P-4a-233

Long Term Observation of ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K in the Air Aerosol and ^{85}Kr in the Air in Prague, Czech Republic

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Basic Information about the Radiation Monitoring Network

Radiation Monitoring Network (RMN, www.suro.cz) of the Czech Republic has been established after the Chernobyl accident in 1986 and it has been developing systematically all the time. It is co-ordinated by the State Office for Nuclear Safety (SONS, www.sujb.cz), which acts as its Management in co-operation with the National Radiation Protection Institute (NRPI, www.suro.cz). The monitoring results are given in Annual Reports on Radiation Situation in the Czech Republic, which are submitted to the Governmental Emergency Commission on Radiation Accidents and to the public through District Administration bodies, health centres and libraries [4].

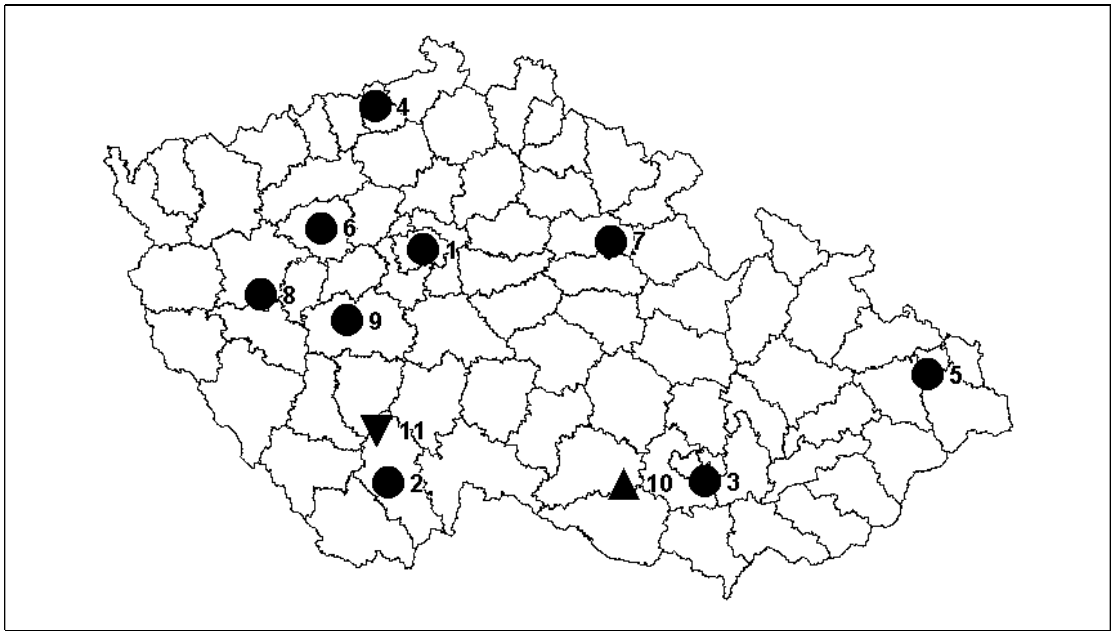
The RMN operates in two regimes: the normal regime, aimed at monitoring the current radiation situation and an early detection of radiation accidents, and the emergency regime aimed at evaluating the consequences of such a radiation accident. The normal regime is executed by permanent bodies engaged in the Network, the emergency regime includes, in addition, emergency bodies. The normal monitoring regime involves several subsystems, in which all permanent RMN bodies are engaged. The subsystems are as follows:

- Early warning network, which comprises 47 photon dose equivalent rate measuring points with automatic transmission of the observed data.
- Territorial network of 184 measuring points equipped with termoluminescent dosemeters (TLD) and local TLD networks with about 90 measuring points in the surroundings of the nuclear power plants Dukovany and Temelín.
- Network of 10 laboratories equipped with gamma-spectrometric and radiochemical analytical instrumentation.
- Territorial network of 23 measuring points of air contamination.

