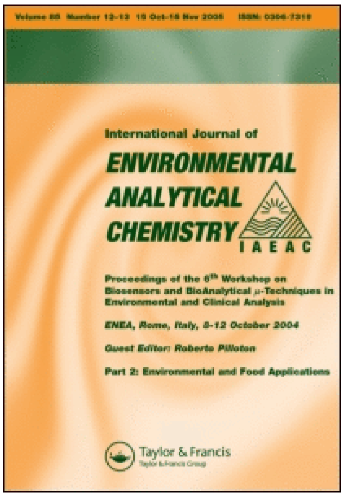

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DISTRIBUTION OF ^{210}Pb AND ^{137}Cs IN SNOW AND SOIL SAMPLES FROM ANTARCTICA

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This paper reports some results on natural and artificial radionuclide distribution in snow samples collected at the Priestly Nevé —Plateau (inland, 1998 m a.s.l.) and the Vegetation Island (near the coast, 200 m a.s.l.) stations, and in a soil sample from Wood Bay station, both near the Italian Base at Terranova Bay in Antarctica. Data on Pb-210 and Cs-137 in snow samples from an 80 cm deep trench and in granulometric fractions of a soil core are discussed.

KEY WORDS: Antarctica, Radionuclides, Snow, Soil, ^{210}Pb, ^{137}Cs.

INTRODUCTION

The research program of our Operative Unit within the National Research Program in Antarctica (Environmental Impact—Chemical Methodologies—1988–90 Campaigns) regards the definition of the nature, distribution and mobility of radionuclides in the Antarctic environment. In fact, several natural and/or man-made radionuclides also find their final end in diverse matrices of this environment where chemical, chemico-physical, biological or mechanical processes may mobilize, transfer and concentrate them among the various phases. The first step of these studies is the establishment of the methodologies for sampling procedures, and the treatment, chemical and radiochemical analyses, including inter-calibrations procedures, of the same.

EXPERIMENTAL

Procedures

Snow samples were collected at Priestly Nevé—Plateau, 01 SN II, (inland, 1998 m a.s.l., 73°38′18″ S lat. and 160°38′32″ E long., drawing temperature −18°C), and at Vegetation

Island, 10 SN II, (near the coast, 200 m a.s.l., 74°47′lat. S and 163°38′long. E), by using the trench method, down to a depth of 80 cm. Four 35x35x20h cm^3 blocks were gathered in each station. The samples were kept at −30°C in 25 l polyethylene containers, previously tested for absence of radionuclide contamination, until their analysis in our laboratory. Starting from the surface: 5.6, 6.1, 6.5, 5.6, and 5.2, 5.0, 7.1, 7.3, litres of equivalent water were determined for each section of the two sampling stations, Pristley Nevé and Vegetation Island stations, respectively. Stable Cs and Pb carriers, were added, while acidifying the samples in the defrosting phase, both to minimize interactions with the container walls and to determine the chemical yields of the overall procedure. The defrosted samples were filtered on 0.45 μm cellulose filters and the concentration step was carried out through distillation on glassware apparatus with an efficient anti-spray device. The global yield for the recovery was determined by analysing the losses due to the entrainment of carriers during distillation. The yields of the concentration steps were determined with a Total Reflection X-Ray Fluorescence Spectrometer (EXTRA II) and resulted >99% both for metals and for all fractions. The final solutions, concentrated at 40 ml and introduced into standard containers, were counted in well defined geometry by high resolution gamma spectrometry. A blank for gamma counting was also prepared using bidistilled water and repeating all treatments of the analyzed samples.

A 10x10x15h cm^3 soil sample was collected at Wood Bay station (30 m a.s.l., 3 km Nord of Edmonson Point, 74°18′ lat.S, 165°05′ long.E). This sample was sectioned into three parts: 0–5, 5–10, 10–15 cm; granulometric distribution was determined after excluding any material larger than 2 mm from each specimen. The following ranges were analysed: 2000–200 μm, 200–100 μm, 100–50 μm and 50–0 μm with the aim of evaluating the distribution of radionuclides among the various fractions. In addition, chemical analyses of these fractions, carried out both in the solid matrix and in solution after a mild leaching, were performed by X-ray fluorescence spectrometry.

