this kind of pollution.

Since 1960 the Laboratory of Radioecology of the University of Parma is involved in the study of marine and terrestrial ecology. In the framework of the Italian PNRA (National Research Program in Antarctica), the laboratory developed from 1987 several researches on terrestrial and marine samples. At first the principal aim was the study and measurement of ^{90}Sr and ^{137}Cs concentrations, together with some natural radionuclides (^{40}K, ^{232}Th and ^{238}U). ^{90}Sr and ^{137}Cs are the most abundant fission products and their half-lives ($t_{1/2}$) are 28.2 and 30.2 years, respectively. Only in a second time other anthropogenic radionuclides were analysed, all characterized by very long $t_{1/2}$ and high toxicity: ^{238}Pu, $^{239+240}Pu$ and ^{241}Am. Tab.2 summarizes all the matrices analysed during 12 years of work in this field.

The present study will try to focus the attention on anthropogenic radioactive contamination levels in terrestrial, lake and marine environment of Antarctica and to observe the evolution of this contamination in 1987 - 1999.

Material and methods. Continental sample sites are represented in Fig.2, whereas Fig.3 shows marine ones. The four lakes analyzed are very little and shallow (from 2 to 4 m depth), and frozen for most time of the year. Terrestrial algae come from the lakes, whereas mosses and lichens were collected in the neighbourhood.

Table 2. Antarctic environmental matrices analysed from 1987 to 1999
Таблица 2. Антарктические образцы, проанализированные в 1987 - 1999 гг.

Terrestrial Ecosystem	Snow Ice cores Lichens (*Usnea Antarctica*, *Lepraria* sp and *Umbilicaria* sp) Mosses Soil under mosses and lichens Soil cores
Lake Environment	Water Macroalgae Superficial Sediments Sediment cores
Marine Ecosystem	Water Water under pack Coastal and open ocean Sediments Macroalgae (*Iridea cordata)* Krill (*Euphasia superba)* Coastal macrofauna (*Adamussium colbecki)* Fishes (*Pagothenia bernacchii* and *Chionodraco amatus)*

