SOME PROPERTIES OF IONIZING RADIATIONS AND RADIOACTIVE IONS IN THE NATURAL ENVIRONMENT

MINORU KAWANO, YUKIMASA IKEBE, YOSHIYUKI NAKASHIMA, TAKEYOSHI NAKAYAMA, MICHIKUNI SHIMO, SUSUMU MINATO, and TAKAO IIDA

Department of Nuclear Engineering

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Abstract

This paper consists of two parts. Chapter 1 is concerned with the results of measurements of ionization intensities due to the beta- and gamma-radiations near the ground carried out from 1963 through 1969. The influences of fallout radioactivity deposited on the ground surface were studied from the view point of atmospheric ionization. The beta-radiation from the fallout radioactive nuclides deposited on the ground surface decreased remarkably during the last 6 years, and recently, a newly designed ionization chamber made of platic plates was used for meauring the beta-radiation. The ionization intensity due to the gamma-radiation didn't change so remarkable as that due to the beta-radiation during the same period. Recently, spectroscopic method with NaI scintllation counter was adopted for obtaining some detailed informations on the terrestrial gamma-radiation. In the beginning of January 1967, sudden increases of the ionization intensities due to the beta- and gamma-radiations from the fallout radioactivity caused by a nuclear explosion test were measured.

Chapter 2 is concerned with the physical properties of RaA ion. To clarify the interaction between RaA ions and condensation nuclei, simultaneous measurements of the concentrations of RaA ions (n_A), radon-222, condensation nuclei (Z) and the diffusion coefficient of condensation nuclei (D) were carried out. It was found that the correlation among them may be well expressed by the simple formula: $q_A=\beta_A n_A Z$. The correlation between β_A and D (or rdius of nuclei) was also obtained. The effective attachment coefficient of RaA ions was compared with that of small ordinary ions, and it was presumed that about 25 per cents of RaA atoms formed on the decay of radon-222 seem to have some charge, and remaining 75 per cents have no charge.

Chapter 1. The ionization intensity due to the beta- and gamma-radiations

1. 1. Introduction

Although the fallout radioactivity caused by nuclear explosion tests was well studied from the view points of radiology, environmental sciences and technology, biology, and medical sciences, almost all of them were mainly concerned with gross counting, nuclide analysis, local anomaly, and their effects on human body, animals, and plants, and the influences of fallout radioactivity on the ionization

intensity near the ground in the natural radiation environment have not been studied so well.

In the previous paper[1.1], the present authors (M. K., Y. I., Y. N.) published the results of measurements on the ionization intensities due to the alpha-, beta-, and gamma-radiations in the atmosphere near the ground in 1963 and 1964. The time variations for several years and the influences of the fallout radioactivity on the ionization intensity due to the naturally occuring radiations were not clarified in the paper. In the present paper, the results of measurements of the ionizations due to the beta- and gamma-radiations from the fallout radioactive nuclides deposited on the ground surface during the last 6 years were described, and the abnormal increases of the beta- and gamma-radiations from the fallout radioactivity were discussed.

As is described below, the rate of ion pair production due to the beta-radiation was so small as considerably hard to measure it with the ionization chamber described in the previous paper. A newly designed ionization chamber used for measuring ionization intensity in low level due to the beia-radiation from the ground surface will be shown. The spectroscopic method with a scintillation counter (NaI $3''\varphi \times 3''$) was used for studying the detailed characteristics of the terrestrial gamma-radiations near the ground, and the response matrix method was used for calculating the real energy spectrum from the pulse height distribution detected with the scintillation counter.

1. 2. Instruments used for measurements

The ionizations due to the beta- and gamma-radiations were described in the previous paper[1.1]. The ionization due to the beta-radiation decreased, and the value of the rate of ion pair prodution is about 1 J recently. Therefore, an improved equipment is necessary to measure precisely the ionization intensity due to the beta-radiation from the ground surface.

A newly designed ionization chamber used for measuring the ionization intensity due to the beta-radiation from the ground surface was designed to minimize the wall effect and other secondary effect of the beta-radiation. The chamber consists of two ionization chambers. One of the two chambers is used for measuring the beta- and gamma-radiations, and another one for the gamma-radiation only. The structures and the dimensions of every parts of two chambers are quite the same each other except materials and thickness of their windows.

Figure 1.1 shows the structure of the ionization chamber used for measuring the beta- and gamma-radiations. The chambers are constructed by acrylite (one kind of polystyene) plates of 20 mm thick with dimensions 440 mm × 440 mm × 225 mm, having a volume of 43.5 litres. All walls except the lower surface are 20 mm thick (2,200 mg/cm^2), and their inner surfaces are coated with carbon acqudac to get static conductivity. The lower surface (window area of 1,936 cm^2) is made of Metalumy sheet (3.4 mg/cm^2). Metalumy is a commercial name of vinyl sheet coated with aluminium. The inner electrode consists of an acrylite plate with dimensions 430 mm × 430 mm, and both surfaces of the plate are perfectly covered with Metalumy sheet.

This electrode is fixed to the wall of the top with four pieces of insulators (polystyrol). Each piece is divided into two parts, and a copper plate of which area is almost the same with that of the electrode is supported by these pieces,

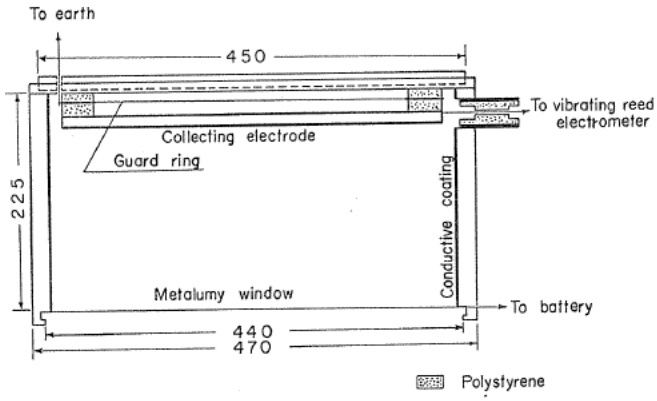

FIG. 1.1. Structure of the ionization chamber used for measuring the ionization intensity due to beta- and gamma-radiation.

and is grounded. Therefore, the copper plate plays a role of guard ring in this chamber. The collecting electrode protruded through an polystyrene plug, and was connected to a vibrating reed electrometer and an automatic recording microammeter. The window of the gamma-chamber is made of an acrylite plate of 10 mm thick which is perfectly covered with Metalumy sheet.

The acrylite plate of 10 mm thick is sufficient to absorb all beta-particles radiated from natural radioactive nuclides on the ground surface. The residual radioactivity, specially alpha-particle, contained carbon acquadac and acrylite is negligibly small as compared with those of any kinds of metals. Furthermore, the scattering of beta-particles by acrylite is very small as compared with that by metals because of its low atomic number.

Figure 1.2 shows the characteristic curves of two chambers; *i.e.*, solid line shows the characteristic curve of the beta- and gamm-chamber, and the dotted line shows that of the gamma-chamber. As is shown in these curves, 45 volts was found to be sufficient for the saturation current at the ionization level of back-ground. The saturated value in the ionization current of beta- and gamma-chamber was larger than that of gamma-chamber as shown in the figure.

