

Correction for Recombination Loss of Ions in Ionization Chambers

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Abstract

The signal charge loss due to initial recombination and that due to volume recombination have been measured separately using parallel plate ionization chambers irradiated with cobalt-60 gamma rays, over a wide range of applied voltage and air humidity. It is shown that both initial and volume recombination depend on the electric field strength and also on humidity through ion clustering reaction with water molecules. In spite of the usual assumptions, a linear relation has not been observed between the inverses of the signal current and applied voltage for initial recombination, nor between the inverses of the signal current and square of the applied voltage for volume recombination. Methods and data which can be used for estimation of initial and volume recombination losses of ions in ionization chambers are proposed.

I. INTRODUCTION

The output current, i , from an ionization chamber is reduced from the saturation current, i_s , by initial recombination, back-diffusion of ions to electrodes, and volume recombination. If the loss due to these phenomena is small, the collection efficiency, $f = (i/i_s)$, can be expressed as

$$f = f_i f_d f_v \quad (1)$$

where f_i , f_d , and f_v are collection efficiencies corresponding to initial recombination, back-diffusion, and volume recombination respectively.

In most cases, volume recombination is the dominant cause for the ion loss and has been studied by many authors. Volume recombination loss is usually estimated by linear extrapolation of the inverse of the signal current expressed as a function of the inverse of the square of the applied voltage, U . This is because the collection efficiency for volume recombination can be expressed as

$$f_v = 1 - f_v \xi^2 / 6 \quad (2)$$

$$\xi = m d^2 q^{1/2} / U \quad (3)$$

$$m = (\alpha / e K_+ K_-)^{1/2} \quad (4)$$

where d is the separation between the two parallel plate electrodes, q the charge of ions of either sign produced by radiation per unit volume per unit time, α the recombination coefficient, e the charge of an ion, and K_+ and K_- the mobilities of positive and negative ions respectively [1,2].

Initial recombination is a phenomenon between positive and negative ions which result from a single ionizing particle and can be separated into cluster recombination and columnar recombination. Cluster recombination is a phenomenon

between ions produced by the same delta electrons with small energy, and columnar recombination is that between ions which are produced in a columnar space around the track of the parent ionizing particle. It is said that the inverse of signal current and that of the electric field strength in a chamber have a linear relation at high electric field strength when initial recombination is dominant [2].

It is known, however, that both initial and volume recombination depend on the humidity of the air in the ionization chambers. This is attributed to the clustering reaction of ions with water molecules. The value of m in Equation (4), which is a physical parameter for volume recombination, has been found by experiment to depend on the lifetime of the ions and also on humidity of the air in the ionization chambers [3, 4]. For initial recombination, the following empirical equation has been proposed:

$$f_i = 1 - a_i [\ln(1/E + b_i) - \ln(b_i)] \quad (5)$$

where E is electric field strength U/d . The symbols a_i and b_i are fitting parameters which depend on humidity [5].

Concerning diffusion loss, equations of f_d have been derived for parallel plate, cylindrical, and spherical ionization chambers [6, 7]. It is known from these equations that ions are lost by back diffusion if they are produced in an electrode vicinity where the electric potential difference from the electrode is less than kT/e , where k is the Boltzmann constant and T the gas temperature. This is correct except at extremely low applied voltages [7]. This means that back diffusion loss can be estimated for any type of ionization chamber by computation of the electric potential distribution in the chamber [8-10]. It is also confirmed by experiment that diffusion loss does not depend on humidity [5].

In the present work, saturation characteristics for initial recombination and volume recombination are measured separately using parallel plate ionization chambers irradiated with cobalt-60 gamma rays over a wide range of applied voltage and of humidity. Improved methods for estimation of initial and volume recombination losses in ionization chambers are proposed.

II. EXPERIMENT

The apparatuses used in the present experiment are the same as those used previously [4, 5]. Collection efficiency for initial recombination was measured irradiating a parallel plate graphite ionization chamber at the lowest exposure rates (0.22 to 25 nCkg⁻¹s⁻¹) in order for volume recombination loss to be small. The ionization chamber used has a collector with a diameter of 34 cm surrounded by a guard plate with an outer

diameter of 40 cm. Volume recombination was measured at high exposure rates (6.3 to $250 \mu\text{Ckg}^{-1}\text{s}^{-1}$) so that diffusion loss and initial recombination loss become relatively small compared to the volume recombination loss. The ionization chamber used for measurement of volume recombination loss has a collector with a diameter of 10 cm surrounded by a guard electrode with an outer diameter of 18 cm. The separation between the collector and polarizing electrodes was fixed at various lengths in a range between 1 and 13 mm by increment of 1 mm (for initial recombination) and 3, 7, 10, 15, 20, or 30 mm (for volume recombination) by changing glass tube spacers.

