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STUDIES ON THE TEMPORAL AND SPATIAL VARIATIONS OF ATMOSPHERIC ELECTRIC PARAMETERS AND THEIR RELATION TO ATMOSPHERIC POLLUTION

A DISSERTATION SUBMITTED
FOR THE DEGREE OF
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THE RESEARCH INSTITUTE OF ATMOSPHERICS
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CONTENTS

PREFACE

This dissertation describes the author's attempt to measure and interpret some of the important characteristics of electrical state of the atmosphere. Even though some of the basic features of the phenomena known popularly as "Atmospheric Electricity" are understood in a general way, many significant aspects are yet explored systematically. Since the electrical state of the atmosphere is influenced by a bewildering range of other geophysical and meteorological phenomena, it is desirable to plan one's experiments with a view to understand certain specific features. This pragmatic approach is essential not only to achieve one's goals in a reasonable amount of time but to avoid confusion which is bound to creep in when one is confronted with a variety of apparently unrelated data-inputs. It is for this reason that the author has narrowed down the choice of objectives to two specific problems. These are : (1) How do the atmospheric electric parameters vary with height ? and (2) How do they vary when one moves from land to mid-ocean ? Essentially, this presentation contains the results of balloon and ship-borne experiments and their interpretation. For convenience, it is devided into three chapters, the contents of which are briefly summarized below.

Chapter I is written as a kind of introduction in which some basic concepts like the 'Global circuit of atmospheric electricity' 'Small ion equilibrium conditions', etc. are described. This chapter also serves as a general background against which the contents of the later chapters could be viewed in their proper perspective. Thus, it sets for forth the rationale for systematic measurements of parameters like the small ion density and electric

conductivity with the air-borne and ship-borne instrumentation.

Chapter II deals mainly with the results obtained through instruments flown on balloons. The basic design considerations and their influence on the accuracy of measurements are also discussed in brief. The data on the small ion density and conductivity, obtained as a function of height, are described in detail and are discussed in the light of the presently accepted theoretical concepts. Also included are the results of a single constant-level balloon flight at stratospheric levels. As the end of the chapter, the relevant theoretical investigations by the author are discussed briefly.

Chapter III constitutes the final part of this dissertation and is concerned with the ship-borne experiments. It describes the author's measurements on the atmospheric electric parameters and the nucleus concentration and discusses in detail how these parameters depend on the distance from shore, on the air stream patterns and on the time-histories of the air masses. The dominant thema in the discussions is, of course, the land pollution, its extent and its influence on the measured parameters. Some measurements carried out in "mid-ocean" have been interpreted in terms of the global circuit. Brief discussion on the "global background level of pollution" is also included. The chapter ends with a comment on the self-consistency of the data collected during different expeditions at varying distances from shore.

Because of the very purpose of the dissertation, the author's contributions are dealt with in detail. At the end of the presentation a list of publications bearing author's contributions is appended.

CHAPTER I

GENERAL INTRODUCTION

1.0. The General Background :

The atmosphere of the earth is not a perfect insulator but has a finite conductivity which shows marked dependence on height. Originally it was thought that the conductivity of the atmosphere was solely due to the ionizing action of the Alpha-, Beta- and Gamma-radiations emitted by the radioactive substances embedded in the earth's crust. If this were true, then one would expect the conductivity of the air to decrease with height. Experiments were made to test this hypothesis by balloon borne instruments. The most historic experiment was made by Hess in 1911 who found that the conductivity at certain heights increased markedly, in sharp contrast to the hypothesis that the ionization was solely due to natural radioactivity. The experiment of Hess marked the beginning of an entirely new and exciting branch of physics called the "Cosmic Ray Physics". Today as the state of affairs stands, the progress and the sophistry achieved in the domain of Cosmic Ray Physics outweigh, by several orders of magnitude, the corresponding accomplishments in the study of Atmospheric Electricity. Yet, the progress achieved by the scientists devoted to the study of Atmospheric Electricity, say, during the last four or five decades has been of such proportions as to justify the division of the subject into many equally important narrow channels of specialization.

A mile-stone in the development of the subject was the introduction of the concept, known as "Global Circuit of Atmospheric Elelctricity" originally conceived by C.T.R.Wilson in 1920's. This concept achieved a unique synthesis of thought and

today we speak freely of the "Generation" of electricity in the disturbed weather through the mechanism of charge-separation inside the thunderclouds and "Consumption" of power in the fair weather areas where the conduction current flows incessantly through the finite resistance of the atmosphere. We now briefly discuss this concept of "Global circuit" in very general terms.

1.1. The Ideal Global Circuit of Atmospheric Electricity :

In an ideal fine-weather (i.e.,when there is no precipitation, no overcasting of the sky, no dusty winds and when the visibility is high and when anthropogenous influences are negligible), there is, in the atmosphere, a vertical electric field which is directed downwards and which has a magnitude of the order of 100 V/m near the ground. This field decreases in magnitude as we go up, almost in an exponential manner. As already mentioned the atmosphere is made conducting by the ionizing influences of radioactive material present in the earth's crust and in the atmosphere and by the cosmic rays. To a first order approximation, the conductivity increases with height almost exponentially. The increasing conductivity with altitude implies that at some, as yet undetermined, height the conductivity is so high that the atmosphere there can be considered as an equipotential surface. The layer of atmosphere at this height, and the earth, which is also an equipotential surface, together can be conceived as two "electrodes" of a giant spherical condenser. The atmospheric filling this condenser system acts as a "leaky dielectric". The leakage is due to the finite conductivity of the atmosphere.

Again to a first order apporoximation, the fair weather current, which can be represented as the product of the field and conductivity, is sensibly constant with altitude. The magnitude of the current density is the order of 10^{-12}-10^{-14} Amp/m^2. In fact it is this constancy of the current density which permits association to be made between the electric field and the conductivity of the air, which are two quantities of different origin. This current is due to the flow of positive charges downwards (or negative charges upwards). The integrated current over the whole globe is in the order of 10^3-$2x10^3$ Amp (for an assumed value of $3x10^{-12}$ Amp/m^2 for the current density the integrated current is $\sim 1.5x10^3$ Amp).

One of the apparently baffling situations which confronted the specialists in the atmospheric electricity, before the concept of Global circuit was introduced, was this : Since the fair weather field is directed downward, there will be negative charges induced on the surface of the earth which is a good conductor. For an assumed value of the fair weather field of 100 V/m, the net negative charge induced over the whole surface of the earth is about $4.5x10^5$ Coloumbs. The fair weather current which is flowing downwards will neutralize the induced charges in a time of about Q/I where Q is the net charge on the earth and I is the net current. For I = $1.5x10^3$ Amp, and Q = $4.5x10^5$ Col., this time is five minutes. Thus the induced charge on the earth and the field which induces this charge, should disappear within a few minutes. But we find that the field is always present. What maintains the fair weather field ? The same question can be framed in a slightly different fashion.

The fair weather field which various with height can be integrated to yield the difference between a specified height and the ground. For heights greater than about 30 km, the potential (with respect to earth) is in the order of 300 KV. (Some authors denote this quantity as the potential of the ionosphere). Then the question is : what mechanism maintains the potential of the ionosphere ?

The concept of Global circuit has been conceived with a view to answer these questions. In this concept, the thunderstorms occurring anywhere in the world pump currents into the ionosphere (more logically, the "equalizing layer") which being a good conductor distributes them all over the Globe. Finally, these currents find their way to ground through the fair weather areas, producing 'fair weather' fields whose magnitude is determined by the "resistance" offered by the local atmosphere. By establishing a link between the fair weather electric parameters and the thunderstorm activity in the disturbed areas, this concept of Global circuit seeks to explain the variations in the former in terms of the corresponding variations in the latter. Thus, for example, the diurnal variations observed in the electric field (in an ideally fair weather) are attributed to the diurnal variations of the thunderstorm activity integrated over all the disturbed areas.

1.2. The Need to Study Deviations from the Global Concept :

The essential correctness of the above hypothesis (i.e. the Global circuit) has been verified, uptill now, by the mean results of a few polar expeditions and the cruises of the Carnegie

Institution. Global circuit has been elaborated by many workers (Brookes,1925, Gish,1944,1951, Gish and Wait,1950, Holzer and Saxon,1952, Dolezalek,1958,1964,1971, Paltridge,1967, Bhartendu, 1971, Koenigsfeld,1971, and others). Though there is little doubt the Global circuit is valid in its essentials, it is very difficult and sometimes almost impossible to detect the Global influences in the data collected near the ground on the continents This is because most, if not all, of the basic parameters controlling the electrical state of the atmosphere are critically dependent on the meteorological and micrometeorological factors. Further, they are influenced markedly by anthropogenous factors. In short, the electrical state of the atmosphere, especially below the "exchange layer", is controlled by all the factors which go into the definition of the word "environment". In a sense, this environmental control is the most challenging problem and to arrive at any possible solution it is essential to study some of the basic processes. A few examples of such basically vital processes are :

(1) The generation and destruction mechanism of small ions, and how these mechanism depend on height.

(2) The influence of natural and artificial (i.e. pollution) aerosols on the delicate ionic balance.

(3) The height dependence of aerosol content.

(4) The influence of meteorological and micrometeorological factors.

(5) The influence of trace elements like O_3, H_2O etc.

(6) The possible influences of any phenomena of extra terrestrial origin.

(7) The physics of charge separation (i.e. thunderstorm electricity) etc.

Evidently, the above mentioned phenomena are too numerous and too wide ranging to be included in this dissertation in a detailed fashion. Thus, we shall, in the following sections discuss only those phenomena which are of immediate relevance to the measurements reported in the latter chapters.

1.3. Ionic Balance in the Troposphere and Stratosphere :

As already mentioned, the fair-weather ionization is produced by the cosmic rays and by the radiations from radioactive substance in the atmosphere and in the earth's crust. Over oceans, polar ice caps and at altitudes above a few kilometers over land the ionization results primary from cosmic rays. Above the first few kilometers, intensity of cosmic rays increases with altitude. Near the ground, the ionization results mainly from Alpha-, Beta- and Gamma-radiations from ground radioactivity which depend strongly on height ; Alpha-radiation is contained in the first few centimeters, Beta-radiation in the first few meters and Gamma-radiation in the first few kilometers. Radon and Thoron gases emitted from the earth's surface also play significant role in the electrification of the lower atmosphere. Bricard (1965) and Ikebe (1970) have evaluated the ionization due to these components and have determined the vertical profiles of each ionization component of the radiations in the atmosphere.

The ions initially formed by the ionizing radiations are "small ions", which disappear by direct recombination between

the ions and by attachment to particles of greater size (aerosols) suspended in the atmosphere. The electric charge of the ions is transfferred to the aerosols and thus "large ions" appear in the atmosphere. The aerosol content and its temporal and spatial variations are as important as the corresponding variations in the ionizing intensity in any study of the ionic processes in the lower region of the atmosphere. With increasing altitude, up to stratosphere, the intensity of ionization increases, while the number density of aerosols decreases rapidly. Thus it is to be expected that the conditions of ionization equilibrium existing at different altitude, up to the stratosphere, show marked variabilty.

In the lower layers of the troposphere, aerosols are usually present in good numbers and hence considerably influence the ionic balance in the atmosphere. The number density of aerosols denoted by Z can be expressed as follows (Bricard,1965):

$$Z = N_0 + \sum_p N_1^{(p)} + \sum_p N_2^{(p)} \qquad (1)$$

where N_0= the number density of uncharged aerosol

$N_1^{(p)}$ and $N_2^{(p)}$= the number densities of positively and negatively charged aerosols with p-elementary charges.

The change in the number density of positive small ions is given by,

$$\frac{dn_1}{dt} = q_1 - \alpha n_1 n_2 - \eta_0 n_1 N_0 - n_1 \sum_p \eta_{12}^{(p)} N_2^{(p)} - n_1 \sum_p \eta_{11}^{(p)} N_1^{(p)} \qquad (2)$$

where q_1= the rate of positive small ion production

n_1 and n_2 = the number densities of positive and negative small ions respectively

α = the recombination coefficient between positive and negative small ions

$\eta_{ij}^{(p)}$ = the attachment coefficients ; i denotes the polarity of large ion

$\eta_{10} n_1 N_0$ = the rate of positive small ions diffusing to uncharged aerosols.

Similarly for the negative small ions :

$$\frac{dn_2}{dt} = q_2 - \alpha n_1 n_2 - \eta_{20} n_2 N_0 - n_2 \sum_p \eta_{21}^{(p)} N_1^{(p)} - n_2 \sum_p \eta_{22}^{(p)} N_2^{(p)} \qquad (3)$$

For large ions with single elementary charge :

$$\frac{dN_1^{(1)}}{dt} = Q_1^{(1)} + \eta_{10} n_1 N_0 + \eta_{21}^{(2)} n_2 N_1^{(2)} - \eta_{21}^{(1)} n_2 N_1^{(1)} - \eta_{11}^{(1)} n_1 N_1^{(1)} \qquad (4)$$

$$\frac{dN_2^{(1)}}{dt} = Q_2^{(1)} + \eta_{20} n_2 N_0 + \eta_{12}^{(2)} n_1 N_2^{(2)} - \eta_{12}^{(1)} n_1 N_2^{(1)} - \eta_{22}^{(1)} n_2 N_2^{(1)} \qquad (5)$$

where $Q_i^{(1)}$ = the rate which the large ions with an elementary charge are formed directly.

The direct formation of large ions occurs in combustion processes in industrial areas, and naturally in frictional charging of blowing dust, and the bubbling and spray electrification associated with surf and white cap activity of lakes and oceans. Usually the ionizing action results solely from radiation, so that we may write :

$$q_1 = q_2 = q \quad \text{and} \quad Q_i^{(1)} = 0 \qquad (6)$$

The multiple charging occurs only by diffusion of an ion

to a similarly charged aerosol. This process is inhibited by the Coulomb fields and thus the number of multiply charged aerosols can be neglected when the number density of aerosols is small. With these two assumptions, the steady state conditions can be expressed as :

$$\left.\begin{aligned} q &= \alpha n_1 n_2 + \eta_{10} n_1 N_0 + \eta_{12} n_1 N_2 \\ q &= \alpha n_1 n_2 + \eta_{20} n_2 N_0 + \eta_{21} n_2 N_1 \end{aligned}\right\} \qquad (7)$$

$$\left.\begin{aligned} \eta_{10} n_1 N_0 &= \eta_{21} n_2 N_1 \\ \eta_{20} n_2 N_0 &= \eta_{12} n_1 N_2 \end{aligned}\right\} \qquad (8)$$

$$Z = N_0 + N_1 + N_2 \qquad (9)$$

In the above treatment we have considered only a single type of aerosols. In the real atmosphere, however, aerosols of different sizes and compositions exist and the equations could, in principle, be generalized to include other types of aerosols. Such a generalized treatment is, however, beyond the scope of the present investigation.

In general, we may expect nearly equal numbers of small positive and negative ions, and assuming equal probability of attachment for both the types, equations (7) and (8) can be written as :

$$\left.\begin{aligned} q &= \alpha n^2 + \eta_0 n N_0 + \eta n N \\ \eta_0 n N_0 &= \eta n N \end{aligned}\right\} \qquad (10)$$

A much simpler formula is as follows (Nolan,1956, Keefe and Nolan,

1962) :

$$q = \alpha n^2 + \beta n Z \qquad (11)$$

where $\beta = \frac{2\zeta\eta_0}{\zeta+2}$ and $\zeta = \frac{\eta}{\eta_0}$

It is generally more convenient and more realistic, in comparing theory and experiment, to view the β's as the effective attachment coefficients associated with the average aerosols. Under conditions when $Z >> (4\alpha q)^{1/2}/\beta$, we can write,

$$q = \beta n Z \qquad (12)$$

Thus, when the number density of aerosols Z is relatively high, we can neglect the term of αn^2 in equation (11). Ikebe (1970) has studied the ionization equilibrium in the atmosphere near the ground surface and over the sea, and has found that the measured values of the parameters q, n and Z were well expressed by equation (12). His measurements were, however, limited to the regions of relatively high aerosol concentration. We do not, therefore, expect equation (12) to be valid for measurements carried out in the regions of relatively clean atmosphere.

At stratospheric heights, the ionization is mainly due to cosmic rays, and we can neglect other processes like photonization, bremsstrahlung ionization and collisional ionization, etc. It is believed that the aerosol number density affecting the ionic balance is negligible and that the conditions of small ion equilibrium prevail in the stratosphere. We can thus write from equation (11),

$$q = \alpha n^2 \qquad (13)$$

Equation (13) represents the ion equilibrium of a "clean" or "aerosol-free" atmosphere. Solutions of equation (13) have been obtained by many workers, e.g. Sagalyn (1965), using the ionization data of Bowen et al. (1938) and Loeb (1960) in the formulation of Thomson theory of volume recombination. There is, however, quite often a wide disparity between experimental results and the predictions of equation (13), even for measurements under supposedly clean atmospheric conditions. This has led to a re-examination of the method of evaluating the recombination coefficient α at various altitudes (Hoppel,1969) and the possible existence of stratospheric aerosols and/or other effects controlling the small ion equilibrium in the stratosphere have been suggested (Whipple,1965, and Morita and Ishikawa,1969a,1970a).

1.4. The Nature of Tropospheric and Stratospheric Ions :

The nature of atmospheric ion has been investigated by several scientists (Langevin,1905, Pollock,1915, Wright,1936, Hogg,1939, and others). The general results of these investigations have led to the discovery of three groups of atmospheric ions ; small, intermediate and large ions. These groups of ions are classified in terms of their "mobility", which is critically dependent on the size and mass of the ions. The wide range of mobilities of atmospheric ions results from the wide range in the sizes of the atmospheric ions, the actual value of being dependent upon the humidity, pressure, the presence of other substances, the age and probably on other factors as well. Under similar conditions it is found that negative ions have

rather greater mobilities than positive ions. No single theory is valid for the entire range of mobilities. For ions which are small enough so that their motion induces no mass motion, the application of kinetic theory is justified. For ions of larger radius, (i.e.,large ion), the motion may induce mass motion and hydrodynamic effects must be included. Considerations of kinetic theory have been described in detail by Loeb (1960). Junge (1963) has derived the analytical expressions for the basic parameters ; fall velocity, diffusion coefficient,mobility and mass. Recently, Hoppel (1968) has reviewed the mobility theory for size ranges of small, intermediate and large ions. The mobility ranges for small, intermediate and large ions are shown in Figure 1.

Attempts to predict the composition of atmospheric ions have been made in recent years. One of the method is to determine the mobility of pure gases by using mass spectrometry in the laboratory, while the other is to measure directly the mobility distribution of atmospheric ions. Many laboratory experiments to determine the mobility of pure gases (such as, Hydrogen, Nitrogen and Oxygen, etc.) have been performed (Loeb, 1960). Especially, the nature of N_2 and O_2 gases has been thoroughly investigated in the laboratory because they are the major constituents of the atmosphere. The results obtained from mass spectrometry may not, however, be valid when applied to atmospheric ions because of extremely short lives, low pressures, and high accelating voltages. Further, there is a possibility that the ion is altered as it passes through the

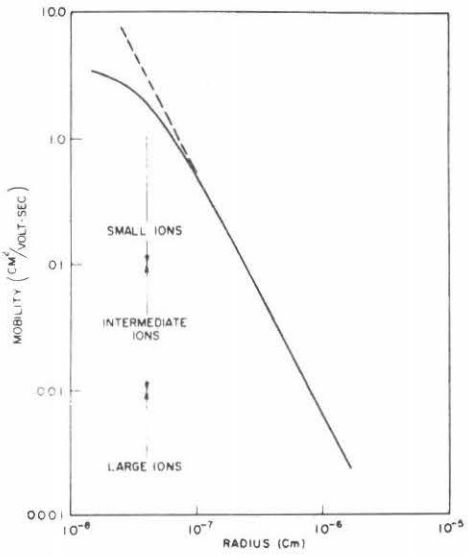

Figure 1. Relation between size and mobility of atmospheric ions (Hoppel,1968).

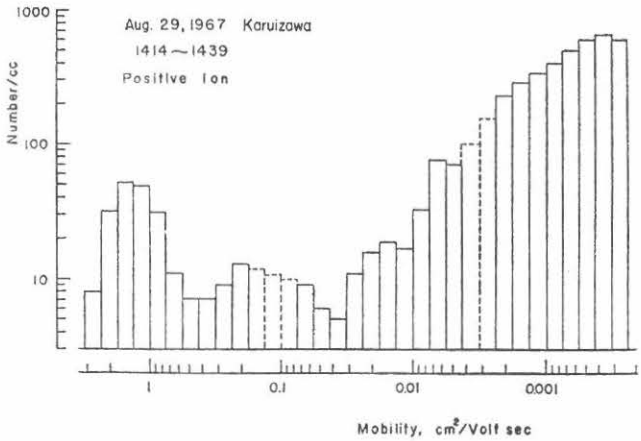

Figure 2. An example of dynamic spectrum of atmospheric ions (Misaki,1970).

focussing slits. The standard method of measuring ionic mobility in the laboratory is not very useful in making tropospheric and stratospheric measurements.

Thus the problem is to the measure mobilities of ions which result from very weak ionizing intensities and have "ages" in the order of several tens of seconds. This is the main reason why only a few investigators have measured the mobility distribution of atmospheric ions (Misaki,1961,1964, and Hoppel and Kraakevik,1965). The results of measurements of mobility distribution in the troposphere show that the atmospheric ions are not confined to a single mobility but are more accurately described by a mobility distribution extending roughly between 0.5×10^{-4} and 3.0×10^{-4} m^2/volt.sec., with a peak around 1.0×10^{-4} m^2/volt.sec., in the small ion region. Recently, Misaki and Kanazawa (1969) have obtained more accurate dynamic spectra of tropospheric ions throughout the mobility range 4.22×10^{-4}-$0.00042 \times 10^{-4} m^2$/volt.sec. by means of three simultaneously operated spectrometers. They have found that the mode of distribution,i.e., the value of mobility at which the maximum in the distribution appears, is independent of the total concentration of small ions. An example of the dynamic spectrum is shown in Figure 2. Mohnen (1969) claims that it is possible to predict the composition and formation mechanism of tropospheric ions by a detailed consideration of charge exchange reaction rates. Thus, most of the ions initially formed would, in the Standard Atmosphere, charge exchange with O_2 and become O_2^+ in less than 10^{-8} seconds. Beyond this

point the reactions and reaction rates are only partially known. However, it is known experimentally that under tropospheric conditions the oxonium ion H_3O^+ does exist, while $[NO(H_2O)_n]^+$ and $[NO_2(H_2O)_n]^+$ probably do not. Since the reactions leading to latter ions are known, certain suppositions regarding the reactions leading to H_3O^+ can be made. H_3O^+ clusters very quickly to form $H_3O^+(H_2O)_n$, where n can be as larger as five or six. Sixna (1969) has also investigated the nature of atmospheric ions and has obtained the composition of hydronium ion, which is depicted in Figure 3.

