DETERMINATION OF ^{210}Pb LOW ACTIVITY BY GAMMA SPECTROMETRY

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INTRODUCTION

The extensive studies aimed at finding out the dose-effect relationship in the area with increased in-door radon content and thus with supposed increased lung cancer incidence risk are carried out in the Czech Republic. The important part of such studies is to estimate the long term dose from radon and its short-lived progeny to lungs for each followed person. Usually, measurements of the activity of ^{222}Rn and its progeny in the in-door air of houses and the extrapolation to the past levels are the base for dose estimation There is also the possibility to assess the intake of short lived radon progeny deposited in the lungs by the in-vivo measurement of long lived radon progeny ^{210}Pb originating by their decay and then transported to other parts of the body and deposited mainly in bones. Annual average intake of ^{210}Pb is estimated to be 44 Bq which results in effective dose equivalent of 0.12 mSv (1). There is an effort to determine the amount of ^{210}Pb both in-vivo and in environmental samples by the detection of its 46.5 keV gamma rays. With regard to the efficiency of measurement the determination of ^{210}Pb deposited in bone surfaces of the skull seems to be the best solution for in-vivo measurement (2). The in-vivo determination is very difficult due to low yield of its gamma rays and also due to their low energy. In addition the activities which can be expected in the human body even with long term inhalation of high activities of radon and its short lived daughters are very low. At present time, the most serious problem is the sensitivity of instruments used. Even if this problem would be solved, the interpretation of activity of ^{210}Pb measured directly in-vivo in skull will be quite complicated because of direct intake of ^{210}Pb through inhalation and ingestion. The monitoring of amount of ^{210}Pb entering the body via direct ingestion and inhalation in areas with increased radon levels and comparison with areas considered as control, i.e. with average levels, is necessary for proper discrimination of individual components of ^{210}Pb retention. Therefore, in addition to the basic problem - calibration of special detectors for the measurement of ^{210}Pb in-vivo - an effort was undertaken to determine ^{210}Pb content in out-door air and particle size distribution of aerosols with attached ^{210}Pb, the content of ^{210}Pb in fallout and in drinking, mineral and table water. During the study the most suitable detector system was sought.

METHOD

The measurements of ^{210}Pb require special equipment with very low background. The equipment consists of special detectors (n type HPGe detectors, HPGe well detectors, HPGe planar detectors, LEGe detectors, special scintillation detectors - phoswichs). Very good shielding is substantial. In the NRPI there are 6 HPGe detectors available for the measurement of ^{210}Pb. Three of them are reverse electrodes detectors with thin Be window (n-type detectors). One well-type detector was used in the study. For the special case of measurement of low energy gamma emitters two LEGe (low energy Ge) detectors with beryllium and carbon-epoxy windows are used. The LEGe detector is a large area detector with a thin boron-implanted outer contact for high sensitivity at low energies. Because of device geometry, the capacitance is low, and this results in excellent resolution at low to moderate energies. The parameters of the detectors are summarized in Table 1. For comparison of capabilities of different detection systems also one p-type coaxial HPGe detector was included in the study. In addition to semiconductor detectors the possibility to use scintillation detectors was studied. Two scintillation NaI(Tl) detectors were used, one 5in x 4in with thin Al window (SC1) and one thin NaI(Tl) detector of 7 mm thickness with Be window (SC2).

RESULTS AND DISCUSSION

Sensitivity of different detection systems was studied. Because of low gamma energy of ^{210}Pb (46.5 keV) and low intensity of this energy line (4.05%), detector with high detection efficiency in this energy region and with low background is suitable. It was found, that all detectors with beryllium windows have approximately the same ^{210}Pb peak in their background, regardless of their detection efficiency (size), so that the source of ^{210}Pb is very near to the detector. As the lowest background in ^{210}Pb peak area was found with well detector (which endcap is from very thin aluminium with no beryllium), it was most probable that the source of ^{210}Pb is in beryllium window. The results of background measurements in the region of interest are summarized for semiconductor detectors in Table 2. There was the attempt to decrease the background by using additional shielding materials in close geometry e.g. copper, tungsten, steel, however with no significant effect. The

effort to decrease minimum detectable activity for ^{210}Pb resulted in complete reconstruction of endcap parts of LEGe detector No. 13, in which the beryllium window was replaced by the window from carbon-epoxy. The background count in the 46 keV peak area decreased approximately 10 times. This conclusion is very important for purchasing HPGe detectors when user has a.o. the aim to detect low activity of ^{210}Pb. The LEGe detector No. 20 was already purchased in special low-background arrangement.

Table 1 Parameters of semiconductor HPGe detectors used for ^{210}Pb measurement

Detector No.	Manufacturer	Type	Rel. efficiency 1333 keV [%]	FWHM 1333keV[keV]	FWHM 46.5keV [keV]	diameter [mm]	length [mm]
13	Canberra	LEGe	-	-	0.60	50.50	15.00
20	Canberra	LEGe	-	-	0.60	50.50	20.00
16	EG&G Ortec	n	38.30	2.00	1.00	59.80	61.60
53	EG&G Ortec	n	20.50	1.82	1.40	49.50	51.90
103	Canberra	n	15.50	1.68	1.10	46.50	39.50
93	Canberra	well	15.80	1.99	1.40	50.00	59.00
43	EG&G Ortec	p	25.00	1.85	1.30	52.90	60.60

Table 2 Summary of background in the region of 46.5 keV and minimum detectable activity for ^{210}Pb

