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RADON–THORON DISCRIMINATIVE MEASUREMENTS IN GANSU PROVINCE, CHINA, AND THEIR IMPLICATION FOR DOSE ESTIMATES

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Indoor radon measurements were carried out in cave dwellings of the Chinese loess plateau in Gansu province, where previously the Laboratory of Industrial Hygiene (LIH), China, and the U.S. National Cancer Institute (NCI) had conducted an international collaborative epidemiological study. The LIH–NCI study showed an increased lung cancer risk due to high residential radon levels, and estimated the excess odds ratio at 100 Bq/m^3 to be 0.19 (Wang et al., 2002). The present study used two types of newly developed passive monitors: One is a discriminative monitor for radon and thoron; the other is a selective monitor for thoron decay products. The arithmetic mean concentrations of indoor radon and thoron were 91 and 351 Bq/m^3, respectively. As reported by our previous study in Shanxi and Shaanxi provinces (Tokonami et al., 2004), the presence of high thoron concentration was confirmed and thoron was predominant over radon in the cave dwellings. However, the mean equilibrium equivalent thoron concentration (EETC) was found to be much lower than expected when assuming the equilibrium factor of 0.1 provided by the UNSCEAR (2000) report. The effective dose by radon and thoron decay products was estimated to be 3.08 mSv/yr. It was significantly lower than the dose of 8.22 mSv/yr estimated from the measurements that did not take into consideration any discrimination between radon and thoron. Excess relative risk of lung cancer per sievert may be much higher than the risk estimated from the LIH–NCI study, considering that discriminative measurements were not used in their study.

It is well recognized that a high level of radon exposure causes lung cancer. Radon has many isotopes, but ^{222}Rn of the uranium decay series and ^{220}Rn (thoron) of the thorium decay series are of concern because of their presence in the human environment and the possibility of their health effects on the public. The contribution of each nuclide to radiation exposure is quite different. Radon's half-life of 3.8 d is long enough for it to enter into the indoor environment and to cause an increase in indoor concentrations, but it is relatively too long to be inhaled into the respiratory tract and to irradiate cells. On

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the other hand, the half-life of thoron is only 56 s, so that its presence is limited to close proximity to the source. We suspect that thoron plays a very important role in risk evaluation for the following two reasons. First, some conventional radon monitors are sensitive to thoron, and thus the values measured as radon concentration may possibly be affected by the presence of thoron (Shang et al., 1997). Second, recent surveys found areas with high levels of thoron (Shang et al., 1997; Wiegand et al., 2000), including the areas we studied (Tokonami et al., 2004). Our study, using newly developed radon–thoron discriminative monitors, found a number of cave dwellings with the thoron levels higher than 200 Bq/m^3 in Shaanxi and Shanxi provinces. We also found that radon levels were significantly lower than what was observed in the large-scale case-control study conducted by the Laboratory of Industrial Hygiene (LIH, China) and U.S. National Cancer Institute (LIH–NCI). Their survey area of Gansu province was very similar to ours. They concluded that high levels of residential radon increase the risk of lung cancer, and that increased lung cancer risks may equal or exceed the extrapolations based on the data for miners (Wang et al., 2002). Since the results of our survey strongly suggest a possibility of overestimating radon concentration when no discriminative measurements are made, the LIH–NCI study might have underestimated the excess relative risk of lung cancer associated with radon exposure.

This article reports the results of our latest radon and thoron survey conducted in Gansu province, where the LIH–NCI epidemiological study was conducted, in addition to our previous study, and discusses problems in dose evaluation.

