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DETERMINATION OF RADON IN SOIL GAS BY AN OPENED COUNTING VIAL AND LIQUID SCINTILLATION COUNTER

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An opened counting vial containing liquid scintillator solution, suspended in a hole of the ground, absorbs a definite fraction of radon present in the soil gas allowing the total radon concentration to be determined. Natural level of radon in soil gas was measured. This method is rapid, simple and sensitive.

Radon has a large partition coefficient between organic solvents, such as toluene, and air or water. Based upon this principle, the authors have previously developed a method for the determination of ^{222}Rn[1)2)] (hereafter radon) or ^{222}Rn[3)4)5)] in natural waters using toluene extraction followed by the integral counting with a liquid scintillation counter[6)2)]. The authors have found that when an opened counting vial containing 0.02 dm^3 of liquid scintillator solution(4g PPO and 0.1g POPOP in 1 dm^3 toluene) is exposed to the atmosphere (hereafter open vial), it absorbs radon in the air effectively. Applying this fact the authors have extended a new determination method of radon in soil gas. This report describes some basic investigations on this method.

A certain amount of ^{226}Ra standardized solution was placed on the bottom of a glass cylinder which was firmly sealed using a polyethylene tube having the same diameter as the mouth of the cylinder (Fig. 1). This tube was hermetically sealed with putty. This closed system was allowed tc stand for a certain period to accumulate radon from radium. An open vial was placed in a holder and suspended in the cylinder. During introduction and removal of vials, a couple of seals on the tube was used to minimize the loss of radon: Seal A was untied and the sample holder was introduced, while seal B remained tightly closed. Seal A was then tied and seal B untied. The holder was then lowered into the cylinder and suspended about 0.1 m from the bottom. It was exposed for a certain time, after which the vial was removed in the reverse procedure described for its introduction. The open vial was then closed with a screw cap. At least 3.2 hrs are needed to establish radioactive equilibrium between radon and its daughters[2)], before the integral counting started.

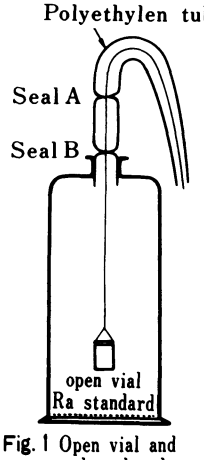


Fig. 1 Open vial and radon closed system

When an open vial is exposed to the atmosphere, radon exclusively dissolves into LS solution. This was confirmed by exposing fourteen different vials for the same exposure time of 1 hour at different times over a period of 5 days. The observed activities of radon are shown in Fig. 2(open circle) where a build-up curve was obtained. The expected activity of radon at the corresponding elapsed times were calculated(triangle) assuming perfect release of radon from the radium sources. A typical build-up curve of radon[7] was obtained.

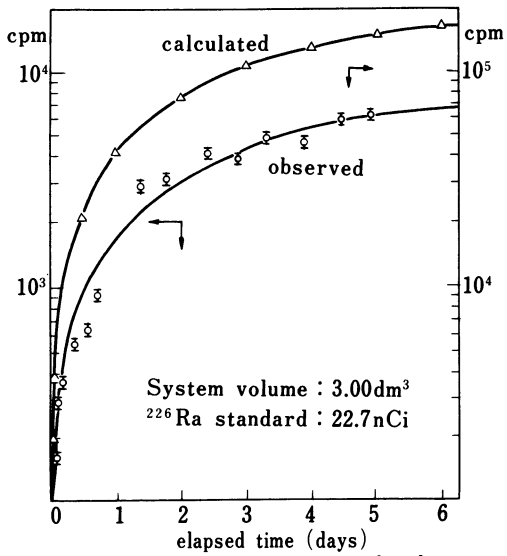


Fig. 2 Build-up of Rn from Ra in a closed system.

The shape of these two curves are quite similar. When the two curves are superimposed, they coincide with each other within experimental error. The discrepancy between the two is considered to arise from the assumption of perfect release of radon or from radon sticking to the walls of the apparatus. These results show that a definite fraction of radon is actually dissolved into LS solution. In other words, the proposed open vial method can dissolve a definite fraction of radon present in the atmosphere.

The effect of exposure time on dissolution of radon into LS solution was examined by the following experiment. A new closed system was prepared for each exposure time. Between each determination, the system was carefully washed with 2N nitric acid, then dried and the air inside changed completely. The observed activities are shown in column I of Table 1. The radon activities, actually dissolved in the LS solution during exposure, are obtained(column II) by applying the factor of 11.1 cpm corresponding to 1 pCi radon. This high sensitivity is an advantage given by the integral counting[2)6)]. The undissolved part of radon (column III) was calculated from the measured counting rate by applying the partition coefficient, after corrections for elapsed time from the removal of vial to the start of counting and for evaporation of LS solution[2)]. The expected activity of radon, assuming perfect release of radon from radium as a function of exposure time, is shown in column IV.

