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Review

Researches and Activities on Radon/Thoron and NORM for Past 30 Years in Japan

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This review paper introduces the concepts and backgrounds of several outstanding activities and researches, selected by us, on radon/thoron and naturally occurring radioactive materials (NORM) for the past 30 years in Japan. The content covers regulations on safety management of radioactive residues based on the graded approach; the development of a radon/thoron measurement tool and its worldwide use; experiences on large-scale inter-comparison calibration tests for radon/thoron laboratories, and on editing review books for experts and beginners; and improvement of public literacy on radiation. We believe Japan has been one of the leading countries in radon/thoron and NORM researches and related activities. We hope the experiences and knowledge of Japan will continue to support and help the next generation's development of researches and activities in fields relating to radon/thoron and NORM around the world.

Key words: radon and thoron, NORM, Uranium mine, regulation, calibration, human resource development

1. Introduction

It is easy to list up the various research viewpoints relating to ^{222}Rn (radon) and ^{220}Rn (thoron), which are the largest sources of natural radiation among naturally occurring radioactive materials (NORMs). Representative examples of associated keywords include: measurement methods, inter-comparison tests and calibration, concentration in the atmosphere and water, dose estimation, human effect, geophysical analysis, relation to earthquakes and faults, relation to nuclear power and so on. The research field continues to attract the interest of radiation scientists. For example, the main topics of presentations in Japan Health Physics Society, especially in the period 1990-2010, were on radon/thoron and NORM. We also believe that contents and topics of radon/thoron and NORM are likely to lead not only in the broader fields of environmental radiation but also in general environmental sciences and technologies, and risk management. We select several researches and activities conducted for the past 30 years in Japan and share their history including the concepts of their designs at the time. We are standing at an important point for the next generation of researches and activities

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on radon/thoron and NORM where the risk conversion factor from radon concentration to annual dose is being revised, the international radioactive waste management strategy is being established, and discussion is ongoing on the general safety management for existing exposure situation after the Fukushima Nuclear Power Plant accident. We hope the contents of the review paper will motivate younger researchers to consider and engage more in radon/thoron and NORM research.

2. Management of Radioactive Residues from A Uranium Mine

The first outcropping of uranium ore in Japan was found in Ningyo-toge, located on the western side of Japan. After the discovery of the outcropping, the "Ningyo-toge Office" of Atomic Fuel Corporation (presently "Ningyo-toge Environmental Engineering Center" of Japan Atomic Energy Agency (JAEA)) was opened for uranium exploration in 1957. This initiated research and development of technology for uranium exploration, mining, milling, refining, and so on. The technology for uranium enrichment, refining and conversion for the production of UF_6 had also been developed since the late 1970's[1]. In earlier days, the monitoring of personal external radiation for workers and the measuring of radon by sampling in the working area were conducted according to Japanese mine safety law.

After uranium exploration in Ningyo-toge was terminated in 1987, the main activities of the Ningyo-toge Environmental Engineering Center shifted to maintenance and management of the mining facilities, including remediation activities at the waste rock pile sites and mill-tailings dam.

The waste rock piles were deposited near the gallery entrance and more than twenty sites were left in both Okayama and Tottori prefectures around Ningyo-toge Pass. Many of these sites had been leased lands, and these were returned to the landowners. In 1988, a citizen group found that the radiation levels were high in some waste rock pile sites. The low-grade ore was left in one of the sites, and the neighboring inhabitants insisted for the Japan Nuclear Cycle Development Institute (JNC: the administrator at that time) to remove 3,000 m^3 waste rock from the site. JNC signed an agreement for its removal with the residents' association, which included the neighboring inhabitants, subject to obtaining consent from a local government of the rock's destination. JNC tried to search and determine a domestic destination, however, the negotiation was not easy. The local government, where the site was located, ordered to take the rock out of the site. The other local governments objected to be a destination of the rock. In other words, JNC could not get the waste rock out of the site due to lack of consent from the local governments at that time[2].

The Japanese mining safety regulation were revised in 1989 according to International Commission on Radiological Protection (ICRP) Publication 26 and the measurement of radon in air and the evaluation of the exposure have since been required. Consequently, various radon concentration measurements in the environment were started in 1989. The measurement points for radon concentration in air were chosen in and around waste rock sites and in residential areas. Long-term concentration measurement by passive methods, continuous measurement by active instruments, and measurement by sampling method were carried out. The concentration of radon in air measured by the passive method exceeded 200 Bq m^{-3} in the waste rock sites but was much lower out of the sites[3]. Later, a continuous radon progeny monitor with a vacuum vessel was developed to measure the variation of the concentration of radon progeny, as Equilibrium Equivalent Concentration of Radon (EEC Rn), by alpha spectrometry[4]. Also, an integrating radon progeny monitor for environmental monitoring was developed to measure the monthly average because the value is directly related to exposure. The data obtained around the waste rock pile sites were comparable to those of controlled places in the city[5]. There are no residents in the vicinity of the sites. However, assuming people lived there, the effective dose outside of the sites and caused by the waste rock pile was estimated, according to Japanese regulation, as less than 1 mSv a^{-1}.

