

METHOD FOR ^{210}Pb MEASUREMENT IN AIR BASED ON LSC

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ABSTRACT. ^{222}Rn decay causes ^{210}Pb and ^{210}Po to be present in the atmosphere. Background levels for both radionuclides average 3.7×10^{-4} and $3.7 \times 10^{-5} \text{ Bq m}^{-3}$, respectively. Some processes and situations cause a local increase of both radionuclides' concentrations of up to 10–100 times the normal levels. Due to the additional irradiation doses to the population, ^{210}Pb has radiological significance. Taking into account these reported levels, we estimate a minimum required sample volume of 300–500 m^3 . We apply 2 aerosol samplers (TF1A-2 and Staplex Inc.) coupled with a 4" diameter filter (synthetic Petrianov fabric) for the sites at Zhovty Wody and Dneprodzerzhinsk, both in the Dnipropetrovs'k region, as well as background samples from Kiev. Sample destruction allows one to prepare and measure LS total beta spectra with observed ^{210}Pb betas. The total beta spectra present varying combinations of beta components that require complicated analyses. We tested and applied beta-spectra decomposition to separate the ^{210}Pb beta peak for calculating corresponding activities.

INTRODUCTION

^{210}Pb ($T_{1/2} = 22.7 \text{ yr}$) and ^{210}Po ($T_{1/2} = 138.39 \text{ d}$) are present in the atmosphere due to ^{222}Rn decay. The National Council on Radiation Protection and Measurements [NCRP] Report 94 (NCRP 1988) estimates the background levels for both radionuclides as 3.7×10^{-4} and $3.7 \times 10^{-5} \text{ Bq m}^{-3}$, respectively. Later publications confirm the average ^{210}Pb background level as $0.5 \times 10^{-4} \text{ Bq m}^{-3}$ (Realo et al. 2004; UNSCEAR 2000). Some processes (fires, tobacco smoke, etc.) and some technological activities (mines, mining enrichment plants, heating and power stations) cause background levels to increase 10–100 times (Cloarec et al. 1995; Lambert et al. 1991; UNSCEAR 2000). Corresponding irradiation doses could be significant and cause health risks.

SAMPLE COLLECTION

We analyzed the ^{210}Pb in air within the framework of radiological estimates of environmental impact and potential irradiation doses to the public from uranium processing sites. The sampling sites selected were Zhovty Wody and Dneprodzerzhinsk (Figure 1), both located in the Dnipropetrovs'k region, and we used samples from Kiev as a background. We used 2 aerosol samplers, TF1A-2 and Staplex Inc., coupled with 4" diameter filter. A synthetic Petrianov fabric was used as filter matter. Taking into account the above-mentioned background levels, we estimated the minimum required air sample volume to be 300–500 m^3 (Buzinny et. al. 2005). Actual air samples collected ranged from 270–1100 m^3 .

SAMPLE PREPARATION

Aerosol filters were processed at least 2–3 weeks after sampling, allowing short-lived radon daughters to decay. Details of the sample preparation technique used for analyzing filters are reported elsewhere (Buzinny et al. 2005). Petrianov fabric was first compacted by acetone dissolving and then dried. Next, filter material residue was ashed in an oven at 500 °C. Ashes were dissolved in 25 mL of 6M HNO_3 while boiling for 30 min in glass vials, and undissolved particles were filtered. The resulting solution was evaporated; the residue was dissolved in 2 mL of 1M HNO_3 and placed into LS vials together with 2 mL of distilled water, which was used for washing the glass. The LS cocktail Optiphase HiSafe 3 was used for simultaneously counting and storing alpha and beta spectra utilizing the LS spectrometer Quantulus 1220TM. Destroying filter material at high temperatures causes additional disequilibrium between ^{210}Pb and ^{210}Po . Yet, we take into account only ^{210}Pb , which is

applicable for most tasks. After preparing and counting the filter samples, we find an intense spectrum shift due to quenching in Teflon® vials. Our research indicates that polyethylene vials are more transparent and thus more applicable for quenched samples.