MATERIALS AND METHODS

A radon and thoron survey was conducted in Qingyan prefecture, Gansu province. Cave dwellings were widely distributed not only in the mountainous area but also in the flat area. A typical underground cave dwelling in the flat area is shown in Figure 1. The number of inhabitants living in such dwellings is

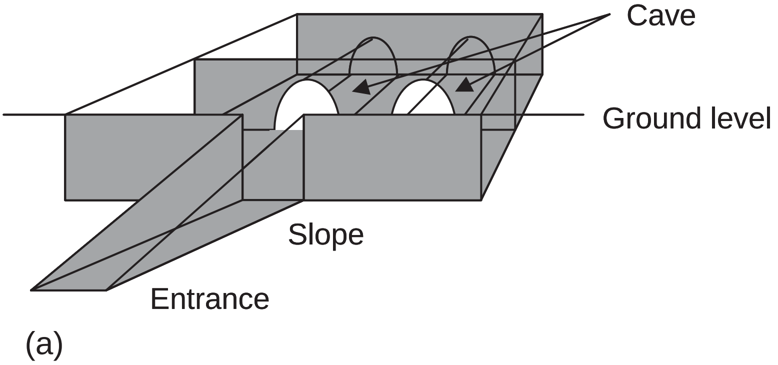

FIGURE 1. Typical underground cave dwelling in Gansu province: (a) perspective view, (b) entrance to the cave dwellings, and (c) overview of the underground cave dwellings.

(b)

(c)

FIGURE 1. *(Continued).*

over 3 million. Long-term measurements for 1 yr were made in 102 dwellings using 2 types of special passive monitors that we newly developed. One type is a discriminative monitor for radon and thoron (Zhuo et al., 2002), and the other is a selective monitor for thoron decay products (Zhuo & Iida, 2000). The discriminative monitor has two diffusion chambers with different air exchange rates. By analyzing sensitivities of the chambers to radon and thoron, their concentrations can be evaluated separately. The lowest detectable limits are 3.5 Bq/m^3 for radon and 13 Bq/m^3 for thoron in the case of an exposure period of 90 days (Tokonami et al., 2003). The selective monitor detects only high-energy alpha particles emitted from ^{212}Po, which is one of the thoron decay products. This monitor uses a Mylar film with a suitable thickness for the discrimination of high-energy alpha particles. By analyzing the deposition rate of the thoron decay products on the monitor surface, the equilibrium equivalent thoron concentration (EETC) can be estimated. The lowest detectable limits of this monitor are 0.08 Bq/m^3 for a 90-d exposure. CR-39 was used as the detector material in both passive monitors. The monitors were suspended from the ceiling in the center of each cave. The distance from the ceiling ranged from 5 to 30 cm.

In a separate short-term survey, size measurements of carrier aerosols attached to radon and/or thoron decay products were tentatively made with a screen-type diffusion battery (SDB). Our SDB system using a CNC (condensation nucleus counter, model 8020, TSI, Inc.) is portable and can make measurements within several minutes (Yamada et al., 2000). It consists of 8 stages with wire screens of 635 mesh. Size distribution on number concentration can be determined in a range from 3 to 1000 nm. Eight indoor and three outdoor measurements were carried out in Gansu province.

RESULTS AND DISCUSSION

The results of the measurements of indoor radon and thoron concentrations and EETC in Gansu province are summarized in Table 1. The arithmetic means of indoor radon and thoron concentrations were 91 and 351 Bq/m^3, respectively. In all dwellings surveyed, thoron was detected. The maximum thoron concentration was 1471 Bq/m^3. In most of the dwellings, the concentration of thoron was higher than that of radon. The maximum concentration of radon was 229 Bq/m^3. Dwellings with their radon levels

TABLE 1. Radon Concentration, Thoron Concentration, and EETC Observed in Gansu Province

	AM (Bq/m^3)	AM (range)	GM (Bq/m^3)	GSD
Radon concentration	91.2	(21–229)	81.1	1.64
Thoron concentration	351	(30–1,471)	261	2.10
EETC	2.77	(0.8–5.7)	2.60	1.45

Note. AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation.

higher than 200 Bq/m^3, which is the action level provided in ICRP Publication 65 (ICRP, 1993), numbered only 2 out of the total of 102. When the U.S. EPA action level of 4 pCi/L (U.S. EPA, 2004) was used as the selection criterion, the number increased to 15. As shown in Figure 2, a and b, both

