

Vertical distributions of beryllium-7 and lead-210 in the tropospheric and lower stratospheric air

Ludwika Kownacka

Abstract Vertical distributions of ^7Be and ^{210}Pb were observed in the troposphere and lower stratosphere over Poland at several altitudes between 0 and 15 km in the period 1987–1998. These two natural radionuclides are formed from gases in the atmosphere by nuclear processes. They are associated with similar size distributions aerosol particles and are removed from the atmosphere with similar efficiencies by scavenging processes of washout and deposition, however, they have various types of sources in the atmosphere. Vertical profiles of ^7Be (cosmogenic origin), characterized by small concentrations near the ground level, increase in the tropospheric levels and at 15 km altitude the concentrations are the highest. The highest concentrations of ^{210}Pb (terrestrial origin) are observed near the ground level, the lowest are below the tropopause, and in the stratosphere the concentration increases slightly. The dependence of the vertical distributions of both radionuclides in the situation of various levels of tropopause are discussed in this work.

Key words atmospheric radioactivity • ^7Be in the atmosphere • ^{210}Pb in the atmosphere • stratosphere • transport in the atmosphere • troposphere

Introduction

Natural radionuclides in the environment are of two general classes, the cosmogenic and the terrestrial. The cosmogenic radionuclides are produced mainly in the nuclear reactions through interaction of the cosmic rays with the air atoms in the atmosphere. The main terrestrial radionuclides are the elements of the two radioactive series headed by the ^{238}U , ^{232}Th , and ^{40}K nuclides, which exist in the earth's crust throughout its history.

The natural radionuclides ^7Be (cosmogenic) and ^{210}Pb (terrestrial) are formed from gases in the atmosphere by nuclear processes and are rapidly associated with the submicron aerosols. The radioactive species are in such microscopic amounts that they do not affect the physical and chemical properties of the aerosols [4]. These two radionuclides have various sources. The source of ^7Be (half-life: 53 days) is a cosmic ray which induces spallation reactions of atmospheric nitrogen and oxygen; about 75% of ^7Be are produced in the stratosphere and the rest in the troposphere. The maximum production of ^7Be occurs near 15 km above the ground [1]. The main source of ^{210}Pb (half-life: 22 years) is the radioactive decay of ^{222}Rn (half-life: 3.8 days) emitted to the atmosphere from the earth's crust; the other artificial sources (burning of coal, use of phosphate fertilizers, cars engine exhaust, fires) in the air are negligible [9, 10].

The atmospheric ^7Be and ^{210}Pb are carried by aerosols. The size distributions of the carrier aerosols for these two isotopes are similar, the (average activity) median aerodynamic diameters measured near the ground level using the

L. Kownacka
Department of Radiation Hygiene,
Central Laboratory for Radiological Protection,
7 Konwaliowa Str., 03-194 Warsaw, Poland,
Tel.: +48 22/ 811 01 95, Fax.: +48 22/ 811 16 16,
e-mail: kownacka@clor.waw.pl

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high-volume cascade impactors were 0.35 μm and 0.37 μm for ^7Be and ^{210}Pb , respectively [3] and 90% or more are attached to the particles with diameter less than 1 μm [8]. These two isotopes are removed from the atmosphere with similar efficiencies by scavenging processes of washout and deposition. Both radionuclides can be treated as sensitive indicators of the transport of masses in the atmosphere and the exchange of the atmospheric masses between the troposphere and stratosphere.

In this paper the results of the measurements of ^7B and ^{210}Pb concentrations at several altitudes in the troposphere and lower stratosphere over Poland during the period from 1987 to 1998 are presented.

Sampling and analytical procedure

During the period of 12 year study between April 1987 and August 1998, aerosol samples have been collected from 5 to 7 levels up to 15 km altitude over north-eastern Poland (Table 1). The measurements of ^7Be and ^{210}Pb concentrations in the air were carried out in aerosol samples collected at the altitudes 1, 3, 6, 9, 12, 15 km and near the ground level.

During the study we collected 185 aerosol samples, which formed 27 vertical profiles of concentrations of radionuclides. The concentrations of ^{226}Ra , ^{90}Sr and radiocesium were also determined [14–18]. The samples of each series were normally collected in a relatively short period of about 8 h under stable weather conditions in a cloudless region of the sky. Thus, the effects of new air masses advection to the sampling region were greatly reduced. Sometimes we could not collect samples from all seven altitudes.

