Experimental determination of attachment function of radon progeny on aerosol particles

Y. Yamada[1], K. Miyamoto[1], S. Tokonami[1], M. Shimo[1] and K. Yamasaki[2]
[1]National Institute of Radiological Sciences, Chiba 263-8555, Japan
[2]Kyoto University, Osaka 590-0494, Japan

ABSTRACT

Aerosol size information is very important for risk estimation of radon exposure. Size measurements of aerosol particles attaching radon progeny were made by using the Dekati's ELPI (Electrical Low Pressure Impactor), and its attachment function was determined. The impactor, which covers a wide size range from 0.03 to 10 μm in diameter, gives number size distribution in real time by an electrical detection method. Prior to those size measurements, effects of impaction substrates on size classification were studied. Tested substrate materials were stainless steel plate and aluminum foil, and those surfaces were as follows: 1) untreated, 2) silicon grease coated, 3) silicon oil coated, 4) adhesive tape stuck. It was suggested that the use of grease- or oil-coated substrates did not interfere for the ELPI's electrical measurements but prevent particle rebound or redispersion. And it was confirmed that the coating gave no damage on alpha energy spectrum analysis by surface-barrier type solid state detector (SSD). Size distribution data on aerosol number and activity were independently taken for each impactor sample. In the size measurements on activity, the correction of sampling loss at the ELPI's charger unit was made. Comparing between number and activity size distributions, attachment function of radon progeny on aerosol surface was estimated. In the lower submicron range, the attachment strongly depend on the aerosol size. The larger the aerosol become, the less the attachment depend on the aerosol size. The attachment function of the submicron aerosols was in a transient region from surface-area proportion to diameter proportion, as shown in the previous studies. It means that the ELPI is useful for measurements on attachment function of radon progeny on aerosol particles. But the ELPI does not demand monodisperse aerosol as carrier aerosol, and the attachment function can be determined for any aerosols in a chamber or field experiment.

INTRODUCTION

Most of natural exposure is well known to be caused by radon decay products (radon progeny). Because of high diffusion, the radon progeny attach to airborne particles in an atmosphere and form to radon progeny aerosols. When the chance of facing to the airborne particles is low, the radon progeny remains in a free atom and some of them form to cluster with water molecules (see Fig. 1). They are generally divided to "attached fraction" and "unattached fraction". The particle size affects the behavior in the atmosphere. For example, the deposition in the human lung depends primarily on their particle size. The size information is one of the most important parameter for risk estimation of radon exposure. In fact, the dose conversion factor (DCF) strongly depend on the aerosol size.

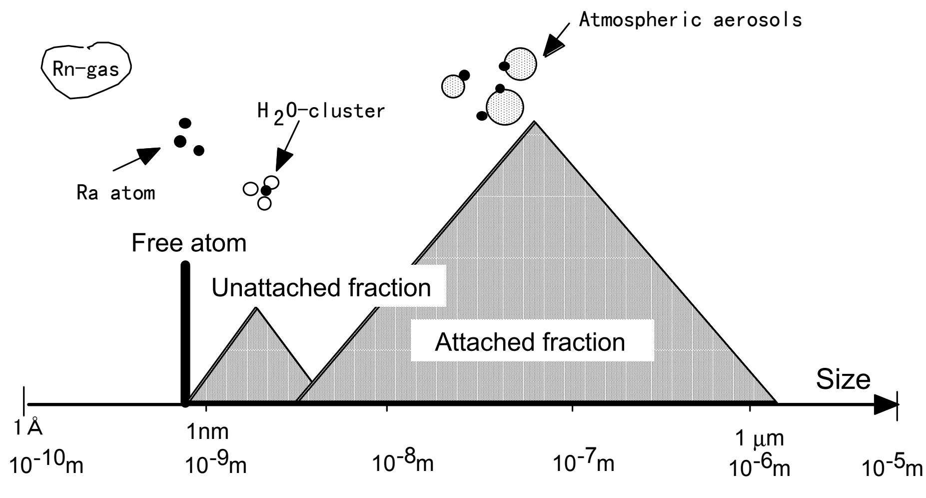


Fig. 1 Schematic of the size distribution of radon progeny aerosol.

