Possibilities of using lead 210 as an atmospheric tracer

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(Manuscript received October 18, 1965)

ABSTRACT

The high sensitivity of the lead 210 measurement through the polonium 210 enables to date the neve layers of the Antarctic continent. Thus, appear some correlations between the concentration of lead 210 in the snow and the solar activity.

The study of the atmospheric concentration of lead 210 above the oceans having proved the stratospheric origin of this nuclide in these regions, the former correlations are therefore due to a possible action of the sun's corpuscular radiations on the stratosphere-troposphere transfers.

1. The interest of measuring the concentration of lead 210 in various environments has been stressed by many authors and particularly in the studies made by PICCIOTO & WILGAIN (1963) and PICCIOTO, CROZAZ & DE BREUCK (1964) on the Antarctic continent neves. These authors have shown that, if the snow is sufficiently cold to avoid fusion phenomena, the elements contained in the ice crystals remain fixed therein. Thus, lead 210 captured by falls, under the Antarctic climatic conditions, remains fixed inside the neve layers. The measurement of its concentration decay as a function of depth, as compared to its radioactive period, makes it possible to determine the time which has been necessary for the accumulation of a given height of the neve. This method does not need such a continuous and minute marking as it is necessary for stratigraphic methods, even when they are associated with the measurement of isotopic ratios O^{18}/O^{16}; or D/H. However, it assumes the hypothesis, certainly a non-strict one, that the yearly fallout of lead 210 is constant. Variations in snow accumulation rate may then cause slight fluctuation on the observed concentrations.

The difficulty of these measurements lies in the very low β radiation energy of this nuclide (0.017 MeV). In fact, lead 210 has mostly been measured by means of its daughter product, bismuth 210, which is always in radioactive equilibrium with the lead owing to its half-life of 5 days and which yields a 1.17 MeV β. The working out of a method for the chemical separation of small traces of polonium 210, a daughter of bismuth and an α emitter (5.3 MeV) has enabled us to obtain a sensitivity of 0.05 dpm per sample (NEZAMI, 1965). This method utilizes as a carrier a sample of lead which is sufficiently old to be free from isotope 210. It also utilizes a semiconductor α spectrometer. Fig. 1 shows, for instance, the α spectrum obtained from a sample of 2 litres of melted snow collected in the Antarctic (NEZAMI, LAMBERT, LORIUS & LABEYRIE, 1964).

2. This high sensitivity has enabled us to study the fluctuations of the lead 210 along a snow core drawn from the border of the Antarctic continent. It is apparent on Fig. 2, using semi logarithmic coordinates that, on the average, concentration decreases exponentially as expected. The average height of falls may thus be worked out to 13.8 ± 1.5 cm of water annually (in the course of the last century) (NEZAMI, LAMBERT, LORIUS & LABEYRIE, 1964). Under the assumption that annual snow fall accumulation variations are comparatively small, or at least cancel one another, it is also possible to determine the epoch of formation of the various neve layers.

It is to be seen that, on the same figure, the lead concentration shows several points for which the fluctuations are very much larger

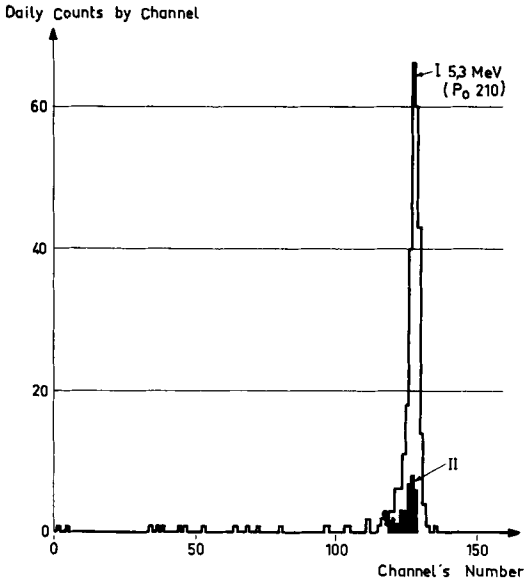

Fig. 1. I. α spectrum of a sample of neve. Sample $n \cdot 12$ (depth 9.6 meters). Activity = 0.8 d.p.m. 10 KeV by channel. Resolution 0.8 %. II. Spectrum of an identical mass of distilled water having had the same treatment.

than the allowed value for the maximum experimental errors. The same fluctuations appear at the same levels for another core drawn a few meters away from the first sample (Fig. 3).

