^{210}Pb-Atmospheric deposition in lichen-carpets in northern Sweden during 1961–1969

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ABSTRACT

The ^{210}Pb-content of lichen-carpets collected in Northern Sweden (62.3° N, 12.4° E) from 1961 until 1969 has been studied. The analytical method used was wet ashing with nitric and perchloric acid and spontaneous deposition of ^{210}Po on silver discs. The α-activity of ^{210}Po was measured with a silicon-surface-barrier detector.

No maximum ^{210}Pb-content in lichen was found in 1963–1965 which could be compared with that previously found for ^{137}Cs, ^{55}Fe and ^{90}Sr, which were released by nuclear-weapons tests during 1961–62. Thus there was no special evidence of any release of significant amounts of artificial ^{210}Pb into the atmosphere by these tests.

The average ^{210}Pb-area-content of lichen during 1962–68 was found to be 16 ± 2 nCi/m^2 and the deposition rate of ^{210}Pb was estimated to be 1.7 ± 0.3 nCi/m^2 a. The effective half life for elimination of ^{210}Pb from the lichen-carpet was estimated to be 7 ± 2 a.

The vertical distribution of ^{210}Pb in the lichen-carpet was also studied, and the highest concentrations were found in the top layer.

Introduction

^{210}Pb in the atmosphere

The main source of ^{210}Pb in the atmosphere is ^{222}Rn, which is exhaled from the ground at a rate of about 0.5 pCi/m^2 s or 1 300 mCi per year, corresponding to a production-rate of atmospheric ^{210}Pb of 0.62 MCi per year, according to Israel (1958), Jacobi (1962) and Jaworowski (1969). The concentration of the long-lived decay products ^{210}Pb ($T_{1/2} = 21a$) and ^{210}Po ($T_{1/2} = 138d$) increases with height and reaches a maximum in the lower stratosphere. Burton & Stewart (1960) found that the concentration gradient of ^{210}Pb increased sharply in the vicinity of the tropopause, similar to fission products from thermonuclear explosions. Thus ^{210}Pb was assumed to descend in a manner similar to fission products. However, the source of the ^{210}Pb in the stratosphere is explained by the ascending air at the equator, according to the Dobson & Brewer circulation model, which carries not only ^{210}Pb but also ^{222}Rn and its daughter products, from which ^{210}Pb will be formed.

A number of investigators, however, have reported evidence for the presence of artificial ^{210}Pb in the atmosphere as a result of a series of nuclear-weapons tests.

Stebbins (1961) who measured ^{210}Pb and ^{181}W collected on filter papers in the atmosphere, first suggested that some of the ^{210}Pb was due to stratospheric injection from the US "Hard-tack" weapons-test series during 1958, and some of it was probably from an earlier test-series. Measurements of ^{210}Pb and ^{210}Po in stratospheric filter samples collected during project "Stardust" performed by Feely et al., (1964) indicate according to Jaworowski (1966) a release of fresh ^{210}Pb in late 1961 or 1962. Krey (1967) found simultaneous anomalously high concentrations of ^{210}Pb and ^{90}Sr in the upper stratosphere during 1966. As the ^{144}Ce/^{90}Sr ratio at this time indicated that the ^{90}Sr-activity resulted from the 1961–62 test-series, he suggested that the ^{210}Pb had the same origin. Bhandari et al. (1966), however, found no evidence for any appreciable production of ^{210}Pb by nuclear-weapons tests during the period 1959–61 when they studied the ^{210}Pb-concentration in certain atmospheric zones.

The concentration of ^{210}Pb and ^{210}Po in the surface-air and in rain-water has been measured continuously since 1961 by Peirson et al. (1966). They found that the ratio of ^{210}Po/^{210}Pb shows a seasonal variation during 1962 and 1963 which might be attributed to artificial production of ^{210}Pb during the nuclear-weapons tests of 1961–62. However, measurement of ^{210}Pb in ground-level air along the 80th meridian performed by Patterson & Lockhart (1964) indicates a lower concentration during Nov. 1961–Oct. 1962 than during Nov. 1960 – Oct. 1961.

Thus different studies of ^{210}Pb in the atmosphere give both positive and negative evidence for production of artificial ^{210}Pb by nuclear-weapons tests. Recently Beasley (1969) searched for anomalous levels of ^{210}Pb in soil and sediments at former test-sites on Bikini and Eniwetok Atolls. His results show concentrations of ^{210}Pb that do not exceed those expected to arise naturally. Thus it seems unlikely that the US tests of 1958–59 or 1961–62 have contributed significantly to the ^{210}Pb-contents of the atmosphere.

