Impact of Radon from Closed Uranium Mine

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INTRODUCTION

In the Ningyo-toge Environmental Engineering Center, research and development projects related to the uranium exploration were carried out from 1967 to 1988 measuring radon concentration in the working places by sampling method using an ionization chamber. "The waste rock pile sites" which were piled up the rocks left by the exploration activities have been maintained following the Japanese Mine Safety Law in the closed mining areas around Ningyo-toge. And the measurements of radon in the environment and the evaluation of the exposure have been necessary by the Japanese regulation since 1989 according to the ICRP Publication26. Therefore, for the purpose of evaluating the environmental impact from the waste rock piles, measurements of environmental radon and dispersion calculation were carried out. The impact was also checked by the tracer gas dispersion experiments at one of the typical waste rock pile site, and the validity of the model calculation was examined.

TRACEABILITY FOR THE MEASUREMENT[1)]

THE STANDARD INSTRUMENT

For the measurement of environmental radon as a standard instrument of radon, gas charging type ionization chamber and vibrating reed electrometer with charge storage method have been used. The calibration of the ionization chamber uses radon which emanates from the radium solution of low concentration with the certifications as a primary standard, and soundness of the electrometer has been checked by the standard current generator periodically. It is confirmed that the ionization chamber has a good agreements by the intercomparison in October, 1995 with U.S. EML that is the radon standard organization of OECD-NEA[2)]. And, the similar result was obtained by the similar intercomparison held in Japan[3)].

RADON CHAMBER

In order to maintain radon monitors the practical radon reference chamber is used. The 7.5m^3 radon reference chamber shown in Fig.1 is cylindrical, lying horizontal, and the radon concentration can be achieved to about 1,000 Bq.m^{-3}, and the temperature and humidity are controllable. Since the atmospheric pressure is about 10% low because the site location is 700m above sea level, the chamber was structurally manufactured to resist pressure of even 1 atm. Solid radon source of natural soil containing about 0.6 Bq.g^{-1} radium sampled from waste rock is used. In addition, there are flanges for sampling the air in the chamber, gloves and a window for the observation on the wall surface of chamber. And tubes to supply radon gas and aerosol are set on the air loop system for the circulation.

EVALUATION BY MEASUREMENTS

INTEGRATING MEASUREMENT

The measuring instrument has adopted electrostatic integrating passive monitors, which was developed by Iida et al.[4)] and is produced by Aloka Co.Ltd.(Japan) in order to measure low concentration radon of the field at the good accuracy. The monitor is set in a naturally ventilated case, the height of which is about from 1 to 1.5 meters. The data are mainly obtained quarterly. The etch pit detector was cellulose nitrate film at first but we changed to CR39 because of the secure quality.

The measurements have been carried out in and around the waste rock pile sites and in residential district such as in the garden of community house or shrine in both Tottori and Okayama prefectures near the Ningyo-toge Engineering Center shown in Fig.2. We have been taken measurements at 2 points near the center and boundary in every waste rock site as a rule, and in comparatively large waste rock site, 5 points measurements are carried out in the north, south, east and west and center. The measurement result in 1996 fiscal year for the environmental monitoring of the waste rock pile site is shown in Fig.3. In the residential district except for Sugegawara, the measurement results are less than 30 Bq.m^{-3}, and seem to be almost same level as reported data[5)] in Japan. Relatively high concentration is observed in Sugegawara compared to other district. In comparison with the residential district the concentrations which exceeds 100 Bq.m^{-3} in the waste rock sites are observed.

The detailed measurements were carried out in Sugegawara district where the a little high concentration in the residential district was observed, in Katamo district as a typical field around the waste rock site, and the districts as a reference field. These results are shown in Fig.4. As for Suegawara in Kurayoshi typical granite zone there is no uranium ore deposit, and higher concentration has been obtained even at the other reference points (R-55, R-58) in the same district, it seems to be the district where the radon concentration is geologically high. In Katamo district, the 2 waste rock piles and ore storage area are in the mountain which left about 1.5km away from the residential district, and an pear orchard ranges to the middle point, and the measurement points are shown in Fig.5. The integrating monitors are placed along the main valley (from R-1 to R-5, from R-14 to R-22), and in the other branch valleys as a reference points (R-23, R-24, R43). The high concentrations are observed inside from the waste rock piles to R-5 boundary point. The figure indicates that the impact is small from the outside of the site boundary to the residential district. The range of data from the same level as the residential district to 58 Bq.m^{-3} is observed from R-7 to R-13 in the site near the boundary. The concentration at R-24 as a reference point is the same level as those of near the boundary of the site. The relatively high concentration might be estimated that in these area air movements are suppressed because the tree grows and the valleys are narrow, in addition to the typical granite zone.

CONTINUOUS MEASUREMENT

The continuous measurement of radon and its progeny are carried out in Katamo district using TEL made by PYLON Co.Ltd. (Canada) and using the monitor developed by the authors[6)] respectively. The continuous progeny monitor for accurately measuring long-term concentration of low environmental radon progeny has been achieved with a large sampling flow rate with 80 liters.min^{-1} and the use of alpha energy spectroscopy in a vacuum cell using a long roll-type membrane filter and a reliable automatic system.