The study of the nature of large ions has led to that of submicron aerosols, which deserve our special attention because of their high concentrations and of their pronounced dependence on the geographic location and local weather. One of the most important problems is to establish the size distribution of submicron aerosols. This distribution could, in principle, lead to a better understanding of the processes of attachment between aerosols (charged or neutral) and small ions. Several investigators have discussed theoretically the mechanism of attachment (Bricard,1965, and others). Experimental estimates of the attachment coefficient have usually been based on the gross counting of aerosol (e.g., Holl and Mühleisen,1955), and the values thus obtained by different workers are not always consistent. Although several methods to determine the size spectrum have been used, the most powerful technique is to measure the ion mobility spectrum. Misaki (1964) has measured the ion mobility spectrum of large

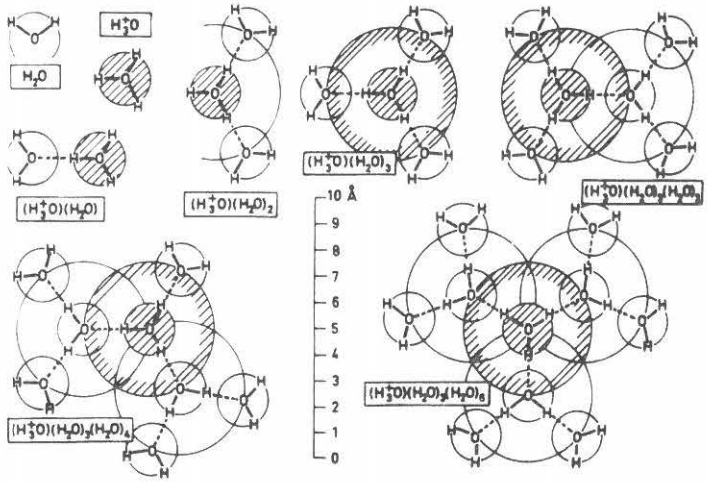

Figure 3. The structure of oxonium and its hydrates-hydronium ions formed in steps (Sixna,1969)

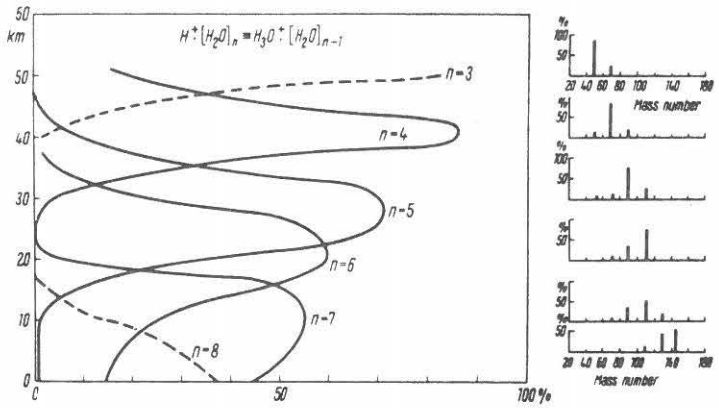

Figure 4. Equilibrium distribution for positive ion clusters $H_3O^+(H_2O)_{n-1} \equiv H^+(H_2O)_n$ as a function of altitude (0-50 km) (Mohnen,1971).

ions and has also estimated the size spectrum of submicron aerosols. Junge (1963) has reviewed the physical properties and the chemical compositions of tropospheric aerosols. Misaki (1960,1966 and 1970) has also summarized the present state of our knowledge of the submicron aerosols. The study of the nature and origin of submicron aerosols is being pursued actively by several workers (e.g.,Mohnen,1971).

The average mobility of atmospheric ions, being inversely proportional to atmospheric density, increases with increasing altitudes while the number density of aerosols and the water vapor pressure decrease rapidly. This behavior implies that the nature of atmospheric ion may change with increasing altitudes. Recent work made by Mohnen (1971) indicates that the most probable positive small ion is hydronium ion, $H^+(H_2O)_n$, with the most likely value of n in the stratosphere being 5 or 6. The degree of hydration depends on the temperature and the amount of water vapor in the atmosphere. The most important negative ion, at stratospheric heights, is $NO_3^-(H_2O)_n$. The equilibrium distribution for positive ion clusters as a function of altitude is shown in Figure 4. However, no direct measurements of mobility spectrum to confirm of the scheme of Mohnen (1971) have been made in the stratosphere. There are only the measurements of the average mobility of small ion calculated from the simultaneous measurements of conductivity and small ion density (Paltridge,1965,Ishikawa et al.,1969, Morita et al., 1971,and Riekert, 1971).

Besides what has been mentioned above, nothing is known

about the nature of stratospheric ions, especially the large and intermediate ions. Though many measurements of stratospheric aerosols have been made directly (Junge et al.,1961, Changnon and Junge,1961, Junge,1961, Rosen,1964,1968, and Bigg et al., 1970, and others) and indirectly (Newkirk and Eddy,1964, Fiocco and Grams,1964, Elterman and Campbell,1964, Dave and Mateer, 1968, Gambling et al.,1971, Bartusek and Gambling,1971, and Shuster,1971, and others),even the mere existence of large and intermediate ions is still unknown. However, the possible role of aerosols in affecting the nature of atmospheric ions and the possible existence of low mobility ions have been suggested by Mohnen (1971). There are various reasons for this suggestion. Firstly, aerosols present at any altitude provide a surface for ion-pair annihilation. Secondly, they can act as condensation nuclei under favorable conditions and thus decrease appreciably the partial pressure of water, which determines the degree of hydration of both positive and negative ions. Thirdly, the aerosols could have been produced originally at any altitude by the positive and negative ions themselves by means of the so-called "gas-to-particle conversion" mechanism. Various atmospheric constituents (ionized or neutral) are transferred from a molecular dispersed state into a colloidal state. This aerosol generation is well known at tropospheric levels and a general survey has been presented by Mohnen and Lodge (1969).

1.5. Temporal and Spatial Variations of Atmospheric Ions :

The electrical parameters in the lower troposphere show significant temporal and spatial variations. For example, the electric conductivity at many places on land, under fair weather conditions, attains a maximum in the dawn hours, and then falls soon after sunrise. This is probably accounted for by the formation of mist or by the increased pollution of the air. As anticipated from the equations of ionic balance, an increase of number of aerosols brings in a corresponding decrease in the small ion density which results in the decrease of conductivity. Thus, we should expect, as is indeed always found, a minimum of conductivity in winter, at places where pollution occurs. Two representative variations of conductivity on land surface are shown in Figures 5(a) and (b).

Over the oceans, on the other hand, the atmosphere is relatively clean so that the variations of the electrical parameters are expected to represent those considered as being typical of the undisturbed atmosphere. Ever since the famous Carnegie cruises during 1915 and 1929, many attempts have been made to measure the electrical properties of the atmosphere over the oceans (Ruttenberg and Holzer,1955, Mühleisen,1967, Buis,1968, Takagi and Kanada,1969,1970, and Morita,1971). Most of these results, if not all, however, indicated that continental pollution and air masses influence the sea surface measurements over long distances from shore. Misaki and Takeuti (1970) have measured the electric field and conductivity over the Pacific Ocean and the Sea of Japan, and their results indicated

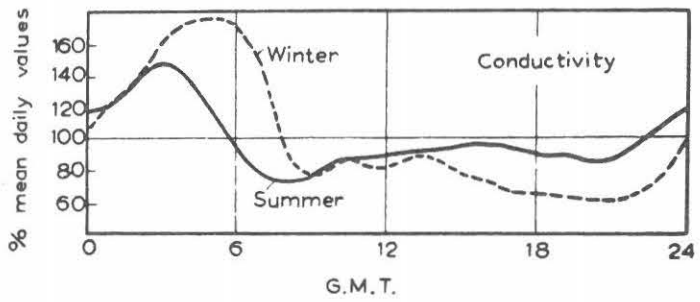

Figure 5(a). Diurnal variation of conductivity at Kew (Chalmers,1957).

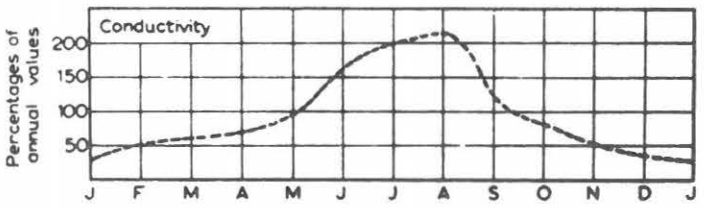

Figure 5(b). Annual variation of conductivity at Kew (Chalmers,1957).

the extension of air pollution from land over the ocean. Cobb and Wells (1970) made measurements of electric conductivity in the global atmosphere over the oceans. The results indicated that the conductivity in the remote South Pacific has remained fairly constant over the past half century but has decreased by at least 20% in the North Atlantic, and the secular conductivity decrease in the North Atlantic is attributed to an increase in the fine-particle aerosol pollution suspended in the atmosphere of the North Heimsphere. Figure 6 shows the secular change in the conductivity from 1907 to 1967 for the two oceanic regions. It thus becomes necessary to study how far the influence of continental pollution continues to exist before one can reasonably assume that a particular set of shipborne measurements represents the atmospheric conditions unaffected by continental pollution. 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, Sagalyn,1958, and Morita et al.,1971). Some measurements of condensation nuclei in the atmosphere over the Pacific, North Atlantic Oceans and the Sea of Japan have been made by Ohta (1950), Hess (1951), O'Connor et al. (1961), O'Connor (1966) and Hogan et al. (1967). 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

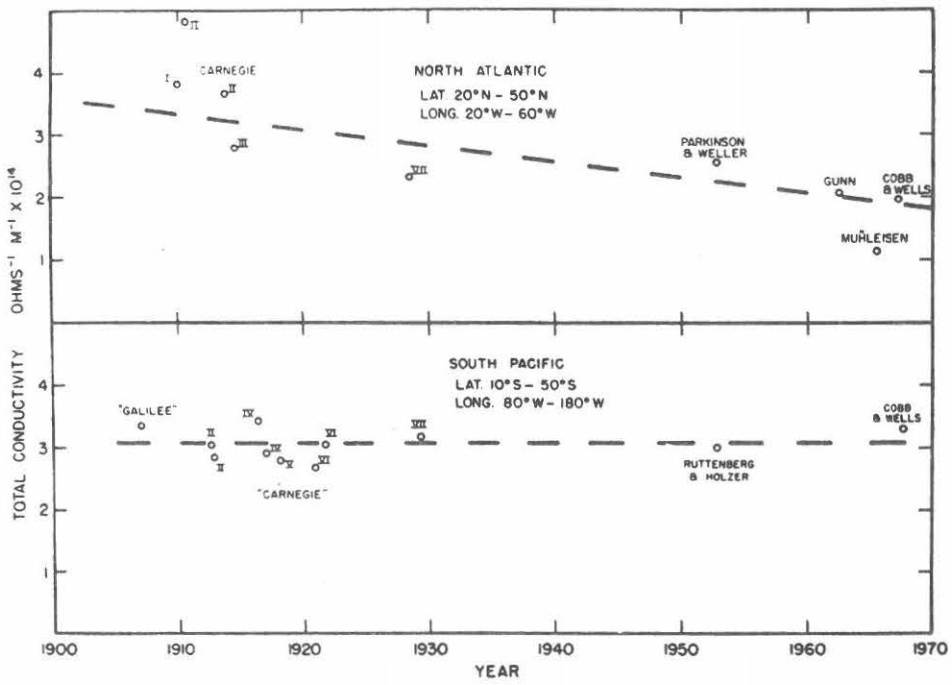

Figure 6. The secular change in atmospheric conductivity from 1907-67 for two oceanic regions. The conductivity in the South Pacific has remained fairly stable ; in the North Atlantic indications are that the conductivity has decreased by 20% in the last 60 years (Cobb and Wells,1970).

to a simpler interpretation primarily because only the ion pairs produced by cosmic rays need to be taken account, thus eliminating the necessity of considering the contribution of radioactive elements in the earth's crust.

The characteristic phenomena in atmospheric electricity are sometimes observed in the region extending from the ground to an altitude of a few kilometers. Continuous mixing of air in the region sometimes causes rapid changes in electric parameters. Measurements show that the thickness of the region (the so-called "exchange layer") may be up to 3 km. There are small-scale convection currents which carry the surface air, together with the ions and aerosols in this region of the atmosphere. Measurements showing such rapid changes of the electric parameters with increasing altitudes near the exchange layer have been reported by many workers (Sagalyn and Faucher,1954, 1956, Clark,1958, Kraakevik,1958b,Koenigsfeld,1958, Curtis and Hyland,1958, and Reiter et al.,1970). For example, Sagalyn and Faucher (1954) made measurements of conductivity, as well as temperature, humidity and large ion content up to an altitude of about 4 km and showed that there was a very marked change in all these parameters on crossing the upper boundary of the exchange layer.

Above the exchange layer the electrical state of the atmosphere is considered to be relatively simple. Coroniti and Heaton (1953) made conductivity measurements in aircraft at various levels up to 6.4 km over the Pacific Ocean and found values of the conductivity agreeing well with those expected from cosmic

ray data. Wide ranging measurements of conductivity up to an altitude of 10 km were carried out by Callaham et al.(1951) on both clear and haze days, with a view to verify the low conductivity values reported by Gish and Sherman (1936). They found a remarkable consistency in their results despite the variability of the conditions under which individual measurements were made. While balloon measurements made by Paltridge (1965) indicated a decrease in the small ion density below theoretical predictions, jet aircraft measurements (Paltridge, 1966a) gave high ion density values which are consistent with insignificant levels of dust in the region studied. Bragin (1967) has obtained the many profiles of ion density of the atmosphere below 90 km and the results suggested that smooth nighttime stratospheric maximum (10-40 km) is sharply cut off during the daytime and that it consists of several secondary maxima. The study of temporal and spatial variations of the ion density in both horizontal and vertical directions in the stratosphere could clarify the behavior of aerosols, the influence of the solar ionizing radiations, etc. Compared with the vertical measurements of ion density in the stratosphere, those seeking to study the temporal and spatial variations at a constant level are, however, relatively few (Paltridge,1966b, and Morita and Ishikawa,1970a).

The results reported by many workers indicate that further and more detailed studies of spatial and temporal variations of the elecriric parameters, especially ion density, in the earth's atmosphere are warranted.

1.6. The Scope of the Present Study :

It is evident from the previous sections that the basic parameters characterising the electrical state of the atmosphere depend very markedly on a bewildering range of natural and artificial phenomena. Even though many attempts to identify these phenomena and to quantitatively estimate their influence on the measurements have been made, the over-all picture today is far from being complete. The measurements so far reported are neither exhaustive (in the sense that they do not constitute even a reasonably representative sampling of the possible situations) nor completely consistent (in the sense that we do not even know whether the disparities in the reported results came necessarily from the difference in the local conditions). Further, the run-away growth of technology has, in recent years, resulted in an unprecedented increase in the atmospheric pollution. The possibility that pollution may even extend upto stratospheric heights is now real (e.g.,the proposed commercialization of SST). Though our understanding of the influence of the various types of pollution on the atmospheric electric parameters is very meagre, the only way to overcome this deficiency is to carry out systematic measurements of the parameters in varied environmental conditions. This is the basic philosophy of the studies reported in this thesis.

The basic idea is to attempt to find out quantitatively, how far has to move in a vertical direction before one could conclude with reasonable certainity that the measured parameters represent a global picture. Similarly, an attempt has also been

made to ascertain the distances in the horizontal from continental landmasses beyond which the global picture is once again 'visible' in the data. Towards this end, relevant data have been obtained by balloon borne equipments at various heights and by shipborne instruments at various distances from the continents. Certainly it cannot be claimed that the present investigations have resulted in a unique determination of the desired results. On the other hand, it can be said that they constitute a systematic set of measurements which, at least in principle, should help better our understanding of the basic processes involved.

CHAPTER II

THE ELECTRICAL STATE OF THE ATMOSPHERE BETWEEN THE GROUND AND THE STRATOSPHERE

2.0. Introduction :

It was emphasized in the previous chapter that the electrical state of the atmosphere is influenced markedly by the meteorological, micrometeorological and anthropogeneous factors. Since these factors, in their turn, depend on the altitudes,it follows that atmospheric electric parameters exhibit considerable height variations. Below the exchange layer considerable mixing of the atmosphere takes place and we should expect this mixing to be reflected in the measurements of the electric parameters in some form of "agitation". For example, the measurements by Sagalyn and Faucher (1954,1956) of the electric parameters show rapid changes with increasing altitude near the exchange layer. Such changes are, under certain conditions, indicative of the existence of the suspended submicron aerosols below the exchange layer. On the other hand, the ionization at stratospheric heights and above is principally due to the cosmic rays and we can reasonably assume the small ion equilibrium conditions to prevail. Thus it can be said that the electrical state of the atmosphere undergoes a kind of transition as we move up from the ground to the stratosphere.

We have also indicated in Chapter I that to understand the complex nature of the dependence of the electrical state of the atmosphere on the height, systematic measurements of electric parameters have to be carried out at as many stations as possible. Two of the important parameters are the small ion density and the conductivity whose measurement is vital to any understanding of the atmospheric electricity.

Many measurements of the small ion density and electric conductivity of the atmosphere to the stratosphere have been made with nearly the same method by different investigators (Gish and Sherman, 1936, Stergis et al., 1955, Kraakevik, 1958a, Woessner et al., 1958, Kroening, 1960, Uchikawa, 1961, Paltridge, 1965, 1966a, 1966b, Takeuti et al., 1966, Ishikawa et al., 1969, and Morita et al., 1971, 1972). There was, however, considerable disagreement between the results reported by different workers. Further, some of the measurements showed significant departures from theoretical predictions. Thus, while the theoretical considerations of many workers (e.g., Cole and Pierce,1965, Morita and Ishikawa,1969a and Shreve,1970) show that the small ion density increases with height to a value of several thousand per cm^3 at about 13 km above which it remains virtually constant, the measurements by Kroening (1960) show extremely low values of negative small ion density at balloon heights. He attributes this low density to the presence of dust particles to which the small ions attach, thereby forming large ions of such low mobilities that lie below the threshold of his apparatus. Similarly, the results reported recently by Bragin et al. (1967) show considerable departure from simple theoretical predictions. Natural and man-made aerosols have been held responsible for some of these results. Whipple (1965), Ishikawa et al., (1969) and Morita and Ishikawa (1969a) have attempted to interpret the discrepancy between the various observations in terms of the possible influence of atmospheric aerosols on the ion density. Suggestions have also been made that the atmospheric

ozone might be responsible for some of the temporal and spatial variations of small ion density in the stratosphere (Morita and Ishikawa,1970a).

The objective of this chapter is to describe the author's attempt to measure the small ion density and conductivity as a function of altitude and to interpret the results in the light of the presently accepted theoretical concepts. Also included are the results of a constant level balloon flight in the stratosphere. Some of the theoretical investigations by the author are described at the end of chapter. In view of the very purpose of the present dissertation, the contributions by the author are dealt with in considerable detail.

2.1. Experiments :

2.1.1. Principles of ion measurements :

The technique of ion density and electric conductivity measurements has remained essentially the same since such measurements were first attempted. It consists of measuring the ionic current in a cylindrical chamber (Gerdien type condenser). The Gerdien type condenser consists of two parts, the inner and outer electrodes. Figure 7 shows the schematic diagram of the Gerdien condenser, along with a trajectory of an ion in an ideal case in which the ion enters the condenser just from the surface of the outer electrode and is trapped at the end of inner electrode under the influence of electric field between outer and inner electrodes. The mobility of the ion is defined as the critical mobility of the system and is given by,

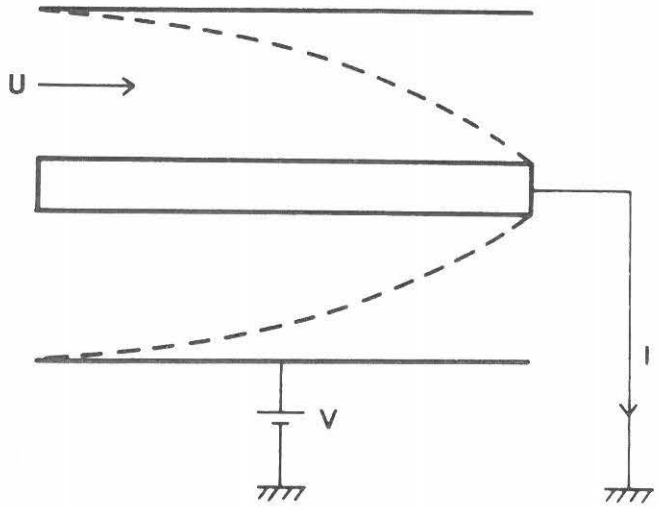

Figure 7. Schematic diagram of the Gerdien condenser showing the trajectory of a trapped ion. The legend is as follows :

U : the air speed

V : the voltage difference between the inner and outer cylinder

I : the current

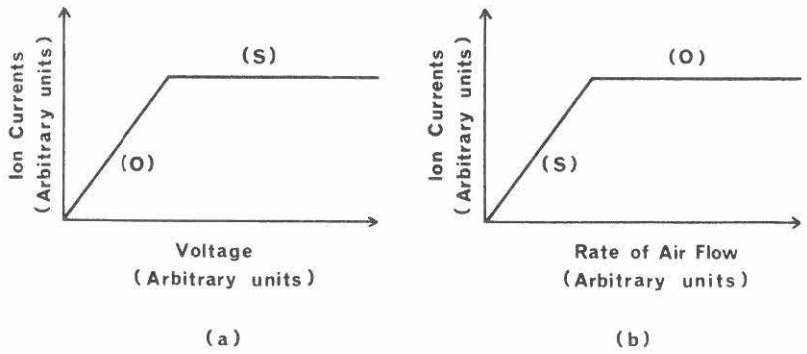

Figure 8. Typical characteristic curves of the condenser.

(a) Illustrating the variation of ion current with voltage difference between the cylinders.

(b) Illustrating the variation of ion current with the rate of air flow.

O : indicates the Ohmic part of the current

S : denotes the saturation part

$$k_c = \frac{\varepsilon}{C} \cdot \frac{\Phi}{V} \qquad (14)$$

where Φ is the rate of air flow, V is the applied voltage between two electrodes, C is the capacity of the condenser and ε is the dielectric constant of air. It can be easily seen that all the ions with mobility larger than k_c should be trapped by the inner electrode during their transit through the condenser. On the other hand, only a fraction of the ions with mobility k less than k_c is trapped for a given value of V. Simple calculation shows that the fraction is k/k_c. The current I flowing into the inner electrode is given by,

$$I = e\Phi\{\int_{k_c}^{\infty} f(k)dk + \frac{1}{k_c}\int_0^{k_c} kf(k)dk\} \qquad (15)$$

where e is the elementary charge and f(k) is the distribution function, that is the number of the ions dn with mobility between k and k+dk is given by f(k)dk.