The demand for the removal of the waste rock by the neighboring inhabitants developed into a legal suit. It was not a matter of safety but a matter of contract. A long time had passed, and the final decision given by a court of law was to remove the 3,000 m^3 waste rock. Eventually, it was decided to adopt an option that combines exporting 300 m^3 low-grade ore to the United States in 2005 and recycling the other waste rock as bricks for domestic use.

The remaining waste rock, with uranium concentration around 1 Bq g^{-1}, were processed to bricks as consumer goods of less than 1 Bq g^{-1} according to Basic Safety Standards (BSS) of International Atomic Energy Agency (IAEA) and the bricks sold out in 2011. The major remedial action was to cover the waste rock sites with weathering granite soil. The action decreased the radon concentrations in air on the sites to around 10 to 40 Bq m^{-3}. On the other hand, after the action the radon concentrations in air around the sites were the same as before[6]. The measurements of external radiation, radon, uranium, and radium concentration in the environment around waste rock pile sites have been carried out periodically, and the results checked by experts and published.

Other facilities are also located in the Ningyo-toge Environmental Center: a conversion plant and two

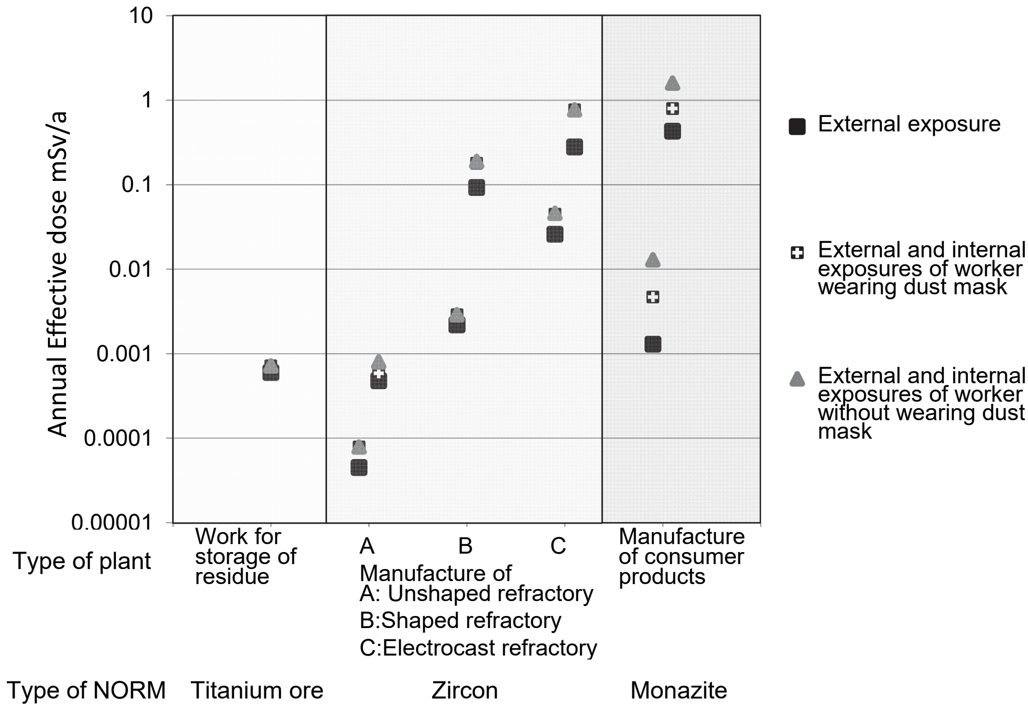

Fig. 1. Assessment of external and internal doses to workers in various NORM industries.

enrichment plants which are under decommissioning; and storage facilities for uranium waste which are being maintained. Now the purpose of the Ningyo-toge Environmental Center is the decommissioning of these facilities including environmental remediation of the remaining mill-tailings dam.

A Technological Advisory Board on the Reclamation of the Ningyo-toge uranium mining and milling facilities consisting of experts and local residents was set up to discuss how to maintain the remaining mining facilities, and it evaluates the activities from an objective point of view since 2001 and reports their findings every year to public[7].