The humidity of the air in these ionization chambers was controlled by circulating air through a trap maintained at a temperature in a range from -75 to -60 °C, or through a gas washing bottle which contained water and was fixed at a constant temperature in a range from 0.5 to 19 °C. The ionization chambers were always maintained at room temperature. The humidity of the air was measured with a dew point hygrometer at about 7 m down the flow from the ionization chamber in a stainless steel pipe. The temperature and pressure of the air were measured in the ionization chambers. The lowest humidity observed was 0.6 and 0.04 % for the measurement of initial recombination and for that of volume recombination respectively. This difference is due to the fact that the ionization chamber used for the measurement of initial recombination was installed in an aluminum container which was not so tight but that for volume recombination was installed in an aluminum chamber which was made of flanges for high vacuum.

The temperature of the air measured in the ionization chambers ranged from 21.0 to 22.7 °C all through the present measurements. The fluctuation of the temperature for one saturation-curve measurement was usually less than 0.03 °C except for several measurements for dry air; in these cases, temperature drifted in a range of 0.2 °C during one saturation-curve measurement. (These conditions were slightly different for measurement in [4, 5]).

Saturation currents were obtained from currents measured for high applied voltages assuming a linear relation between $1/f$ and $1/U$ or $1/U^2$. Results with an error of less than 2% are shown for both the initial recombination loss and m values in the present paper.

III. EXPERIMENTAL RESULTS

A. Initial Recombination

Figure 1 shows experimental results for f_i as a function of the inverse of the electric field strength, $1/E$ obtained for humidity of 56%. The results were obtained from mean values of absolute currents measured for both polarities and corrected for diffusion loss and volume recombination loss. Correction for volume recombination loss was made using the results for m which will be discussed later in the present paper. The solid line in the figure was obtained fitting Equation (5) to the experimental results. The equation was applied because of its

simplicity among several equations that pass through the point of $f_i = 1$ at $1/E = 0$ m/V and also because it agrees relatively well with the experimental data in a wide range of $1/E$.

Figure 2 shows the inverse of f_i obtained by fitting Equation (5) to the experimental results for the various values of humidity. The values of the fitting parameters a_i and b_i for each line are listed in Table 1. The ranges of humidity and pressure during measurement are also shown in the table. Some of the lines in Figure 2 are not in the order of the humidity values. This is due to large scatter of experimental results and also because some measurements were made only in a small range of electric field strength. The results for the humidity of 1.6, 30, and 74 % were obtained from data measured in a range less than 0.014 m/V; these data are the same as those shown in [5]. In the present paper, however, they are corrected not only for diffusion loss but also for volume recombination loss before fitting Equation (5). Some of the results were not used in the present paper because their errors exceeded 2%.

It is known from Figures 1 and 2 that there is not a linear relation between the inverse of the signal current and that of the electric field strength in the range of the electric field

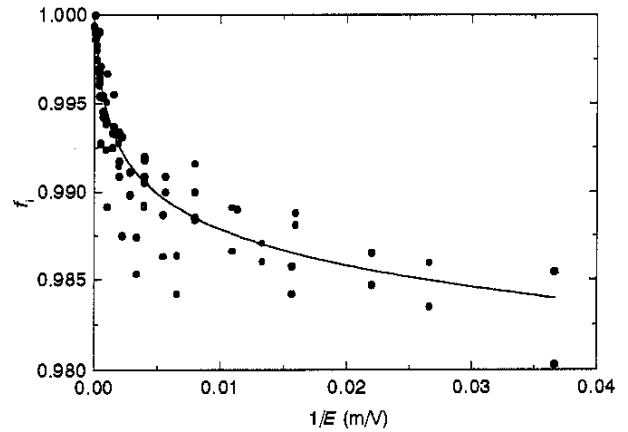


Figure 1: Experimental results of f_i for humidity of 56%.