This type of fluctuations can be caused only by a definite variation in the concentration of snow while it was falling and not by an annual change in snow height. This latter phenomenon could not, it is obvious, account either for fluctuations of this magnitude or above all for the observed concentration inversions.

The periodic character of the fluctuations (of period 11 years approximately) has induced us to compare the lead concentration to the sun activity as determined from the mean annual number of recorded spots. It is seen, on Fig. 4, that the 3 first maximums correspond well to the maximum solar activity. Further on, for samples over 30 years old, the accuracy of measurement and, in particular, the uncertainty on the half-life of lead 210, do not make it possible to hope that such a good correlation

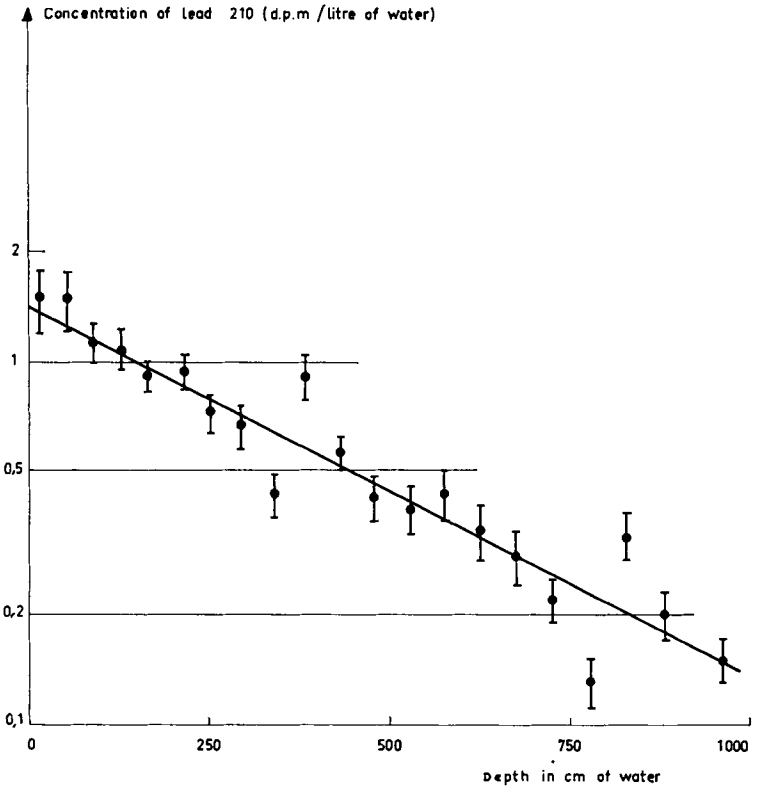

Fig. 2

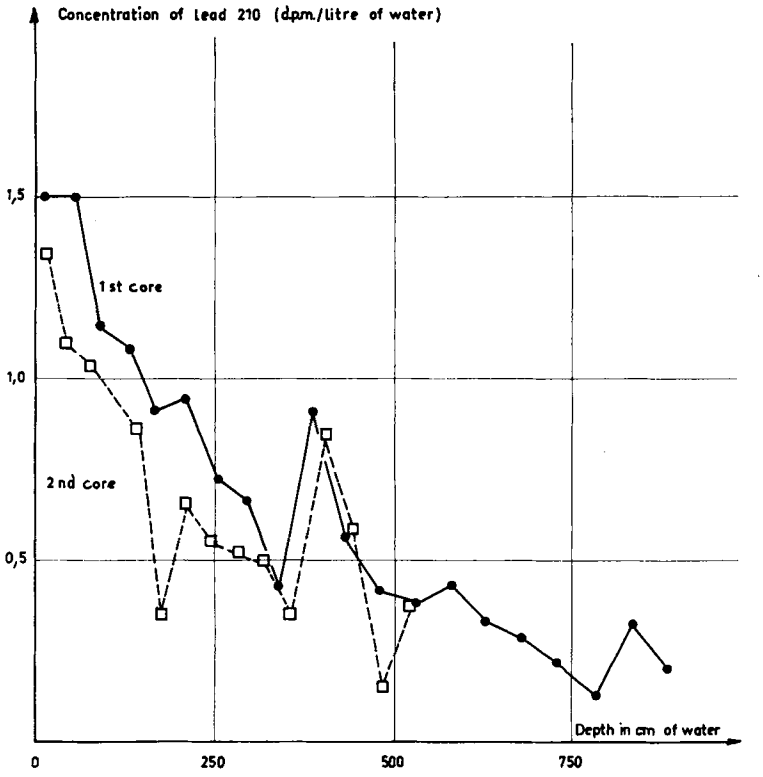
Concentration of lead 210 (dpm./litre of water)
1,5
1,0
0,5
1st core
2nd core
Depth in cm of water
0
250
500
750

