Radon Decay Product Aerosols in Ambient Air

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ABSTRACT

The aerodynamic size distributions of radon decay product aerosols, i.e. $^{214}$Pb, $^{212}$Pb, and $^{210}$Pb were measured using low-pressure (LPI) as well as conventional low-volume 1-ACFM and high-volume (HVI) cascade impactors. The activity size distribution of $^{214}$Pb and $^{212}$Pb was largely associated with submicron aerosols in the accumulation mode (0.08 to 2.0 $\mu$m). The activity median aerodynamic diameter “AMAD” varied from 0.10 to 0.37 $\mu$m (average 0.16 $\mu$m) for $^{214}$Pb-aerosols and from 0.07 to 0.25 $\mu$m (average 0.12 $\mu$m) for $^{212}$Pb-aerosols. The geometric standard deviation, $\sigma_g$ averaged 2.86 and 2.97, respectively. The AMAD of $^{210}$Pb-aerosols varied from 0.28 to 0.49 $\mu$m (average 0.37 $\mu$m) and the geometric standard deviation, $\sigma_g$ varied from 1.6 to 2.1 (average 1.9). The activity size distribution of $^{214}$Pb-aerosols showed a small shift to larger particle sizes relative to $^{212}$Pb-aerosols. The larger median size of $^{214}$Pb-aerosols was attributed to $\alpha$-recoil depletion of smaller aerosol particles following the decay of the aerosol-associated $^{218}$Po. Subsequent $^{214}$Pb condensation on all aerosol particles effectively enriches larger-sized aerosols. Pb-212 does not undergo this recoil-driven redistribution. Even considering recoil following $^{214}$Po $\alpha$-decay, the average $^{210}$Pb-labeled aerosol grows by a factor of two during its atmospheric lifetime. Early morning and afternoon measurements indicated that similar size associations of $^{214}$Pb occur, despite humidity differences and the potential for fresh particle production in the afternoon. In estimating lifetimes of radon decay product aerosols in ambient air, a mean residence time of about 8 days could be applied to aerosol particles in the lower atmosphere below precipitation cloud levels.

Keywords: Radon; Decay products; $^{214}$Pb; $^{212}$Pb; $^{210}$Pb; Radioactive aerosols; Cascade impactors; 1-ACFM; AMAD; Residence time.

INTRODUCTION

The decay of radon, $^{222}$Rn and thoron, $^{220}$Rn in the atmosphere produces low vapor pressure decay products that coagulate with other nuclei or condense on existing aerosol particles. These decay products include $^{218}$Po (3.0 min), $^{214}$Pb (26.8 min) and $^{212}$Pb (10.64 h). A long-lived decay product in the $^{222}$Rn decay chain, $^{210}$Pb (22.3 y) is produced about an hour after attachment of $^{218}$Po onto aerosol particles. Very little data exist on the ambient aerosol associations of the decay products of both radon isotopes with respect to their aerodynamic size distributions relative to the formation and growth mechanisms of aerosols.

Fig. 1 presents the decay scheme for radon, $^{222}$Rn and thoron, $^{220}$Rn, illustrating the short-lived as well as the long-lived decay products, their mode of radioactive decay and their half-lives.

Diffusion batteries have been used to investigate aerosol size associations of gross $\alpha$-activity from the short-lived radon decay products (Soilleux, 1970; George and Breslin, 1980; George et al., 1984) in outdoor air situations, and high-volume cascade impactors for gross $\beta$- and $\gamma$-activity from the long-lived radon and thoron decay products (Röbig et al., 1980). The residence time of atmospheric aerosol particles in the lower atmosphere assuming that the air in the troposphere is considered as a well-mixed reservoir (closed system) is a function of various removal processes, the most important being:

\begin{enumerate}
  \item dry deposition by impaction, diffusion and sedimentation,
  \item wet deposition by rain drops (precipitation scavenging) as a result of processes occurring both within and below the rain cloud.
\end{enumerate}

The residence time of atmospheric aerosol particles can be estimated by means of radioactive nuclides as tracers, which become attached to aerosol particles and are removed with them as they are scavenged by precipitation or undergo dry fallout. Several methods have been used for estimating the mean residence time of atmospheric aerosol particles. These include measurements of the activities and ratios of radioactive decay products of radon, $^{222}$Rn which emanate from continental surfaces into the atmosphere, such as $^{210}$Pb (22.3 y), $^{210}$Bi (5.01 d) and $^{210}$Po (138.38 d) (Francis et al., 1970; Poet et al., 1972; Lambert et al., 1980; 1983; Marley et al., 2000, Baskaran and Shaw, 2001). However, there is disagreement between the

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![Decay schemes of $^{222}$Rn and $^{220}$Rn illustrating short-lived and long-lived decay products.](image)
derived values of the residence times due to various processes, including the fact that they refer to different portions of the atmosphere.