Figure 1 Sampling sites of uranium ore deposits and processing plants in Ukraine

SPECTRA ANALYSES

Predictably, most of our attention was focused on spectra analysis. We found that most samples have a clear ^{210}Pb peak. As expected, samples are in disequilibrium between ^{210}Pb and ^{210}Bi , as previously reported (Buzinny et. al. 2005) in sample spectra for the Kiev site with the presence of other radionuclides. We require separation of the ^{210}Pb contribution in the sample beta spectrum itself (Buzinny et. al. 2005). To accomplish this, we subtract the smooth part of the beta spectrum produced by ^{210}Bi or other radionuclides, i.e. ^{137}Cs or even $^{90}\text{Sr}+^{90}\text{Y}$, from the total beta spectrum. For spectra decomposition, we apply a similar approach to what we have published previously (Buzinny et al. 1993). We practiced with the spectra of other radionuclides for different quenching conditions, since a pure ^{210}Bi spectrum could hardly be measured without chemical separation, i.e. ^{40}K and ^{137}Cs spectra. ^{137}Cs spectra were preferable; we describe below the ^{137}Cs quenched spectra set.

ALGORITHM

All sample spectra processing was performed using the Matlab® package by Mathworks, Inc. Sample processing begins by measuring calibrated samples of ^{210}Pb and ^{137}Cs as well as background spectra for different conditions of sample quenching. Next, we measured sample spectra. Each sample spectrum was prepared for mathematical processing (i.e. formatting to 1 column), and during processing, we normalized all spectra to be expressed as counts per second (cps).

To analyze each sample spectrum, we determined the sample quenching level SQP(E) by selecting the nearest background and ^{137}Cs spectra pair. We then subtract the background (see Equation 1) and perform the spectrum decomposition:

$$S = S_q - BG \quad (1)$$

where S is the net sample spectrum; S_q is the sample's measured spectrum; and BG is the corresponding background spectrum.

We then recalculate the complete modeled spectrum for channels 1–1024 according to the smooth spectrum shape between channels 350–500 and the corresponding ^{137}Cs spectrum interval (see Equations 2 and 3).

$$X = B(\text{rng},1) / S(\text{rng}) \quad (2)$$

$$S1 = B(:,1) \times X \quad (3)$$

where X is the calculated count rate for the “corresponding” radionuclide in the sample; B is the spectra matrix, which includes the calibrated normalized spectra for some radionuclides of possible interference (^{137}Cs , ^{40}K , $^{90}\text{Sr}+^{90}\text{Y}$) (we used ^{137}Cs); and rng is the region of interest, the part of the spectrum between channels 350 and 500.

To obtain the net peak of ^{210}Pb betas, we subtract the modeled spectrum from the sample spectrum:

$$S2 = S - S1 \quad (4)$$

Finally, we calculate the optimized window for ^{210}Pb calculation to be 130–320. This range is wide enough for high efficiency and sharp enough to prevent interference of possible sample luminescence. The spectra decomposition rate is shown in Figure 1 for the ^{210}Pb calibration source and for 1 aerosol filter sample. The electron peak of the calibrated ^{137}Cs spectrum is clearly visible in each figure.

SPECTRA STANDARDIZATION

Sample quenching corresponds to the variation of counting efficiency and spectra shape, which are standardized by the external standard principle and corresponding parameter SQP(E) used in Quantulus 1220. Our experience in finding the low-energy spectrum range corresponding to ^{210}Pb betas has taught us to pay close attention to changes in efficiency. Spectrum shape changes are smooth enough to use quenched spectra ranged for 10 units of SQP(E), corresponding to 8 spectra sets for the applicable SQP(E) range of 720–800. After selecting the appropriate spectra set, we proceeded with each sample spectrum. Precise spectrum standardization includes a shift to the left or right side for a range of up to 8 channels. The spectra shift actually plays a minor role in spectra decomposition and can be ignored for channels 1–3.

Counting efficiency is well defined in the wide SQP(E) range for the ^{210}Pb standard solution in equilibrium up to ^{210}Po , which could be precisely measured on Quantulus 1220 using alpha-beta separation. The corresponding ^{210}Pb counting efficiency trend for sample quench is shown in Figure 2.

OTHER APPLICATIONS

When we applied the spectra decomposition to the Chernobyl filters, we were able to separate and clearly see the ^{241}Pu spectrum. We thus could compare the ^{241}Pu spectrum to the sample's total alpha spectrum, or even see both ^{241}Pu and ^{210}Pb betas at the same time for another sample (see Figure 3).

