The Distribution and Deposition of Radionuclides over Antarctica

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ABSTRACT

The production of artificial radionuclides from the atmospheric thermonuclear testing of the 1950s onwards has been detected in Antarctica. The presence of these pollutants has been utilised by science to produce meteorological information about the Antarctic continent. Accurate analysis of radionuclide stratospheric residence times, air to ice radionuclide concentration ratios, fallout characteristics of radionuclides and snow accumulation rates are made possible by their presence in Antarctica. The artificial radionuclides of ^{137}Cs and ^{90}Sr and natural radionuclide ^{210}Pb have ideal decay properties for analysing the meteorological applications. By understanding the processes that current pollutants follow in Antarctica we can plan effectively for new and emerging pollutants that threaten the continent.

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1 INTRODUCTION

Artificial radionuclides are radioactive nuclei that are not naturally found in nature. They are produced through the anthropogenic processes of nuclear reactions, particle accelerators and radionuclide generators. Naturally occurring radionuclides are the daughter nuclei of long lived radioisotopes that were formed at the time of planet Earth's accretion. In the Antarctic continent the deposition, concentration and distribution of artificial radionuclides such as caesium 137 and strontium 90 and the natural radionuclide lead 210 can be analysed from snow and ice core samples. Artificial and natural radionuclide distribution can also be determined from non-ice covered region samples of soil, plant and sediment (Pourchet, Magand, Frezzotti, Ekayin & Winther, 2003). Koide, Michel, Goldberg, Herron and Langway. (1979) published a comprehensive depositional history of artificial radionuclides in the an ice shelf region.

Caesium 137 is an artificial radioactive isotope of caesium that undergoes beta decay and is a common fission product in nuclear reactors, weaponry and testing. The half-life of approximately 30 years makes ^{137}Cs a good candidate for chronological dating while also implying a significant amount of geographical distribution before all the caesium decays to its stable daughter product, barium. Caesium has high solubility and readily dissolves in water on account of most of its chemical compounds being salts. Strontium 90 is another nuclear fission product, has a half-life of approximately 28.8 years and also decays by beta emission. ^{90}Sr exhibits biochemical behaviour very similar to its other periodic group two element, calcium. The half-life similarity to ^{137}Cs warrants similar geographical distribution concerns.

Lead 210 is a global naturally occurring radionuclide and daughter product of radioactive radon gas. ^{210}Pb decays by alpha and beta emission with a half-life of approximately 22.3 years. The lack of radon sources in Antarctica (uranium ores, phosphate rock, shale and some igneous and metamorphic rocks) in conjunction with the long transit times for non-Antarctic air masses to reach the continent result in very low natural ^{210}Pb levels in Antarctica. ^{210}Pb levels in Antarctica appear to be constant and have not been altered by hydrogen bomb testing (Bull, 1971). The consistent natural flux of ^{210}Pb deposition in Antarctica results in the ability for it to be used as an accurate dating mechanism for the last hundred years (Eisen et al., 2008). The differentiation between these radionuclides can be conducted with high resolution gamma spectrometry by detecting the different decay energies of each radionuclide. Careful analysis of the concentration, ratios and radioactivity of these nuclei can lead to useful meteorological information about the Antarctic continent.

The atmospheric thermonuclear experiments carried out over the 27 year period from 1953 to 1980 resulted in the deposition of artificial radionuclides in Antarctica through circulated transport in the upper stratosphere and atmosphere (Pourchet et al., 2003). The radioactivity detected on the Antarctic continent as a result of the nuclear testing has been measured for various years in the past and has been shown at times to be 20 times higher than the natural background radioactivity levels. Figure 1 displays the beta decay radioactivity in disintegrations per hour as a function of year for data recorded at the South Pole Station, which is considered representative of other Antarctic research bases (Pourchet, Pinglot & Lorius, 1983).