3. Dose Assessment of Workers and Pubic Using NORM and Regulatory Framework of Japan

Japan is deficient in mineral resources and most mines for various resources, except limestone and small number of precious metal, were closed more than 20 years ago. The significant issue on NORM regulation would be exposure of workers handling various types of industrial raw materials such as monazite, zirconium, titanium, bastnaesite, phosphate ore, etc. Several studies were carried out in order to investigate the exposure situation for workers handling industrial NORM. In the studies, measurements of dose rate in air and concentration of airborne natural radionuclides including radon/thoron decay products at typical points in workplaces were performed[8, 9]. The annual effective dose for each group of workers was estimated using results of the measurements and information on the working conditions with parameters used in the report of European Commission, RP122[10]. Although exposure conditions such as working time, distance between worker and materials, and volume of the materials were quite different for different workplaces and types of worker, the average effective doses of the highest exposure group and lowest exposure group were assessed for every type of plants except for workplaces involving storage of titanium ore.

Results of the dose assessment are summarized in Figure 1. The results reveal that the annual effective doses vary widely due to differences in materials and conditions of work such as volume of the material, distance between source and worker, and working time. In most cases, levels of annual effective dose were estimated to be less than the order of 1 mSv a^{-1} which is the dose criterion provided in the International BSS[11] for exemption of bulk amounts of materials containing natural radionuclides. From the results of the studies, annual effective dose exceeded 1 mSv a^{-1} only in the most critical case where the worker handles material containing more than several hundred Bq g^{-1} of natural nuclides from ^{232}Th series.

In Japan, some people believe that there is a health effect of low dose radiation exposure and that various

Table 1. Results of dose assessment of user of consumer goods containing NORM

Consumer goods	Activity concentration of NORM		Annual effective dose of user	Equivalent doses to the skin.
	^{238}U(^{238}U series)	^{232}Th(^{232}Th series)		
	Bq g^{-1}	Bq g^{-1}	mSv a^{-1}	mSv a^{-1} in mm^2
Bedclothes [12]	0.043 – 0.26	0.01 – 2.3	0.11	-
Underwear [12]	1	8.8	0.22	-
Radon spa bathing apparatus [12]	10 – 34	81 – 270	0.11	-
Wall material [12]	0.58	3.1	0.01	-
Cosmetics (powder) [13]		68	5.5 (Internal exposure due to inhalation of powder)	-
Bracelet(mineral) [14]	41 – 140	20 – 810	< 0.01 – 0.3	4 – < 20
Jewel(mineral) [14]	0.42 – 300	5 – 6.8	0.02 – 0.05	70 – 280

Table 2. Regulatory criteria of nuclear source material

·Activity concentration:
> 74 Bq g^{-1} (liquid or gas) or > 370 Bq g^{-1} (solid)
·Amount of uranium and thorium (*) > 900 g
(*) the sum of the amount of thorium and 3 times of the amount of uranium

type of consumer goods containing NORM such as monazite are distributed to public. The studies for dose assessment of the public using such kinds of consumer goods have been carried out. Representative examples of the results of the studies[12-14] are shown in Table 1.

It is difficult for workers in the NORM industry to monitor their own exposure dose and measure activity concentrations of the materials because they are not experts of radiation protection. Thus, a database was developed to distribute the integrated information to them[15]. For this database, industrial raw materials were collected, and the activity concentrations of natural nuclides such as ^{238}U, ^{232}Th series and ^{40}K were measured by using inductively coupled plasma mass spectrometry and gamma ray spectrometry. The database includes the information of the measured results as well as data in the literature.

Regarding the regulation of NORM in Japan, the regulatory criteria for nuclear source material except nuclear fuel material are provided by "the law for the Regulations of Nuclear Source Material, Nuclear Fuel Material and Reactors" as shown in Table 2.

Issue of NORM regulation became a subject of deliberation of the Administrative Group established under Radiation Council, and "Report on Exemption of Regulations on Naturally Occurring Radioactive Materials (NORM Report)"[16] was issued in 2003. The report showed regulatory criteria for 8 groups of NORM categorized according to amenability of regulation.

In 2009, "Guideline for Ensuring Safety of Raw Materials and Products Containing Uranium or Thorium"[17] was developed by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) based on the principle of the NORM Report. The guideline does not define radioactive concentration as the criteria for control but designates the category of materials subject to the guideline. The designated category for materials was selected from materials for which the activity concentration exceeds 1 Bq g^{-1}. The criterion of 1 Bq g^{-1} was proposed in the document of IAEA RS-G-1.7[18] from viewpoint of exclusion. Information on the dose assessment is not sufficient to establish the regulation system. The guideline is not legally binding and requires manufacturers and importers to conduct self-management for handling and disposal of the materials, and for exposure of their workers. If the dose to workers evaluated by manufacturers and importers exceeds 1 mSv a^{-1}, countermeasures to reduce the exposure of their workers are required by the guideline. In the current regulation system, there is a problem that workers in NORM industries who receive more than 1 mSv a^{-1} would not be subject to legal regulation as occupational exposure. The NORM report and the guideline should be reviewed for revision based on the new system provided by ICRP publication 103[19] with concept of exposure situations.