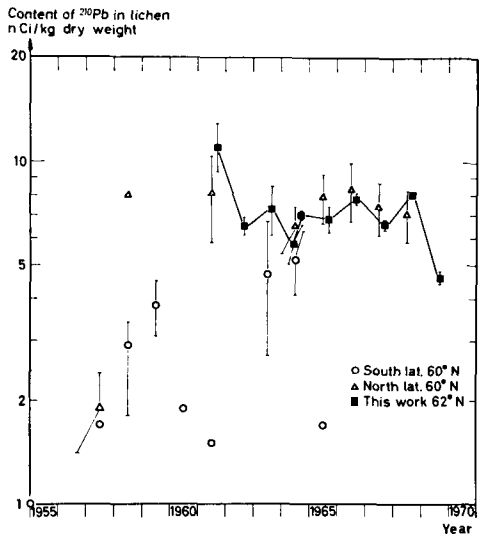

Fig. 1. The content of ^{210}Pb in lichen samples collected North and South of the latitude of 60° N during 1955-69. The triangles represent the average of all published data North of 60° N latitude and the filled squares represent the results of this work. The circles represent the results from South of 60° N latitude. Details about the previously published data are given in Table 1.

^{210}Pb in lichen

Lichen is a unique combination of an alga and a fungus which covers a great deal of the ground. Species of the Cladonia group which are common in Arctic and Sub-Arctic areas have been found to retain radionuclides from air and rain-water. From the standpoint of radiological health, this is of special interest in view of the food-chain lichen–reindeer–man (Svensson & Lidén, 1965).

The ^{210}Pb-contents of various lichen species collected on Svalbard and in Poland have been reported by Jaworowski (1966). He found a maximum content of ^{210}Pb during 1958–59 and he suggested that this could have been caused by attachment of naturally present ^{210}Pb to the particulate injected into the stratosphere by nuclear explosions. Unfortunately, there has been no other systematic investigation of the ^{210}Pb-level in lichen during the entire decade of 1960–1970. The few data that are available from different countries have been summarized in Table 1. Ramzaev et al. (1969) and Blanchard & Moore (1970) recently reported some measurements of the ^{210}Pb-content in lichen from the period prior to 1945 before any nuclear-weapons tests had been performed. All other published data are from the period after 1945 and they do not indicate any obvious increase in the ^{210}Pb-level. Some results of the ^{210}Pb-content of *Cladonia alpestris* collected in Finland during 1964–1967 are given by Kauranen & Miettinen (1969), and recently published data from the period 1966–68 in the USSR have been presented by Ramzaev et al. (1969). Some other results from samples collected in Alaska, the USA, Canada, Northern Finland and Britain have been published by different authors: Blanchard (1967), Hill (1965) and Holtzman (1966). The average ^{210}Pb-levels in lichen collected both North and South of 60° N latitude have been determined on the basis of the published data which are summarized in Table 1, and which are shown in Fig. 1. Owing to the great geographical, seasonal and biological spread of the samples, the data available are not satisfactory. However, they are assumed to correspond to the magnitude of the ^{210}Pb-content in the lichen.

The lichen-samples for radionuclide studies in Northern Sweden (62.3° N, 12.4° E) have been collected since 1962 by Lidén and his

Table 1. *Summary of published data on the content of ^{210}Pb and ^{210}Po in lichen collected all over the North Hemisphere*

The lichen species are given in abbreviations: Cl, *Cladonia;* a, *alpestris;* ce, *cenotea;* m, *mitis;* r, *rangiferina;* s, *silvatica;* su, *subulata;* u, *uncialis;* Ce, *Cetraria;* d, *delisei;* i, *islandica;* ri, *richardsonii;* Ca. e, *Caloplaca elegans;* Sp. g, *Sphaerophorus globosus*