Using computer program of "RADEP", Solomon (1) showed that the ICRP (International commision on Radiological Protection) 66 model (2) produces values for the dose per unit exposure for ultrafine (unattached) radon progeny with an activity median thermodynamic diameter (AMTD) ~ 1 nm, that is up to 25 times higher than for attached radon progeny, with an AMTD in the range 100 ~ 300 nm.

Numerous studies on the attachment of radon progeny to ambient aerosol particles have been made. Major attachment theory basically consist of a diffusion theory and a kinetic theory. The attachment is proportional to the particle size for large particles > 1 μm and to the particle surface area for small particles < 0.1 μm (3). In most of the experimental studies, monodisperse particles have been used as carrier aerosol of radon progeny. The monodisperse particles might be useful in size determination for the attachment coefficient, but it limits the variety of carrier aerosol and cannot exclude differences in aerosol samples between for particle size measurement and for activity measurement. Recently, new particle sizing instrument for a real-time measurement was developed (4). It is one of the cascade impactor, particle samples are obtained for each collection stage. Evaluation of both particle number and activity size distributions will allow us to understand an attaching mechanism of radon progeny to aerosol particles. Therefore, the new impactor was applied to experimental determination of the attachment function.

Experimental Method

Size measurements of aerosol particles attaching radon progeny were made by using the electrical low pressure impactor (ELPI-2000, Dekati). The impactor, which covers a wide size range from 0.03 to 10 μm in diameter, gives number size distribution in a real time by an electrical detection method with a multi-channel electrometer. It consists of a corona charger, low pressure impactor and multi-channel electrometer as shown in Fig. 2. The aerosol particles in the sample air are charged electrically at an corona charger. The charged particles collected in a specific impactor stage produce an electrical current which is proportional to the number of the collected particles. Although the number measurement is made in a real-time, the impactor unit must be disassembled for activity measurement. In this study, radioactivity of radon progeny is measured by alpha detector such as a solid state detector (SSD). In size measurements, backup filter has been added just after the ELPI-unit. The alpha counting system (Alpha-Analyst, Canberra) of twelve detectors was used for alpha energy spectrum and radioactive decay analyses. From those above measurements, air concentration and size distribution was determined against each radon progeny of ^{218}Po (RaA), ^{214}Pb (RaB) and ^{214}Bi (RaC).

Prior to those size measurements, effects of impaction substrates on size classification were studied. Tested substrate materials were stainless steel plate and aluminum foil, and those surfaces were as follows: 1) untreated, 2) silicon grease coated, 3) silicon oil coated, 4) adhesive tape stuck. Keskinen et al. had stated electrical particle loss caused by the charger (4). It has been evaluated as a collection efficiency of each stage in an electrical operation, and the correction has been made in the ELPI's software for size analysis (6,7). But this correction is not applicable to the ELPI for taking the normal inertial impactor sample in non-electrical operation. Therefore the, the wall loss caused by an electrical field at the corona charger in an inlet of the ELPI was studied by using radon progeny aerosols. The loss is divided to the wall loss caused by particle motion and the electrical loss caused by strong electric field. Impactor samples were taken with or without a charger to determine the wall loss efficiency. And they were taken in an operation of charger power on or off.