From 1950 to 1955 the radioactivity levels are reasonably constant at 80-100 disintegrations per hour per kilogram. A vast spike is seen in the spring

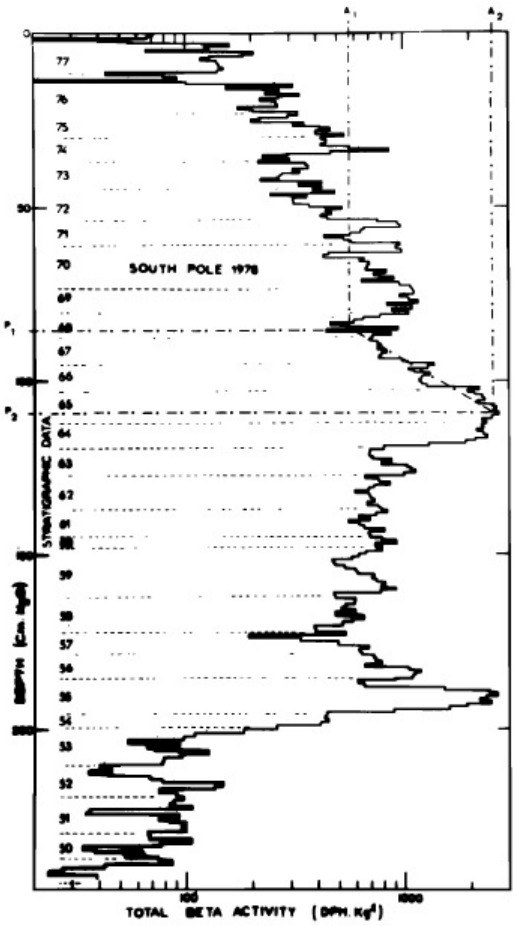

Figure 1: Total beta activity for South Pole Station with snow expressed in water equivalent (Pourchet et al., 1983).

of 1955 which is attributed to the radionuclide distribution throughout the world from Operation Castle, a series of high energy thermonuclear tests conducted at Bikini Atoll in the Marshall Islands which commenced on March 1 1954. From 1956 to 1964 a number of atmospheric tests caused the radioactivity levels in Antarctica to float around values approximately 10 times larger than values for the years previous to 1954. 1965 saw another large spike attributed to stratospheric transport of radioactive material from the Dominic and U.S.S.R test series (Pourchet et al., 1997). From 1966 onwards the radioactivity levels slowly trend downwards and return to original natural background quantities. The small spikes in radioactivity during the downward trend are attributed to smaller scale nuclear testing conducted primarily by China and France.

The presence of natural and anthropogenic radionuclide ratios in the Antarctic in conjunction with known thermonuclear events can lead to specific scientific determinations of: radioisotope stratospheric residence times; concentration ratios of radionuclides in the air and ice; radionuclide fallout's distribution characteristics; and the important annual snow accumulation rates. This report will focus on these four areas and briefly draw conclusions on the implications they hold for science.

2 METEOROLOGICAL APPLICATIONS OF RADIONUCLIDE FALLOUT

2.1 Radioisotope stratospheric residence time

Stratospheric residence time for radioisotopes is a parameter used to describe how long radionuclides will circulate in the atmosphere from their time of creation in a nuclear event until their deposition on the lithosphere. Comparison between the years of known spikes in thermonuclear testing and the corresponding snow layer radioactivity measurements allow the stratospheric residence time for particular radionuclides to be measured. The snow layer activities are most commonly measured in continuous core samples. Radionuclides with a short half-life will decay to their daughter products while still in the atmosphere and it can be difficult to measure and conclusively determine an origination event, especially in situations where the daughter product occurs naturally. Radionuclides with long half-lives will have a scarcity of disintegrations and require long observation periods to detect radioactivity levels and draw conclusions about their origin.