High altitude samplers were constructed inside fuel tanks of the MIG-type aircrafts. The samplers were equipped with the mechanism to control the opening and closing of the air inlet and the grid supported the filter [13]. Those for the sampling up to 12 km were suspended underneath the wing and the second for the sampling up to 15 km was situated underneath the fuselage. The active surface of the filter in the “wing” device is 1640 cm^2 [11] and those in “fuselage” is 1380 cm^2 . The samplers are powered and operated from the pilot cockpit.

The “wing” samplers were used last time in 1993, because this type of MIG aircrafts which lift the “wing” devices went out of use. In the following years we have had the opportunity to use as a platform only MIG-21 aircraft with the “fuselage” samplers in all sampling levels.

During horizontal flights samples were collected by means of the ram pressure on filter fastened to the grid inside the tanks. We used a chlorinated PVC Petrianov filter FPP-15-1.5, which is characterized by large mechanical strength, high efficiency and small aerodynamic drag for high flow velocities. This filter was tested at linear velocity from 0.25 to 4 m/s using monodisperse particles of diameter 0.32, 0.9 and 1.25 μm . The collection efficiency of the filter varied between 95.5 and 99.99% at the linear air speed between 1.5 and 4 m/s, which is typical for our high altitude sampling [19]. Other measurements indicate that the efficiency of this filter become improved for the particles of radii from

Table 1. Number of analyzed samples.

Year	Altitude (km)							Total
	Ground level	1	3	6	9	12	15	
1987	3	2	2	3	3	3	2	18
1988	3	3	3	3	3	3	3	21
1989	2	2	2	2	2	2	2	14
1990	3	3	3	3	3	3	3	21
1991	2	2	2	2	2	2	2	14
1992	4	4	4	4	2	4	4	26
1993	2	2	2	3	1	2	1	13
1994	1	1	1	1	–	1	1	6
1995	1	1	1	1	1	1	1	7
1996	1	1	1	1	1	1	1	7
1997	3	3	3	2	2	3	2	18
1998	3	3	3	2	3	3	3	20
Total	28	27	27	27	23	28	25	185

0.026 to 1.71 μm at various velocities up to 6 m/s, with decreasing temperature and pressure, i.e. at the conditions prevailing at the higher altitudes [22].

The samplers were calibrated in the wind tunnel of the Institute of Aviation in Warsaw. Assuming the standard atmospheric conditions, the amount of air passing through the filter in unit time was measured for each simulated altitude. A special computer program was used for calculations of air passing through the filter of various altitude level and plane speed. The volume of air samples ranged from about 100 m^3 STP at 15 km to about 3700 m^3 STP at 1 km (STP – standard temperature and pressure at the earth surface, i.e. 1013 hPa and 15°C). The error in air volume determination was 5–12% depending on the altitude.

The ground level sampler, described by Bilkiewicz *et al.* [2], was equipped with an air blower, a gas meter and a filter holder protected against the atmospheric influence. The active surface of the filter is 1250 cm^2 . The volume of the filtered air in one sampler was about 1500 m^3 .

Concentrations of ^7Be were determined by γ -spectrometry using an HPGe detector and a Canberra 90 analyser, from the area of the photopeak at 477.7 keV. Concentrations of ^{210}Pb were determined by radiochemical separation using electrodeposition of ^{210}Bi on a nickel disk. The activity of ^{210}Bi was counted immediately after spontaneous elec-

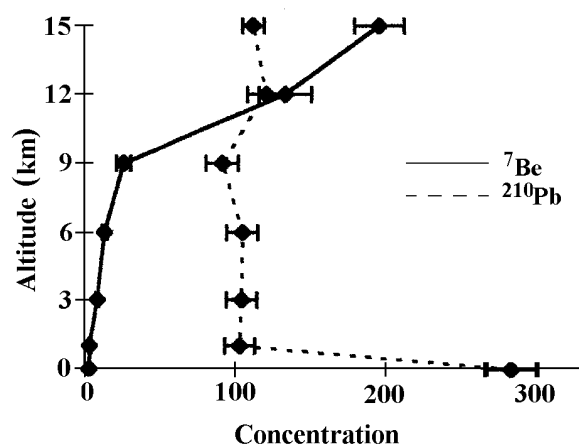


Fig. 1. Vertical distributions in the atmosphere of average concentrations of ^7Be (mBq/m^3 STP) and ^{210}Pb ($\mu\text{Bq}/\text{m}^3$ STP) and the standard errors of the mean between 1987 and 1998.