Measuring points of air contamination

The measuring points of the territorial network of air contamination (Fig.1) are equipped, besides other instruments, by aerosol samplers with throughput from 40 to 900 m^3/h. The aerosol filters, on which aerosol is continuously collected, are measured by semiconductor gamma-spectrometry. The aim of these measurements is not only to detect an abnormal radiation situation in co-operation with Early Warning Network but also to follow the long time course of the aerosol activity concentration. At present, the aerosol activity concentration of ^{137}Cs is so low that it can only be detected regularly using an aerosol sampler with high throughput and sensitive spectrometric equipment with low detection limit. Such equipment is at the NRPI in Prague (Fig.1, locality 1). The results of the aerosol activity concentrations of ^{137}Cs, ^{7}Be, ^{210}Pb, ^{40}K collected in NRPI and ^{85}Kr, sampled on the territory of the Department of Radiation Dosimetry of the Nuclear Physics Institute at Prague are presented in the paper. The measurement of ^{7}Be ^{210}Pb and ^{40}K can be used as a QA/QC check-up.

Fig.1: Aerosol sampling localities of the Radiation monitoring network in the Czech Republic



1. Prague
2. České budějovice
3. Brno
4. Ústí nad Labem
5. Ostrava
6. Rakovník
7. Hradec Králové
8. Plzeň
9. Kamená
10. NPP Dukovany (in operation, 6 measuring points in the surroundings)
11. NPP Temelín (in construction, 8 measuring points in the surroundings)

Sampling and measurement

The aerosols, the analysis of which is presented here, have been collected at NRPI in Prague (locality 1) on the aerosol filters by an aerosol sampler with flow rate at first 60 m^3/h (up to 1992), then with flow rate 200 m^3/h (1993 - 1996) and than with 900 m^3/h (1997- up to now). The contemporary aerosol sampler "Snow White" is shown in Fig.2. The filters are changed twice a week because of relatively high air pollution. The filters are measured without any previous treatment by semiconductor gamma-spectrometry (HPGe detector with relative efficiency 55%) in one week intervals (it means 2 filters combined) 3 days after the termination of the sampling in order to reduce contribution of the activity of the short-lived radon progeny. During the sampling a charcoal cartridge for measurement of the radioactive gaseous iodine is placed under the aerosol filter. The charcoal cartridge is changed every month. It is measured only in the case of aerosol iodine presence on the aerosol filter. The measuring time of the filter is about 250 000 s.

Besides an artificial radionuclide ^{137}Cs, coming from higher levels of atmosphere and from the resuspension of the original fallout on the ground surface, the activity concentration of the cosmogenic ^{7}Be, of the radon progeny ^{210}Pb and natural ^{40}K are evaluated by gamma-spectrometry routinely. The evaluation of these natural radionuclides helps to confirm the quality of sampling and measurements in individual laboratories. After the gamma-spectrometric analysis ^{238}Pu, $^{239+240}Pu$ and ^{90}Sr were determined in the quarterly combined filters by radiochemical methods.

The method of determination of the global ^{85}Kr activity concentration in the air originating from the tests of nuclear weapons in atmosphere and from nuclear fuel reprocessing plants is used at one site in Prague. It is based on separation of krypton from air by cryogenic adsorption on beds of active charcoal and radiometry of ^{85}Kr by $CaF_2(Eu)$ scintillation detector. Afterwards, the analysis of the separated sample for krypton element by gas chromatography aimed at the separation efficiency evaluation is performed.

Fig.2: Aerosol sampler “Snow White” with the flow rate of 900 m^3/h.

Results

The time course of the month averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentrations in NRPI in Prague in the time period from June 1986 to December 1999 is presented in Fig.3, together with the dust concentration. The time course of the month averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentrations recalculated on becquerel per gram of the aerosol is in the Fig.4.

Decrease of the activity concentration of ^{137}Cs in the end of the eighties and in the beginning of the nineties was, besides slow radioactive transformation, caused mainly by 2 reasons: the fallout of the aerosol particles from atmosphere and the migration of transferred ^{137}Cs in the soil causing lower activity of ^{137}Cs in the resuspended particles. Presently the activity concentration of ^{137}Cs in the air is about 1.0 μBq/m^3 and it is almost constant. This can be very well seen in the Fig.5 and 6, where there are the time courses of the year averages of the followed radionuclides.