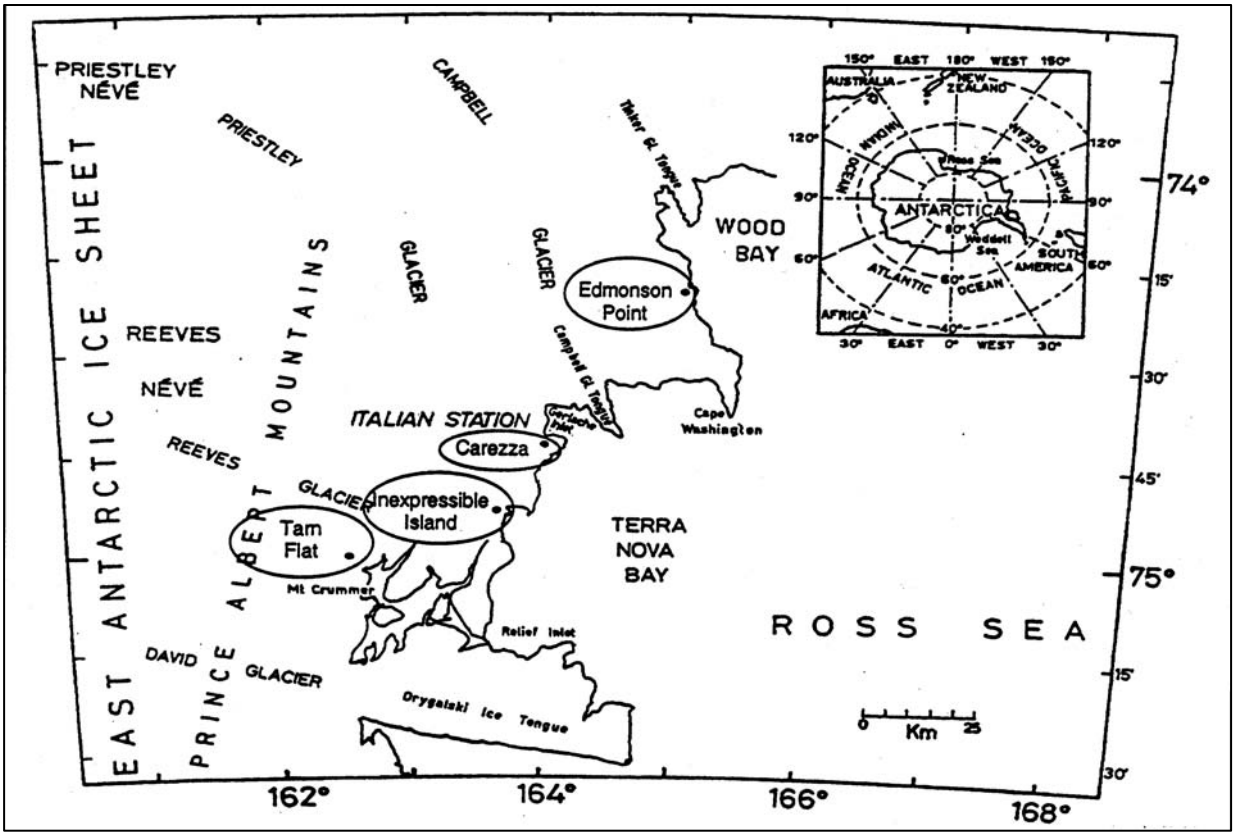
