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DETERMINATION OF AEROSOL MEAN RESIDENCE TIME USING ^{210}Pb AND ^{7}Be RADIONUCLIDES IN THE ATMOSPHERE OF ISLAMABAD

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The concentrations of ^{210}Pb, ^{214}Pb and ^{7}Be radionuclides (having radioactive half lives of 22.3 years, 27 min. and 53.29 days, respectively) in 184 surface level air samples of Islamabad (33.38° N, 73.10° E) were measured using the technique of non-destructive gamma-spectrometry on high purity germanium (HPGe) detector. The mean concentrations of ^{210}Pb, ^{214}Pb and ^{7}Be in the samples were found to be 0.284 ± 0.012 mBq m^{-3}, 631.06 ± 155.47 mBq m^{-3} and 3.171 ± 0.084 mBq m^{-3}, respectively. The aerodynamic size distribution of ^{7}Be in the atmospheric aerosols was also determined, using Anderson 1-ACFM cascade impactors, which resulted in the mean diameter of ^{7}Be-laden aerosols as 0.79 ± 0.12 µm. The mean residence times (MRT) for ^{210}Pb and ^{7}Be-laden aerosols, thus obtained, were 5.29 ± 0.91 days and 7.18 ± 0.97 days respectively.

Keywords: Radionuclides, Spectroscopy, Scavenging, Aerosols, Concentration, Vertical mixing.

1. Introduction

Aerosols present in the atmosphere, is a result of a large number of chemical and physical processes [1]. These chemical and physical processes depend on the particles number density and their residence time. The boundary layer aerosols, defined as those particles, liquid or solid, lie at the interface between a solid surface and a thin fluid layer, extending to a height of about 100 m above the ground surface, that is static because of friction between the molecules of the fluid and the solid surface, are the main carriers of natural and artificial, radioactive and non-radioactive substances released into the atmosphere. The radioactive, water soluble and aerosol-borne substances act as tracers in the atmosphere and are considered as ideal tools for the study of atmospheric transport processes. The source distribution of these tracers is well known, they are removed from the atmosphere by the radioactive decay as well as by dry and wet deposition to different complex structures. Once produced, these tracers instantaneously and irreversibly attach themselves to aerosols of the relevant sizes (the radii of these aerosols lie between 0.02 to 1.0 µm) and are distributed by the general atmospheric circulation [2, 3].

Radioactive tracers in the environment can be divided into three main groups, based on their origin [4]. Primordial are those radionuclides present during the formation of earth and are still present due to their long half lives, cosmogenic are those radionuclides produced by cosmic rays in the atmosphere and anthropogenic are those radio-nuclides produced in the environment by nuclear tests and nuclear incidents. We choose ^{210}Pb from primordial and ^{7}Be from cosmogenic radionuclides as tracers in the study of submicron sized aerosol deposition and their residence times in the atmosphere using air measurements.

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^{210}Pb ($T_{1/2}$=22.3 years) is a naturally occurring radionuclide. It is formed from the decay of ^{222}Rn and belongs to ^{238}U decay series. ^{7}Be ($T_{1/2}$=53.3 days) is a cosmogenic radionuclide. It is formed in the lower stratosphere and upper troposphere as a result of spallation reactions, disintegration of nitrogen, oxygen and carbon nuclei that have been hit by cosmic ray neutrons and protons [5].

The residence time is an important parameter of aerosols that determines their transport in the atmosphere and can be used to study the dispersal of aerosols on regional and global scale. Its determination in the different layers of atmosphere has been attempted by several researchers around the world, using concentrations of fission products (^{90}Sr, ^{137}Cs etc.), cosmic rays produced radionuclides (^{7}Be, ^{32}P, ^{35}S etc.) and activity ratios of ^{222}Rn and its decay products.