Year	Place	^{210}Pb nCi/kg d.wt	$^{210}Po/^{210}Pb$	No. of samples and species	Ref.
Pre-1900	USSR	15	—	1 ?	(1)
1900–1945	USSR	21 ±8	—	3 ?	(1)
1923	Alaska	5.8±1.3	—	3 Cl, su, u, ce	(7)
1948	Alaska	15 ±3	—	2 Ce, i, ri	(7)
1949	Alaska	11 ±1	—	2 Ce, d+Sp.g	(7)
1955	Poland	1.0	—	1 Cl, m	(2)
1956	—	—	—	—	
1957	Svalbard	1.9±0.5	—	2 Cl, m+r	(2)
	Poland	1.7	—	1 Cl, s	(2)
1958	Svalbard	8.0	—	1 Cl, m	(2)
	Poland	2.9±0.5	—	3 Cl, a, m, s	(2)
1959	Poland	3.8±0.7	—	2 Cl, m, s	(2)
1960	Poland	1.9	—	1 Cl, s	(2)
1961	Poland	1.5	—	1 Cl, s	(2)
	Alaska	11.5	—	1 Cl, s	(3)
	Finland	4.5	—	1 Cl, a	(3)
	Finland	8.3	—	1 Cl, a	(4)
1962	—	—	—	—	
1963	Poland	4.7±2.0	—	2 Cl, a, m	(2)
1964	Alaska	4.6±1.1	0.96±0.07	2 ?	(5)
	Denmark	4.3	0.9	1 ?	(5)
	Ohio, USA	6.3±0.7	0.88	2 ?	(5)
	Canada	3.5	1.0	1 Cl, a	(6)
	Finland N.	7.4±0.9	1.0	3 Cl, a	(6)
	Britain	8.9±1.1	1.0	2 Ca, e	(6)
	Poland	3.2	—	1 Cl, a	(2)
	Finland	7.4	0.86±0.06	2 Cl, a	(4)
1965	Alaska	7.6	0.96	1 Cl	(3)
	Finland	6.0	0.92	2 Cl, a	(4)
	Poland	1.7	—	1 Cl, s	(2)
	Svalbard	10.0	—	3 Cl, m	(2)
1966	Finland N.	7.9±2.3	0.96±0.03	2 Cl, a	(4)
	Finland S.	6.0±0.6	0.85±0.03	3 Cl, a	(4)
	USSR	10.9	0.86	1 Cl	(1)
1967	USSR	7.4±1.3	0.86±0.06	4 Cl	(1)
1968	USSR	7.0±1.2	—	4 Cl	(1)

Ref.:
(1) Ramzaev et al. (1969)
(2) Jaworowski (1966)
(3) Holtzman (1966)
(4) Kauranen & Miettinen (1969)
(5) Blanchard (1967)
(6) Hill (1965)
(7) Blanchard & Moore (1970)

collaborators using a well standardized technique. Svensson & Lidén (1965), Lidén & Gustafsson (1967), Lidén (1969) and Lidén & Mattsson (1970) These samples are thus suited for making a reliable investigation of the ^{210}Pb-level during the last decade. The species used was *Cladonia alpestris* and each sample was collected in an area of 0.25 m², marked by a frame, except in 1961 when the sample was collected in an area of 4 m².

Analysis and measurements

The most widely used technique for determination of low levels of ^{210}Pb and ^{210}Po in environmental materials is spontaneous deposition of ^{210}Po on silver or nickel discs from slightly acid solutions. The method described by Flynn (1968) was found to be the most suitable one for analysing ^{210}Pb and ^{210}Po in lichen because of quantitative rapid deposition with no interference from the iron or other ions present. The lichen-samples were dried at 105°C for 12 hours and then pulverized and mixed into a homogeneous powder. A suitable quantity (10 g) was wet-washed by using concentrated nitric acid and perchloric acid in a Kjelldahl apparatus. The dissolved sample was then transferred to a 400 ml beaker and the interference of iron (III), chromium (VI) and other oxidants was eliminated by adding 1.0 g of hydroxylaminehydrochloride. The deposition of tellurium was avoided by adding 0.5 g of sodiumcitrate as a complexing agent. Addition of 10 mg bismuth-carrier prevented ^{212}Bi, decay-product of ^{232}Th, from depositing. The pH-value was adjusted to 2.0 with concentrated ammonia-solution and after dilution to 50 ml, the beaker was placed on a magnetic stirrer and heated to 85–90°C for 2–3 min to reduce any iron (III) or other oxidants present. A plastic holder with a mounted silver disc was placed in the beaker for 2 hours at 85–90°C. The disc was then removed and washed with demineralized water and methanol. ^{210}Pb was deposited on an area of the silver disc having a diameter of 22 mm. The radiochemical yield of the whole procedure was determined as 98 ± 2% by using samples with known content of ^{210}Po.

The ^{210}Po-activity of the sample was then measured with a silicon-surface-barrier detector with an area of sensitivity of 4.5 cm^2 (⌀ 24 mm) which was connected to a multi-channel analyser. The fractional width at half maximum "FWHM" of the 5.30 MeV α-peak was 85 keV in the evacuated measuring-chamber. The counting efficiency was determined by calibrated sources of ^{239}Pu and ^{241}Am electrodeposited on silver discs as above. The efficiency thus determined was 0.38 counts per disintegration.