This paper summarizes results of an investigation designed to characterize the aerodynamic size distributions and the residence time of atmospheric aerosols in the context of\(^{222}\text{Rn}\) and \(^{220}\text{Rn}\) decay products for better understanding of how radioactive aerosols behave in the atmosphere, and of the aerosol growth mechanisms.

**EXPERIMENTAL METHODS**

Measurements on aerodynamic sizes of atmospheric aerosols associated with radioactive nuclides, such as radon decay products \(^{214}\text{Pb}, {212}\text{Pb}\) and \(^{210}\text{Pb}\) were carried with Anderson type 1-ACFM ambient cascade impactors with or without the Andersen model 2000 low-pressure modification, as well as with Sierra model 236, six-stage high-volume cascade impactors, HVI.

The 1-ACFM design operated at 28 L/min (1 ft\(^3\)/min). The eight stages had effective cutoff diameters, ECDs of 0.4, 0.7, 1.1, 2.1 3.3, 4.7, 7.0 and 11.0 \(\mu\)m. The low-pressure modification, which alters the impactor’s operation by increasing the resolution in the submicron region, involves a regulated flow rate of 3 L/min, five low-pressure (114 mm Hg) stages for the submicron region and eight atmospheric pressure stages for separating aerosol particles above 1.4 \(\mu\)m. The ECDs of the low-pressure stages were 0.08, 0.11, 0.23, 0.52 and 0.90 \(\mu\)m, whereas for the upper stages they were 1.4, 2.0, 3.3, 6.6, 10.5, 15.7, 21.7 and 35.0 \(\mu\)m. A schematic diagram of a low-pressure cascade impactor, LPI complete system is shown in Fig. 2. The stainless steel plates supplied by the manufacturer were used for aerosol particle collection. Either polycarbonate or glass-fiber backup filters were used to collect all particles below the 0.08-\(\mu\)m collection plate.

The length of each collection period varied from about 3 h for \(^{214}\text{Pb}\) to 30 or 40 h for \(^{212}\text{Pb}\) with low-pressure impactor measurements and from 1 to 24 h for \(^{210}\text{Pb}\) and \(^{210}\text{Bi}\) for high-volume impactor measurements. The samples were collected 13 m above the ground on the roof of the Environmental Sciences Division building at the Oak Ridge National Laboratory, ORNL, in Oak Ridge, Tennessee (35°58'N, 84°30'W).

The deposits on the stainless-steel collection plates of the low-pressure cascade impactor were leached with a solution of 1 M HNO\(_3\) and the leachate rapidly evaporated on 5.08-cm stainless steel plates using a hot plate. The concentrations of \(^{214}\text{Pb}\) and \(^{212}\text{Pb}\) in the impactor plate leachates were measured using seven ZnS (Ag) alpha scintillation counters with backgrounds averaged 0.025 cpm. The 1 \(\sigma\) counting uncertainty was 5% or less for the stages corresponding to aerosols below 0.52 \(\mu\)m and < 15% for the stages collecting to aerosols above 0.52 \(\mu\)m. The calculated activities of \(^{212}\text{Pb}\) and \(^{211}\text{Pb}\) were used to determine the activity size distribution of each radionuclide and then the activity median aerodynamic diameter “AMAD” of the \(^{214}\text{Pb}\) and \(^{212}\text{Pb}\)-aerosols. The high-volume impactor plates were leached with 0.1 M HCl.