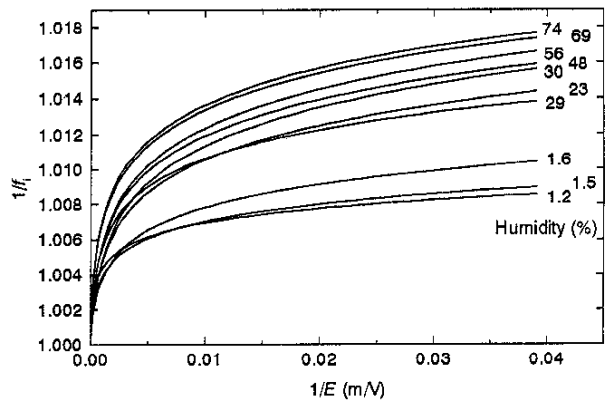


Figure 2: Inverse of f_i obtained for various values of humidity.

Table 1.
Values of fitting parameters in Equation (5) for the lines
shown in Figure 2.

Humidity (%)	Pressure (hPa)	a_i (10^{-3})	b_i (10^{-6}m/V)
1.2 ± 0.6	1008 ± 14	1.37 ± 0.10	64 ± 13
1.5 ± 0.2	1006 ± 4	1.12 ± 0.13	22 ± 10
1.6 ± 0.2	1003 ± 11	1.86 ± 0.27	158 ± 60
23.1 ± 0.3	1007 ± 7	3.03 ± 0.18	261 ± 36
29.0 ± 0.8	1009 ± 6	2.28 ± 0.20	101 ± 24
30.0 ± 0.4	1003 ± 13	2.72 ± 0.23	221 ± 45
48.0 ± 1.4	1013 ± 10	2.81 ± 0.14	160 ± 21
56.7 ± 1.3	1007 ± 10	3.05 ± 0.17	186 ± 25
69.4 ± 2.1	1011 ± 8	2.85 ± 0.15	99 ± 14
74.6 ± 0.4	1004 ± 11	2.87 ± 0.26	95 ± 25

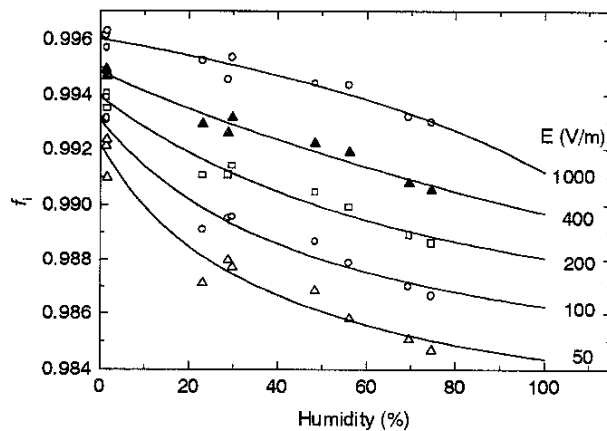


Figure 3: Symbols: values of f_i obtained from Equation (5). Solid lines: result of Equation (6) fitted to the values for the symbols.

strength measured in the present experiment. The steep change in the signal current near the zero of $1/E$ is attributed to cluster recombination and the slow change in the range of larger $1/E$ to columnar recombination [5].

Figure 3 shows the values of f_i as a function of humidity for several different values of E . The values for the symbols in the figure are obtained from Equation (5) with the values for a_i and b_i in Table 1. The solid lines in Figure 3 show the results obtained by fitting the following equation to the values for each electric field strength

$$f_i = (c_2 + c_3 h) / (1 + c_1 h) \quad (6)$$

where h is the humidity and c_1 , c_2 , c_3 are fitting parameters. Several other formulas such as linear and quadratic equations were also applied; Equation (6) showed the most reasonable results over the entire range of humidity except for $E = 1000$ V/m, for which it is slightly concave down. By the method mentioned above, we can estimate the initial recombination loss of signal current from parallel plate ionization chambers for any electric field strength and for any

humidity.

B. Volume Recombination

Figure 4 shows the values of m obtained for various values of humidity as a function of $d^2/U(P_{\text{H}_2\text{O}}/P_{\text{total}})$, where $P_{\text{H}_2\text{O}}$ and P_{total} are water vapor pressure and total pressure respectively. The value of the function is proportional to the probability of collision with water molecules for ions in their lifetime drifting in an ionization chamber. The open symbols (\circ , \triangle , and \square) and $*$ show the results for the data used in [4] but are corrected for initial recombination and diffusion loss. The symbols $*$, which are for humidity of 5-10%, are scattered in a relatively wide range. This is attributed to a large fluctuation of humidity during measurements of signal current for saturation curves [4]. We have not measured for the range of humidity in the present work. All the results shown in Figure 4 were obtained from saturation curves after being corrected not only for initial recombination loss and diffusion loss, but also for applied voltage effect [11, 12]. The correction procedure for the applied voltage effect is described in Appendix A.