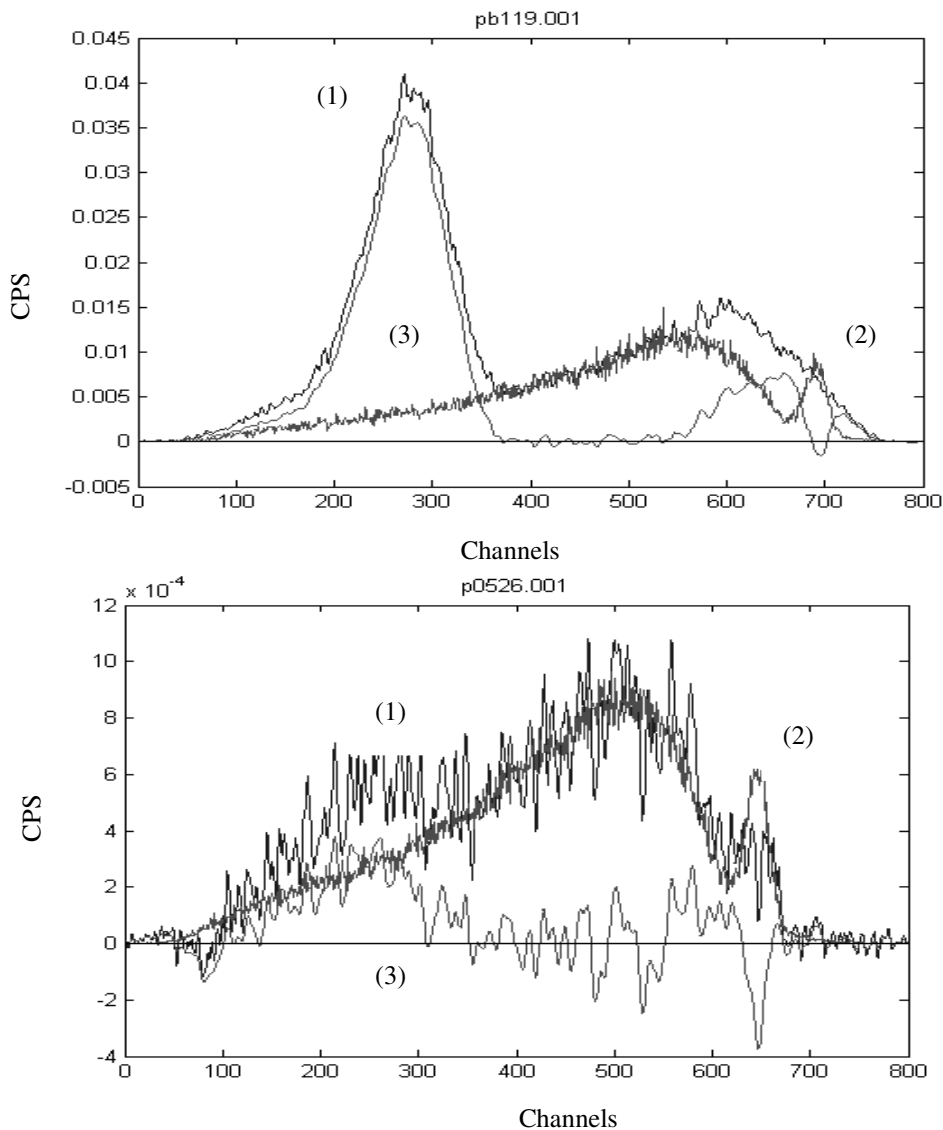


Figure 2 Decomposition of LS beta spectra. Top: 1) $^{210}\text{Pb}+^{210}\text{Bi}$; 2) corresponding ^{137}Cs used to fit smooth shaped ^{210}Bi ; and (3) fitted ^{210}Pb peak. Bottom: 1) sample total beta; 2) corresponding ^{137}Cs ; and 3) fitted ^{210}Pb peak. The conversion electron peak for the calibrated ^{137}Cs spectrum is clearly visible in each figure.

RESULTS

We tested an approach for sample LS spectra by analyzing ^{210}Pb . This method is based on the decomposition of sample spectra using the corresponding ^{137}Cs beta spectrum shape for low energies, i.e. between of 350–500 channels. We tested the spectra decomposition system for some spectra of ^{210}Pb calibration source for different quenching conditions, as well as for spectra of measured aerosol filters from the sampling sites of Kiev, GW, and PCP site. The ^{210}Pb calibration source provides results that allow the quench correction for the measured filters' spectra. By knowing the different radionuclides that cause interference for ^{210}Pb determination, as well as levels of interfering