4. Development of Radon and Thoron Discriminative Monitors, and Their Applications

The presence of radon and thoron in the indoor environment creates a major public health risk[20]. In the framework of public health protection from radon and thoron, many surveys have been performed to estimate radon and thoron concentrations in many countries. In Japan, there were 3-period of the nationwide and regional radon surveys using passive radon and thoron monitors.

The first nationwide radon surveys were conducted at 5,700 dwellings in the late 1980s and early 1990s [21, 22] using a Karlsruhe-type passive radon detector (a KfK-type radon monitor)[23] which could not discriminate radon from thoron. The arithmetic mean of residential radon level was 20.8 ± 18.8 Bq m^{-3}. Moreover, radon concentrations in wooden houses show regional differences closely related to granite distribution, whereas radon concentrations in non-wooden houses show no typical pattern related to geology. However, the radon measurements in the first survey were found to contain significant thoron interference due to the high air exchange rate of KfK-type radon monitor[23]. Moreover, an electrostatic integrating radon monitor was developed for the environmental radon monitoring in 1988 by Iida *et al.*[24] The collecting electrode method was used based on calculating the internal electric field and a drying agent of phosphorus pentoxide (P_2O_5), which was placed in the bottom of the monitor. This monitor has a high sensitivity and low detection limit (1.2 Bq m^{-3} for an exposure time of 2 months). However, its use extended only to performance tests in six locations: in the plains of Nagoya and Kasugai; in the mountainous regions of Nakatsugawa and Shidara; on the seaside of Saku island; and in Sapporo, Hokkaido, which has a cold climate. In this small radon survey, the annual mean radon concentrations for indoor and outdoor air ranged from 3.7 to 9.5 Bq m^{-3} and 6.4 to 11.9 Bq m^{-3}, respectively. This device is not suitable for nationwide surveys due to the limitation of the method which include requirements for a high voltage and a drying agent, and the dependence of the $^{218}Po^{+}$ electrostatic collection efficiency on the humidity of the air.

The second nationwide indoor radon survey was carried out at 900 houses in 47 prefectures during 1993-1996[25] with radon-thoron discriminative passive radon detectors known as UFO detectors[26]. The annual mean indoor radon concentration was found to be 15.5 ± 18.8 Bq m^{-3} which was 5 Bq m^{-3} lower than the results obtained by the first nationwide indoor radon survey. The data in the second survey were used only for estimating the average radon concentration in each region in Japan. Seasonal variation of the radon concentration was found to be high in October to December and low in summer from July to September. The variability of the mean radon concentration among the house types was found that wooden houses (12.9 ± 18.8 Bq m^{-3}) have lower radon concentration than concrete (23.1 ± 15.5 Bq m^{-3}) or concrete block houses (42.5 ± 55.4 Bq m^{-3}). Likewise, the nationwide outdoor radon concentrations in 47 prefectures in Japan were also measured by UFO detectors in 1997-1999[27]. More than 70% of radon monitors were installed at the bare ground. The outdoor radon concentrations ranged from 3.3 Bq m^{-3} in the Okinawa region to 9.8 Bq m^{-3} in the Chugoku region due to the geological characteristics. The arithmetic mean of outdoor radon in Japan were evaluated as 6.1 ± 1.9 Bq m^{-3}, which was 40% of the indoor radon concentration in Japan. Additionally, a nationwide indoor-workplace radon survey was carried out at 705 sites of offices, factories, schools, and hospitals in 2000-2003[28] using UFO detectors. The indoor radon concentrations in workplace ranged from 1.4 to 182 Bq m^{-3} with the arithmetic mean of 20.8 ± 19.5 Bq m^{-3}. The arithmetic mean radon concentrations evaluated at offices, factories, schools, and hospitals were 22.6 ± 17.0, 10.1 ± 8.3, 28.4 ± 24.5, and 19.8 ± 24.4 Bq m^{-3}, respectively. However, the major drawbacks of UFO detector were the large size (The two opposed hemispherical stainless-steel diffusion chambers of 120 mm and 75 mm diameter) and a complicated structure, and the entry of thoron had not been studied well.