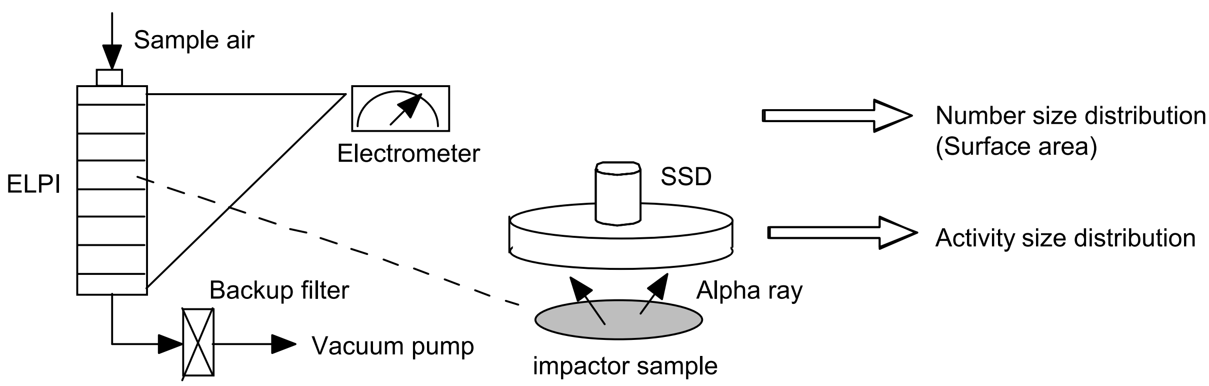


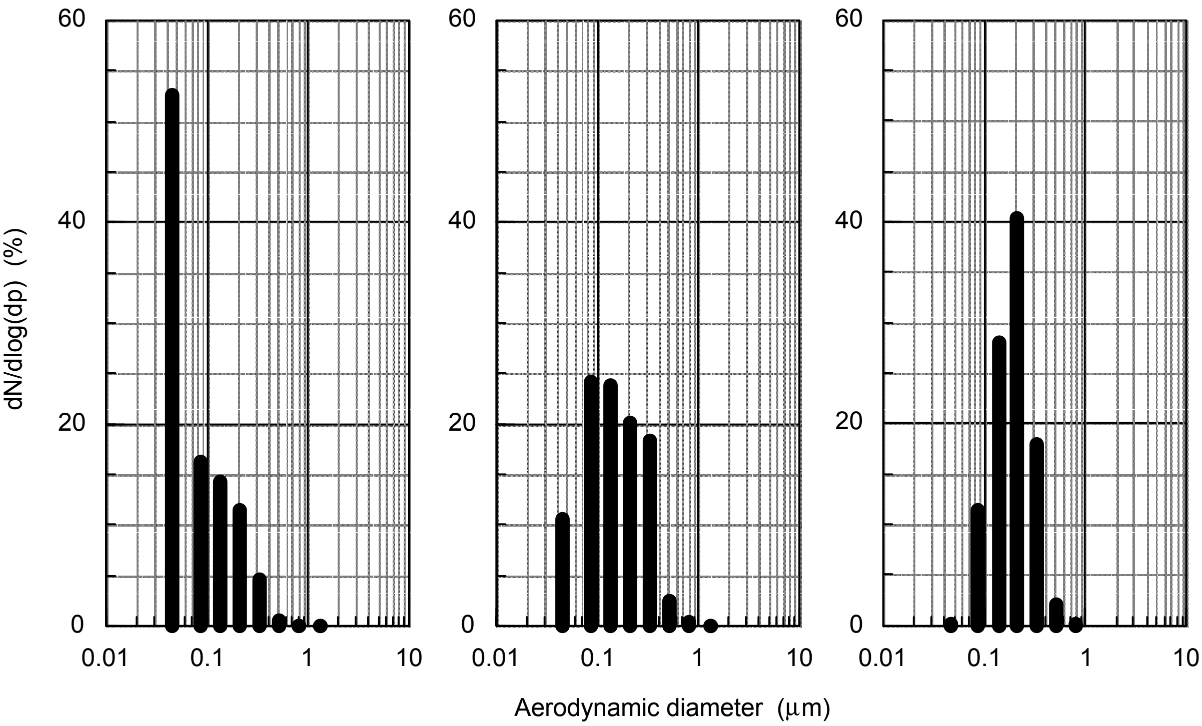
Fig. 2 Experimental setup for the size measurements of radon progeny aerosol.