In [4], it is suggested that the value of m can be expressed as a function of $d^2/U(P_{\text{H}_2\text{O}}/P_{\text{total}})$. It is noted in Figure 4, however, that the value depends on humidity even for a fixed value of $d^2/U(P_{\text{H}_2\text{O}}/P_{\text{total}})$. It follows from this fact that at a constant value of $d^2/U(P_{\text{H}_2\text{O}}/P_{\text{total}})$, more water molecules are attached to ions by clustering reaction when humidity is higher. This may be explained if it can be assumed that water molecules stay not as isolated molecules but as larger water clusters when humidity is higher, and that the attachment factor of the cluster becomes larger with the size of the cluster.

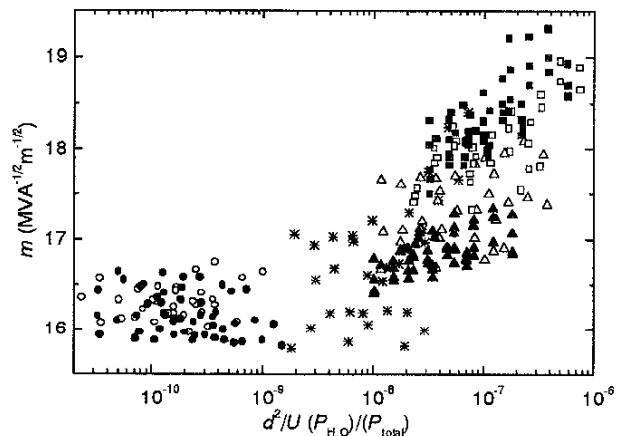


Figure 4: Values of m obtained for air with humidity of \circ , 0.10%; \bullet , 0.11%; $*$, 5-10%; \blacktriangle , 23%; \triangle , 39%; \blacksquare , 73%; and \square , 81%. The value of the abscissa, $d^2/U(P_{\text{H}_2\text{O}}/P_{\text{total}})$, is proportional to the number of collisions of ions with water molecules during drift time in the ionization chamber.

Table 2
Values for parameters in Equation (7) for different values of humidity.

Humidity (%)	Pressure (hPa)	m_0 (MVA ^{-1/2} m ^{-1/2})	A_1 (MVA ^{-1/2} m ^{-1/2})	t_1 (10 ⁻⁶ m ² V ⁻¹)
0.11 ± 0.1	1007 ± 12	16.22 ± 0.03	-	-
22.9 ± 1.0	1010 ± 7	16.40 ± 0.15	0.62 ± 0.14	4.8 ± 2.0
39.3 ± 1.5	1010 ± 13	17.18 ± 0.15	1.12 ± 0.41	55 ± 263
73.4 ± 1.5	1005 ± 9	17.70 ± 0.16	1.26 ± 0.16	8.0 ± 3.0
80.7 ± 3.0	1014 ± 8	17.67 ± 0.11	2.18 ± 2.15	46 ± 66

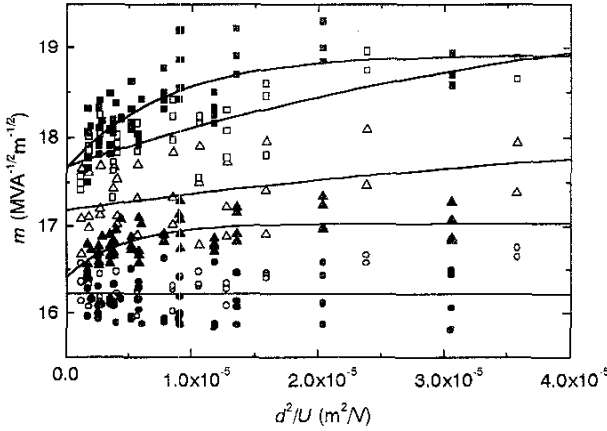


Figure 5: Values of m shown in Figure 4, except those for humidity of 5-10 %, are plotted as a function of a factor d^2/U , which is proportional to the lifetime of ions in ionization chambers. The solid lines show results of Equation (7) for parameter values listed in Table 2.

