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Natural and artificial radionuclides in the tropospheric and lower stratospheric air over Poland

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Abstract The high altitude aircraft sampling of aerosols have been carried out at four to seven levels up to 15 km over Poland from 1973 to 1998, and 117 vertical concentration profiles of natural radionuclides and fission products were determined. It was found that the atmospheric concentrations of ^{226}Ra increased after large volcanic eruptions. The vertical concentrations profile of ^{226}Ra had a characteristic quasi-parabolic distribution, with the highest concentrations near the ground level and in the stratosphere. Concentrations of ^{210}Pb had a more homogeneous vertical distribution, due to the quiescent ascent of its gaseous ^{222}Rn precursor from the ground. Vertical concentrations of fission products revealed different types of profiles. After nuclear explosions, the highest concentrations were observed in the stratosphere. The Chernobyl accident reversed the aerial vertical profiles; the main source was near the ground level, but part of the radioactive debris entered also the lower stratosphere. Because of resuspension, the stratospheric residence time of radiocesium from the Chernobyl accident was about three times longer than that of the fallout from nuclear explosions. The resuspension effects and the atmospheric mass transport in the boundary layers are also responsible for the increased concentrations of radiocesium at 1 km altitude observed since 1987 in the situations of atmospheric mass advection from the highly contaminated Chernobyl region.

Key words atmospheric radioactivity • Chernobyl • nuclear tests • stratosphere • transport in the atmosphere • troposphere

Introduction

The vertical distributions of concentration of natural radionuclides and fission products have been studied since 1973 in the troposphere and stratosphere over the north-eastern part of Poland. The natural radionuclides (^{7}Be and the members of uranium and thorium chains) enter the atmosphere from two types of sources: 1) the ground level sources emitting continuously (dust from industry, burning of coal, automobiles, emanation of ^{222}Rn from the ground and resuspension from the land and sea surface) and 2) the stratospheric source of a cosmogenic radionuclide ^{7}Be and volcano eruptions which may introduce radionuclides into the stratosphere [4]. Between 1945 and 1980 vast amounts of fission products were introduced into the troposphere and stratosphere by nuclear explosions and in 1986 by the Chernobyl accident [5].

Methods

Atmospheric particles samples have been collected from 1, 3, 6, 9, 12 and 15 km on a polyvinyl-chloride Petrianov filter using specially constructed samplers suspended underneath the aircrafts, and a stationary device for sampling near the ground level. The samples for each profile were normally collected over a period of about 8 h, under stable weather conditions, from cloudless regions of the sky.

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Since 1973, 689 particulate samples have been collected from the tropospheric and lower stratospheric air; they formed 117 vertical profiles of radionuclides concentrations. The samples were analysed by γ-spectrometry using an HPGe detector and a multi-channel analyser Camberra 90. The concentrations of ^{90}Sr, ^{210}Pb and ^{226}Ra were determined by radiochemical methods [3]. The measured ^{134}Cs and ^{137}Cs activities on our air filter in most of the samples were low and, therefore, the radiochemical method was used for $^{137+134}$Cs determination. The volume of air samples ranged from about 100 m^3 STP at 15 km to about 3700 m^3 STP at 1 km altitude (STP-standard temperature and pressure at the earth surface, i.e. 760 mm Hg and 15°C).

Results and discussion

During the long time study, several natural and man-made events have occurred which have influenced the levels of radionuclides concentration at particulate altitudes in the atmosphere. The individual vertical distributions of radionuclides in the atmosphere were strongly dependent on the short-term changes in weather conditions and the tropopause height and also on the activity of their sources.

Natural radionuclides

The long-term mean and not typical vertical distributions of natural radionuclides in the atmosphere are presented in Fig. 1. The cosmogenic ^{7}Be produced in spallation reactions of cosmic rays with atmospheric nitrogen and oxygen has the main source (~80%) in the stratosphere and the maximum of concentration in our geographical region is normally at about 15 km altitude [1]. The vertical distributions of ^{7}Be showed a small concentration near the ground level, rather small changes in tropospheric air and greatly increased in the stratosphere. Changes of the concentrations observed in the higher troposphere and stratosphere are connected with the changes of tropopause level in and before sampling time. In the situation of lower tropopause level, the concentration at 15 km may be lower than at 12 km altitude.