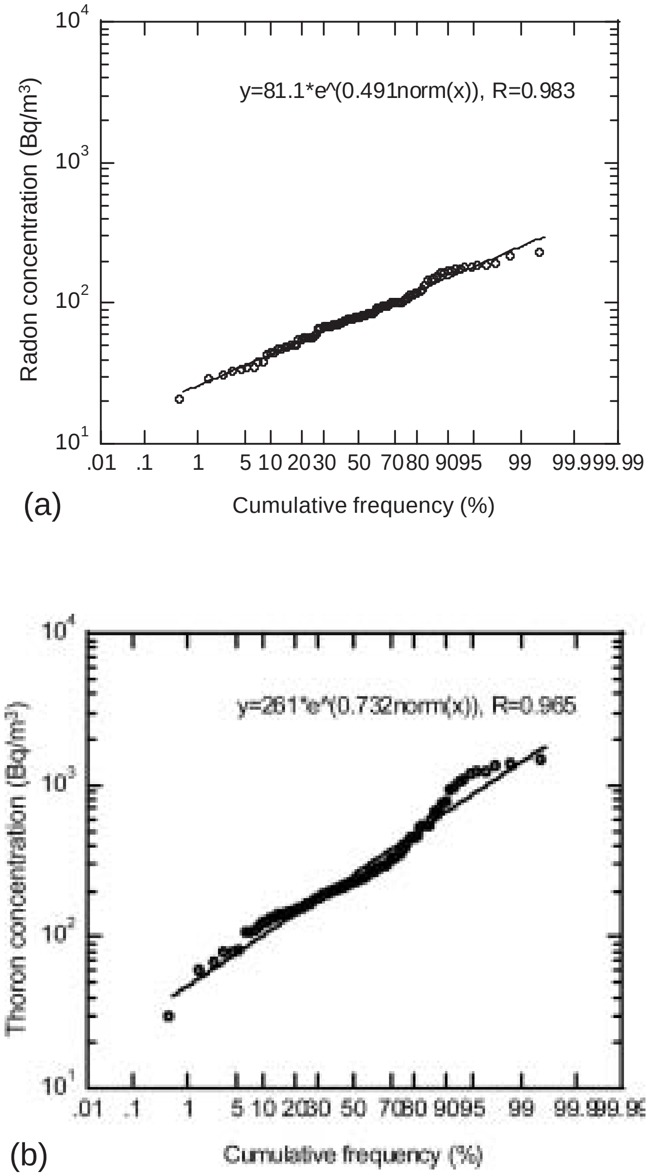

FIGURE 2. Lognormal cumulative frequency distributions: (a) radon concentration, (b) thoron concentration, and (c) EETC.