The third nationwide radon survey was conducted from 2007 to 2010 at 3500 houses in 47 prefectures[29] by the National Institute of Public Health (NIPH) using radon-thoron discriminative monitor (RADUET) which developed by Tokonami *et al.*[30] The objective of this survey was to confirm the increasing radon concentration in homes constructed after the mid-1970s. After correcting for seasonal fluctuation, the arithmetic mean of indoor radon concentration was 14.3 ± 14.7 Bq m^{-3}, which was similar to the second survey, was the highest in the houses constructed in the mid-1980s and decreased thereafter.

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in 2011, the radon concentrations were measured by passive radon-thoron monitor (RADUET) and a pulse-type ionization chamber at temporary housing facilities, apartments and detached houses in Fukushima Prefecture by Hosoda *et al.*[31] The mean radon concentrations at temporary housing units, apartments and detached houses, were 5 ± 3, 7 ± 3 and 9 ± 8 Bq m^{-3}, respectively. The overall mean value of indoor radon concentrations was 7 ± 3 Bq m^{-3} (Ranged: < 3-28 Bq m^{-3}). According to the third nationwide radon survey[29], the mean of the indoor radon activity concentrations in Fukushima Prefecture dwellings was ~10 Bq m^{-3} after adjusting for seasonal fluctuation. Moreover, the average value of indoor radon concentration obtained in this study was close to the reported mean value of outdoor radon activity concentration in Fukushima Prefecture: 6.3 ± 2.6 Bq m^{-3} [27]. Assuming the inhabitants lived in these accommodations for one year, the mean annual effective doses due to indoor radon in all housing types was 0.22 ± 0.12 mSv (ranged < 0.12 - 0.83 mSv). These doses were all lower than the reported Japanese average: 0.45 mSv (15.5

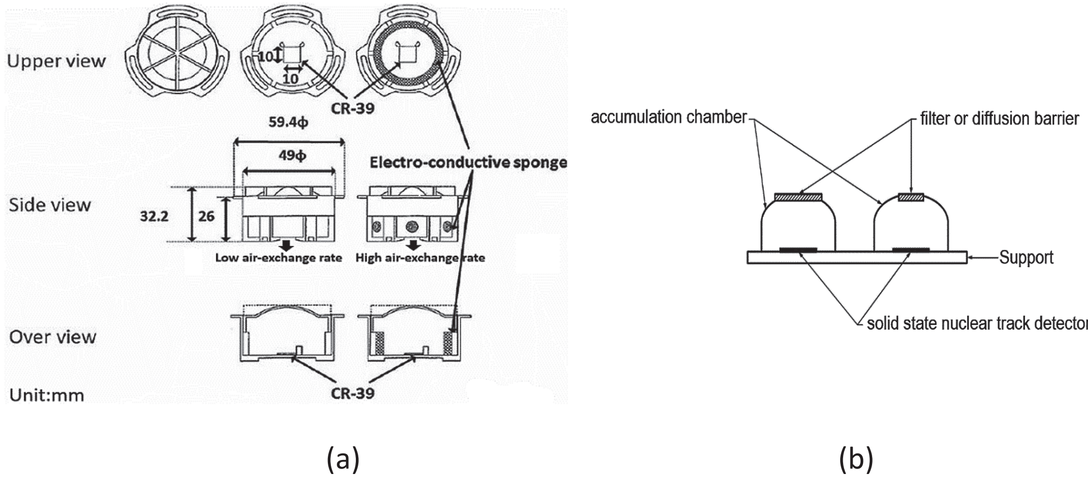

Fig. 2. Overview of (a) RADUET (b) design of Radon-Thoron discriminative measuring device.

Bq m^{-3}) in the second nationwide survey[25].

After the success in previous radon surveys, Japan has been one of the leading countries to develop radon/thoron detection instruments. REDUET has been one of the most popular radon and thoron discriminative detectors used for small and large national survey in various countries. Examples of such countries are China[32, 33], India[34, 35], Indonesia[36], Cameroon[37-41], Australia[42] and Hungary[43] which conducted long term measurement using RADUET for measuring radon and thoron concentration at high natural background radiation (HNBR) areas. In addition, RADUET were deployed for assessing indoor radon levels in dwellings of Chiang Mai, Thailand where high numbers of new cases of lung cancer patients[44] had been observed. Moreover, RADUET was used for the nationwide indoor radon and thoron survey in Canada[45, 46]. Furthermore, the national survey for radon and thoron in public schools and local governmental offices has been done using RADUET in South-Korea[47]. Besides, the regional survey of radon and thoron levels in different types of modern houses in Kenya[48, 49] was also conducted by using RADUET monitors. In additional application, RADUET was deployed for measuring radon and thoron in soil gas[50] around the south-west of Belgrade in Serbia.