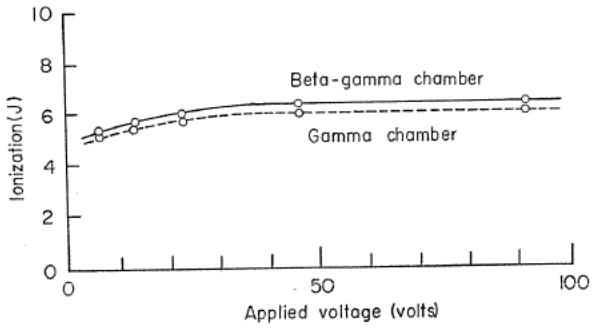

FIG. 1.2. Characteristic curves of ionization chambers used for beta- and gamma-radiations, and gamma-radiation, respectively.

The ionization intensity q (measured in the unit of ion pair/cm^3/sec, J) is usually given by the relation

$$qeV_e = i \tag{1}$$

where, all quantities are measured in the proper units, and e is the electronic charge 1.6×10^{-19} coul., i is the current measured with a vibrating reed electrometer, and V_e is the effective ionizing volume of the chamber. The effective ionizing volume of the chamber is obtained by the experimental and theoretical methods, and described in Reference (1. 2). The results of this study show that the effective ionizing volume of the chamber amounts to 0.60 V for the beta-radiation from the ground surface (in the case of 100 cm high above the ground), and amounts to 0.48 V for the beta-radiation from the atmosphere. V is the volume of the chamber, and is 3.11×10^4 cm^3.

Substituting the effective volume into the relation (1), the ionization intensity measured with the chamber may be given as follows;

$$q = \frac{i}{1.6 \times 10^{-19} \times 0.6 \times 3.11 \times 10^4} = \frac{i}{2.99 \times 10^{-15}} \tag{2}$$

where, q is expressed in the unit of ion pair/cm^3/sec. The rate of ion pair production shown in Fig. 2 were calculated by using the relation (2).

The difference in the saturated ionization current between the two chambers shown in Fig. 1. 2 gives the value of the ionization intensity to be 0.8 J. The difference seems to be caused by the beta-radiations from the floor inside a concrete building.

Figure 1. 3 shows the arrangement of the two chambers to measure the ionization intensity due to the beta-radiation from the ground surface. The collecting electrodes of two chambers are joined with a brass rod which was provided with "T" fitting to allow connection with the pre-amplifier of a vibrating reed electrometer. The interconnecting "T" arrangement was enclosed in grounded metal shield making contact with both guard rings. The electrometer was operated in conjunction with an TOA-DEMPA recording microammeter. As described above, 45 volts was found to be sufficient for saturation at these levels of ionization.

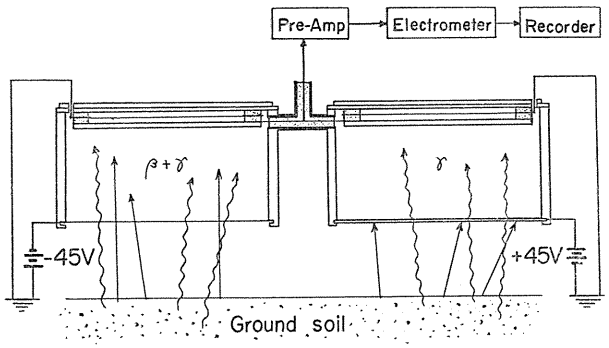

FIG. 1. 3. Arrangement of the two chambers for measuring the ionization intensity due to beta-radiation from the ground.

Opposite polarity was applied to each chamber, so that the measured current was proportional to the difference in ioization current between the two chambers. To ascertain that the difference in the ionization current between the two chambers caused by the irradiation of the gamma-radiation is negligibly small, the joint chambers were irradiated by the gamma-radiation from Ra-226 of 5 mCi. The difference in ionization current between the two chambers was found to be always less than 1.5 per cents of the ionization current in each chamber. This result seems to show that the joint chambers can be used for measuring the difference in ionization current between the two chambers. Therefore, the current measured with the joint chambers is proprtional to the ionization intensity due to the beta-radiation penetrated into the chamber.

1.3. Measurements and Results

i) Results of measurements for 6 years

The measurements of the ionization intensities due to the beta-, and gamma-radiations from the ground surface were continuously carried out inside and outside of the campus of Nagoya University from 1963 to 1969. The amplitudes of diurnal variation curves of both ionization intensities were small. As shown in

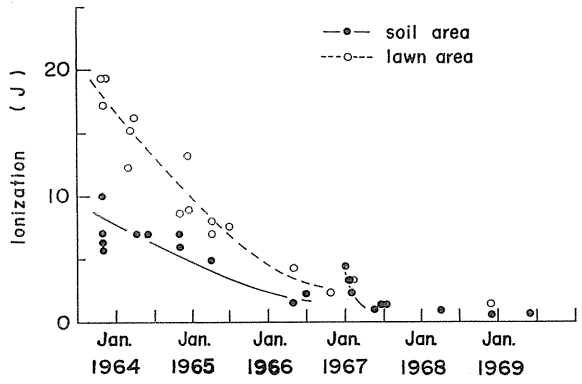

FIG. 1.4. The variation of the ionization intensity due to the beta-radiation.

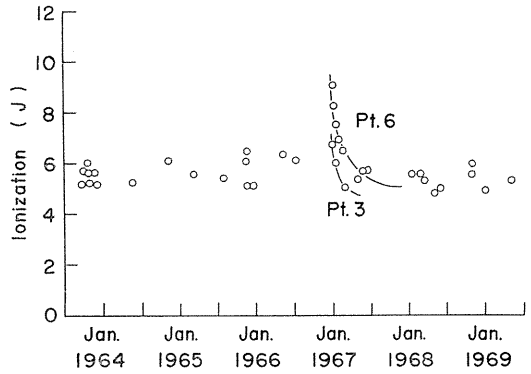

FIG. 1.5. The variation of the ionization intensity due to the gamma-radiation.