The activity of \(^{210}\text{Pb}\) and \(^{210}\text{Bi}\)-associated aerosol particles was measured by a low-background phoswitch scintillation detector system having a background of 2 cpm and efficiency higher than 40% for counting \(\beta\)-radiation. This system consisted of a thin CaF\(_2\) (Eu) primary crystal with a decay time of 0.23 \(\mu\)s. The samples were counted for long enough to obtain a statistical accuracy better than 5%. It must be noted that \(^{210}\text{Pb}\) and \(^{210}\text{Bi}\) were chemically separated and measured for their activities. Detailed description of the analytical method and technique was established and presented elsewhere (Jaworowski, 1963). The activity of \(^{210}\text{Pb}\) was also measured by a surface barrier Ge detector through its gamma-ray peak of 46.5 keV. The activity size distribution and the AMAD of the \(^{210}\text{Pb}\)-aerosol particles were determined upon the measurement of the activities of the aerosol-associated radioactive nuclides. Fig. 3 shows a typical gamma-ray spectrum of a glass fiber air filter obtained by a Ge detector, in which the 46.5 keV \(\gamma\)-ray peak of \(^{210}\text{Pb}\) is clearly shown.

![Fig. 2. A schematic diagram of a low-pressure cascade impactor complete system.](image-url)
RESULTS AND DISCUSSION

Activity Size Distribution of Radon Decay Product Aerosols

Fig. 4 shows representative plots from 46 low-pressure impactor measurements made over a ten-month period, illustrating the aerodynamic size distributions of 214Pb- and 212Pb-aerosols. R is the radioactivity and Dp is the particle diameter, in μm.

About 46% of the measurements showed a radioactivity peak in the 0.11- to 0.23-μm region (subplot a), while 39% showed a peak in the 0.23- to 0.52-μm region (subplot b). The remaining 15% of the measurements resulted in activity size distributions similar to those in subplot c (8.7%) or subplot d (6.5%), where 214Pb- and 212Pb-activities were highest in different size ranges in the same spectrum. On the average, about 76% of the 214Pb-activity and 67% of the 212Pb-activity was found to be associated with aerosol particles in the 0.08- to 1.4-μm size range. The activity-associated aerosol particles smaller than 0.08-μm can also be substantial, as indicated in Fig. 4. Fig. 4 illustrates a peak in the 0.23- to 0.52-μm region (subplot b). The remaining 15% of the measurements resulted in activity size distributions similar to those in subplot c (8.7%) or subplot d (6.5%), where 214Pb- and 212Pb-activities were highest in different size ranges in the same spectrum. On the average, about 76% of the 214Pb-activity and 67% of the 212Pb-activity was found to be associated with aerosol particles in the 0.08- to 1.4-μm size range. The activity-associated aerosol particles smaller than 0.08-μm can also be substantial, as indicated in Fig. 4. Fig. 4 illustrates that 214Pb is generally enriched in the accumulation mode aerosol, particularly between 0.11 and 0.52 μm, where most of the surface area and mass occurs, while the Aitken nuclei fraction (below 0.08 μm) contained a higher percentage of 212Pb-activity compared with 214Pb-activity in 69.6% of the measurements.

The radon decay products become associated with atmospheric aerosol particles by condensation or coagulation processes that are surface-area related. The activity size distribution of 214Pb-aerosols showed a small shift to larger particle sizes relative to 212Pb-aerosols. The shift of 212Pb to a slightly higher size distribution compared to 214Pb was also found using 1-ACFM and high-volume cascade impactors (Fig. 5). The higher flow rates of these impactors, as well as the ability to measure high-volume activity by gamma-ray spectrometry, made us confident that this shift was real and not a data analysis artifact. The larger median size of 212Pb-aerosols was attributed to α-decay depletion of smaller aerosol particles following the decay of the aerosol-associated 218Po. Subsequent 218Po condensation on all particles effectively enriches larger-sized aerosols. Pb-212 does not undergo this recoil-driven redistribution. Even considering recoil following 212Po to α-decay, the average 210Pb-labelled aerosol grows by a factor of two during its atmospheric lifetime. Early morning and afternoon measurements indicated that similar size associations of 214Pb occur, despite humidity differences and the potential for fresh particle production in the afternoon.