Figure 2 shows and illustrates an overview of (a) RADUET (b) designs of Radon-Thoron discriminative measuring device[51], which was originally developed in Japan. RADUET and other designs of Radon-Thoron discriminative detectors compose of dual accumulation chambers with the same geometry but with different air-ventilation or air-exchange rates. Each chamber is equipped with a solid-state nuclear track detector fixed at the bottom. RADUET uses the allyl diglycol carbonate chip (CR-39) to detect alpha particles emitted from thoron, radon, and their decay products. The dual accumulation chambers of RADUET are made of electro-conductive plastic with the inner volume of 30 cm^3 [30]. Each accumulation chamber has a filter through which thoron and radon diffuse (Fig. 2 b). This filter is set to prevent the access of aerosol in air and thoron and radon progenies at the time of sampling. Radon diffuses into the accumulation chamber with low air-exchange rate through a strong diffusion barrier: the invisible air gap between the lid and the chamber in the design of RADUET; consequently only negligible thoron gets into the chamber because of its short half-life of 55.6 sec when compared to radon (3.82 days). For RADUET accumulation chamber, with high air-exchange rate, both thoron and radon gases can go through by natural air exchange due to the chamber's structure which has 6 holes of 6 mm in diameter that are covered with an electro-conductive sponge and opened at the side wall of the chamber. On the one hand, the high air-exchange rate should be as high as possible (thoron and radon calibration factors are nearly the same). On the other hand, the low air-exchange rate should be as low as possible by using a strong diffusion barrier; nonetheless, it should be mentioned that the diffusion barrier or the filtering medium must not block the air renewal. The air-exchange rates of two accumulation chambers of RADUET differ by the two orders of magnitude[52, 53] as a consequence of structural differences, so radon and thoron can be discriminated.

For analysis of CR-39 in paired low and high air-exchange rate chambers of RADUET, the CR-39 detectors are chemically etched according to manufacturer's instructions such as: a 6.25M NaOH solution at 90C for 6 hours (CR-39s manufactured by Radosys Ltd.

Hungary) or a 6M NaOH solution at 60 °C for 24 h (CR-39s manufactured by Nagase Landauer, Ltd. Japan). Subsequently, the detectors are washed with water several times and kept for 5 min in the distilled water (with ultrasonic) to remove all unwanted waste materials from the surface of the detectors. After that, a commercial alpha-track reader or optical microscope and imaging software are used to obtain the total track density of each CR-39. With these raw data (the total track densities) of low (N_L) and high (N_H) air-exchange rate chambers and the calibration coefficients determined by exposure to known radon and thoron concentrations in a reference laboratory, the radon and thoron concentrations are calculated as the solution of Equations 1 and 2.

$$N_L = R_n \times CF_{Rn1} \times T + T_n \times CF_{Tn1} \times T + B \tag{1}$$

$$N_H = R_n \times CF_{Rn2} \times T + T_n \times CF_{Tn2} \times T + B \tag{2}$$

where Rn and Tn are the mean concentrations of radon and thoron during the exposure period in kBq m^{-3}. CF_{Rn1} and CF_{Tn1} are the radon and thoron calibration coefficients for the low air-exchange rate chamber expressed as tracks of cm^{-2} kBq^{-1} m^3 h^{-1}. CF_{Rn2}, and CF_{Tn2} are the radon and thoron calibration coefficients for the high air-exchange rate as tracks cm^{-2} kBq^{-1} m^3 h^{-1}. T is the exposure time in hours and B is the background track density of CR-39 detector in cm^{-2}. The strength of RADUET detector is cost effective, easy to handle and transport and hence often employed for radon and thoron measurements in nationwide and regional surveys. The weakness can be found in long-term measurements due to the basic tenacity of the registered alpha tracks because this type of monitor can provide only an average activity concentration for the corresponding period. In addition, as the response of RADUET to thoron depends on wind field[54], caution must be paid to the measured value of thoron.

5. Calibration for Radon Measurement

Calibration and Intercomparison Exercises

Calibration is important for radon concentration detectors as well as other radiation detectors. Radon is a noble gas and alpha-emitter nuclide with 3.8 days half-life. Therefore, an ionization chamber is often used as a standard instrument in calibration for radon monitors. The introduction of radon gas generated from a radium source into an ionization chamber in order to calibrate radon concentration detectors is complicated[55] and, therefore, strongly needs high knowledge on and skills for handling radon.

On the other hand, radon progeny (^{218}Po, ^{214}Pb and ^{214}Bi) are solid, short-lived nuclides in air, easily influenced by air quality and circumstance. Radon progeny are generally sampled by filtering method, and a calibration test for progeny measurement needs more detailed attention than a test for radon measurement.