TABLE 1.1. The results of measurements of the ionization intensities after 1964

Places		Period	Surface Conditions	Beta-radiation (J)	Gamma-radiation (including Cosmic Ray) (J)	Gamma-radiation (including Cosmic Ray) (μR/hr)
Kyoto Univ.		April. 1965	soil	5.2	7.0	12.1
			moss	6.8	7.0	12.1
			lawn	7.6	7.3	12.6
Nagoya Univ. Pt	6	June, 1964	soil	7.2	5.1	8.8
	〃	Nov., 1964	〃	7.0	6.0	10.3
	2	Dec., 1964	moss	13	—	—
	〃	〃	grass	8.6	—	—
	6	April, 1965	soil	—	5.5	9.5
	〃	Aug., 1965	〃	—	5.4	9.3
	〃	Dec., 1955	〃	—	6.0	10.3
	5	〃	〃	—	6.4	11.0
	3	〃	lawn	—	5.1	8.8
	2	〃	grass	—	5.1	8.8
	6	May, 1966	soil	1.7	6.3	10.9
	2	〃	grass	4.2	—	—
	6	July, 1966	soil	2.5	6.0	10.3
Kyoto Univ.		Nov., 1966	lawn	2.3	7.2	12.4
Nagoya Univ. Pt.	6	Jan. 6, 1967	soil	4.5	9.0	15.5
	〃	Jan. 11, 1967	〃	—	8.3	14.3
	〃	Jan. 14, 1967	〃	3.3	8.3	14.3
	〃	Jan. 18, 1967	〃	3.3	7.5	12.9
	〃	Jan. 24, 1967	〃	—	7.1	12.2
	〃	Jan. 30, 1967	〃	2.3	6.9	11.9
	〃	Feb. 14, 1967	〃	—	6.5	11.2
	3	Jan. 14, 1967	lawn	3.3	6.7	11.5
	〃	Jan. 18, 1967	〃	—	6.0	10.3
	〃	Jan. 24, 1967	〃	3.0	5.9	10.2
	〃	Jan. 30, 1967	〃	—	5.9	10.2
	〃	Feb. 14, 1967	〃	—	5.0	8.6
	6	May 17, 1967	soil	1.0	5.4	9.3
	〃	June 18, 1967	〃	1.3	5.8	10.0
	〃	June 23, 1967	〃	1.3	5.8	10.0
	〃	June 26. 1967	〃	—	5.8	10.0
	〃	Feb. ε, 1968	soil	—	5.6	9.6
	8	Feb. 6, 1968	lawn	—	5.6	9.6
	2	Feb. 9, 1968	grass	—	5.6	9.6
	1	March 14, 1968	soil	1.0	5.4	9.3
	10	May 2, 1968	soil	—	4.9	8.5
	3	May 9, 1968	lawn	—	5.1	8.8
	8	Nov. 20, 1968	lawn	1.4	6.0	10.3
	11	〃	soil	0.60	5.6	9.6
	10	Dec. 28, 1968	soil	—	4.9	8.5
	11	May 9, 1969	soil	0.72	5.3	9.2

3rd Chinese nuclear test explosion: May 9, 1966.
5th Chinese nuclear test explosion: Dec. 28, 1966.
6th Chinese nuclear test explosion: June 17, 1967.

Fig. 1.4, the mean value of the ionization intensity due to the beta-radiation from the ground surface decreased gradually from 1963 through 1969. It was about 20 J in the grass area and 10 J in the soil area in the fall of 1963, and decreased to 2 J in the grass area and 1 J in the soil area in the beginning of 1969.

The decrease of the beta-radiation seems to be due to the decrease of fallout radioactivity dpeosited on the ground surface, because the additional nuclear ex-

plosion tests were not carried out often after 1663. The fact that the intensity of the beta-radiation in the grass area was higher than that in the soil area seems to be due to the accumulation of the fallout radioactivity caused by the nuclear explosion tests on the leaves and roots of the grasses.

Figure 1.5 shows the variation of the ionization intensity due to the gamma-radiation on the campus from 1963 through 1969. The variation is not so remarkable as that due to the beta-radiation. The results of measurements of the ionization intensity in several places after 1965 are shown in Table 1.1 (The results before 1964 were described in reference (1.1)).

ii) Sudden increase of the ionization due to the beta-, and gamma-radiations from the fallout radioactivity caused by the nuclear explosion tests

As is evident from Figs. 1.4 and 1.5, sudden increases of the ionizations due to the beta-, and gamma-radiations were observed in the beginning of Jan., 1967. The variations of the ionization intensities in Jan., 1967 are shown in detail in Fig. 1.6. The 5th Chinese nuclear explosion test was carried out at Lop Nor (90° E, 40° N), Chinese People Republic on Dec. 28, 1966. Radioactive particles produced by the test were transported by the Jet Stream, and reached the central area of Japan on Dec. 30, 1966[1,3].

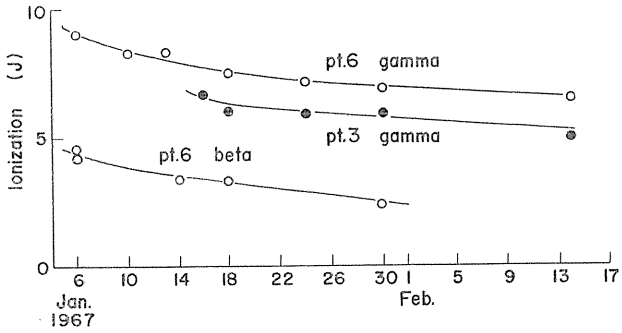

FIG. 1.6. Variations of the ionization intensities due to the beta- and gamma-radiations after the 5th Chinese nuclear explosion test.

It happened to rain on Dec. 30, and the radioactive particles were washed out by the rain and deposited on the ground surface. Unfortunately, no measurements were carried out on the campus before Jan. 6, 1967.

The ionization intensity which increased suddenly in the beginning of January decreased gradually, and reached ordinary level before March, 1967. The measured values of the ionizations due to the beta-, and gamma-radiations seems to be rather constant after March, 1967. The ionization due to the beta-radiation at one meter high above the ground surface amounts to about 1 J at present, while that due to the gamma-radiation (including the cosmic ray) amounts to about 5~6 J. According to Okano[1,4], similar sudden increases of the dose rate due to the gamma-radiation were detected in and around Tokyo on Dec. 30.

On the occation of the 3rd Chinese nuclear explosion test (May 9, 1966), dry fallout particles were detected with a G-M counter on the roof of the building of this department. Average number of dry fallout particles (higher than 10^3 cpm/

particle) on the roof was found to be about 18 particles/cm^2, and the maximum intensity of the radioactivity hold in one particle amounted to about 5×10^5 cpm. However, the ionizations due to the beta-, and gamma-radiations near the ground surface didn't increase remarkably. On the occations of the 6th and 7th Chinese nuclear explosion test (June 17, 1967 and Dec. 27, 1968 respectively), neither the deposition of fallout particles caused by the tests nor the increase of the ionizations were found at all.

iii) Nuclides of the fallout radioactivity

As described above, the ionization intensities due to the beta-, and gamma-radiations decreased gradually from 1963 through 1966, and the ionization intensity due to the beta-radiation in the area coverd with grass or lawn exceeded that in the soil area. The intense beta-radiation seems to be caused by highly concentrating fallout radioactive debris on the grass or lawn. To study th radioactive nuclides concentrated on grass and lawn, the energy spectra of the beta-, and gamma-radiations from the grass samples on the campus were measured.

About 5 gr. of grass was sampled at Pt. 3 (one of the stations on the campus) on June 14, 1964. Figure 1.7 shows the absorption curve of the beta-rays from the grass sample. ^{137}Cs and ^{90}Sr+^{90}Y were detected distinctly. A lump of grass with

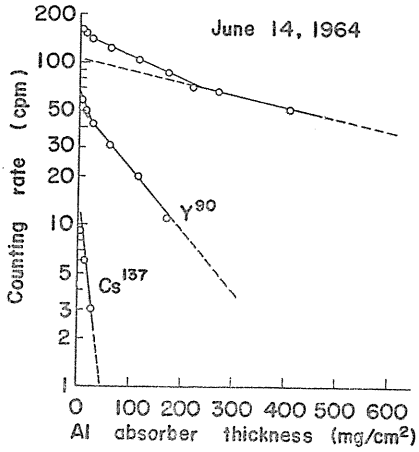

FIG. 1.7. The absorption curve of beta-rays from a lump of grass and soil sampled on the campus in 1964.