Fig. 3

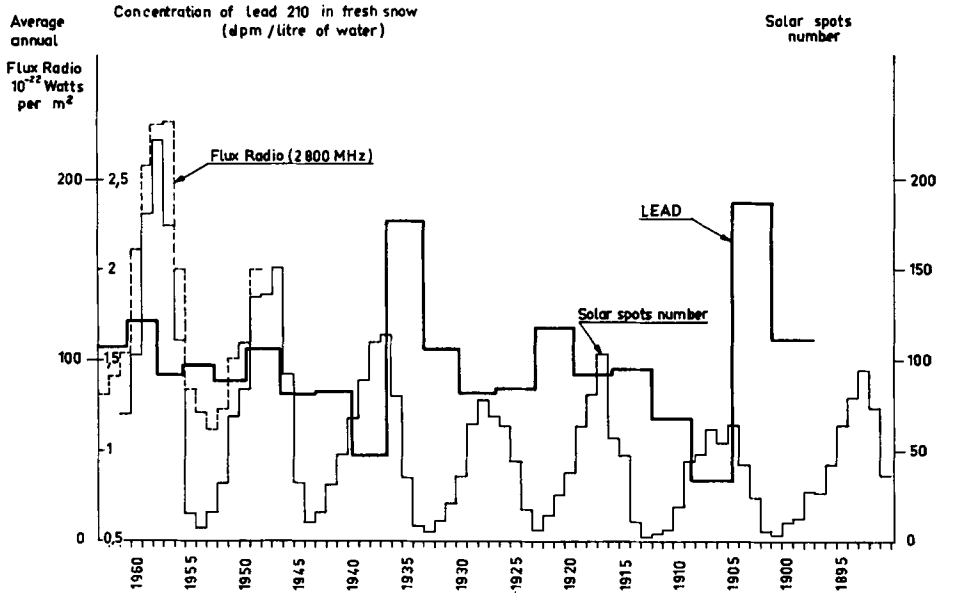
Average annual
Flux Radio 10^{-22} Watts per m²
Concentration of lead 210 in fresh snow (dpm / litre of water)
Solar spots number
Flux Radio (2800 MHz)
LEAD
Solar spots number
200
100
0
2,5
2
1,5
1
0,5
200
150
100
50
0
1960
1955
1950
1945
1940
1935
1930
1925
1920
1915
1910
1905
1900
1895

Fig. 4

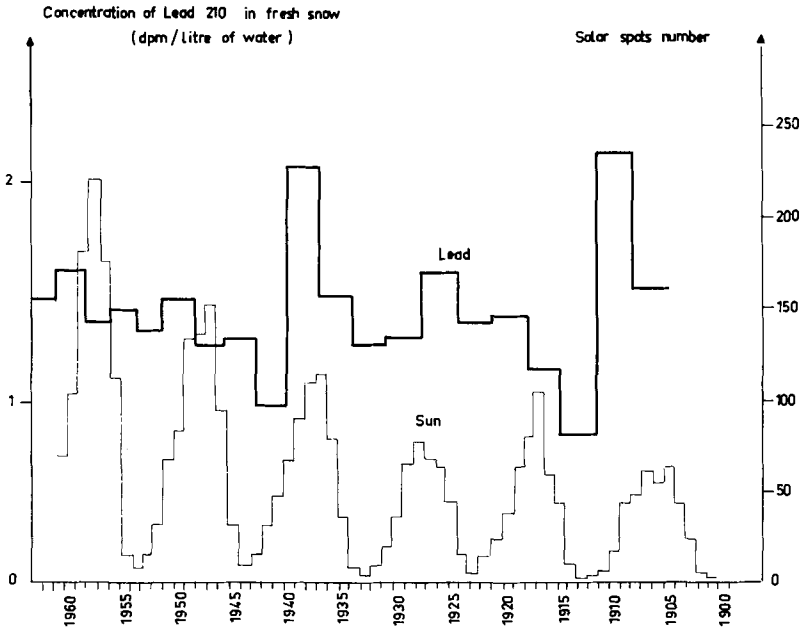

Fig. 5

may be attained. Yet, a proper choice of parameters enables the working out of Fig. 5, in which the correlations remain valid for the 6 last solar cycles (LAMBERT, NEZAMI & LABEYRIE, 1965).

3. Whatever the character and the cause of these fluctuations are, they raise the question of the origin of Antarctic lead 210 and of its mode of transfer from the surface of continents, which constitutes the source of radon.