Fig.3: The time course of the month averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentration (in Bq/m^3) and the dust concentration in the air (in g/m^3) in the air

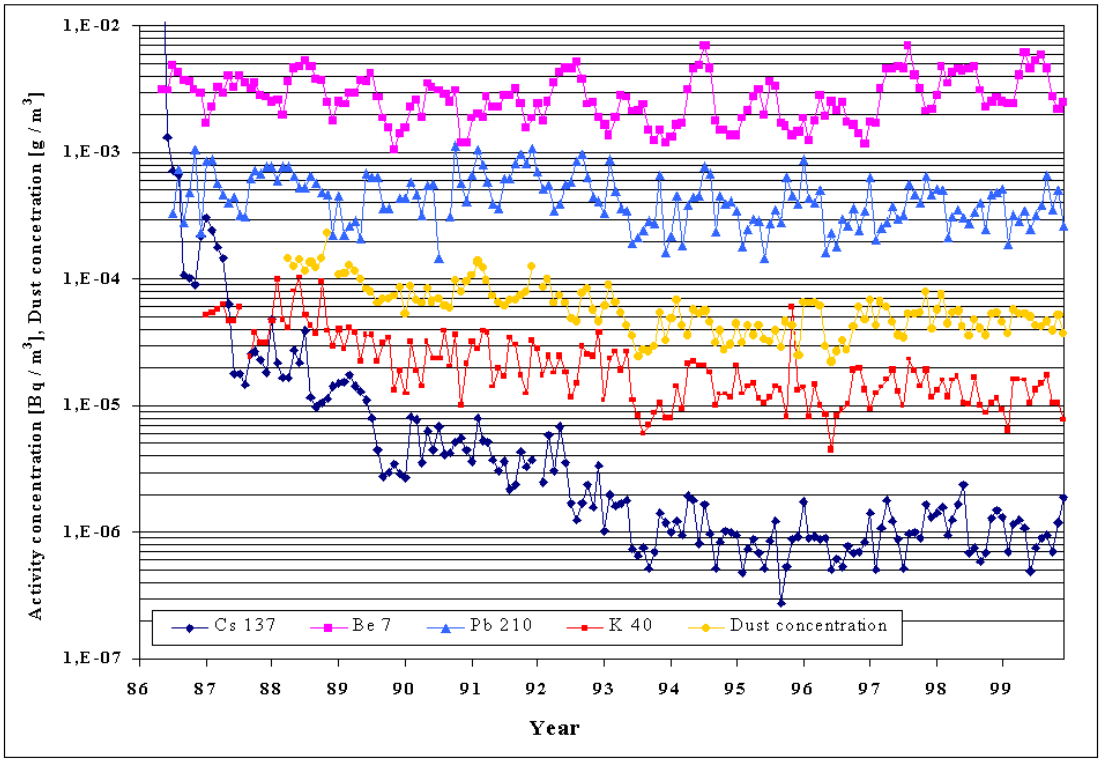


Fig.4: The time course of the month averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentration (in Bq/g)

