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^{7}Be Concentrations in Air, Rain Water and Soil in Cantabria (Spain)

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^{7}Be concentrations present in air, rain water and soil have been measured in the region of Cantabria (Spain) during the last 4 yr. There was a relatioship between rainfall and the deposited areal activity of the nuclide at the study site which was consistent with observed annual global rainfall and fallout. © 1997 Elsevier Science Ltd. All rights reserved

Introduction

The massive release of radiation into the atmosphere caused by the Chernobyl nuclear power station accident on the 26 April 1986 demonstrated the need for different countries, through official institutions, to establish and maintain an environmental radiological surveillance network.

In this context, the Department of Medical Physics of the University of Cantabria signed an agreement of collaboration in 1991 with the Board of Nuclear Security with the aim of starting an environmental radiological surveillance plan in the autonomus region of Cantabria. This is integrated in the network of Environmental Radiological Surveillance stations (REVIRA), which can be found throughout Spain. The methods used in developing the above mentioned Surveillance Program have made it possible to measure the ^{7}Be concentration in air, rain water and soil for the last 4 yr. The results obtained have been compared to values found in other latitudes and regions of the planet.

^{7}Be is a radionuclide that has been studied in a number of areas in different latitudes in both the northern and southern hemispheres. This isotope is produced by cosmic ray induced spallation on O and N in the atmosphere resulting in the formation of BeO or $Be(OH)_2$ molecules. These molecules spread through the atmosphere where they come in contact with atmospheric aerosoles which makes them useful in the study of the behaviour of those same aerosols. Different atmospheric processes lead to their appearing in the troposphere and their later precipitation back to the earth's surface (Olsen *et al.*, 1985).

Cosmic radionuclide concentration can vary greatly according to latitude and altitude. Approximately 70% of the ^{7}Be is produced in the stratosphere and the other 30% in the troposphere. The quantity produced in the stratosphere usually stays there for about a year before entering the troposphere where it remains for about 6 weeks. Its transfer to the earth's surface is achieved by gravitational settling and precipitation. Its half life (53 days) is long enough for it to be used as an atmospheric tracer in the study of air movements, plus dry and humid deposition on the earth's surface. The study of the possible seasonal changes in ^{7}Be concentrations due to an increase of its migration from the stratosphere into the troposphere (which is related to the rainfall and other meteorological factors), gives useful information about its distribution and behaviour. At the same time, the measurement of ^{7}Be deposition on the ground is useful for studies on erosion in which this element is used as a tracer (Wallbrink and Murray, 1993).

Material and Methods

Environmental samples of air and rain water were taken from the roof of the medical school. For the air samples a suction pump was used. Filters can be attached to capture aerosols (dust particles) and active carbon traps for atmospheric radioiodine. Pump flow is about 30–35 L/min.

Rain water samples are collected on a monthly basis with the help of a stainless steel pluviometer which has an area of 1 m^2.

The filters used for capturing the atmospheric aerosols were glass fibre filters, with a retention efficiency of close to 100% for particles larger than 0.8 μm. The filters were changed once a week after having filtered air volumes of around 350 m^3.

Table 1. Concentrations of ^{7}Be in aerosols, rain water and soil

	1992	1993	1994	1995
Aerosols (mBq m^{-3}) (weekly)	—	(Lid* 6)	(1–7.4)	(1–9)
Aerosols (mBq m^{-3}) (quarterly)	3	(2.2–3.4)	(4–4.5)	(3–4.4)
Rainwater (kBq m^{-3})	(3.4–4.1)	(Lid† 5.0)	(2.3–5.4)	(1.5–1.7)
Soils (Bq m^{-2})	230	470	275	200

*Lid = 0.1 mBq m^{-3}.
†Lid = 0.73 kBq m^{-3}.

Measurement of ^{7}Be in the air filters was done on a weekly basis and every 3 months a sample collection taken from all the filters in the same period was gathered and placed in a 5 cm dia Petri dish.

Rain water samples were collected on a weekly basis in 1992–1993 and on a monthly basis during 1994. When the volume of water collected was less than 2 L it was considered to be dry deposition. Distilled water was poured over the surface of the pluviometer in order to collect the accumulated dry deposition. Once a total volume of 10 L was reached a quota of 1 L was taken. When the volume of rainfall was higher than 2 L, a 1 L sample of all rain water is collected. The rain water or dry deposition is acidified after being taken to prevent absorption from the walls of the recipient. 0.5 L of the water taken during each month is used to make a sample corresponding to a 3 month period. The sample will later be evaporated on a flexible plastic dish and introduced into a Petri capsule 9 mm dia.

