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# RADIOACTIVE AND STABLE ISOTOPES IN ABIOTIC AND BIOTIC COMPONENTS OF ANTARCTIC ECOSYSTEMS SURROUNDING THE ITALIAN BASE

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Results concerning the analysis of natural (K-40, Th-232, U-238) and anthropogenic (Sr-90, Cs-137, Pu-238, Pu-239, 240) radioactivity determined in samples collected during the PNRA (National Program for Antarctic Research) 1990–91 and 1991–92 Scientific Expeditions, are presented. The data refer to samples of the terrestrial, lacustrine and marine ecosystems surrounding the Italian Base in the Terra Nova Bay (Ross Sea) territory with special emphasis on the Cs-137 biogeochemical behaviour. In particular, the role of the organic substance in the radionuclide transfer has been evaluated through statistical correlation analysis between the Cs-137 concentrations and organic matter, organic carbon and nitrogen contents determined in samples of marine and lacustrine sediments.

**KEY WORDS:** Antarctica, Terra Nova bay, artificial radioisotopes, natural radioisotopes, radioecology.

## INTRODUCTION

Natural and artificial radioactivity measurements have been executed during the last five years on environmental samples collected in the terrestrial, lacustrine and marine ecosystems surrounding the Italian Terranova Base in Antarctica.

The results concerning the PNRA (National Program for Antarctic Research) 1987–88, 1988–89 and 1989–90 Scientific Expeditions have already been reported in previous papers<sup>1–4</sup>. In this work the unpublished results obtained during the next 1990–91 and 1991–92 Campaigns are presented.

The major part of the work has been oriented to the evaluation of the Cs-137 and the natural K-40, Th-232, U-238 presence in different environmental components of the three ecosystems. Moreover, data of H-3, Pb-210 and Po-210 was obtained in the past in some ice and compact snow cores<sup>5</sup>.

The biogeochemical behaviour of Cs-137 in the Antarctic environment is also discussed. In particular the role of the organic substance in the radionuclide transfer has been evaluated through statistical correlation analysis between the Cs-137 concentrations and organic matter, organic carbon and nitrogen contents determined in samples of marine and lacustrine sediments.

**Table 1** Sampling stations of the 1990–91 and 1991–92 campaigns not reported in previous publications.

<i>Code</i>	<i>Sample</i>	<i>Latitude (S)</i>	<i>Longitude (E)</i>
<i>Terrestrial ecosystem</i>			
SN 17	Snow	74°43'	164°01'
VO 14 Lich.	Lichens	74°04'	165°19'
VO 14 MOSS	Moss	74°04'	165°19'
SL 8	Soil	74°20'	165°07'
SL 14	Soil	74°04'	165°19'
SL 20	Soil	74°43'	164°01'
<i>Lacustrine ecosystem</i>			
LW 17/3	Lake water	74°43'	164°01'
AG 8	Algae	74°20'	165°07'
AG 12	Algae	74°59'	162°33'
AG 17	Algae	74°43'	164°01'
LS 8	Sediment	74°20'	165°07'
LS 12	Sediment	74°59'	162°33'
LS 17	Sediment	74°43'	164°01'
LS 20A	Sediment core	74°43'	164°01'
LS 20B	Sediment core	74°43'	164°01'
<i>Marine ecosystem</i>			
PW 5	Pack water	74°41'20"	164°06'56"
PW 6	Pack water	74°38'04"	164°17'17"
PW 7	Pack water	74°40'05"	164°13'38"

Some preliminary results on Sr-90 and Pu-239 + 240 concentrations obtained by radiochemical separations carried out on terrestrial and lacustrine samples collected during the 1990–91 Expedition, are also presented. Such determinations were executed as part of a collaboration activity with Prof. C. Testa and Dr. C. Roselli of University of Urbino.

Furthermore, part of this research was oriented towards the determination of the earth-alkaline elements calcium, magnesium, barium and strontium, besides the contents of potassium and uranium in seawater samples collected in coastal stations.

All sampling stations refer to the classic Italian study area at Terra Nova Bay and its inlet as described in the 1990–91 and 1991–92 Expedition final reports<sup>7</sup> and in previous papers for the marine environment<sup>8</sup>. The list of sampling stations is provided in Table 1.