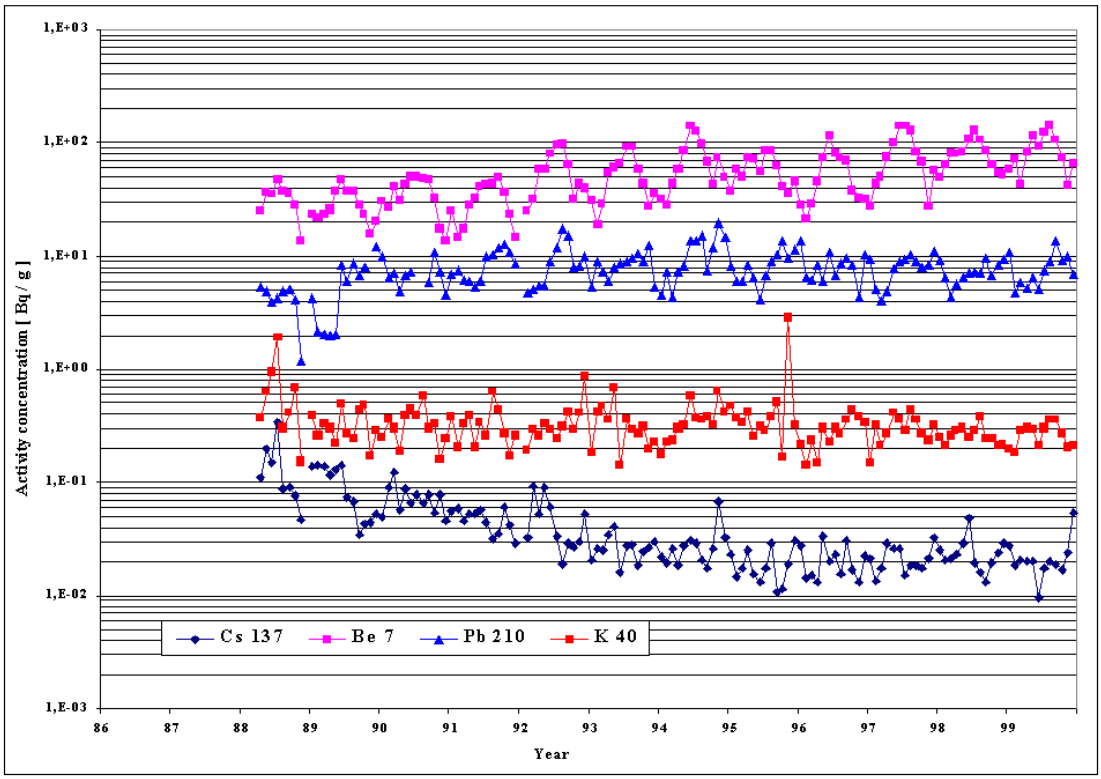
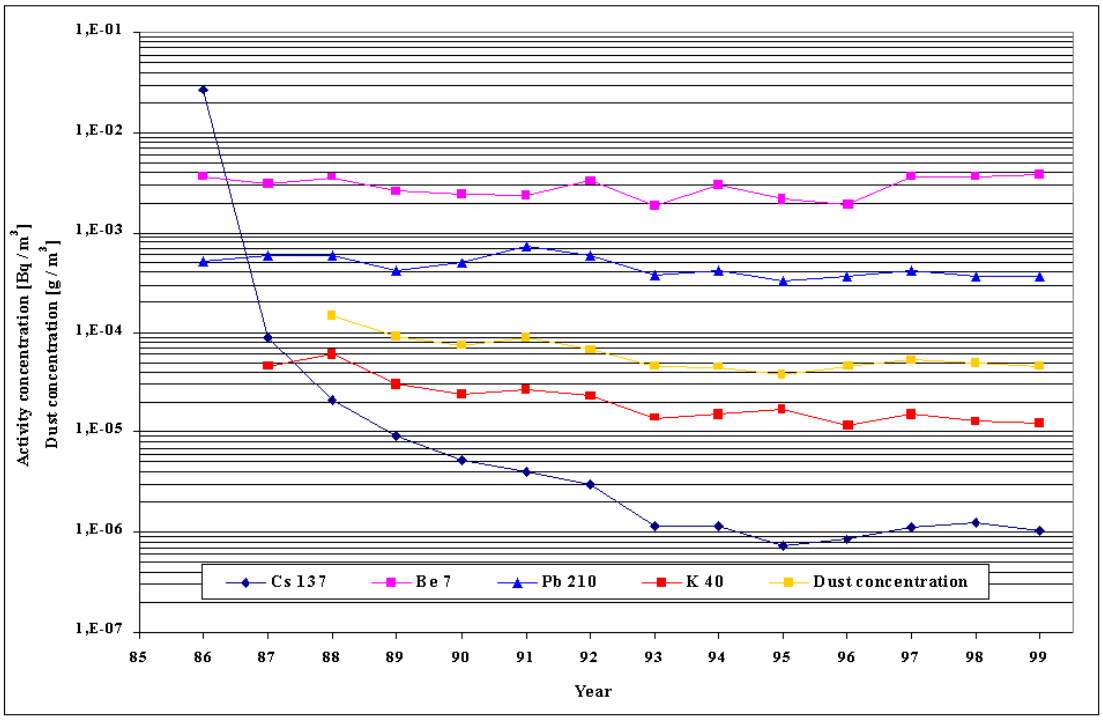