If we consider a simple situation in which all the ions have the same mobility k^*, we have the two following cases :

(1) $k^* < k_c$ —— When the mobility of ions is less than the critical mobility of the system, the current can be expressed, from equations (14) and (15), as :

$$I = \frac{C}{\varepsilon} \cdot ek^*n \cdot V, \quad \text{and} \quad \lambda = ek^*n \qquad (16)$$

where n is the ion density and λ is the polar conductivity.

(2) $k^* > k_c$—— When the mobility of ions is larger than the critical mobility of the system, equation (15) becomes,

$$I = e\Phi n \qquad (17)$$

The typical characteristic curves in Figure 8(a) and (b) summarize the relations mentioned above. Figure 8(a) shows the voltage-current characteristics, while 8(b) indicates the rate of air flow-current characteristics. The characteristic curves consist of two parts : Ohmic (O) and Saturation (S) current. In the saturated state, all ions entering the condenser contribute to the current which is a function of the ion density and the rate of air flow as shown in equation (17). If the voltage applied to the condenser is reduced (or the rate of air flow is increased), the condenser becomes unsaturated. That is, the current becomes independent of the rate of air flow and is a function only of the ion density and the mobility (i.e.,of the electric conductivity). By suitably adjusting the physical dimensions of the condenser, the rate of air flow and the voltage applied to it, one can therefore measure either the ion density or the conductivity.

2.1.2. Experimental procedure :

Under the auspices of the Research Institute of Space and Aeronautical Science, University of Tokyo, five balloon flights have been carried out during the years 1967 and 1970. The relevant data pertaining to these flights are shown in Table 1. Though five experiments have been made, we concern ourselves mainly with the results obtained from the last three flights, because the reliability of data obtained from the first two flights has been rather questionable. For example, in the experiments of Flight I, the voltage-current characteristics of the Gerdien condenser have sometimes changed irregularly during

Table 1. Data describing the scope of the balloon flights.

Flight NO.	I	II	III	IV	V
Date	Oct. 21, 1967 Nighttime	Sep. 5, 1969 Nighttime	Sep. 6, 1969 Daytime	Sep. 21, 1970 Daytime	Sep. 21, 1970 Nighttime
Parameters measured	Small ion density Conductivity	Small ion density Mobility spectrum	Small ion density Conductivity	Small ion density	Small ion density
Altitude, km	24.8	21.7	19.0	24.0	30.5
Reliability	Not good	Not good	Good	Good	Good

the flight, and in the case of Flight II, the records were very much disturbed by undesirable factors. Thus we are obliged to exclude the results of Flights I and II in the present discussion.

The average mobility of small ion, k, is inversely proportional to atmospheric density and can be written as :

$$k = k_0 \left(\frac{\rho_0}{\rho} \right) \qquad (18)$$

where k_0 is the appropriate average ionic mobility at N.T.P. and ρ_0 and ρ are the average atmospheric densities at N.T.P. and at the point of measurement respectively. Usually, the average ionic mobility lies between 1.0×10^{-4} and 2.0×10^{-4} m^2/volt. sec. at ground level and increases by a factor of about 100 times at an altitude of 30 km. The applied voltage to the electrodes of the condenser and the rate of air flow were so arranged that the condenser would be saturated for all ions of mobility greater than an appropriate critical mobility k_{c1} for the ion density measurements, and for the conductivity measurements the condenser would be unsaturated for all ions of mobility less than a critical mobility k_{c2}. Figure 9 shows the mobility profiles predicted by equation (18) for three values of k_0. Typical critical mobilities k_{c1} and k_{c2} used in the baloon experiments are also shown.

Except for some minor modifications, the basic design considerations for the entire equipment including the Gerdien condensers were similar to those adopted by earlier workers (e.g., Paltridge, 1965,1966a). Table 2 summarizes the design of instrumentation. The symbols shown in Table 2 have the following meanings : D and d are the diameters of the outer and inner

Table 2. Data relevant to the instrumentation used in the balloon flights. See text for an explanation of the symbols.

Flight NO.	Small ion density				Electric conductivity			
	D, mm	d, mm	L, mm	Kc_1, $m^2/v.s.$	D, mm	d, mm	L, mm	Kc_2, $m^2/v.s.$
III	80	20	200	1.1×10^{-4}	80	10	100	105×10^{-4}
IV	80	20	200	1.1	80	10	100	105
V	130	20	300	1.0	—	—	—	—

electrodes and L is the length of the electrode. Only in the case of Flight V the condenser had a micro-blower. The ion density and conductivity have been obtained by using downward vertical airflow relative to the condensers. As examples, the schematic diagrams of the Gerdien condensers used are shown in Figures 10(a) and 10(b). Figure 10(a) shows the Gerdien condensers for the small ion density and electric conductivity measurements used in Flights III and IV, while Figure 10(b) indicates that for the small ion density measurements in Flight V. The ion density which can be measured with the Gerdien condenser depends directly on the rate of air flow which, in its turn, decreases with altitude.

It may be mentioned here that a number of laboratory experiments has been made, prior to the flights, to calibrate the equipments and to ensure the stability of the entire arrangement during the flights. Thus, wind tunnel experiments have been performed to calibrate for the rate of air flow in the case of Gerdien condensers without the blower. For the equipment on Flight V, when the blower type condenser was used, the blower current had been checked periodically during the flight. Also the characteristics of ion current versus applied voltage to the condenser have been calibrated in the laboratory. The small ion density and electric conductivity measured simultaneously and independently by using two Gerdien condensers showed a fairly good agreement. The entire assembly of the equipment used in the all experiments was consisted of the Gerdien condenser system, temperature sensors, swichting barometer, radio sonde, and

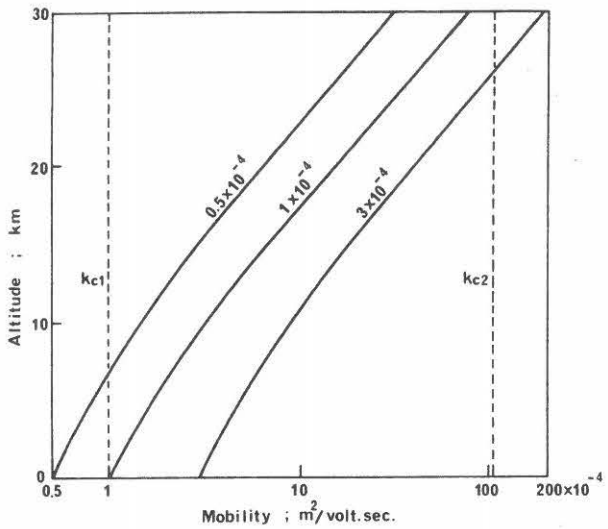

Figure 9. The mobility profiles predicted by equation (18). The figures characterizing the curves denote the values of k_0 in equation (18). k_{c1} and k_{c2} are the typical critical mobilities appropriate for the balloon measurements.

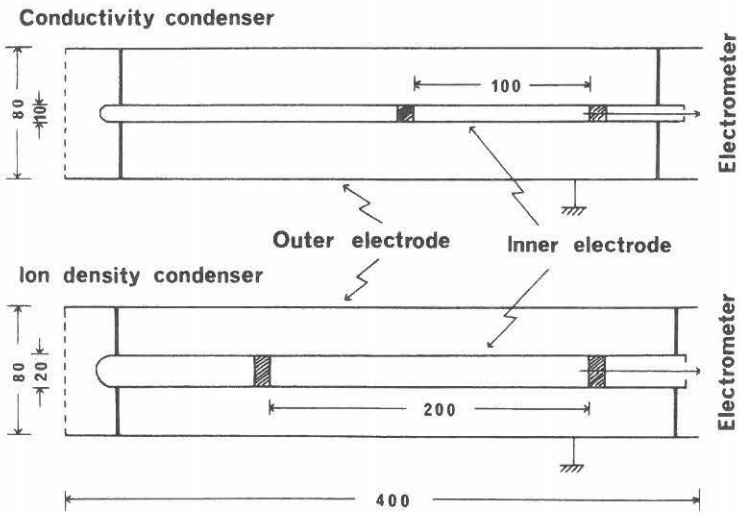

Figure 10(a). Schematic diagram of the Gerdien condensers used in Flights III and IV. All the dimensions are in millimeter.

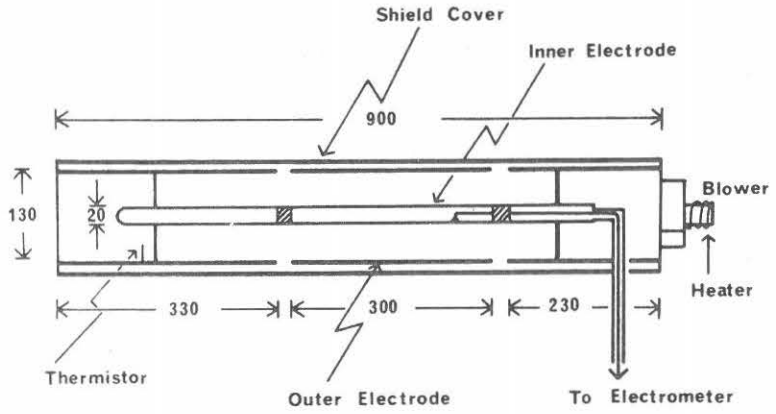

Figure 10(b). Same as Figure 10(a) for Flight V.

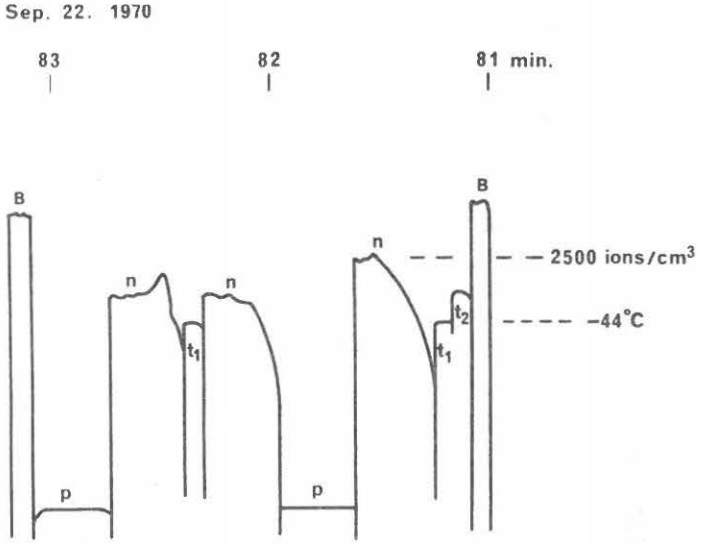

Figure 11. An example of the actual record obtained during Flight V, showing the values of blower current (B), temperature of the air both outside (t_1) and inside (t_2) the gondola, ion density (n) and pressure (p). The record corresponds to an altitude range from 24.7 to 25.6 km.

other accessories. An example of the actual record obtained from Flight V is shown in Figure 11. The time taken to complete one set of measurements of all parameters (blower current, temperatures of the air both inside and outside the gondola, ion counts and pressure) was about 2 minutes. The zero level of the condenser was checked automatically every 20 minutes by applying zero voltage to the outer electrode of the condenser. The telemetering frequency used was 1680 MHz.

2.1.3. Experimental results :

The results of Flight III are presented in Figures 12 and 13, which show the variations of positive small ion density and positive polar electric conductivity with altitude respectively. Figure 14 shows the variation of positive small ion density obtained from Flight IV. All the results in these figures are obtained under daytime conditions. The small ion densities shown in the figures have been derived from equation (17) with $\Phi = \pi u(R^2 - r^2)$ where u is the balloon ascent velocity, and R and r are the radii of outer and inner electrodes of the condenser respectively. The variation of negative small ion density with altitude obtained during the nighttime is shown in Figure 15, which indicates the result of Flight V.

In general, the above results can be compared very favorably with the independent measurements of other workers like Paltridge (1965). Though the variation of conductivity shows the usually exponential increase with altitude, the decrease in the small ion density at high altitudes is observed in the

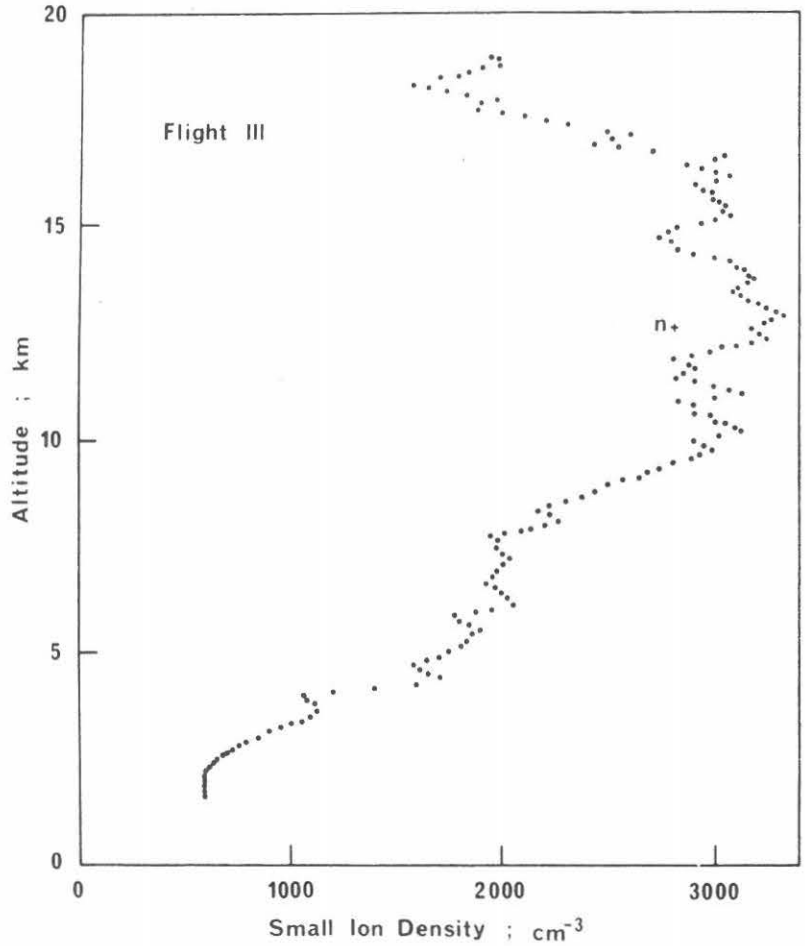

Figure 12. Showing the distribution of positive small ion density with altitude obtained from Flight III.

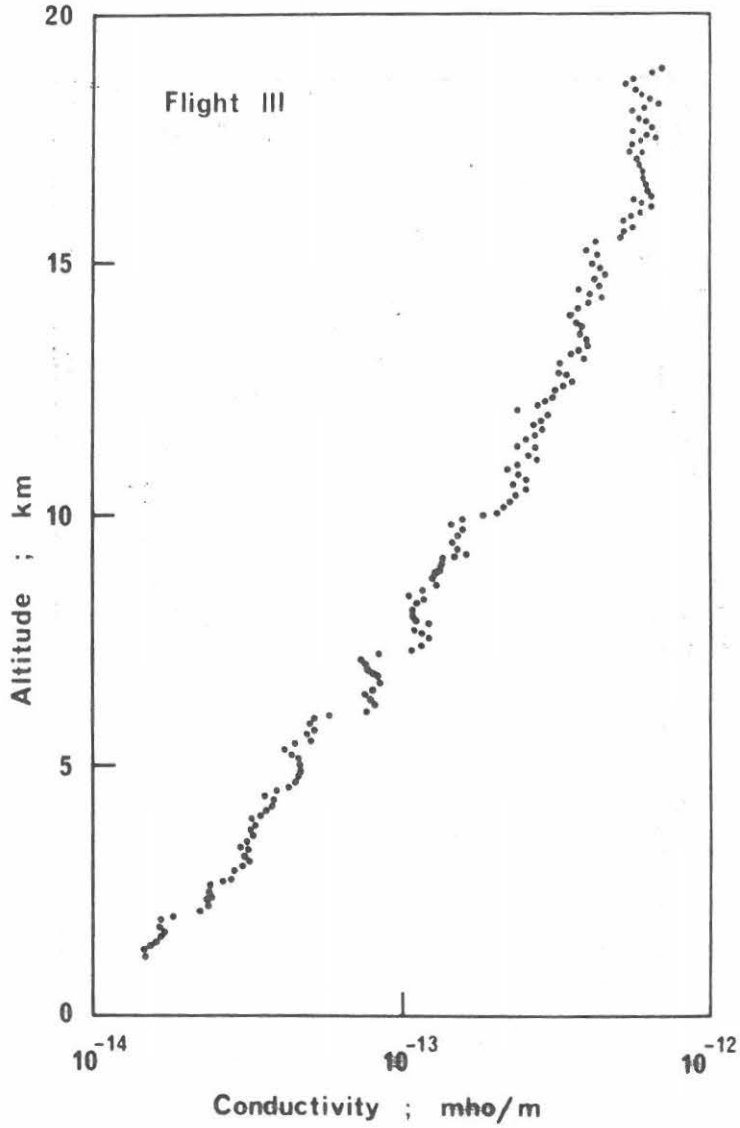

Figure 13. Showing the variation of positive polar conductivity with altitude. Flight III.

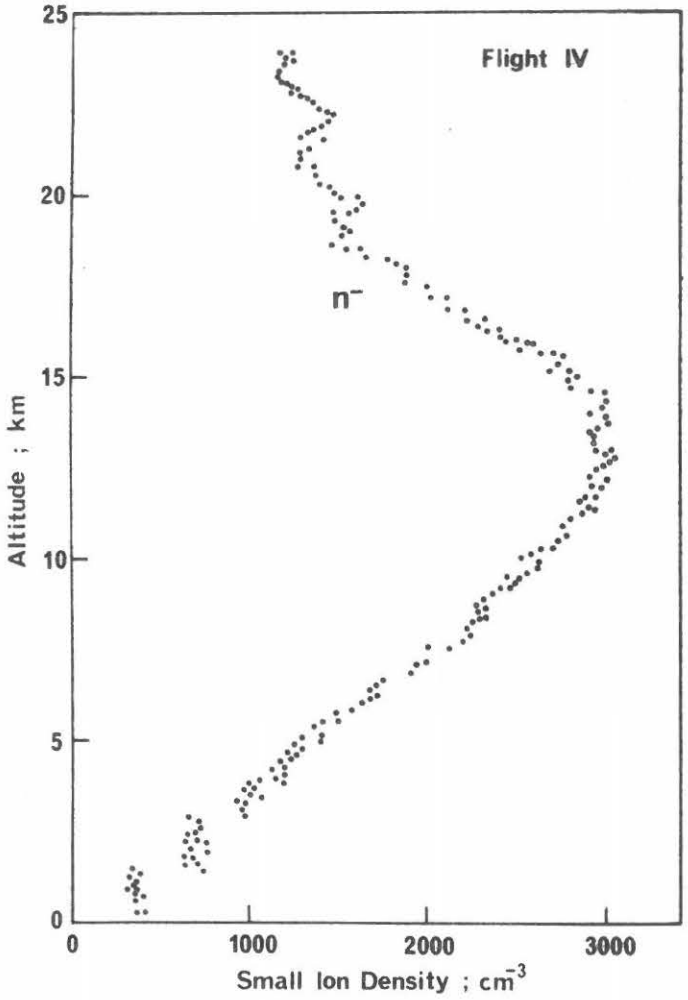

Figure 14. Showing the distribution of negative small ion density. Flight IV.

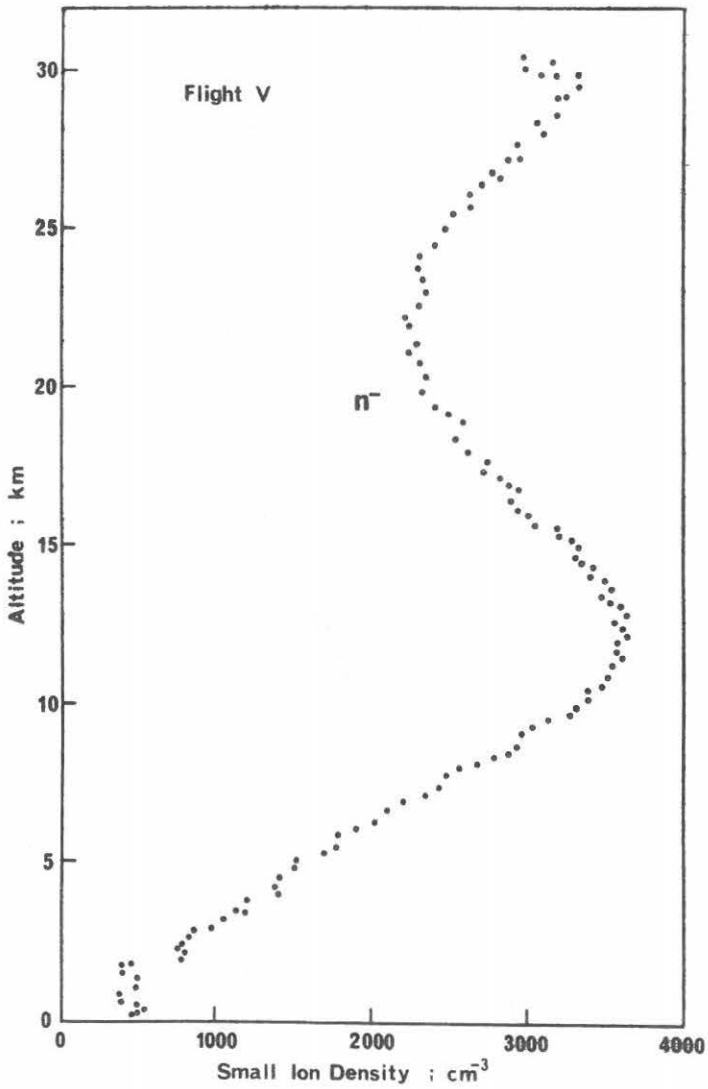

Figure 15. Same as Figure 14. Data obtained from Flight V.