Figure 5 shows the results of m measured for different values of humidity, as a function of d^2/U , which is a parameter proportional to the lifetime of ions in ionization chambers. It does not show the results for humidity of 5-10%. The solid lines in the figure show the values of the following equation obtained by adjusting the fitting parameters, m_0 , A_1 , and t_1 to each experimental result for different humidity:

$$m = m_0 + A_1(1 - \exp(-t/t_1)) \quad (7)$$

where $t = d^2/U$. Table 2 shows the values of the parameters for each line in the figure, humidity, and pressure during measurements. The value of m_0 for humidity of 0.11 % in the table was obtained assuming $A_1 = 0$ for both the experimental results measured for humidity of 0.1 and 0.11 %.

Symbols in Figure 6 show the values of m calculated by Equation 7 for several different values of d^2/U . The solid lines in the figure show approximate linear functions for each result at different values of d^2/U . Although the errors of each value for the parameters are relatively large, it is possible to get reasonable values of m for any humidity.

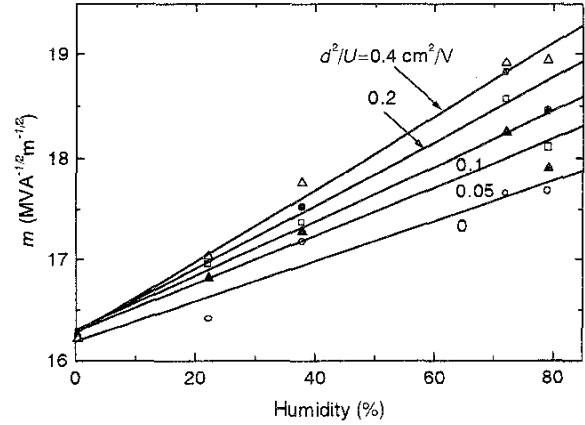


Figure 6: Values of m obtained by Equation (7) with the parameter values listed in Table 2, for several different values of d^2/U . Solid lines show the results of fitting a linear function to each set of data.

IV. APPLICATION OF THE DATA FOR RECOMBINATION LOSS

To show the applicability of the results obtained in the present work, we compare the ion loss in a cylindrical ionization chamber experimentally obtained and that calculated from the parameters obtained using parallel plate ionization chambers. Figure 7 shows various collection efficiencies for a cylindrical ionization chamber as a function of the applied voltage, U . The chamber has glassy carbon electrodes and guard electrodes around both ends of the inner electrode. The charge collection volume of the chamber has an inner and an outer radius of 2.5 and 13.1 mm respectively, and it has a length of 282 mm [13]. The measurement was made for a temperature of 21.1 ± 0.1 °C, atmospheric pressure of 1007 ± 9 hPa, and humidity of 26.4 ± 0.6 %. The solid and broken lines in the figure respectively show collection efficiencies, f_d^c , for diffusion loss in the cylindrical ionization chamber calculated by exact and approximate equations derived in [7]. The circles in the figure show experimental results for the collection efficiency. The

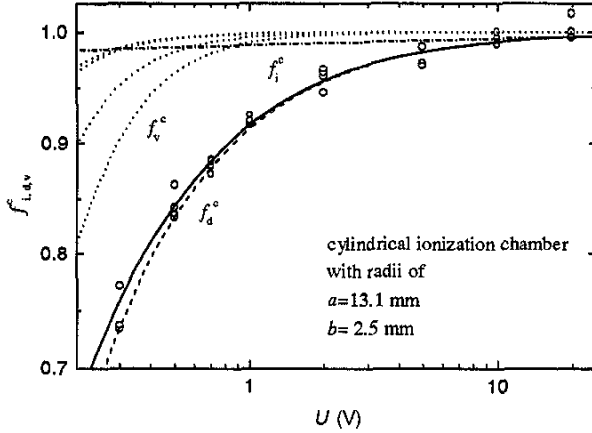


Figure 7: Various collection efficiencies for a cylindrical ionization chamber when the applied voltage is U . The solid and broken lines respectively show collection efficiencies, f_d^c , for diffusion loss calculated with exact and approximate equations derived in [7]. The circles in the figure show experimental results for the collection efficiency. The chain line and dotted lines show collection efficiencies for initial recombination, f_i^c , and those for volume recombination, f_v^c , respectively. These values were used to obtain the experimental result for f_d^c .

experimental values were obtained from results measured for total collection efficiency, f^c , by collecting for initial recombination loss and volume recombination loss calculated as follows.

