Chapter III
PRELIMINARY WORK

Chapter III presents the previous work on mitigation of the effective dose of radon decay products through the use of an air cleaner in a dwelling in Okinawa, Japan [45]. The concentration of radon and its decay products and the activity size distribution of attached aerosol particles were measured with and without the use of an air cleaner in a dwelling with a high radon concentration.

3.1 Study Location

Concentrations of indoor radon and its decay progeny were measured in the first-floor bedroom of a dwelling in Yomitan village, Okinawa prefecture, Japan. The study took place from 19 to 21 November 2007. The volume and surface area of the room are about 72 m^3 and 24 m^2, respectively. The walls and the floors are made from gypsum board and wood, respectively. The bedroom was selected for experiments because relatively high levels of radon concentration had been previously recorded in this room [46].

3.2 Measurement Techniques

Indoor radon concentration and equilibrium equivalent concentration of radon (EECRn) were measured continuously at every one hour with a pulse ionization chamber (AlphaGUARD, Genitron Instruments GmbH, Germany) and a Working Level monitor (Pylon Electronics Inc., Canada), respectively.

The total activity concentration of radon progeny was measured after sampling radon progeny on a glass microfiber filter (47 mm diameter, Whatman®, England) that had been operated at a flow rate of 10 L min^{-1} for 5 min. Gross alpha particles were counted with a ZnS(Ag) scintillation detector (Ludlum Instrument Inc., USA) for a period of 40 min at 5-min intervals. The concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi were calculated with the method described by Thomas [47].

The unattached fraction of radon progeny was measured by using an alpha spectrometry method. Unattached progeny was collected on a 400-mesh metal wire screen (47 mm diameter, TETKO Inc., USA), and they were considered as a single type for convenience. The diffusion coefficient was assigned to be 0.065 $cm^2 s^{-1}$ [48].

The alpha activities were detected during and after air sampling by a continuous air monitoring PIPS detector (Model CAM1700, Canberra Inc., USA), which was set opposite to the metal wire screen [49]. The measurement system was set up as shown in Figure 3-2. ^{241}Am was used as a calibration source, and the counting efficiency was found to be 30.1 ± 0.5%. The detector has an active area of 1700 mm^2 (46.5 mm diameter) and an alpha energy resolution of about 70 keV. This resolution enables the discrimination between the alpha particle energies of 6.0 MeV from ^{218}Po and 7.8 MeV from ^{214}Po. To determine the activity concentration of unattached radon progeny (^{218}Po, ^{214}Pb and ^{214}Po), the measurements were performed in two steps. First, the alpha spectrum was acquired during a sampling period of 20 min. Second, after waiting for a period of 5 min without sampling, the alpha spectrum was measured again (during decay) for a period of 15 min. The air flow rate during the measurement was 4 L min^{-1}. From the measured alpha-counts of ^{218}Po and ^{214}Po during the sampling period and the ^{214}Po counts during the decay period, the activity concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi could be calculated by using the build-up and decay method [50]

For the determination of the particle size distribution of the attached radon progeny, a Micro Orifice Uniform Deposit Impactor (Model 110 MOUDI^{TM}, USA) was used. Efficiency curves and interstage losses of the impactor stages were determined [51]. The impactor consisted of eight size fractionating stages and a back-up filter holder, and it was operated at a flow rate of 30 L min^{-1} for 10 min. The measured 50% cut-off diameters for the eight stages were 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, and 18 μm. A 400-mesh metal wire screen was set at the top of the MOUDI to prevent invasion of unattached progeny, and the collection efficiency was estimated to be 89.1%, based on fan model filtration theory [52]. After sampling, the collected activity on each impactor stage was simultaneously measured with the ZnS(Ag) scintillation detectors. The activity concentrations of radon progeny for each stage were analyzed by using the decay method. The particle size distributions were described in terms of a log-normal distribution, defined by the activity median aerodynamic diameter (AMAD) and geometric standard deviation (σ_g). In addition to this numerical evaluation, the impactor data were also evaluated by the Expectation-Maximization algorithm [53].