Soil samples are collected once a year in clear areas with a total surface area of about 10 m^{2} next to the Medical Faculty building. The samples obtained, after being dried, sieved and weighed, are introduced into a sample holder (Marinelli geometry).

The measurement of ^{7}Be activity in the different samples was done by using a high-purity germanium coaxial detector surronded with shielding material to reduce the background counting rate. The system is previously calibrated in energy and efficiency for each of the geometries used. The detection efficiency of the gamma spectrometer was calculated by preparing a model sample that contained ^{137}Cs and ^{226}Ra in known quantities and which was later poured over a white filter introduced into a 5 cm Petri dish. The ^{7}Be detection limit for a 24 h time count was 0.1 mBq m^{-3}.

The detection efficiency shown when analysing rain water was carried out by using a standard sample of known quantities of ^{137}Cs and ^{226}Ra which is later evaporated on a plastic stand and then packed in a 9 cm Petri dish. The obtained detection limit fo 24 h time count is 730 Bq m^{-3}.

To calculate the detection efficiency in measurement of concentrations of gamma emitt radionuclides in soil a sample, with the sa geometry and density as the samples to be analy was used containing a known quantity of ^{152}Eu. the measuring conditions used the limit of detection for a 24 h count for ^{7}Be was 2.2 Bq k (66 Bq/m^{2}).

Results

The region of Cantabria is situated on the nort coast of Spain at a latitude of 43°N. The rair averages between 1000 and 1500 mm. which is ev distributed throughout the year although sum usually tends to be drier than the rest of the seas Table 1 shows the variation in ^{7}Be concentrat measured in air samples taken on a weekly quarterly, rainfall and soil during the 1992–1 period.

The range of concentrations measured in quarterly mixed samples obtained from the air fi was betwwen 2.2 and 4.5 mBq m^{-3}. These re turned out to be similar to those obtained by o authors also situated in the northern hemisphere at different latitudes (see Table 2). Our we measurements gave a wider range of result 0.1 mBq m^{-3} (Lid) and 9 mBq m^{-3}.

Considering that ^{7}Be production in the troposp tends to be relatively constant depending on latitude (Lal *et al.*, 1958; Young and Silker, 1 1980), then the amount of ^{7}Be in the air colum that latitude may also remain approximately stant, and subtractions to this would occur prim by wet precipitation processes. So, an increas precipitation would lead to a decrease in concentrations in the air. Figure 1 gives the valu ^{7}Be concentrations obtained every 3 months July 1992 to June 1995 in mBq m^{-3} and also the fall, in mm, for every 3 month period. In Fig. 1

Table 2. Average annual fallout rates at different locations

Fallout (Bq m^{-2} yr^{-1})	Location	Latitude	Rainfall (mm)	Authors
867	Arkansas, U.S.A.	38 N	1070	Lee *et al.* (1985)
898	Chilton, U.K.	51 N	822	Peirson (1963)
1030	Canberra, Australia	35 S	660	Wallbrink and Murray (1994
1249	Heidelberg, Germany	52 N	810	Schumann and Stoeppler (196
1267	Bombay, India	19 N	2277	Lal *et al.* (1979)
1618	Milford Haven, U.K.	51 N	1328	Peirson (1963)
2850	Bermuda	33 N	1700	Turekian *et al.* (1983)

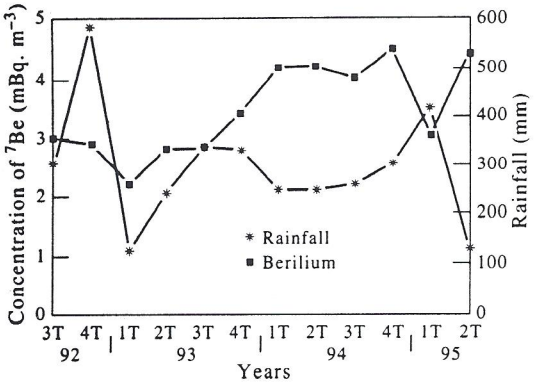

Fig. 1. Variation of ^{7}Be concentration in atmospheric aerosols vs rainfall.

