

# DETERMINATION OF ATMOSPHERIC $^{210}\text{Pb}$ AND $^7\text{Be}$ -LADEN AEROSOLS' MEAN RESIDENCE TIMES OVER ISLAMABAD.

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The concentrations of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  radio-nuclides (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  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  in the samples were found to be  $0.284 \pm 0.012$  mBq m<sup>-3</sup>,  $631.06 \pm 155.47$  mBq m<sup>-3</sup> and  $3.171 \pm 0.084$  mBq m<sup>-3</sup>, respectively. The aerodynamic size distribution of  $^7\text{Be}$  in the atmospheric aerosols were also determined, using an Anderson 1-ACFM cascade impactors, which resulted in the mean diameter of  $^7\text{Be}$ -laden aerosols as  $0.79 \pm 0.12$  μm. The mean residence times (MRT) for  $^{210}\text{Pb}$  and  $^7\text{Be}$ -laden aerosols, thus obtained, were  $5.29 \pm 0.91$  days and  $7.18 \pm 0.97$  days respectively.

Keywords: Radio-nuclides, Spectroscopy, Scavenging, Aerosols, Concentration, Vertical Mixing.

## 1. Introduction

Aerosols present in the atmosphere, are a result of a large number of chemical and physical processes [1]. These processes depend on the particle number concentration and residence time. The boundary layer aerosols are the main carrier 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 relatively 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 (those present during the formation of earth and are still present in the atmosphere due to their long half lives): Cosmogenic (those produced by cosmic rays in the atmosphere and lithosphere) and Anthropogenic (those produced in the environment by nuclear tests and nuclear incidents). We choose

$^{210}\text{Pb}$  from primordial and  $^7\text{Be}$  from cosmogenic radio nuclides as tracers in the study of submicron sized aerosol deposition and their residence times in the atmosphere using air measurements.

$^{210}\text{Pb}$  ( $T_{1/2}=22.3$  years) is a naturally occurring radio nuclide, formed from the decay of  $^{222}\text{Rn}$ , belonging to  $^{238}\text{U}$  decay series and  $^7\text{Be}$  ( $T_{1/2}=53.3$  days) is a cosmogenic radio nuclide, 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 ( $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  etc.) from nuclear oriented activities, cosmic rays produced radio nuclides ( $^7\text{Be}$ ,  $^{32}\text{P}$ ,  $^{35}\text{S}$  etc.) and activity ratios of  $^{222}\text{Rn}$  and its decay products.

For the residence time of  $^{210}\text{Pb}$  laden aerosols, the  $^{222}\text{Rn}$  and its decay product ratios namely,  $^{210}\text{Po}/^{210}\text{Pb}$ ,  $^{210}\text{Bi}/^{210}\text{Pb}$ ,  $^{222}\text{Rn}/^{210}\text{Pb}$  and  $^{214}\text{Po}/^{210}\text{Pb}$  have been used in the air and rain water samples of atmosphere [6-9]. Since the ratios of  $^{210}\text{Po}/^{210}\text{Pb}$  and  $^{210}\text{Bi}/^{210}\text{Pb}$  have either overestimated the values of mean residence time or underestimated them due to the large variations in the concentrations over oceans and continents, different values of  $^{210}\text{Pb}$  in the upper and lower atmosphere, the addition of soil particles and upper atmospheric aerosols, the labor, difficulty and large errors in the radio chemical separation and long waiting times after sampling for the determination of  $^{210}\text{Bi}$  activities [6, 10-14]. So, we used the ratio  $^{214}\text{Po}/^{210}\text{Pb}$  for the estimation of residence time of  $^{210}\text{Pb}$ -laden aerosols in the surface level air, following the best suited method used by Ref. [8], as both these nuclides have the same origin and both are gamma emitters.

For the residence time of  $^7\text{Be}$  laden aerosols, the method adopted by 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 near future.

## **2. Materials and Methods**

### **2.1 Air Sampling and Measurement Technique**

In order to measure the  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  concentrations in the surface air, analysis were carried out on Gelman type A/E Glass Fibre filters (47 mm diameter) that were used in portable air sampler (model DF-AB-75L) installed in the open lawn near Physics Department, COMSATS Institute of Information Technology (CIIT), Chak Shehzad Campus (Latitude  $33^\circ 38'$  N, Longitude  $73^\circ 10'$  E), about 06 feet above the ground. The height of 06 feet was chosen to see the effect of polluted aerosols on humans and to minimize the chance of choking of filter at relatively low heights.

The air sampler was used for fixed time, 15 hours at a flow rate of 26 LPM (Litres per Minute) and it was established that 15 hours of air sampling by the air sampler at this flow rate is best suited without any chance for choking of filter even in dusty seasons.

The sampling programme started on 16<sup>th</sup> of July 2007 and ended on 15<sup>th</sup> of January 2009. A total of 184 samples, about thrice a week, were collected and measured by  $\gamma$ -ray spectrometry using a high resolution (2.2 keV at 1.33 MeV  $^{60}\text{Co}$ ), high efficiency (52%), low background, N-type HPGe detector. The  $\gamma$ -ray activities of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  radioisotopes were detected for about 24 hours in the detector in order to have a reasonable number of counts in the spectrum, with their efficiencies of about 5.6%, 22.6% and 13.5% at energies of 46.50 keV, 351.92 keV and 477.56 keV respectively. The data were analyzed by a PC using an MCA card MCDWIN containing a commercial software Gamma-W supplied by DSG, Germany.

## 2.2 Determination of Mean Residence Times

The concentrations of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  in air samples were determined using an N-type HPGe (D.S.GmbH) detector having a 52.3% relative efficiency, from the 46.50 KeV, 351.92 KeV and 477.56 KeV  $\gamma$ -ray lines, respectively. 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  $^{60}\text{Co}$  source for the calibration. Each sample was directly placed on the top of the detector, following identical geometry of the standard reference material. It is to be mentioned here that the background was periodically measured, observing the same sample matrix in the detector.

