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SOME EXPERIMENTAL RESULTS ELABORATING THE EQUILIBRIUM CONDITIONS OF SMALL IONS IN THE ATMOSPHERE OVER THE PACIFIC OCEAN

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Abstract

This paper presents some results of measurements of small ion density and concentration of condensation nuclei over the Pacific Ocean. The results obtained relatively near shore seem capable of being explained in terms of the simplified equilibrium equation which takes into account only the attachment of small ions to condensation nuclei. At greater distances, both the processes of recombination of small ions and attachment of small ions to condensation nuclei to be considered. The difference in the value of the attachment coefficient estimated from the near and distant measurements seems to be consistent qualitatively with the theory of coagulation which predicts an increase in the particle radius with time.

1. Introduction

Compared with the measurements of electric elements in the atmosphere on the land, those seeking to clarify the interaction between ionization, small ions and condensation nuclei over the oceans are relatively few (Parkinson and Weller, 1953, and Sagalyn, 1958).

Some measurements of condensation nuclei in the atmosphere over the Atlantic and North Atlantic Oceans have been made by Hess (1951), O'Connor et al. (1961), O'Connor (1966) and Hogan et al. (1967). Recently, Junge (1969) has made a systematic study of aerosols and has described three types of atmospheric aerosols in Pacific air masses of middle and lower latitudes.

In principle, combined measurements of electric elements and condensation nuclei in the atmosphere over the oceans could lead to a better understanding of the interaction between ionization, small ions and condensation nuclei. The sea surface measurements have the advantage of being amenable to a simpler interpretation primarily because only the ion pairs produced by cosmic rays needs to be taken into account, thus eliminating the necessity of considering the contribution of the radioactive elements in the earth's crust.

This paper presents some results of measurements of small ion density and condensation nuclei over the Pacific Ocean and discusses briefly the inter-relationship between the two parameters.

2. Collection of Data

The measurements were made by installing suitable instruments on the research vessel "Hakuho-Maru" of the University of Tokyo. The expedition (code number KH-70-1) was carried out during the period of February 3 to March 5, 1970 and covered the latitudes of 10°N to 40°N and the longitudes of 140°E to 160°E.

The small ion density was measured by means of ordinary aspiration-type Gerdien condensers. The basic design of the condensers is the same as that described by Ishikawa et al. (1969), except for some minor changes in certain dimensions. The applied voltages to the condensers were so arranged that for the rate of airflow of 300 liters per minutes, the condensers would be saturated for all ions of mobility greater than 0.4 cm^2/volt sec.

The condensation nuclei were measured by means of the Pollak-type counter. The counter was used with an overpressure of 16 cm Hg (adiabatic expansion of 1.2 to 1.0 atm). The corresponding supersaturation in the cylinder was calculated to be 3.2 at 27°C, and thus, the measurements refer to condensation nuclei of radii larger than about 0.90 $m\mu$.

Since wind measurements were made continuously, it is reasonably certain that the data chosen for the analysis were not contaminated directly by the ship's exhaust.

3. Some Results and Discussion

In Fig. 1 are shown the measurements of nucleus concentration in the neighborhood of Tokyo. The minimum distance from the shore is indicated in the diagram. The nucleus concentration tends to decrease with increase in distance. Similar results showing the dependence of nucleus concentration on distance from shore have been reported by other workers (Sagalyn, 1958, and Hogan et al., 1967).

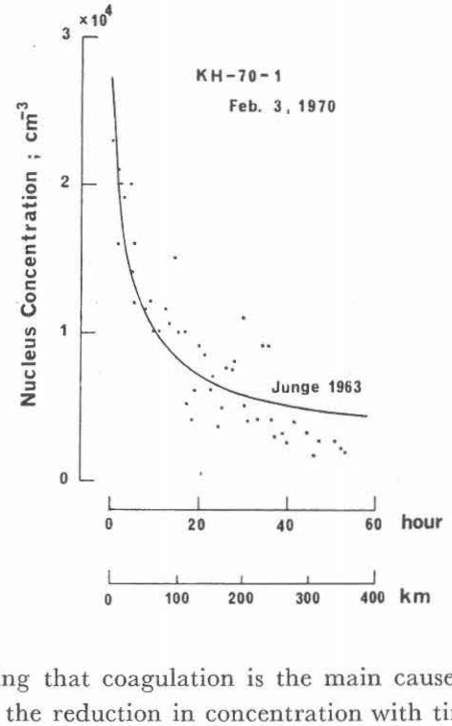

Fig. 1. Showing the measurements of nucleus concentration in the neighborhood of Tokyo. The distance shown is the minimum distance from shore. For the details, see text.