The similarity of the half-lives of ^{137}Cs and ^{90}Sr in conjunction with their same decay method, beta decay, make them ideal candidates for sampling and are often combined and interpreted for simplicity as a single radioisotope having a half-life of 29 years. This manner of simplification was utilised in stratospheric residence time research conducted by Pourchet et al. (1983) and Pourchet and Pinglot (1979). To measure their stratospheric residence times relatively simple mathematical relationships are used with a small number of assumptions. Assuming the radioactivity measured in snow core samples is proportional to the amount of radioactivity in the air at the time of deposition then radioactivity in the snow at the time of measurement, A is the product of the proportionality, ρ and the radioactivity level in the atmosphere at the time of measurement, N.

$$A = \rho N$$

Radioactive decay is accounted for using straightforward exponential decay equations to deduce radioactivity levels at time of deposition. Two layers located at differing depths with different radioactivities can be measured, sample 1 and 2. A simple equation relating the natural log of radioactivities, A and the time of deposition of each sample, t is then derived.

$$\alpha = \frac{\ln A_1 - \ln A_2}{t_2 - t_1}$$

Taking the inverse of this result produces an atmospheric residence time.

The research conducted by Pourchet and Pinglot. (1979) concluded stratospheric residence time for the caesium strontium combined sample at 1.63 years. This was the time taken from the formation, due to the previously mentioned nuclear events, to the deposition on the Antarctic continent. Reiter (1977) produced a value of 1.69 years for ^{90}Sr complemented by Fabian, Libby and Palmer. (1968) who found a 1.6 year residence time. Updated research in 1983 conducted by Pourchet et al. (1983) put the value at 1.5 ± 0.2 years for the caesium strontium sample. In these studies it was assumed that the radioactivity deposited in the snow layers was proportional to the radioactivity in the air at the same time.

2.2 Air and snow radionuclide concentration

The ice, snow and glaciers of the Antarctic environment contain radionuclide fragments in measureable concentrations. Ice and snow core measurements give a sequence of strata that can be aged. By combining the ice core data with measured radioactivities in the air for the same year it becomes possible to produce profiles of previous atmospheres. We can draw conclusions about how much atmospheric radioisotope pollution is deposited on the continent and how much remains in the stratospheric circulation systems. A quantitative research approach conducted by Pourchet et al. (1983) utilises a simple ratio. The ratio is expressed as the concentration of the sample of interest in air over the concentration of the same sample of interest in snow.

$$\phi = \frac{c}{k} = \frac{\text{air concentration } (g\ m^{-3})}{\text{concentration in snow } (g\ g^{-1})}$$

Snow and ice core samples are often processed by melting, weighing, acidifying and filtering on ion exchange paper, where all radionuclides become trapped in the paper. The filters are analysed by gamma spectrometry (Delmas Pourchet, 1977). Bendezu (1978) found the air and snow concentration ratio of ^{210}Pb to be 2.4 at Dumont d'Urville for the 16 year period between 1955 and 1971. The four year period between 1959 and 1963 had a concentration ratio of 1.3 at South Pole station. The concentration ratios for ^{90}Sr conducted as part of the same research were recorded at 1.8 for Dumont d'Urville and 0.7 for South Pole station over similar time periods.

2.3 Fallout and distribution of radionuclides

After transport in the stratosphere just prior to deposition airborne radionuclides above Antarctica are subject to a large array of environmental factors. Upon reaching the Antarctic the radionuclides do not deposit in a well-defined, uniform manner. The radionuclide fall-out is subject to factors such as snow precipitation and the local geographical parameters of temperature, altitude, distance to coast and wind profiles. Over the Antarctic continent 20% of the total land area receives less than 50 mm of water per year and 40% of the land area receives less than 100 mm per year (Pourchet et al., 1983).

Pourchet et al. (1997) provides a comprehensive analysis of the radionuclides ^{137}Cs, ^{90}Sr and ^{210}Pb regarding their spatial distribution and fall-out over Antarctica. Over the entire Antarctic continent for the period of 1955 to 1980 the radioactivity levels of ^{137}Cs ranged from 17 Bq m^{-2} to 174 Bq m^{-2}. The mean value for dry fallout, particles leaving air masses on their own accord, was put at 82% of the total fallout mass (Pourchet et al., 1997). The caesium 137 distribution was shown to have correlation to the local temperature and altitude with the most concentrated fallouts of caesium found at the high altitudes of more than 3000 metres and temperature of less than -40 degrees Celcius. There was a general increase in radionuclide deposition as altitude increased and temperature decreased. The highest concentration of 174 Bq m^{-2} was recorded at DB station at an altitude of 3400 metres, at a significant distance from the coastline, located the central area of the Antarctic Plateau.