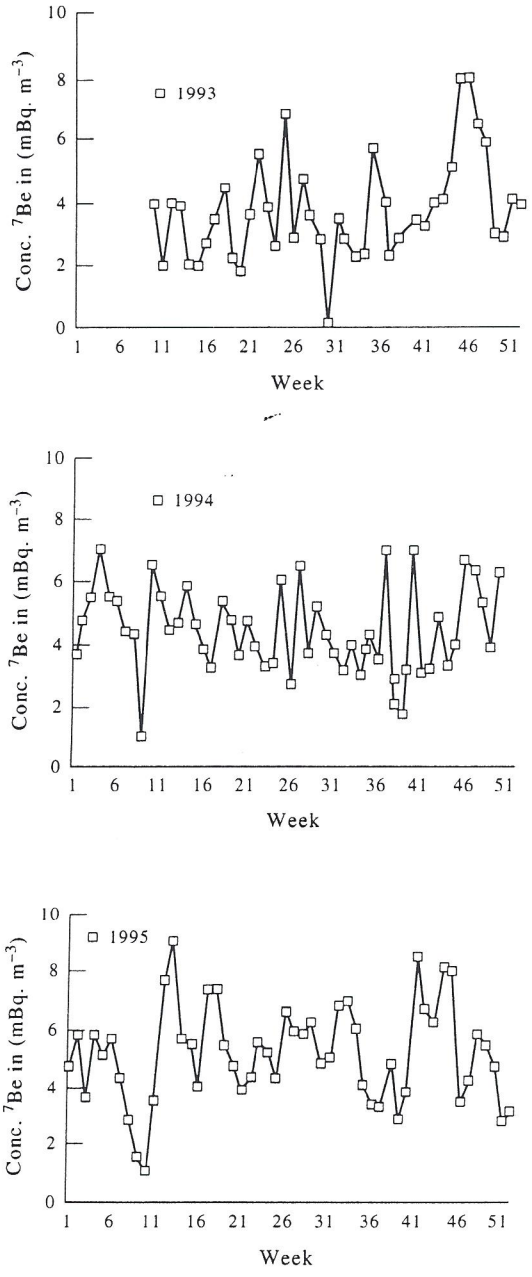

Fig. 2. Annual variations of ^{7}Be in air samples.

can observe qualitatively the inverse correlation that exists between these two variables; it is possible, with the exception of two points (1 T and 2 T of 1993) to see a decrease in ground-level air activity concentration as net quarterly rainfall increases. Figure 2 shows the changes detected in the air samples taken on a weekly basis during the 3 yr studied. No seasonal changes could be detected. The observed values show erratic variations within each year. This seems to indicate that air ^{7}Be concentrations are affected basically by local weather conditions such as humid precipitation and air stability. These are important factors considering the average rainfall of about 1000 mm in this area of Spain.

The concentration range of ^{7}Be in rain water samples was between the detection limit of 0.73 Bq L^{-1} and 5.05 Bq L^{-1}. This range was similar to those obtained by other authors, e.g. 0.59–2.74 Bq L^{-1} (Peirson, 1963) or 0.02–5.9 Bq L^{-1} (Wallbrink and Murray, 1994).

No relationship was found between the ^{7}Be concentrations in rain water samples taken quarterly and the amount of wet precipitation that fell in the same period of time.

The ^{7}Be coming from the dry deposition is inversely related to the amount of rain water collected during the same period of time. The contribution of dry to the total fallout was similar to that found by Brown *et al.*, 1988, which was between 5 and 8% of the total amount of ^{7}Be deposited. This values fall within the uncertainties estimated for total inventories. For this reason and the fact that we used weekly and monthly measurements (humid precipitation is to be expected), that this component was not determined on a systematic basis.

The concentrations of ^{7}Be in soil vary from 230 to 470 Bq m^{-2}. From this we can deduce that the annual fallout of ^{7}Be, considering its radioactive desintegration, is between 1100 and 2240 Bq m^{-2} yr^{-1}, similar to those found in other latitudes (Table 2). From the data given in Table 1 we can once again deduce that annual berylium deposition has a closer relationship to variations in precipitation, in the form of rain, than the difference in latitudes.

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