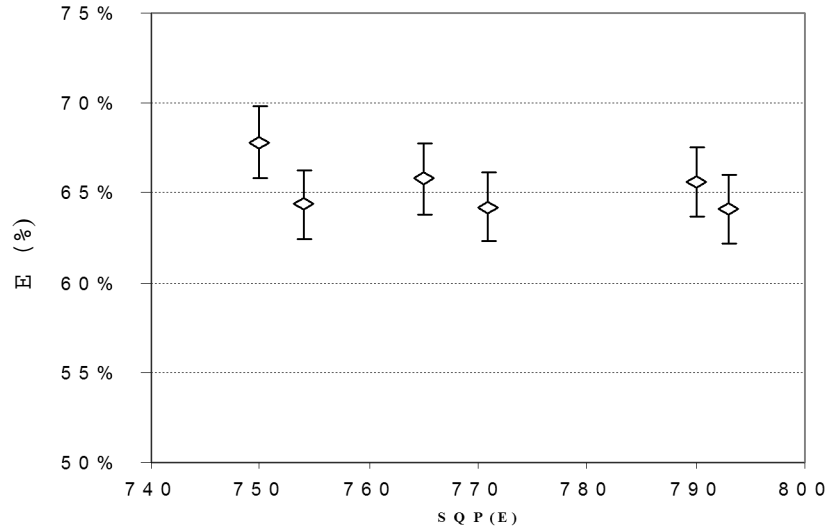


Figure 3 ^{210}Pb counting efficiency trend for filter sample quench

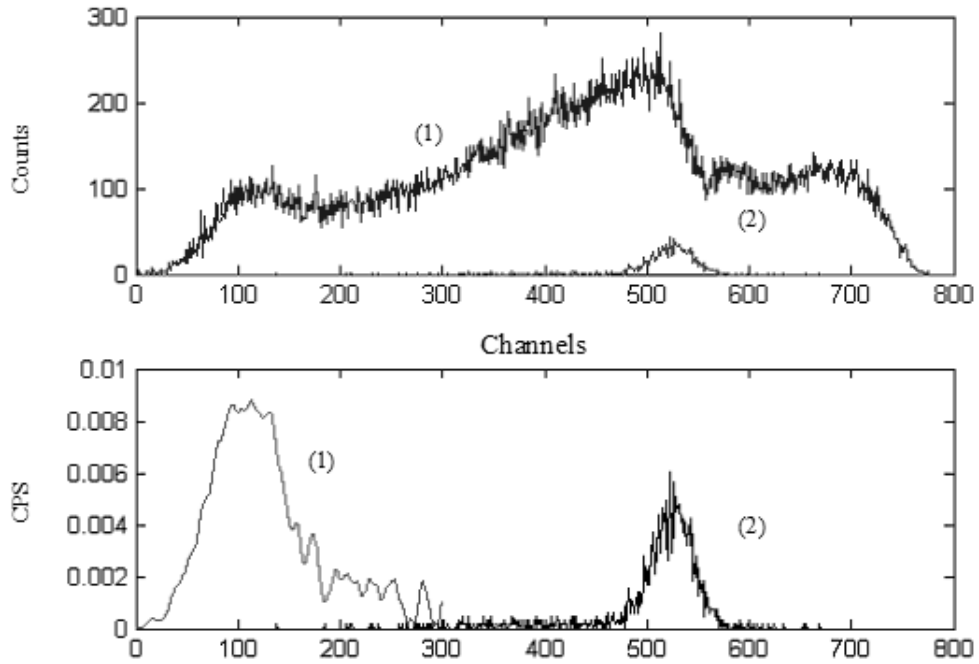


Figure 4 Decomposition of LS beta spectrum for the Chernobyl aerosol filter. Top: 1) total beta and 2) total alpha spectra. Bottom: 1) decomposition of low-energy beta spectrum ($^{241}\text{Pu}+^{210}\text{Pb}$) compared to 2) corresponding sample's total alpha ($^{239+239+240}\text{Pu}$ and ^{210}Po).

radionuclides for different sites when we process with spectra, we are able to determine the corresponding estimates of total beta activity, ^{210}Pb level, ^{210}Bi level, ^{137}Cs level if appropriate, and discover the presence high beta emitters in total beta spectrum, i.e. $^{90}\text{Sr}+^{90}\text{Y}$.

All filter activity data obtained are presented at Table 1 and Figure 5. A wide range of ^{210}Pb is seen in air-specific activity variation.

Table 1 ^{210}Pb activity (Bq m^{-3}) histogram for aerosol filters analyzed.