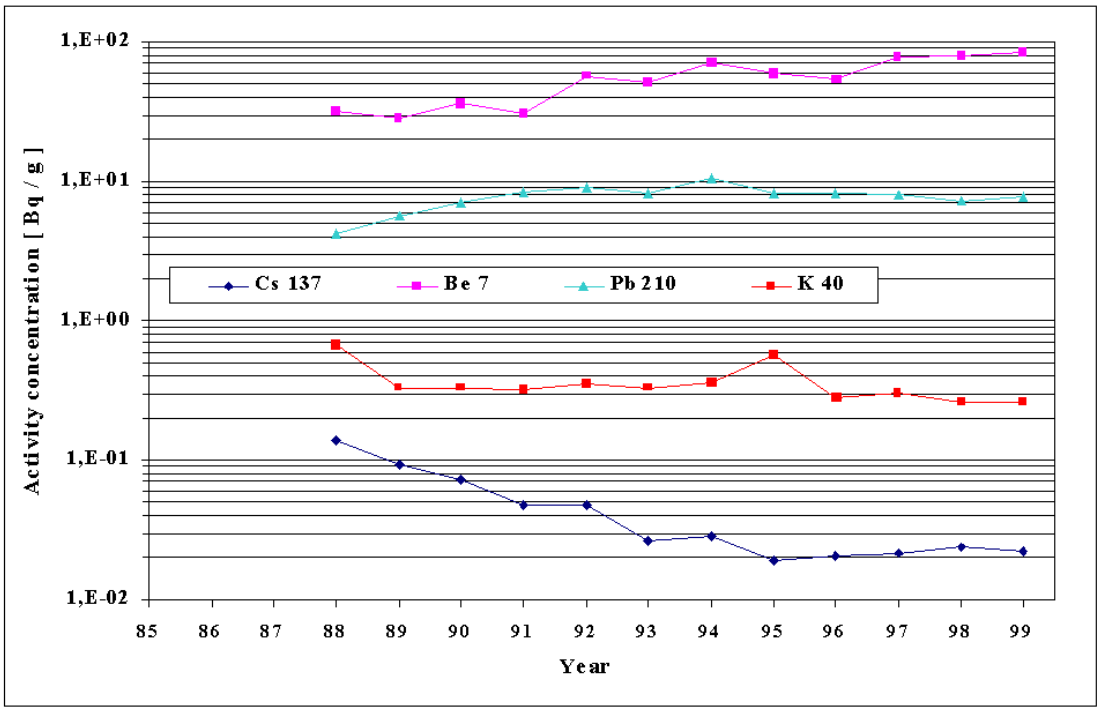