Table 1 The effect of exposure time on dissolution of Rn (25°C)

Exposure time days	I Observed activity cpm	II Rn dissolved into LS pCi	III Undissolved part of Rn pCi	IV Rn built-up from Ra* pCi	V Ratio II+III/IV %
0.21	31.1±1.1	2.8±0.1	27.0±1.0	116	25.7
0.29	43.4±1.2	3.9±0.1	37.7±1.0	159	26.1
0.42	63.1±1.2	5.7±0.1	54.8±1.0	228	26.5
0.56	71.4±1.3	6.4±0.1	62.0±1.0	300	22.8
0.58	77.4±1.3	7.0±0.1	67.2±1.0	310	23.9
0.58	76.9±1.3	6.9±0.1	66.8±1.0	310	23.8
0.71	99.6±1.4	9.0±0.1	86.5±1.0	375	25.4
System volume: 2.05 dm³		* assumed 100% release		average	24.9±1.3
²²⁶Ra activity:3.11 nCi				coeff. of variance	5.1%

The activity of radon dissolved increases proportionally with the duration of exposure as shown in Table 1, but will reach a definite value after the establish-

ment of the radioactive equilibrium between radium and radon. The ratio of total measured activity(II + III) to the expected radon activity(IV) is shown in column V. The average value of the ratio is 24.9 ± 1.3% with a C.V. of 5.1%. These figures have no practical meaning because perfect release of radon, which does not actually occur[8)9)], is assumed. However this constant ratio means that a definite fraction of radon present in the atmosphere dissolves into LS solution and the exposure time has no effect on dissolution.

The reproducibility of this method was tested as follows. Three open vials were simultaneously exposed for 44.4 hrs in a closed system, followed by integral counting. A satisfactory results were obtained as shown in Table 2(a).

Table 2 The reproducibility of the open vial method

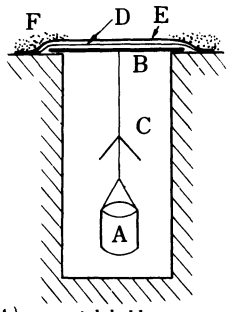
a) ^{226}Ra activity: 20.8 nCi (25°C)

Exposure time hours	Observed activity cpm	Rn dissolved into LS pCi
	1,033±8	93.1±0.7
44.4	1,048±8	94.4±0.7
	1,044±8	94.1±0.7
System volume: 3.80 dm³	average	93.9±0.6
	C.V.	0.6%

b) soil of uranium mine (200g, 25°C)

Exposure time hours	Observed activity cpm	Rn dissolved into LS pCi
21.6	26.8±1.0	2.4±0.1
21.6	23.4±1.0	2.1±0.1
21.6	28.3±1.1	2.6±0.1
System volume: 2.65 dm³	average	2.35±0.18
	C.V.	7.7%

Three open vials were separately placed in a closed system containing some soil from a uranium mine. Three vials were removed separately, after the same exposure time of 21.6 hrs (Table 2(b)). The average amount of radon determined was 2.35 ± 0.18 pCi with a C.V. of 7.7%. Though the experiment with the soil from uranium mine contains large errors, the reproducibility obtained with this method shows to be satisfactory for determination of radon in soil gas as the field experiment.



A) open vial holder
B) wood stick C) filter paper
D) polyethylene sheet
E) paper dish F) soil
Fig. 3 Open vial in a test hole

This method was applied to measure the natural level of radon in soil gas. A test hole was made on the ground surface as shown in Fig. 3. An exposure time of about five days was applied. The observed activities of six different trials are shown in column II of Table 3, together with the curie unit(column III). The undissolved part of radon(IV) was calculated as described above. The sum of

Table 3 Natural background of Rn exhaled into the hole of university ground

Date (1977)	I Exposure time days	II Observed activity cpm	III Rn dissolved into LS pCi	IV Undissolved part of Rn pCi	Total Rn present in the hole pCi	Rn concentration in soil gas pCi dm^{-3}
Oct.14-20	5.0	23.3±1.1	2.1±0.1	21.9±1.0	24.0	8.7
Oct.20-25	5.0	55.8±1.2	5.0±0.1	52.1±1.0	57.1	20.8
Oct.25-30	4.8	70.8±1.3	6.4±0.1	66.7±1.0	73.1	26.6
Oct.30-Nov.4	5.3	56.7±1.2	5.1±0.1	53.2±1.0	58.3	21.2
Nov.4 - 9	4.9	57.8±1.2	5.2±0.1	54.2±1.0	59.4	21.6
Nov.20-25	5.0	72.6±1.3	6.5±0.1	67.8±1.0	74.3	23.4

test hole: ø 1.0 x 3.5 dm (2.75 dm³)

(III + IV) represents the total activity of radon actually present in the atmos-

phere of the hole, which was found to vary between 8.7 to 26.6 pCi dm^{-3} during this period. These results are considered to be reasonable compared with those of other reports[10)11)]. It deserves special emphasis that the open vial method does not need any device for collection of radon in soil gas, but requires only an open vial suspended in a test hole for a certain time.

Finally the effect of naturally occuring ^{220}Rn and ^{218}Rh on this method must be mentioned, since they are simultaneously dissolved and counted. The effect of ^{218}Rn(half life 3.92 sec) is negligible due to its low natural abundance and short half lives of its daughter products[3)]. Our experience of field application of this method shows that as the counting was performed after at least 100 hrs from removal of the vial, ^{220}Rn has no effect on the counting rate, because of the rapid decay of its longest lived daughter(^{212}Pb half life 10.64 h). If determination of ^{220}Rn was required, the vial was counted several times within 80 hrs after its removal. The amount of ^{220}Rn can be determined by resolving the composite decay curve obtained[12)].

The proposed open vial method may be summarized as follows. Although the open vial does not absorb all the radon present in the atmosphere, it always dissolves a definite fraction of the total amount of radon according to its partition coefficient. The exposure time appears to have no effect on the dissolution of radon. The undissolved part of radon is calculated from the dissolved part by applying the necessary factor and corrections. The total amount of radon can be easily obtained from the sum of the dissolved and undissolved parts.

This method gives good reproducibility for measuring radon in soil gas, and is very sensitive because it can measure the radon inclusive of its five daughter products by applying the integral counting with a liquid scintillation counter. Additionally it is very simple because radon in the air can be determined without the need of sampling. These characteristics are advantageous and promising for field works such as prospecting uranium mine or detecting faults and fissures in geological survey[13)].

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