The simultaneous measurements have been carried out at R-5 of the site boundary and at a point between R-18 and R-19 in the pear orchard in the Katamo district. The result from April 1993 to March 1996 can be seen in Fig.6. As the figure indicates, the concentration on the site boundary which is closer to the waste rock tends to be higher. As a seasonal variation, high concentration is observed in the summer. In general it is known that radon concentrations in regions that face to the Sea of Japan are high in winter and low in summer[7)]. The data measured at the Katamo tends to be similar to this pattern of variation in those areas. As for the mean value of the equilibrium equivalent radon concentration from all the data, 8.0 Bq.m^{-3} and 4.7 Bq.m^{-3} were obtained on the boundary of the site and in the pear orchard respectively. Although the equilibrium equivalent radon concentration on the boundary tended to be high, according to the documents, the value of 4.1 Bq.m^{-3} (Nagoya Japan[8)]), and the arithmetic mean 8.6 Bq.m^{-3} and geometrical mean 6.9 Bq.m^{-3} (Germany[9)]) were reported as outdoor results. Judging from the above data, it may safely be assumed that the result in the pear orchard can at least be recognized as a normal level. Therefore, the 3.3 Bq.m^{-3} difference seems to be the long term impact from the waste rock, if the value in the pear orchard is assumed a background of this district. Next, for the purpose of the evaluation of the annual effective dose, this 3.3 Bq.m^{-3} of equilibrium equivalent radon concentration can be converted into the effective dose of 0.38 mSv.y^{-1} using the ALI from ICRP Pub.32[10)] and the respiratory volume of adult man from ICRP Pub.23[11)]. This result on the boundary is smaller than public dose limit 1 mSv.y^{-1}

SAMPLING MEASUREMENT

Radon concentration and equilibrium equivalent radon concentration by sampling method have been measured at the several integrating measurement points. As for the radon concentration the air was sampled about 1 hour using a small pump in plastic bag and it was measured by the gas charging type ionization chamber afterwards. And, as for the equilibrium equivalent radon concentration was measured using working level monitor WLM200 by TracerLabo Co.Ltd.(Germany). The measurements were carried out every month at the relatively large waste rock pile site, and quarterly or once a year at other places.

The results were summarized and shown in table 1. Though the radon concentrations tend to be high, the equilibrium equivalent radon concentrations are low, and very small equilibrium factor has been obtained. A similar place is also reported in another document[12)]. It can be estimated that in the vicinity of radon source, the progeny nuclides have not sufficiently grown and that the source is locally limited. It might be expediently possible to estimate the equilibrium equivalent radon concentration using the obtained equilibrium factor (F) by sampling method and the results of integrating measurement. These values were described as Estimated EECRn in the table. The results indicated that the equilibrium equivalent radon concentrations were also low from this table.

EVALUATION BY CALCULATION[13)]

EVALUATION CONDITION

As the radon exists naturally, the concentration changes greatly in different atmospheric conditions, and the relatively high concentration is expected because of the granite zone, it is very difficult to judge whether the impact from waste rock pile or from natural component by measurements. Therefore, for the purpose of evaluating the environmental impact from the waste rock piles only, the radon emanated from the surface of the piles were measured, and radon concentration and equilibrium equivalent radon concentration out of the site were obtained by the dispersion calculation. The effective dose was obtained according to ICRP Pub.32[10)] and

ICRP Pub.23[11]. The evaluation was carried out with representative emanation data which were average measured emanation data at each waste rock pile. The radon emanation from the mouth at Katamo-shimo 1 which has been temporarily closed by the sandbags was also measured, and it was added to the calculation. It was assumed that only the radon itself emanated from the ground surface of the waste rock piles, and the progeny nuclides were calculated according to the Betheman equations. The deposition of the progeny nuclides on the ground surface or the obstacles during the dispersion was disregarded in order to evaluate conservatively. In Japan the equilibrium equivalent radon concentration is regulated, but the exposure by the radon gas itself is not regulated. However, in this study, as the equilibrium factor was small and the exposure by radon gas can not be disregarded either, the evaluation was carried out including the effective dose by radon gas.

The model in which the basic dispersion formulation assumed the Gaussian distribution is well used in document[14] to evaluate radon effect from the uranium mill tailing piles. In this evaluation, we assumed that the emission height and the calculation points are ground surface for using the plume model, since the plume model with the Gaussian distribution is widely adopted by the many guidelines or manuals[15] as a simple model. The land form of the waste rock piles was assumed to be a plane rectangle, which has the same size as the waste rock pile area. Since the lower limit of the usable wind velocity of the plume model is empirically $1 m.s^{-1}$, in the manual as a reference, the wind velocity of $1 m.s^{-1}$ was also adopted in this calculation. As for the wind direction, it was conservatively assumed that the wind blows only in one direction throughout a year, which means that the wind direction distribution was disregarded. Therefore, the directions of the calculation points were along the short and long axes of the rectangle, and the shortest distance from the edge of the waste rock pile to the site boundary was used in the calculation. Since the purpose of the calculation is to obtain the annual mean, as for the atmospheric stability, neutral D was adopted because the frequency of D accounts for more than a half of the annual total and more unstable category than D holds 80% according to the weather observation.

Because the source of radon is a plane, it can be approximated as a cluster of point sources. The area of the waste rock pile was divided into 1m×1m meshes for the calculation, and the numerical integration was carried out by assuming that the radon gas was emitted from the each center.