Assuming that coagulation is the main cause of this decrease of the nucleus concentration, the reduction in concentration with time can be computed from the theory of coagulation for a given aerosol distribution. The smooth curve shown in Fig. 1 was obtained from Junge's (1963) calculations which assume an initial concentration of 27000 particles per cm^3 and an initial average radius of 32 $m\mu$. The time scale shown in the figure is so chosen that the theoretical curves fit reasonably well the experimental results. Though the agreement between the computed and measured results is reasonable for distances below 130 km, there is a tendency for the experimental points to lie systematically below the computed curve for greater distances. This is probably due to the presence of two or more air masses with different origins and histories. Though it is evidently rather difficult to calculate the simultaneous influence of a number of air masses, it can perhaps be said that the decrease in the

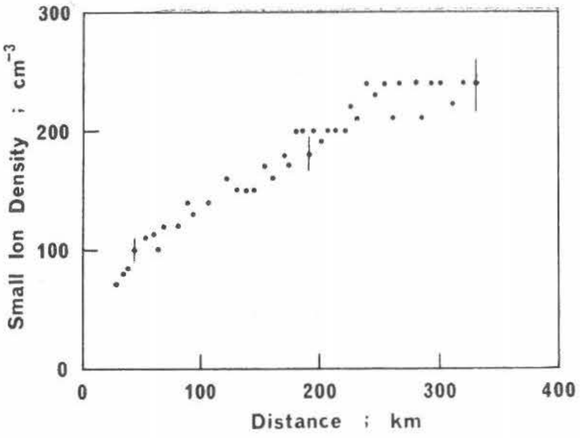

Fig. 2. Showing the variation of small ion density with distance. The measurements refer to the neighborhood of Tokyo. The bars indicate the probable errors in measured parameters.

nucleus concentration with distance shown in Fig. 1, is possibly due to the coagulation processes.

The small ion density measured simultaneously with the nucleus cencentration is shown in Fig. 2. The abscissa is the same as that in Fig. 1. The errors in measured quantities are shown at three selected points. The increase in small ion density with distance shown in Fig. 2 can easily be related to the corresponding decrease in nucleus concentration (Fig. 1) through the well-known equilibrium equation for small ions:

$$q = \alpha n^2 + \beta n Z \quad \cdots\cdots (1)$$

where,
q = the rate of ion-pair production
α = the recombination coefficient for small ions
n = the small ion density (assumed equal for positive and negative ions)
β = the effective attachment coefficient between small ions and condensation nuclei
Z = the nucleus concentration

Under conditions when $Z \gg (4\alpha q)^{1/2}/\beta$, we can write

$$q = \beta n Z \quad \cdots\cdots (2)$$

Thus, when the nucleus concentration Z is relatively high, we would expect a linear

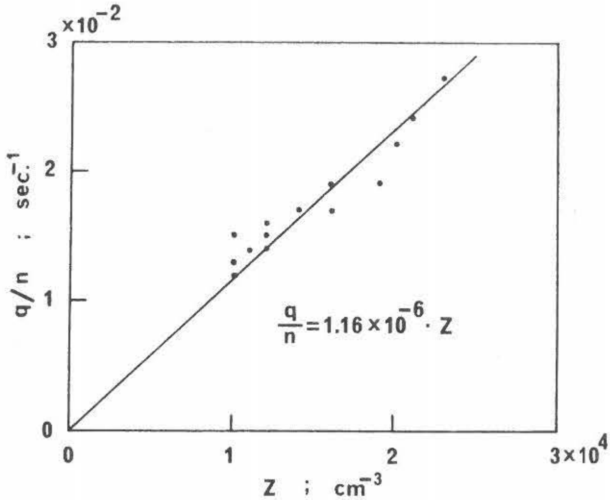

Fig. 3. Showing the relation between q/n and Z. The estimated value of β is equal to 1.16 $\times$ 10^{-6} cm^6/sec.

relationship between q/n and Z with the slope representing β. The scatter plot of q/n versus Z for all measurements within a distance of 130 km is shown in Fig. 3. Following Parkinson and Weller (1953), we have assumed q = 1.9 ion-pairs per cm^3 per sec. The straight line shown in Fig. 3 is the linear least-square fit between the two parameters. The straight line is represented by the equation:

$$\frac{q}{n} = 1.16 \times 10^{-6} Z \quad \cdots\cdots \quad (2a)$$

Kawano et al. (1970) have reported that, over land, their measurments of q, n and Z could be interpreted by the simple relation (2). Thus it seems probable that for distances about 100 km from shore, the equilibrium conditions governing the relation between the small ion density and condensation nuclei, are the same as those over land.

