Residence time of tropospheric aerosols in association with radioactive nuclides

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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:

(a) dry deposition by impaction, diffusion and sedimentation, and

(b) wet deposition by rain drops (precipitation scavenging)

as a result of processes occurring both within and below the rain cloud. There could be variations in the removal rates at different continental locations of the globe, over the oceans, and at high altitudes of the atmosphere due to changes in meteorological conditions. There is also a dependence of the tropospheric aerosol residence time on the latitude.

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 (i) cosmic-ray produced radionuclides, such as ^{7}Be (TI/2 =53.3 d), (ii) radioactive decay products of radon, ^{222}Rn and thoron, ^{210}Rn which emanate from continental surfaces into the atmosphere, such as ^{210}Pb (Ti/2 = 22.3 y), ^{210}Bi (TI/2 = 5.01 d) and ^{210}Po (TI/2 = 138.38 d), and (iii) fission product radionuclides from man-made explosions from nuclear weapons testing or reactor accidents at nuclear facilities, such as ^{89}Sr (TV2 = 50.5 d), ^{90}Sr (TI/2 = 28.8y) and. ^{140}Ba (TI/2 = 12.79 d). However, there is disagreement between the derived values of the residence times due to various processes, including the fact that they refer to different portions of the atmosphere, e.g. cosmic-ray produced isotopes refer to upper troposphere or lower stratosphere, such as ^{7}Be, and radon decay products, such as ^{210}Pb, ^{210}Bi, and ^{210}Po that refer to lower troposphere and also due to the existence of different sources for some isotopes, such as ^{210}Po.

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The collection of atmospheric aerosol particles was carried out with high volume jet air samplers, type TFIA-2 of Staplex having glass-fiber filters type TFAGF 810 of Staplex, highly retentive for particulate material, 20.32 cm x 25.40 cm (8" x 10") in dimensions and 99.28 % collection efficiency for submicron particles as small as 0.3 urn and over. The air-flow rate of these samplers was regulated from 1.7 m^3 $min"^1$ (60 cfm) to 1.92 m^3 $min"^1$ (68 cfm), (average 1.84 m^3 $min"^1$ or 65 cfm).The length of each collection period was 24 hours. The size fractionation of atmospheric aerosol particles was carried out with aerosol cascade impactors, type Andersen 2000, as follows: (i) the 1-ACFM design was operated at air-flow rate of 28 1 $min"^1$ (1 ft^3 $min"^1$). Its stages had effective cut-off diameters (BCD) of 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 7.0 and 11.0 urn, (ii) the low-pressure modification which alters the impactor's operation by increasing the resolution in the submicron region, involved a regulated air-flow rate of 3 1 $min"^1$, five low-pressure (114 mm Hg that is the absolute pressure downstream of the critical orifice) stages for the submicron region and eight atmospheric pressure stages for separating aerosol particles above 1.4 urn The ECDs of the low-pressure stages were 0.08, 0.11, 0.23,0.52. and 0.90 urn, whereas for the upper stages they were 1.4, 2.0, 3.3, 6.6,10.5, 15.7, 21.7 and 35.0 urn (iii) the high volume cascade impactors had a regulated air-flow rate either of about 0.57 m^3 $min"^1$ (20 cfm) or 1.13 m^3 $min"^1$ (40cfm) and the ECDs were 0.41, 0.73, 1.4, 2.1,4.2 and 10.2 urn for the 20 cfm configuration or 0.49, 0.95,1.5, 3.0 and 7.2 urn for the 40 cfm configuration at the standard temperature and pressure atmospheric conditions (250° and 760 mm Hg). The stainless steel plates supplied by the manufacturer were used for collection of the aerosol particles. Either polycarbonate or glass-fiber backup filters were used to collect all particles below the 0.08-um collection plate for the low- pressure impactors, below the 0.4- urn collection plate for the 1-ACFM impactors, and below the 0.41- urn or 0.49- urn collection plate for the high volume impactors.

The length of each collection period varied from 1 to 24 hours for ^{7}Be, and ^{210}Pb,^{210}Bi and ^{210}Po, depending on impactor type and objective. The activity of ^{7}Be-associated aerosol particles was measured through its gamma-ray peak of 477.6 kev using a high resolution (1.9 keV at 1.33 MeV of ^{60}Co), high efficiency (42 %) low-background HP Ge detector. The uncertainty of the gamma counting system for ^{7}Be measurements was better than 8.5 %. The activity of ^{210}Pb- and ^{210}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 us. The samples were counted for long enough to obtain a statistical accuracy better than 5 %. It must be noted that ^{210}Pb and ^{210}Bi were chemically separated and measured for their activities. Detailed description of the analytical method and technique was established and presented elsewhere. The activity of lead-210 was also measured by a surface barrier Ge detector through its gamma peak of 46.5 keV. The activity size

* Harsaw model TASC-12-A6

distribution and the activity median aerodynamic diameter (AMAD) of the aerosol particles were determined upon the measurement of the activities of the aerosol-associated radioactive nuclides.

It was found that a mean value of about 8 days could be applied to aerosol particles in the lower atmosphere below precipitation cloud levels, as determined based on measurements of the activities and ratios of activities of cosmic-ray produced radionuclides, such as ^{7}Be, the radioactive decay products of radon-222 emanated from soil into the atmosphere, such as ^{210}Pb, ^{210}Bi and ^{210}Po and the fission product radionuclides released into the atmosphere during the explosions either from nuclear weapons testing or nuclear reactor accidents, such as ^{89}Sr, ^{90}Sr and ^{140}Ba. Increase with altitude within the troposphere by a factor of 3 or less and dependence of the mean tropospheric aerosol residence time on the latitude were also reported and discussed.