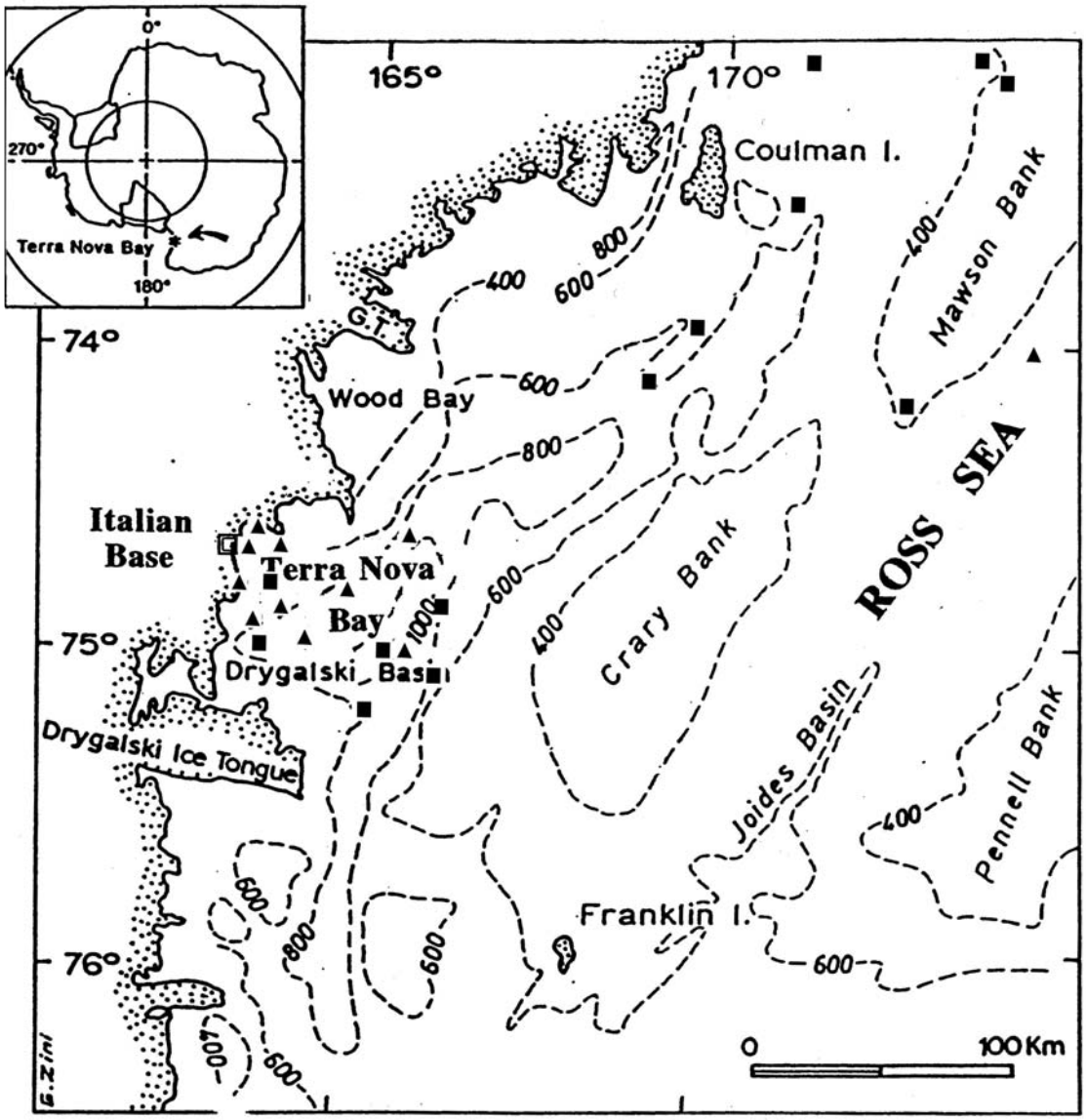