In research regarding the distribution of ^{90}Sr conducted by Pourchet et al. (1997) the radioactivity levels presented three main spikes: the 1953 increase attributed to the 1952 Ivy bomb test; the 1955 increase due to the Operation

Castle test series; and the late 1962 increase accredited to the re-engagement of nuclear tests in the atmosphere in 1961. A strong correlation was found between the amount of ^{90}Sr deposited and the water accumulation rate. Roi Baudouin Station had a ^{90}Sr radioactivity of 88.8 Bq m^{-2} for the 10 year period between 1953-1963 and South Pole Station had a radioactivity of 37 Bq m^{-2} for the same period. Roi Baudouin Station had a water accumulation rate of 40 cm yr^{-1} compared to the lower 6.8 cm yr^{-1} for South Pole Station. Scott Base recorded ^{90}Sr radioactivities of 70.3 Bq m^{-2} with a water accumulation rate of 11.4 cm yr^{-1}. The study goes on to declare more observations are required to provide conclusive correlation. Wilgain, Picciotto and De Breuck. (1965) support these findings by concluding that the total deposition of ^{90}Sr for the 10 year period between 1953 and 1963 would be in the range of 37 – 111 Bq m^{-2}.

Pourchet et al. (1997) analysed the deposition of the naturally occurring ^{210}Pb and found radioactivities of 0.9 – 8.2 Bq m^{-2} per year which is in agreement with the research conducted by Roos, Holm, Persson, Aarkrog and Nielsen. (1994). The elevated concentrations of ^{210}Pb activity in McMurdo Sound were attributed to the close proximity of Mount Erebus (Pourchet et al., 1997). Roos et al. (1994) however found that the samples from Deception Island did not have increased ^{210}Pb radioactivities despite the presence of a volcano (last eruption in 1964) and ice-free areas.

2.4 Annual snow accumulation rates

Manned presence in Antarctica is relatively modern history and the time since reputable scientific endeavours begun on the continent is even more recent. It is comprehensible then that there is few directly measured annual snowfall accumulation studies. Positive and reliable results are only available from stations that have been established for many years (Picciotto Wilgain, 1963). A strong aid in the determination of annual snow accumulation rates is the presence of radioactive isotopes. Picciotto and Wilgain (1963) state that finding a reference level in the snow corresponding to a definitive date is the best method for measuring mean snowfall accumulation. There are different methods for determining snow accumulation depending on the radionuclide of choice. This section will focus on the method used for the radionuclides ^{137}Cs and ^{90}Sr and the method utilised for ^{210}Pb.

Determination of snow accumulation rates using ^{137}Cs and ^{90}Sr involve the volume of snow accumulated between two known dates, shown by the snow radioactivity. Certain amounts of extrapolation to other time periods can be undertaken. Previously mentioned radioactivity reference levels of 1955 and 1965 make it possible to analyse accumulation rates for two periods; 1955 to 1965, and from 1965 onwards (Pourchet et al., 1983). Pourchet et al. (1983) took beta activity profiles for a large number of different stations in East Antarctica and a small number of stations in West Antarctica in addition to stations from Dumont d'Urville to Dome C as illustrated by figure 2.

Figure 3 illustrates a beta profile of radioactivity as a function of depth for one such station. The finding of this research illustrated that for the period of 1965 onwards there was a mean annual snow accumulation rate increase of 35% compared to the 1955 to 1965 period over a large area of East Antarctica.