The longer half-life of 212Pb (10.64 h) compared to 214Pb (26.8 min) might favor the presence of larger aerosol associations of 212Pb if coagulation rates are fast relative to radioactive decay rates, although cascade impactor measurements may not be sensitive enough to record this effect. Instead, the measurements indicated that the shorter-lived chain (218Po, 214Pb, etc) was more often associated with larger aerosol particle sizes than the longer-lived chain (218Po, 212Pb, etc). The high-volume impactor measurements reported by Röbig et al. (1980) also indicated a large particle shift of 214Pb relative to 212Pb (Fig. 6). They stated that this was due to shifts of the activity size distributions from night to day-time to smaller particle sizes by photochemical reactions and to shifts to larger particle sizes when the relative humidity increases during rainfall, and therefore depends on sampling time. They also observed that the activity size distributions of the long-lived radionuclides 214Pb and 212Pb are shifted to larger particle sizes compared with the activity size distributions of the shorter-lived 212Pb and 214Pb. That shift was due to larger residence times for 210Pb and 212Pb in the atmosphere and for 214Pb due to a different size distribution of stratospheric and tropospheric aerosols.

The 214Pb shift might be explained by the fact that a significant fraction of the 3.05-min 218Po atoms, precursor of 214Pb atoms, should attach to an existing aerosol particle or coagulate with other nuclei during their lifetime, as mean attachment half-lives are of the order of a minute less (Porstendörfer and Mercer, 1980). When this attached 218Po α-decays, the recoiling 214Pb atom, decay product of 210Pb, can escape the aerosol particles. Complete recoil loss would occur if the particle diameter, Dp of the aerosol were smaller than the range of the recoiling nucleus. In water, this recoil range is 0.13 μm (Mercer, 1976) and should be somewhat less in the atmospheric aerosols that have a density closer to 1.5 g cm⁻³ (Friedlander, 2000). By contrast, very little of the 0.146 μs-212Po would attach before decaying to 212Pb, because of its short life relative to attachment times. A considerable fraction of the 214Pb should undergo recoil detachment, particularly from aerosol particles with diameters smaller than 0.1 μm (diameter approximating the recoil range). The probability of loss would decrease with increasing particle radius (Mercer, 1976). If the recoiling 214Pb behaves like the original 214Po, the net effect would be a shift of 214Pb to a larger particle size distribution.

Since the subsequent β-decays of 212Pb and 214Pb + 214Bi do not result in significant recoil (Mercer, 1976), the α-measurement of...
Fig. 4. Aerodynamic size distributions of $^{214}$Pb- and $^{212}$Pb-aerosols obtained with low-pressure cascade impactors. R is the radioactivity, and $D_p$ is the particle diameter. (a) Results occurred 46% of the time, (b) 39% of the time, (c) 8.7% of the time, and (d) 6.5% of the time. Lower $D_p$ limits are arbitrary.

Fig. 5. Aerodynamic size distributions of $^{214}$Pb- and $^{212}$Pb-aerosols obtained with 1-ACFM and high-volume cascade impactors, illustrating the large particle shift of $^{214}$Pb. R is the radioactivity, and $D_p$ is the particle diameter. Lower $D_p$ limits are arbitrary.

$^{214}$Po and the $^{212}$Pb-decay products is in reality tracing the aerosol size distribution of a $^{220}$Rn-decay product atom ($^{212}$Pb) which has condensed only once and a $^{222}$Rn-decay product atom ($^{214}$Pb) that has probably condensed more than once. This stability of the Pb-isotopes is the basis for the generic reference to $^{212}$Pb and $^{214}$Pb size distributions.

The AMADs of $^{214}$Pb-aerosols determined with the low-pressure impactor data varied from 0.10 to 0.37 $\mu$m (average 0.16 $\mu$m) and for $^{212}$Pb-aerosols from 0.07 to 0.25 $\mu$m (average 0.12 $\mu$m) (Table 1). For the $^{210}$Pb-aerosols the AMADs varied from 0.28 to 0.49 $\mu$m (average 0.37 $\mu$m). Lead-210 is produced by $\alpha$-decay of $^{214}$Po, the event used to quantify $^{214}$Pb distributions on the low-pressure cascade impactors. While the relationship between the aerodynamic sizes of $^{211}$Pb and $^{210}$Pb is complicated because of the large differences in their atmospheric lifetimes, $^{210}$Pb has always found to be associated with aerosols larger than...
214Pb, as indicated by the differences in AMADs reported in Table 1. Table 1 shows data for the AMADs of the radon decay product aerosols. The results were obtained assuming lognormal distributions. The reported from the literature data for radon decay product aerosols are also presented in Table 1 and analyzed as follows:

Grundel and Porstendörfer (2004) more recently carried out measurements in outdoor air at a northern latitude (51°32'N, 9°55'E) in Göttingen, Germany using an on-line alpha cascade impactor (OLACI) for a period of about one month and showed that the activity size distributions of the short-lived radon (222Rn) and thoron (220Rn) decay products are about 12-19% in the accumulation mode (Aitken nuclei, 0.05-60 nm), 81-88% in the nucleation mode (particle diameter > 1000 nm). The AMAD of the accumulation mode varied between 332 nm for 210Po and 347 nm for 212Po for the short-lived radon decay products and between 382 nm for 210Po and 421 nm for 212Po for the thoron decay products. In comparison to the short-lived radon decay products, the AMADs of the thoron decay products were shifted significantly to larger values. For the long-lived radon (222Rn) decay products 210Pb and 212Pb they showed that these radionuclides are almost 93-96% adsorbed on aerosol particles in the accumulation size range and only 4-7% of their activities are attached on nuclei with diameters smaller than 60 nm. AMAD-values of 558 nm for 210Pb and 545 nm for 212Pb were measured, i.e. significantly larger than those of the short-lived radon and thoron decay products.

In El-Minia, Egypt (28°04'N, 30°45'E), El Hussein and Ahmed (1995) by using a low-pressure Berner-type cascade impactor showed that the activity size distributions of the 210Pb- and 214Bi-attached aerosols were nearly identical and that most of the activities were associated with aerosol particles of the accumulation mode. The mean AMAD of 210Pb-aerosols (range 190-620 nm) and 214Bi-aerosols (range 190-620 nm) had the same value of 380 nm, but the relative geometric standard deviation, $\sigma_g$, of the log-normal distribution of 210Pb-aerosols ($\sigma_g$: 1.67-2.25, average 2.05) shows a broader activity size distribution than for 214Bi-aerosols ($\sigma_g$: 1.67-2.25, average 2.05). Later, El Hussein et al. (1998) from measurements in El-Minia, Egypt, found that the AMAD, varied from 175 to 485 nm (average 330 nm) and the geometric standard deviation, $\sigma_g$, varied from 2.5 to 2.9 (average 2.7), while for its decay product 212Pb-aerosols the AMAD varied from 296 to 360 nm (average 320 nm) and the $\sigma_g$ varied from 2.1 to 2.88 (average 2.7), which means that they are quite similar. For the relatively longer-lived thoron decay product 212Pb-aerosols, the AMAD of the accumulation mode varied from 249 to 390 nm (average 360 nm) and the $\sigma_g$ varied from 2.1 to 3.2 (average 2.7) (Mohammed et al., 2000).

Winkler et al. (1998) by using a nine-stage low-pressure Berner-type cascade impactor in Neuherberg, Germany (48°13'N, 11°36'E) reported that the activity size distribution of 210Pb in ambient aerosols was unimodal (log-normal) and associated with submicron aerosols of about 0.5 to 0.6 $\mu$m. On average, the AMAD of 210Pb-aerosols (0.53 $\mu$m) has been found to be significantly lower than the average mass median aerodynamic diameter “MMAD” (0.675 $\mu$m), and higher than or at most equal to the respective surface median aerodynamic diameter “SMAD” (0.465 $\mu$m) of the aerosols, i.e. SMAD < AMAD < MMAD. Variation of the atmospheric processes resulted in a variability of the AMAD, between 0.28 and 0.74 $\mu$m for 210Pb-aerosols. While in the winter period (October to April) the AMAD values of 210Pb-aerosols averaged 0.595 $\mu$m, in the summer period 210Pb was associated with significantly smaller-sized aerosols (AMAD: 0.43 $\mu$m).

Suzuki et al. (1999) by using a 40-cfm high-volume cascade impactor with five stages reported that 77% of 210Pb- and 70% of 214Po-activities were measured in size-fractionated aerosols with a diameter smaller than 0.70 $\mu$m from the west coast of the Japanese Islands (38°46'N, 139°44'E).