CALCULATION RESULT AND DISCUSSION

For each site, the results of radon concentration, equilibrium equivalent radon concentration, and effective dose calculated by the plume model (atmospheric stability=D, wind velocity=$1.0 m.s^{-1}$) are shown in table 2. In this table, the results from the multiple waste rock piles within the same site are simply summed, and it was confirmed that the effective doses do not exceed 1 $mSv.y^{-1}$ at each site boundary. In addition, the similar results were obtained even at a wind velocity of $0.5 m.s^{-1}$, assuming that the dispersion is weak at night and dawn, using not the shortest distance but realistic one from the wind direction estimated by the measured radon concentrations.

The calculation results were compared with the radon concentration and equilibrium equivalent radon concentration. As for the radon concentration, calculated value and measured value agree well, considering that the measurement points are near the boundary within the site, the radon concentration of the background is about 20 $Bq.m^{-3}$, and the ununiformity of emanation brings about localized radon concentration. However, as for the equilibrium equivalent radon concentration between calculated and measured value, poor agreement is observed with one order difference, because the measured values are not determined by long term measurements but by grab samplings. The calculated results seems to be too small to discriminate from the background level in the normal environment reported in other reports[8) 9)].

DISPERSION EXPERIMENT

The dispersion experiments were conducted three times by using SF_6 tracer gas in 1992 and 1994 in order to obtain the radon influence from the waste rock piles to the residential area at Katamo as a demonstration field shown in Fig.5. In case of such land form, it is known that the mountain breeze (nocturnal drainage flow) arises at night. In order to know how much of radon is flowing to the residential area, the tracer gas was emitted from the pile while the mountain breeze was observed at night, and the air samples from the site to the residential area were collected by automatic samplers and analyzed by gas chromatography. In addition, the radon concentrations in the collected samples were also measured by ionization chambers, and the effect from the waste rock piles was examined with the radon emanation.

EXPERIMENTAL RESULT AND DISCUSSION

It is observed that the result of the dispersion experiments by the continuous emission has a good agreement with the estimated concentration on the down wind axis by the plume model (atmospheric stability=D, wind velocity=0.5, 1 $m.s^{-1}$), as shown in Fig.7. Although several experimental concentrations which exceeded the model results were observed, those were only temporal phenomena which occur when the mountain breeze was broken up in the morning. And the transport speed of tracer gas toward residential area was estimated to be about 0.5 $m.s^{-1}$ by the puff emission experiment. It can be said that the dispersion experiments were carried out conservatively considering that the mountain breeze is not always observed even at night. Therefore, the weather conditions of the model, such as atmospheric stability D, 1 $m.s^{-1}$ wind velocity, and one wind direction, seem to be valid for the purpose of annual evaluation. And, it seems to be also adequate to assume atmospheric stability D and 0.5 $m.s^{-1}$ wind velocity from transport speed by the puff emission experiment as the calm condition at night. On the basis of three time experiments, the maximum effect of radon concentration on the site boundary was estimated to be about 20 $Bq.m^{-3}$ from the radon samples collected out of the site at dawn as shown in Fig.8, assuming that the behavior of the radon is the same as the tracer gas.

CONCLUSION

The environmental impact of radon and its progeny from the waste rock piles have been investigated. Although the relatively high concentrations are observed inside of the site, the results outside of the site indicate that the impacts are small compared with the natural background level. And it was confirmed that the effective doses from the waste rock piles are less than 1 $mSv.y^{-1}$ by the evaluation based on both measurement and calculation out of the site boundary.

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Table 1 Summary of radon and equilibrium equivalent radon concentration by sampling method

Site	Radon [Bq/m^3]				EECRn [Bq/m^3]				F-value [-]				Estimated EECRn
	Ave.	Max.	Min.	N	Ave.	Max.	Min.	N	Ave.	Max.	Min.	N	
Asabatake-1WR	119.4	754	15	13	4.3	10.4	1.3	16	0.095	0.29	0.006	12	3.6
Asabatake-1WR(B)	96.2	184	18	10	3.7	11.9	0.9	10	0.051	0.11	0.008	10	2.2
Asabatake-2WR	85.0	330	8	37	8.1	12.6	3.9	45	0.085	0.65	0.012	37	5.5
Asabatake-2WR(B)	77.4	371	7	34	3.7	22.9	0.5	44	0.087	0.39	0.013	34	9.5
Asabatake-2BWR	64.8	155	13	8	3.6	10.4	1.0	9	0.093	0.23	0.013	8	5.4
Asabatake-3WR	146.4	280	68	9	3.3	8.0	0.7	12	0.032	0.088	0.016	9	2.1
Katamo-1,2WR(s1)	4786.7	66300	14	48	77.4	339	0.6	49	0.067	0.29	0.001	47	24.5
Katamo-1,2WR	20.0	57	6	24	3.6	7.9	0.7	25	0.27	0.65	0.037	24	6.4
Katamo-1,2WR(R-5)	28.7	130	6	33	3.2	10.8	0.9	37	0.15	0.63	0.032	32	6.8
Katamo-1,2WR(B4)	21.3	56	9	8	2.9	6.5	1.1	9	0.17	0.33	0.039	8	1.9
Katamo-S2WR	54.7	176	9	7	3.7	9.7	0.9	8	0.085	0.17	0.038	7	3.0
Katamo-3WR	37.3	67	14	6	2.0	2.9	1.3	7	0.071	0.14	0.021	6	1.2
Ayumidani-WR	22.9	48	6	11	3.4	5.9	2.0	12	0.18	0.35	0.071	11	9.1
Kan'nokura-1WR	44.3	107	6	9	2.2	4.6	0.6	9	0.092	0.35	0.022	9	2.4
Kan'nokura-1WR(B)	83.1	135	37	9	3.6	5.5	0.8	9	0.043	0.073	0.022	9	2.9
Kan'nokura-2WR	437.7	1210	15	9	7.7	26.1	0.9	9	0.047	0.25	0.008	9	6.0
Kan'nokura-2WR(B)	59.2	297	6	6	3.5	12.0	1.1	8	0.22	0.37	0.040	6	3.8
Nakatsugo-WR	11.7	21	5	10	2.4	5.8	0.2	32	0.29	0.67	0.049	14	4.1
Nakatsugo-WR(B)	10.8	20	5	9	2.9	5.4	1.5	12	0.35	0.78	0.15	10	5.0
Choja-WR	23.0	91	5	10	1.8	3.4	0.6	15	0.19	0.49	0.012	10	2.8
Choja-WR(B)	41.7	127	5	6	2.0	3.1	0.6	8	0.18	0.62	0.018	6	7.9
Sugegawara	14.7	42	5	37	3.7	12.0	0.4	47	0.27	0.67	0.031	37	15.8
Controls(others)	22.9	191	5	26	2.8	8.4	0.2	60	0.36	0.70	0.031	28	5.0