^{210}Pb is a prime radionuclide candidate for radiochronology in the determination of snowfall accumulation calculations. As expected ^{210}Pb activity

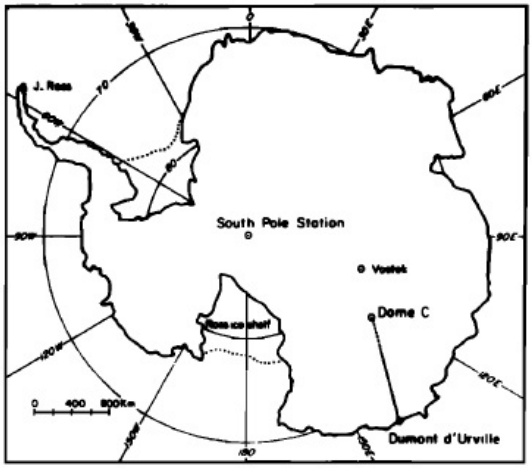

Figure 2: Map of Antarctica showing sites of snow accumulation studies (Pourchet et al., 1983).

decreases as the depth of firn and ice layers increases. The age of a firn layer is given in Eisen et al. (2008) by the equation

$$t = \frac{1}{\lambda} \ln \frac{A_0}{A_z}$$

Where λ is the decay constant of ^{210}Pb and activities A_0 and A_z are the activities at the surface and depth z respectively. The average snow accumulation of snow above this depth is given by

$$b = \frac{z\lambda}{\ln \frac{A_0}{A_z}}$$

The important assumptions for ^{210}Pb analysis are: the average ^{210}Pb precipitation has remained constant for the last two centuries; the parent product ^{226}Ra concentrations are considered negligible; no diffusion of parent product ^{222}Rn has permeated into the tiny air spaces in the ice; and that the ^{210}Pb remains in its location of deposition (Eisen et al., 2008). Despite these assumptions snow accumulation calculations from this method are considered reliable.

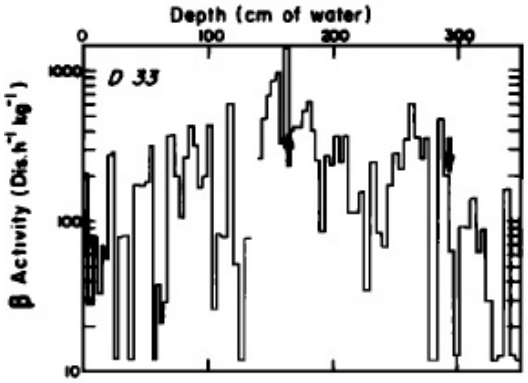

Figure 3: Beta activity for D33 Station as a function of depth of water(Pourchet et al., 1983).

3 CONCLUSIONS

The most indicative signal of pollutants reaching the Antarctic is from thermonuclear atmospheric testing. The events produced nuclear debris in the stratosphere which were deposited on the Antarctic continent after a stratospheric residence time.

The stratospheric residence time for the radionuclides ^{137}Cs and ^{90}Sr are considered equal due to their similar decay properties and chemical compounds. The most up to date stratospheric residence time is given as 1.5 ± 0.2 years (Pourchet et al., 1983). ^{210}Pb is a naturally occuring radionuclide with low levels constantly found in the Antarctic. It is not formed as a product of nuclear testing.

Air and snow radionuclide concentration ratios are dependent heavily upon fall-out and distribution characteristics but overall show a propensity to have a ratio value greater than 1, indicative of the air being more saturated with the radionuclide than the snow at the time of deposition.

The distribution and fall-out of radionuclides that occurs over Antarctica has been shown to be heavily influenced by geographical parameters such as the case of the inland DB station at an altitude of 3400m having the highest deposition of 174 Bq m^{-2}. Supported by Pourchet (1997) who found the highest ^{90}Sr depositions in locations of the highest water accumulation rates. Volcanic proximity is not always a precusor to higher levels of ^{210}Pb.

The techniques used to analyse snow accumulation rates are highly dependent on the physicsl characteristics of the chosen radionuclide. Prior knowledge of the radionuclide's presence in Antarctica is imperative if accurate radiochronology is to be achieved.

All four meterological applications of radionuclides in the Antarctic are strongly linked together and cannot be isolated. Simple radioactie dating of layers provides insight into stratospheric residence times and snowfall accumulation rates. Utilising the physical decay properties of radionuclides allows for conclusions to be drawn about their concentrations in the past which is a precusor to fall-out and distribution patterns. Understanding the pollutants that are present in the Antarctic now and how we utilise them for the benefit of scientific understanding provies foundations for understanding emerging pollutants to the Antarctic continent.

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