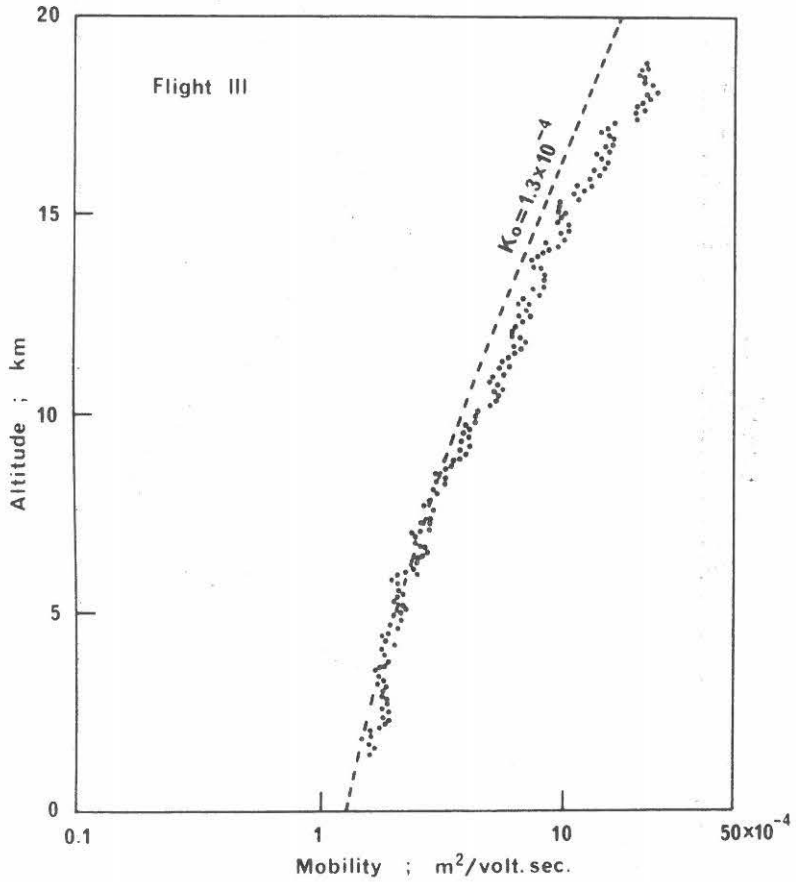

Figure 16. Average ionic mobility distribution derived from the measured values of small ion density and conductivity (shown in Figures 12 and 13 respectively). The broken line indicates the mobility predicted by equation (18) with $k_0 = 1.3 \times 10^{-4}$ m^2/volt.sec.

results of all flights and for both polarities.

Using equation (16) and the measured values of the small ion density and conductivity given in Figures 12 and 13, one can derive the altitude distribution of average ionic mobility shown in Figure 16. The broken line is the mobility profile predicted by equation (18) for a value of $K_0 = 1.3 \times 10^{-4}$ m^2/volt. sec. It is clearly seen from Figure 16 that the data points deviate significantly and systematically from the mobility profile predicted by equation (18) at 9 km and above. This deviation is in all probability due to the fact that a complete aerodynamic analysis has not been included in the reduction of data obtained by the Gerdien condenser. It is, in principle, possible to include such an analysis with the help of the wind-tunnel experiments and to improve the agreement between the data points and the broken line in Figure 16. The following section is devoted to such aerodynamic considerations.

2.2. Analysis of the Results and Discussion :

2.2.1. Aerodynamic investigations :

It is well known that the ion density obtained by using a Gerdien condenser depends directly on the rate of air flow in the condenser. The rate of air flow, when no blower is used, is a function of the Reynolds number. The dependence of the Reynolds number on the altitude necessitates the use of a correction factor which, in the present cases, has been determined following the procedures described by Paltridge (1965) and Morita and Ishikawa (1969b).

The flow pattern through along straight pipe is determined

uniquely by its Reynolds number, that is, by the dimensionless quantity uD/ν where u is the velocity and ν the kinetic viscosity of the fluid, and D is the pipe diameter. The dependence of the fully developed flow on the Reynolds number is a well documented phenomena. However, in the case of a short Gerdien condenser, such as has been used in Flights III and IV , the relatively simple theory of fully developed flow does not apply. The type of flow corresponds roughly to that in the inlet length of a long pipe, and is a highly complicated function of the Reynolds number and tube length. For the typical balloon-borne instruments, the associated Reynolds number during ascent varies enormously from very high values near the ground to very low values at stratospheric heights. This is shown in Figure 17, where the Reynolds number associated with the present Gerdien condenser is plotted as a function of altitude for a typical balloon ascent rate of 4.0 m/sec.

Wind tunnel experiments have been performed to calibrate the rate of air flow as a function of altitude (or the Reynolds number). For this purpose the Gerdien condenser model was placed in a low speed wind tunnel. Measurements of the actual air speed across the condenser have been made by using a hot wire anemometer and the results of actual rate of air flow were compared with those in the absence of the condenser. The required variation in the Reynolds number was achieved by varying the flow speed in the wind tunnel itself. The actual velocity profiles across the condenser obtained from the wind tunnel experiments are shown in Figure 18. Depending on the Reynolds number, the following patterns could be obtained :

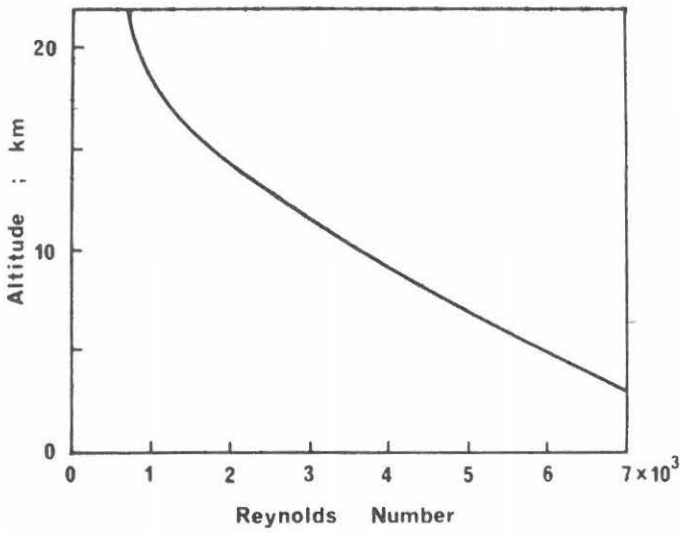

Figure 17. Showing the altitude dependence of the Reynolds number associated with the Gerdien condenser.

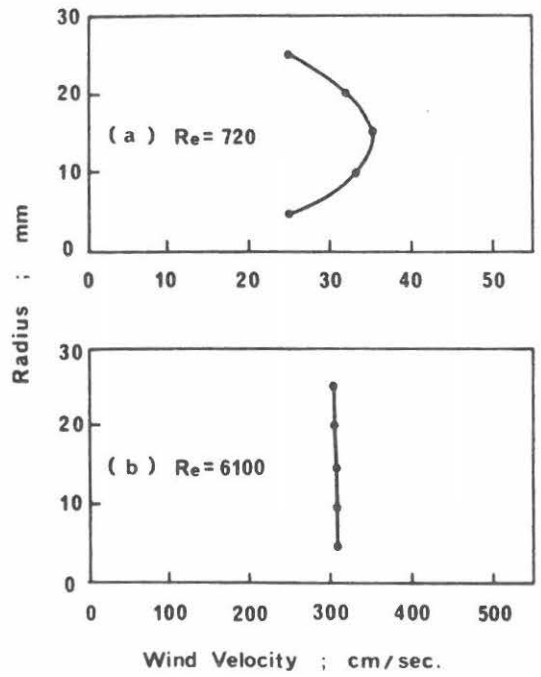

Figure 18. Showing the actual wind velocity profiles across the condenser from wind tunnel experiments. Re stands for the Reynolds number.

(a) For low Reynolds number(Figure 18(a)), the velocity profile in the condenser at the exit assumes parabolic shape of the fully developed laminar flow. Viscous energy loss is high, and the actual flow rate is strongly dependent on the Reynolds number and the length of the condenser.

(b) At higher Reynolds number (Figure 18(b)), the velocity profile is constant across the entrance, and remains practically unaltered at the exit. The flow pattern is virtually unaltered by the presence of the condenser.

It is well known that turbulent flow in the Gerdien condenser could be a source of serious error in the determination of the ion density. By introducing smoke into the air stream in the wind tunnel experiments, we have observed that turbulence was negligible even when the Reynolds number was high. Thus, in our balloon experiments the influence of turbulence has been ignored.

From these wind tunnel experiments whose results are accurate to within five percent, a correction factor has been obtained and is shown in Figure 19 as a function of altitude. The values of small ion density obtained from Flights III and IV (and shown in Figures 12 and 13) have thus to be multiplied by the correction factor shown in Figure 19. It may be mentioned that the present estimates of the correction factor are in excellent agreement with those reported by Riekert (1971) recently.

The measured values of the small ion density and conductivity usually show small-scale fluctuations throughout the altitude range, which are particularly evident in the case of

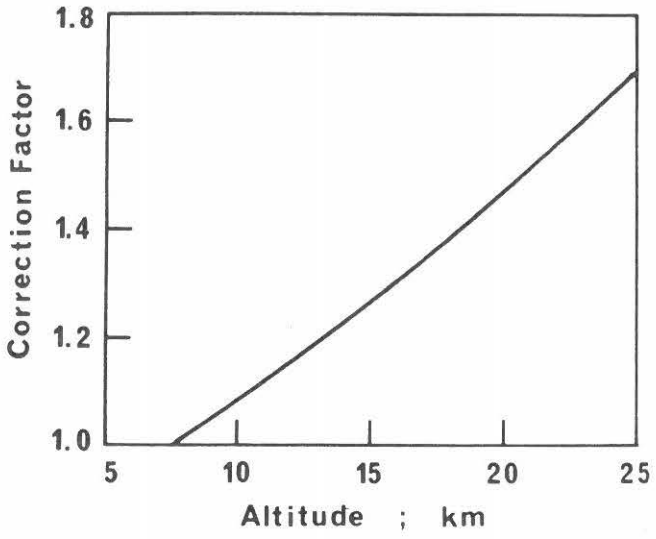

Figure 19. The correction factor versus altitude estimated from the wind tunnel experiments.

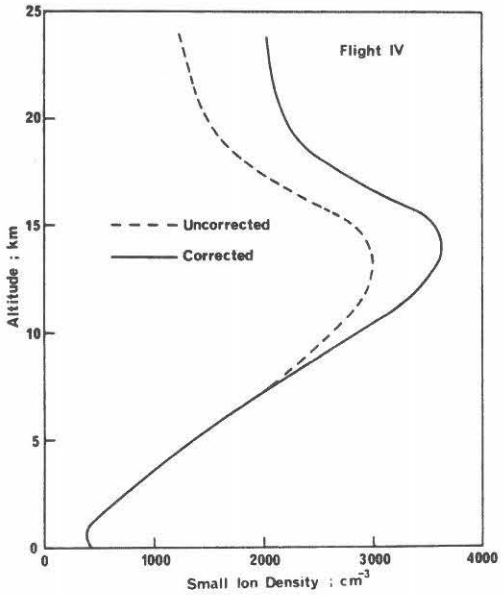

Figure 20. The uncorrected and corrected (obtained by multiplying the uncorrected values by the correction factor shown in Figure 19) ion density profiles. Flight IV.

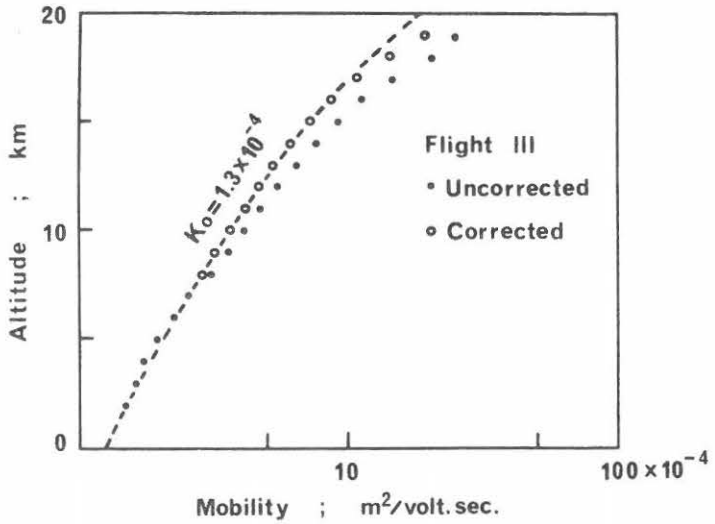

Figure 21. Uncorrected (closed circles) and corrected (open circles) mobility profiles. The broken line indicates the mobility predicted by equation (18) with $k_0 = 1.3 \times 10^{-4}$ m^2/volt.sec.

Flight III (see, Figures 12 and 13). While we could ascertain that the rotation and swinging of the gondola were negligible in the lower atmosphere, we could not do so at higher altitudes where sudden changes in the vertical ascent velocity might have also occurred. If present, these effects could cause high variability in the measured values of ion density and conductivity. We thus confine our discussion mainly to the smoothed up profiles of the measured parameters ignoring the small-scale fluctuations at higher altitudes.

Both the corrected (i.e. using the data in Figure 19) and uncorrected ion density profiles are shown in Figure 20, which is the result of Flight IV. The revised mobility distributions have been obtained by using the corrected values of ion density, and are shown in Figure 21. The agreement between the corrected mobility distribution and that predicted by the inverse density rule is now excellent upto 17 km, and this shows the reliability of present data. There is, however, some tendency in the measured profile to level off above 17 km. This is probably due to the non-uniformity of the velocity profile across the condenser when the balloon reaches these altitudes. Such non-uniform velocity profiles are likely to render the conductivity readings inaccurate (Paltridge, 1965). Our own wind tunnel experiments attest to this conclusion.

2.2.2. The vertical profiles of small ion density and electric conductivity :

A. Interpretation of the results :

The electrical state of the atmosphere sometimes shows

rapid changes in the lower atmosphere. Measurements showing such rapid changes of the electrical parameters with increasing altitude near the exchange layer have been reported by many workers (e.g., Sagalyn and Faucher, 1954 and 1956)and have been interpreted mostly in terms of the influence of the meteorological conditions.

Our results also show sudden changes in the small ion density at certain altitudes as shown in Figures 22(a), (b) and (c). Only data below an altitude of 5 km are shown in these figures because, as already mentioned in the previous section, we could be certain that within this height range the rotation and the swinging of the gondola were negligibly small. Rapid and significant changes in the measured parameters are easily discernible at an altitude of about 4 km in Figure 22(a) and at altitudes of 1.5 and 3 km in Figure 22(b). Similarly, the nighttime measurements plotted in Figure 22(c) show such changes around about 2 km. It can be seen that these changes correspond roughly to the temperature inversions. The rapid changes of the ion density could probably be attributed to the influence of the suspended submicron aerosols in the lower atmosphere. Though the number density of aerosols usually shows more or less an exponential decrease with altitude, the distribution of aerosols does not change systematically in the exchange layer. It is also known that the top of the exchange layer is characterized usually by the discontinuity in the temperature distribution and a sharp decrease in the aerosol concentration. The height where such discontinuous changes occur depends strongly on the

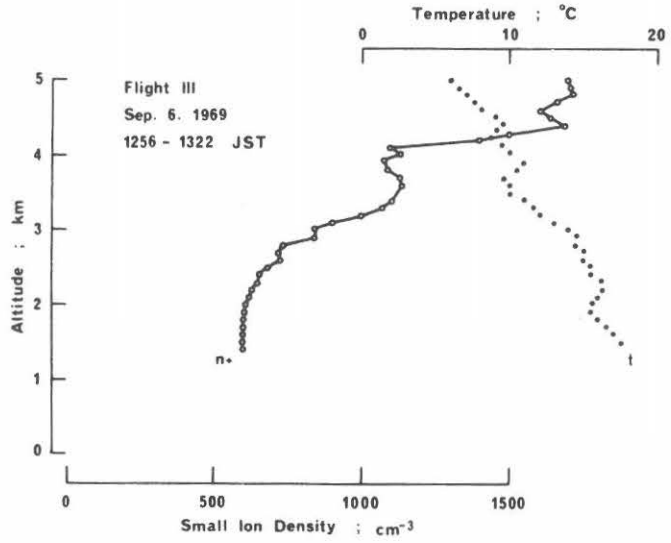

Figure 22(a). Showing rapid changes in positive ion density (n_+) and air temperature (t). Only data below 5 km altitude are shown. Flight III.

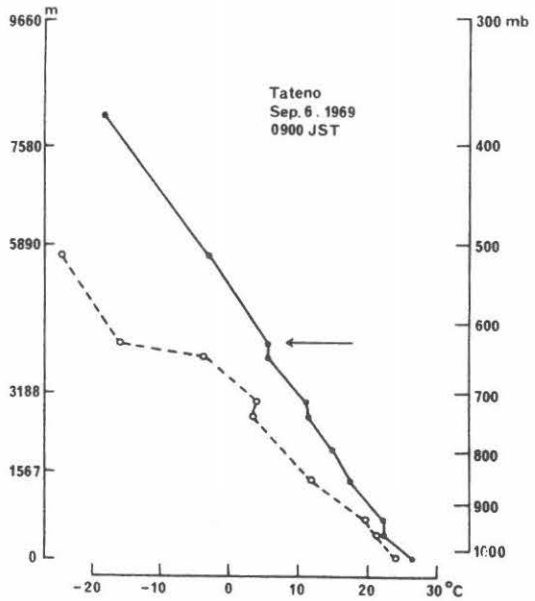

Figure 23(a). Meteorological data relevant to Flight III. Full line shows the air temperature while the broken line gives the dew point. The arrow indicates the altitude where rapid changes in ion density have occurred (see Figure 22a).

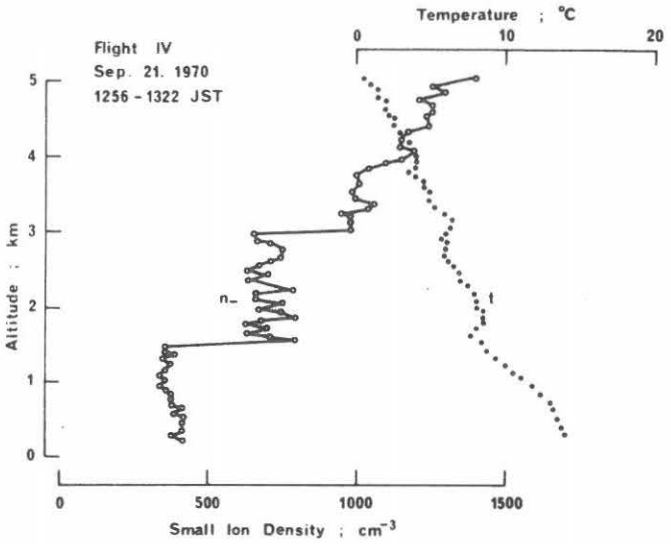

Figure 22(b). Showing rapid changes in negative ion density (n_-) and air temperature (t) during daytime. Only data below 5 km altitude are shown. Flight IV.

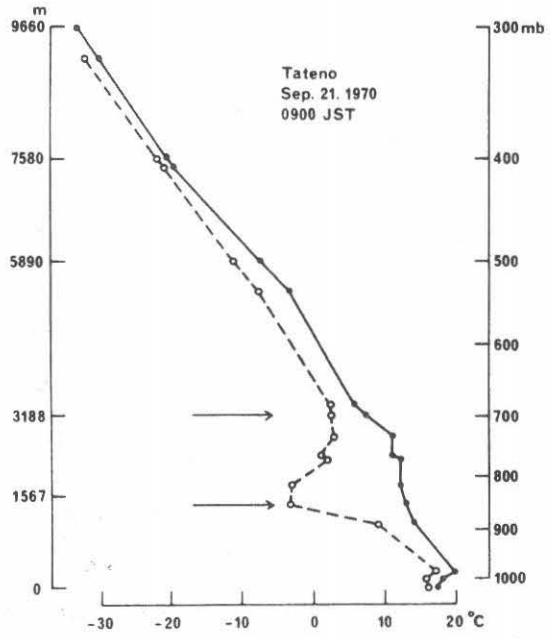

Figure 23(b). Meteorological data relevant to Flight IV.

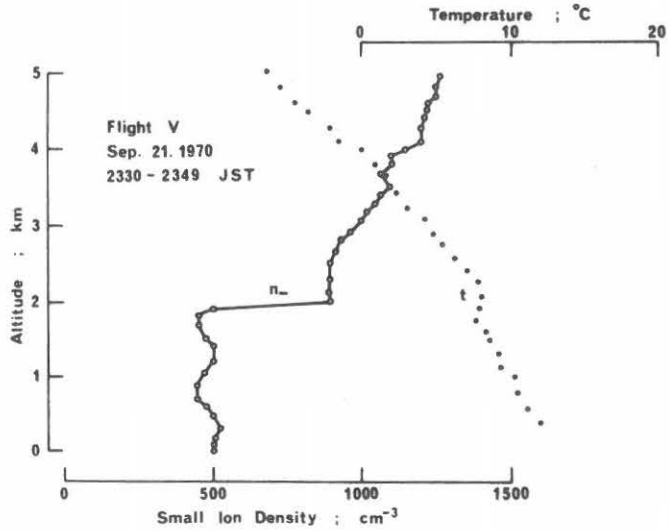

Figure 22(c). Same as Figure 22(b) for nighttime.

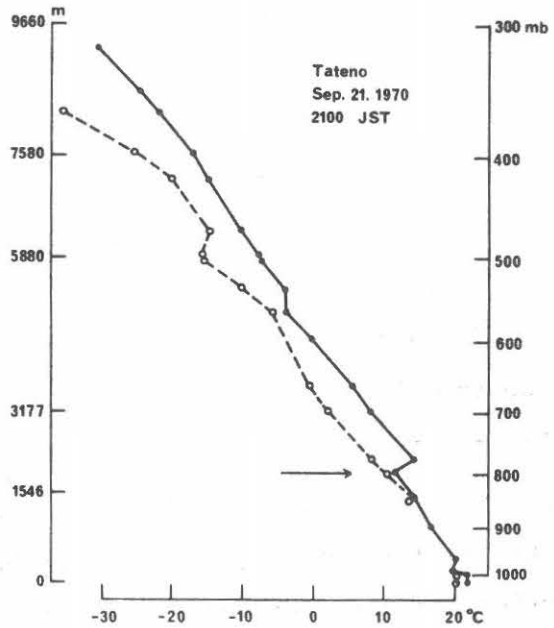

Figure 23(c). Meteorological data relevant to Flight V.

meteorological conditions and varies considerably from day to day and with season. It can be further seen from Figures 22(a), (b) and (c) that the altitude characterizing rapid changes in the ion density is higher in the daytime than in the nighttime. This could perhaps be attributed to the fact that convection is weaker in the nighttime than in the daytime. With a view to substantiate the connection between the rapid changes in the ion density shown in Figure 22 and the meteorological conditions, we have studied the emagrams obtained Tateno and Sendai Aerological Observatories. The meteorological data relevant to Flights III, IV and V are shown in Figures 23(a), (b) and (c) respectively. The arrows in these figures indicate heights where the ion density in Figures 22(a), (b) and (c) changes rapidly.

A detailed study of the emagrams has suggested that during the periods of balloon flights, possible meteorological conditions conducive to the existence of different air masses are likely to have existed at these altitudes.