Of course, under the circumstances when the condition $Z \gg (4\alpha q)^{1/2} / \beta$ is not valid, Eq. (1) needs to be combletely satisfied. In Fig. 4 we show the measurements carried out at distances of about 1000 km or more from the shore. Again q is assumed to be 1.9 ion pairs per cm^3 per sec. The data points in Fig. 4 indicate the half-hourly averages of the parameters. The straight line is the linear least-square fit and is given by

$$\frac{q}{n^2} = 3.76 \times 10^{-6} + 4.11 \times 10^{-6} \frac{Z}{n} \quad \cdots\cdots \quad (1a)$$

This value of α is greater than that expected from the Thomson theory by a factor

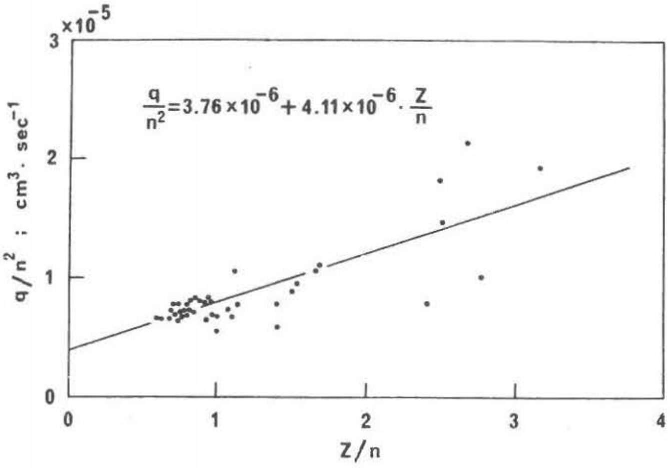

Fig. 4. Showing the relation between q/n² and Z/n. The estimated values of α and β are equal to 3.76×10^{-6} cm³/sec. and 4.11×10^{-3} cm³/sec.

of about 2. Similarly, the value of β in (1a) is greater than that obtained for near distances (Eq. 2a) by a factor of about 4. Since coagulation results in an increase of the radius of the condensation nuclei, an increase in β with distance seem physically justified, provided we assume that air masses originating on the continents could, under favorable circumstances, travel great distances. In fact, Sagalyn (1958) reported, from measurements in the exchange layer, that the average radius of the pollution particles is about four times greater than that observed over land. The model distribution of Junge (1963) also predicts that the average radius of the aerosols could increase by a factor of about 3, due to coagulation effects. Thus our present results seem to be qualitatively in confirmity with the theoretical model of Junge (1963) and with the measurements of Sagalyn (1958).

	Altitude	Nucleus concentation ; cm^{-3}		
		Average	Minimum	Maxmum
Hakuho KH-70-1	Surface	650	280	1200
Carnegie VII	Surface	810	300	1700
Aircraft	500 ft	1150	340	2720

Table 1. Comparison of the various estimates of the nucleus concentration. The aircraft measurements and the Carnegie results are taken from Sagalyn (1958).

Finally, the average, the maximum and the minimum values of the nucleus concentration at about 5 meters above the sea surface are shown in Table 1. It is believed that these values correspond to open sea surface conditions (distance greater than 1000 km) and are determined from the measurements spread over five days of ideally fair weather. We were not able to detect any regular daily variation either in the small ion density or in the nucleus concentration. For comparison, the values obtained at 500 ft above the Atlantic Ocean and the Carnegie results (Sagalyn, 1958) are also shown in the table.

It may be mentioned that the average values of positive and negative small ion density over the mid-ocean in fair weather have been found to be 470 cm^{-3} and 430 cm^{-3}, respectively. These values are greater than those reported by Mühleisen (1967) from measurements on the "Meteor" by a factor of about 1.6 to 2.0. This discrepancy may have been due to the difference in background pollution in the atmosphere over the Atlantic and the Pacific Ocean.

4. Concluding Remarks

Our measurements in the Pacific Ocean show that the values of the recombination and the attachment coefficients are 3.76×10^{-6} cm^3/sec. and 4.11×10^{-6} cm^3/sec., respectively, when the distance from the shore is about 1000 km or more. When the distance is relatively smaller, in the order of 100 km, the value of the attachment coefficiont is seemingly reduced by a factor of about 4.

All our results are in reasonable agreement, at least qualitatively, with existing theories of coagulation and ionization equilibrium.

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