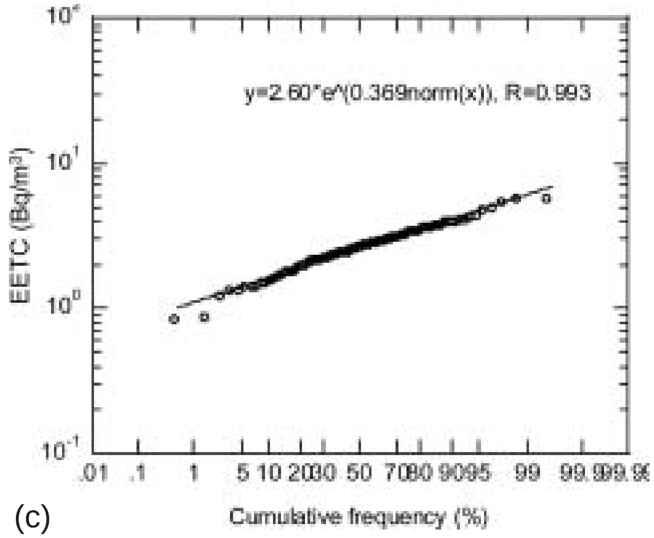

FIGURE 2. *(Continued).*

radon and thoron concentrations showed a lognormal distribution. Geometric standard deviation of radon concentration was evidently larger than that of thoron concentration. Radon and thoron concentrations did not show a significant correlation, and their correlation coefficient, *R*, was .04 (see Figure 3a). While both radon and thoron emanate from the soil and/or wall materials, their concentrations might be quite different since only thoron concentration is strongly dependent on the distance from the emanation point (Doi et al., 1994). Therefore, the thoron measurement data taken by such passive monitors should not be used for estimating the exposure dose due to thoron. The thoron concentration obtained by radon–thoron discriminative measurements should be used for evaluation of a potential thoron emanation. For this reason, we used selective monitors for EETC measurements in indoor environments. EETC levels in cave dwellings of Gansu province showed a lognormal distribution (see Figure 2c), and ranged from 0.8 to 5.7 Bq/m^3. We expected the presence of correlation between thoron concentration and EETC, but our survey showed only a very weak correlation between them ($R = .16$) as shown in Figure 3b, suggesting that EETC cannot be estimated from the observed concentration of thoron by assuming an equilibrium factor for thoron. Furthermore, the mean ratio of EETC to thoron concentration was calculated to be 0.00789 (=2.77/351) using the data obtained from our survey, and the ratios were found to be below 0.03 in 98.7% of the measured dwellings. It should be noted here that the observed ratios are much lower than the frequently assumed value of 0.1, which is provided by the UNSCEAR report (UNSCEAR, 2000), and that small values below 0.01 had been observed. In

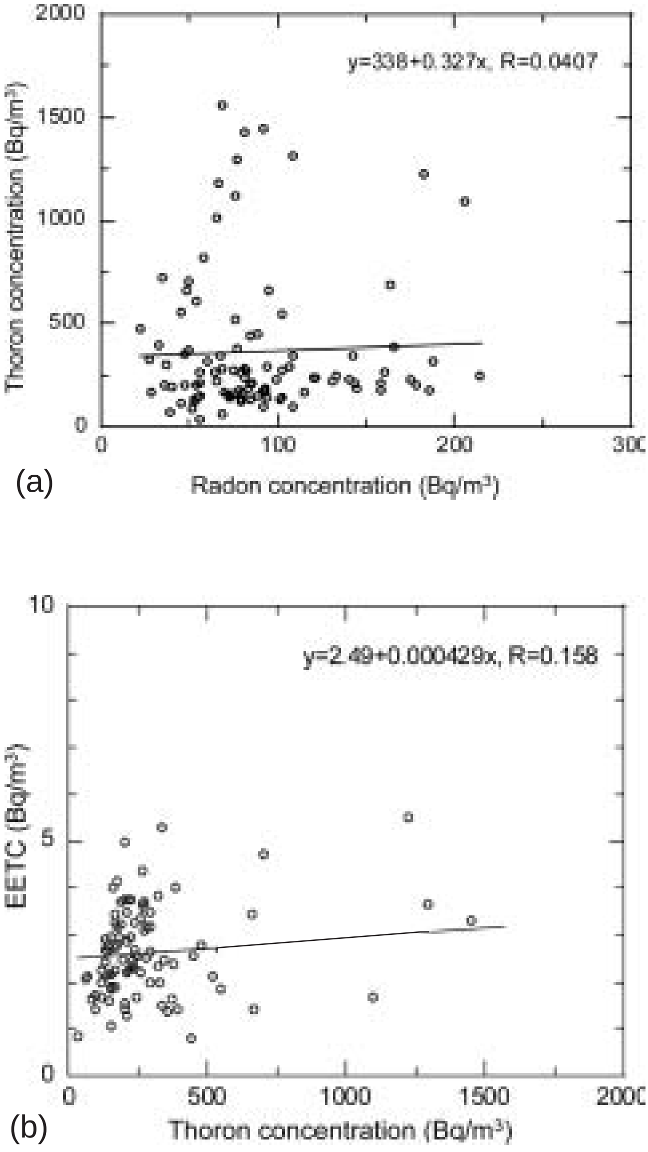

FIGURE 3. Correlation relationships among radon-related parameters: (a) between radon and thoron concentration, (b) between thoron concentration and EETC, and (c) between radon concentration and EETC.

our previous study (Tokonami et al., 2004) conducted in the neighboring provinces of Shanxi and Shaanxi, a positive correlation between radon concentration and EETC was shown. In Gansu province, however, the correlation was very weak (see Figure 3c). High thoron concentration and low levels of the equilibrium factor for thoron were common to both studies.