Note1: "EECRn" means equilibrium equivalent radon concentration
Note2: "N" means the number of data.
Note3: "Estimated EECRn" were obtained from the integrated radon concentration in fiscal 1996.

Table 2 The results of radon concentration, equilibrium equivalent radon concentration and effective dose calculated by the plume model (atmospheric stability =D, wind velocity =1.0m/s)

waste rock	conditions of the estimation				results of the estimation			
	size of waste rock area	waste rock area	down wind distance	radon emanation	radon concentration	equilibrium equivalent radon concentration	effective dose	total of the site
Nakatsugo	120 m × 100 m	12,000 m^2	2 m	0.22 $Bq/m^2/s$	12.7 Bq/m^3	0.157 Bq/m^3	0.033 mSv/y	0.033 mSv/y
Choja	12 m × 26 m	312 m^2	14 m	0.78 $Bq/m^2/s$	11.4 Bq/m^3	0.111 Bq/m^3	0.026 mSv/y	0.026 mSv/y
Kan'nokura-1	39 m × 130 m	5,070 m^2	2 m	0.47 $Bq/m^2/s$	27.7 Bq/m^3	0.363 Bq/m^3	0.075 mSv/y	0.075 mSv/y
Kan'nokura-2	72 m × 89 m	6,408 m^2	2 m	1.10 $Bq/m^2/s$	57.9 Bq/m^3	0.567 Bq/m^3	0.134 mSv/y	0.134 mSv/y
Asabatake-1-1	33 m × 43 m	1,419 m^2	2 m	0.59 $Bq/m^2/s$	24.3 Bq/m^3	0.137 Bq/m^3	0.044 mSv/y	0.086 mSv/y
Asabatake-1B	27 m × 34 m	918 m^2	4 m	0.76 $Bq/m^2/s$	21.9 Bq/m^3	0.137 Bq/m^3	0.042 mSv/y	
Asabatake-2	25 m × 80 m	2,000 m^2	5 m	0.77 $Bq/m^2/s$	30.0 Bq/m^3	0.352 Bq/m^3	0.076 mSv/y	0.076 mSv/y
Asabatake-2B	16 m × 25 m	400 m^2	4 m	0.30 $Bq/m^2/s$	7.5 Bq/m^3	0.039 Bq/m^3	0.013 mSv/y	0.013 mSv/y
Asabatake-3	32 m × 50 m	1,600 m^2	4 m	0.50 $Bq/m^2/s$	17.1 Bq/m^3	0.138 Bq/m^3	0.036 mSv/y	0.036 mSv/y
Katamo-1	22 m × 91 m	2,002 m^2	5 m	0.71 $Bq/m^2/s$	28.9 Bq/m^3	0.366 Bq/m^3	0.076 mSv/y	0.294 mSv/y
Katamo-2	29 m × 59 m	1,711 m^2	2 m	0.71 $Bq/m^2/s$	32.7 Bq/m^3	0.233 Bq/m^3	0.065 mSv/y	
Ore storage area	16 m × 24 m	384 m^2	32 m	0.04 $Bq/m^2/s$	0.3 Bq/m^3	0.006 Bq/m^3	0.001 mSv/y	
Storage-area	11 m × 26 m	286 m^2	45 m	0.02 $Bq/m^2/s$	0.1 Bq/m^3	0.002 Bq/m^3	0.000 mSv/y	
Shimo-1 mouth	point	-	30 m	824 Bq/s	59.9 Bq/m^3	0.704 Bq/m^3	0.151 mSv/y	
Katamo-3	21 m × 24 m	504 m^2	4 m	0.21 $Bq/m^2/s$	5.1 Bq/m^3	0.026 Bq/m^3	0.009 mSv/y	0.009 mSv/y
Katamo-shimo2	20 m × 65 m	1,300 m^2	4 m	0.32 $Bq/m^2/s$	12.2 Bq/m^3	0.116 Bq/m^3	0.028 mSv/y	0.028 mSv/y