Fig.5: The time course of the year averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentration (in Bq/m^3) and the dust concentration (in g/m^3) in the air



Note: The average values for the year 1986 are calculated only from the data of the 2nd half of the year.

Fig.6: The time course of the year averages of the ^{137}Cs, ^{7}Be ^{210}Pb and ^{40}K activity concentration (in Bq/g)

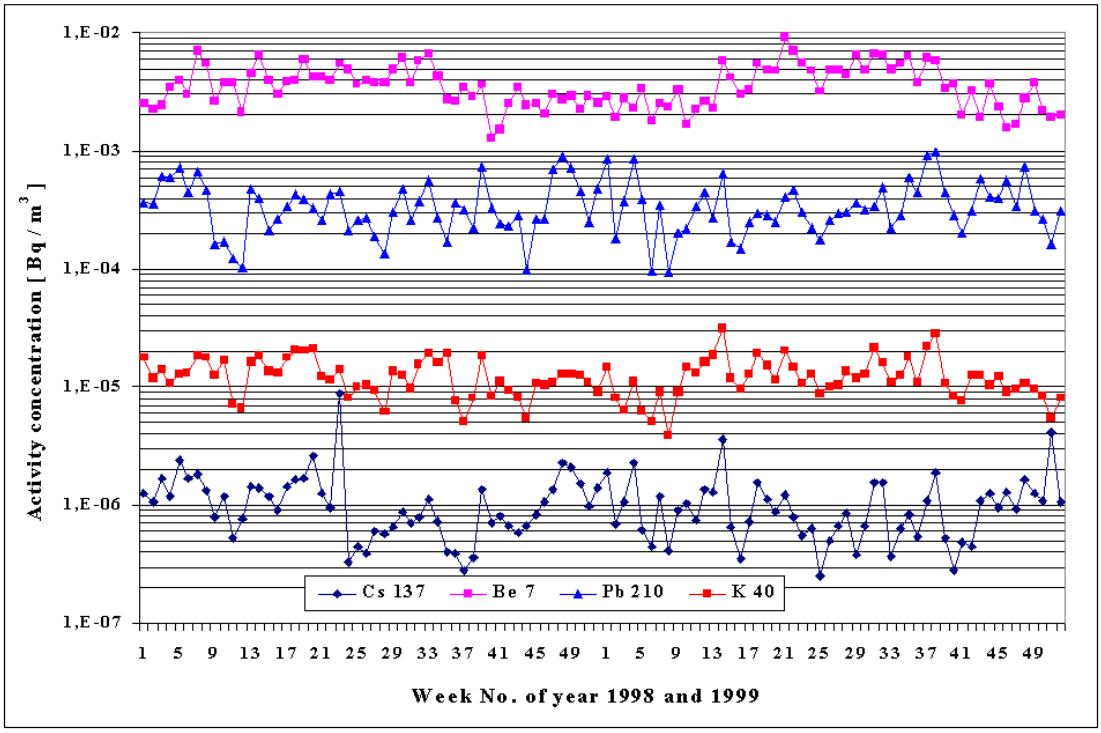


Berylium 7 shows typical seasonal variation with the maximum in summer due to thermal convection of the air masses. The mean value of activity concentration of ^{7}Be for the locality 1 from 1986 to 1999 is 2900 μBq/m^3. The activity concentration of ^{7}Be in Bq/g (Fig. 4, 6) slightly increases, in contrast to other radionuclides. The increase in the end of the eighties and in the beginning of the nineties is probably caused by the decrease of dust concentration at the sampling site. This effect overlays the influence of the solar cycle. (Solar activity has the 11 years cycle with the variation in maxima and minima). In the second half of the nineties it is possible to explain the increase of the activity concentration by the decreasing solar activity.

The mean value of ^{210}Pb activity concentration from 1986 to 1999 is 480 μBq/m^3. This value is in accordance with the value 500 μBq/m^3 given by UNSCEAR Report for the same geographical latitude. No seasonal variation of the activity concentration on the 0.01 significance level was observed. The slow decrease of the activity concentration in the beginning of the nineties is probably caused by the decrease of the observed dust concentration at the sampling site. The explanation of the behaviour of ^{40}K is similar to that for ^{210}Pb.

