Measurement of Bithmuth (²¹⁴Bi) in Indoor Air and Evaluation of Deposition Fraction

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Abstract: Activity size distribution of unattached as well as attached 214 Bi to aerosol particles was estimated in indoor air of physics department at Minia University, Minia City, Egypt. The samples were collected using a wire screen diffusion battery technique and a low pressure Berner cascade impactor. The mean Activity Median Thermodynamic Diameter (AMTD) of unattached 214 Bi was determined to be 1.25 nm with a relative mean Geometric Standard Deviation (GSD) of 1.29. A mean unattached fraction (f_{un}) of 0.08 ± 0.05 was obtained. The average activity concentration of 214 Bi was found to be 4.9 ± 0.42 Bq m⁻³. Most of the attached activities of 214 Bi were associated with aerosol particles of the accumulation mode. The GSD of the accumulation mode for 214 Bi was determined to be 3 with an Active Median Aerodynamic Diameter (AMAD) of 350 nm. Based on the obtained measured data values, deposition fraction of 214 Bi has been evaluated by using a stochastic deposition model. The bronchial deposition efficiencies of particles in the size range of attached 214 Bi were found to be lower than those of unattached progeny.

Keywords: Attached and unattached radon progeny, Deposition fraction, activity size distribution.

1. Introduction

Deposition theoretical modeling of radon containing aerosol in human lung represents a useful tool to interpret health effects from inhaled particular and to study the effectiveness of different inhalation procedure. Deposition is the process that determines what fraction of the inspired particles is caught in the respiratory tract and, thus, fails to exit with expired air. It is likely that all particles that touch a wet surface are deposited, thus, the site of contact is the site of initial deposition. Distinct physical mechanisms operate on inspired particles move them toward respiratory tract surfaces. Major mechanisms are inertial forces, gravitational sedimentation, Brownian diffusion, interception and electrostatic forces.

A major factor governing the effectiveness of the deposition mechanisms is the parameters of the activity size distribution (active median thermodynamic diameter, AMTD, active median aerodynamic diameter, AMAD, geometric standard deviation, GSD) of unattached and attached radon progeny in indoor air. Radon progeny have a particular size as molecular species but then attach to particles with a wide range of sizes. Since size helps to determine particles within the size range that can penetrate the oropharynx. The physics of air flow in human airways and the resulting deposition pattern of inhaled aerosol particles throughout the respiratory tract are determined by the size of the inhaled particles and by the breathing pattern for a given airway generation [1]. These airways form the trachea to the alveolar sacs form a structure similar to the crown of tree. Physical and mathematical of these models are usually based on airway geometry which approximated by a sequence of straight cylindrical tubes [2-7]. These models always require some information about the parameters of activity size distribution. Therefore, it is necessary to measure the activity size distribution of radon progeny, ²¹⁴Bi, to compute the deposition fraction of ²¹⁴Bi through the human lung.

The activity size distribution of radon progeny has been determined by tagging the natural aerosol particles with radon progeny. However, some measurements of radon progeny, for attached and unattached fraction, have been performed in indoor and atmospheric air [8-38]. Most of the observed data of these literatures shown that the size distribution consisted of ultrafine clusters with median diameters between 0.5 and 5 nm (unattached activity) and progenies associated with ambient aerosol particles in sizes ranging between 100 and 500 nm (attached activity). The unattached fraction is deposited nearly completely in the respiratory tract during inhalation, whereas 80 percent of the attached are exhaled without deposition. The amount of unattached activities up to about 10 percent of the total activity, but is considered to yield about 50 percent of the total radiation dose [39].

Wire screens are commonly used to estimate unattached radon progeny fractions. The use of wire screens for the separation of the unattached fraction was first studied by James et al [40] Tomas and Hinchcliffe [41] and George [42]. Measurements of activity size distribution of the unattached and attached ²¹⁴Bi show that the unattached fraction can be well separated from aerosol particle attached activities ([8–38]. For the measurement of activity size distribution of attached aerosol particles, cascade impactors are commonly used. Low pressure cascade impactors are useful for the size separation of small particles, where the low pressure effect reduces the drag force on the particle. This reduction in drag allows smaller particle sizes to be collected in low pressure impactors compared with impactors which are operated at the normal pressure (sierra impactor).

Based on obtained measurements, deposition fraction of radon progeny has been evaluated by using a stochastic deposition model [5]. It has been also discussed the effect of radon progeny deposition fraction by adult male for various levels of physical exertion.

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2. Materials and Methods

2.1. Attached activity fraction

A low pressure Berner cascade impactor (Model, 20/0.015) was used to determine the activity size distribution of attached short-lived radon progeny (214 Bi) [43]. It consisted of eight size fractionating stages and a back-up filter holder, [44] and operated at a flow rate of 1.7 m³ h⁻¹. Aluminum foils were used as collection media and a glass fiber filter as the backup filter. Therefore, the collection efficiencies as well as the cut-off diameters of the impactor stage could be calculated. The measured 50% cut-off diameters were 82, 157, 270, 650, 1100, 2350, 4250 and 5960. The collected activity on each impactor stage was measured with a well-type 3×3" NaI (Tl) detector. The total inters stage losses of aerosol particles were less than 2% of the total activity [43] see figure.1. More description about this procedure was given by Mohamed [45]-[46].