It is known that lead 210 present in the lower atmosphere or, which amounts to the same, fallen on the ground, may originate either from the desintegration of radon in the lower atmosphere or from the injection into the troposphere of lead 210 accumulated in the stratosphere after a vertical diffusion of radon. However, numerous measurements of lead 210 atmospheric concentrations made in continental stations, show up the prevalent part of lead 210 which was yielded locally by the first daughter products of radon 222 (PATTERSON & LOCKHART, 1964). One may hope to obtain more representative values for the general circulation of lead 210 by studying its configuration above the oceans. Therefore, in order to study whether the atmospheric concentration of lead 210 obeyed the laws governing artificial radioactive aerosols of stratospheric origin, ships transporting French expeditions to the Antarctic and to the sub-Antarctic islands were used to study the distribution of this nuclide as a function of latitude.

The samples were collected at a height of approximately 10 meters above sea level after being filtred through cellulose filters of approximately 10 cm^2, replaced at the rate of one filter a day. This made up about 360 m^3 daily. On each filter, the α radioactivity was measured either instantaneously or after a delay ranging from one hour to a year. It is very difficult to know the efficiency of such a measurement, carried out without any chemical separation. However, we were able to calibrate our sampling and measuring equipment by means of series of alternate α and β measurements, during the decay of the short lived daughter products of radons 220 and 222.

The β counting efficiency being known, it is thus possible to determine a gross α counting efficiency which makes allowance for the uneven penetration of aerosols inside the filter and which, fortunately, proved quite constant for all samplings. The β radioactivity was also measured, after delays of one week. The mean

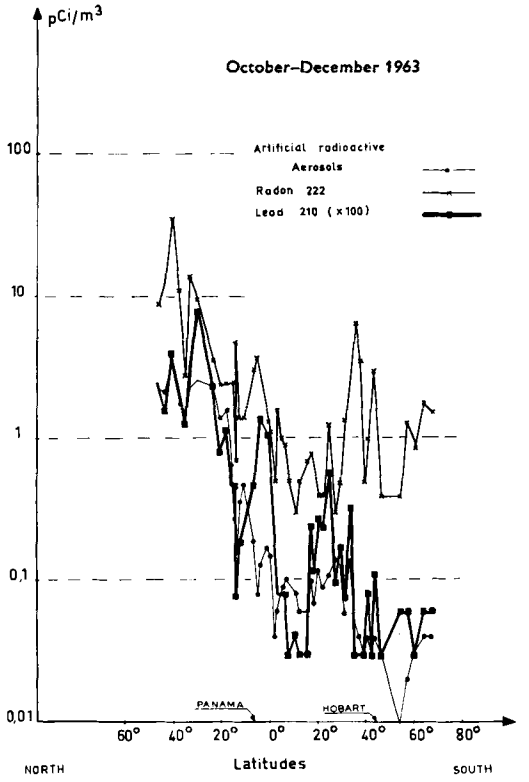

Fig. 6

concentrations at various latitudes for radon, thoron, polonium 210, lead 210 and artificial radioactive aerosols were thus obtained.

It appears on Figs. 6 and 7 that, when the concentration of radioactive artificial aerosols is constant, the variations in lead 210 concentration are correlated to those of radon. Inversely, when the radon concentration is constant, that of the lead 210 varies together with the artificial radioactivity. However, as a whole, the order of magnitude for atmospheric lead 210 radioactivity varies in accordance with the total β radioactivity. This makes possible to state that lead 210 has essentially a stratospheric origin. Thus, for the years 1963–1964, the radioactivity due to lead 210 was approximately 1/100th of the total β radioactivity.

If the sampling is made over a continent, the high concentration of radon and lead 210 produced directly, completely hides this result. This may be seen on Fig. 8, on a voyage from France to the Kerguelen islands, across the Mediterranean, the Red Sea and along the African coast of the Indian Ocean, i.e. in regions where the radon concentration remains close to its continental value (50 to 100 pCi/m³).

4. If the main part of lead 210 measured in the Antarctic originates from the stratospheric reservoir, it may be inferred that the fluctuations observed in the fallout must be caused by variations in the rate of injections of stratospheric aerosols into the troposphere. If it is accepted that the fluctuations we have observed are effectively correlated to the solar activity, it must then be admitted that the solar activity and particularly corpuscular emissions may cause or amplify such injections.