For the residence time of ^{210}Pb laden aerosols, the ^{222}Rn and its decay product ratios namely, ^{210}Po/^{210}Pb, ^{210}Bi/^{210}Pb, ^{222}Rn/^{210}Pb and ^{214}Pb/^{210}Pb have been used in the air and rain water samples of atmosphere [6-9]. Since the ratios of ^{210}Po/^{210}Pb and ^{210}Bi/^{210}Pb have either overestimated the values of mean residence time or underestimated them. The reasons for the overestimation or underestimation are the large variations in the concentrations over oceans and continents, different values of ^{210}Pb in the upper and lower atmosphere and the addition of soil particles and upper atmospheric aerosols. Moreover, the labor, difficulty and large errors in the radio-chemical separation and long waiting times after sampling for the determination of ^{210}Bi activities have reduced the use of ^{210}Bi/^{210}Pb ratio for the calculation of aerosol residence time [6, 10-14]. So, we used the ratio ^{214}Pb/^{210}Pb for the estimation of residence time of ^{210}Pb-laden aerosols in the surface level air (surface level air is defined as the air prevailing near the ground surface) following the method used in Ref. [8], as both these nuclides have the same origin and both are gamma emitters. For the residence time of ^{7}Be laden aerosols, the method adopted in Ref. [15], based on the aerosol particle growth rate measured on Anderson 1-ACFM, has been used in this study.

The data regarding different parameters of aerosol particles are still scarce or even not existing in this region of Asia, particularly, in Pakistan. So by having such data (18 months measurements in the air from Islamabad) on aerosols, we will not only provide the baseline data for the region but will also be able to study the atmospheric transport phenomena in the locality with the help of residence time index in the near future.

2. Materials and Methods

2.1. Air sampling and measurement technique

In order to measure the ^{210}Pb, ^{214}Pb and ^{7}Be concentrations in the surface air, sampling was carried out on Gelman type A/E Glass Fibre filters (47 mm diameter) that were used in the portable air sampler (model DF-AB-75L) installed in the open lawn near Physics Department, COMSATS Institute of Information Technology (CIIT), Chak Shehzad, Islamabad (Latitude 33^{o} 38′ N, Longitude 73^{o} 10′ E), at a height of 6 feet above the ground level. The height of 6 feet was chosen to see the effect of aerosols on humans.

The air sampler was used for a fixed time of 15 hours at a flow rate of 26 litres per minute (LPM). Earlier, it was established that 15 hours of air sampling and 26 LPM flow rate on the air sampler are the optimum parameters, for having enough particulates on the filter without any chance of choking even in dusty seasons.

The sampling programme started on 16th of July 2007 and ended on 15th of January 2009. A total of 184 samples, about thrice a week, were collected and measured by γ-ray spectrometry using an N-type HPGe detector of resolution 2.2 keV at 1.33 MeV of ^{60}Co and relative efficiency of 52%. The samples were counted for 24 hours in order to have a reasonable number of counts in the spectrum. The absolute efficiencies of the detector for ^{210}Pb (at 46.50 keV), ^{214}Pb (at 351.92 keV) and ^{7}Be (at 477.56 keV) were calculated to be about 5.6%. 22.6% and 13.5% respectively as shown in Fig.1. The data were analyzed by a PC using an MCA card MCDWIN, having a commercial software Gamma-W supplied by DSG, Germany.

2.2. Determination of mean residence times

The procedures for energy calibration and geometric efficiency determination were adopted from the technical report series [16]. The counting efficiency of the detector was determined, using a

^{166m}Ho source for the calibration. Each sample thus collected on filter was directly placed on the top of the detector. It is to be mentioned here that the background was periodically measured, observing the same sample matrix in the detector.

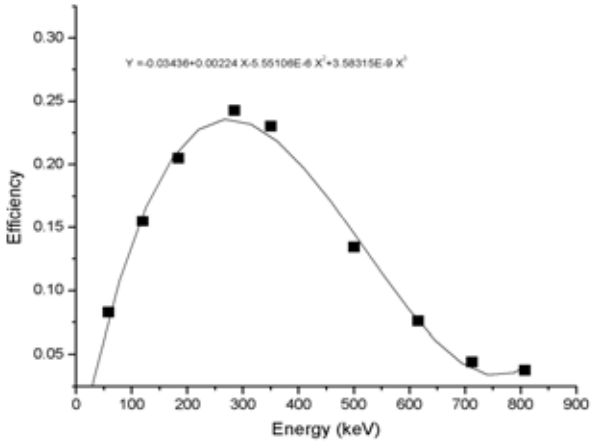

Figure 1. Efficiency response curve of HPGe detector with energy.