Apart of this, Papastefanou (2008) estimated the AMAD of another radionuclide in ambient aerosols, that is 7Be ($T_\text{½} = 53.3$ days) of cosmogenic origin, varying between 0.76 and 1.18 $\mu$m (average 0.90 $\mu$m) in the accumulation mode of the activity size distribution of aerosol particles. The aerodynamic size
distribution of $^7$Be in atmospheric aerosols and then the AMAD was achieved by using Andersen 1-AFM cascade impactors, in association with a gamma-ray spectrometer linked with a Ge coaxial-type detector for the $^7$Be activity measurement of the collection plates and the back up filter of the impactor.

**Residence Times of Radon Decay Product Aerosols**

The method for estimating the residence time of atmospheric aerosol particles associated with the radon decay product radionuclides is based on the radioactivity of a pair of genetically related radioisotopes, such as $^{210}$Pb, $^{210}$Bi and or $^{210}$Pb, $^{210}$Po, according to the sequential disintegrations in the $\beta$-decay scheme

$$^{210}\text{Pb} (\beta^-, 22.3 \text{ y}) \rightarrow ^{210}\text{Bi} (\beta^-, 5.0 \text{ d}) \rightarrow ^{210}\text{Po} (\alpha, 138.38 \text{ d}) \rightarrow ^{206}\text{Pb} (\text{stable})$$

and the Eq. (2) by the formula

$$\tau_R = \frac{1}{\lambda_{Po} N_{Po} / \lambda_{Pb} N_{Pb}} \left( \frac{1}{\tau_R} + \frac{1}{\lambda_{Po}} - \frac{1}{\lambda_{Pb}} \right)$$

where $\lambda_{Po}$ is the decay constant of $^{210}$Po and $\lambda_{Pb}$ is the decay constant of $^{206}$Pb.

The residence time, $\tau_R$ is described by the formula

$$\tau_R = \frac{1}{\lambda_{Bi} N_{Bi} / \lambda_{Pb} N_{Pb}} - \left( \frac{1}{\lambda_{Bi} + \lambda_{Po}} \right) N_{Bi} = 0$$

where $\lambda_{Bi}$ is the first-order rate constant for the removal of aerosol particles from the atmosphere by all processes, that is the inverse of residence time, $\tau_R$.

The ratio of the activities $\lambda_{Bi} N_{Bi} / \lambda_{Po} N_{Po}$ in Eq. (2) varied from 0.40 to 0.68 (this work) or from 0.42 to 0.85 (Moore et al., 1972). The reported from the literature $\tau_R$ values are also shown in Table 2 and analyzed as follows:

Poet et al. (1972) estimated tropospheric aerosol residence times ranging from 1.59 to 13 days (average 5.4 days) when based on the $^{210}$Bi/$^{210}$Pb activity ratios and from 11 to 77 days (average 24 days) when based on the $^{210}$Po/$^{210}$Pb activity ratios for twenty measurements of aerosol samples carried out in surface air during a 4 ½-year period at Boulder, Colorado (40°01'N, 105°17'W). They concluded that, a mean tropospheric residence time of about 4 days could be applied for aerosol particles in the lower troposphere and about a week for aerosol particles in precipitation. They also found that the mean aerosol residence time increases with altitude within troposphere less than by a factor of 3 (Moore et al., 1973).

Francis et al. (1970) examined aerosol residence times for $^{210}$Po of 9.6 days ± 20% based on the $^{210}$Po/$^{210}$Pb activity ratios from the dust of filtrate (> 0.22 μm) collected during a 3-month period (May-August) at Madison, Wisconsin (43°05'N, 89°22'W).

Marley et al. (2000) estimated residence times for seven aerosol samples collected at Argonne, Illinois (41.7°N, 88.0°W) during a 2-year period, three aerosol samples collected at Phoenix, Arizona, one sample collected at Socorro, New Mexico, and five samples collected at Mexico City, Mexico. Based on the $^{210}$Bi/$^{210}$Pb activity ratios, they resulted in residence times of 6 to 67 days, while based on the $^{210}$Po/$^{210}$Pb activity ratios, they suggested residence times of 33 to 66 days for the aerosols below 2 μm in size.