Apparatus and measurements

All samples were first oven-dried to constant weight (110°C) and then introduced into suitable containers of different volumes but of definite disc geometry for high resolution gamma spectrometry, using an Intrinsic Germanium Detector with an energy resolution of 1.8 keV at 1330 keV, for the high energy side of the spectrum and an Intrinsic Germanium Planar Detectors, [energy resolution: 0.6 keV at 122 keV] for the lower energy side. The obtained activity data was processed with the Silgamma (Silena-Milano) analyzer system. Energy and efficiency calibrations were performed using a Mixed Radionuclide Gamma-Ray Reference standard solution (QCY 44 and Eu-152) from Amersham (U.K.). Due to the very low activity levels of samples, the measurement time needed to reach good counting statistics was about 80 hours. Activity data was properly corrected for matrix self-absorption at the different energies, taking into account the different geometries of the small samples obtained from the diverse granulometric fractions. All data was decay-corrected to the collection time, and data errors reported in the tables refer only to counting statistics.

The X-Ray Fluorescence Spectrometer consists of an EXTRA II module (R. Seifert, D-2070 Ahrensbourg) incorporating two total reflection beam lines. Two X-ray tubes with different anode materials (molybdenum and tungsten) are used to maximize sensitivity for all detectable elements. The spectrometer is equipped with a Si(Li) detector, pulse processor, automatic sample changer and main computer controlling a multichannel analyzer (Link System AN 10000).

RESULTS AND DISCUSSION

The aim of our research demands the knowledge of natural and anthropogenic fluxes of aerosol particles deposited onto the Antarctic ground. Because only the last century is the period which interests us, the use of Pb-210 and Cs-137 as radiocronological tracers appears to be suitable. Due to the very low levels of radionuclide activity in the various matrices large amounts of sample must be used. Unfortunately, the sampling methods were initially not suitable for providing all the required samples and in sufficient quantities. In particular, the snow samples collected were only 80 cm deep and for this reason present data should be considered cautiously until deeper and larger sample amounts become available.

Snow

Table 1 shows the unsupported Pb-210 activities for both the Priestley Nevé—Plateau and the Vegetation Island stations.

Absolute activity data and specific data in function of the water equivalent contained in each section are reported. The water equivalent content of the snow sections of the inland station was rather constant with depth, while at Vegetation Island station, near the coastline, the snow presented significant compactness effects, confirmed by increasing weight of water equivalent in depth, probably a consequence of a seasonal re-melting of the snow. According to the Goldberg method[1], utilized by several authors[2-6], on the average admitting a "constant rate of supply of Pb-210" from the atmosphere, it is possible to date the deposit through the Pb-210 n.s. content of the snow, which, in this matrix, is solely of atmospheric origin as the

Table 1 Pb-210 n.s. Activity Data on Snow Samples Collected at 01 SN II (Plateau—Priestly Nevé) and 10 SN II (Vegetation Island) Stations, Referred to the Sampling Date. The Activity Reported as Bq abs. Refers to 35×35×20h cm^3 Samples.

	01 SN II *Lat. 73° 38′ S, Long. 160° 38′ E*			*10 SN II* *Lat. 74° 47′ S, Long. 163° 38′ E*		
Fraction cm	*Snow Water Equivalent* dm^3	*Pb-210 Bq abs. (error %)*	*Pb-210* Bq/m^3	*Snow Water Equivalent* dm^3	*Pb-210 Bq abs. (error %)*	*Pb-210* Bq/m^3
0–20	5.6	0.142 (9)	25.4	5.2	0.387 (10)	74.4
20–40	6.1	0.105 (9)	17.2	5.0	0.256 (15)	51.2
40–60	6.5	0.080 (8)	12.3	7.1	0.295 (15)	41.5
60–80	5.6	0.052 (9)	9.3	7.3	0.273 (15)	37.4
TOTAL	23.8	0.379	64.2	24.6	1.211	204.5