The values of collection efficiency for initial recombination loss, f_i^c , and that for volume recombination loss, f_v^c , in the cylindrical ionization chamber are shown in Figure 7 by chain and dotted lines respectively. However, the correspondence of the dotted lines to the circle symbols is not shown in the figure. The collection efficiency for initial recombination loss in a cylindrical ionization chamber can be obtained by the following equation because initial recombination depends only on the electric field strength at the place where the ion pairs are produced.

$$f_i^c = \frac{\int_b^a 2\pi r f_i[E(r)] dr}{\int_b^a 2\pi r dr} = \frac{2 \int_b^a r f_i[E(r)] dr}{(a^2 - b^2)} \quad (8)$$

where a and b are respectively the outer and inner radius of the charge collection volume of the cylindrical ionization chamber, and f_i the collection efficiency obtained with parallel plate ionization chambers for the electric field strength, E , at a radius of r in the cylindrical ionization chamber.

For volume recombination in the cylindrical ionization chamber, the following equations are proposed [1,2] instead of Equation (3):

$$\xi = m \frac{[(a-b)\kappa]^2 q^{1/2}}{U} \quad (9)$$

$$\kappa = \left[\frac{a/b + 1 \ln(a/b)}{a/b - 1} \right]^{1/2} \quad (10)$$

To take into account the dependence of m value on the lifetime of ions and humidity, we used a value of m which was obtained using parallel plate ionization chambers for the mean lifetime of ions and humidity which are respectively the same with those in the cylindrical ionization chamber. The mean lifetime used here is that of ions which are lost by volume recombination in each ionization chamber. In the derivation of an equation which defines the relation between the mean lifetime of ions in each ionization chamber, it is assumed that the mobility of positive and negative ions is the same (Appendix B).

The good agreement between the experimental and theoretical values of f_d^c shown in Figure 7 not only confirms the validity of equations for diffusion loss of ions in cylindrical ionization chambers but also justifies the calculation method of recombination loss of ions in cylindrical ionization chambers.

V. DISCUSSION

In the present work, the space charge effect has been ignored. The effect is small for the range of exposure rates in the present experiment because no systematic dependence of m value on exposure rates was found. The m value obtained in the present experiment shows an average of values throughout the lifetime of ions for each experimental condition. More accurate equations may be derived for the relation between the average lifetime of ions in parallel plate ionization chambers and that in cylindrical ionization chambers if precise values of mobility for both ions with different polarities are obtained. The dependence of the cluster size of water molecules on humidity must be an important problem in cloud physics; however, we could not find data about this phenomenon.

Niatel [14] obtained initial recombination loss and m values which depend on the electric field strength using a free air ionization chamber. Although humidity is not mentioned in [14], the results for initial recombination loss agree with values of the present result for humidity in a range from 20 to 60 % if the ion loss at the largest electric field strength i.e. 200V/cm, which she has assumed to be a saturation range, is neglected. If we take into account this loss, the humidity is estimated to be in the range from 70 to 90 % for her experiment. The m values, which are obtained from the results in Table 1 of [14], are distributed in a range from 20.2 to $18.1 \times (\text{MVA}^{-1/2} \text{m}^{-1/2})$ for electric field strength from 27.8 to 77.8 V/cm. A large value such as $20.2 \text{ MVA}^{-1/2} \text{m}^{-1/2}$ is out of the range of the present results for m values obtained from Equation (7) as shown in Figure 6.

Quite recently, Boutillon [15] obtained a value $19.9 \text{ MVA}^{-1/2} \text{m}^{-1/2}$ for m value at 20 °C, 1013.25 hPa, and humidity of 50% using free air ionization chambers. She has also measured dependence of initial recombination loss of ions on

humidity and that of recombination loss on air density. It is suggested that m value depends on 1.215 power of air density [15]. According to the suggestion, the m value of the present result must be increased by 2 % at most to normalize the value for the air density of [15]. This increase is not enough for the large discrepancies between the results of m values reported in [14, 15] and that of the present experiment. The reason for these large discrepancies is not clear.