Above the altitudes characterized by sudden changes, the ion density increases upto a height of about 14 km in the daytime (Flights III and IV)and 12 km in the nighttime (Flight V). Figure 24 shows the smoothed up vertical profiles from all flights. The full and broken lines in the figure show the negative ion density profiles obtained during nighttime (Flight V) and daytime (Flight IV) respectively, while the chain line indicates the daytime positive ion density profile from Flight III. It may be mentioned here that for heights above 25 km the simple

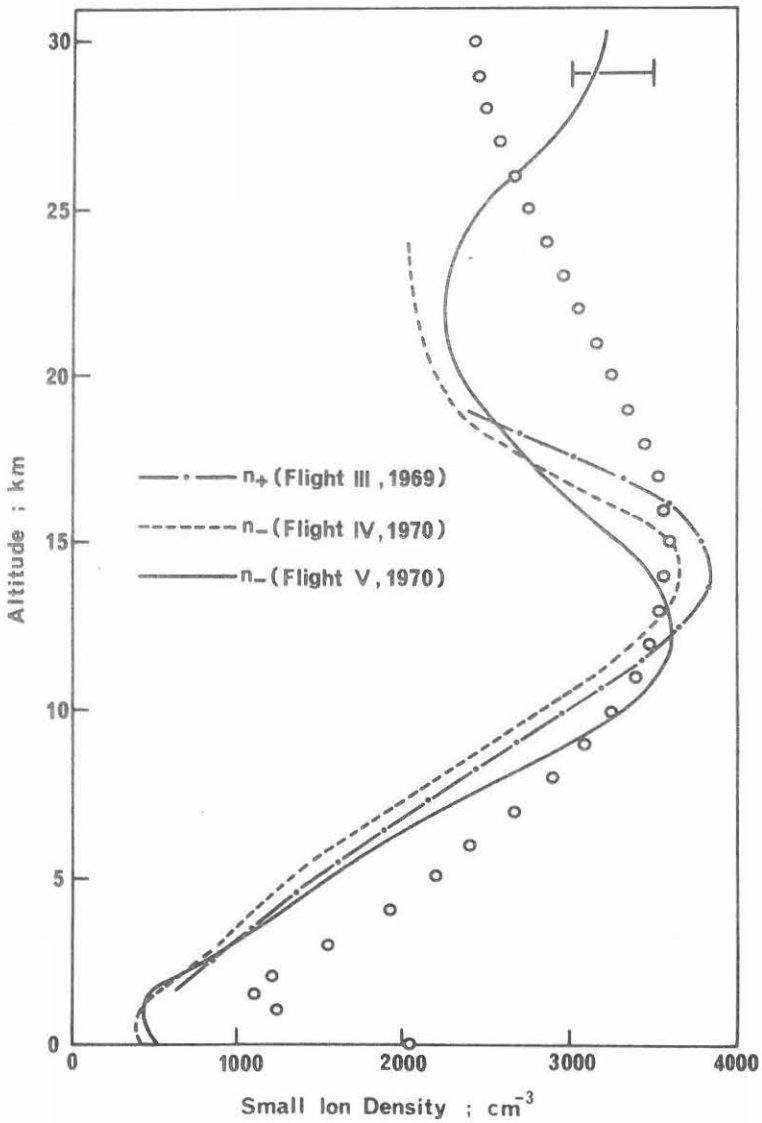

Figure 24. Superposed plots of ion density measurements from all the flights. The legend is as follows :

Full line : negative small ion density in the night (Flight V).

Broken line : negative small ion density in the day (Flight IV).

Chain line : positive small ion density in the day (Flight III).

Open circles : small ion density determined from the ionization measurements of Ikebe et al. (1970).

See text for details.

aerodynamic considerations given in the previous section are not sufficient to measure the ion density and conductivity with reasonable accuracy. For Gerdien condensers without blowers, at such heights, the rate of air flow correction to the ion density readings is grossly inaccurate, and the conductivity measurements are unreliable. In order to obtain reliable values of the ion density above 25 km, Gerdien cendensers with the blower must be used, as was done in the case of Flight V (See Figure 10(b)).

B. Discussion in the light of others'work :

In this sub-section we shall discuss the implications of our results in the light of the researches carried out by other workers.

A few days before our balloon flights at Haranomachi Balloon Base (geomagnetic latitude 27.7^{o}N) Ikebe et al. (1970) measured the height variation of ionization by means of an air-borne ionization chamber at the same base. The profile of ionization is shown in Figure 25. It is evidently desirable to see how far these two independent sets of experiments are mutually consistent.

The ionization measurements of Ikebe et al. (1970) can be converted into an ion density profile, assuming a reasonable profile for the recombination coefficient between positive and negative ions. Assuming the small-ion theory of Thomson to hold (Stergis et al., 1955), ion densities at various heights have been calculated from the ionization measurements of Ikebe et al. (1970), and are shown in Figure 24 as open circles at

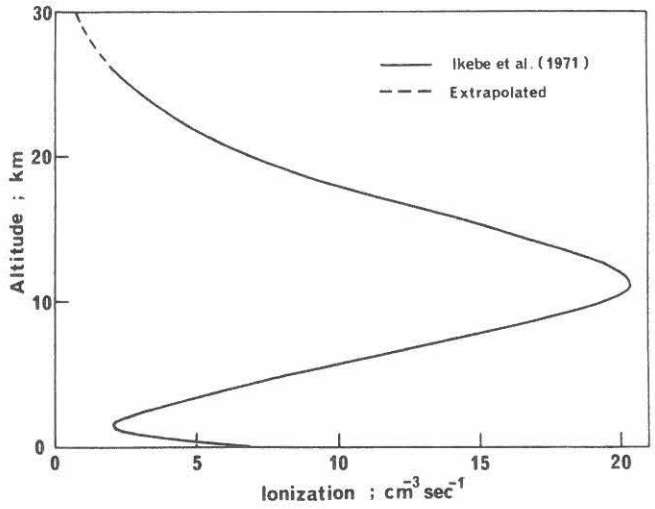

Figure 25. Ion production rates reported by Ikebe et al. (1970). Broken line indicates the region of extrapolation.

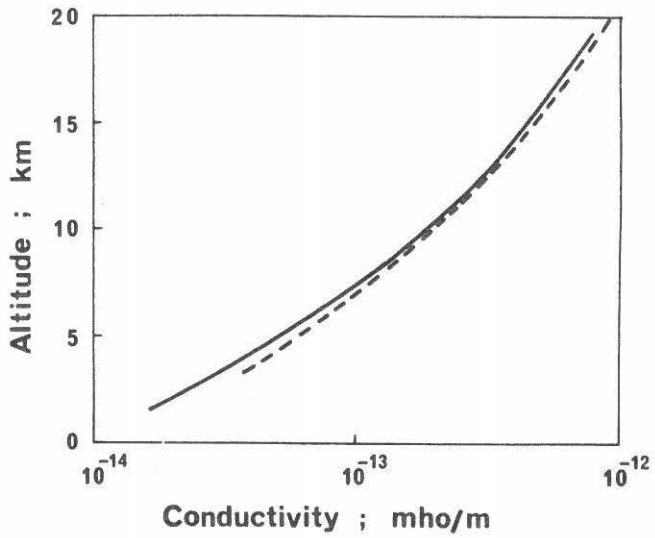

Figure 26. Comparison between the present conductivity measurements (full line) and those deduced from the ionization measurements of Ikebe et al. (1970) (broken line).

3 km-intervals. It is evident from Figure 24, that the agreement between the measured nighttime profile (continuous line) and that deduced from the ionization data of Ikebe et al. (1970) is reasonably good only in the limited altitude region of 9 to 13 km. The discrepancies between the two profiles are very significant both above and below this altitude range. Similarly, the measured daytime profiles (broken and chain lines) show agreement only in the region between 11 and 15 km. It may be mentioned here that one of the possible reasons for the disagreement between our results and those of Ikebe et al. (1970) is the existence of aerosols which could influence, to a marked degree, the small ion density. The author has studied this possibility and in section 2.2.3 is given a rather detailed account of the influence of the stratospheric aerosols on the small ion density.

Just as we have compared, in the above, our measurements of ion density with those deduced from the observations of Ikebe et al. (1970), we can also compare the conductivity profiles. The ionization measurements of Ikebe et al. (1970) can be converted into conductivity profiles assuming a reasonable value for the average ionic mobility (K_0). It is seen from Figure 21, that $K_0 = 1.3 \times 10^{-4}$ m^2/volt.sec. is quite satisfactory. The comparison between the conductivity profile obtained from Flight III and that deduced from the results of Ikebe et al. (1970) is illustrated in Figure 26. As is the case with the ion density measurements, the agreement between conductivity profiles is good only in the altitude region of 9-13 km. The

reasons for the disagreement outside this altitude region are, of course, similar to those discussed in relation to ion density measurements and will not be repeated here.

The recent calculations by Shreve (1970) show that the theoretical ion density increased with altitude reaching a nearly constant value in the stratosphere. However, the profiles obtained by us (see Figure 24) do not show any tendency to level off at stratospheric heights. Indeed, the daytime profiles show significant decrease above 15 km. Further, the nighttime profile shows a conspicuous minimum around 22 km above which it again increases tending probably towards a second maximum around 29 km. Our results show that the daytime peak in the ion density is roughly 2 km higher than the nighttime peak. This is contrary to the finding of Bragin (1967) who reported that during daytime the lower boundary of the stratospheric ion maximum is situated 5-10 km closer to the earth's surface than in the nighttime. It does not seem possible to offer a simple explanation for these discrepancies between the theory and experiment in terms of any spatial and temporal changes in the parameters concerned. For example, at stratospheric heights, the ionization is mainly due to cosmic rays and shows little temporal and spatial variations, particularly in the region of lower geomagnetic latitudes. The recombination coefficient between positive and negative ions, which depends on the temperature and pressure in the ambient atmosphere, cannot also show significant variation since the changes of temperature and pressure in the stratosphere are very small. Thus the

simple small-ion theory, as expressed in equation (13), does not predict any significant temporal and spatial variations in the ion density above the exchange layer. The factors controlling the temporal and spatial variations of the ion density in the stratosphere are still unknown at the present time. It is, however, to be hoped that an exact knowledge of the chemistry of the small ions and its dependence on the minor constituents, the existence and the nature of aerosols, etc., will lead to a better understanding of the disagreement between the theoretical and experimental ion density profiles. But presently our knowledge in this regard is rather limited. For example, the scheme of Mohnen (1971) suggests that the most probable positive small ion is the hydronium ion, $H^+(H_2O)_n$, with a likely value of n in the stratosphere of about 5 or 6, the actual degree of hydration depending on the temperature and the amount of water vapor in the atmosphere. The most important negative ion at the stratospheric heights is $NO^-_3(H_2O)_n$ the formation mechanism of which involves the atmospheric ozone. But the details have not been worked out. In any case, it seems essential to investigate the detailed chemistry of the small ion equilibrium, possibly including the influence of minor constituents, before any comprehensive understanding of the electric state of the stratosphere is achieved. In this regard the author has attempted to investigate the possible role of the atmospheric ozone in controlling the temporal variations in the stratospheric ion density. The details of these investigations are incorporated in section 2.2.4.

2.2.3. The small ion density at a constant level :

As mentioned in the introduction, we have conducted a constant level balloon-flight experiment on September 22, 1970, the purpose being to study the temporal variations at a specified height (in this case, 29.5 km). Within the estimated accuracy of the experiment, the instruments operated well on the whole and the results are shown in Figure 27. The parameters measured were the negative ion density and temperature. We could conduct only one such flight and as such the results cannot be considered conclusive. Yet, the following inferences could be drawn from the results

(a) There was considerable temporal variation in the ion density, the variation itself becoming smaller as the dawn approached.

(b) The measured ion densities were much higher than the theoretically predicted values (2500 cm^{-3}). It may be recalled here that even in the case of vertical sounding experiments, as mentioned in the previous sections, the measured ion densities at stratospheric heights were higher than the theoretical values.

It may be mentioned that in an ideal situation, we would expect a small decrease in the negative ion density following the stratospheric sunrise because of the photodetachment of electrons, especially in the higher regions. It is, however, doubtful if the photodetachment really plays any significant role at 30 km (Cole and Pieice, 1965). Of course, the Thomson theory predicts a small decrease in the densities of both the

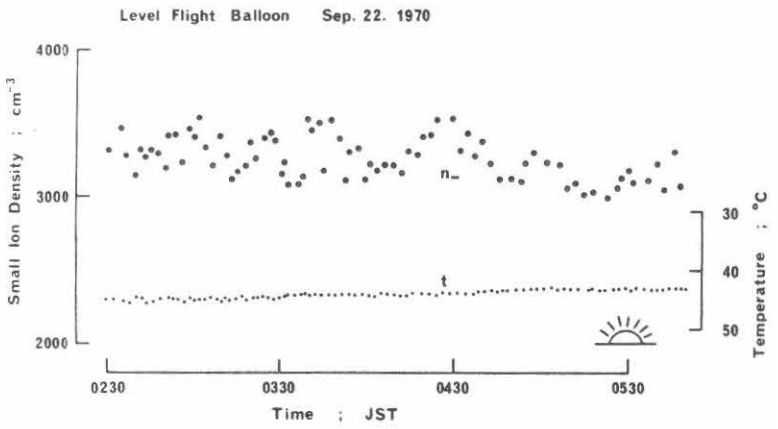

Figure 27. Showing the temporal variations in negative small ion density (n^-) and air temperature (t) at a constant level of 29.5 km.

positive and negative ions caused by the increase in the recombination coefficient when the temperature decreases but then temperature variations in the stratosphere are very small as shown in Figure 27. Thus, the predicted decrease in the ion density should be small. Recently, Coroniti (1971) suggested that the evaporation of water vapor from the surface of the balloon or from the instrumental package itself could cause a decrease in the ion density and points out that shortly after sunrise the water vapor pressure increased by a factor of a thousand. Inspite of the above discussion, it is not possible to pinpoint the reasons for the observed fluctuations in the ion density at stratospheric heights. It is rather difficult to associate these fluctuations with any changes in the ionizing flux (i.e. cosmic rays). The results are inconclusive, if only because they are not exhaustive, and, it is futile, at least at this state, to speculate on the factors responsible for temporal changes in the ion density in the stratosphere. Further experimental researches are clearly warranted.

2.2.4. The effect of stratospheric aerosol :

It was stated in Chapter I that to calculate the small ion density exactly the complete set of equations describing the ionic equilibrium must be solved. Recent investigations by several workers (e.g., Cole and Pierce,1965) indicate that practically no free electrons exist in the stratosphere so that the processes of attachment of electrons to neutral molecules, recombination between electrons and positive ions, and detachment

of electrons to form negative ions may be left out of consideration without much loss of generality.

When aerosols are present, and when eddy diffusion is negligible equations (2) and (3) may be used in the discussion of ionic equilibrium. Assuming that ionization in the stratosphere is solely due to cosmic rays and that charge neutrality is maintained, equations (2) and (3) may be written, under steady state conditions, as :

$$\left.\begin{aligned} q &= \alpha n^2 + nF(p) \\ F(p) &= \eta_0 N_0 + \sum_p (\eta_{12}^{(p)} + \eta_{11}^{(p)}) N^{(p)} \end{aligned}\right\} \qquad (19)$$

The total number density of aerosols can be expressed from equation (1), as :

$$Z = N_0 + 2\sum_p N^{(p)} \qquad (20)$$

From equations (19) and (20),

$$F(p) = \frac{\eta_0 + \sum_p (\eta_{12}^{(p)} + \eta_{11}^{(p)}) N^{(p)}/N_0}{1 + 2\sum_p N^{(p)}/N_0} \cdot Z \qquad (21)$$

Thus, when all the quantities in equation (21) are known, the small ion density at any height can be easily calculated from equation (19).

Several attempts have been made to estimate the attachment coefficients and the results at ground level vary, between different experiments, by a factor than 10 (Bricard,1965).

At higher altitudes, since the attachment coefficient depends theoretically on the size of aerosols, which is again

largely unknown, the problem is even more difficult. Though several theoretical treatments of this subject are available (Whipple,1933,1965, Junge,1955, Bricard,1965, and others), we follow the method of Bricard (1965) whose expressions for η_0, $\eta_{12}^{(p)}$ and $\eta_{11}^{(p)}$ are written as :

$$\left.\begin{aligned}
\eta_0 &= \frac{\pi R^2 v}{1 + \frac{vR^2}{4D(R+\Delta)}} \\
\eta_{12}^{(p)} &= \frac{p\pi R^2 v \frac{e^2}{\kappa T} \exp \frac{-pe^2}{\kappa T(R+\Delta)}}{\frac{pe^2}{\kappa T} + \frac{v^2}{4D} R^2 \{1-\exp \frac{-pe^2}{\kappa T(R+\Delta)}\}} \\
\eta_{11}^{(p)} &= \frac{p\pi R^2 v \frac{e^2}{\kappa T} \exp \frac{pe^2}{\kappa T(R+\Delta)}}{\frac{pe^2}{\kappa T} - \frac{v^2}{4D} R^2 \{1- \exp \frac{pe^2}{\kappa T(R+\Delta)}\}}
\end{aligned}\right\} \qquad (22)$$

where D and v are the diffusion coefficient and thermal velocity of the small ions, R is the radius of the aerosol, κ is the Boltzmann's constant, and Δ is expressed in terms of the mean free path δ as :

$$\Delta = \frac{1}{3R\delta} \{(R+\delta)^3 - (R^2+\delta)^{3/2}\} - R \qquad (23)$$

We can thus evaluate the attachment coefficients for a radius of aerosols at any height from equation (22).

Though many vertical measurements of size and number density of aerosol have been made in recent years, those relevant to

the region under the present study are relatively few. There are the measurements of Junge (1961), Chagnon and Junge (1961) and Rosen (1964). Junge (1961) has measured the concentration of aerosols of radius $\leq$ 0.1 micron in the altitude range of about 5 and 30 km, and Chagnon and Junge (1961) have obtained the vertical distribution of aerosols with an average radius of 0.15 micron. Rosen (1964) has measured aerosols with radii larger than 0.3 micron in the atmosphere to about 30 km. The measurements of Junge (1961) and Chagnon and Junge (1961) were made a few years before the eruption of Mt. Agung, Bali in March, 1963. The results of Rosen (1964) on the other hand, seem to have been perturbed by the eruption at Bali. According to Volz (1970) the Agung and subsequent volcanic eruptions have created a significant level of aerosols in the upper atmosphere since 1963. We thus may use the measurements of Junge (1961) for small aerosols as being representative for the middle latitudes, and those of Rosen (1964) and Chagnon and Junge (1961) as upper and lower limits respectively for large aerosols during the period of 1969 to 1970. Figure 28 shows the profiles of aerosol number density reported by these authors. The profile denoted by (a) in the figure also includes the data of Weickmann (1955) for the regions below 5 km.

It is necessary to know the ratio of neutral and ionized aerosols carrying p-elementary charges, $N^{(p)}/N_0$ in equation (21), for our computations. Keefe et al. (1959) and Bricard (1965) showed the distribution of charges follows a Boltzmann type law so that we may write :

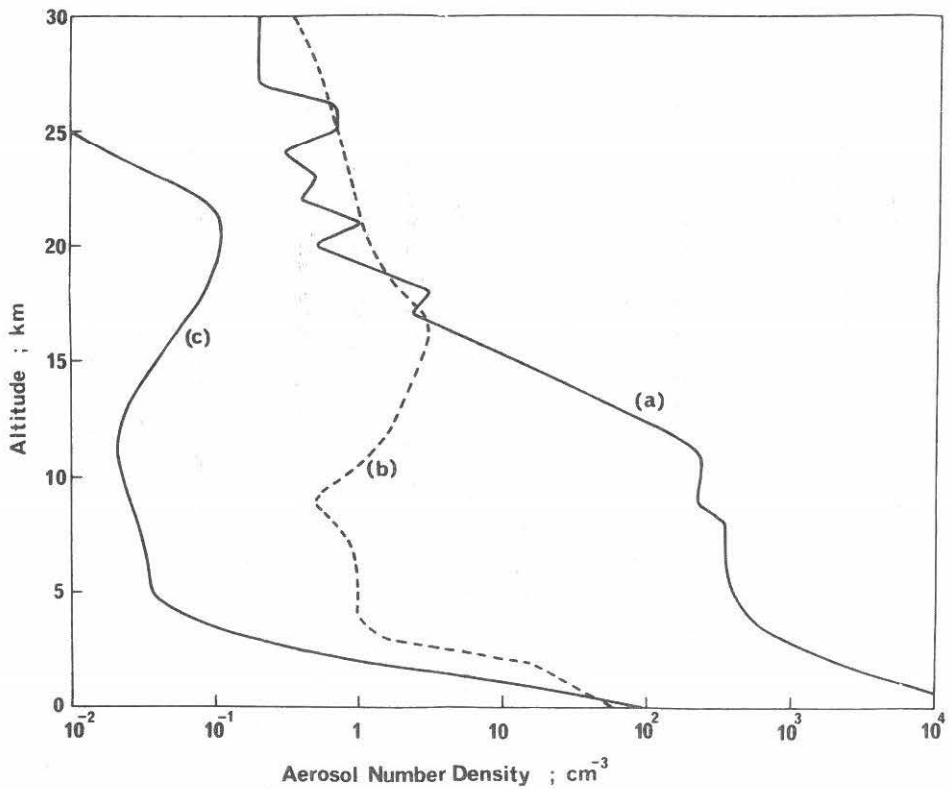

Figure 28. Some experimental profiles of aerosol number density.

(a) In the height range 5-30 km from Junge (1961). Below 5 km from Weickmann (1955).

(b) From Rosen (1964).

(c) From Chagnon and Junge (1961).

$$N^{(p)}/N_0 = \exp\left(-\frac{p^2e^2}{2RkT}\right) \qquad (24)$$

Equation (24) indicates that the ratio is about 0.45 for R = 1 μ and p = 5 and is about 0.07 for R = 0.3 μ and p = 5. What has been said so far is sufficient to allow computations regarding the influence of aerosols on the ion density, provided the production function q in equation (19) is known. For this purpose, we have made use of the ionization measurements of Ikebe et al. (1970). The results of computation, for the aerosol profiles (a) and (b) of Figure 28, are plotted in Figures 29 and 30 respectively. The results for the case in which both the types of aerosol distributions (i.e. (a) and (b) in Figure 28) are simultaneously present, are shown in Figure 31. Results depicted in Figures 29, 30 and 31 show that the influence of aerosols is significant at low altitudes. For example, we see from Figure 29 that at 10 km the aerosols could decrease of ion density by about 10%. At higher altitudes, the effect is very small and we do not anticipate any perceptible influence of aerosols at stratospheric heights.