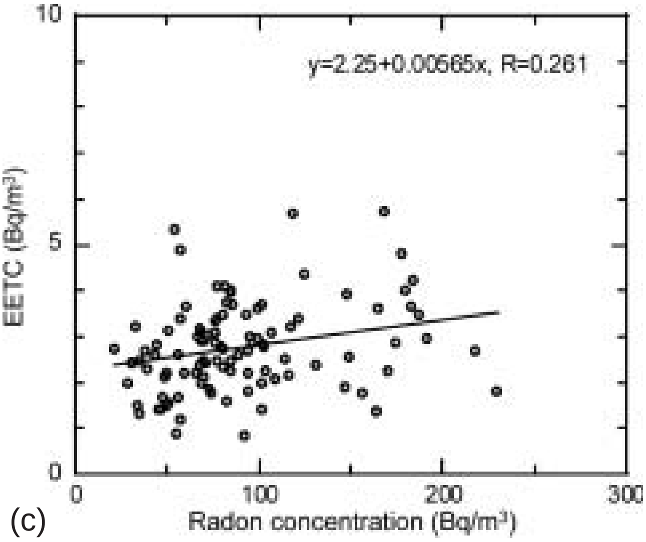

FIGURE 3. *(Continued).*

The mean radon concentration of 91 Bq/m^3 observed in this study was somewhat higher than what was reported in our previous study (64 Bq/m^3), but it was significantly lower than 223 Bq/m^3, a value reported by the LIH–NCI study conducted in the same area of Gansu province. Since the LIH–NCI study used "Radtrak" type detectors, which are sensitive to thoron, the reading might be affected by the presence of thoron (Pearson & Spangler, 1991; Tokonami et al., 2001). Thus we strongly suspect that the study's radon concentration might be overestimated.

The effective dose caused by exposure to the decay products of radon and thoron was estimated using the following equations:

$$H_{\mathrm{Rn}} = \mathrm{EERC} \times t \times \mathrm{DCF}_{\mathrm{Rn}} \tag{1}$$

$$H_{\mathrm{Tn}} = \mathrm{EETC} \times t \times \mathrm{DCF}_{\mathrm{Tn}} \tag{2}$$

where H_{Rn} is the annual effective dose for radon decay products (mSv/yr), H_{Tn} is the annual effective dose for thoron decay products (mSv/yr), EERC is the equilibrium equivalent radon concentration (Bq/m^3), EETC is the equilibrium equivalent thoron concentration (Bq/m^3), t is the time in hours of indoor exposure in a year (=7000 h), DCF$_{\mathrm{Rn}}$ is the dose conversion factor for radon (=9 nSv/h/Bq-m^3), and DCF$_{\mathrm{Tn}}$ is the dose conversion factor for thoron (=40 nSv/h/Bq-m^3). These dose conversion factors are provided in the UNSCEAR report (UNSCEAR, 2000). In this study, EERC was calculated using an equilibrium factor of 0.4 for radon, which is also provided by the UNSCEAR report. Using the data shown in Table 1, the effective dose in indoor environments was estimated to be 2.30 mSv/yr (=91.2 × 0.4 × 7000 × 9/1,000,000) for radon decay products and 0.776 mSv/yr (=2.77 × 7000 × 40/1,000,000) for thoron decay products. The total effective dose was 3.08 mSv/yr. If radon–thoron discriminative measurements had not been carried out in the study, the estimated effective dose might have been quite different from the above value. If Radtrak monitors were used, the indicated radon concentration could have been 326 Bq/m^3 (=91.2 + 351 × 0.669), using Tokonami's conversion factor of 0.669 (=1.88/2.81) (Tokonami et al., 2001) for the calculation. In this case, there is no data on thoron concentration; so the effective dose would be estimated to be 8.22 mSv/yr (=326 × 0.4 × 7000 × 9/1,000,000), which is higher than our estimate by a factor of 2.67 (=8.22/3.08). If discriminative measurements were made without incorporating EETC data, the effective dose for such a case could also be calculated. Assuming an equilibrium factor of 0.1 for thoron provided by the UNSCEAR report, the effective dose for thoron decay products was calculated to be 9.83 mSv/yr (=351 × 0.1 × 7000 × 40/1,000,000). By adding the effective dose of 2.30 mSv/yr for radon decay products, the effective dose in the third case was estimated to be 12.13 mSv/yr (=2.30 + 9.83). There were quite large differences in the estimated effective dose among the measurement methods. As mentioned earlier, the presence of thoron affects both radon measurements and dose evaluation. The significance of thoron measurements, especially EETC measurements, should be taken into consideration more carefully.