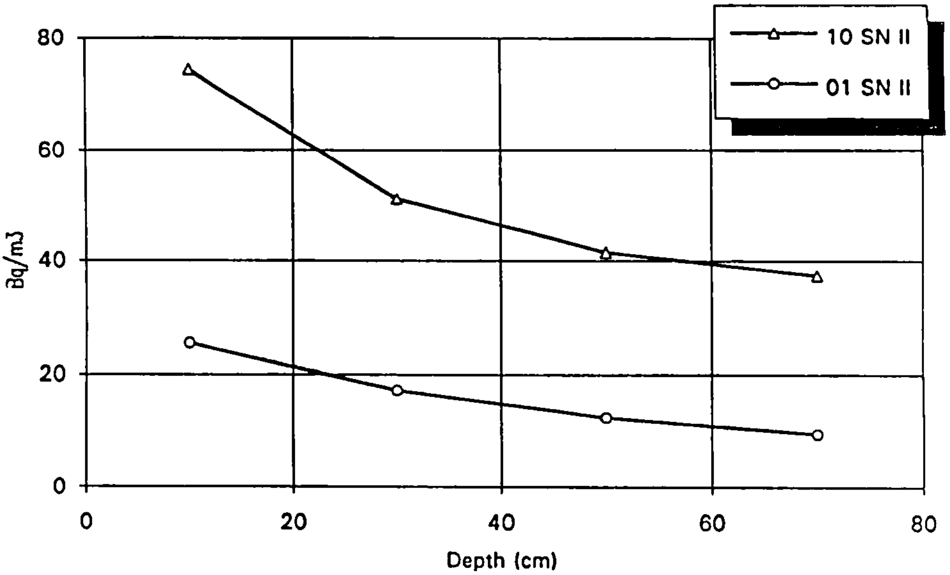

Figure 1 Pb-210 n.s. activity (Bq/m^3) *vs.* depth of snow water samples from 01 SN II (Priestly Nevé—Plateau) and 10 SN II (Vegetation Island) stations.

contribution from the sea and from volcanic dust was found to be negligible[7-10]. The specific activity of Pb-210 n.s. in the snow should decrease exponentially with depth according to the law of radioactive decay. The use of specific activity also avoids the misleading variations of snow density with depth, which may depend upon the diverse, climatic and/or geographic conditions varying from site to site.

The Pb-210 n.s. specific activities and logarithmic profiles, versus depth for both stations are reported in Figures 1 and 2 respectively. The observed activities regularly decrease with depth, without considerable fluctuations as expected from c.r.s. hypotheses. The logarithmic plots appear to be linear with regression coefficients of 0.99 for the Priestly Nevè station, and 0.97 for the Vegetation Island, at 95% confidence level.

On this basis, a possible equivalence cm/years could be calculated, once confirmed by the results from deeper sections. The average snow accumulation rates, considering the slope of Pb-210 n.s. constant, would be 4–5 kg/m^{2}y for Priestly Nevé, and 6–7 kg/m^{2}y for Vegetation Island. Literature data report values rather unhomogeneous for snow deposition rates in Antarctica, thus indicating a high variability of precipitation events in different sites 11–15.

No data on the Cs-137 content, for each specific section, were obtained, because of the low activity levels of this radionuclide. However, putting together all the concentrated solutions, total Cs-137 activity was found to be 0.206 and 0.322 Bq/m^2 for the first 80 cm snow layer, at the Priestly Nevé and Vegetation Island stations, respectively. The Pb-210

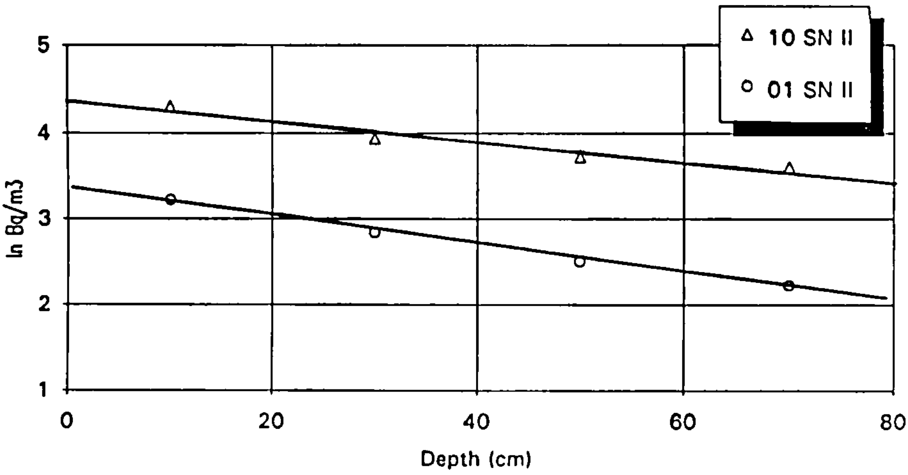

Figure 2 Pb-210 n.s. activity (ln Bq/m^3) *vs.* depth of snow water samples from 01 SN II (Priestly Nevé—Plateau) and 10 SN II (Vegetation Island) stations.