The mean vertical profile of ^{226}Ra and ^{210}Pb concentrations tends to decrease with altitude in the troposphere, the highest concentrations being at ground level and the lowest below the tropopause at 9 km. In the stratospheric air, the concentrations of these two radionuclides increase again. These quasi parabolic vertical profiles are the results of combination effects of stratospheric and ground level sources and the transport of contaminations in the atmosphere. Increase of the radionuclide concentrations in stratospheric air is in agreement with other measurements, because in the lower stratosphere, upper the tropopause, there is a layer with higher aerosol concentration [2]; this was confirmed by balloons, lidar and satellite observations. On these three diagrams (Fig. 1) examples of mean and atypical vertical profiles, which were produced by a strong vertical transport of massy in the spatial meteorological situations (well-developed baric systems, atmospheric fronts, thunderstorms, folding and lapse in the tropopause and short-term changes of the tropopause level) [3], are shown. The very untypical vertical distribution of ^{210}Pb on 23 May 1990 was produced by the upward transport of atmospheric mass connected with the well-developed baric system and atmospheric front before and in the sampling time.

The average concentrations of ^{226}Ra were used for the estimation of temporal changes of ^{226}Ra content in a column of 1 m^2 of the air between the ground level and 15 km altitude (Fig. 2). All the volcano eruptions marked (Fig. 2) are situated in the North Hemisphere, in the tropical or polar zones, and the contamination are transported to our region about half a year. After the big volcanic eruptions of Fuego, St.Helens, El Chichon, Nevado del Ruiz and Pinatubo, the higher content of ^{226}Ra was observed over Poland for the column 0–12 and 0–15 km. In the high levels of the troposphere (6–12 km) it was not clearly observed, because the residence time in troposphere is very low.

Fission products

The other type of radionuclides measured for twenty six years in the troposphere and stratosphere were the fission products. In the first 8 years of our study, the fission products were introduced into the stratosphere and troposphere by Chinese and French nuclear weapon tests and later in 1986 by the Chernobyl accident. Between 1973 and 1980 nine Chinese nuclear explosions in Lop Nor were conducted in the atmosphere with the energy yield ranging from 0.02 to 4 megatons. Before the Chernobyl accident (1973–1985), the vertical distributions of fission products in the troposphere increased steadily with altitude and there were an abrupt increase in the stratosphere. All Chinese

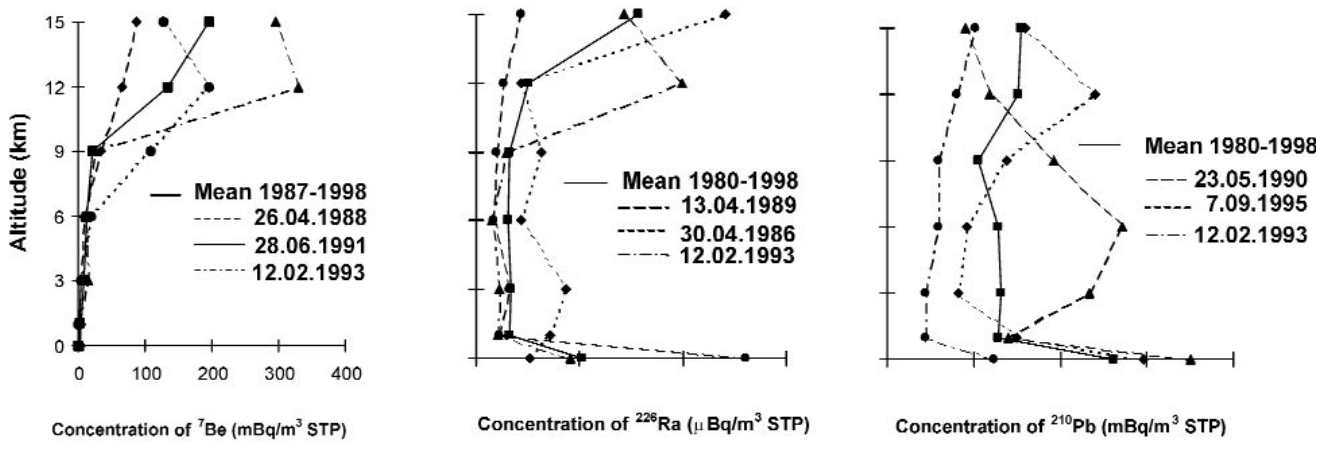

Fig. 1. Vertical concentration profiles of ^{7}Be, ^{226}Ra and ^{210}Pb in the atmosphere.