In 1980s, radon concentration had been measured in many countries. For the purpose of guarantee of reliability in radon measurement, Environmental Measurement Laboratory of Department of Energy, US (DOE-EML), Nuclear Radiation Protection Board, UK (NRPB) and Australia Radiation Laboratory (ARL) at the time were nominated as the standard laboratories by IAEA. Other laboratories in the world were requested to calibrate their detectors with one of the standard laboratories[56]. The inter-comparison test was performed by the grab sampling method. A laboratory received a standard radon gas collected in a good airtight chamber from one of the standard laboratories, and the laboratory estimated the concentration of the radon in the test.

In Japan, the radon calibration using ionization chambers was independently carried out by Nagoya University (NU)[57], Kyoto University (KU), Waseda University (WU) and Power Reactor and Nuclear Fuel Development Corporation (PNC) at that time[58]. Results of calibration tests among the four laboratories agreed well[57]. Several laboratories in Japan have repeatedly compared with one of the four laboratories. Many laboratories participated in domestic comparison tests for radon measurement[58-62] as well as in international inter-comparison exercises hosted by world standard laboratories. As a result, each laboratory in Japan demonstrated its reliability on radon measurement[57]. However, a passive radon monitor (UFO detector) that was widely distributed by National Institute of Radiological Sciences (NIRS) (currently National Institutes for Quantum and Radiological Science and Technology: QST) in Japan showed systematic difference of 20-30 percent from EML and ARL.

Under this condition, eleven laboratories in Japan planned an exercise of radon measurements at EML of US. This was not a calibration exercise by the blind method, but an intercomparison exercise to clarify the degree of agreement of values among participants based on the EML guide. The results of measurements obtained by the Japanese laboratories agreed well, but their average value showed 5 percent less than the EML's value[63].

We would like to list three points for a calibration exercise on radon measurement. First, participants must select either a blind test or an intercomparison exercise based on the purpose of the calibration. Second, all the necessary conditions and observational matters for the calibration should be accepted among all participants. Third, participants should openly show the results of their estimation, and it is important to discuss the results frankly among them.

Radon Facility of NIRS

In Japan, NU, WU and PNC had facilities only for radon concentration calibration, but not for radon exposure. In 2000, NIRS built the Radon Facility for radon exposure exercises with a walk-in type chamber (observation room)[64, 65]. This chamber consists of the following: a radium source for supplying high radon concentration, a monitor to control radon concentration, a system supplying aerosols for radon progeny experiments, and a control system for temperature and humidity in the chamber. Researchers and technicians in Japan have used this facility for calibration of radon detectors and various experiments on radon and its progeny[66-72].

6. Editing Review Books for Experts and Beginners for Radon Research

From 1970s to 1990s, many recommendations and reports on the effect of exposure to radon in environment were published by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR)[73-76], the International Commission on Radiological Protection (ICRP)[77-79], the National Council on Radiation Protection and Measurements (NCRP)[80, 81], the National Research Council (NRC)[82, 83], the World Health Organization (WHO)[84] and the Nuclear Energy Agency of the Organization for Economic Co-operation and Development (OECD/NEA)[85]. Thus, interest in radon was generated worldwide and many researches and surveys on radon measurement and/or dose estimation were carried out. Research on radon was also very active in Japan at the time, and this situation became a motivation for establishing a system of collaboration and cooperation among researchers and experts. In addition, radon and/or thoron symposia, seminars and workshops were frequently held after 1977; those are recorded chronologically in the preface of the seminar report "Radon and Thoron —Opportunities, Properties and Health Effects—"[86].

These activities can, individually or as a compilation in a book, provide an outlook on development and calibration of radon, concentration measurement in outdoor and indoor environments, and dose evaluation. Reports of symposia and/or seminars, reports from universities and institutes were the main focus of the current compilation. Topics were extracted from two big symposia that were held in 1991 and 1997: the Symposium on Environmental Radon[87] by the Committee on Radon, and the Radon and Thoron in The Human Environment[88] by the 7th Tohwa University International Symposium respectively. Other reports written by Japanese were "Radon and Its Daughter Nuclides -Measurement and Calibration-" by Japan Health Physics Society (1988); "Radon, Occurrence and Measurement of Thoron and Their Daughter Nuclides" by Japan Nuclear Regulation Authority (1991); "Inter-comparison of Radon, Thoron and Their Daughter Nuclides" by Electron Science Institute (1993); "Characteristics and Behaviour of Thoron and Their daughters" by Research Reactor Institute, Kyoto University (1995); "Occurrence, Measurement and Exposure Evaluation for Radon Families", by ibid (1996); "Report of Radon Concentration Measurement and Dose Estimation" by National Institute of Radiological Sciences (1998); and "Exposure Evaluation of Human to Radon" by Japan Health Physics Society (1998). We could identify examples of radon researches at the time shown in their titles and years.