Figure.1. Air sampling, using Berner impactor, vacuum pump and Setup of gamma spectroscopy

2.2. Unattached activity fraction

In the present work, a wire screen diffusion battery similar to that employed and calibrated by Cheng et al [47] was used. It was constructed with the same screen characteristics to determine the size distribution of unattached radon progeny. The diffusion battery consisted of five stainless-steel screens with 24, 35, 50, 200, and 635 mesh numbers. The screens were calibrated with monodisperse silver aerosol particles. The measured 50% cut-off diameters of the screens are 0.9, 1.3, 1.9, 4.0 and 7.9 nm. Complete setup explained in figure.2. More description about this procedure was given by Mohamed [45]-[46].

2.3. Log-normal distribution parameters

The parameters of the attached and unattached activity size distribution, Active Median Aerodynamic Diameter (AMAD), Active Median Thermodynamic Diameter (AMTD) and Geometric Standard Deviation (GSM), were obtained from the lognormal distribution method by Hinds, [48] as:

$$\ln AMD = \frac{\sum n_i \ln d_i}{\sum n_i}$$

$$\ln(GSD) = \left[\frac{\Sigma n_i (\ln d_i - \ln AMD)^2}{\Sigma n_i}\right]^{\overline{2}}$$
(2)

1

Where AMD is the Active Median Diameter, n_i is the fraction in stage or screen i, d_i is the cutoff diameter of the stage or screen i and GSD is the Geometric Standard Deviation.



Figure 2: The screen diffusion battery, filter holder and surface barrier detector arrangement for the measurements of the unattached fractions of radon progeny

Deposition of inhaled aerosols in any given region of the human respiratory tract depends upon particle size, shape, density and subject breathing pattern. Deposition calculations here were based on the following set of aerosol characteristics pattern. Each component or mode of the radon progeny aerosol was assumed to be represented by a log normal distribution of activity with particle size, in which the geometric standard deviation (GSD) is related to the activity median diameter (AMD) by [23].

$$GSD = 1 + 1.5[1 - (100AMD^{1.5} + 1)^{-1}]$$
(3)

In the present investigation, the AMD of the size distributions were 20, 150, 250 and 350 nm for attached radon progeny, while for unattached fraction was 1.25 nm. The standard deviation was calculated for each corresponding AMAD from equation (1). In the present work, deposition fractions represent aerosol mass fraction deposited within specified regions during a complete breathing cycle (consists of equal inspired and expired times, 2 sec for each with no pause) normalized to the mass entering the trachea.

A total lung volume 3000 cm³, a tidal volume (V_t) 1000 cm³ and a spherical particles having a density equal 1g cm⁻³ were considered. Airway geometry was selected randomly and deposition was calculated deterministically by using the deposition formulas which were described by Koblinker and Hofmann [5]. Ventilation rates were taken as 0.54 and 1.5 m³ h⁻¹ for rest and light exercise, respectively. These values have been substituted in the stochastic deposition model to calculate the amounts of radon progeny deposited in each airway generation, as function of aerosol size and breathing rate, for each subject.

(1)

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3. Results and Discussions

Total activity concentration (C) of short-lived radon progeny 214 Bi, Activity median aerodynamic diameter in nm (AMAD), relative geometric standard deviation (GSD) of attached log-normal size distribution of 214 Bi, activity median thermodynamic diameter (AMTD), relative geometric standard deviation (GSD) of unattached log-normal size distribution of 214 Bi and the unattached log-normal size distribution of 214 Bi and the unattached fraction (f_{un}) of short-lived radon progeny measured in indoor air of physics department, Minia university, Minia City, Egypt are listed in Table 1.

The mean activity concentration of ²¹⁴Bi was found to be 4.9 \pm 0.42 Bq m⁻³ with range 0.46 to 16.65 Bq m⁻³. Frequency of activity concentration of ²¹⁴Bi is shown in figure.3. The Active Median Aerodynamic Diameters, AMAD, of the accumulation mode for ²¹⁴Bi was found to be varied between 253and 427 nm with mean value 350 nm and with corresponding Geometric Stander deviation, GSD, of 3.00 with range from 2.58 to 3.59. The activity size distribution of the attached fraction of ²¹⁴Bi is shown in Figure. 4. The attached activity fractions are plotted as histograms vs the cut-off diameters of the stages. It's clear that all the activity size distributions of the attached ²¹⁴Bi could be approximated as unimodal log normal distributions. The attached particles of the accumulation mode (size: 100-2000 nm) were found to be the dominant part of the measured activity size distributions.

From Table.1 the mean values of AMTD of 214 Bi were determined to be 1.25 varied between 1.06 and 1.47 nm with mean GSD of 1.29 ranges from 1.19 to 1.37. The unattached fraction (fun) varied between 0.01 and 0.21 with a mean value of 0.08±0.05%. The activity size distributions of the unattached 214 Bi are shown in Figure.5. The unattached activity fractions collected in each wire screen are plotted as histograms against the cutoff diameter.