Now we can easily explain the disparities at tropospheric heights, in the ion density profiles obtained from our flights and those computed from the ionization measurements of Ikebe et al. (1970), (see Figure 24). We can attribute these disparities to the presence of aerosols. For example, an aerosol distribution of the type marked (a) in Figure 28, would yield a good agreement between our results and those deduced from ionization measurements of Ikebe et al. (1970) within the tropospheric heights. This suggests the possibility that small ion density

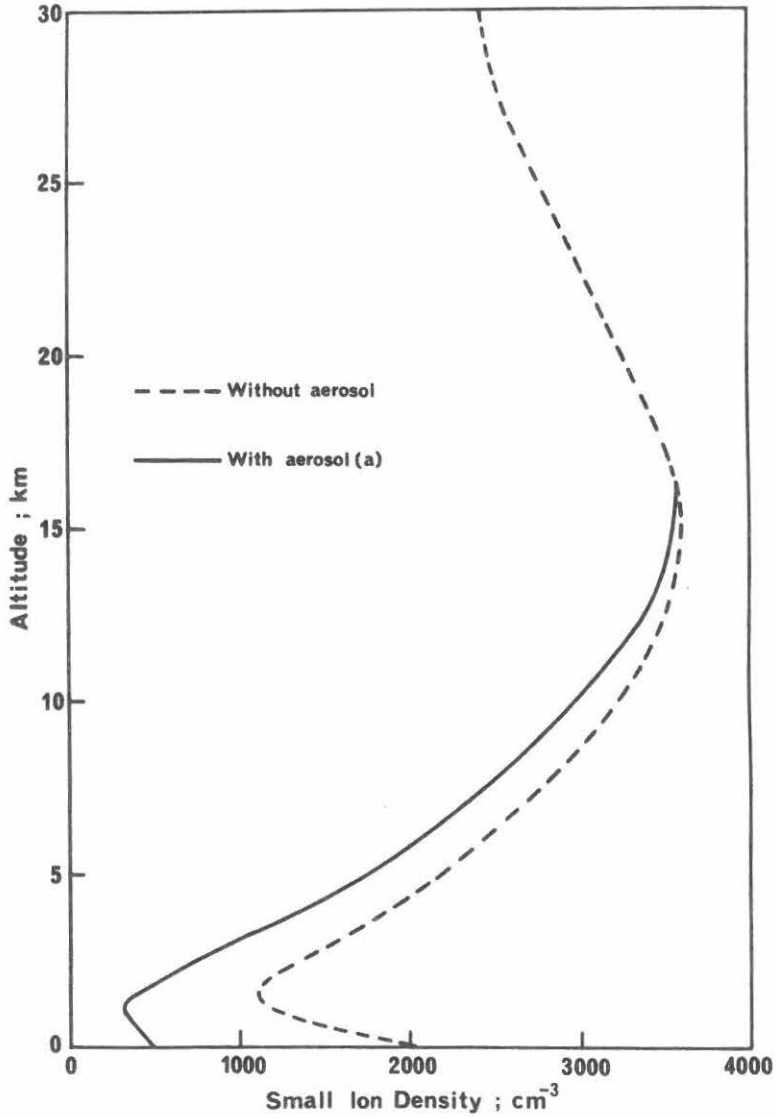

Figure 29. Illustrating the influence of aerosols on the small ion density. The full and broken lines show the ion density profiles with and without the aerosols. The aerosol number density profile assumed for these calculations is that shown in Figure 28(a). See text for details.

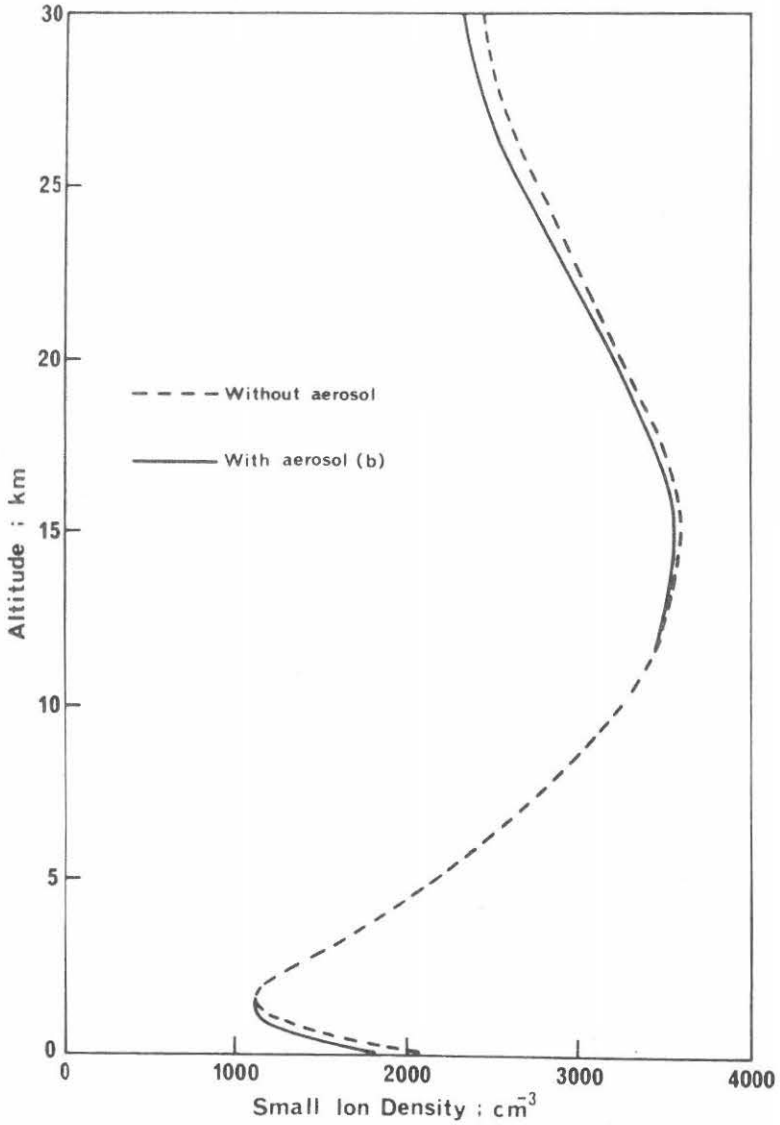

Figure 30. Same as Figure 29, for the aerosol number density profile shown in Figure 28(b).

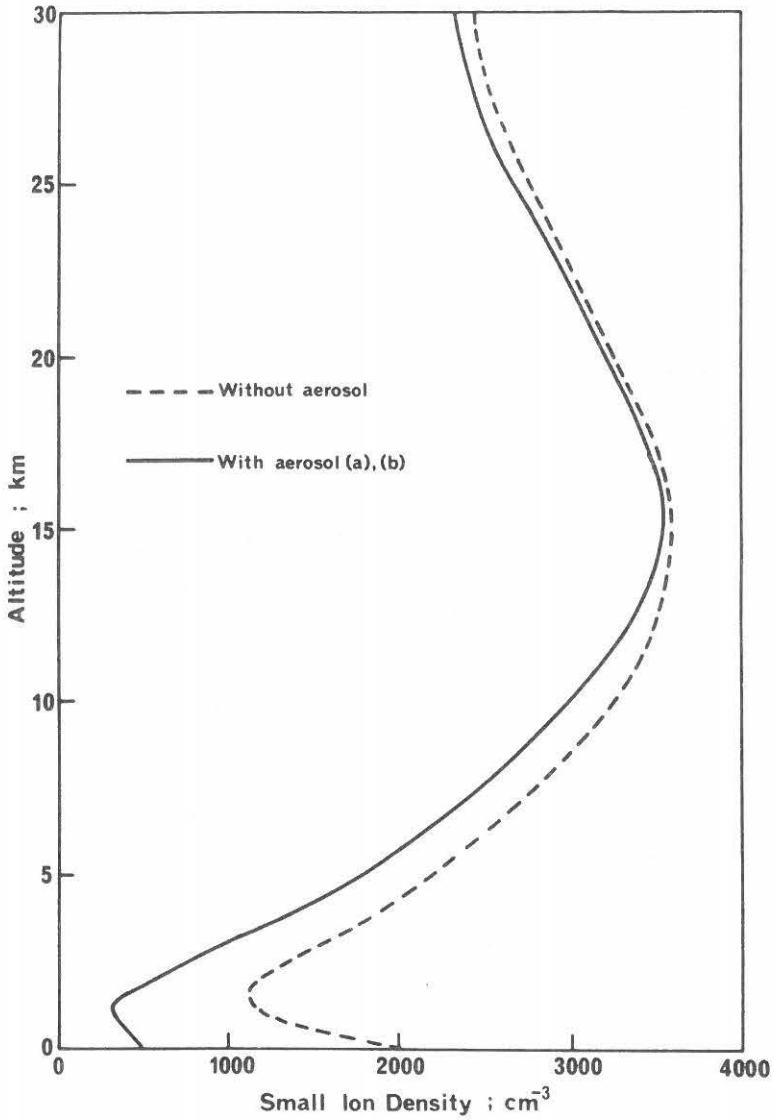

Figure 31. Same as Figure 29, when both types of aerosols distributions shown in Figure 28(a) and (b) are simultaneously present.

measurements at lower altitudes could be used to trace the existence of aerosols. The observed decrease in ion density at stratspheric heights, which contrasts with the theoretical predictions, cannot be so easily explained in terms of the aerosol distributions so far described. Recently, Rosen (1968) has reported aerosol densities greater than his previous measurements (Rosen, 1964) by a factor of 5 to 10 in the equatorial region at altitudes in the range of 15-25 km. Further, as we can see from equation (22), aerosols of larger radii would influence the ion density more markedly than those of smaller radii, for a given number density. Thus existence of a few 'large' aerosols might have dramatic effects of the small ion density. Also, Mohnen (1971) suggests the possible formation and existence of metastable condensation embryos in the upper atmosphere. According to Rosen (1967) one micron diameter aerosols in the equatorial stratosphere could be easily produced by coagulation of 0.05 micron diameter aerosols in the presence of trace gas molecules of 10^{-12} g/cm^3 concentration. He further suggests that horizontal eddy diffusion can easily account for the maintenance of the aerosol concentrations at higher latitudes. If, any or all of, the above suggested mechanisms were to be operative, the ion densities at stratospheric heights could surely be influenced to a marked degree.

2.2.5. The possible role of atmospheric ozone :

In this section, we study the possibility that, under certain circumstances, the atmospheric ozone might influence perceptibly the electrical state of the stratosphere. This study

assumes certain ion-chemical reactions to be effective and is based on the schemes worked out by Fehsenfeld et al. (1967,1968) and Mohnen (1971).

Cosmic rays, which are the main source of ionization in the stratosphere, are thought to create primary positive ions and free electrons. The primary positive ions will lead to the formation of stable positive ions like $H^+(H_2O)_n$ (e.g. Mohnen, 1971). Following Fehsenfeld et al. (1967), we write a simplified version of the possible processes for electron loss and positive ion formation as,

$$\text{Cosmic rays} \rightarrow \begin{cases} O_2^+ \ , \ N_2^+ \rightarrow X^+ \\ e \rightarrow O_2^- \rightarrow X^- \end{cases}$$

where, X^+ and X^- are the most probable positive and negative ions respectively. The following equations are obtained for the steady state :

$$\left.\begin{aligned} q &= \alpha_1 n_2^- n_x^+ + \alpha_2 n_x^- n_x^+ \\ q &= \alpha_1 n_2^- n_x^+ + \alpha_3 n_2^- n_3 \\ n_x^+ &= n_2^- + n_x^- \end{aligned}\right\} \qquad (26)$$

where, n_2^-, n_x^-, n_x^+ and n_3 are the densities of O_2^-, X^-, X^+ and O_3 respectively, and q is the rate of ion production. A reasonable profile for q could be that shown in Figure 25. The recombination coefficients α_1 and α_2 can be written as,

$$\left.\begin{aligned} \alpha_1 &= \alpha_1' + \alpha_1'' \, n_M \\ \alpha_2 &= \alpha_2' + \alpha_2'' \, n_M \end{aligned}\right\} \qquad (27)$$

where, n_M is the number density of the third neutral molecule in three body collisional reactions. The rate coefficient α_3 of the charge exchange process from O_2^- to X^- was given by Fehsenfeld et al. (1967) experimentary, and by Bragin et al. (1967) hypothetically. From the above equations, we obtain the cubic equation,

$$\alpha_2\alpha_3(\alpha_2-\alpha_1)n_3(n_x^-)^3 - \alpha_2\{(\alpha_3 n_3)^2-\alpha_2 q\}(n_x^-)^2 - 2\alpha_2\alpha_3 q n_3(n_x^-) + (\alpha_3 n_3)^2 q = 0 \qquad (28)$$

which can be solved for n_x^- assuming that all other quantities are known. The values of n_3 whose profiles are shown in Figure 32, are taken from the Handbook of Geophysics (1961), and correspond nearly to the same latitude where our balloon measurements of ion density were carried out. The following rate coefficients are perhaps reasonable :

$\alpha_1' = 1.0 \times 10^{-8}$ cm^3/sec. (Nawrocki,1961)

$\alpha_1'' = 5.0 \times 10^{-26}$ cm^6/sec.
$\alpha_2' = 1.0 \times 10^{-7}$ cm^3/sec.
$\alpha_2'' = 5.0 \times 10^{-25}$ cm^6/sec.
$\alpha_3 = 1.0 \times 10^{-15}$ cm^3/sec. (Bragin et al,1967)

Taking these values, n_x^- is obtained from (28) and n_x^+ from (26). It may be mentioned here that Fehsenfeld et al.(1967) gave a value α_3 which is supposed to be valid at the region of 70 km altitude, while the value given by Bragin et al.(1967) is believed to apply to the region between 10-60 km. Hence we have preferred the latter in the present calculation for the strato-

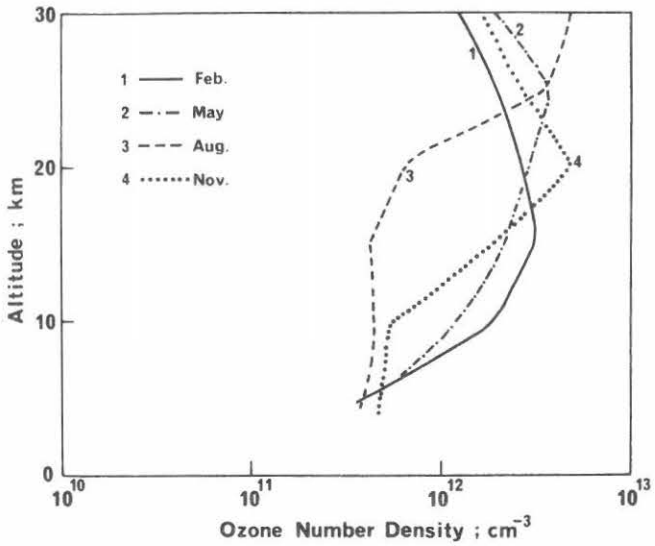

Figure 32. Ozone number density profiles (Handbook of Geophysics, 1961).

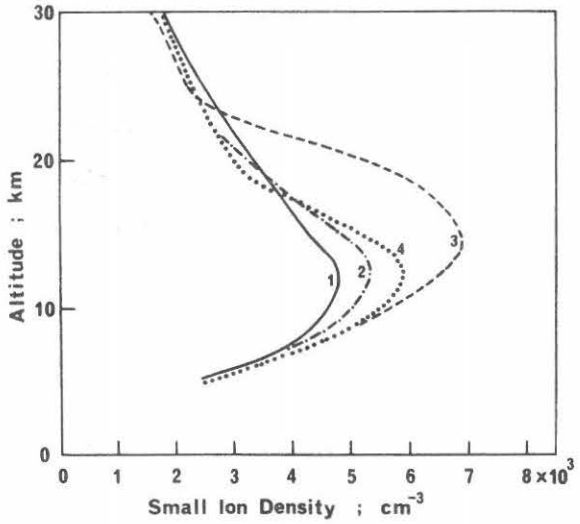

Figure 33. Computed ion density profiles corresponding to ozone density profiles shown in Figure 32.

sphere. The final profiles thus obtained for different months are shown in Figure 33, indicating the significant influence of ozone profiles (Figure 32) on the ion density distributions (Figure 33). All the ion density profiles show a remarkable decrease above about 13 km and thier maxima vary with the number density of ozone. We therefore suggest that one of the possible reasons for differences between the individual measurements of small ion density shown in Figure 24 is that the ozone density profile might have changed.

2.3. Summary and Conclusions :

With a view to improve our understanding of the electrical state of the atmosphere, measurements of small ion density and electric conductivity from the ground to the stratosphere have been made during the period 1969 to 1970 by means of balloon borne instruments.

The small ion density in the lower regions of the atmosphere showed sudden changes at certain altitudes which seem to correspond to temperature inversions in all the cases. A careful check of the relevant meteorological data with the help of the emagrams has revealed that the during the period of balloon flights, conditions conducive to the existence of different air masses are likely to have existed in these regions.

The daytime and nighttime profiles have been found to agree well, except for the following features : (a) The daytime peak occurs at a height greater than that for the nighttime by about 2 km. (b) The nighttime profile shows a second peak around 30 km. All the profiles however show a remarkable decrease

in ion density in the altitudes between about 16 and 25 km.

The observed profiles have been compared with those calculated from the ionization measurements, assuming ionization equilibrium with small ions. The influence of aerosols on the ion density profile has been investigated to account for the disagreement between the measured and calculated profiles. The results of calculation suggest that the influence of atmospheric aerosol can explain the disagreement only within the troposphere. No definitive conclusions could be reached regarding the disagreement at stratospheric heights.

The results of small ion density obtained at a constant level of about 30 km seem to show a decreasing tendency towards the dawn hour.

As one of the possible factors controlling the small ion density in the stratosphere, the influence of atmospheric ozone has been investigated. The results of calculation suggest that the variation in atmospheric ozone, under suitable conditions, could cause similar variations in the measured ion density profiles.

CHAPTER III

THE LAND-TO-OCEAN TRANSITIONAL BEHAVIOR OF THE ATMOSPHERIC ELECTRIC PARAMETERS

3.0. Introduction :

It is generally believed that the measurements of electric parameters of the atmosphere over the oceans will lead to a better understanding of the various physical processes involved than similar measurement over land masses. For example, the electric field measured at mid-ocean,rather than over land is more likely to yield a better insight into the global atmospheric phenomenon. In fact, the results of such mid-ocean measurements form the bedrock on which the concept of the global atmospheric electric circuit is based. Ever since the famous Carnegie expeditions during 1915 and 1929, many attempts have been made to measure the electric properties of the atmosphere over the Atlantic and North Atlantic Oceans. Among such attempts mention may be made of those by Parkinson and Weller (1953), Sagalyn (1958) and Mühleisen (1967). Similarly, Ruttenberg and Holzer (1955), Takagi and Kanada (1969,1970) and Morita (1971) have reported some measurements of electric field and conductivity in the atmosphere over the Pacific Ocean.

It was emphasised in Chapter I that the main purpose of mid-ocean measurements is to obtain values of the various parameters which could be considered as being typical of the undisturbed atmosphere. Even though atmospheric pollution originates mainly over land areas, its influence on the electrical state of the atmosphere over oceans could, under suitable conditions, be felt for considerably long distances from shore. For example, Takagi and Kanada (1969) have reported that the daily averages of the electric field measured within a distance of about 150 km from shore consistently tend to be higher than those obtained

at greater distances. Recently, Misaki and Takeuti (1970) have studied the behavior of electric field and conductivity over the Pacific Ocean and the Sea of Japan, and have discussed the extent to which land pollution affected their measurements. More recently, Cobb and Wells (1970) have made measurements of electric conductivity in the global atmosphere over the oceans, and have discussed the correlation between their measurements and the global atmospheric pollution. They have reported that the conductivity measured in the Indian Ocean, even several hundred miles from land, appeared to be affected by airborne pollution from South Asia. Similar results indicating the extent to which continental influence can be traced in sea surface measurements have been reported by other workers (e.g.,Mühleisen,1967).

It thus becomes necessary to study systematically how far influence of the continental pollution continues to exist before one can reasonably assume that a particular set of ship-borne measurements represents the atmospheric conditions unaffected by the land pollution. Thus, the variation of electric parameters could become a measure of the extension of land pollution to the oceanic atmosphere and a systematic measurements of these parameters would acquire new meaning in the fields of meteorology, atmospheric electricity and in "Aerosol Climatology". Though there are many individual measurements of electric parameters over the oceans, systematic studies of the transitional behavior of the electric parameters of the atmosphere from the land to the ocean are very few. For such studies, of course, combined measurements of electric parameters and aerosol concentration would be of great value.

Few measurements seeking to clarify the interaction between ionization, small ions and aerosols over the oceans have been reported (Ikebe,1970 and Morita et al.,1971). 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 (1968) and Hogan et al. (1967). Similarly, Ohta (1950) and Ohta and Ito (1971) have reported some measurements of condensation nuclei over the Sea of Japan and the Pacific Ocean. Junge (1969) has made a systematic study of aerosols at cape and described three types of aerosols in Pacific air masses of middle and lower latitudes. Recently, Junge and Jaenicke (1971) have made measurements of aerosol size distribution in the atmosphere over the Atlantic Ocean and have discussed atmospheric background aerosols. The sea surface measurements have the advantage of being amenable to a simpler interpretation because only the ion pair produced by cosmic rays need to be taken into account, thus eliminating the necessity of considering the contribution of the radioactive elements in the earth's crust (Ikebe, 1970).

In view of the above, the author has undertaken a systematic study of the nature of the variations of the electric parameters of the atmosphere from land to the oceans. For this purpose several expeditions have been carried out, the details of which are described in the following sections.

3.1. Collection of Data :

Under the auspices of the Ocean Research Institute, University of Tokyo, and Tokai-Kisen Co. Ltd., nine expeditions have been carried out during the years 1968 to 1971. The relevant

data pertaining to these expeditions are shown in Table 3. Expeditions I and III which charted the oceanic regions of the South and North Pacific, covered such long distances from the shore that the measurements could be considered to represent the global aspects. On the other hand, the courses of the other expeditions concern mainly the regions which could be influenced directly by the land pollution. The courses of the vessel during these expeditions are shown in Figures 34 and 35.