In the LIH–NCI study conducted in Gansu province, it was concluded that high levels of residential radon increase the risk of lung cancer. If the actual radon concentration is much lower than the study results show and if thoron is present, the effective dose to cave dwellers would be much lower than their estimates. If both the relative risk of 1.19 at 100 Bq/m^3 estimated by LIH–NCI and the effective dose of 3.08 mSv/yr estimated by this current study are relevant, this would be extremely important as it would mean that there would be a high risk associated with low doses and this in turn would mean an increase in the risk factor for lung cancer.

In order to consider the reason why the relative risk for lung cancer is high in spite of the low exposure dose, exposure conditions related to dose were examined. First, effective doses due to radon and thoron were evaluated. When the dose conversion factors of 0.17 nSv/h/Bq-m^3 for radon and 0.11 nSv/h/Bq-m^3 for thoron provided by the UNSCEAR report were used, the effective doses were calculated to be 0.109 mSv/yr (=91.2 × 7000 × 0.17/1,000,000) and 0.270 mSv/yr (=351 × 7000 × 0.11/1,000,000), respectively. The total dose due to radon and thoron was 0.379 mSv/yr, and it was only about one-eighth of the dose due to their decay products. Second, the dose due to gamma radiation was examined. Gamma dose rates on the order of 100 nGy/h were observed in the cave. The level was slightly higher than average, but the effective dose due to gamma radiation