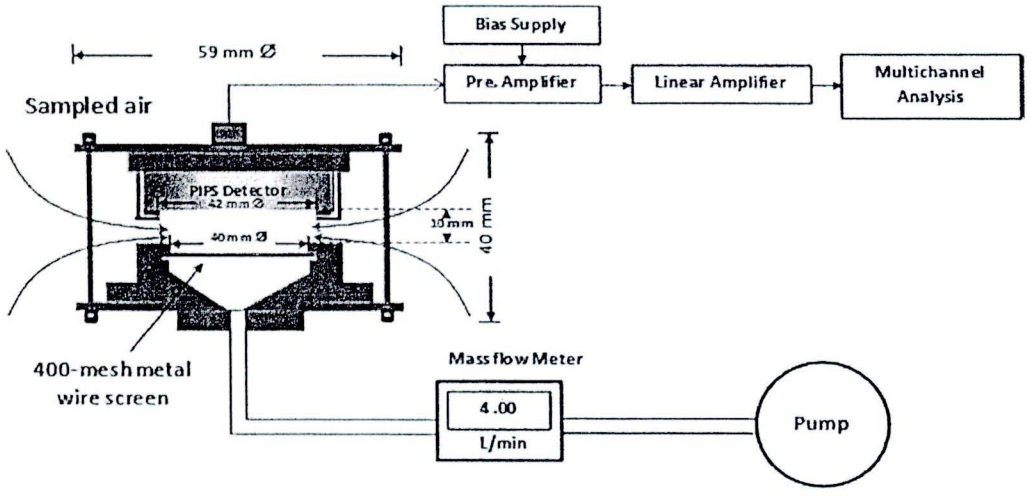

Figure 3.1 The schematic diagram of unattached radon progeny measuring system.

3.3 Measurement Conditions

The air cleaner (Hitachi model EP-X31, Japan) was equipped with a HEPA filter and a deodorizing activated carbon filter, and was set at the highest flow rate (6.5 m^3 min^{-1}) for faster and more powerful room cleaning. The air cleaner is suitable for a room of up to about 50 m^2 in size, according to its specifications. Measurements were conducted without the operation of the air cleaner (case I, 16:00 on 19/11/07 to 16:00 on 20/11/07) and with it (case II, 16:00 on 20/11/07 to 16:00 on 21/11/07). Human activities were kept as a minimum as possible during the measurement periods.

Ambient conditions in the room, including temperature, relative humidity, and air pressure were continuously measured, along with the radon concentration, by the AlphaGUARD monitor.

3.4 Results and Discussion

3.4.1 Measurement Results

Figure 3-2 shows the temporal variations for radon concentration and EECRn (Figure 3-2a), temperature and relative humidity (Figure 3-2b), and air pressure (Figure 3-2c) for cases I and II. The radon concentration level and EECRn show the same tendencies in both cases. In this measurement, the differences in temperature

and air pressure between indoor and outdoor were about 2-5 °C and 1-3 hPa, respectively. These differences were not so significant. However, the mean radon concentration in case II (373 Bq m^{-3}) was higher than in case I (229 Bq m^{-3}) (Table 3-1), whereas the mean EECRn was lower by about 57%. A possible explanation is that the difference resulted from the human activities in the experimental room and from the indoor-outdoor air exchange process on radon entry into or escape from the experimental room. The air exchange process is the results of a complex interplay of many factors, such as wind velocity and indoor-outdoor temperature and air pressure differences. In addition, these factors are time-varying [54]. The cause of difference in radon concentration could not be specified from our short-term survey. Long-term measurements of radon concentration and related parameters will be necessary. The mean radon concentration during the measurement was more than 300 Bq m^{-3} (Table 3-1), much higher than the average annual indoor radon concentration in Japan (15.5 Bq m^{-3}). The effectiveness of the air cleaner in reducing exposure can be observed from the values for the equilibrium factor (F, the ratio of EECRn to radon concentration): it was about one-third the size of case I in case II (decreased by about 71%). Li and Hopke [55] also reported that the radon concentration was unchanged but F was reduced by about 50-60 % when the air cleaning system was operated.

Table 3-1 Mean (± standard deviation) concentrations of radon and its progeny.

Case	Radon (Bq m^{-3})	EECRn (Bq m^{-3})[a,c]	F[b]
I	229 ± 15	33.1 ± 3.1	0.14 ± 0.01
II	373 ± 17	14.4 ± 0.7	0.04 ± 0.01
Mean	301 ± 17	23.8 ± 1.6	0.08 ± 0.01

[a]EECRn is the equilibrium equivalent concentration of radon, that was calculated using "Correction Factor (CF)", and the CF was estimated assuming a certain activity concentration ratio: ^{218}Po, ^{214}Pb and ^{214}Bi = 1:1:1. However, in actual environment, the concentration ratio of ^{218}Po, ^{214}Pb and ^{214}Bi is always different from it.
[b]F is the equilibrium factor as a ratio of EECRn to radon concentration.