n.s./Cs-137 activity ratio, for this surface layer, shows that this value is lower (1.8) at the Priestly Nevé station than that found at Vegetation Island (3.8), because the activity data for Pb-210 n.s. is lower at the first station. In fact, the flux of Cs-137, coming from the stratosphere, can reasonably be considered almost constant, while the concentration of Pb-210 n.s. can be considered practically constant in the troposphere, this due to the homogeneous diffusion of Radon-222, which can present different concentrations in the snow, in relation to the height of the air column scavenged by the falling snow crossing this column. So, the Pb/Cs activity ratio should decrease with altitude, because of the different radionuclide proportions scavenged by the snow. A similar trend was also found in Canada by Monaghan[16].

The above calculated values of snow accumulation rates result much lower than those found in other Antarctic areas. However, it should be noted that our data, while limited to the 80 cm surface layers, show a regular decrease of Pb-210 activities with depth, which is, on the top snow layer of the same order of magnitude as those reported by several other authors (e.g. Crozaz[5], Sanak & Lambert[11], Koide et al.[12]).

The low accumulation rate values, resulting from the Pb-210 n.s. activities of the top 80 cm, may also be due to particular orographic and geographic conditions of the sampling stations, where strong winds and/or melting processes at low altitude near the sea may partially remove the falling snow. The constancy in depth of Pb-210 n.s. activity slopes is to be confirmed in future campaigns. In addition, the Pb/Cs ratio observed for the snow sampling stations, seems to suggest a higher mobility of Cs-137 in the snow with respect to Pb-210 n.s., probably due to leaching or to its greater solubility; a very important phenomenon in the summer percolation, only for the stations at low altitude and near the coast.

Table 2 Granulometric distribution (%) referred to both total amount and <2mm fraction, in three soil sections of Wood Bay sample.

GRANULOMETRIC FRACTIONS	*PERCENTAGE* *0–5 cm*		*5–10 cm*		*10–15 cm*	
	TOTAL	*<2mm*	*TOTAL*	*<2mm*	*TOTAL*	*<2mm*
> 2 mm	38.5	-	37.4	-	46.3	-
2000–200 µm	58.5	95.0	60.2	96.4	52.3	97.4
200–100 µm	2.0	3.4	1.5	2.3	0.9	1.7
100–50 µm	0.4	0.7	0.4	0.6	0.2	0.4
< 50 µm	0.6	0.9	0.5	0.7	0.3	0.5

Soil

In Table 2 the granulometric distribution (%) in the three sections of soil collected at the Wood Bay station, near the Melbourne volcano, are reported.

No relevant variations of granulometric composition with depth were observed in this ground sample, mainly formed by a rather coarse matrix, probably originating from desegregation of volcanic rocks. In fact, the composition of this material is quite constant and presents a typical high iron content. The concentration ratios of various elements with respect to iron, as principal component of the sample, are the following: Fe/Ca = 3, Fe/K = 3, Fe/Ti = 9, Fe/Mn = 45, Fe/Zn = 333.

The granulometric fractions were analyzed for radionuclide content and their specific activities are reported in Table 3.

As expected, the smaller fractions, with higher specific area, are much more enriched in both Cs-137 and Pb-210 n.s. (of atmospheric origin), as reported, for example, in Figure 3, for the 0–5 cm section. But the radionuclides activity observed for the 200–2000 µm fractions, in spite of the low activity, accounts for the main fraction of radionuclides.

Table 3 Activity data in the granulometric fractions of soil sections of Wood Bay sample.