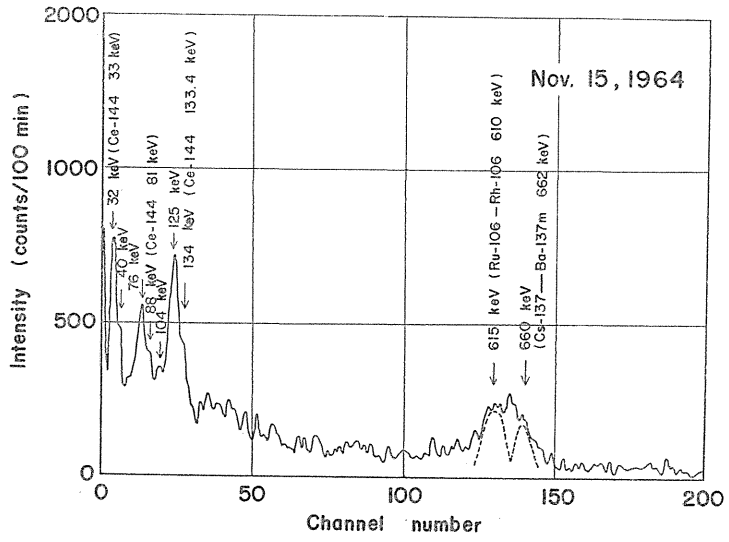

FIG. 1.8. The energy spectrum of gamma-rays from a lump of grass and soil sompled on the campus in 1964.

soil (30 $cm^2 \times 2$ cm in depth, 200 gr.) was also sampled at Pt. 3 on Nov. 15, 1964, and analyzed with a NaI(Tl) scintillation spectrometer. Figure 1.8 shows the gamma-ray spectrum of the sample. Photo-peaks due to ^{144}Ce, $^{106}Ru(+^{106}Rh)$, and ^{137}Cs $(+^{137m}Ba)$ were found. These nuclides were contained in the fallout radioactive debris. The obtained results coincide fairly well with those obtained by Fujita[1,5].

Shortly after the 3rd Chinese nuclear explosion test on May 9, 1966, dry fallout particles were collected on the roof of the building of this department, and radioactivity was analysed. Figure 1.9 shows the absorption curve of the beta-ray from the sample prepared by dissolving three particles in HCl solution. As is shown in the figure, ^{144}Ce and ^{140}La were detected. Figure 1.10 shows the gamma-ray spectrum of the fallout particles. Photo-peaks due to $^{95}Zr+^{95m}Nb$, ^{132}I, ^{132}Te, ^{140}La, and ^{144}Ce were detected. However, as described above, radiations from these highly radioactive debris had no influence on the ionization intensity of the air near the ground surface.

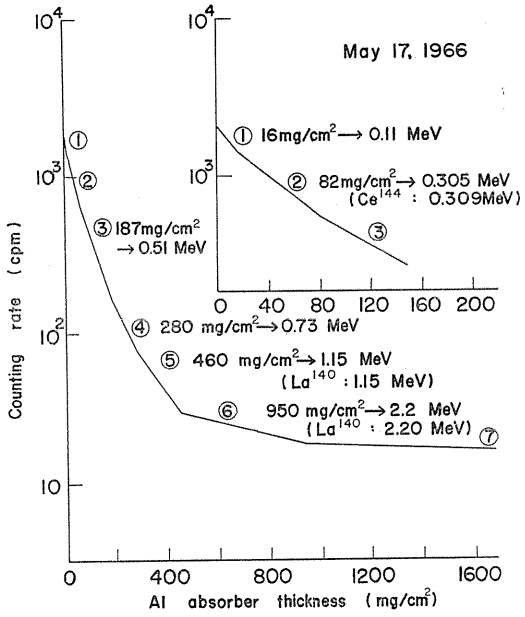

FIG. 1.9. The absorption curve of beta-rays from fallout particles caused by the 3rd Chinese nuclear explosion test.

1.4. Spectrometric method used for estimating the ionization intensity due to terrestrial gamma-radiation

The results described above were obtained by measurements with the ionization chamber without nuclide analysis. The ionization intensity due to the gamma-radiation described above includes that due to the cosmic ray. The ionization intensity due to the cosmic ray on the geomagnetic latitude of Nagoya was measured as 1.9 J. Therefore, the ionization intensity due to only terrestrial gamma-radiation may be estimated by subtracting 1.9 J from the value of the gamma-radiation described above.

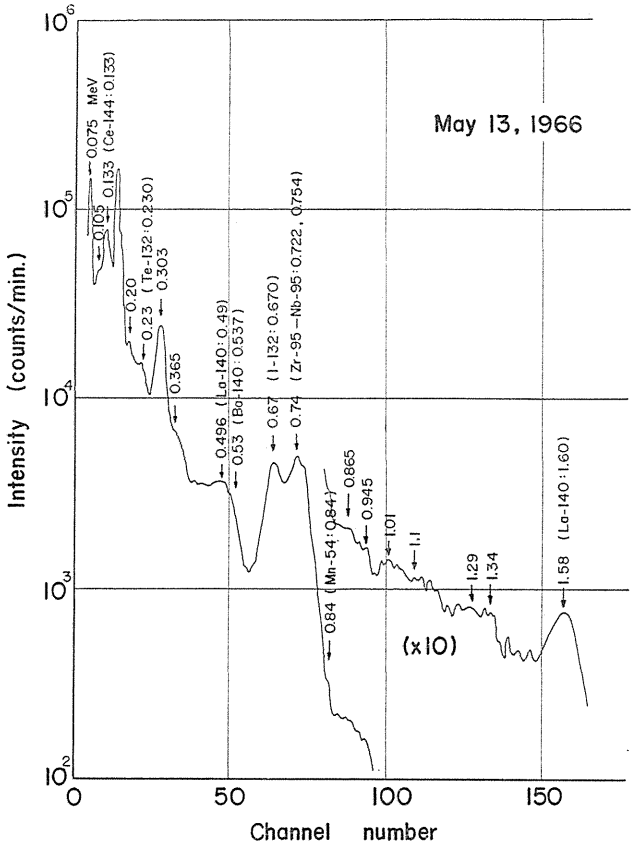

FIG. 1.10. The energy spectrum of gamma-rays from fallout particles caused by the 3rd Chinese nuclear explosion test.

To study the energy spectrum of terrestrial gamma-ray is important for detaied knowledges on terrestrial gamma-radiation. In general, it is considerably hard to estimate an energy spectrum in the air from a pulse height distribution measured with a scintillation spectrometer. The present authors used the method of transformation of the pulse height distribution obtained with a NaI scintillation spectrometer into real energy spectrum in the air by the response matrix method.

Correcting the response spectra calculated by Zerby *et al.*[1,6], we obtained the response spectra available to analyze the experimental results. Applying the response spectra, we made a response matrix for the broad beam incident to the rectangular direction of the axis of a cylinder ($3''\varphi \times 3''$ NaI scintillator), and analyzed the observed pulse height distribution by the response matrix[1,7]. We denote the pulse height distribution of terrestrial gamma-radiation, response matrix, and real spectrum by P_0, R, and N_0, respectively. The following relation may be consisted among these three functions.

$$P_0 = R \times N_0 \tag{1}$$

If we adopt pulse height distribution for the first approximation of spectrum N_0, the following relation is obtained.

$$N_1 = P_0 \tag{2}$$

Multiplying N_1 by the response matrix R we denote the product by P_1 as follows:

$$P_1 = R \times N_1 \tag{3}$$

The ith element of the second approximation of spectrum, N_2, is as follow:

$$(N_2)_i = \frac{(N_1)_i}{(P_1)_i} \times (P_0)_i \tag{4}$$

Multiplying N_2 by the response matrix R, we denote the product by P_2 as follow:

$$P_2 = R \times N_2 \tag{5}$$

The ith element of the third approximation of spectrum, N_3, is as follow:

$$(N_3)_i = \frac{(N_2)_i}{(R_2)_i} \times (P_0)_i \tag{6}$$

The iterative cycle is repeated untill the series of trial vectors, N_m, achieves a satisfactory degree of convergence, and real spectrum of the terrestrial gamma-ray can be obtained.