A summary of radon progeny concentrations is shown in Table 3-2. The unattached progeny of ^{218}Po and ^{214}Bi increased by about 50% during the operation of the air cleaner, and the attached progeny decreased as compared with case I. The potential alpha energy concentration (PAEC) of unattached progeny increased by about 87 %, while the air cleaner was operated (case II). The unattached progeny

increased because aerosol particles were removed from the room air by the air cleaner. Thus, newly formed radon decay products have fewer particles to which they can attach, and the concentration of unattached progeny can increase during the operation of an air cleaner with a high-efficiency filter [56]. Consequently, there is also a high value for f_p in case II, which increased by about 174% as compared with case I. However, the activity concentrations of ^{218}Po and ^{214}Pb and the PAEC of the whole fraction (attached and unattached fractions) in case II are lower than in case I. This resulted in a low F value for case II (Table 3-1).

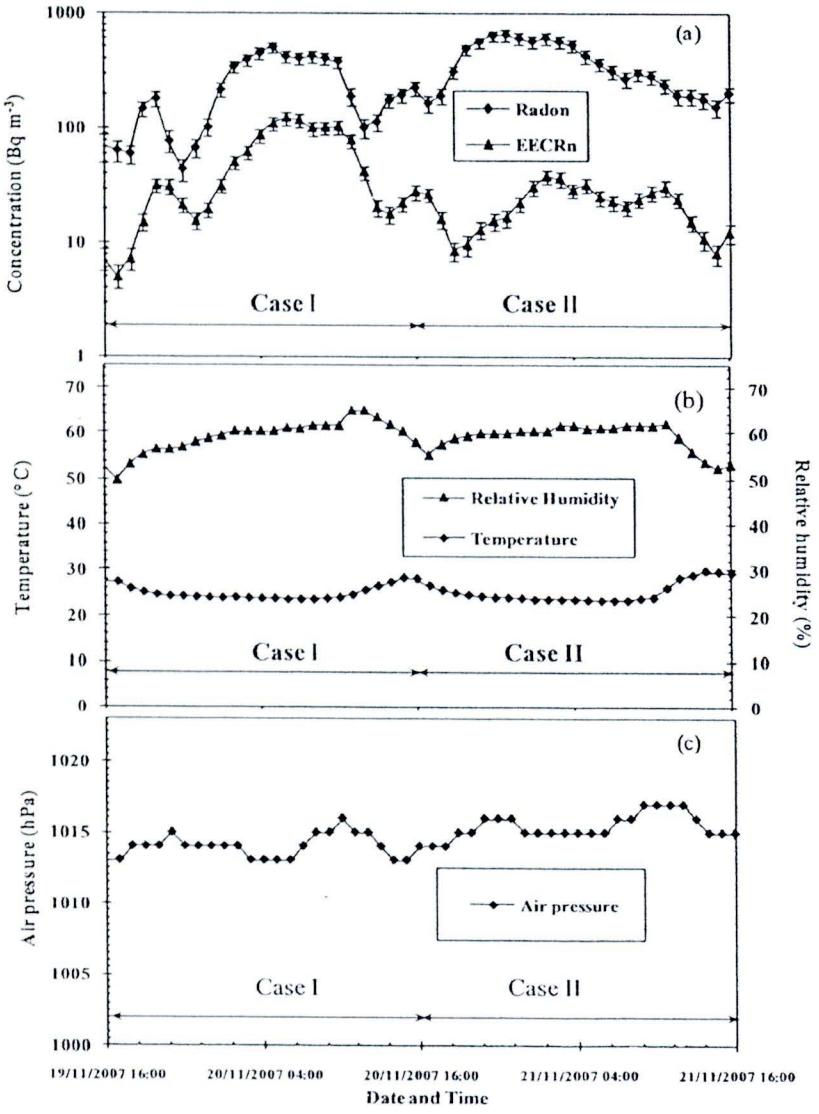

Figure 3-2 Temporal variations of (a) radon and EECRn, (b) temperature and relative humidity, and (c) air pressure.

Table 3-2 Mean (± standard deviation) concentrations of the unattached and total activity concentrations (ranges are given in parentheses) [a, b].