Section	*Granulometric fractions*	*Activity (Bq/kg)* *Bi-214*	*Pb-210tot*	*Pb-210 n.s.*	*Cs-137*	*Total Activity (Bq)* *Pb-210 n.s.*	*Cs-137*
0–5 cm	0–50 µm	23.6	117.9	94.3	11.1	0.61	0.07
	50–100 µm	10.7	92.9	82.2	5.9	0.42	0.03
	100–200 µm	30.0	62.9	32.9	4.2	0.91	0.12
	200–2000 µm	22.8	36.5	13.7	1.6	11.08	1.26
5–10 cm	0–50 µm	28.4	115.6	87.2	8.5	0.46	0.04
	50–100 µm	17.9	106.9	89.0	5.9	0.37	0.02
	100–200 µm	27.8	74.4	46.6	2.0	0.76	0.03
	200–2000 µm	22.8	39.8	17.0	1.1	11.60	0.72
10–15 cm	0–50 µm	22.7	95.5	72.8	7.4	0.52	0.05
	50–100 µm	14.6	88.5	73.9	6.3	0.37	0.03
	100–200 µm	27.3	52.0	24.7	1.4	0.59	0.03
	200–2000 µm	23.7	32.4	8.7	0.5	11.43	0.66

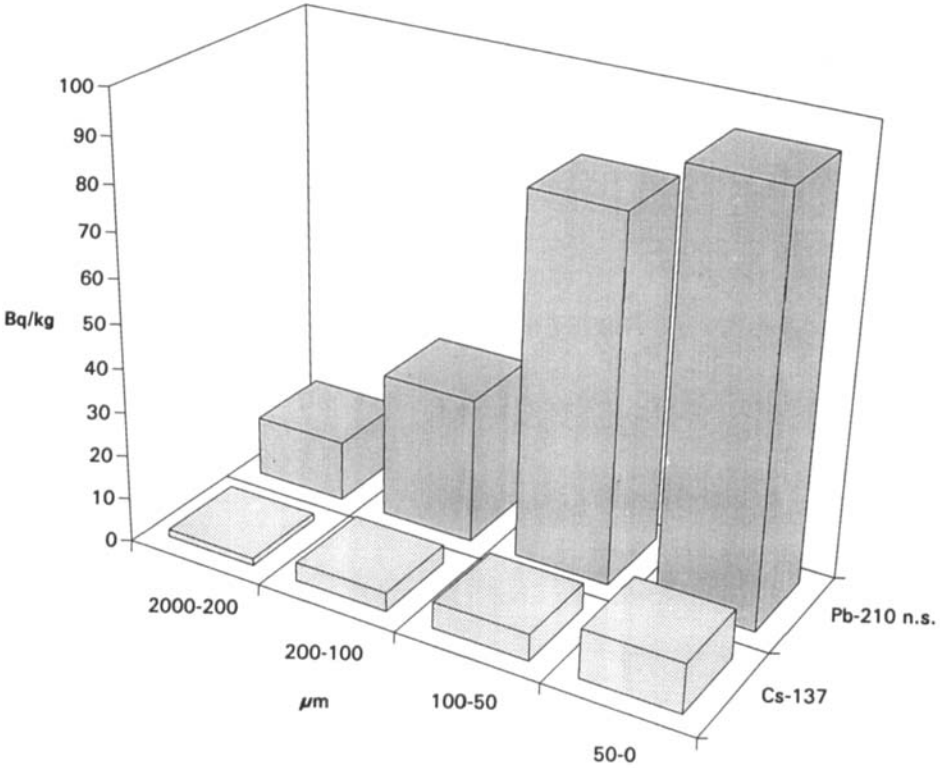

Figure 3 Pb-210 n.s. and Cs-137 activity (Bq/kg d.w.) in the granulometric fractions of the 0–5 cm section of Wood Bay soil core.

A very complete "air washing" of the coarser granules, thus lowering the radioactivity, mainly for Cs-137, but doesn't change the importance of its contribution to the total activity of the coarser fraction in the overall balance. This is only an apparent discrepancy because the Cs-137 distribution, among the various fractions, simply reflects a homogeneous surface distribution. In fact, the very large amount of the coarser fraction compensates its low specific surface area.

The Pb-210 n.s./Cs-137 ratio in this soil does not show any definite trend regarding granule dimension. It is to be noted that the finer fractions represent only a small percentage of total soil mass, and the total amount of Pb-210 n.s. and of Cs-137 doesn't change very much with depth. So, the high porosity of these samples allows the permeation of the radionuclides coming from the surface layers, down to a relevant depth. This fact must be taken into proper account for dating purposes, and also for evaluation of anthropogenic pollution phenomena. In fact, recently deposited elements may not always remain trapped in the same place, and the case study is quite relevant in this respect. A somewhat similar situation may be found in snow deposit of low accumulation rate where only a fraction of the total precipitations may remain.

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