Figure 1.11 shows the pulse height distribution obtained at 1 m high above the ground surface on the campus in Dec., 1968. Figure 1.12 shows the result of analyzing the distribution shown in Fig. 1.11. The tranformation of the result shown in Fig. 1.12 into the ionization intensity is given by the following relation.

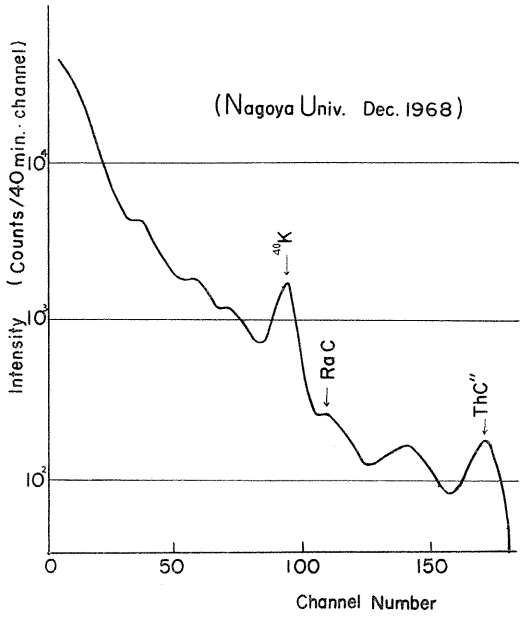

FIG. 1.11. Pulse height distribution of terrestrial gamma-ardiation.

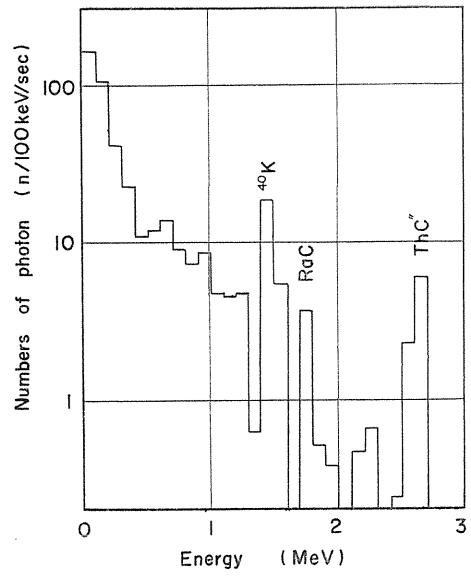

FIG. 1.12. The terrestrial gamma-ray spectrum analyzed by the responce matrix method.

$$q = \frac{\sum_{i=1}^{30} E_i \times N_i \times \mu_i \times L}{V \times W \times t}$$

where,

q: the ionization intensity (J)
E: the incident photon energy (MeV)
N: the number of incident photons (n/100 keV)
μ: the energy absorption coefficient of the air (cm^{-1})
L: the effective length of NaI(Tl) (cm)
V: the volume of a NaI(Tl)
W: W-value of the air (MeV/ion pair)
t: the measuring time (sec)

The comparison between the ionization intensity measured with the ionization chamber of plastic wall, 30 litres in volume, and that measured with the scintillation spectrometer is made. The result is shown in Table 1.2. As shown in this table, the former coincides fairly well with the latter. Subtracting the primary components of Thorium series, Uranium series, and ^{40}K successively, we can obtain the scattered component of terrestrial gamma-radiation by soil. The contribution of each series to the total ionization intensity can be also obtained from the result. Their contributions are shown in Table 1.3 and Fig. 1.13. The dose build up factor of soil, shown in Table 1.3, coincide well with that obtained by Hulqvist[1,8].

TABLE 1.2. Comparison of observed values obtained by the chamber method with values obtained by the present meihod

Location	Present method	ion chamber	
		(including cosmic-ray)	(γ-radiation Only)
No. 1	2.53 (J)	4.6 (J)	2.7 (J)
2	3.50	5.6	3.7
3	2.45	4.8	2.9
4	2.65	4.7	2.8
5	2.85	5.0	3.1

TABLE 1.3. Constitution of terrestrial gamma radiation obtained by the dresent method

		Number of Photons* (sec^{-1})	Total energy* (MeV. sec^{-1})	ionization (J)
Primary component	^{40}K	19.6	28.6	0.439
	U series	30.5	26.0	0.450
	Th series	21.5	29.5	0.413
	total of P.C.	71.6	84.1	1.302
Scattered component		371	85	1.41
Build up factor		6.2	2.0	2.1

* Numbers and total energies of photons passing through NaI Scintillator (3″φ×3″).

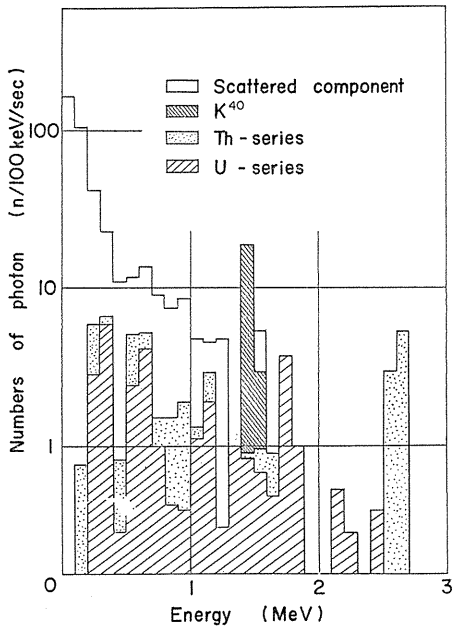

FIG. 1.13. Detailed profile of terrestrial gamma-radiation.

Reference

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Chapter 2. Interaction between RaA ions and condensation nuclei

2.1. *Introduction*

The RaA ions formed on the decay of radon-222 seem to be the most fundamental form of the naturally occuring radioactive aerosols. The mobility distribution of the naturally occuring radioactive ions were measured by Wilkening[2.1], Wilkening, Kawano, and Lane[2.2]. Bricard[2.3] also measured the mobility distribution of RaA ions formed on the decay of radon-222 which was emanated from Ra-solution. According to the result obtained by Bricard, the mobilities of primary ions were measured to be about 2.2 cm^2/sec. volt, which coincide fairly well with the value assumed by Chamberlain and Dyson[2.4].

These radioactive ions attach to aerosol particles, and form "radioactive aerosols". The attachment theory of radioactive ions has been made by several authors[2.5~2.9], and the size distribution of natural radioactive aerosols have also been measured by many authors[2.10~2.14]. However, few measurements have been made concerning the attachment process of natural radioactive ions[2.15].

In the previous paper[2.16], we reported the interaction between the natural radioactive ions and condensation nuclei. The correlation among the concentration of RaA ions (n_A), the concentration of condensation nuclei (Z), and the production rate of RaA ions (q_A) were well expressed by the following simple formula: $q_A=\beta_A n_A Z$, where β_A is the effective attachment coefficient between RaA ions and condensation nuclei.