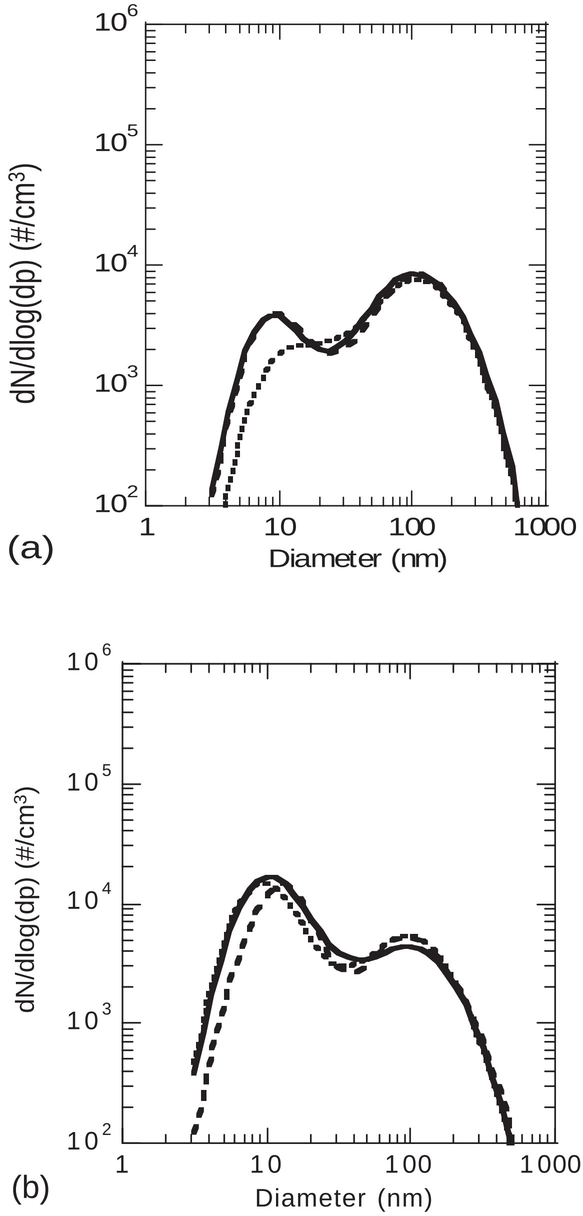

FIGURE 4. Size distributions of indoor aerosols observed in different dwellings.

remained around 1 mSv/yr. Third, the particle sizes of aerosols attached to radon and thoron decay products were examined. The dose conversion factor, DCF, which is used to calculate exposure dose from radon concen-

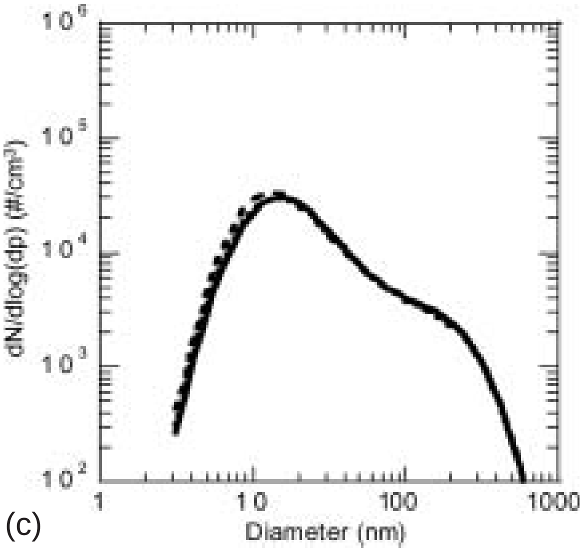

FIGURE 4. *(Continued)*

TABLE 2. Size Distributions of Indoor Aerosols Observed in Gansu Province

		Major fraction			Minor fraction		
Dwelling type	Mode	*N* (/cm^3)	CMD (nm)	GSD	*N* (/cm^3)	CMD (nm)	GSD
Underground cave	Bimodal	5.73×10^3	108	1.82	1.77×10^3	13.4	1.83
Underground cave	Bimodal	7.96×10^3	11.2	1.62	3.06×10^3	89.1	1.79
Aboveground cave	Weak bimodal	2.09×10^4	15.9	1.84	1.37×10^3	103	1.70

Note. *N*, number concentration; CMD, count medium diameter; GSD, geometric standard deviation.

tration, is dependent on the size of the aerosols. According to the calculation using LUDEP (Jarvis et al., 1996), the DCF at 10 nm is about 10 times larger than that at 100 nm (Ishikawa et al., 2001). Indoor aerosols observed in Gansu province had distinctive size distributions. The three distribution curves measured in each dwelling are shown in Figure 4. Most of them were bimodal, and the sizes at the peak were almost constant. The distribution parameters after the peak separation are summarized in Table 2. These values are not activity-weighted but number-weighted. Considering the attachment theory (Porstendorfer et al., 1979) for decay products, the activity-weighted size would increase about two times [$= \exp(2 \times \ln_2 1.8)$] when GSD is 1.8. Even if attachment was considered, smaller size fractions still existed at around 20–30 nm. These smaller size fractions would elevate the exposure dose. At the present time, the contribution of this size effect on the exposure dose has not been estimated with adequate accuracy. However, it is certain that the existence of the small size fraction in decay products elevates the exposure dose. Compared with the gas fraction and gamma radiation, the contribution to dose might be large. If the dose contribution were confirmed, the high risk factor of lung cancer observed in Gansu province might be explained without contradiction.