The small ion density and electric conductivity have been measured by the ordinary aspiration Gerdien condensers. The basic design of the instruments is the same as that used for balloon measurements (see Figure 10(b)), except for some minor changes in certain dimensions. The applied voltages to the condensers were so arranged that for the rate of air flow of 300 liters per minute, the ion density condensers would be saturated for all ions mobility greater than 0.4×10^{-4} m^2/volt.sec., and the conductivity condensers would be unsaturated for all ions of mobility less than 4×10^{-4} m^2/volt.sec. As can be seen from Table 3, the conductivity measurements have been made during all the nine expeditions, while the small ion density results are available only for two expeditions. Even though the conductivity measurements alone could be carried out to yield absolute values, simultaneous measurements of conductivity and small ion density were very useful in checking the systematic errors of the instruments. This is because the average ionic mobility determined from these two parameters is reasonably 'invariant' under the conditions at ground level. An example of actual record obtained from Expedition III is shown in Figure 36. The zero level of the condenser was checked automatically

Table 3. Data describing the scope of the expeditions

Expedition NO.	Period	Latitude & Longitiude	Parameters measured
I	Nov. 14, 1968 to Feb. 27, 1969	$35^{\circ}N-70^{\circ}S$ $140^{\circ}E-170^{\circ}W$	Electric field Conductivity
II	June 30, 1969 to July 10, 1969	$31^{\circ}N-35^{\circ}N$ $127^{\circ}E-140^{\circ}E$	Electric field Conductivity Small ion density
III	Feb. 3, 1970 to Mar. 5, 1970	$10^{\circ}N-40^{\circ}N$ $140^{\circ}E-160^{\circ}E$	Electric field Conductivity Small ion density Condensation nuclei
IV	Oct. 22-24, 1971	$33.5^{\circ}N-35^{\circ}N$ $140^{\circ}E-142^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient
V	Oct. 6-7, 1971	$33^{\circ}N-35^{\circ}N$ $139.5^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient
VI	Oct. 12, 1971	$33^{\circ}N-35^{\circ}N$ $139.5^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient
VII	Nov. 17-18, 1971	$33^{\circ}N-35^{\circ}N$ $139.5^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient
VIII	Dec. 8-9, 1971	$33^{\circ}N-35^{\circ}N$ $139.5^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient
IX	Dec. 16, 1971	$33^{\circ}N-35^{\circ}N$ $139.5^{\circ}E$	Conductivity Condensation nuclei Diffusion coefficient

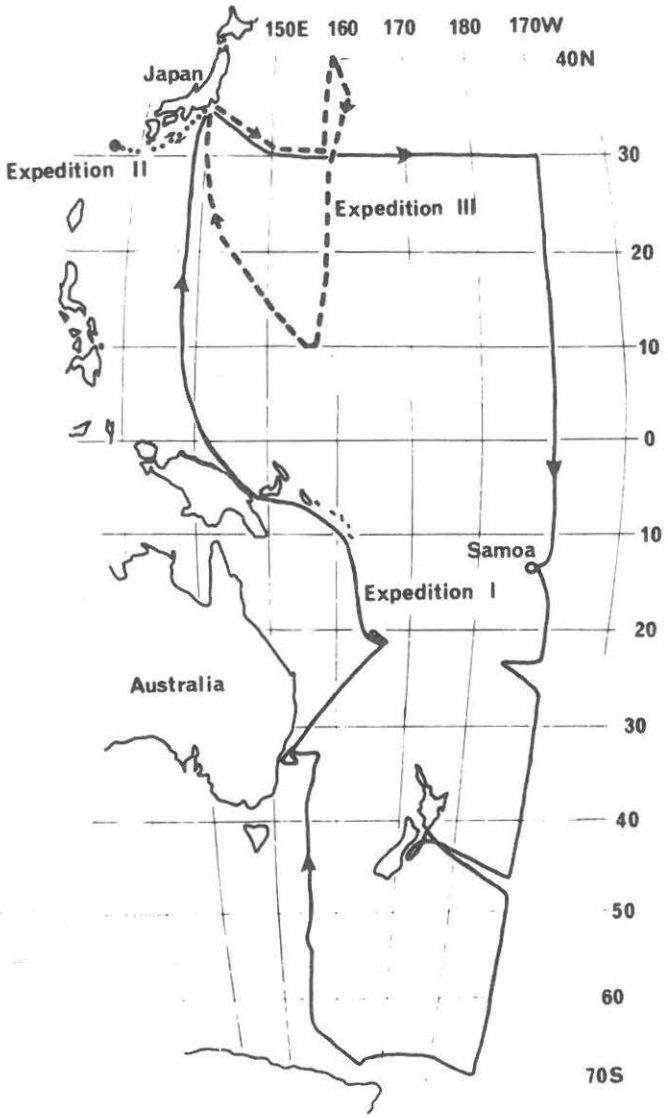

Figure 34. The routes followed during the three expeditions. The legend is as follows : Expedition I ———, Expedition II ·····, Expedition III -----. For other details, see Table 3.

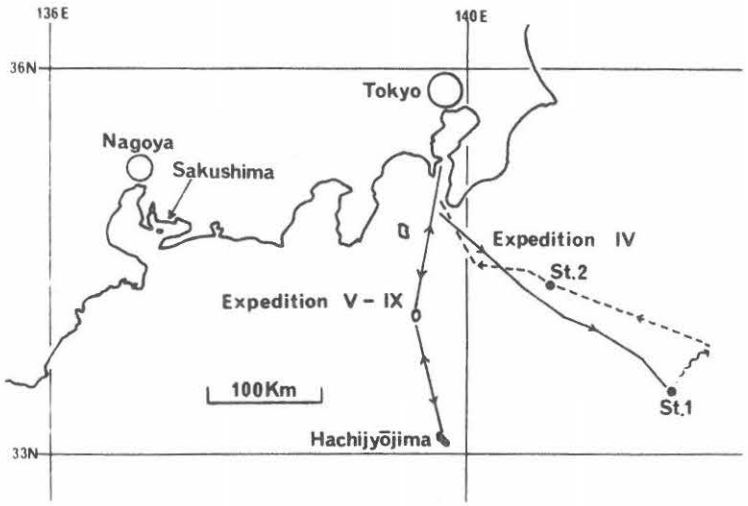

Figure 35. The routes of Expeditions IV to IX. For other details, see Table 3.

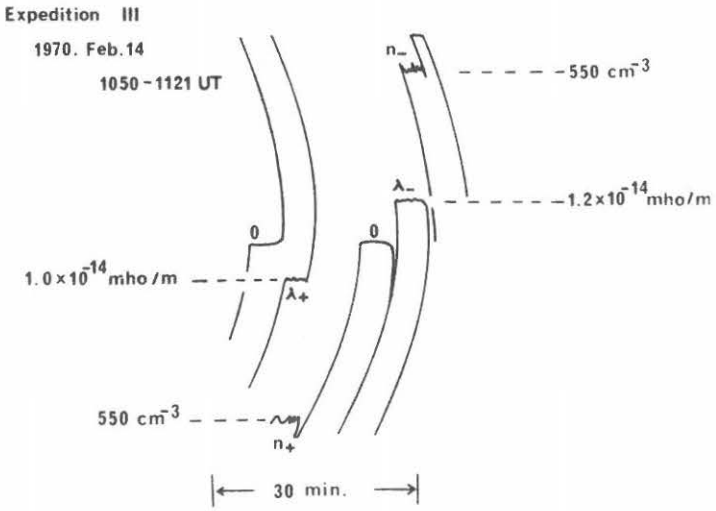

Figure 36. An example of the actual record obtained during Expedition III, showing the small ion density (n) and electric conductivity (λ).

every 12 minutes by applying zero voltage to the outer electrode of the condenser.

The condensation nuclei and their diffusion coefficient were measured during Expeditions IV-IX. 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 0.90 milli-micron. The calibration curve obtained by Nolan and Pollak (1946) was used for the estimation of concentration of nuclei from the measured value of extinction. In the present case the electronically improved technique in which the peak of extinction was held and amplified electronically, was employed. This enabled low concentrations to be measured. The performance of the counter was compared with those used by other workers like Kawano and his collaborators in the Nagoya University. The results obtained simultaneously and independently with the different counters showed good agreement. The diffusion coefficient of nuclei was measured by the diffusion battery of the type developed by earlier workers (Nolan and Nolan, 1938). The dimensions of the metal diffusion battery used are :

Distance between plates : 0.10 cm
Width of plates : 20 cm
Length of plates : 40 cm
Channel number : 30

The total flow rate in the present work was 3 liter per minute. The diffusion coefficient of nuclei was estimated by using

Gormley's formula (Nolan and Nolan, 1938) and the equivalent radius corresponding to the diffusion coefficient measured was determined by using Stokes-Chunningham's relation.

One of the main difficulties encountered in practice was the influence of pollution from the ship itself, which, under suitable wind directions, could make the data unreliable. Regarding this, Misaki et al. (1972) have discussed the influence of the contamination from the ship on the conductivity measurements in the Pacific Ocean. To avoid such influence, in the present measurements, the air intakes of the instruments were stuck out of the bridge, and only the data obtained when the wind blew directly from the open sea to the intakes were used in the analysis. Since wind measurements have been made continuously, we could make it reasonably certain that the data chosen for the analysis have not been contaminated directly by the ship's exhaust.

3.2. Results and Discussion :

3.2.1. The extension of land pollution to the atmosphere over the ocean and its influence on the sea surface measurements :

Figure 37 shows an example of the variation of conductivity with distance from shore. For convenience, the courses of the vessel and sky conditions are also shown. The minimum distance (closed circles) from the ship to the shore is indicated in the graph. It is at once evident from Figure 37 that a reasonably good correlation exists between the distance and conductivity. The vessel at that time was heading towards Wellington

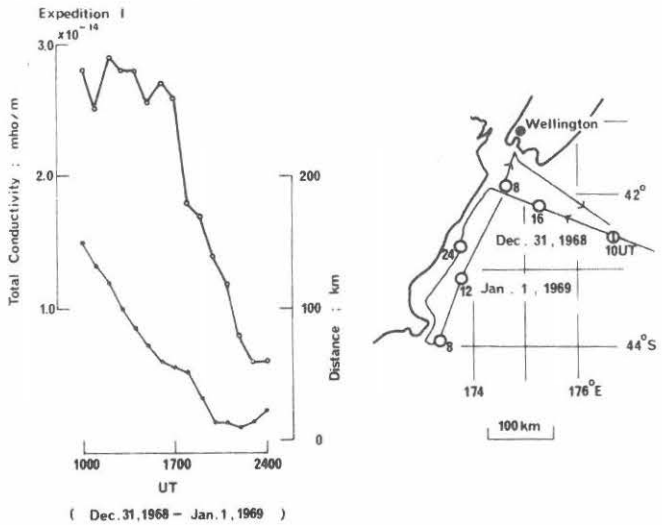

Figure 37. Conductivity measurements in the neighborhood of New Zealand. The distance shown is the minimum distance from shore. The route of the cruise, the relevant days and sky conditions are also shown. The open circles present the total conductivity, while the closed circles indicate the distance.

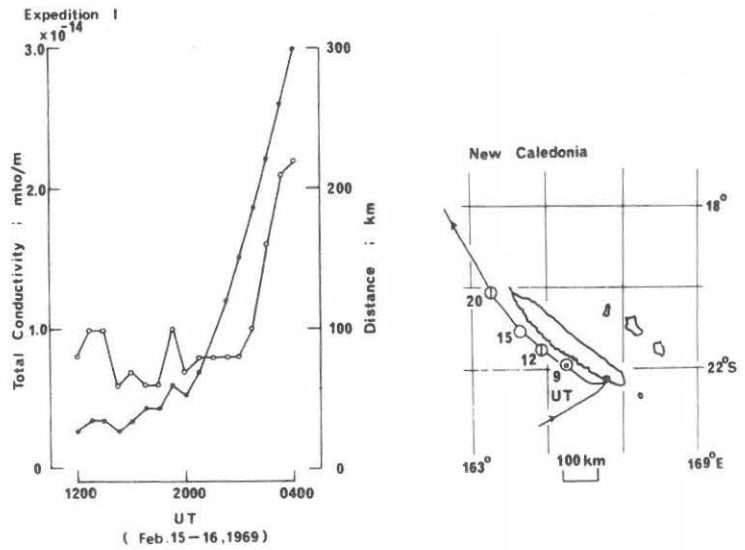

Figure 38. Measurements in the neighborhood of New Caledonia. Notation is same as in Figure 37.

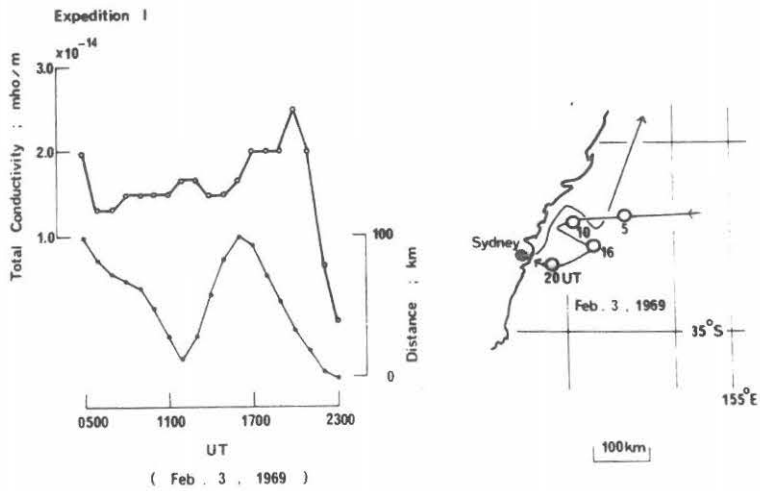

Figure 39. Measurements in the neighborhood of Australia. Notation is same as in Figure 37.

in New Zealand. It seems reasonable to suggest that the dependence of conductivity on distance is probably due to the extension of land pollution to these distances. It seems that for distance greater than about 100 km, the change in the conductivity with distance is very small. At least, within the experimental error, we can say that the conductivity tends to "level off" at a distance of the order of 100 km.

The marked dependence of the conductivity on distance from the shore is also found in the neighborhood of New Caledonia. The conductivity apparently depends on the distance upto about 300 km as shown in Figure 38. Considering the industrial potential of New Caledonia, it is however difficult to appreciate that the distance-dependence of conductivity shown in Figure 38 is due to the pollution originating on the main land.

It may be mentioned here that Misaki and Takeuti (1970) have also reported a similar dependence of conductivity on distance, from their measurements in the Pacific Ocean and the Sea of Japan. Assuming that the pollution originating on land is uniform distributed in space, despite of the actual non-uniform distribution of industries, Misaki and Takeuti (1970) interpreted their results in terms of diffusion. Such an interpretation, however, is not likely to be valid if we consider other complications like the movement and the life-history of air masses travelling from the continents to the sea. As an example, we show in Figure 39, the dependence of conductivity on distance near Sydney. We see that conductivity varies with distance in a very complicated manner. Evidently, one would have to consider many other factors like the movement and the life of the continental air masses.

In fact, in many cases the dependence of the electric parameters on the distance from the shore is far more complicated than that shown in Figures 37 and 38.

In Figure 40 we show another example of the variation of the conductivity with distance from shore, which is the results of Expedition II. The points marked A and B in the figure indicate the average values of the conductivity obtained from data recorded continuously for over a week at these two fixed points. The bars over the points A and B show the spread in the experimenta data. The results of small ion density obtained simultaneously at these two points also showed a similar behavior. There was practically no dependence of the conductivity on distance and measurements over land and ocean showed no significant difference. During the period the weather conditions were very unstable and complicated, and the conductivity measurements seem to reflect these weather conditions. A representative surface weather chart during the expedition is shown in Figure 41. The values of conductivity consistently tended to be low throughout the expedition, while the nucleus concentration measured simultaneously tended to be high (Shimo et al., 1972). Even though the measurements made at a distance of about 300 km from shore, did not seem to represent the typical undisturbed atmosphere over the ocean.

On the other hand, as shown in Figure 42, the results obtained in the neighborhood of Tokyo show marked dependence of the small ion density and conductivity on the distance from shore. Both the small ion density and conductivity increases rather smoothly with the increase in distance and finally

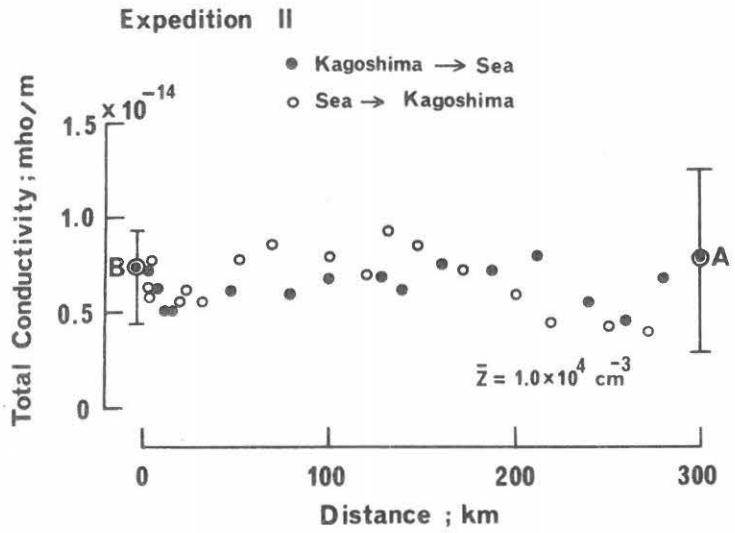

Figure 40. Showing the variation of the total conductivity with distance in the neighborhood of Japan. Z denotes the average nucleus concentration during this expedition (Ikebe,1970). See text for an explanation of points of A and B.

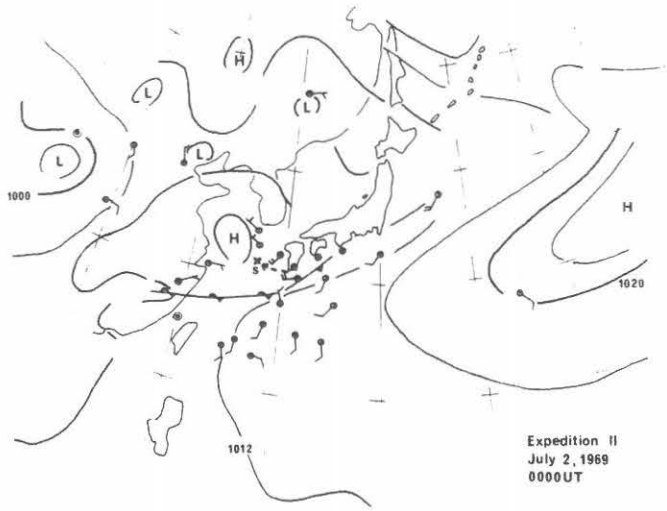

Figure 41. A representative surface weather chart during Expedition II. S denotes the ship's position.

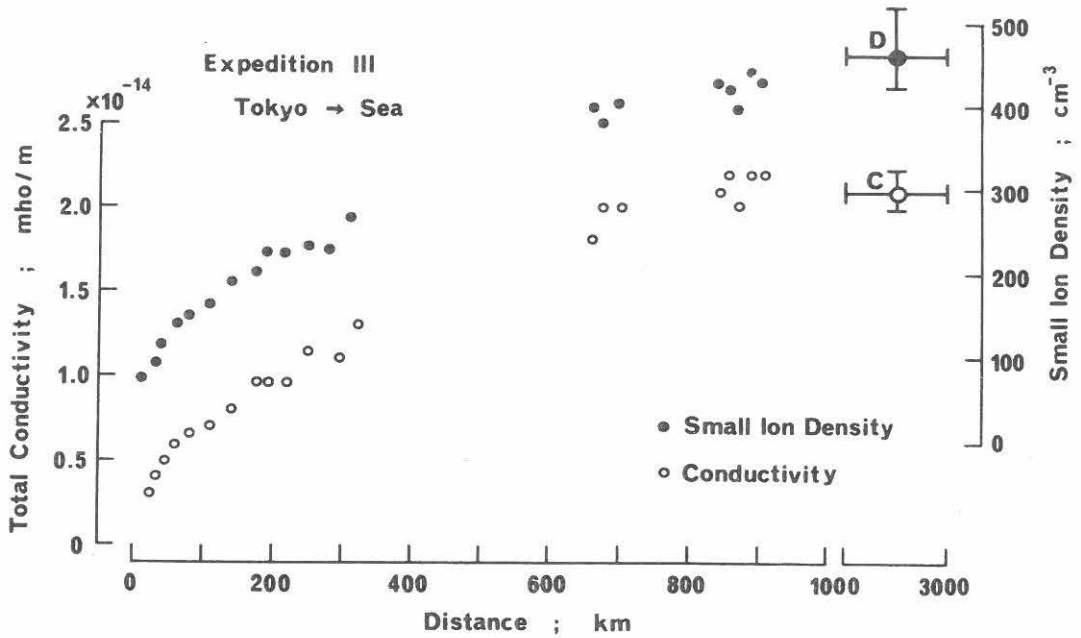

Figure 42. Showing the dependence of total conductivity and small ion density on the distance from shore. Measurements made in the neighborhood of Japan. Open circles refer to the conductivity while closed circles indicate the small ion density. See text for an explanation of points of C and D.

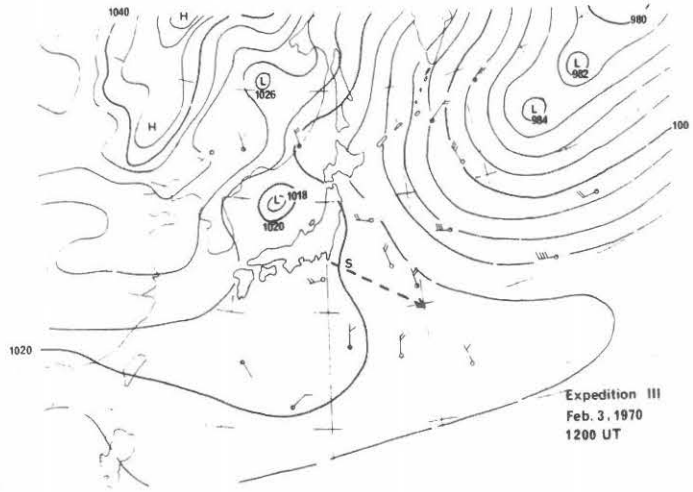

Figure 43. A representative surface weather chart during Expedition III. S denotes the ship's position.