In the present paper, we report the results of simultaneous measurements of q_A, n_A, Z, and D (the diffusion coefficient of condensation nuclei). The dependencies of the effective attachment coefficient between RaA ions and condensation nuclei (β_A) on the diffusion coefficient of condensation nuclei (D) and the size of condensation nuclei (r) are also discussed. Finally, β_A are compared with the effective attachment coefficient between small ordinary ions and condensation nuclei, β_0.

2.2. *Instruments used for measurements*

The instruments used for the present work are as follows:

i) Concentration of RaA ions (n_A)

The apparatus used for collecting radioactive ions is the same with the one described in the previous paper[2.17]. In the present work, the copper wire is maintained at negative potential of 225 volts. The critical mobility, denoted by k_c, of the radioactive ions trapped on the wire is calculated to be about 2.2 cm^2/sec. volt. The radioactive ions are collected for one hour, and at the end of the collection period, the wire is wound upon a flat spool for activity counting. A ZnS scintillation counter is used for alpha-counting, and the counted values are automatically recorded for 90 minutes after the end of the collection. As is mentioned in the previous paper, it was clarified that the radioactive nuclides trapped on the wire are mainly RaA. Then the time variation of alpha-activity of the wire may be expressed as follows:

$$I_\alpha = \lambda_A N_A + \lambda_C N_C$$
$$= \eta Q n_A \left[\frac{\lambda_B \lambda_C (1-e^{-\lambda_A T})}{(\lambda_B-\lambda_C)(\lambda_C-\lambda_A)} (e^{-\lambda_A t} - e^{-\lambda_C t}) \right.$$

$$- \frac{\lambda_C \lambda_A (1 - e^{-\lambda_B T})}{(\lambda_C - \lambda_B)(\lambda_B - \lambda_A)} (e^{-\lambda_B t} - e^{-\lambda_C t})$$
$$+ \left\{ (1 - e^{-\lambda_C T}) + \frac{\lambda_A \lambda_C}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_B)} (e^{-\lambda_B T} - e^{-\lambda_C T}) \right.$$
$$\left. - \frac{\lambda_B \lambda_C}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} (e^{-\lambda_A T} - e^{-\lambda_B T}) \right\} e^{-\lambda_C t}$$
$$+ (1 - e^{-\lambda_A T}) e^{-\lambda_A t} \Big] \qquad (1)$$

where,

I_α: alpha-counting rate at the time t (dps)
η: collection efficiency for RaA ions
Q: flow-rate (cm^3/sec)
n_A: concentration of RaA ions (cm^{-3})
N_A and N_C: numbers of RaA and RaC atoms collected on a wire respectively
λ_A, λ_B, and λ_C: decay constants of RaA, RaB, and RaC respectively (sec^{-1})
T: collection time (sec).

Substituting the values $\eta=1$, $Q=\left(\frac{25}{2}\right)^2 \times 200$ cm^3/sec, $\lambda_A=3.79\times10^{-3}$ sec^{-1}, $\lambda_B=4.31\times10^{-4}$ sec^{-1}, $\lambda_C=5.86\times10^{-4}$ sec^{-1}, and $T=7{,}200$ sec into formula (1), and using the value 0.080 as a counting efficiency of the scintillation counter, we can estimate the concentration of RaA ions (n_A) in the air from the counting rate.

ii) Concentration of condensation nuclei (Z)

A Pollak photoelectric nucleus counter was used for the present work. The calibration curve obtained by Nolan[2.18] was used for estimating the concentration of condensation nuclei from the measured value of extinction.

iii) Diffusion coefficient of condensation nuclei (D)

The diffusion battery methed[2.19][2.20] was used for measuring the diffusion coefficient of condensation nuclei. A metal diffusion battery with ten channels was used. The dimensions are:

Distance between two plates	=0.10 cm
Width of plates	=20 cm
Length of plates	=56 cm.

The total flow rate in the present work is about 0.8~1.2 l/min. The diffusion coefficient of condensation nuclei was estimated by using Gormley's formula[2.20]. Corresponding particle diameter (r) was calculated by using Stokes-Cunningham formula.

iv) Rate of RaA ion production (q_A)

Althongh it is well known that positively charged RaA atoms are present in the atmosphere, it has not been perfectly clarified as to how many per cents of RaA atoms are positively charged and how many per cents are neutral. Therefore, we assume hereafter that "$100\times f$ per cents of RaA atoms formed on the decay of radon-222 have a single positive charge and the remaining RaA atoms have no charge". Then we may estimate the rate of radioactive (RaA) ion pro-

duction using the concentration of radon gas in the air:

$$q_A = f q_{Rn} = f \lambda_{Rn} n_{Rn} \tag{2}$$

where,

q_{Rn}: rate of RaA atom production ($cm^{-3} sec^{-1}$)
λ_{Rn}: decay constant of Rn-222 (sec^{-1})
n_{Rn}: concentration of Rn-222 (cm^{-3}).

In the present work, the concentration of Rn-222 was measured by the charcoal trap method[2,17] or the filter pack method[2,21].

v) **Concentration of small positive ions**

A Gerdien type ion counter is used for measuring the concentration of small positive ions. The diameters of the outer cylinder and the inner electrode are 8 cm and 1 cm respectively, and the length of the inner electrode is 25 cm. The velocity of air flow is 200 cm/sec, and the potential difference between the cylinder and the electrode is 100 volts. For the values described above, the critical mobility of the ions is calculated to be about 1.3 cm^2/sec. volt.

2.3. Results of measurements

Simultaneous measurements of the concentrations of radon-222, RaA ions, condensation nuclei, and the diffusion coefficient of condensation nuclei were made in Feb., 1967 on the campus of this university. The concentration of small ordinary ions and the rate of small ion pair production were also measured.

In the case of RaA ion equilibrium, the following relation may be given:

$$\beta_A = \frac{q_A}{n_A Z} \tag{4}$$

where,

β_A: effective attachment coefficient between RaA ions and condensation nuclei ($cm^3 sec^{-1}$).

The rate of RaA ion production (q_A) cannot be directly measured. We can only determine the values of $q_{Rn}=q_A/f$ or β_A/f. However, as will be described in section 2.5, the value of f is estimated to be about 0.25.

According to the formula (4), n_A/q_A is expected to be inversely proportional to Z, if we assume β_A to be constant. In actual, the value of β_A is considered to be mainly controlled by the size of condensation nuclei ($2r$), and the time variation of r, hence β_A are remarkable. Figure 2.1 shows the correlation between n_A/q_A and Z. The data shown in the figure are classified into five groups* accrding to the values of D measured simultaneously. In each group, the values of D, hence β_A are rather constant. As shown in this figure, n_A/q_A is almost inversely proportional to Z. Thus, if we take the variation of D into consideration, the correlation among n_A, Z, and q_A is expressed by the simple formula: $q_A = \beta_A n_A Z$.

The effective attachment coefficient between RaA ions and condensation nuclei

* $D=(5\sim10,\ 11\sim40,\ 41\sim60,\ 61\sim90,\ >91)\times10^{-6}$ cm^2/sec.