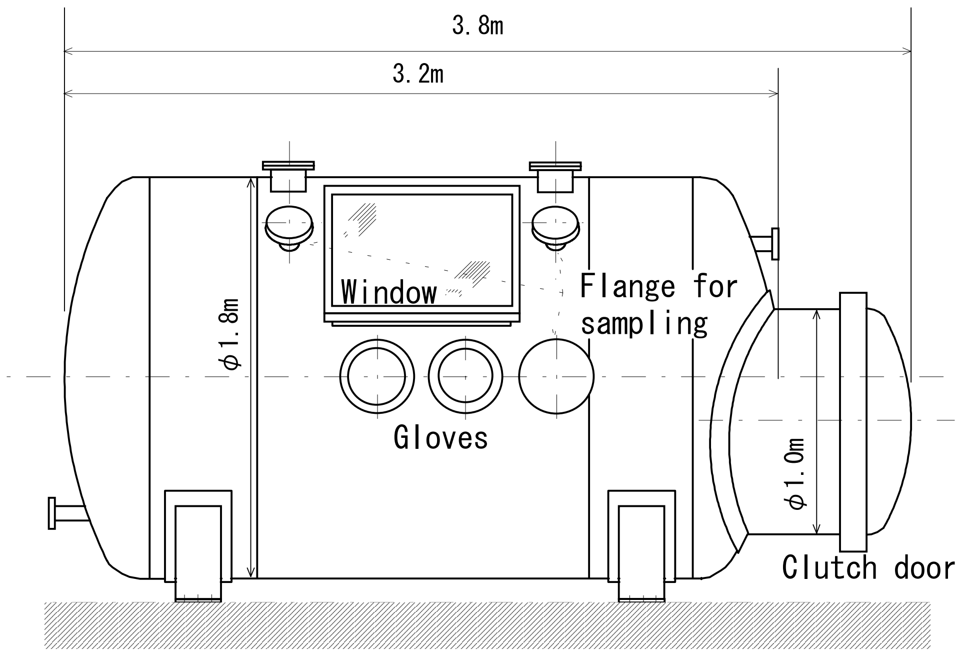

Fig.1 Radon Chamber

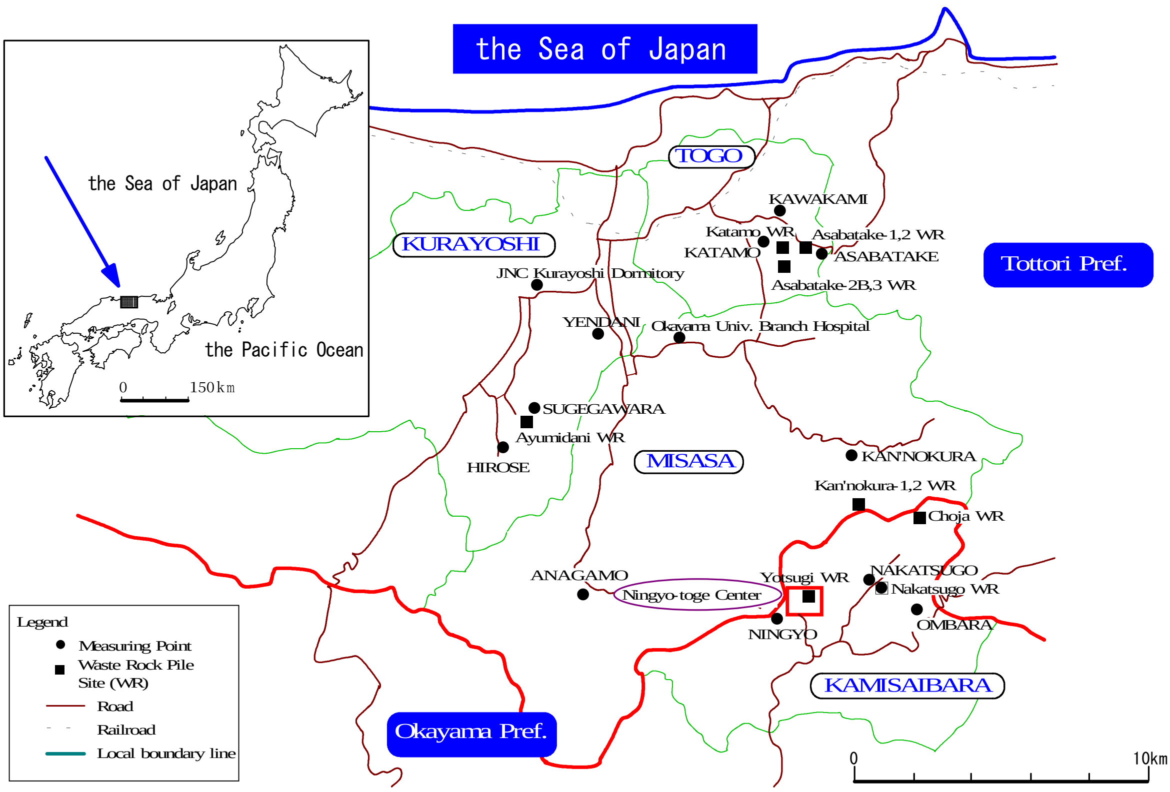

Fig.2 Location of the waste rock pile sites and the measuring points

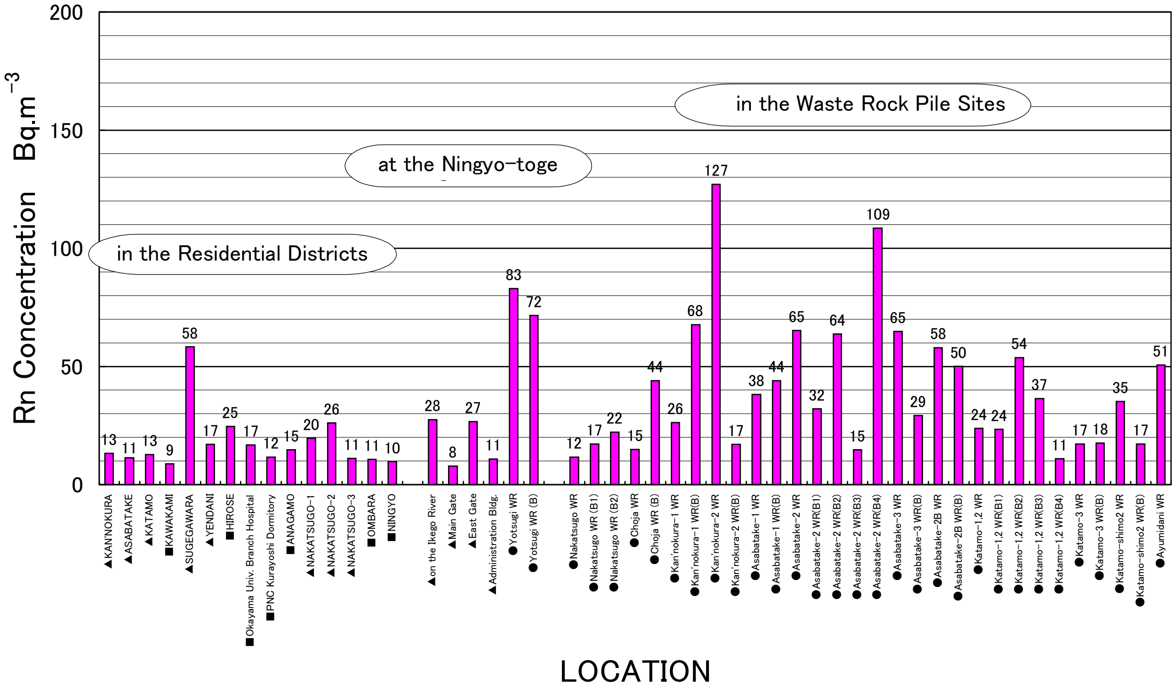

Fig.3 Radon concentration in and around the waste rock pile sites in fiscal 1996