"Atmospheric Radon Families and Environmental Radioactivity I, II and III"[89-91] were respectively published in 1985, 1990 and 1995 by the Research Committee on Atmospheric Radon Families. The main radon subjects in the books were: measuring methods, inter-comparison and calibration, concentration data in atmosphere and water, dose estimation, human effect, geophysical analysis, relation to earthquakes and faults, and relation to nuclear power.

The reports clearly show the trends of radon research, identify unknown problems, and suggest the latest direction of related researches. Therefore, we should extensively watch out not only for peer-reviewed papers but also other reports in the field of radon research.

According to the above description, interest for radon continued until 2000, and many reports have been produced at intervals of a few years. However, no symposium and/or seminar was held in Japan after 2000 until now, except for special committees on radon in water (2004) and on protection level of radon (2012), both held by Japan Health Physics Society[92, 93]. We expect that an international symposium or seminar on radon, thoron and their progeny will be held in the near future.

7. Radon and NORM for Improvement of International Radiation Literacy

We hope that radon and NORM will play a major role not only from the viewpoint of improving radiation literacy but also from viewpoints of general risk management and improving environmental risk literacy. Here, we will introduce a case where radon/thoron and NORM are effectively applied in the secondary education of nuclear science and technology in the Asia-Pacific region.

IAEA introduces one of their activities in an article[94]: "The International Atomic Energy Agency (IAEA) has been strengthening the education of NST in secondary schools to support sustainability of applications of nuclear technology in member states. The implementation of the

IAEA Technical Cooperation (TC) project RAS/0/065 during 2012-2016 provided valuable proficiency in successfully introducing NST in secondary schools in the Asia-Pacific region in selected pilot countries. ⋯ The project led to development of education materials, hands-on exercises, as well as co-curricular activities which made nuclear concepts more interesting to students." In this project which is still continuing as RAS/0/079 (2018-2021), Team JAPAN, consisting of several Japanese experts led by The University of Tokyo, have been supporting activities and developing several educational tools and modules with "WOW factor", which is for example defined by Cambridge Dictionary as a quality or feature of something that makes people feel great excitement or admiration, using the STEAM (Science, Technology, Engineering, Arts and Mathematics) education concept[95]. Feedback from official pilot activities in the eight countries (the Philippines, Indonesia, Malaysia, Thailand, Sri Lanka, Jordan, Oman and Mongolia) has motivated Team JAPAN to expand its perspective to develop new educational tools and modules adopting radon and NORM[96]. One of main development examples is the Peltier cooling type cloud chamber, with a large area of view being capable of accommodating five to six students at a time, whose main radiation sources are radon/thoron gases and their decay products[97]. It inspires a "WOW" effect in the observers as they realize the existence of natural radionuclides and the ensuing radiation. They also learn the type and range of radiation, the half-life of radioactive materials, and the decay series of radon and thoron. They will study the history of Nobel Prize related to the cloud chamber and will have a longing for scientific and technological progress. In addition, a next-generation environmental radiation survey meter named as KIND-pro and KIND-mini also became effective as educational tools for identifying and understanding natural radiation and its dose-rate level. These are based on Csl (Tl) or plastic scintillator modules designed to fit Information and Communication Technology (ICT) education. It is important to observe the spatial variation of radiation dose in natural environments, and to analyze and compare various risk sources including radiation. Radiation sources made of NORM, consisting on pressed powder of coffee beans, mineral deposits from hot springs, etc., could become simplified calibration tools for educational survey meters. These can be used anywhere by anybody, even outside of radiation-controlled areas. All of the above have been developed by Team JAPAN and the technical knowledge and concepts were shared with pilot countries.

We consider that the how-to for obtaining educational tools and radiation sources suitable for schools and for safely handling them is an important subject for future study. Radon/thoron and NORM would be simple, convenient, and informative items for public education.

8. Conclusions

The review paper selected and introduced several outstanding activities, concepts, and backgrounds on radon/thoron and NORM researches for the latest 30 years in Japan. The content covers safety management of radioactive residues; regulations based on the graded approach; a radon/thoron measurement tool used worldwide; experiences of large-scale inter-comparison calibration tests and of editing review books for experts and beginners; and improvement of public literacy on radiation. We believe Japan has been one of the leading countries in the radon/thoron and NORM researches. We hope the experiences and knowledge of Japan could continue to support and help the next generation's development of researches and activities in the field relating to radon/thoron and NORM in the world.

Conflicts of Interest

The authors declare no conflict of interest.

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