Results and discussion

The concentration of ^{210}Pb in the lichen-samples was determined by ^{210}Po-analyses several years after they had been collected. During this period the ^{210}Po present at the time of sampling had almost completely decayed, while fresh ^{210}Po had reached radioactive equilibrium with the ^{210}Pb present. All samples were analysed twice and the standard error of the mean was less than ± 2%. There is, however, a systematic error for ^{210}Pb inherent in the sampling procedure, arising from the fact that the lichen also contains ^{226}Ra and its short-lived decay-products, which decay to form more ^{210}Pb in the samples after collection. Because ^{210}Pb takes some 80 years to reach radioactive equilibrium with ^{226}Ra, and that the ^{226}Ra/^{210}Pb activity-ratio in lichen is only about 0.01–0.08 according to Ramzaev et al. (1969) and Blanchard & Moore (1970), the error introduced by the presence of ^{226}Ra is negligible in this investigation. Measurements of the dried lichen-samples with a high-resolution γ-spectrometer also show a negligible presence of ^{226}Ra and its short-lived daughter products in comparison to ^{210}Pb, which was analysed by means of the 46.5 keV photo-peak (Mattsson, 1969).

The ^{210}Pb-content in lichen-carpets collected from 1961–1969 in the vicinity of Lake Rogen (62.3° N, 12.4° E) in Northern Sweden are given in Fig. 1 together with the summarized data from other countries. There is a surprisingly good agreement between the data from Northern Sweden and other data North of latitude 60° N. These values indicate a constant level during the last decade. The low value during 1969 could possibly be due to insufficient ingrowth of ^{210}Po, but the ^{210}Po/^{210}Pb activity-ratio at the time of sampling has, however, been reported by Kauranen & Miettinen (1969) and Ramzaev et al., to be about 0.8–1.0, which indicates an almost established radioactive equilibrium. Any significant increase of the ^{210}Pb-content in lichen for any certain year which would indicate a high artificial release of ^{210}Pb, is not indicated.

The area-content of ^{210}Pb in lichen collected from 1961 until 1969 in Northern Sweden are given in Fig. 2. The slope of the dotted line in Fig. 2, which is adapted to the experimental values, depends strongly upon the extreme

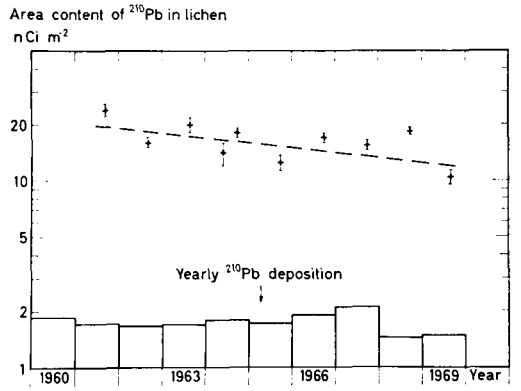

Fig. 2. The area-content of ^{210}Pb in lichen collected in Northern Sweden from 1961 until 1969. The line is deduced from the experimental values using the method of least squares. The uncertainty indicated is 1 S.E. The histogram shows the yearly deposition of ^{210}Pb from the atmosphere as calculated from precipitation data.

values in 1961 and 1969. Because of the non-standardized sampling technique used in 1961, the value for that year is not quite comparable with the other values. The extremely low value in 1969 can possibly be explained by the unusually low amount of precipitation during 1968 and 1969. Taking this into consideration, the area-content of ^{210}Pb in lichen was fairly constant during the period 1962–68 with an average of 16 ± 2 nCi/m^2. The yearly amount of precipitation at the place where the lichen were collected has been extrapolated from the precipitation measured at two meteorological stations, Myskelåsen och Tännäs, in the vicinity of Lake Rogen. The data for these localities were kindly supplied by the Swedish Meteorological and Hydrological Institute. The average radioactivity concentration of ^{210}Pb in rainwater at middle latitudes in the Northern Hemisphere has been estimated to be 2.5 ± 0.5 pCi/l according to the data given by Burton & Stewart (1960) and Jacobi (1963). As the ^{210}Pb in the atmosphere is deposited with 90% by precipitation according to Jacobi (1963), the yearly deposition of ^{210}Pb was estimated from these data and is given as histogram in Fig. 2. Consequently, the mean value of ^{210}Pb deposition-rate during the period 1960–69 was estimated to be 1.7 ± 0.3 nCi/m^2a. Thus under normal conditions, with the equilibrium value of 16 ± 2 nCi/m^2, the long-term elimination of ^{210}Pb from the lichen-carpet corresponds to an effective half-time of 7 ± 2 *a*.