Note: "WR" and "(B)" means the measuring point in the waste rock pile site and near the site boundary, respectively.

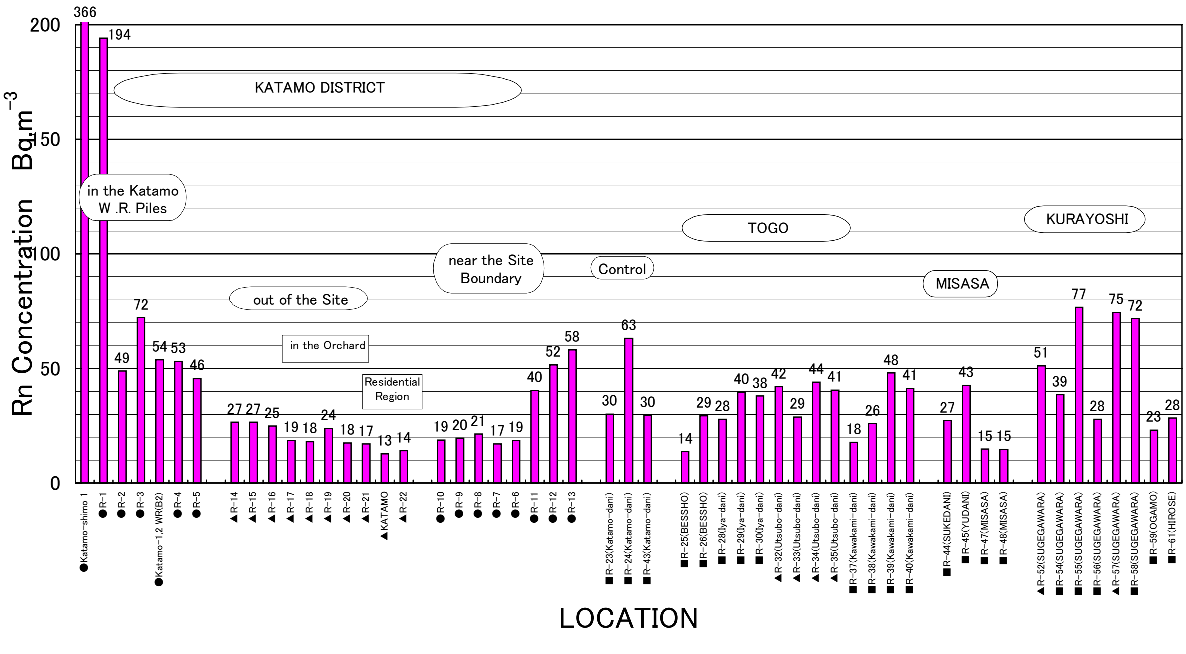

Fig.4 Radon concentration obtained by the detailed measurements in fiscal 1996.
Note: In Fig.3 and Fig.4, ●, ▲ and ■ means the measuring point in the waste rock pile site, around the site and in the control area, respectively.