Case	Number of measurement	C_1^{un} (Bq m^{-3})	C_1^{att+un} (Bq m^{-3})	C_2^{un} (Bq m^{-3})	C_2^{att+un} (Bq m^{-3})	C_3^{un} (Bq m^{-3})	C_3^{att+un} (Bq m^{-3})	$PAEC^{un}$ (nJ m^{-3})	$PAEC^{att+un}$ (nJ m^{-3})	f_p [c]
I	5	26.4 ± 1.4 (3.3–51.7)	75.4 ± 2.9 (46.5–88.2)	0.9 ± 0.4 (0–3.0)	23.0 ± 0.8 (13.7–33.6)	2.1 ± 0.3 (0–5.9)	5.7 ± 1.0 (0.04–11.3)	22.3 ± 1.3 (7.6–42.2)	119.7 ± 3.5 (65.2–161.1)	0.19 (0.05–0.21)
II	6	52.2 ± 0.4 (37.6–63.7)	60.0 ± 1.8 (48.2–90.1)	0.8 ± 0.2 (0–3.8)	12.8 ± 0.5 (5.8–20.4)	4.4 ± 0.1 (2.5–5.8)	4.8 ± 0.6 (1.5–9.1)	41.7 ± 0.6 (37.6–47.8)	80.6 ± 2.1 (62.6–125.6)	0.52 (0.30–0.71)

[a] "un" and "att+un" refer to unattached progeny and total progeny, respectively.
[b] C_1, C_2 and C_3 refer to the activity concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi, respectively.
[c] f_p is the unattached fraction, which was calculated using the equation $f_p = PAEC^{un}/(PAEC^{att+un})$.

Figure 3-3 shows the activity size distribution of radon progeny for the two cases. The parameters for the activity size distributions of the attached radon progeny are summarized in Table 3-3, where it can be seen that σ_g is the same for both cases in the nucleation, accumulation, and coarse modes. The lowest activity fractions of the coarse mode in both cases are observable. In addition, the activity concentration of attached radon progeny (Figure 3-3) in the accumulation mode (50–2000 nm) in case II was obviously deceased by about 42% as compared with that of case I. This decrease is related to the EECRn level (Figure 3-2a), which was lower in case II than in case I. Furthermore, the deposition velocity of accumulation mode particles is less than that of nucleation and coarse mode particles [57]. Particles in the accumulation mode can therefore no longer be in suspension in air. Thus, it is feasible that the decreased EECRn in case II is a result of a reduction of particles in the accumulation mode.

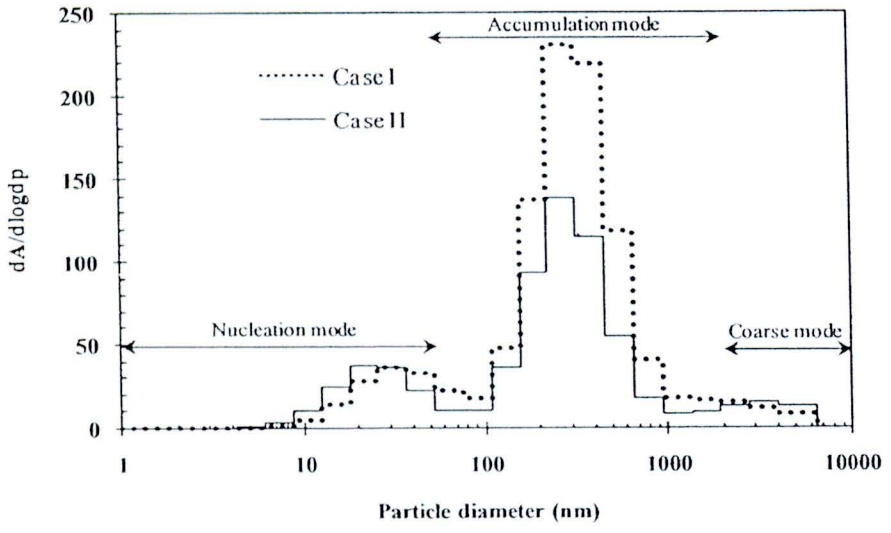

Figure 3-3 The activity size distribution of radon progeny (averaged over the study period).

Table 3-3 Mean values of the activity median aerodynamic diameter ($AMAD_i$), the geometric standard deviation (σ_{gi}), and the activity fraction (f_i) of log-normal activity size distribution of attached progeny (ranges are given in parentheses). The nucleation mode (aerosol particle size range: < 50 nm), accumulation mode (50–2000 nm), and coarse mode (> 2000 nm) are represented by Mode 1, Mode 2, and Mode 3, respectively [58].