Fig. 2. Continental sample sites. Circles indicate lake areas
Рис. 2. Места отбора континентальных проб. Точками обозначены районы озер

Most of the marine samples come from Terra Nova Bay and some other samples were collected in the subantarctic zone (see Fig.3).



■ Станции отбора проб донных отложений

▲ Станции отбора проб поверхностной воды

Fig. 3. Marine sample sites (σ - superficial water samples, ν - superficial sediment samples)
Рис. 3. Места отбора морских проб

Terrestrial sediments have been collected by hand, whereas for marine and lacustrine ones were used grab hooks. As far as organisms are concerned, we considered *Pagothenia bernacchii, Adamussium colbecki, Chionodraco hamatus, Euphausia superba* and *Iridea cordata.* The species of lichens that have been recognized were *Usnea antractica*, *Candelaria* sp., *Lepraria* sp.

and *Umbilicaria* sp. Mosses and lacustrine algae were not classified.

Radiometric and radiochemical methods are summarized in Fig. 4. For the specific details see [5, 6].

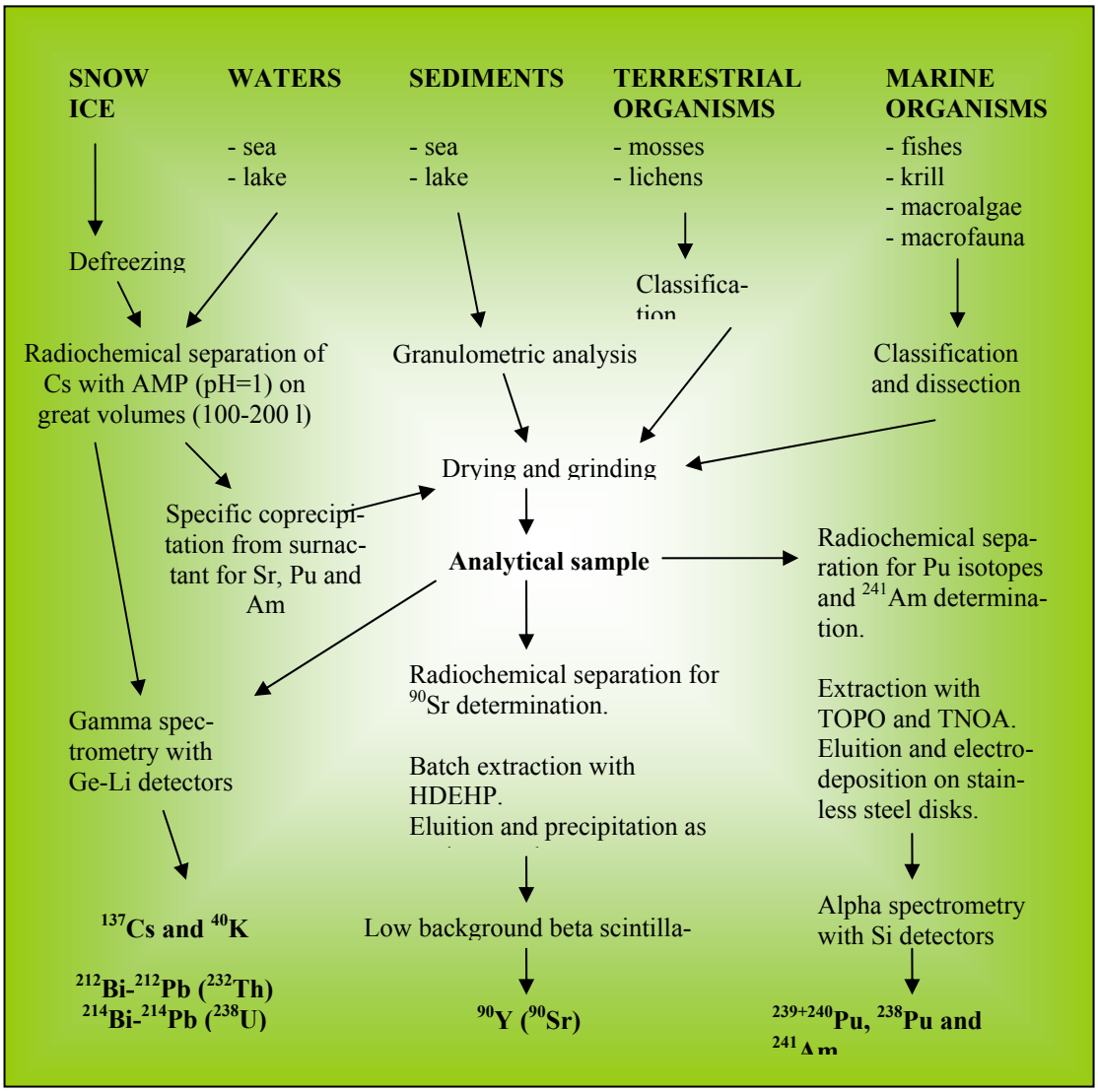


Fig. 4. Radiometric and radiochemical methods
Рис. 4. Радиометрические и радиохимические методы

Only seawater samples need to be pre-treated before any radiochemical analyses, con-centrating great volumes of water using different techniques: for ^{137}Cs we used the Ammonium-phosphomolibdate method, whereas, for $^{239+240}$Pu, ^{238}Pu and ^{241}Am we used the Iron hydroxide (III) one [7].

For ^{137}Cs and all natural radionuclides the determination of activity in all sample types is made directly with gamma spectrometry by means of high resolution (FWHM 1,8 Kev at 1.33 Mev), Ge(Li) and Ge hih purity detectors (PGT Silena).For the analyses of ^{90}Sr, activity is deter-mined after a complex radiochemical separation: batch extraction with HDEHP, precipitation

with yttrium oxalate and measurement of Y-90 (descendant of ^{90}Sr) activity through low level beta scintillation.

Isotopes of Pu and ^{241}Am are determined through a specific method, which consist in a previous ashing at 600°C, then acid digestion, passage through Microtene-TNOA and/or HDEHP columns and electrodeposition on stainless steel disks. Deposition conditions (voltage and time) are different between Pu and Am. Alfa counting was carried out by means of two EG&G Ortec silicon chambers. To calculate the efficiency of separation we added to the analytical samples the following spikes: ^{85}Sr, ^{134}Cs, ^{242}Pu and ^{243}Am.