Carnuba wax and incense smoke were used as carrier aerosolof radon progeny. In a chamber experiment with a volume of ~ 2 m^3, the carnauba wax aerosols were produced by an vaporization-condensation type aerosol generator (MAGE, Lavoro Ambiente). The condensation nuclei was NaCl particles nebulized from the solution. In an indoor experiment using the big space of ~ 6000 m^3, incense smoke aerosols were simply produced by a combustion. The radon gas in the chamber experiment was fed from soil source, and that in the room experiment was emanated from building materials.

The sampling time of the ELPI was 2 min or 5 min. The loss time for sample transfer from the aerosol sampling to the alpha counting was just 2 min. The 9 counting for each sample was taken at every 5 min. In order to confirm particle collection characteristics of the ELPI or to monitor a status of aerosol generation, comparison measurements was made with a particle sizing system using a screen type diffusion battery method with a condensation nucleus counter (SDB/CNC). Screens of the SDB was stacked in the same way as the commercial version of TSI-3040, but the sampling procedure and data analyzing method were modified for the fast measurement (8). The sizing performance of the SDB/CNC had been confirmed in a comparison measurement with the scanning mobility particle sizer (SMPS-3934, TSI) at the Environmental Measurements Laboratory (EML, US-DOE).

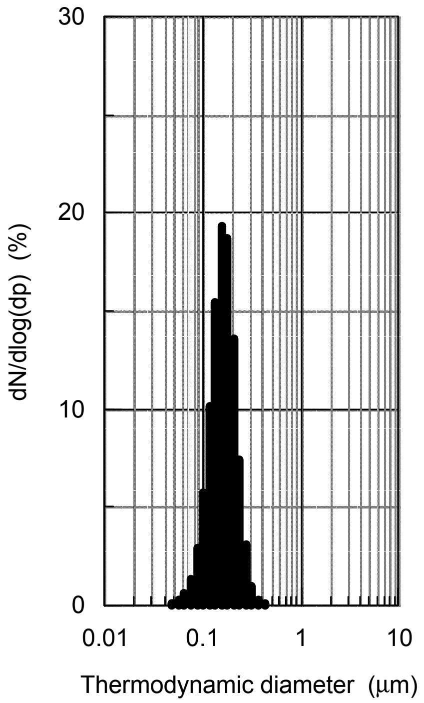
RESULTS AND DISCUSSION

Prior to size measurements by the ELPI, effects of impaction substrates on size classification were studied. Tested substrate materials were stainless steel plate and aluminum foil, and those surfaces were as follows: 1) untreated, 2) silicon grease coated, 3) silicon oil coated, 4) adhesive tape stuck. As shown in Fig. 3, there were a big difference in the measured size distributions among the various surface conditions. From the comparison measurement (see Fig. 4) with the SDB/CNC, the grease or oil coating was found to be good for the surface of substrate. No differences were significantly observed between in the substrate materials of stainless steel plate and aluminum foil. And also the thickness of coating did not affect the collection efficiency of impactor stage. Figure 5 shows one of the typical alpha energy spectrum of the ELPI sample of radon progeny aerosol. Alpha rays emitted from RaA and RaC' were clearly separated and formed two sharp peaks in 6.0 and 7.7 MeV. It means that the coating to prevent particle rebound and/or redispersion gives no damage for the analysis of alpha energy spectrum. In the ELPI impactor, a



(a) Untreated (b) Adhesive taped (c) Grease coated

Fig. 3 Difference in size distribution of the incense smoke aerosol collected on various substrates of the ELPI.