## MATERIALS AND METHODS

Samples of superficial unfiltered sea and lacustrine water collected during the IVth Expedition (1990–91) were submitted to the NCFE extraction method. This technique

allows higher Cs-137 radiochemical separation yields (almost 100%) compared to the ones obtained with the classical AMP method<sup>1,2</sup>. In this technique, the liquid sample (300–400 liters for sea water and 95 liters for lake water) is pumped through NCFC-bed columns (ammonium hexacyanocobalt (II) ferrate (II) supported on silica gel), equipped with a flux counter (constant flux 100 ml/min), whose property is capturing the cesium isotopes<sup>6</sup>.

Lacustrine and marine sediments, soil, lichens and moss, lacustrine algae, ichthyofauna and macrofauna organisms were collected during the different scientific expeditions. The different matrices were oven-dried at 105°C, minced and then introduced into standard source containers to be counted for gamma spectrometry. For what concerns the marine organisms, biometric analysis, sex recognition and anatomical dissection were executed before the drying procedure.

The analytical source obtained were counted for gamma spectrometry by means of two PGT Silena high resolution germanium detectors (FWHM 1.8 KeV at 1.33 MeV) for quantitative evaluation of the gamma emitters. All instruments had been previously calibrated through an intercalibration exercise with the IAEA- Marine Environment Laboratory (Monaco).

The gamma energy considered ranged between 0.08 and 1.5 MeV with direct detection of Cs-137 (0.662 MeV) and K-40 (1.461 MeV) besides determination of Th-232 and U-238 through descendants of the natural families: respectively Bi-212 (0.727 MeV) and Ti-208 (0.583 MeV) for thorium, Pb-214 (0.352 MeV) and Bi-214 (0.609–1.120 MeV) for uranium.

The data obtained were elaborated with an IBM personal computer equipped with the EG&G-Ortec programme "Quantitative analysis software programme for gamma spectrometry". Because of the low levels of Cs-137 contamination, measurements time was between 50 and 150 hours. Errors reported referred to standard deviation (1 sigma) and was due only to the counting statistics. All data were decay corrected to sampling time and are herein reported as Bq/m<sup>3</sup> for water samples and Bq/kg dry for solid samples.

The determination of the plutonium isotopes was carried out through a particular radiochemical separation<sup>9</sup> after a sample pre-treatment based on the matrix composition. Sediments were certainly the most analyzed samples and the procedure can be summarized in the following steps.

Ashed sediments were added of Pu-242 (4 dpm) as internal tracer and submitted to a HNO<sub>3</sub> attack. Different passages on microtene TOPO (oxyde of tri-n-octylphosphine) were executed and then a classical electrodeposition on steel plates in acid solution was carried out.

Electrodeposited plates were hence counted for alpha-spectrometry by means of two EG&G-Ortec silicon chambers and one ionizing chamber for the quali-quantitative identification of Pu-238 (5.5 MeV), Pu-239 + 240 (5.1 MeV) and Pu-242 (4.9 MeV).

The data obtained were elaborated with an IBM personal computer equipped with the EG&G-Ortec programme "Quantitative analysis software programme for alpha spectrometry" following the same operative procedures reported above for gamma counting.

Strontium was determined from the liquid residue of one passage of the plutonium extraction. In this technique yttrium (Sr-90 descendant) was extracted with HDEHP (di-(2-ethylhexyl)-phosphate) and precipitated with oxalic acid. Y-90 was therefore measured by means of an ASPN beta counter.

The organic matter contained in the sediment samples was determined as difference between ash (550°C) and dry weight; nitrogen with the Kjeldahl method; phosphorus through spectrophotometry and organic carbon with the COD method.

## RESULTS AND DISCUSSION

The data detected allowed a complete and satisfactory mapping of the distribution of natural and artificial radioactivity in different ecosystem components of the Terra Nova Bay inlet and the forelying sea. In particular, higher concentrations of artificial radioactivity were measured in lacustrine algae samples (Tarn Flat, AG 12) and in mosses and lichens collected at Kay Island (VO 14). These samples were collected during the 1990–91 Campaign and the results are reported in Table 2.

Both aquatic and terrestrial vegetable organisms demonstrated to be efficient bioindicators of environmental radiocontamination of the 1990–91 study area. Stream waters derived from ice melting, besides their collection and stagnation in the small lake-pond system seemed to play a fundamental role in the transport and accumulation of Cs-137 and Pu-239,240 in lake sediments and algae. Their concentrations were generally higher than the ones detected in soil samples collected in areas surrounding the same lakes during the previous Campaigns<sup>1-3,10</sup>.