Nr	Site	Sampling date	Air volume (m^3)	$^{210}\text{Pb} \times 10^{-4}$ (Bq m^{-3})
1	Kiev	28.04.2004	425	1.63
2	Kiev	30.04.2004	613	0.77
3	Kiev	25.05.2005	233	15.2
4	Kiev	26.05.2005	539	4.61
5	Kiev	27.05.2005	479	4.31
6	Kiev	30.05.2005	533	6.55
7	Kiev	31.05.2005	910	8.46
8	Kiev	01.06.2005	667	2.83
9	Kiev	02.06.2005	554	2.38
10	Kiev	03.06.2005	336	4.96
11	Kiev	16.06.2005	446	7.87
12	Kiev	21.06.2005	1090	2.63
13	Kiev	22.06.2005	580	2.65
14	Kiev	23.06.2005	589	6.06
15	Zhovty Wody	22.07.2005	279	11.3
16	Zhovty Wody	22.07.2005	321	2.52
17	Dnieprodzherzhinsk	28.07.2005	271	12.1
18	Dnieprodzherzhinsk	28.07.2005	312	0.74
19	Dnieprodzherzhinsk	27.07.2005	186	1.10
20	Dnieprodzherzhinsk	29.07.2005	261	5.14
21	Dnieprodzherzhinsk	29.07.2005	301	0.85
22	Dnieprodzherzhinsk	25.08.2005	516	6.87
23	Zhovty Wody	14.10.2005	441	16.4
24	Zhovty Wody	14.10.2005	508	15.4
25	Dnieprodzherzhinsk	09.11.2005	392	23.6
26	Dnieprodzherzhinsk	09.11.2005	451	20.4

CONCLUSIONS

We developed and tested a comprehensive LS-based tool for ^{210}Pb beta spectra separation, allowing us to calculate the activity in the aerosol filters. The approach allows the ^{210}Pb determination in the presence of unstable ^{210}Bi or even other radionuclides, such as ^{137}Cs and $^{90}\text{Sr}+^{90}\text{Y}$.

A similar approach could be applied for ^{241}Pu beta separation from the total beta LS spectra of aerosol filters from Chernobyl, which have a high activity of ^{137}Cs and $^{90}\text{Sr}+^{90}\text{Y}$. The main advantage in analyzing the total beta spectra is the ability to discover any new component of beta activity.

Calculated ^{210}Pb activities for the studied filters vary from 8.4×10^{-5} to $2.3 \times 10^{-3} \text{ Bq m}^{-3}$. The measured data are well defined, showing a wide contrast and wide uncertainty. Thus, to define the origin of ^{210}Pb using short-collection-time filters requires gathering 2 separate filters collected at the same time using the same technique in 2 different locations under similar weather conditions, with 1 used as a background and 1 to be studied.

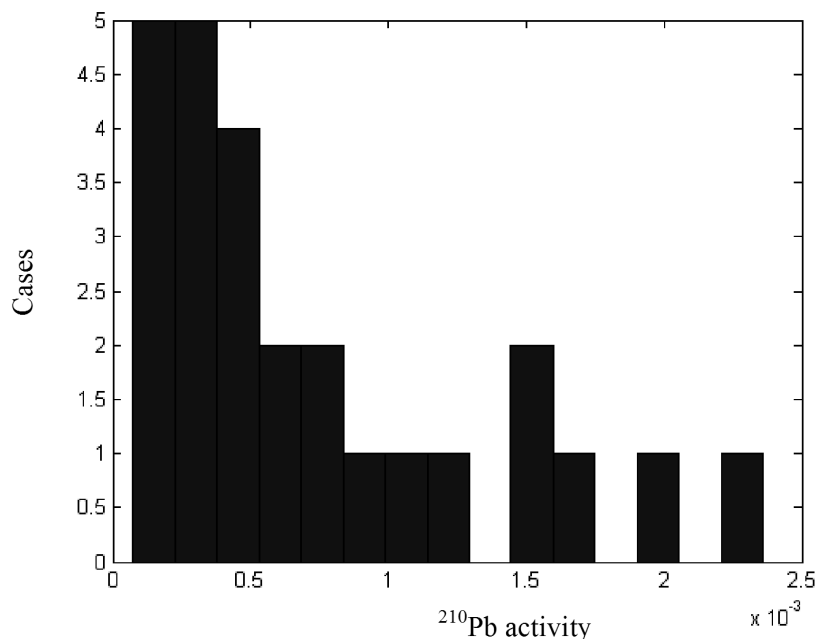


Figure 5 ^{210}Pb activity (Bq m⁻³) histogram for aerosol filters analyzed

In the near future, we plan to compare measurements performed by LS counting total beta spectra decomposition to measurements by direct gamma spectroscopy.

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