Results and discussion. In general, for ^{137}Cs, contamination seems to be higher in the continental environments than in the marine one (see Fig. 5) and the highest values of all were measured in mosses and lichens. As a consequence these organisms can be considered good bioindicators of radioactive pollution, principally because they accumulate these elements from the environment during growth.

High activity values are due to the specific behavioural and physiological characteristics of these organisms: high surface/volume ratio, low growth rate and long life.

Tab. 3 resumes ranges of contamination levels measured from 1987 to 1999.

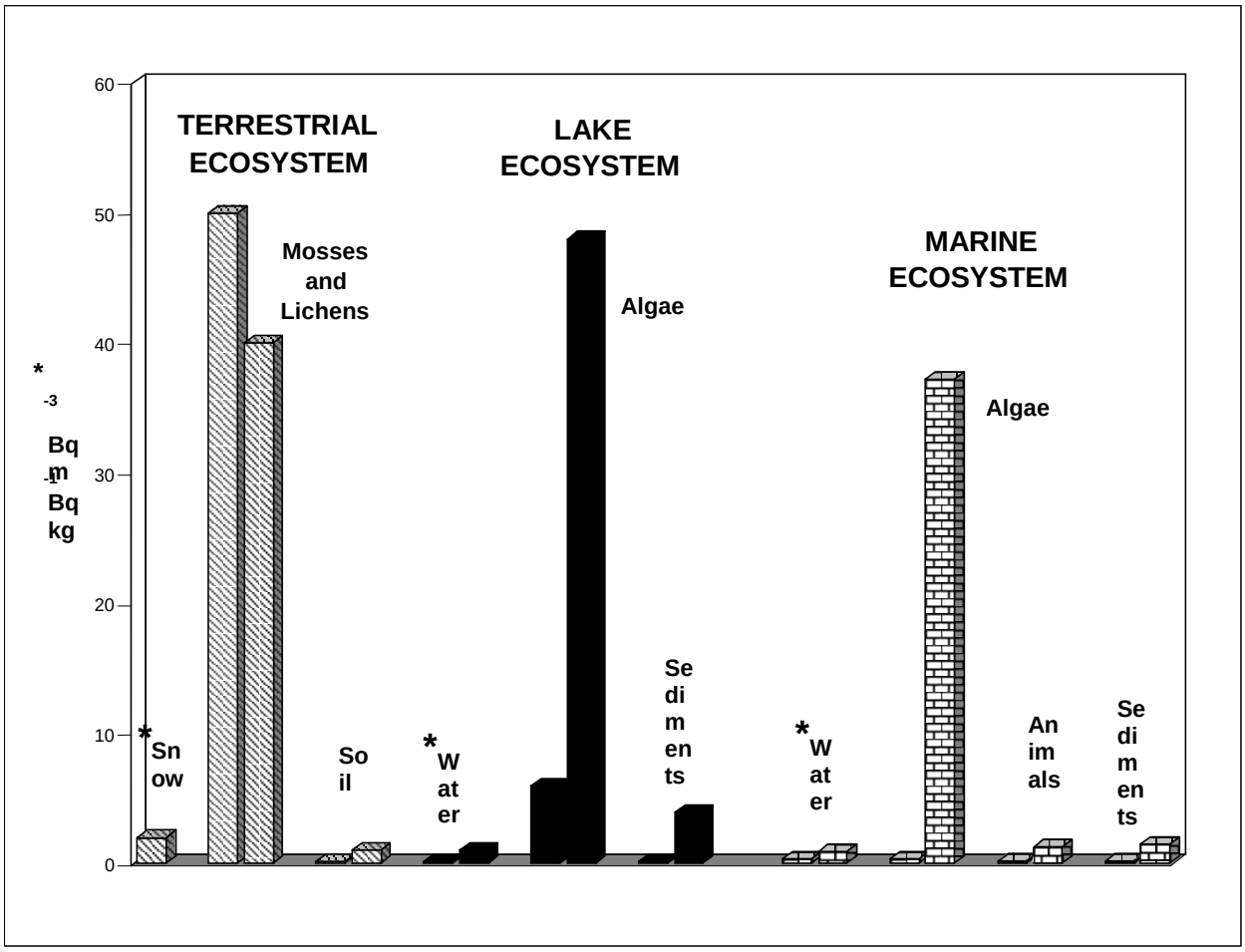


Fig. 5. Maximum and minimum concentration levels of ^{137}Cs measured in terrestrial, lacustrine and marine samples
Рис. 5. Максимальные и минимальные уровни содержания ^{137}Cs в сухопутных, озерных и морских пробах

Table 3. Anthropogenic radioactive contamination ranges measured in the matrices collected in Antarctica in 1987 - 1999 (Bq m^{-3}, Bq Kg^{-1} of dry weight)
Таблица 3. Уровни радиоактивного загрязнения в антарктических образцах, собранных в 1987 - 1999 гг. (Бк $м^{-3}$, Бк $кг^{-1}$ сухого веса)

	Matrices	^{137}Cs	^{90}Sr	$^{239+240}Pu$	^{241}Am
Marine	Sup. Water	0.44-0.89	ND	0.0058-0.0067	0.0005-0.0025
Ecosystem	Sup. Sediment	0.14-1.50	<0.10-0.36	0.0056-0.0908	0.0024-0.0440
	Algae	0.40-37.2	ND	0.2010-0.6580	0.0400-0.3050
	Animals	0.18-1.28	0.08-0.11	<0.0022-0.0455	0.0006-0.0089
Lake	Sup. Water	0.19-1.01	<0.001	ND	ND
Ecosystem	Algae	7.1-41.8	2.5-19.3	ND	ND
	Sup. Sediment	0.11-3.93	0.18-0.76	0.0051-0.0969	0.0023-0.0412
Terrestrial	Mosses	11.3-49.9	9.20-24.40	0.3280-0.9080	0.1780-0.6140
Ecosystem	Lichens	47.5-162	ND	4.64	1.94
	Soil	<0.12-1.4	0.08-0.20	0.0010-0.0190	0.0010-0.0130