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

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

Where N is the number of disintegrations of the nuclide during detection time,  $\lambda$  is the decay constant ( $\text{s}^{-1}$ ), 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,  $P_\gamma$  is the transition probability of the nuclide and  $\varepsilon_\gamma$  is the detector efficiency of nuclide at the specified energy and Q is the flow rate ( $\text{m}^3\text{s}^{-1}$ ) 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  $^{210}\text{Pb}$  was calculated by using the equation:

$$C = \frac{(C_\gamma)_E}{(P_\gamma \times \varepsilon_\gamma)_E \times Q} \quad \text{Eq. (2)}$$

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  $^{210}\text{Pb}$  and  $^{214}\text{Pb}$  were then incorporated to the mean residence time (MRT) for aerosols suspended in the air by using the well known approximation mentioned in Ref. [8]:

$$MRT = \frac{C_{(^{210}\text{Pb})}}{C_{(^{214}\text{Pb})}} \times \lambda_{(^{210}\text{Pb})}^{-1} \quad \text{Eq. (3)}$$

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

For the estimation of residence time of  $^7\text{Be}$  laden aerosols, aerodynamic size distribution of  $^7\text{Be}$  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  $0.4 \mu\text{m}$  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  $9.0 \mu\text{m}$ . Aerosols through dry deposition were collected on stainless steel plates and glass fiber filters ( $47 \text{ mm}$  diameters) were used as back-up filters to collect particles below  $0.4 \mu\text{m}$  collection plate. These filters and stainless steel plates were then treated as radioactive sources and the activity concentrations of  $^7\text{Be}$  through  $477.6 \text{ KeV}$   $\gamma$ -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  $0.94 \mu\text{m}$  (with mean  $0.79 \pm 0.12 \mu\text{m}$ ) and the average value of activity median aerodynamic diameters for aiten nuclei  $(AMAD)_{\text{Aitken}}$  has been reported as  $0.015 \mu\text{m}$  [17], which were then incorporated in the approximation used for the determination of aerosol residence time, as

$$\tau_R = \frac{(AMAD)_{\text{mean}} - (AMAD)_{\text{Aitken}}}{MGR} \quad \text{Eq. (4)}$$

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

### 3. Results and Discussions

Surface air measurements were performed by pumping and filtering the air at an elevation of 06 feet above the ground ( $\sim 1760 \text{ ft asl.}$ ) in the open lawn near Physics Department, COMSATS Institute of Information Technology (CIIT), Chak Shehzad Campus Islamabad.

The activity concentrations ( $\text{mBq m}^{-3}$ ) of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  in surface air obtained for each filter by  $\gamma$ -ray spectrometry, using N-type HPGe detector varied from  $0.056 \pm 0.006 \text{ mBq m}^{-3}$  to  $0.761 \pm 0.023 \text{ mBq m}^{-3}$ , with mean value of  $0.284 \pm 0.012 \text{ mBq m}^{-3}$ ,  $179 \pm 32 \text{ mBq m}^{-3}$  to  $1520 \pm 105 \text{ mBq m}^{-3}$ , with mean value of  $640 \pm 127 \text{ mBq m}^{-3}$  and  $0.772 \pm 0.051$  to  $6.847 \pm 0.125 \text{ mBq m}^{-3}$ , with mean value of  $3.171 \pm 0.084 \text{ mBq m}^{-3}$  respectively for all the 184 measurements 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  $\leq 4$  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  $^{210}\text{Pb}/^{214}\text{Pb}$  ratio calculation, the mean residence time of  $^{210}\text{Pb}$ -laden aerosols in the surface level air was found as  $5.29 \pm 0.91$  days and the mean residence time of  $^7\text{Be}$ -laden aerosols, from the activity median aerodynamic diameters (AMAD) based calculation, was obtained as  $7.18 \pm 0.97$  days. Though both the residence times of  $^{210}\text{Pb}$  and  $^7\text{Be}$  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 variation in these values are a result of a 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 MRT for both the radio-nuclides seem to be due to the removal process of wet scavenging, as heavy rain falls happen during monsoon in July and August and the higher MRT values are due to vertical transport in stratosphere and troposphere, specially for  $^7\text{Be}$ -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 radio-nuclides 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 as the collection were made about 6 feet above the ground level where the aerosols tend to condensation and coagulation 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 samples were collected on portable air sampler and measured by  $\gamma$ -ray spectrometry using a high resolution, low background, N-type HPGe detector. The air samples of 15 hours sampling period, about twice a week, were collected and the  $\gamma$ -ray activities of  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  radioisotopes were detected for about 24 hours in the detector. For the size fractionation of  $^7\text{Be}$ -laden aerosols, aerodynamic size distribution of  $^7\text{Be}$  in the atmospheric aerosols were determined, using an 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  $0.4\ \mu\text{m}$  collection plate. These filters and stainless steel plates were then measured for the activity concentrations and size fractionation respectively of  $^7\text{Be}$ -laden aerosols.

The mean values of residence times of  $^{210}\text{Pb}$  and  $^7\text{Be}$  laden aerosols in the surface level air thus obtained from different methods in this work are  $5.29\pm 0.91$  days and  $7.18\pm 0.97$  days respectively. These MRT values agree well with the reported MRT values and are fully acceptable for studying atmospheric transport phenomena through surface level air of the region.

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Statistics of the  $^{210}\text{Pb}$ ,  $^{214}\text{Pb}$  and  $^7\text{Be}$  concentrations ( $\text{mBq m}^{-3}$ ) and MRT in the air samples of Islamabad.

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