Much earlier, Fry and Menon (1962) in twelve measurements carried out during a 7-month period (spring and summer) at Fayetteville, Arkansas (36°03'N, 79°54'W), indicated apparent aerosol residence times ranging from 2.4 to 25.6 days (average 8.5 days), based on the $^{210}$Bi/$^{210}$Pb activity ratios in Arkansas rains. Gavini et al. (1974) also estimated residence times varied from 3 to 240 days (average 29 days) from the $^{210}$Bi/$^{210}$Pb activity ratios in Arkansas rains also.

Baskaran and Shaw (2001) based on the $^{210}$Po/$^{210}$Pb activity ratios estimated residence times of arctic haze aerosols, from the upper atmosphere to the Arctic atmosphere, varied between 12 and 32 days from the measurements carried out during an 1 1/2 – year period (winter) at Poker Flat (65.1°N, 147.5°W) and between 0 and 39 days from eight measurements carried out during a 2-month period (winter) at Eagle (69.5°N, 141.2°W) in Alaska.

Very early, Lehman and Sitkus (1959) estimated aerosol residence times of 20 days from the $^{210}$Po/$^{210}$Pb activity ratios in air at Freiburg, Germany (47°59'N, 7°51'E), while Peirson et al. (1966) estimated aerosol residence times of 40 days from the $^{210}$Po/$^{210}$Pb activity ratios in air at Milford Haven, Wales (51°40'N, 5°02'E). Lambert et al. (1980) estimated aerosol
residence times varied from 8.8 to 10.5 days based on the 210Bi/210Pb activity ratios in air, while later in another work they estimated aerosol residence times varied from 7 to 9 days (average 8.58 days) based on the 210Bi/210Pb activity ratios in air, in near Paris, France area during a 6-year period (Lambert et al., 1983).

Gäggeler et al. (1995) during an one-year period carried out measurements on the activities of 210Pb, 212Pb and 222Rn in air at high altitude in Jungfraujoch, Switzerland (46°32'N, 7°59'E) as high as 3450 m by using an epiphaniometer. They estimated aerosol residence times varied between 1 and 12 days (average 6 days) from the 210Bi/210Pb activity ratios.

Finally, Rangarajan (1992) based in the 210Bi/210Pb activity ratios in air estimated a mean residence time of the natural radioactive aerosols in the planetary boundary layer at Bombay (Mumbai), India (18°58’N, 72°50’E) around 8 days from forty three measurements carried out during the long dry period, from October to May.

The data of Table 2 admit residence times of lower values as low as 1.59 days and higher values as high as 320 days. Mostly, the lower τA-values were resulted from the 210Bi/210Pb activity ratios and higher τA-values from the 222Rn/210Pb activity ratios. Low τA-values were also resulted from the 222Rn/210Pb activity ratios. Poet et al. (1972) concluded that longer apparent residence time values were based on the 210Po/210Pb activity ratios, than those based on the 210Bi/210Pb activity ratios. The difference may be explained by the presence of a mixture of aerosols of various apparent ages, of which older aerosols (those of greater ages) contribute most of 210Po. On the other hand, the solid products of radon-decay, that is 210Po, 210Bi and 210Po might arise from sources other than radioactive decay within the atmosphere. Poet et al. (1972) showed that up to 85% of the 210Po in the atmosphere is of terrestrial origin, and the vertical profile of 210Po was found to differ appreciably from that expected from the decay of 222Rn. Lambert et al. (1979) indicated that volcanic gases are very rich in long-lived radon decay products, especially in 210Po relative to 210Pb. Soil particles are the most likely contributors, since a part of the tropospheric aerosols originates at the Earth’s surface. Coal burning and forests fires presumably are additional sources of radionuclides.

Apart of this, Papastefanou (2008) estimated the residence times of another radionuclide in ambient aerosols, that is 1Be, while for the 212Pb-aerosols, the mean AMAD was found to be 0.16 m diameter particles in size. Based on the 46% low-pressure cascade impactors, the mean activity median aerodynamic diameter “AMAD” of 214Pb-aerosols was found to be 0.12 m, while for the 212Pb-aerosols, the mean AMAD was found to be 0.12 m. The slightly larger size of 214Pb-aerosol particles confirmed with operationally different cascade impactors, was attributed to α-recoil-driven redistribution of 214Pb following the decay of the aerosol-associated 214Po. Even considering recoil following 214Po α-decay, the average 212Pb-labelled aerosols grow by a factor of two during its atmospheric lifetime. Estimated lifetimes of radon decay product aerosols in ambient air resulted in a mean residence time of about 8 days that could be applied to aerosol particles in the lower atmosphere below the boundary layer.