Concentrations for all nuclides are higher in unfiltered seawater samples than in filtered ones [7]. This fact can be explained with the high affinity of these elements for the particulate matter present in the water column. As a consequence, the radionuclides are absorbed on the surface of these particles and sink with them to the bottom.

Regarding ^{137}Cs, we registered a progressive decrease in seawater concentrations in the last 12 years (87 - 99), passing from a mean value of 0.9 Bq m^{-3} to 0.5 Bq m^{-3} (see Fig. 6a). The decrease is equal to ca 50 % and part of it (ca 22 %) is due to the natural decay of ^{137}Cs. The remainder can be explained with dilution and auto depuration phenomena due to the oceanic circulation. Same negative trend was observed for lake waters and lake algae (see figures 6b and 6c). The decrease is really high (up to 80 % for lake waters), probably totally due to natural decay. In fact these lakes can not develop auto depuration processes because they are closed environments. Tarn Flat algae seem to be more contaminated than those of Edmondson Point, and Carezza Lake algae are the less polluted of all.

As pointed out before, values are generally higher in the lacustrine sediments than in the marine ones or soils. This is an evidence of what assessed before defining a more contaminated terrestrial-lacustrine environment. Soils collected under mosses and lichens present a considerably high activity, for ^{137}Cs above all. Also for superficial marine samples we have noticed a progressive decrease of ^{137}Cs contamination levels, passing from 0.4 - 1.5 Bq Kg^{-1} of dry weight in 1988 to 0.3 - 0.4 Bq Kg^{-1} of dry weight in 1998.

As to marine organisms, it seems that auto depuration processes of the ecosystem are well working and only for *Pagothenia bernacchii* and *Euphausia superba* we could still detect small activities for ^{137}Cs. On the contrary, concentrations of ^{137}Cs in the macroalga *Iridea cordata* are very low.