Cs-137 and natural radionuclides concentrations measured in two soil cores (SL 8, Edmonson Point; SL 20, Skua Lake) and two lake sediment cores (LS 20 A e B, respectively center and side of Skua Lake) collected during the 1991–92 Expedition are illustrated in Table 3. The Cs-137 contribution to the core contamination was higher in the top layer (0–5 cm) whereas decreasing values were observed in the underlying strata (5–10 and 10–15 cm). In spite of this, level zero of radiocesium contamination was not detected due to the core shortness. For this reason inventory data were not calculated.

As far as natural radioactivity is concerned K-40 was the most abundant primordial radionuclide in all samples analyzed and concentrations of Th-232 and U-238 were in the ranges of previous data<sup>1-3</sup> in relation to the natural composition of soil and sediments.

**Table 2** Concentrations of artificial and natural radioisotopes in environmental matrixes of the 1990–91 Campaign. Values in Bq/kg dry (data quoted with (\*)) are in Bq/mc).

Sample	Cs-137	Sr-90	Pu-238	Pu-239,240	K-40	Th-232	U-238
<i>Lacustrine ecosystem</i>							
LW 17/3 (*)	1.01 ± 0.65	< 0.001	< 0.001	< 0.001	ND	ND	ND
AG 8	5.97 ± 0.36	ND	ND	ND	406.6 ± 9.71	ND	ND
AG 12	46.8 ± 0.54	ND	ND	ND	568.5 ± 10.8	ND	ND
AG 17	10.4 ± 0.41	19.3 ± 1.5	0.03 ± 0.01	0.12 ± 0.01	204.3 ± 10.6	ND	ND
LS 8	3.21 ± 0.16	5.90 ± 0.4	< 0.02	0.08 ± 0.01	1130 ± 10.02	46.3 ± 1.5	53.3 ± 0.9
LS 12	0.14 ± 0.09	12.9 ± 1.1	< 0.02	0.02 ± 0.01	912.2 ± 7.91	58.1 ± 0.9	79.5 ± 1.2
LS 17	0.27 ± 0.19	0.3 ± 0.2	< 0.04	< 0.03	1014 ± 12.2	25.3 ± 1.2	56.9 ± 0.6
<i>Terrestrial ecosystem</i>							
SN 17 (*)	2.50 ± 1.12	0.01 ± 0.001	< 0.0001	< 0.0001	ND	ND	ND
VO 14 Lich.	47.5 ± 2.15	12.2 ± 1.0	0.05 ± 0.01	0.22 ± 0.03	239.8 ± 47.5	ND	ND
VO 14 Mos.	39.0 ± 0.64	13.3 ± 0.9	0.08 ± 0.01	0.30 ± 0.04	383.6 ± 13.3	ND	ND
SL 14 Mos.	1.49 ± 0.17	2.90 ± 0.2	< 0.04	< 0.03	771.8 ± 07.9	35.1 ± 1.3	42.3 ± 1.5

**Table 3** Concentrations of Cs-137, K-40, Th-232 and U-238 in soil and lake sediment cores collected during the 1991–92 campaign. Values in Bq/kg dry.