The ion loss due to initial recombination and that due to diffusion become relatively large compared to volume recombination loss for low exposure rate at high applied voltages and for small separation between electrodes of ionization chambers. For these conditions, it is difficult to obtain, by experiment, the loss of ions due to each phenomenon separately. In conclusion, a method was discussed which makes it possible to estimate the amount of ion loss due to initial recombination, volume recombination, and diffusion for various types of ionization chambers for a wide range of applied voltage and humidity, taking into account the effect of humidity.

VI. APPENDIX A

When a parallel plate ionization chamber is irradiated from the front side (polarizing electrode) with cobalt-60 γ rays, the output current increases and decreases linearly with the positive and negative applied voltage (beyond the recombination region) respectively [11, 12]. This increase/decrease is attributed to the change in the stopping power of the air due to acceleration/deceleration of secondary electrons in the applied electric field. Although the change in the output current due to this effect is small per unit of applied voltage, the effect can not be cancelled out by using mean values of the absolute values of the output current for both polarities. This is because the increase in the output current depends on the distance between the two electrodes, but the decrease is almost constant on the distance if it is in a range less than 0.03 m.

The current, I_0 , which is corrected for the applied voltage effect is obtained from a current, I , measured at an applied voltage of U by the following equation:

$$I/I_0 = 1 + gU$$

where g is a coefficient for increase/decrease of the current due to the applied voltage effect. It has a constant value of 1.84×10^{-6} 1/V for negative applied voltage and has a value obtained by the following equation for positive applied voltage:

$$g = (19 \times 10^{-6} + 0.71 \times 10^{-3}d) / (1 + 732d)$$

where d is the distance between the two electrodes in units of m. All these values were obtained from experimental data shown in [11].

VII. APPENDIX B

The value of m which is used for volume recombination loss in a cylindrical ionization chamber must be the same as that obtained for parallel plate ionization chambers for the

mean-lifetime identical with that in the cylindrical ionization chamber. This mean-lifetime should be the average of lifetimes of ions which are lost in each ionization chamber by volume recombination. The number of ions which are lost by volume recombination is proportional to the product of ion densities of the positive and negative ions at each point. If it is assumed that the mobility does not change with time and is the same for both ions with charge of different polarity, viz. $K_+ = K_- = K$, the mean-lifetime T of ions in a parallel plate ionization chamber is the same for both ions and is expressed as

$$T = \frac{\int_0^d \frac{dx}{2KU} \alpha N_+(x) N_-(x) dx}{\int_0^d \alpha N_+(x) N_-(x) dx} = \frac{d^2}{4KU}$$

where $N_+(x)$ and $N_-(x)$ respectively represent densities of positive and negative ions at a position x in the parallel plate ionization chamber. They are assumed to be linear functions of x ; i.e., they are not affected by recombination loss.

The average drifting time of ions which pass through a position of radius r from the inner side toward the outer direction, in a cylindrical ionization chamber, is $\ln(a/b)(r^2 - b^2) / (4KU^c)$. The mean-lifetime, T_+^c , of ions which drift toward outside and are lost by volume recombination in a cylindrical ionization chamber to which is applied a voltage U^c , is derived by the following equation:

$$T_+^c = \frac{\int_b^a \frac{\ln(a/b)(r^2 - b^2)}{4KU^c} \alpha N_+^c(r) N_-^c(r) dr}{\int_b^a \alpha N_+^c(r) N_-^c(r) dr} = \frac{\ln(a/b)(4b^7 - 28a^2b^5 + 35a^3b^4 - 14a^5b^2 + 3a^7)}{4KU^c(7a^5 - 35a^3b^2 + 35a^2b^3 - 7b^5)}$$

The equation for mean-lifetime of negative ions, T_-^c , has a similar formula, but a and b in the right hand parenthesis of the numerator are replaced. As the values of T_+^c and T_-^c are not the same, their average value, $T^c = (T_+^c + T_-^c) / 2$, was used for the comparison between the mean-lifetime of ions in a cylindrical ionization chamber and that in parallel plate ionization chambers; from the assumption, $T^c = T$, we get

$$\frac{\ln(a/b)[(a^7 + b^7) - 6(a^5b^2 + a^2b^5) + 5(a^4b^3 + a^3b^4)]}{(2a^5 - 10a^3b^2 + 10a^2b^3 - 2b^5)U^c} = \frac{d^2}{U}$$

The m value for a cylindrical ionization chamber to which is applied a voltage of U^c should be the same as that for parallel plate ionization chambers which is applied a voltage, d^2 / U , which satisfies the above equation.

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