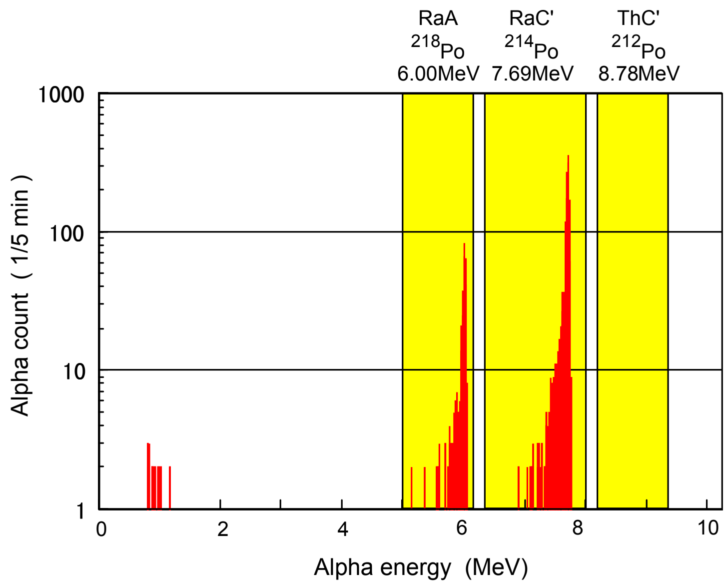


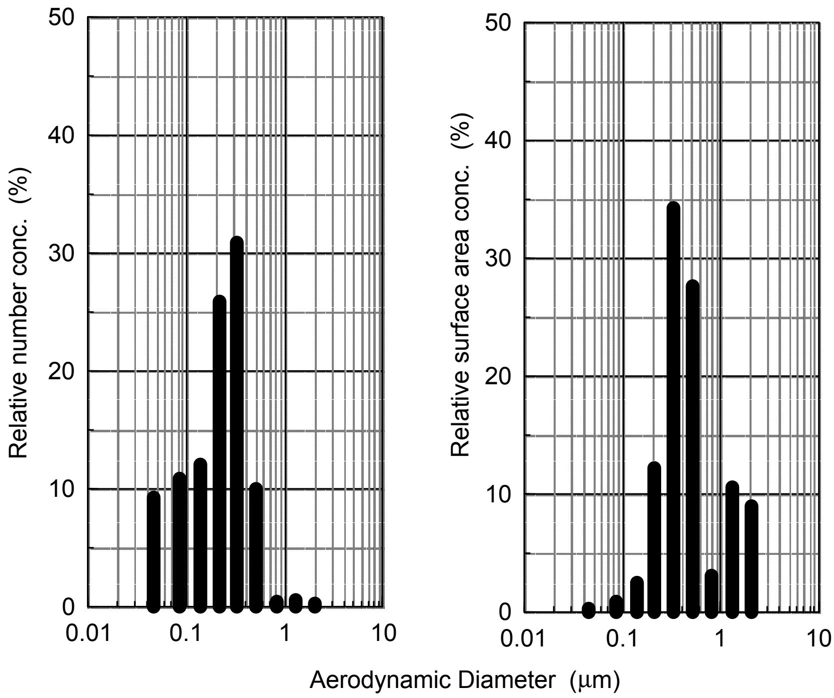
Fig. 4 Size distribution of the incense smoke aerosol measured with the SDB /CNC system.

Fig. 5 Alpha energy spectrum of the ELPI sample collected in the grease coated substrate.

special device of the charger unit was installed at inlet of the impactor unit, and the sampling loss at the charger unit was investigated. As a significant loss was observed there, the correction of the sampling loss was made in evaluation of radioactivity collected in impactor stage.