Detector No.	Count rate in 46.5 keV peak [cps]	Integral count rate in the 46.5 keV region [cps]	Efficiency for planar source of ^{210}Pb	MDA [Bq]
13	0.00670	0.0104	0.151	0.20
13a[1]	0.00065	0.0026	0.151	0.10
20	0.00044	0.0031	0.137	0.12
16	0.00670	0.0273	0.159	0.30
53	0.00530	0.0220	0.106	0.40
103	0.00650	0.0099	0.113	0.33
93	0.00130	0.0146	0.381[2]	0.09
43	0.00130	0.0100	0.016	1.80
SC1	-	0.4000	0.169	1.20
SC2	-	0.1250	0.007	1.85

[1] after reconstruction of endcap

[2] efficiency for source inside well

Minimum detectable activity for different types of semiconductor and for scintillation detectors was calculated for typical geometry used for measurement of environmental samples, i.e. planar source with diameter of 5.2 cm. The results for 100000 s measurement time and for 5 % risk of error type I and II are included in Table 2. The determination concentration of ^{210}Pb in out-door air was the first attempt to estimate the amount of ^{210}Pb in various components of environment (3). High volume aerosol samplers (with air throughput from 60 $m^3.h^{-1}$ to 200 $m^3.h^{-1}$) were used for the aerosol collection altogether in four different places. Aerosol filters (PC-S, produced in SLZ, Hnúšťa, Slovakia), on which aerosol was collected one week, were measured without any treatment by semiconductor gamma spectrometry. Filters were measured in close geometry, using measuring time at least 100000 s. The average concentration of ^{210}Pb in the aerosol in out-door air in Prague for the whole sampling period 9 years (1986-1994) was found 540 $\mu Bq/m^3$. This value is in accordance with values given by UNSCEAR Report for the same geographical latitude. Similar values were found on other places in Czech republic, too. The ratio of activity of short lived progeny of ^{222}Rn and to the activity of radon itself to the activity of ^{210}Pb is approximately one order of magnitude higher in in-door air that in out-door air. It means that in-door aerosol with ^{210}Pb has much shorter mean time of life than the one in out-door air. There is higher

is higher probability of attachment of the particles on the walls and surfaces in-door than out-door. The ventilation of the room could influence ^{210}Pb content in in-door air, too.
Particle size distribution of aerosols with attached ^{210}Pb on their surfaces was studied with the use of 6-stage cascade impactor (Sierra Andersen, Model 236) attached to a high-volume air sampler Sierra Misco with a flow control system (Model Sierra Misco 5000). Flow-rate 0.565 $m^3.min^{-1}$ was used. Slotted fibreglass filter, sucked with solution of silicone grease to reduce re-entrainment and bounces of aerosol particles was placed on each stage. After the 6th stage the back-up filter for catching the rest of the particles was placed. Samplings with cascade impactor were performed both out-door and in-door. Main part of ^{210}Pb is attached both in out-door and in-door air to the small aerosol with aerodynamic diameter under 0.4 μm. Particle size distribution does not differ significantly from aerosol particle size distribution of short lived radon progeny. No difference in aerosol particle size distribution for the out-door and in-door air was found.
The yearly intake of ^{210}Pb by inhalation from out-door air is about 4 Bq (in accordance with UNSCEAR value). In the houses with elevated radon concentration, yearly intake by inhalation is from 70 to 700 Bq.
Table and mineral waters could be important contributors to the ingestion intake especially if their source is in places with high amount of uranium series radionuclides in the bedrocks. Water samples were measured in native form, after evaporation of 1 to 10 litres of water and after radiochemical separation of ^{210}Pb . Altogether 54 samples of potable, mineral and table water including one sample of pilsner beer were measured. The volume activity of ^{210}Pb ranged from detection limit (approx. 0.003 Bq/l) to 1.4 Bq/l. Majority of samples had the volume activity of ^{210}Pb below 0.1 Bq/l. Activity of ^{210}Pb in potable water was from less than 0.005 Bq/l to 1.4 Bq/l, activity in 16 samples of mineral and table water was lower with maximum of 0.2 Bq/l. Assuming the yearly consumption of potable water to be about 770 l for an average adult, the maximum intake of ^{210}Pb in exceptional cases could be about 1kBq which results in the dose of 1.3 mSv. The average yearly consumption of mineral and table water is for an adult about 14 l so the dose from ingestion of ^{210}Pb in this water is negligible.
A semiconductor LEGe detectors No.13 before and after reconstruction and No. 20 were used for in-vivo determination of ^{210}Pb in skull of people. Assumed distribution of ^{210}Pb on the skull bone surfaces was simulated by planar source moved on outer and inner surfaces of the skull. . The minimum detectable activity of ^{210}Pb in skull was determined for 5400 s measuring time to be about 20 Bq , i.e. about 130 Bq in total skeleton.

CONCLUSION

The study proved that the high resolution semiconductor gamma spectrometry is promising method for determination of ^{210}Pb in various kinds of environmental samples. Its sensitivity can be further improved using concentration and separation radiochemical methods for sample pretreatment. However, the most important field of application of semiconductor gamma spectrometry in measurement of ^{210}Pb is in-vivo determination of its amount in human body. For the detection of ^{210}Pb in-vivo in human skull further effort to decrease background in the 46.5 keV region is needed. The sensitivity can also be increased using geometry for simultaneous measurement with several LEGe detectors. Interpretation of measured values of ^{210}Pb in-vivo needs also the correction for enhanced direct intake of ^{210}Pb by inhalation in radon houses, which is enabled by the sampling and measurement of in-door air.

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