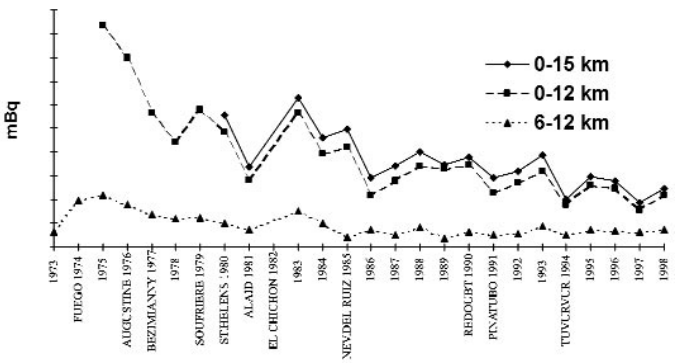

Fig. 2. Temporal changes of the content of ^{226}Ra in a 1 m^3 column of the air between the ground level and 15 km altitude.

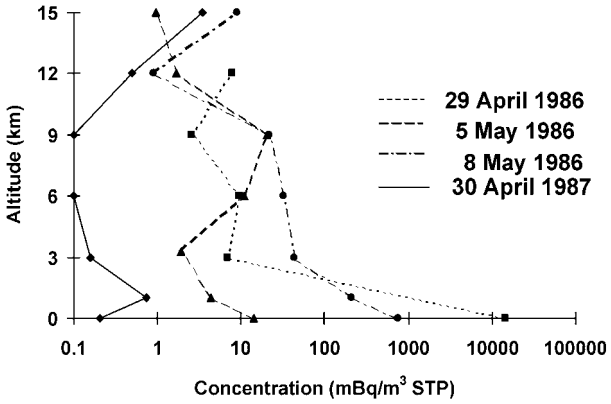

Fig. 3. Vertical concentration profiles of radiocesium after the Chernobyl accident.

explosions in the megaton range were easily detected over Poland. The decreasing of mean annual concentration of fission products in all levels in the atmosphere by the next years after the bigger nuclear explosions have been observed. After the last Chinese atmospheric nuclear explosion on 16 October, 1980, we observed first appearance of the radioactive cloud over Poland and by the next two years the descending of radionuclides from higher altitudes in the atmosphere to the lower ones [5].

The Chernobyl reactor accident (26 April, 1986) reversed the earlier vertical profile of fission products in the atmosphere; the highest concentrations were found near the ground level and the lowest ones in the stratosphere. But it was observed only for a short period of time because in the first eight days of May there was an upward transport in the troposphere and on 5 May the highest concentration of radiocesium was below the tropopause, and later we observed slightly increased concentrations in stratospheric air, indicating on upward transport through the tropopause (Fig. 3).

The high concentration of radiocesium near the ground level in Warsaw on 8 May was connected with the new release of radionuclides from the Chernobyl sarcophagus on 6–7 of May. Because of resuspension, the stratospheric residence time of radiocesium from the Chernobyl accident was about three times longer than that of the fallout from nuclear explosions (240 and 770 days, respectively) [5].

One year after the Chernobyl accident we observed an increase of radiocesium and ^{90}Sr concentrations at 1 km altitude in situations of mass advection from the east. This phenomenon was observed until 1998. The observed effect was explained, by a simple transport model, assuming that the source of this phenomenon is the resuspension of radionuclides from the highly contaminated by the Chernobyl fallout regions of Ukraine, Belarus and Russia, and that the transport of contaminations was in the atmospheric boundary layer [6].

In 1998, the high concentrations of radiocesium were observed in the troposphere (27 May at 3 km and on 20 August at 9 km altitudes). Gamma-spectrometry analysis indicated that only ^{137}Cs was present in the samples, other fission products were not detected. This high concentrations of ^{137}Cs were originated most probably from the radiological incident which occurred in Algeciras, (S-W) Spain, at the end of May in which sources of ^{137}Cs ($\sim 3\times10^{11}$ -3×10^{12} Bq) were melted in the Accerix steel factory [7]. It is to add that increases of ^{137}Cs in air near the ground level were observed in many countries.

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