The geographical distribution of ^{210}Pb in lichen collected under the 20th meridian has been studied by Jaworowski (1966). His results are seen in Fig. 3 together with the results of this study and those from North and South Finland published by Kauranen & Miettinen (1969). The values North of latitude 60° N seem to successively increase up to Svalbard. This distribution is, however, in disagreement with the latitudinal profiles of average ^{210}Pb-concentrations in the ground-level air, which were obtained during 1960 and 1961 by Patterson & Lockhart (1964), who found a distinct maximum at latitude 40° N. Their profile has been normalized to the value in lichen at latitude 40° N and is indicated in Fig. 3 by a dotted line. There it can be seen that the values from Poland agreed well with this distribution, but the value from Svalbard is nearly an order of magnitude higher. This could possibly be explained by a faster elimination of ^{210}Pb in lichens at lower latitudes.

The distribution and transport of ^{210}Pb within the lichen-carpet has been studied after the samples (0.25 m^2) were divided into different layers according to Fig. 4. The top-layer (*A*) is the fresh part which is eaten by reindeer.

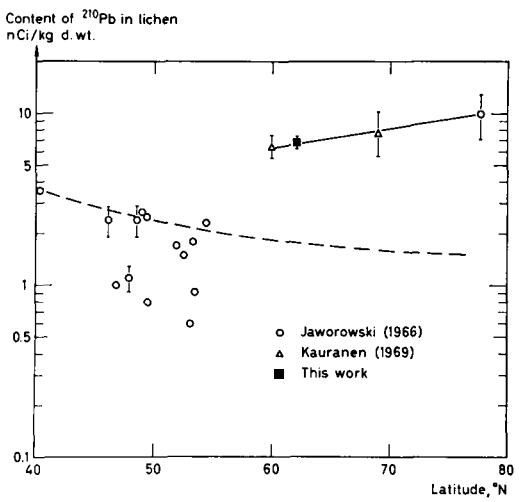

Fig. 3. The geographical distribution of the ^{210}Pb content in lichens during the year 1965 along the 20th East Meridian according to Jaworowski (1966) with data from Finland, published by Kauranen & Miettinen (1969), and this work included.

Table 2. *^{210}Pb-content of different layers in the lichen carpet according to Fig. 4 expressed partly as nCi/kg dry weight and partly as nCi/m²*

Part of the lichen carpet	^{210}Pb content 1967	1968	1969
	nCi/kg d.wt		
A 3 cm	7.16 ± 0.06	9.7 ± 1.1	4.62 ± 0.15
B 3 cm	4.96 ± 0.37	7.5 ± 0.8	2.52 ± 0.22
C 6 cm	4.00 ± 0.25	5.7 ± 0.4	1.72 ± 0.36
Litter & leaves	38.4 ± 1.7	—	18.0 ± 4.0
Jelly 2 cm	—	7.6 ± 0.4	—
Soil 3 cm	—	4.4 ± 0.1	—
Roots from other vegetation	—	2.23 ± 0.04	—
	nCi/m²		
A 3 cm	4.54 ± 0.15	5.3 ± 0.3	3.20 ± 0.05
B 3 cm	2.92 ± 0.07	3.5 ± 0.2	1.43 ± 0.16
C 6 cm	4.19 ± 0.05	5.4 ± 0.2	1.43 ± 0.40
Litter & leaves	3.0 ± 0.7	3.0 ± 1.0	4.4 ± 0.2
Jelly 2 cm	—	8.7 ± 0.3	—
Soil 3 cm	—	9.4 ± 1.8	—
Roots from other vegetation	—	0.31 ± 0.01	—

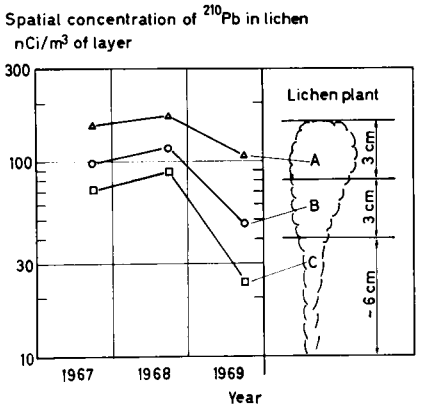

Fig. 4. The vertical distribution of ^{210}Pb in the lichen-carpet divided as shown by the picture to the right in the figure. The average spatial concentration of ^{210}Pb in the different layers is given for each of the years 1967–69. The experimental points in the figure represent the mean value of two different samples. The spread is less than is indicated by the dots.