The activity concentrations of ^{214}Pb and ^{7}Be were calculated from the expression

$$C = \frac{N\lambda^2}{(1-e^{-\lambda t})\times(e^{-\lambda t_1}-e^{-\lambda t_2})\times P_\gamma \times \varepsilon_\gamma \times Q} \qquad (1)$$

Where N is the net area under the full energy peak of gamma-ray, λ is the decay constant (s^{-1}), t is the time of air sampling (s), t_1 is the time difference between the end of air sampling and the start of detection, t_2 is the time difference between the end of air sampling and the end of detection, P_γ is the transition probability of the nuclide and ε_γ is the detector efficiency of nuclide at the specified energy and Q is the flow rate (m^3s^{-1}) of air sampler. Here t_1 and t_2 were introduced for the decay correction in activity of the samples during counting in the detector.

Similarly the concentration of ^{210}Pb was calculated by using the equation:

$$C = \frac{(C_\gamma)_E}{(P_\gamma \times \varepsilon_\gamma)_E \times Q} \qquad (2)$$

Where $(C_\gamma)_E$, $(P_\gamma)_E$ and $(\varepsilon_\gamma)_E$ are count rate, transition probability and detector efficiency of the radionuclide at energy E respectively.

The measured values of activity concentrations of ^{210}Pb and ^{214}Pb were then incorporated to the mean residence time (MRT) for aerosols suspended in the air by using the approximation mentioned in Ref. [8]:

$$MRT = \frac{C_{(^{210}Pb)}}{C_{(^{214}Pb)}} \times \lambda^{-1}_{(^{210}Pb)} \qquad (3)$$

Where $\lambda = \frac{\ln 2}{T_{\frac{1}{2}}(^{210}Pb)}$, $T_{\frac{1}{2}}$ is the half life of ^{210}Pb.

For the estimation of residence time of ^{7}Be laden aerosols, aerodynamic size distribution of ^{7}Be in the atmospheric aerosols were determined, using an Anderson 1-Actual Cubic Foot per Minute (1 ACFM) cascade impactors. A flow rate of 30 LPM (Litres per Minute) and eight atmospheric pressure stages were adjusted for the collection of aerosols above 0.4 μm in the impactor for the purpose. The effective cut-off diameters of this instrument involve 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 5.8 and 9.0 μm. Aerosols through dry deposition were collected on stainless steel plates and glass fiber filters (47 mm diameters) were used as back-up filters to collect particles below 0.4 μm collection plate. These filters and stainless steel plates were then treated as radioactive sources and the activity concentrations of ^{7}Be through 477.6 KeV γ-rays were measured in the HPGe detector. The activity median aerodynamic diameters (AMAD) obtained in this work, from 15 measurements each of one week collection time about 6 feet above the ground, ranged from 0.48 to 0.94 μm (with mean 0.79±0.12 μm) and the average value of activity median aerodynamic diameters for aitken nuclei $(AMAD)_{Aitken}$ has been reported as 0.015 μm [17], which were then incorporated in the approximation used for the determination of aerosol residence time (τ_R), as

$$\tau_R = \frac{(AMAD)_{mean} - (AMAD)_{Aitken}}{MGR} \qquad (4)$$

Where $(AMAD)_{mean}$, $(AMAD)_{Aitken}$ are the mean and aitken sized nuclei activity median aerodynamic diameters respectively, and MGR is the Mean Growth Rate (0.004-0.005 μmh^{-1}) of aerosol particles [18], from which the mean value was taken.

Table 1. Statistics of the ^{210}Pb, ^{214}Pb and ^{7}Be concentrations (mBq m^{-3}) and mean residence time of aerosol over Islamabad.