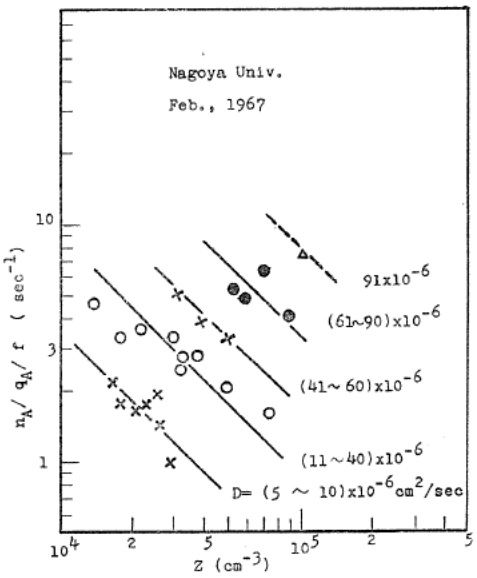

FIG. 2.1. The correlation between $n_A/(q_A/f)$ and the concentration of condensation nuclei. Data are classified according to the value of D.

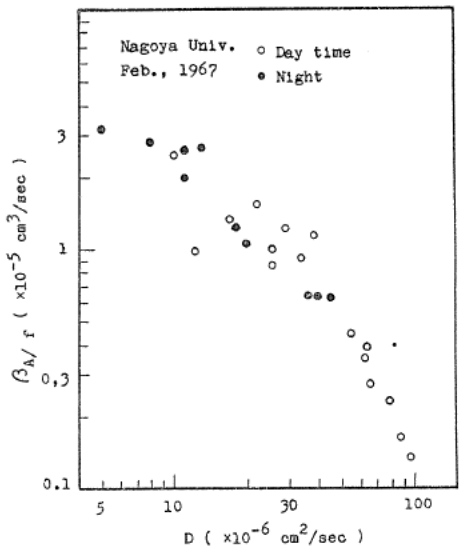

FIG. 2.2. The correlation between the attachment coefficient of RaA ions (divided by f) and the diffusion coefficient of condensation nuclei.

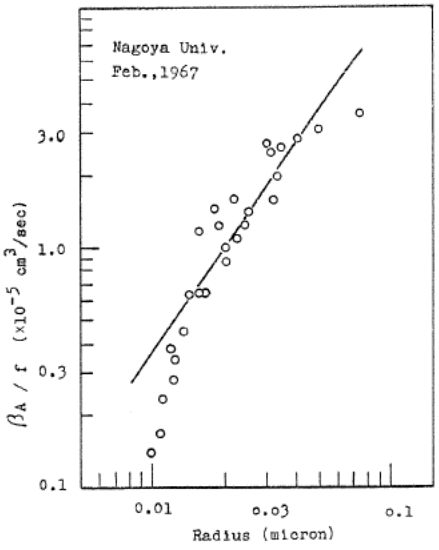

FIG. 2.3. The correlation between the attachment coefficient of RaA ions (divided by f) and the radius of condensation nuclei.

(β_A) may be estimated from the formula (4). β_A is considered to be a function of the diffusion coefficient of nuclei (D). Figure 2.2 shows the correlation between β_A/f and D. As is shown in this figure, β_A/f correlates fairly well with D.

The average diameter of condensation nuclei ($2r$) may be calculated by using Stokes-Cunningham formula. In Fig. 2.3, the correlation between β_A/f and r is shown. Similar correlations were also obtained at Tadeshina, in Aug., 1966.

According to Keefe and Nolan[2.22], the effective attachment coefficient between small ions and nuclei (b) is expressed theoretically as a function of that between ions and uncharged nuclei (η_0), and η_0 is given as follows:

$$\eta_0 = \frac{4\pi r D_0 (1+\sqrt{\pi y})}{1+\frac{4D_0}{rC}} \tag{5}$$

where,

r: radius of the nucleus (assumed spherical)
C: average speed of the small ions
D_0: diffusion coefficient of the small ions
$y = \frac{\varepsilon^2}{2rkT}\left(= \frac{2.88\times10^{-6}}{r} \text{ for a temperature of } 17°\text{C}\right)$

where ε is electronic charge, T is absolute temperature, and k is Boltamann's constant.

Our results of observations of small ions measured simultaneously with RaA ions suggested the excess of nucharged nuclei (or the lack of charged nuclei) in the atmosphere during the observation period[2.23]. In this case, b closes η_0.

In the case of RaA ion, $D_0=0.054$ cm^{2}sec^{-1} and $C=1.75\times10^4$ cm.sec^{-1}. Inserting these values into formula (5), we obtain η_0 as a function of r. The solid line in Fig. 2.3 represents η_0 (arbitrary unit). The measured values shown in the figure agree fairly well with the solid line in the range of r larger than 0.015 μ.

Lassen and Rau[2.5] obtained a similar equation, but without the image term, to describe the attachment of ThB to aerosol particles. Our measured values seem to agree well with the theory of Lassen and Rau in the range of r smaller than 0.03 μ.

2.4. *Comparison of the attachment coefficient of RaA ions (β_A) with that of small ordinary ions (β_0)*

As is mentioned above, the concentration of small ordinary ions (n_0) and the rate of small ion pair production (q_0) were also measured simultaneously. Therefore, we can calculate the effective attachment coefficient between small ions and condensation nuclei (β_0) using the following formula: $\beta_0 = \frac{q_0}{n_0 Z}$.

On the other hand, concerning RaA ions we can only calculate the value of β_A/f, and the values of f, therefore β_A remain unknown.

Keefe and Nolan calculated η_0 for small ordinary ions of which mobilities are 1.34 cm^2/sec. volt as a function of the radius of nuclei. We can also calculate η_0 for RaA ions of which mobilities are 2.2 cm^2/sec. volt using formula (5). We denote the former by η_{00} and the latter by η_{0A}. Calculations show that η_{0A} coincide almost with η_{00} in the range of radius from 0.91 μ to 0.6 μ:

$$\frac{\beta_A}{\beta_0} \equiv \frac{\eta_{0A}}{\eta_{00}} = 1.0.$$

Thus, β_A is considered to coincide well with β_0.

In actual, β_A/f is found to be almost proportional to β_0. Figure 2.4 shows the correlations between β_A/f and β_0 obtained on Nagoya University. As is shown in the figure, β_A/f correlates fairly well with β_0. The value of the ratio $(\beta_A/f)/\beta_0$ is about 4.0 (Fig. 2.4). In the figure, the solid line represents the correlation between 4.0 η_{0A} and η_{00}.

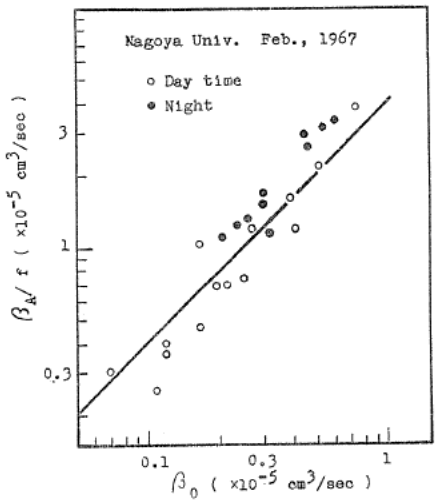

FIG. 2.4. The correlation between the attachment coefficient of RaA ions (divided by f) and that of small ordinary ions, obtained on the campus of this university.

Comparing the observed result with the theoretical one, we can obtain the following relation: $(\beta_A/f)/\beta_0=1/f=4.0$. Thus, the value of f should be estimated at about 0.25 on the average; therefore, in general, about 25 per cents of RaA atoms formed on the decay of radon-222 may have some charge, and the remaining 75 per cents may have no charge.