Another hypothesis could be that part of the collected lead 210 directly originates from the Sun and so adds up to lead 210 yielded by the Earth. Only the balance of lead 210 fallouts on the Earth surface makes it possible to see whether the number of lead atoms deposited appreciably exceed the number of radon atoms introduced into the atmosphere from the continents surface, and on the other hand if the sun

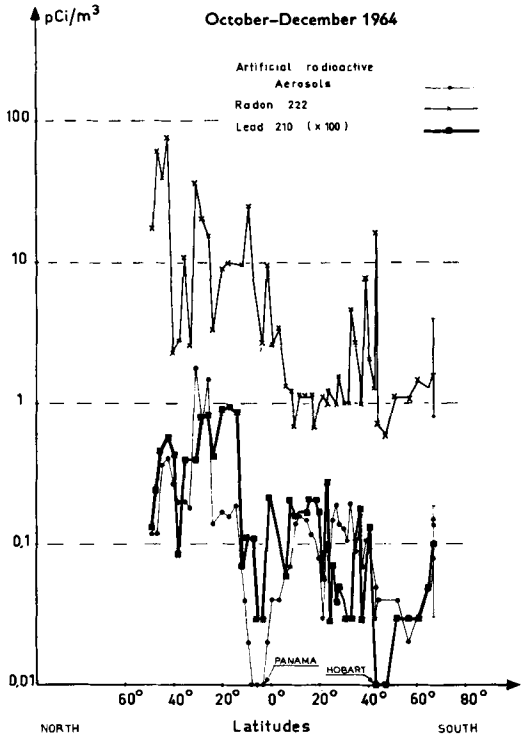

Fig. 7

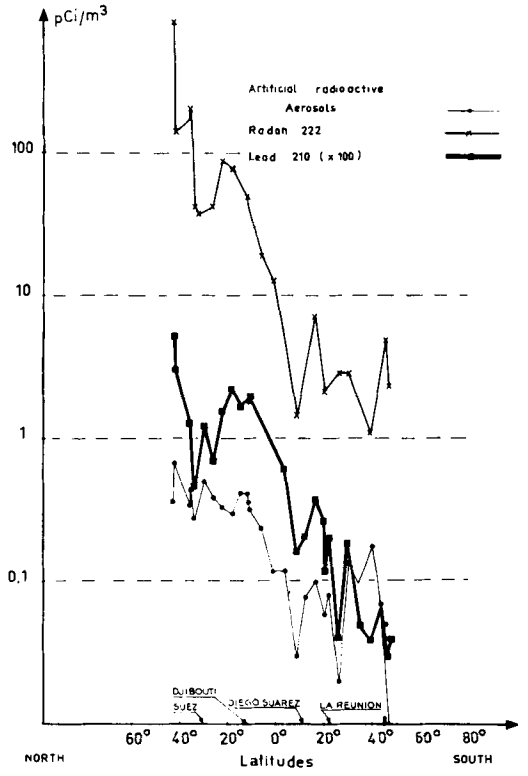

Fig. 8

is likely to yield such a vast quantity of this nuclide.

The working out of this balance is extremely hazardous, as it is necessary to make allowance, particularly for dry and wet fallout account being taken of their unequal distribution in function of rainfalls, latitude and season as well as for the possible unequalities between the fallout on the continents and on the oceans. Moreover, the numerical data which we have obtained are smaller in number, and are insufficiently completed by the existing publications. The same applies, similarly, to the radon balance.

Allowance being made for these restrictions, we have shown that the number of lead atoms deposited on the whole Earth is to be estimated at 2.10^{25} annually, practically in equilibrium with the $2.5{,}10^{25}$ radon atoms yielded each year by the continents (LAMBERT & NEZAMI, 1965). These figures are anyway too high to include a non negligible part originating from the Sun, taking into account what is known of the solar wind, and the relative abundance of hydrogen and of heavy metals on the Sun surface.

It therefore seems more reasonable to investigate henceforward how the corpuscular emission from the Sun could cause injections into the troposphere of stratospheric aerosols.

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ВОЗМОЖНОСТИ ИСПОЛЬЗОВАНИЯ СВИНЦА 210 В КАЧЕСТВЕ ТРАССЁРА АТМОСФЕРЫ

Высокая чувствительность измерения свинца 210 посредством полония 210 позволяют датировать слои фирсового льда в Антарктиде. Таким образом, появляются некоторые связи между концентрацией свинца 210 в снеге и солнечной активностью. Изучение концентрации свинца 210 в атмосфере над океаном показало стратосферное происхождение его ядер в этих областях, поэтому указанные корреляции возможно обусловлены воздействием солнечного корпускулярного излучения на стратосферно-тропосферный перенос.