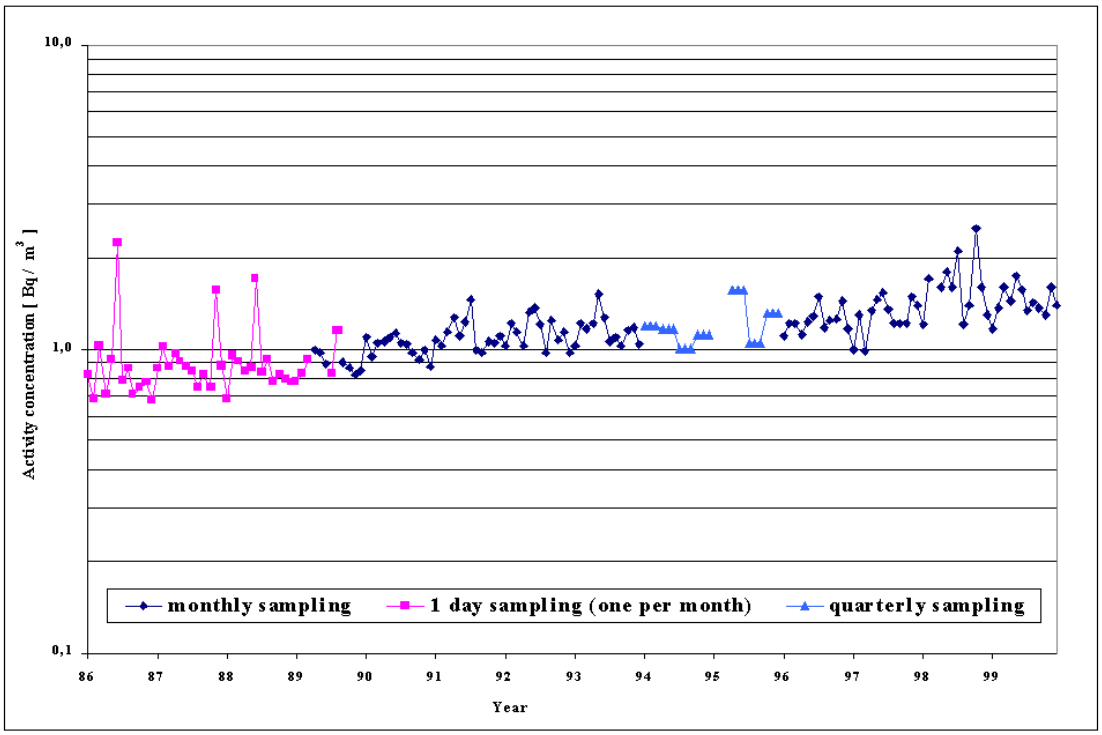
Fig.7 shows the time course of the followed-up radionuclides for the last 2 years at week intervals in order to illustrate the variations of the individual values. The increase of the ^{137}Cs activity concentration in the 23rd week of 1998 due to the melting up of a source in Algeciras, Spain can be clearly seen.

Fig.7: The time course of the week averages of the ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K activity concentration for the last 2 years



Monthly averages of activity concentration of ^{85}Kr are presented in Fig.8. They increase all the time and the actual value lies at about 1.4 Bq/m^3. The increase is smaller than the forecasts from the eighties, because of the slower development of the nuclear energy production and the smaller quantity of the reprocessed nuclear fuel than expected. The time courses of the average year activity concentrations of ^{85}Kr are presented in Fig.9.

Fig. 8: The time course of the month averages of the ^{85}Kr activity concentration