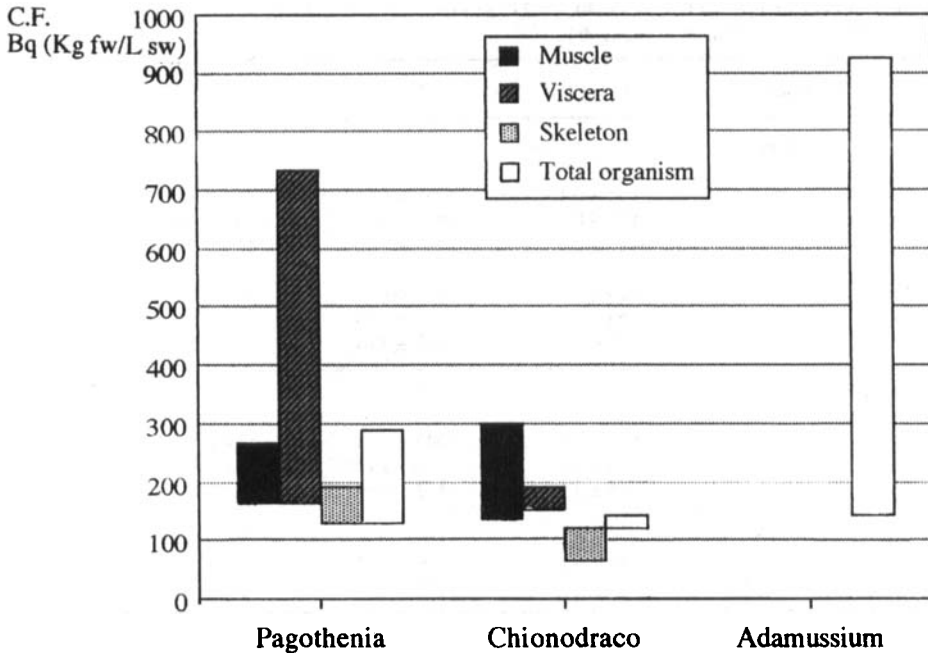
Sample	Stratum (cm)	Cs-137	K-40	Th-232	U-238
<i>Terrestrial ecosystem</i>					
SL 8	0–5	0.71 ± 0.1	1334 ± 10.1	49.5 ± 1.0	64.2 ± 1.6
SL 8	5–10	0.22 ± 0.1	1066 ± 8.50	56.4 ± 0.9	46.1 ± 0.9
SL 8	10–15	0.15 ± 0.1	1153 ± 8.70	43.2 ± 0.8	53.5 ± 1.1
SL 20	0–5	0.79 ± 0.1	942 ± 06.9	38.3 ± 0.7	23.5 ± 0.4
SL 20	5–10	0.63 ± 0.1	1087 ± 7.66	41.2 ± 0.7	30.6 ± 0.5
SL 20	10–15	0.27 ± 0.1	945.9 ± 8.01	30.7 ± 0.6	23.7 ± 0.6
<i>Lacustrine ecosystem</i>					
LS 20A	0–5	0.23 ± 0.1	1042 ± 7.30	32.6 ± 0.6	24.5 ± 0.4
LS 20A	5–10	0.18 ± 0.1	985.6 ± 13.6	25.6 ± 0.7	20.5 ± 0.3
LS 20A	10–15	0.15 ± 0.1	1132 ± 10.9	25.9 ± 0.7	25.0 ± 0.3
LS 20B	0–5	0.26 ± 0.1	1189 ± 15.3	21.2 ± 0.7	25.7 ± 1.1
LS 20B	5–10	0.14 ± 0.1	1089 ± 16.2	23.9 ± 1.3	24.3 ± 0.9
LS 20B	10–15	0.12 ± 0.1	1150 ± 15.9	25.1 ± 0.8	23.5 ± 0.3

Cs-137 concentrations detected in marine samples of the 1990–91 Campaign were generally lower than the ones observed in the lacustrine and terrestrial ecosystems. All analyses performed in the marine ecosystem were detailly described in previous papers<sup>8,11</sup>. In particular, ranges of concentrations of this radionuclide were 0.20–0.42 Bq/m<sup>3</sup> for sea water, 0.11–0.20 Bq/kg dry for coastal and off-shore sediments, 0.16–0.77 Bq/kg dry for the organisms different anatomical components.

Concentration factors (C.F.) for Cs-137 measured in marine organisms collected during the 1988–89, 1989–90 and 1990–91 Campaigns were generally high in the bivalve *Adamussium colbecki* in relation to the filter—feeding habit of this species. On the contrary, the ones determined in *Pagothenia bernacchii* and *Chionodracco hamatus* were lower and reflected different feeding habits than *Adamussium colbecki*. Ranges of the C.F. determined in total organisms and single components are reported in Figure 1. Nevertheless, the concentration factors determined in *Adamussium* seemed quite well related to the seasonality of the ecosystem with higher values during the austral summer periods of intense primary production.

This might suggest a major role of the particle form of Cs-137 in transporting this isotope to the Antarctic benthic compartment, although it is well known that radiocesium in temperate waters is mostly in the soluble form.

The trends of Cs-137 and organic matter in marine coastal and off shore sediments seemed to support this idea. In fact, higher Cs-137 concentrations were detected in samples containing higher values of organic matter (5.5–8.0% dry weight; average value other samples 3.86%), organic carbon (3.61–7.88 mg/g; average value other samples 1.86 mg/g) and nitrogen (3.01–4.60 mg/g; average value other samples 1.11 mg/g). Concentrations of Cs-137 were quite well correlated with the contents of the above elements. In fact, their correlation coefficient ranged between 0.65 and 0.95.



**Figure 1** Ranges of concentration factors determined in single components and total organisms of Antarctic marine fauna.

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