2.5. Correlations between each element obtained on Mt. Norikura

In the previous paper[2.17], we reported the correlations among concentrations of RaA ions, small ions, and condensation nuclei. We can proceed with our studies concerning these correlations.

Figure 2.9 in Ref. (2.17) shows the correlation between the concentration of RaA ions and that of small ions. The concentration of RaA ions correlates fairly well with that of small positive ions. A similar correlation was reported previously by one of the authors[2.2]. Now we assume an ionization equilibrium of small positive ions. Then the following relation may be given[2.23]:

$$q_0 = \beta_0 n_0 Z \tag{6}$$

where,

q_0: production rate of small positive ions ($cm^{-3}\,sec^{-1}$)
β_0: effective attachment coefficient between small positive ions and condensation nuclei ($cm^3\,sec^{-1}$)
n_0: concentration of small positive ions (cm^{-3}).

Taking the formulae (4) and (6) into consideration, we get the following relation:

$$n_A = \left(\frac{\beta_0}{\beta_A}\frac{q_A}{q_0}\right). \tag{7}$$

The concentrations of radioactive ions and that of small ions are measured "simultaneously". Accordingly, the average size of the nuclei which are attached by small ions may be considered to be almost the same as that by radieactive ions. Therefore, in formula (7), we can consider the ratio β_A/β_0 to be almost constant. The production rate of small ions, denoted by q_0, is almost independent of time, and we obtained:

$$\begin{aligned} &\text{Nagoya Univ.: } q_0 = 14.9 \text{ J} \\ &\text{Mt. Norikura: } q_0 = 14.5 \text{ J}. \end{aligned} \tag{8}$$

According to the formula (7), the concentration of radioactive ions is expected to be proportional to that of small positive ions, if we assume the production rate of radioactive ions (q_A) to be constant (see Fig. 2.9 in Ref. (2.17)). However, the time variation of q_A is considerably wide actually. Therefore, according to the formula (7), it is probable that the ratio n_A/q_A is proportional to the concentration of small positive ions (n_0). Figure 2.5 shows the correlation between n_A/q_A and n_0. The correlation shown in Fig. 2.5 is better than that shown in Fig. 2.9 in Ref. (2.17).

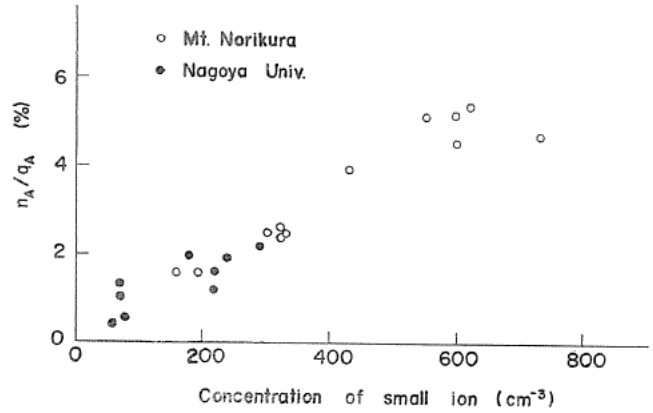

FIG. 2.5. The correlation between n_A/q_A and the concentration of small ordinary ions.

Fig. 2.10 in Ref (2.17) shows the correlation between n_A and Z. The correlation is not so evident as is expected by the formula (4).

According to the formula (4), n_A/q_A is expected to be inversely proportional to Z, if we assume β_A to be constant. However, as the time variation of the size of nuclei is considerably remarkable, that of β_A is also remarkable. Unfortunately, we did not measure the size of condensation nuclei on Mt. Norikura. But we can presume the value of β_A from the value of β_0, because β_A is considered to be almost proportional to β_0. Figure 2.6 shows the correlation between n_A/q_A and

Z. The data shown in the figure are classified into five groups* according to the values of β_0 estimated from the equation (6). In each group, the value of β_0 are rather constant. As shown in this figure, n_A/q_A is almost inversely proportional to Z. Thus, if we take variation of β_0 (hence β_A) into consideration, the relation among n_A, Z, and q_A is expressed by the simple formula: $q_A=\beta_A n_A Z$.

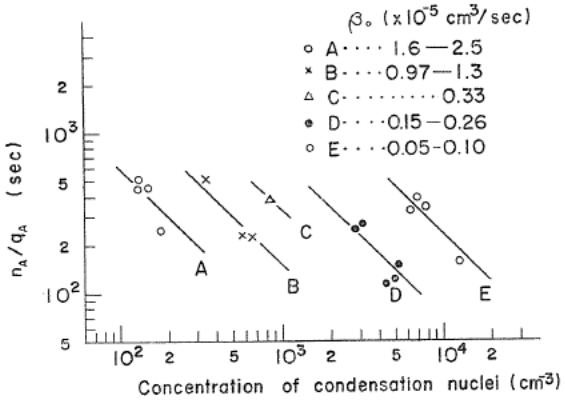

FIG. 2.6. The correlation between n_A/q_A and the concentration of condensation nuclei.

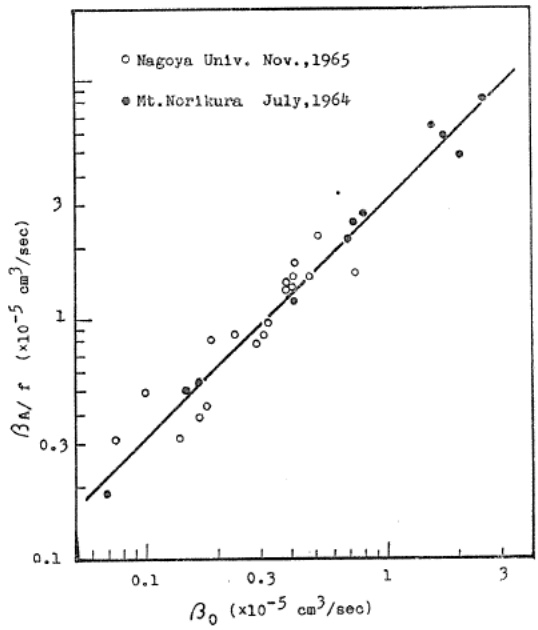

FIG. 2.7. The correlation between the attachment coefficient of RaA ions (divided by f) and that of small ordinary ions obtained on Mt. Norikura.

* $\beta_0=(1.6\sim2.5,\ 0.97\sim1.3,\ 0.33,\ 0.15\sim0.26,\ 0.05\sim0.01)\times10^{-5}$ cm³/sec.

Figure 2.7 shows the correlation between β_A/f and β_0 obtained on Mt. Norikura. The correlation is similar to that shown in Fig. 2.4.

2.6. Conclusion

To study the interaction between RaA ions and condesation nuclei, the simultaneous measurements of the concentrations of RaA ions, condensation nuclei and radon-222 were carried out. The diffusion coefficients of nuclei were also measured. It was found that the correlation among them is well expressed by the simple formula: $q_A=\beta_A n_A Z$. The correlation between β_A and D or r was also obtained. The functional form of β_A coincide fairly well with those obtained theoretically by Keefe and Nolan in the range of radius larger than 0.015 μ.

The effective attachment coefficient of RaA ions was compared with that of small ordinary ions. Consquently, it was presumed that about 25 per cents of RaA atoms formed on the decay of radon-222 seemed to have some charge, and remaining 75 per cents have no charge.

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