Our results prove once again that even the Antarctic Continent suffers from radioactive pollution. Despite that, ^{137}Cs concentrations show a progressive decrease in time due both to natural decay and auto depuration phenomena. We can assess that in 100 years ^{90}Sr and ^{137}Cs will practically disappear from Antarctica, if no other explosions will take place in the meanwhile.

Tab. 4 shows the comparison between contamination levels measured in Antarctica with other data obtained by our research group in the

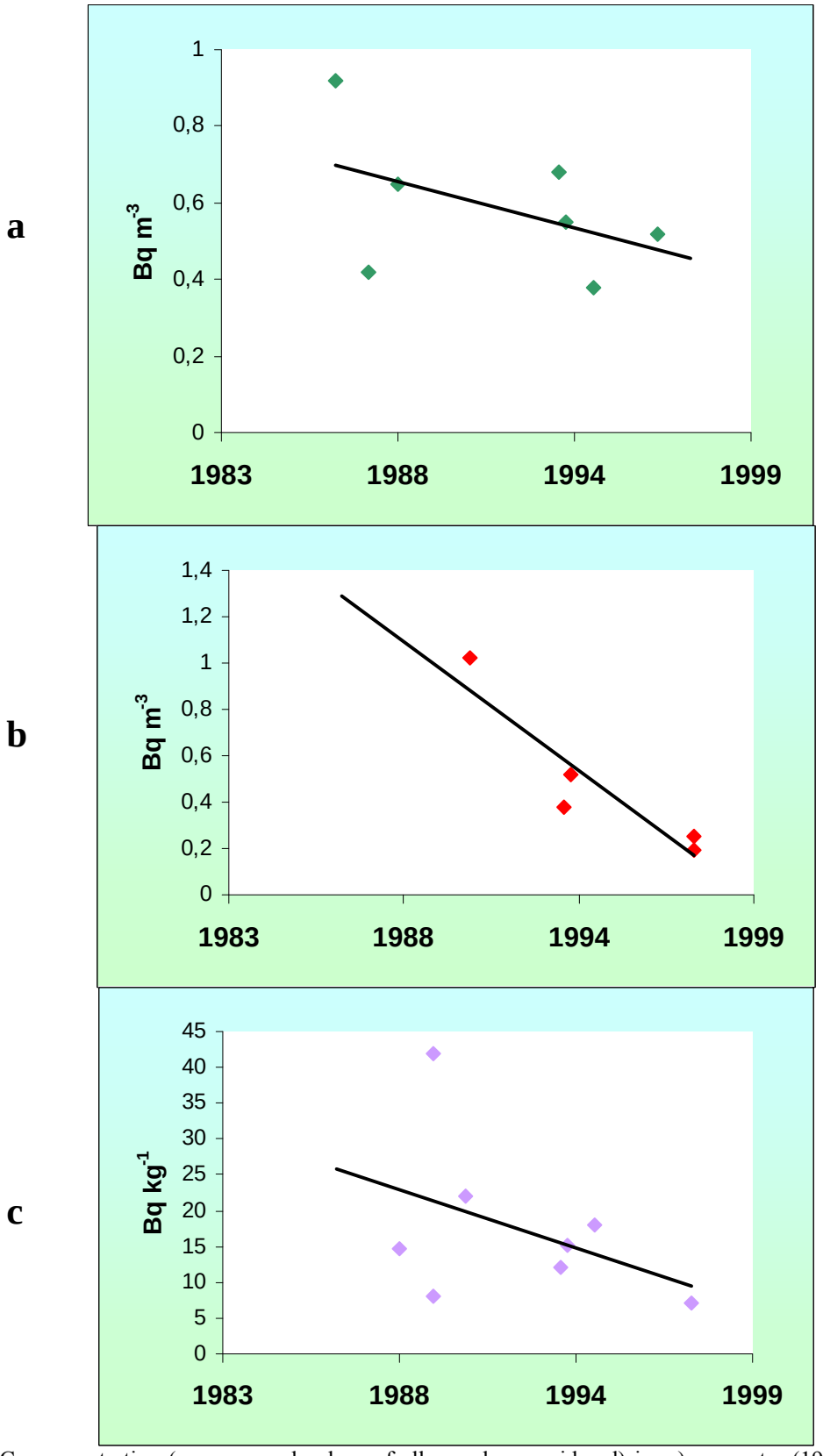


Fig. 6. ^{137}Cs concentration (mean annual values of all samples considered) in: a) sea water (1988 - 1998); b) lake water (1991 - 1998); c) lake algae (1991 - 1998).

Рис. 6. Динамика содержания ^{137}Cs (средние значения по всем измеренным пробам) в: a) морской (1988-1998 гг.) и b) озерной воде (1991-1998 гг.); c) в озерных водорослях (1991 - 1998 гг.).

Mediterranean Sea, whereas the mean surface values for ^{137}Cs in some marine areas of the world are resumed in Tab. 5.

Table 4. Anthropogenic radionuclides concentration levels (Bq m^{-3}, Bq Kg^{-1} of dry weight) in marine matrices from Antarctica and some areas of the Mediterranean Sea
Таблица 4. Уровни содержания антропогенных радионуклидов (Бк $м^{-3}$, Бк $кг^{-1}$ сухого веса) в морских образцах из Антарктиды и некоторых районов Средиземного моря

	^{137}Cs	^{90}Sr	$^{239+240}Pu$	^{238}Pu	^{241}Am
Antarctica			(B.T.N. – Ross Sea 1990 - 1996)		
Sup. Water	0.44-0.89		0.0058-0.0067	0.00013	0.0005-0.0025
Sup. Sediment	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)		
Sup. Water	3.36-5.18		0.013-0.025	0.0009-0.0074	
Sup. Sediment	1.50-18.5	<0.16-0.51	0.080-0.683	0.004-0.0022	0.036-0.188