Radionuclide	Quartiles					Mean	Uncertainty	MRT (days)
	Min.	1st	Median	3rd	Max.	(μ)	(σ)	
^{210}Pb	0.056	0.172	0.246	0.390	0.761	0.284	0.012	5.29±0.91
^{214}Pb	200.56	385.35	554.37	875.77	1371	631.06	155.47	
^{7}Be	0.772	2.321	2.977	3.672	6.847	3.171	0.084	7.18±0.97

3. Results and Discussion

The activity concentrations of ^{210}Pb, ^{214}Pb and ^{7}Be in all air particulate samples obtained by γ-ray spectrometry varied from 0.056±0.01 to 0.761±0.02 mBq m^{-3}, 179±32 to 1520±105 mBq m^{-3} and 0.772±0.05 to 6.847±0.13 mBq m^{-3} respectively. The mean values of these radionuclides were calculated as 0.284±0.01 mBq m^{-3}, 640±127 mBq m^{-3} and 3.171±0.08 mBq m^{-3} respectively as given in Table 1. The mean residence times of tropospheric aerosols has been reported in the literature to vary from 4 to 60 days [19], depending on the origin of the aerosols, their size distribution, the frequency of rain events and the dry deposition. In Ref. [10], the authors have reported the MRT of aerosols in the lower atmosphere of USA to be ≤ 4 days and showed its increased trend with the increasing altitude within the troposphere. Ref. [20] has mentioned the deduced mean residence times,based on modeled results, for the aerosols of northern and southern tropospheres, as 5.88 days and 8.79 days respectively. Based on the ^{210}Pb/^{214}Pb ratio calculation, the mean residence time of ^{210}Pb-laden aerosols in the surface level air was found as 5.29±0.91 days and the mean residence time of ^{7}Be-laden aerosols, from the activity median aerodynamic diameters (AMAD) based calculation, was obtained as 7.18±0.97 days. Though both the residence times of ^{210}Pb and ^{7}Be laden aerosols obtained here in this work have not a wide range of values 3.28 to 6.52 days and 4.31 to 8.56 days respectively but surely the variations in these values are a result of few factors, namely, wet deposition due to washout effect, mass exchange between the stratosphere and the troposphere, vertical transport in the troposphere due to increase of temperature in the summer season and horizontal transport due to different pressure areas. The lower values of the MRT range for both radionuclides seem to be due to the removal process of wet scavenging, as heavy rain falls happen during monsoon in July and August and the upper values of MRT range are due to vertical transport (downwards or upwards) of these radionuclides in stratosphere and troposphere, specially for ^{7}Be-laden aerosols in spring seasons. Moreover, the hot summer seasons bring westerly and south-westerly winds from mostly barren lands, full of dust and smoke to the area and are probably responsible for the increased concentrations of radon decay products in the atmosphere which in turn transfer these radionuclides to the upper atmospheric boundary, hence having long MRT values. Although these MRT values are comparable to the reported values in the literature e.g. see Ref. [8, 10, 15 and 20], but somewhat smaller than the expected values were found in this study. The reason may be the fact that samples were collected about 6 feet above the ground level where the aerosols tend to condense and coagulate near the earth surface due to comparatively high relative humid air, thus increasing the size of aerosol and the probability of deposition increases which in turn shortens the MRT of these aerosols.

4. Conclusions

A total of 184 air particulate samples were collected on glass fibre filters, using portable air sampler and measured by γ-ray spectrometry using a high resolution, low background, N-type HPGe detector. The air samples of 15 hours sampling period about thrice a week, were collected and the γ-ray activities of ^{210}Pb, ^{214}Pb and ^{7}Be radioisotopes were determined for 24 hours. For the size fractionation of ^{7}Be-laden aerosols, aerodynamic size distribution of ^{7}Be in the atmospheric aerosols were determined, using Anderson 1-Actual Cubic Foot per Minute (1 ACFM) cascade impactors. Aerosols through dry deposition were collected on stainless steel plates

and glass fiber filters were used as back-up filters to collect particles below 0.4 μm size. These filters and stainless steel plates were then measured for the activity concentrations and size fractionation respectively of ^{7}Be-laden aerosols.

The mean values of residence times of ^{210}Pb and ^{7}Be laden aerosols in the surface level air thus obtained from different methods in this work are 5.29±0.91 days and 7.18±0.97 days respectively. These MRT values agree well with the reported MRT values found in the literature.

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