Case	Number of measurements	Radon progeny								
		Mode 1			Mode 2			Mode 3		
		$AMAD_1$ (nm)	σ_{g1}	f_1	$AMAD_2$ (nm)	σ_{g2}	f_2	$AMAD_3$ (nm)	σ_{g3}	f_3
I	4	29 (23–42)	1.6 (1.5–1.7)	0.14 (0.09–0.21)	267 (234–308)	1.7 (1.6–1.9)	0.81 (0.73–0.86)	2082 (1858–2520)	1.5 (1.4–1.6)	0.05 (0.05–0.07)
II	3	23 (17–28)	1.6 (1.5–1.7)	0.20 (0.05–0.32)	249 (217–275)	1.7 (1.6–1.8)	0.71 (0.73–0.86)	2576 (2170–3143)	1.5 (1.4–1.6)	0.08 (0.03–0.17)

$_i$ refer to the mode of particles.

These results show that the air cleaner used in this study is effective in removing particles in the accumulation mode, that in agreement with the Trust Science Innovation [59], that reported the mechanisms of HEPA filter is high efficiency (99.97%) for 300 nm particles at a flow rate of 0.085 m^3 min^{-1}. Therefore, F (Table 3-1) decreased and f_p (Table 3-2) increased in case II as a result of the removal of particles in the accumulation mode. Several studies [55, 60, 61] have shown that air cleaners are effective in reducing total radon progeny, although the concentration of unattached radon progeny can increase.

3.4.2 Estimation of Effective Dose

To estimate the effective dose from radon progeny, the effective dose per unit exposure to radon progeny (the dose conversion factor, DCF) is needed. The DCF can be derived by using either a dosimetric or an epidemiological approach. Although the International Commission on Radiation Protection (ICRP) recommends that the DCF should be based on epidemiological studies, it also states that a dosimetric approach is useful for comparing the doses that result from different exposure conditions [33]. In the present study, aerosol conditions are quite different between the two cases. Thus, the effective dose was compared between the two cases, using the ICRP 66 dosimetric model.

Porstendörfer [41] proposed that a dominant factor affecting the DCF is f_p, and that it can be expressed as the following function of f_p:

$$DCF = DCF_{ae} + C f_p \qquad (3\text{-}1)$$

where DCF_{ae} is the DCF for the attached fraction and C is a constant derived from DCF for the unattached fraction (DCF_{un}).

Ishikawa et al. [38] calculated DCF values for reference conditions in places that have different aerosol characteristics. Using these values, the DCF_{ae} was calculated to be 20.2 nSv/(Bq h m^{-3}) for case I and 26.1 nSv/(Bq h m^{-3}) for case II. Also, the DCF_{un} was calculated to be 86.6 nSv/(Bq h m^{-3}) for cases I and II, assuming that AMAD and σ_g of a typical particle size distribution for the unattached fraction are 1 nm and 1.3, respectively [38]. On the basis of these values, DCF can be expressed as the following functions of f_p:

$$\text{Case I: } DCF = 20.2 + 66.4 f_p \text{ nSv/(Bq h m}^{-3}) \quad (3\text{-}2)$$

$$\text{Case II: } DCF = 26.1 + 60.5 f_p \text{ nSv/(Bq h m}^{-3}) \quad (3\text{-}3)$$

Using the above formulas and the unattached fraction (Table 3-2), the DCF of case I was calculated to be 32.8 nSv/(Bq h m^{-3}) and that of case II was 57.6 nSv/(Bq h m^{-3}). That is, the DCF for case II is about 1.8 times the size of that for case I.

The Radiological Protection Regulation [33] recommends that the minimum exemption level for exposure situations to radon in dwellings is 200 Bq m^{-3}. Using this radon concentration and estimated equilibrium factors (Table 3-1), the EECRn was assumed to be 28 in case I and 8 in case II. Consequently, the effective dose (rate) for the two cases can be compared as follows:

Case I: 28 Bq m^{-3} × 32.8 (nSv/(Bq h m^{-3})) = 918 nSv/h

Case II: 8 Bq m^{-3} × 57.6 (nSv/(Bq h m^{-3})) = 461 nSv/h

Therefore, operation of the air cleaner decreased the effective dose by 50%. This result was in general agreement with the 39% dose reduction found by Li and Hopke [55].

3.5 Conclusions

This study was conducted to investigate the effects of an air cleaner on radon mitigation, including the dose mitigation effects. The air clearer was operated at the highest flow rate (6.5 m^3 min^{-1}) in the room of 72 m^3 in volume. The results show that the EECRn and the equilibrium factor decreased, but the unattached fraction increased. According to our dose calculations, the effective dose resulting from the radon

progeny was reduced by operating the air cleaner. Hence, the use of an air cleaner was found to be an effective method for mitigating the health hazards of exposure to indoor radon.