Animals	1.00-2.57		<0.01-0.034	0.0020	
			(Po Delta 1990 - 1996)		
Sup. Water	2.80-4.30				
Sup. Sediment	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	
			(Aeolian Islands 1994 - 1995)		
Sup. Water	2.92-3.20		0.012-0.020		
Sup. Sediment	0.32-7.80	0.50-1.50	0.11-0.23	0.0066-0.0089	0.029-0.161

Basin	Mean ^{137}Cs - Bq m^{-3}
Baltic Sea	125
Irish Sea	55
Black Sea	52
North Sea/Barents Sea	10-12
Arctic Sea	7.6
Mediterranean Sea	5.4
North Pacific Ocean	4.0
North Atlantic Ocean	2.9
Indian Ocean	2.9
South Pacific Ocean	1.6
South Atlantic Ocean	1.4
Antarctica	0.4

Table 5. Mean ^{137}Cs concentrations in some oceanic superficial waters in 1990 [1, 4, 11, 12]
Таблица 5. Средние концентрации ^{137}Cs в поверхностных водах разных акваторий в 1990 г [1, 4, 11, 12]

It is evident that in the Mediterranean Sea values are 10-12 times higher than in Antarctica [2]. As an example, the Po Delta seems to be quite radioactive: we detected 31 Bq Kg^{-1} of dry weight in the superficial sediments, mainly because the river collect everything flowing through the Padana Plain and deposits it when debouching in the Adriatic Sea.

In the Baltic Sea and northern marine areas in general, contamination is 300-100 times higher and the Antarctic Sea is the less polluted at all (0.4 Bq m^{-3}). In the Black Sea we can notice the Chernobyl effect: values for ^{137}Cs are 52 Bq m^{-3} and they were much lower before the accident. The other three northern seas (Baltic Sea, Irish Sea and Barents Sea) also belong to the category of the contaminated sites, mainly because they receive the inputs of Russian rivers which collect all the radioactive wastes produced by the Russian nuclear power stations.

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