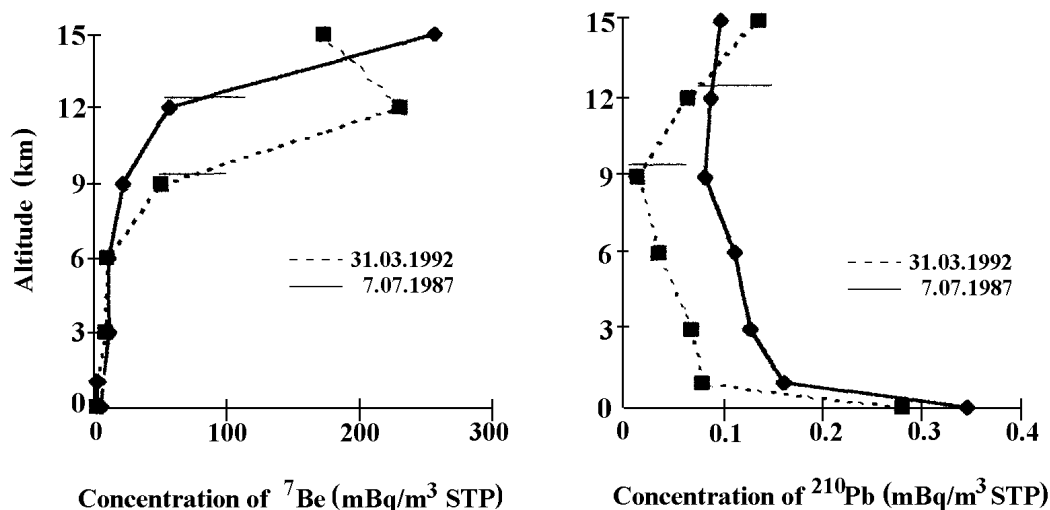


Fig. 2. Individual vertical distributions of atmospheric concentrations of ^7Be and ^{210}Pb .

trodeposition using a low-level anticoincidence beta GM counter. All ^7Be and ^{210}Pb activities were decay-corrected.

The tropopause height for the sampling region was determined for each sampling period on the basis of the vertical temperature distribution in the atmosphere received from the Institute of Meteorology and Water Management. The method recommended by the World Meteorological Organisation was used for the calculation.

Results and discussion

The average of vertical concentrations distribution of ^7Be and ^{210}Pb in the atmosphere between the ground level and 15 km for the period 1987–1998 shows typical patterns (Fig. 1). The mean concentrations of ^7Be rapidly decrease from the high values in the stratosphere to low values below the tropopause, with rather small changes in their tropospheric profile and the lowest concentrations near the ground. The concentrations of ^7Be in the troposphere and near the ground level show seasonal variations, which are connected with exchange of air between the stratosphere and the troposphere in situation of tropopause folding events [6]. The mean pattern of ^{210}Pb has a different shape: the highest concentrations are near the ground level. They tend to decrease with altitude in troposphere. The lowest concen-

trations are below the tropopause at 9 km, and in the stratosphere the concentrations are little higher. An increase of ^{210}Pb concentrations in the stratosphere were also observed in 12 vertical profiles over middle west of the United States [21]. In 11 situations the concentrations of ^{210}Pb at 15 km were lower than the lower limit of detection (17 mBq per sample), because the volume of air samples taken from 15 km (in STP) are rather small in comparison with the samples from other altitude levels.

The main source of ^{210}Pb in the atmosphere is a quiescent ascent of its gaseous ^{222}Rn parent from the ground and upward transport of subspecies mobilized by resuspension and motion with the atmospheric masses. In spite of the ground level sources, enough ^{222}Rn and its daughter isotopes enter the stratosphere creating a significant stratospheric ^{210}Pb reservoir above the tropopause [5, 7]. Other sources are the volcanic eruptions, which may introduce the ^{210}Pb to the stratospheric level [15, 20].