### Table 2. Residence times, τA, of tropospheric aerosols (days).

<table>
<thead>
<tr>
<th>Investigation</th>
<th>210Bi/210Pb</th>
<th>210Po/210Pb</th>
<th>222Rn/210Pb</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oak Ridge, Tennessee (35°58’N, 84°30’W)</td>
<td>4.8-15.3 (avg. 8.2)</td>
<td>-</td>
<td>-</td>
<td>This work</td>
</tr>
<tr>
<td>Boulder, Colorado (40°01’N, 105°17’W)</td>
<td>1.59-13.0 (avg. 5.4)</td>
<td>11-37</td>
<td>-</td>
<td>Poet et al. (1972)</td>
</tr>
<tr>
<td>Argonne, Illinois (41.7°N, 88.0°W)</td>
<td>6-67</td>
<td>33-66</td>
<td>2.2,3.4</td>
<td>Moore et al. (1972)</td>
</tr>
<tr>
<td>Madison, Wisconsin (43°05’N, 89°22’W)</td>
<td>-</td>
<td>9.6</td>
<td>-</td>
<td>Marley et al. (2000)</td>
</tr>
<tr>
<td>Fayetteville, Arkansas (35°3’N, 78°54’W)</td>
<td>2.4-25.6 (avg. 8.5)</td>
<td>-</td>
<td>-</td>
<td>Fry and Menon (1962)</td>
</tr>
<tr>
<td>oker Flat, Alaska (65.1°N, 147.2°W)</td>
<td>3-240 (avg. 20)</td>
<td>2-320 (avg. 40)</td>
<td>-</td>
<td>Gavini et al. (1974)</td>
</tr>
<tr>
<td>Eagle, Alaska (69.5°N, 141.2°W)</td>
<td>-</td>
<td>0-38.9</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Jungfraujoch, Switzerland (46°32’N, 07°59’E)</td>
<td>-</td>
<td>1-12 (avg. 6)</td>
<td>-</td>
<td>Gäggeler et al. (1995)</td>
</tr>
<tr>
<td>Freiburg, Germany (47°59’N, 7°51’E)</td>
<td>-</td>
<td>20</td>
<td>-</td>
<td>Lehmann and Sittkus (1959)</td>
</tr>
<tr>
<td>Gif-sur-Yvette, France (48°52’N, 2°20’E)</td>
<td>8.8-10.5</td>
<td>-</td>
<td>6.5</td>
<td>Lambert et al. (1980)</td>
</tr>
<tr>
<td>Milford Haven, Wales (51°40’N, 5°02’W)</td>
<td>7-9</td>
<td>-</td>
<td>6.77</td>
<td>Lambert et al. (1983)</td>
</tr>
<tr>
<td>Bombay, India (18°58’N, 72°50’E)</td>
<td>-</td>
<td>40</td>
<td>-</td>
<td>Peirson et al. (1966)</td>
</tr>
<tr>
<td>El-Minia, Egypt (28°04’N, 30°45’E)</td>
<td>4.3-12.8 (avg. 9.83)</td>
<td>-</td>
<td>-</td>
<td>Ahmed et al. (2000)</td>
</tr>
</tbody>
</table>
NOMENCLATURE

\begin{itemize}
  \item 1-ACFM normal flow rate cascade impactor
  \item AMAD activity median aerodynamic diameter
  \item cfpm cubic feet per meter
  \item Dp particle diameter
  \item ECD effective cutoff diameter
  \item HVI high-volume cascade impactor
  \item LPI low-pressure cascade impactor
  \item M molar solution
  \item MMAD mass median aerodynamic diameter
  \item N number of radioactive nuclei
  \item OLACI on-line alpha cascade impactor
  \item SMAD surface median aerodynamic diameter
\end{itemize}

Greek symbols

\begin{itemize}
  \item \( \lambda \) radioisotope disintegration rate
  \item \( \sigma_g \) geometric standard deviation
  \item \( \tau \) radioisotope mean life
  \item \( \tau_R \) residence time of aerosol particles
\end{itemize}

REFERENCES


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