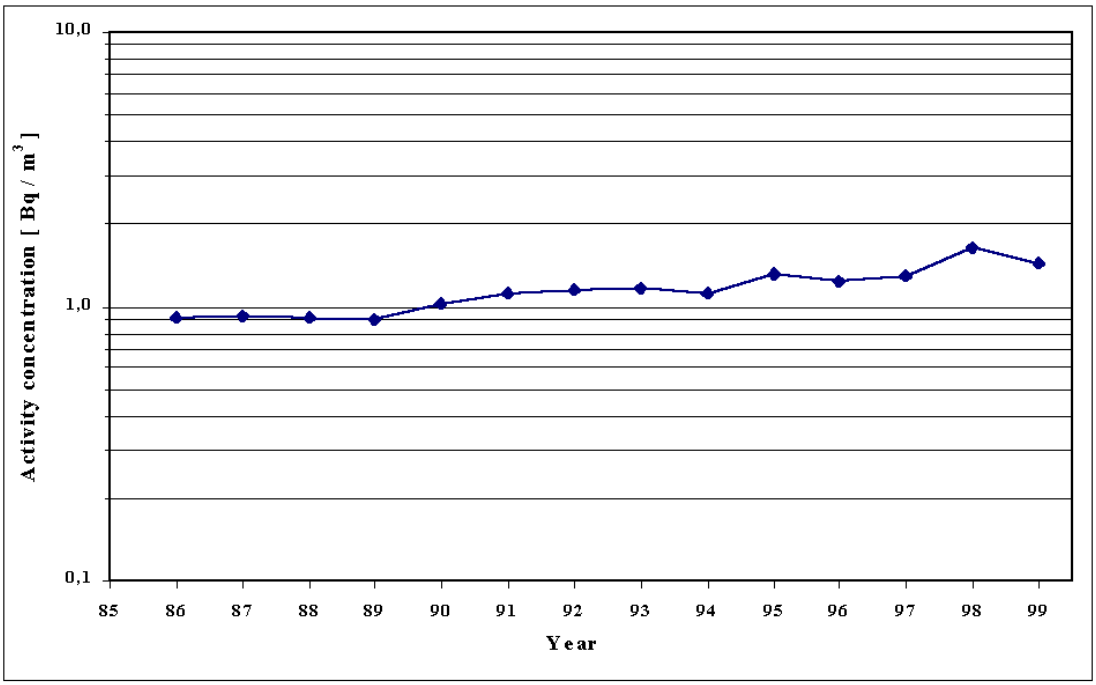


Fig. 9: The time course of the year averages of the ^{85}Kr activity concentration

The average values of activity concentration over the Czech Republic territory are not significantly different from the values acquired on the site in Prague. The estimation of the annual committed effective doses from inhalation of ^{137}Cs, ^{7}Be, ^{210}Pb and ^{85}Kr for an adult from population of Czech Republic for the year 1998 is in Tab.1.

Tab.1: The estimation of the annual committed effective doses from inhalation of ^{137}Cs, ^{7}Be, ^{210}Pb and ^{85}Kr for an adult from population, conversion coefficient for conversion of the dose to the unit of the inhaled activity [3] and the values of the mean activity concentrations in the Czech Republic territory in the year 1998.

Nuclide	Activity concentration	Conversion coefficient	The annual committed effective dose
	[Bq / m^3]	[Sv / Bq]	[Sv]
^{137}Cs	1.54 E-06	3.9 E-8	4.38 E-10
^{7}Be	2.91 E-03	5.5 E-11	1.17 E-09
^{210}Pb	4.86 E-04	5.6 E-6	1.98 E-05
^{85}Kr$^{*)}$	1.62 E+00	2.2 E-11	2.60 E-07

$^{*)}$ ^{85}Kr was measured at the location in Prague only.

The radionuclides ^{238}Pu, $^{239+240}$Pu and ^{90}Sr in the combined filters were determined quarterly. The activities were as a rule very low; in the year 1998 $^{239+240}$Pu in the range from 0.53 to 2.4 nBq/m^3, ^{90}Sr in the range from 70 to 150 nBq/m^3 and ^{238}Pu under the detection limit.

The aerosol particle size distribution

The estimation of the inhalation dose depends on the radionuclide aerosol particle size distribution. Therefore a pilot study of aerosol particle size distribution for individual radionuclides using a six stages cascade impactor was performed. Because of very low activities of ^{137}Cs, only ^{210}Pb and ^{7}Be aerosol particle size distribution was measured. The results i.e. dependence of the activity concentration of the radionuclides on the particle aerodynamic diameter (under the assumption of their logarithmic-normal distribution) were expressed by the use of 2 parameters: activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD).

It was found that the main part of ^{210}Pb and ^{7}Be is attached both in out-door and in-door air to the fine aerosol with aerodynamic diameter under 0.4 μm. Particle size distribution does not differ significantly from the aerosol particle size distribution of short lived radon progeny. No difference was found in aerosol particle size distribution for the out-door and in-door air. In most cases, the AMADs were in the range from 0.05 to 0.35 μm [5].

Conclusions

- The activity concentrations of the evaluated radionuclides are very small and resulting committed effective doses from inhalation of them are insignificant in comparison to committed effective doses from natural background, namely from inhalation of short lived radon progenies (2-3 mSv annually).
- The time behaviour of the ^{7}Be activity concentration can be related to 11 year solar cycle; when the solar activity is at its maximum, the activity concentration of ^{7}Be will be near the minimum.
- The expected slow increase of the ^{85}Kr activity concentration is in the agreement with prediction and it is caused by the development of the nuclear energy production.
- The activity concentrations of ^{137}Cs, ^{7}Be, ^{210}Pb and ^{40}K have been almost constant over the last 5 years period.

Acknowledgement

The assistance of Mr. I. Světlík, who made the sampling and evaluation of ^{85}Kr in the last year is greatly appreciated.

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