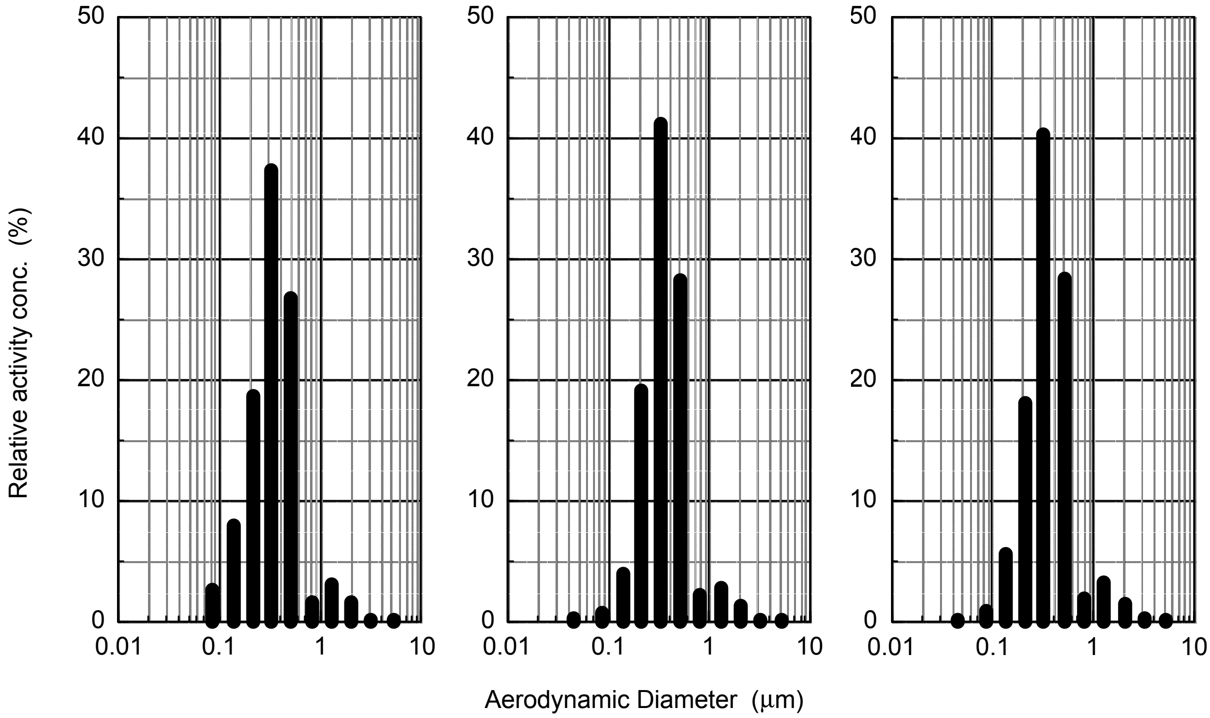
The experimental results on size distribution of radon progeny aerosol are shown in Fig. 6. According to the measurement with the SDB/CNC system, number concentration of the incense smoke aerosol was measured to be 2.4 x 10^3 particles/cm^3. The count median diameter (CMD) and the geometric standard deviation (GSD) were

0.238 µm and 1.74, respectively. The ELPI was operated in the normal mode using charger unit. The surface-area size distribution, which was calculated from the number size distribution, moved to the right side, but the peak remained in the same 5th stage of 0.261 ~ 0.402 µm as the peak of number size distribution was (see Fig. 6(a) and (b)). On other hand, the activity size distributions of RaA, RaB and RaC were estimated from the alpha energy spectrum analysis of each impactor stage. The distributions were almost the same among them (see Fig. 6(c), (d) and (e)). And the peaks of the activity size distributions also were observed in the same 5th stage as the peaks of number and surface-area size distribution were. The shape of the activity size distribution were different from those of the number and the surface-area size distributions. In order to clear how radon progeny attach to aerosol particle, radioactivity per particle was calculated and its dependence on the particle size was evaluated. In other words, size distribution data on aerosol number and activity were independently taken for the same impactor sample. Comparing between number size distribution and activity one, attachment function of radon progeny on aerosol surface was estimated. In the lower submicron range, the activity per particle increased in proportion to surface-area of aerosol as shown in Fig. 7. Namely, the attachment function was very closed to squared one of aerosol size. In the upper range, the activity per particle was proportion to aerosol size. And the middle range was in a transient region from aerosol surface-area to aerosol size. These results do not contradict with the experimental results using monodisperse aerosols (5). It means that the ELPI is useful for measurements on attachment function of radon progeny to aerosol particle. Since the measurement with ELPI does not demand monodisperse aerosols, the attachment function can be determined for any aerosols in a chamber or field experiment.