The other layers (*B* and *C*) consist of older parts of the plant. The distribution of the ^{210}Pb-content between the different layers of the lichen-carpet, "jelly" (decomposed plant material) and soil are given in Table 2. From this table it is seen that there is a small fraction of litter and leaves in the lichen-carpet which has a high content of ^{210}Pb. This fraction introduces an element of uncertainty in the results from unfractionated samples which are contaminated with an unknown fraction of litter and leaves. In this investigation, however, this fraction is rather constant and therefore without any greater significance. The uncertainty introduced by the contamination of the samples with soil and "jelly" is, however, negligible because of their low ^{210}Pb-content. The vertical distribution of ^{210}Pb in the lichen-carpet is given in Fig. 4 as the ^{210}Pb-activity per unit volume (nCi/m³) of lichen-carpet layer. This distribution was very similar in 1967 and 1968, but the total ^{210}Pb-area-content was slightly higher in 1968. The distribution recorded in 1969, however, is somewhat different and the total area-content was lower, probably due to the unusually low amount of precipitation during both 1968 and 1969.

Conclusion

The content of ^{210}Pb in lichen collected in Northern Sweden (62.3° N, 12.4° E) was rather constant during the period 1961–69. No maximum was found in 1963–1965 similar to those

previously found for the artificial fallout radionuclides ^{137}Cs, ^{90}Sr and ^{55}Fe. (Lidén & Gustafsson, 1967; Persson, 1969). Thus there was no special evidence of significant amounts of ^{210}Pb released by nuclear-weapons tests during 1961–62. The ^{210}Pb-content in lichen-carpets from Northern Sweden was found to be 7 ± 1 nCi/kg d.wt during 1961–68 which is in good agreement with the values 8 ± 1 nCi/kg d.wt found by Ramzaev et al. (1969) in the USSR during 1967–1968 and 7 ± 1 nCi/kg d.wt found by Kauranen & Miettinen (1969) in Finland during 1961–66. The average of the ^{210}Pb-area-content in lichen during the period 1962–68 was found to be 16 ± 2 nCi/m^2. The normal deposition-rate of ^{210}Pb was estimated to be 1.7 ± 0.3 nCi/m^2a. Under normal conditions the effective half-time for long-term elimination of ^{210}Pb from the lichen-carpet was consequently 7 ± 2 *a*. The vertical distribution of ^{210}Pb in the lichen carpet shows a maximum concentration in the top layer. The distributions were similar during 1967 and 1968, which indicates normal conditions disturbed in 1969 probably by the unusually low amount of precipitation during 1968 and 1969.

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ОТЛОЖЕНИЯ СВИНЦА-210 В ЛИШАЙНИКАХ СЕВЕРНОЙ ШВЕЦИИ В ТЕЧЕНИИ 1961–1969 ГОДОВ

Изучается содержание ^{210}Pb в лишайниках, собранных в северной Швеции (62,3° с. ш., 12,4° в. д.) в период с 1961 по 1969 гг. В качестве аналитического метода использовалось влажное сжигание с азотной и хлорной кислотами с одновременным отложением ^{210}Po на серебряные диски. Измерялась α-активность ^{210}Po с помощью порогового детектора с силиконовым покрытием. Никакого максимума содержания ^{210}Pb не было найдено в лишайниках для периода 1963–1965 гг., который мог бы быть сравним с максимумом, найденным ранее для ^{137}Cs, ^{55}Fe и ^{90}Sr, которые были введены в атмосферу во время испытаний ядерного оружия в период 1961–1962 гг. Таким образом, нет специальных указаний на ввод в атмосферу значительных количеств искусственного ^{210}Pb во время этих испытаний. Средняя концентрация ^{210}Pb в лишайниках для периода 1962–1968 гг. была найдена равной 16 ± 2 нанокюри/m^2, а скорость его осаждения оценивается как $1{,}7 \pm 0{,}3$ нанокюри/m^2 в год. Эффективный полупериод выведения ^{210}Pb из лишайников оценивается как 7 ± 2 года. Изучалось также вертикальное распределение ^{210}Pb в лишайниках, и наибольшие концентрации были найдены в верхних слоях.