Figure 3: Activity concentration Frequency of ²¹⁴Bi

Table 1: Total activity concentration of ²¹⁴ Bi, the parameters
of unattached and attached ²¹⁴ Bi activity size distribution
(AMAD, AMTD, GSD) and unattached fraction, fun, in
indoor air in physics department, Minia university.

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No	C Bq m ⁻³	AMAD nm	GSD	AMTD nm	GSD	\mathbf{f}_{un}
1	2.10	427.27	2.83	1.09	1.29	0.03
2	4.23	261.91	3.18	1.21	1.22	0.06
3	1.79	372.68	3.08	1.17	1.37	0.05
4	1.61	405.55	3.02	1.43	1.27	0.03
5	3.38	362.44	3.09	1.23	1.21	0.04
6	6.15	361.31	2.58	1.06	1.19	0.01
7	16.65	363.76	3.00	1.33	1.26	0.09
8	2.63	390.35	3.32	1.44	1.37	0.01
9	4.87	291.23	2.64	1.09	1.22	0.08
10	11.98	314.97	3.00	1.35	1.36	0.036
11	4.49	322.69	2.78	1.10	1.31	0.08
12	5.39	380.97	3.59	1.44	1.37	0.03
13	6.81	253.17	2.71	1.16	1.25	0.07
14	3.95	368.29	2.97	1.47	1.23	0.06
15	3.24	347.31	2.84	1.20	1.31	0.1
16	0.46	371.62	3.16	1.18	1.34	0.16
17	6.81	320.56	2.90	1.31	1.32	0.21
18	2.94	367.31	3.30	1.39	1.30	0.12
19	3.26	322.77	2.67	1.06	1.23	0.13
20	4.70	386.16	3.19	1.19	1.30	0.11
	4.9±0.4	350 nm	3.00	1.25 nm	1.29	0.08±0.05



Figure 4: Activity size distribution of attached ²¹⁴Bi in indoor air



Figure 5: Activity size distribution of unattached ²¹⁴Bi in indoor air

Figure 6 represent the variation in deposition fraction with particle size that is predicted for each airway generation of the adult male lung over the size range of concern for deposition of radon progeny. Figure 6 relate the number of particles deposited in each airway generation to the number that enter the trachea on inhalation. It can be seen that the deposition fraction of particles with diameter of 1.4 nm is calculated to be uniformly high throughout the bronchi (generation 1 to 8). By the time the inspired air reaches the bronchioles (generation 9-15), the number of airborne 1.4 nm particles available for deposition was low. Therefore, deposition is found to decrease rapidly in succeeding generations of the bronchiolar airways.



Figure 6: Deposition fraction calculated with varying particle diameters

If the particle size is increased to 20 nm, the deposition fraction is found to be about an order of magnitude lower in the bronchi and reach a broad beak in the human respiratory airways. Particles of this size are typical of the radon progeny growth mode produced by human activities such as cooking or vacuum cleaning (23). It is assumed that the size distributions of both unattached and growth modes of radon progeny are not affected by the humid environment of the respiratory tract (23). The radon progeny aerosol produced by cooking/vacuuming has three size modes (5 % of potential alpha energy is unattached, 15 % has an AMAD of 20 nm and 80 % has an AMAD of 150 nm). The 20 nm mode is

hydrophobic and does not increase in the size within the respiratory tract [23].

The bronchial deposition fraction of particles in the size range of attached radon progeny were found to be about two orders of magnitude lower than those of unattached progeny. This clears the disproportionately large contributions to the dose from exposure to small fraction of radon progeny in the unattached state. The attached or so-called accumulation mode of the radon progeny aerosol has a median size in ambient air that ranges from about 150 to 400 nm diameter17. However, the carrier aerosol particles are considered to be partly hygroscope and grow in the respiratory tract to about double their ambient size. This figure also compares the profiles of deposition throughout the lung that were calculated for particles that attain equilibrium sizes of 250 and 350 nm within the respiratory tract. It is seen that deposition is expected to be relatively independent of particle size over this limited range (at least in resting subjects) [49].

Another significant factor that must be accounted for in calculating the deposition profile of radon progeny within the respiratory tract is the typical variability or dispersion in size of the aerosol particles. Unattached radon progeny have a relatively narrow distribution of particle size, whereas the activity-size distribution of progeny attached to ambient aerosol particles is typically broad.

4. Conclusion

- The mean AMTD of the unattached fraction of ²¹⁴Bi was found to be 1.25 nm with a mean GSD of 1.29.
- The mean AMAD of the attached fraction of ²¹⁴Bi was found to be 350 nm with a mean GSD of 3.
- The unattached fraction (fun) of short lived radon progeny was found to be 0.08±0.05.
- The obtained parameters of the activity size distribution (AMTD, AMAD and GSD) are necessary to calculate the deposition through the human lung.
- For best estimation of doses, an accurate data of these parameters are necessary. Simultaneous measurements ²¹⁴Bi in different environments (outdoor, indoor, mines) are necessary in order to model time-dependent behaviors, deposition, attachment and to determine particle size distribution.

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