(a) Number

(b) Surface area

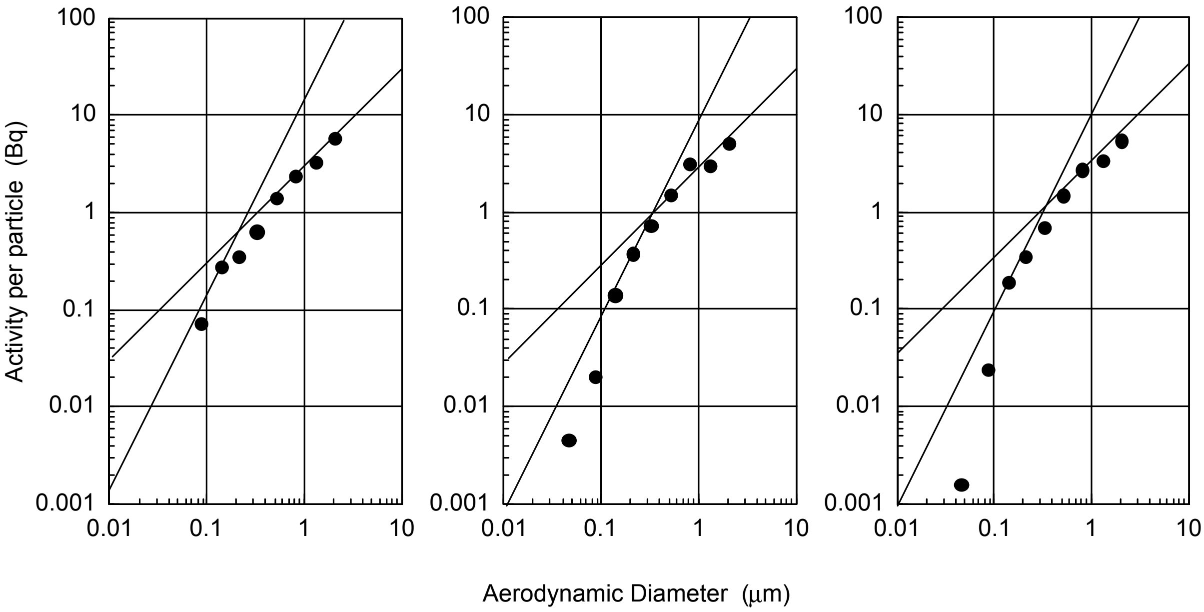


(c) Activity of RaA

(d) Activity of RaB

(e) Activity of RaC

Fig. 6 Number, surface area and activity size distributions of the incense smoke aerosol measured with the ELPI.



(c) RaA (d) RaB (e) RaC

Fig. 7 Size dependance of activity per particle on particle size

REFERENCES

1. S.B. Solomon, *A radon progeny sampler for the determination of effective dose*, Radiat. Prot. Dosim. 72, 31-42 (1997).
2. International Commission on Radiological Protection (ICRP), *Human respiratory tract model for radiological protection*, Publ. 66 (1994).
3. J. Porstendorfer and T.T. Mercer, *Adsorption probability of atoms and ions on particle surfaces in submicrometer size range*, J. Aerosol Sci., 9, 469-474 (1978).
4. J. Keskinen, K. Pietarinen and M. Lehtimaki, *Electrical low pressure impactor*, J. Aerosol Sci., 23, 353-360 (1992).
5. J. Porstendorfer, G. Robig and A. Ahmed, *Experimental determination of the attachment coefficients of atoms and ions on monodisperse aerosols*, J. Aerosol Sci., 10, 21-28 (1979).
6. M. Marjamaki, J. Keskinen, D.R. Chen, et al., *Performance evaluation of the electrical low-pressure impactor (ELPI)*, J. Aerosol Sci., 31, 249-261 (2000).
7. J. Porstendorfer, G. Robig and A. Ahmed, *Experimental determination of the attachment coefficients of atoms and ions on monodisperse aerosols*, J. Aerosol Sci., 10, 21-28 (1979).
8. Y. Yamada and Radon Group for Intercomparison, *The 2nd intercomparison experiment of radon , its progeny and particle sizes at EML (2)*, The 33rd annual meeting of Japan Health Physics Society in Hamamatsu, A-22 (1998).