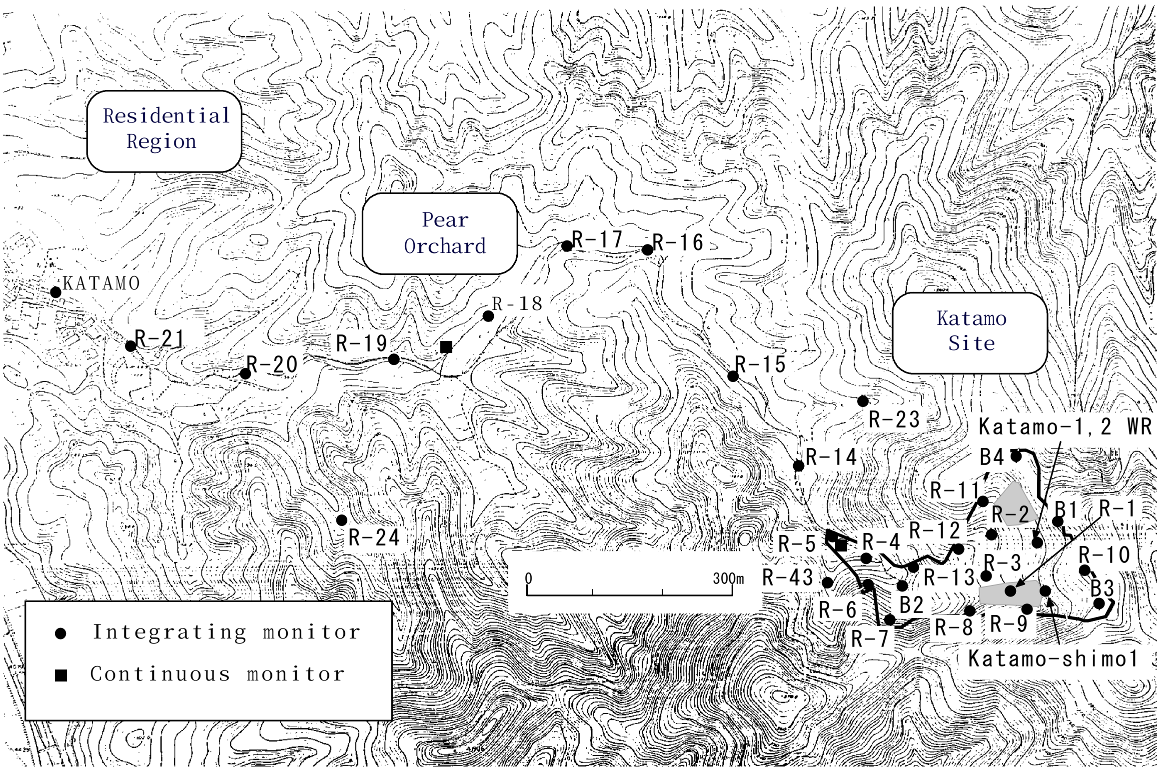

Fig.5 Measuring points in the Katamo district

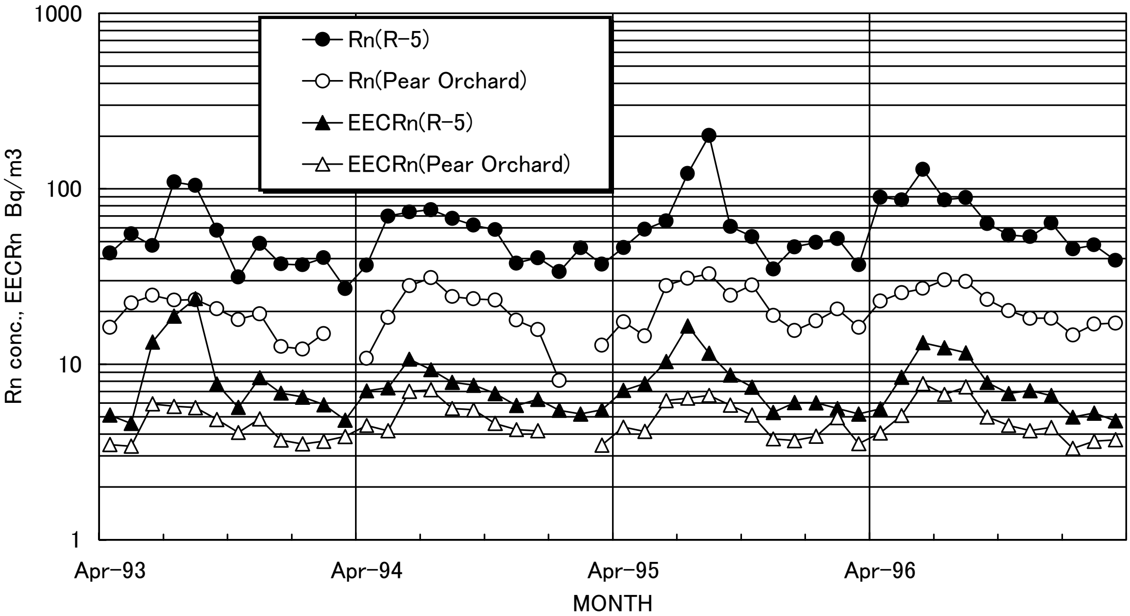

Fig.6 Radon and its progeny concentration obtained by continuous monitors