REFERENCES

Doi, M., Fujimoto, K., Kobayashi, S., and Yonehara, H. 1994. Spatial distribution of thoron and radon concentrations in the indoor air of a traditional Japanese wooden house. *Health Phys.* 66:43–49.

ICRP. 1993. ICRP Publication 65, Protection against Radon–222 at home and at work. *Ann ICRP* 23 (2).

Ishikawa, T., Tokonami, S., Yonehara, H., Fukutsu, K., and Yamada, Y. 2001. Effects of activity size distribution on dose conversion factor for radon progeny (in Japanese). *Jpn. J. Health Phys.* 36:329–338.

Jarvis, N. S., Birchall, A. C., James, A. C., Baily, M. R., and Dorrian, M. D. 1996. LUDEP 2.0, personal computer program, NRPB-SR287. NRPB, Chilton, UK.

Pearson, M. D., and Spangler, R. R. 1991. Calibration of α-track monitors for measurement of thoron (^{220}Rn). *Health Phys.* 60:697–701.

Porstendorfer, J., Robig, G., and Ahmed, A. 1979. Experimental determination of the attachment coefficients of atoms and ions on monodisperse aerosols. *J. Aerosol Sci.* 10:21–28.

Shang, B., Wang, Z., Iida, T., Ikebe, Y., and Yamada, K. 1997. Influence of ^{220}Rn on ^{222}Rn measurement in Chinese cave dwellings. In *Radon and thoron in the human environment*, eds. A. Katase, and M. Shimo, pp. 379–384. Singapore: World Scientific.

Tokonami, S., Sanada, T., and Yang, M. 2001. Contribution from thoron on the response of passive radon detectors. *Health Phys.* 80:612–615.

Tokonami, S., Zhuo, W., Ryuo, H., Yonehara, H., Yamada, Y., and Shimo, M. 2003. Instrument performance of a measuring system with the alpha-track detection technique. *Radiat. Protect. Dosim.* 103:69–72.

Tokonami, S., Sun, Q., Akiba, S., Zhuo, W., Furukawa, M., Ishikawa, T., Hou, C., Zhang, S., Narazaki, Y., Ohji, B., Yonehara, H., and Yamada, Y. 2004. Radon and thoron exposures for cave residents in Shanxi and Shaanxi provinces. *Radia. Res.* 162:390–396.

U.S. Environmental Protection Agency. 2004. 402-K02-006. A Citizen's Guide to Radon: The Guide to Protecting Yourself and Your Family from Radon. Washington, DC: U.S. EPA.

UNSCEAR. 2000. *UNSCEAR 2000 report.* New York: United Nations.

Wang, Z., Lubin, J. H., Wang, L., Zhang, S., Boice, J., Cui, H., Zhang, S., Conrath, S., Xia, Y., and Kleinerman, R. A. 2002. Residential radon and lung cancer risk in a high-exposure area of Gansu province, China. *Am. J. Epidemiol.* 155:554–564.

Wiegand, J., Feige, S., Quingling, X., Schreiber, U., Wieditz, K., Wittmann, C., and Xiarong, L. 2000. Radon and thoron in cave dwellings (Yan'an, China). *Health Phys.* 78:438–444.

Yamada, Y., Tokonami, S., Fukutsu, K., and Shimo, M. 2000. Improvement of the SDB/CNC aerosol sizing system for fast measurement at field. *Radiat. Protect. Dosim.* 88:329–334.

Zhuo, W., and Iida, T. 2000. Estimation of thoron progeny concentrations in dwellings with their deposition rate measurements. *Jpn. J. Health Phys.* 35:365–370.

Zhuo, W., Tokonami, S., Yonehara, H., and Yamada, Y. 2002. A simple passive monitor for integrating measurements of indoor thoron concentrations. *Rev. Sci. Instrum.* 73:2887–2881.