The individual vertical distributions of radionuclides in the atmosphere depend strongly on the short-term changes in weather conditions, and the short-term changes in tropopause height [11, 15]. Examples of two individual vertical profiles of ^7Be and ^{210}Pb are given in Fig. 2. Profiles on 7 July 1987 represent a type similar to the average long term profile in Fig. 1. Rather low concentration of ^7Be at the 12

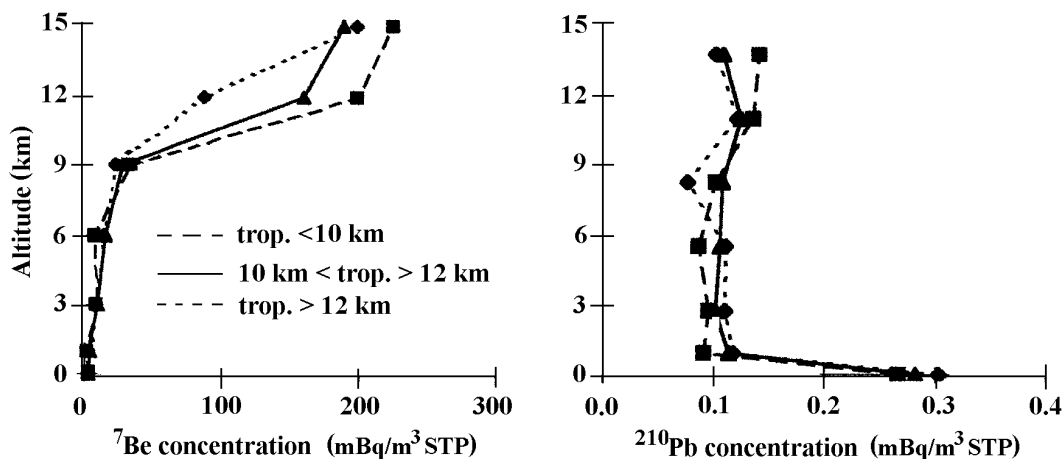


Fig. 3. Dependence of vertical distribution of ^7Be and ^{210}Pb (mBq/m³ STP) in the lower stratosphere and troposphere on the height of tropopause in the years 1987–1998.

km altitude in comparison with the concentration at the 15 km level is due to the high altitude level of tropopause in the sampling day (12.4 km). The low level of tropopause (9.5 km) in 31 March 1992 caused this atypical vertical pattern of ^{210}Pb . The lowest concentration was observed below the tropopause. In the stratosphere the concentration increased and at the 15 km altitude the concentration expressed in $\mu\text{Bq}/\text{m}^3$ STP was about 50% of our ground level value. When expressed in ambient air units ($\mu\text{Bq}/\text{m}^3$), the concentration at the 15 km altitude is several times lower than at the ground level, but it is higher than in the higher troposphere. This could have been connected with the Pinatubo volcano eruption (9 June 1991), which introduced the contaminations to the stratosphere up to the 24 km altitude. The increase of ^{210}Pb and other natural radionuclide concentrations in the stratosphere were also observed after the large volcanic eruptions in the Northern Hemisphere (Fuego, 1974; St. Helens, 1980; Ell Chicon, 1982; Nevado del Ruiz, 1985 and Pinatubo, 1991) [12].

Individual vertical distributions of radionuclides change their shape depending upon the tropopause height on the sampling day and the day before. This is also seen in vertical profile of mean concentrations of ^7Be and ^{210}Pb in three various ranges of the tropopause height observed on the day of the sampling, i.e. below 10 km, between 10 and 12 km, and above 12 km (Fig. 3). In our geographical region the tropopause is usually observed between 10 and 12 km. In most cases the samples from 12 km represent aerosols from the lowest part of the stratosphere. In the situations where the tropopause level is below 10 km (Fig. 3) the concentration of ^7Be at 12 km is very high, similar to the concentration measured at 15 km. If the tropopause is going up, the concentration of ^7Be at 12 km is decreasing, when the tropopause is higher than 12 km, the samples contain a mixture of tropospheric and stratospheric air. At all levels in the troposphere the concentrations are similar. At the situation of the lowest tropopause level (below 10 km) the lowest concentrations of ^{210}Pb near the ground level and in the middle troposphere were observed. If height of the tropopause is higher than 12 km, the concentrations near the ground and in the middle troposphere increased, but in the higher level of troposphere (at 9 km), the concentration went down. At the low altitude level in the stratosphere the concentration of ^{210}Pb grows up because the stratospheric reservoir is above the tropopause. Short-term changing of the tropopause height caused an increase in the exchange of atmospheric masses between the troposphere and stratosphere.

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