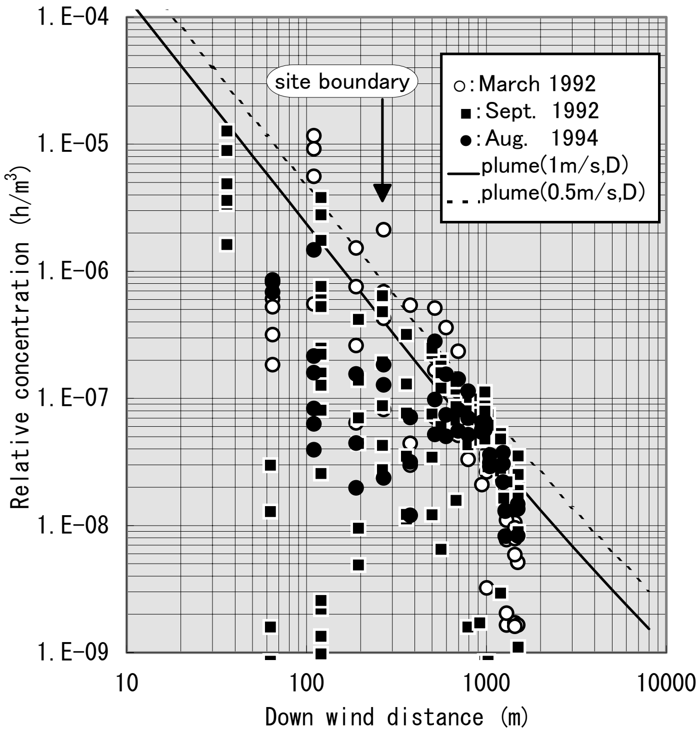

Fig.7 Results of dispersion experiments

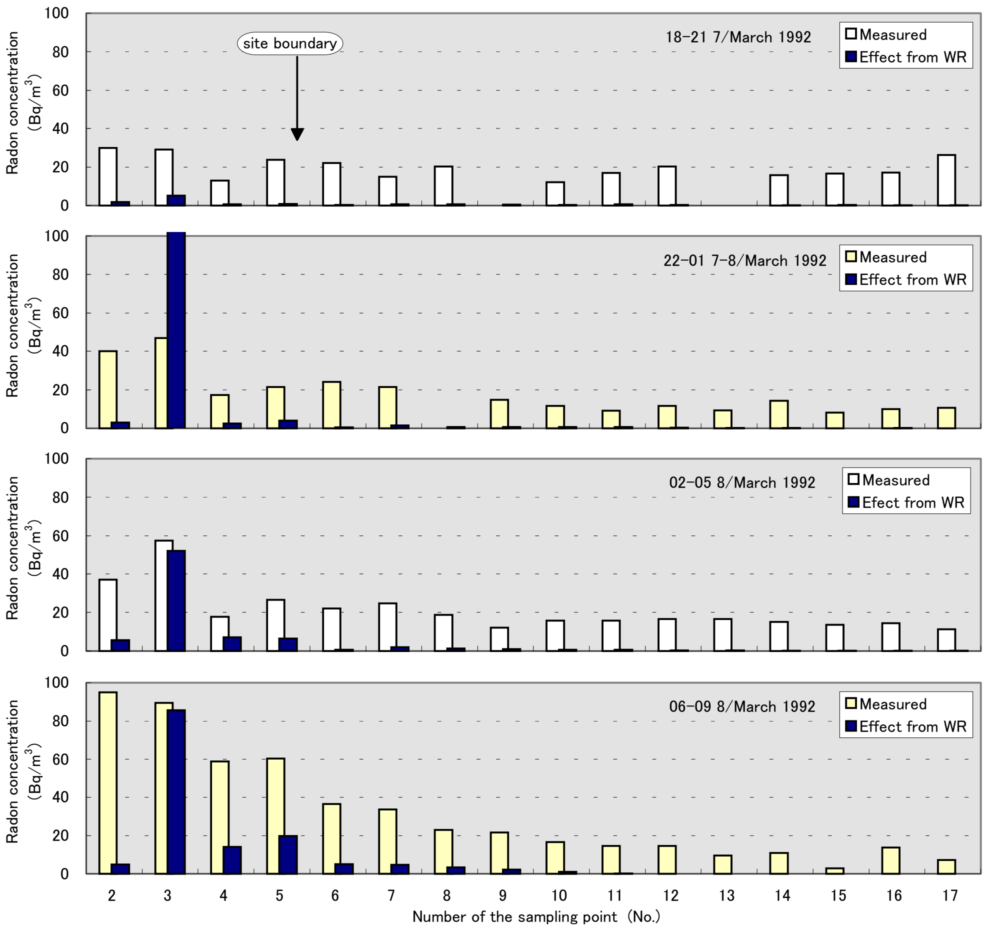

Fig.8 Measured radon concentration and estimated effect from waste rock piles by the dispersion experiments