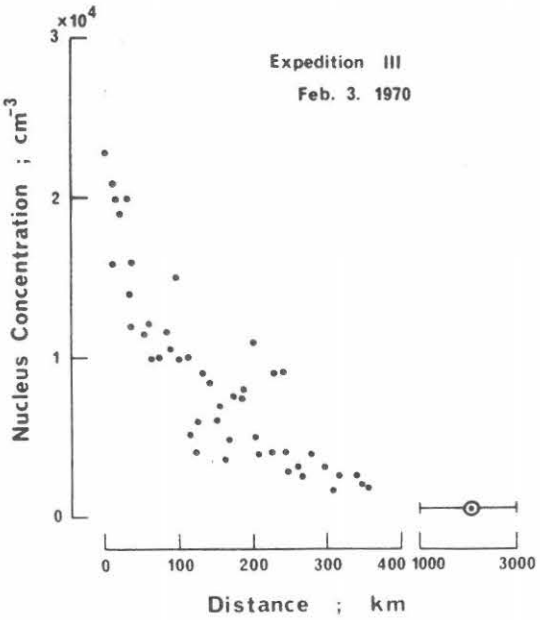

Figure 44. Showing the variation of nucleus concentration with distance in the neighborhood of Japan. Data obtained from Expedition III.

attain nearly constant values at a distance of about 1000 km. Apparently, these results are consequence of high industrial activity in the neighborhood of Tokyo. Further, the meteorological conditions prevalent at these times were favourable for the pollution to spread over large oceanic regions. A representative surface weather chart during the period is shown in Figure 43. The marked dependence on the distance of the nucleus concentration simultaneously measured is also shown in Figure 44. The points marked C and D in Figure 42 and the points with flags in Figure 44 indicate the values obtained at distances of the order of 1000 km or more from shore. Figure 45 shows the variation of conductivity with distance obtained from the results of Expedition IV. The stations marked St.1 and St.2 are the same as shown in Figure 35 which depicts the route of Expedition IV. In Figure 45 open circles refer to measurements made on the 'forward' journey from Tokyo to St.1 while the closed circles refer to the 'return' journey from St.1 to St.2. It is evident from Figure 45 that the variation of conductivity are significantly different in the two cases. The conductivity consistently increased on the forward journey tending to level off between 150-200 km, while on the return journey when the vessel approached the shore the next day after drifting at St.1, it decreased almost linearly with distance. The corresponding variations in nucleus concentration in the two cases are shown in Figure 46 in which the points marked T and E indicate the values obtained at Tokyo-and Tateyama-bays respectively. The bars indicate the spread in the experimental data. Though the nucleus concentration showed marked variability in the bay regions, it remained sensibly

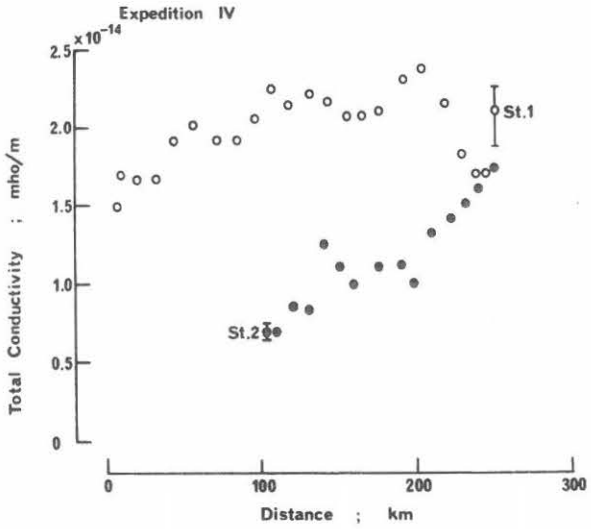

Figure 45. Showing the dependence of conductivity on distance. Open circles refer to values obtained on the 'forward' journey from Tokyo to St.1 while the closed circles refer to those on the 'return' journey from St.1 to St.2. Data from Expedition IV.

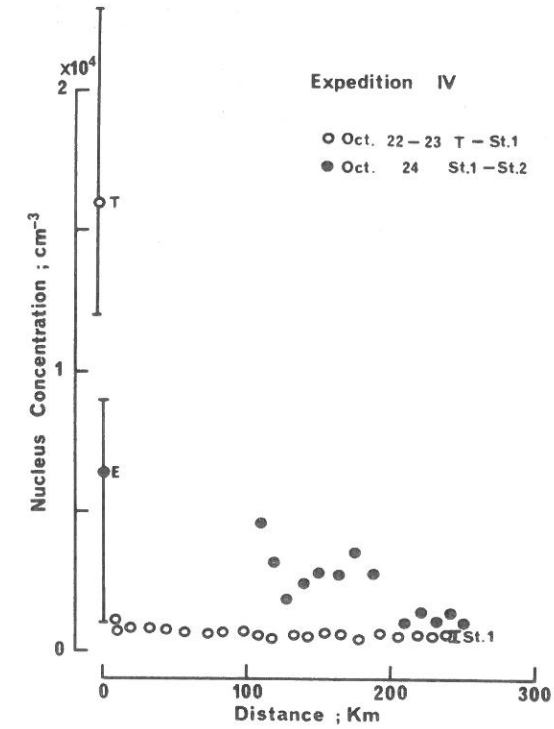

Figure 46. Variations in nucleus concentration corresponding to the conductivity measurements shown in Figure 45. The limits of variation recorded at Tokyo (T) and Tateyama (E) bays are also shown. Other notation is same as in Figure 45.

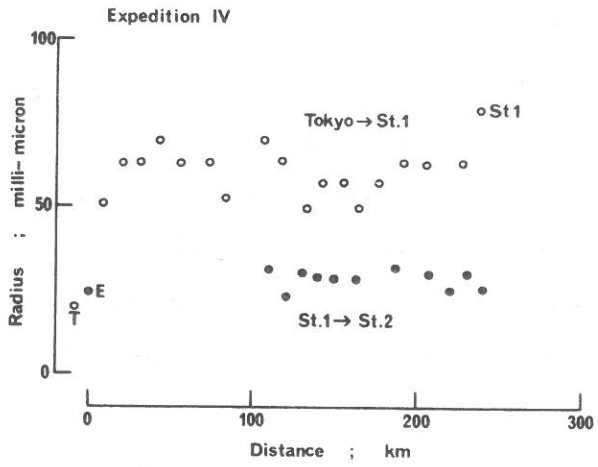

Figure 47. Showing the variation of the equivalent radius with distance. Data from Expedition IV. Other notation is same as in Figure 45.

constant on the forward journey to St.1 (open circles). On the other hand, the return journey measurements (closed circles) showed much variation with an over-all tendency to increase as the shore was approached.

In Figure 47, we show how the equivalent radius, estimated from the diffusion coefficient of nuclei, changed during the forward and return journeys. Once the vessel was out of Tokyo and Tateyama bays, the equivalent radius showed little change with distance even though the values on the forward journey (open circles) were consistently higher than those on the return journey (closed circles).

The results of Expeditions V to IX are summarized in Figures 48, 49 and 50, which show the variation of the conductivity, nucleus concentration and the equivalent radius of nuclei with distance respectively. As seen from Figure 48 which show the results of Expeditions V, VII and IX, conductivity remained low upto a distance of about 100 km after which it increased rapidly (Expeditions V and VII). The nucleus concentration, as can be seen from Figure 49, shows the corresponding over-all decrease with distance even though on some expeditions (e.g.,VI and IX) it exhibits rapid and irregular variations within 100 km. In Figure 50, we show how the equivalent radius varies with distance. As in the earlier figure, data from Expeditions V through IX have been superposed. If we give equal weight to all the points in Figure 50, it follows that the equivalent radius does not depend much on the distance from shore. If, on the other hand, we consider each expedition as being unique in its own way, we find that there are, in general three-different types of variation.

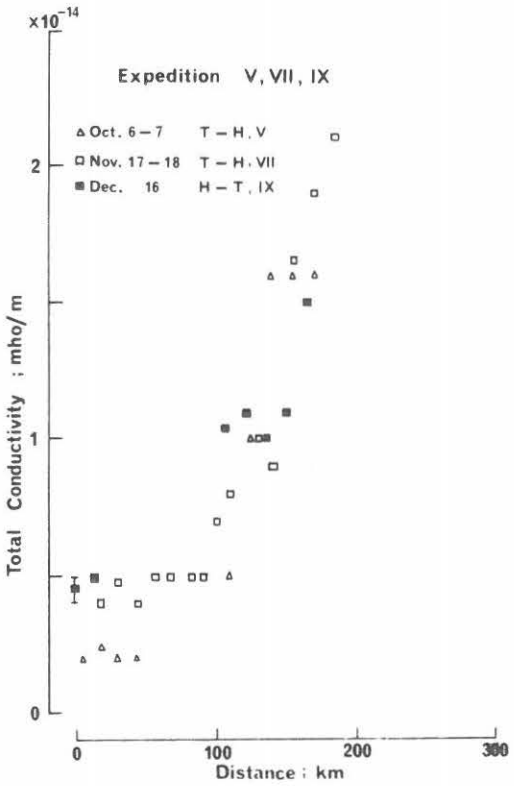

Figure 48. Showing the variation of electric conductivity with distance. Data from Expeditions V, VII and IX have been superposed.

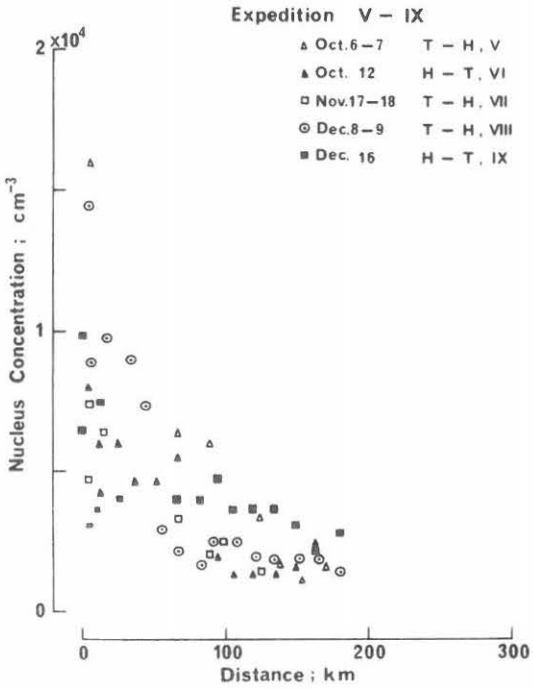

Figure 49. Superposed plot of nucleus concentration data obtained from Expeditions V through IX.

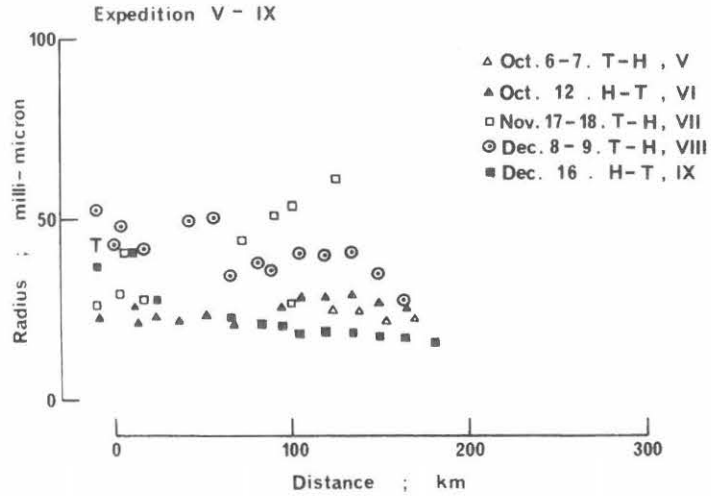

Figure 50. Superposed plot of equivalent radius of nuclei data obtained from Expeditions V through IX.

In the first category, which includes Expeditions VIII and IX, the equivalent radius tends to decrease with the increase in distance. In second (Expedition VII) it tends to increase with the distance and finally, in the third type, there is no perceptible dependence on distance (Expedition VI).

So far we have presented the data obtained on different expeditions without attempting to explain in detail the significant differences between the results of individual expeditions. Indeed, it appears at first sight that there is little systematics in the data. In the next sub-section we attempt to seek a meaningful interpretation of the apparently 'random' results so far described.

3.2.2. Interpretation of the data :

Even though we have briefly mentioned above that land pollution, extending well over large areas of the oceanic atmosphere, might be responsible for the observed dependence of the measured parameters on the distance from shore, we did not present any supporting evidence to show that conditions were indeed favourable for such a spreading of pollution. Moreover, any given set of measurements is a function of the local small-scale meteorological environment, the movement and time-histories of the air masses, etc. We now study these factors in some detail. We first note that the local weather conditions were quite stable during Expeditions IV, VI, VIII and IX but that they were very unstable during Expeditions V and VII. The unstable meteorological conditions sometimes cause scatter in the measured values.

It is of course necessary to consider, apart from local

meteorological conditions, other factors like the movement and the life of air masses to understand fully the results of measurements. Towards this end, it is essential to investigate the air stream-lines and air masses trajectories in a manner similar to that described by Petterssen (1956). Since the surface wind data are much influenced by geophysical features (e.g., friction introduced by the ground surface) it is desirable to choose the wind data at a higher level. It would seem that any level chosen within the exchange layer would serve the purpose since the physical states of the atmosphere at such a level and at the ground level would not be much different. This is because of the fact that the atmosphere within the exchange layer is usually thoroughly mixed by convection. We have therefore chosen 850 mb level as the reference. Typically, this corresponds to an altitude of about 1.5 km.

In Figure 51 are shown the air stream lines at 850 mb level during Expedition III. It is seen from the figure, in which S denotes the location of ship, that the vessel proceeded in a direction nearly parallel to the air stream lines. Thus we might expect that land pollution would, in this case, spread over long distances from shore. Indeed, we note from Figure 42 that both the small ion density and conductivity continue to depend on distance upto about 900 km. We can therefore attribute, the observed dependence of the parameters on distance to the pollution originating on the main-land.

Similarly, the peculiar behavior of the conductivity and nucleus concentration shown in Figures 45 and 46 could be qualitatively explained. We recall that the conductivity values

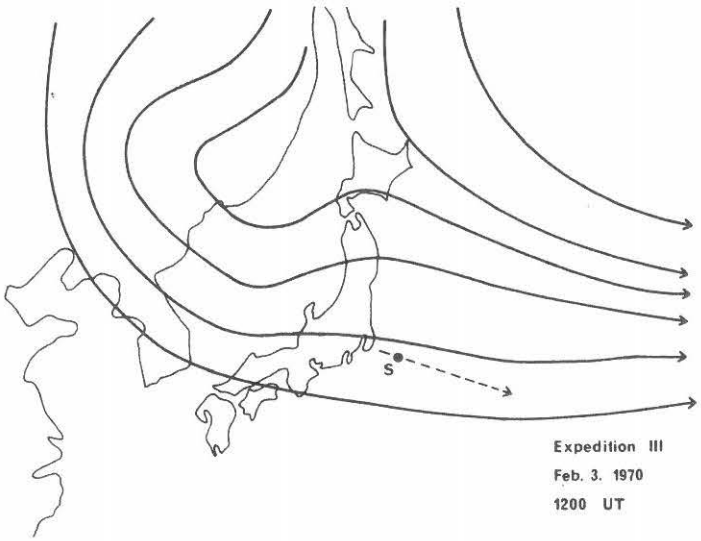

Figure 51. Air stream lines at 850 mb level during Expedition III. S denotes the position of ship.

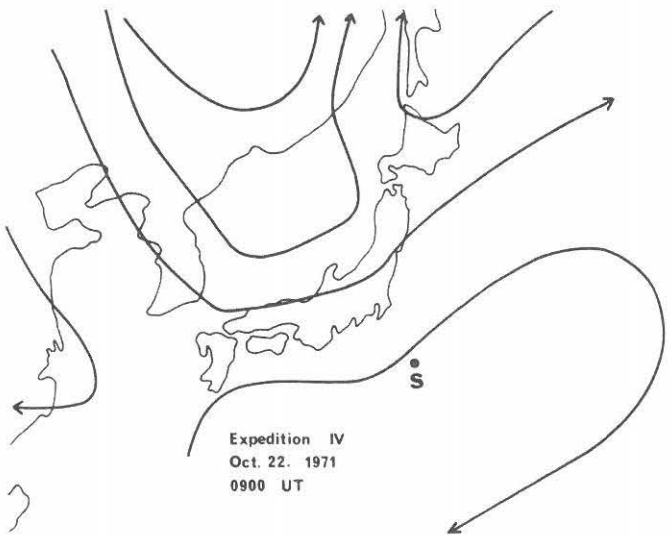

Figure 52(a). Same as Figure 51. Expedition IV.

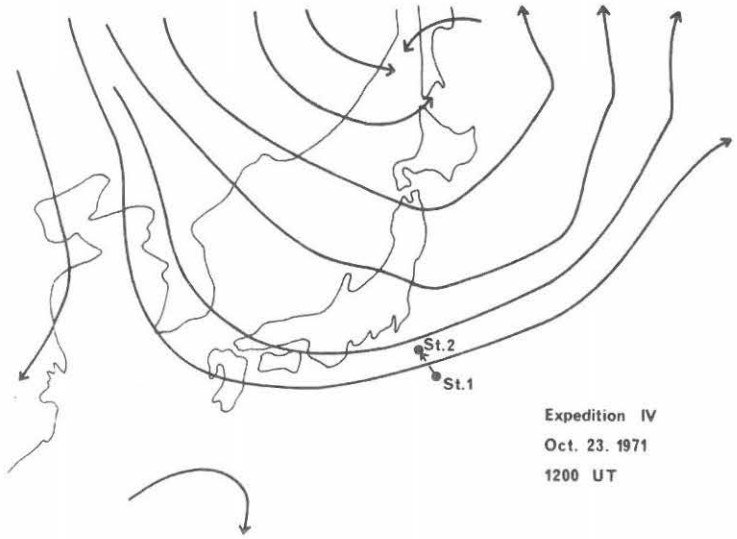

Figure 52(b). Same as Figure 51. Expedition IV.

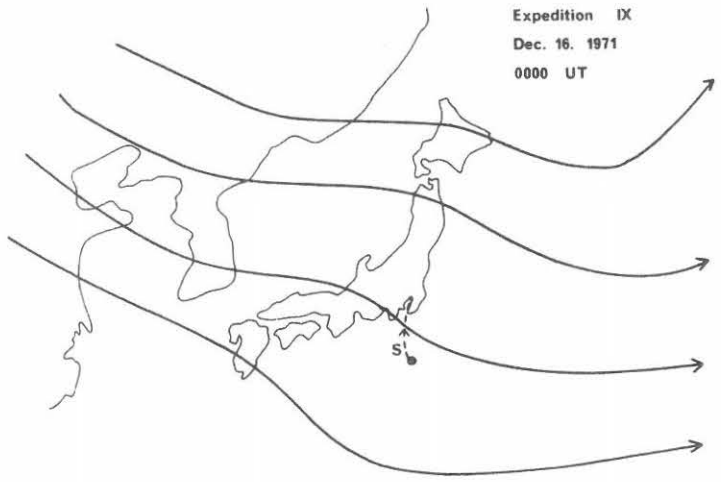

Figure 53. Same as Figure 51. Expedition IX.

measured on the 'forward' journey from Tokyo to St.1 (refer Figure 35) were higher than those measured on the 'return' journey from St.1 to St.2, the reverse being true in the case of nucleus concentration. The relevant patterns of air streams corresponding to these two journeys are shown in Figures 52(a) and (b). We note that during the forward journey the relevant winds came essentially from the sea while on the return journey it came directly from land. Thus we could reasonably expect that the measurements on the return journey were much more influenced by land pollution than those on the forward journey. Thus it seems possible to explain qualitatively the data presented in Figures 45 and 46, in terms of the land pollution affecting the sea-borne measurements.

In the case of Expeditions V through IX the relevant winds came generally from the land masses and the ship's course crossed the stream lines. Roughly speaking, the patterns of air stream lines during these expeditions were all very similar. As an example, we show in Figure 53 the stream line patterns corresponding to Expedition IX. We note the wind was blowing from land and , in a qualitative sense, we can explain the distance-dependence of conductivity and nucleus concentration shown in Figures 48 and 49 in terms of the extension of the land pollution.

Even though the air stream line patterns help in understanding the probable processes responsible for the observed behavior of some of the parameters, they cannot lead to any quantitative estimates. For a more quantitative and physical interpretation of the various features of the data presented above, a knowledge of the time-histories and trajectories of air masses will be

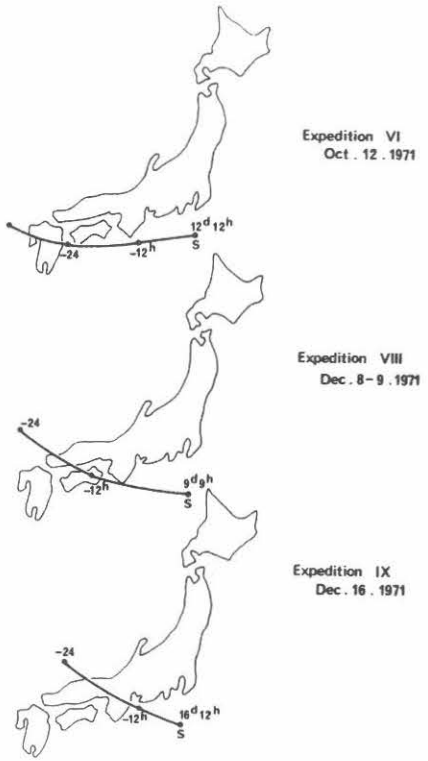

Figure 55. Same as Figure 54. Expeditions VI, VIII and IX.

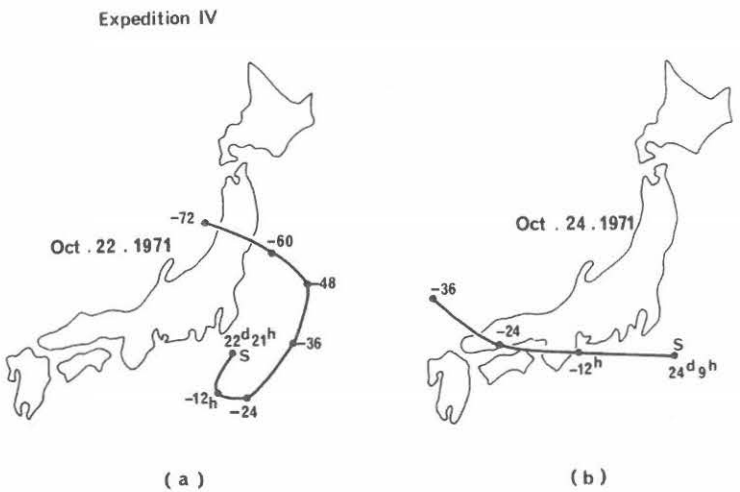

Figure 54(a) and (b). Air mass trajectories in steps of 12h during Expedition IV.

necessary. In Figures 54 and 55 we show a few examples of the air-mass trajectories in steps of 12 hours each. The trajectories in Figure 54 correspond to Expedition IV while those in Figure 55 correspond to Expeditions VI,VIII and IX. Going back to Figures 45, 46 and 47, we find that the data on the forward journey (i.e. open circles in these figures) have been measured in the air masses of long history (i.e.very aged). On the other hand, the measurements on the return journey (closed circles) correspond to air masses of relatively short history. Similarly, we find from Figure 55 that the measurements made on Expeditions VI and IX also correspond to relatively low time-histories of land air-masses.

It would seem desirable, in view of the above, to attempt to interpret the data using the age of the air masses as a parameter. The results of such an analysis are shown in Figures 56 through 58 in which the time axis refers to the age of the air masses. It is at once clear from Figures 56 and 57, which show the dependence of total conductivity and nucleus concentration on time, that age of the air masses is certainly a better parameter than the minimum distance from shore. This fact is clearly brought out by a comparison of Figures 56 and 45. While in Figure 45, for some distances, (above 100 km), the conductivity is "double valued", in Figure 56 we find that the open and closed circles are well separated in time to yield 'unique' values. The 'ambiguity' in Figure 45 is completely absent in Figure 56. We can state from an inspection of Figures 56 and 57 that among the many factors which could influence the conductivity and nucleus concentration measurements, the age of the air masses is vitally

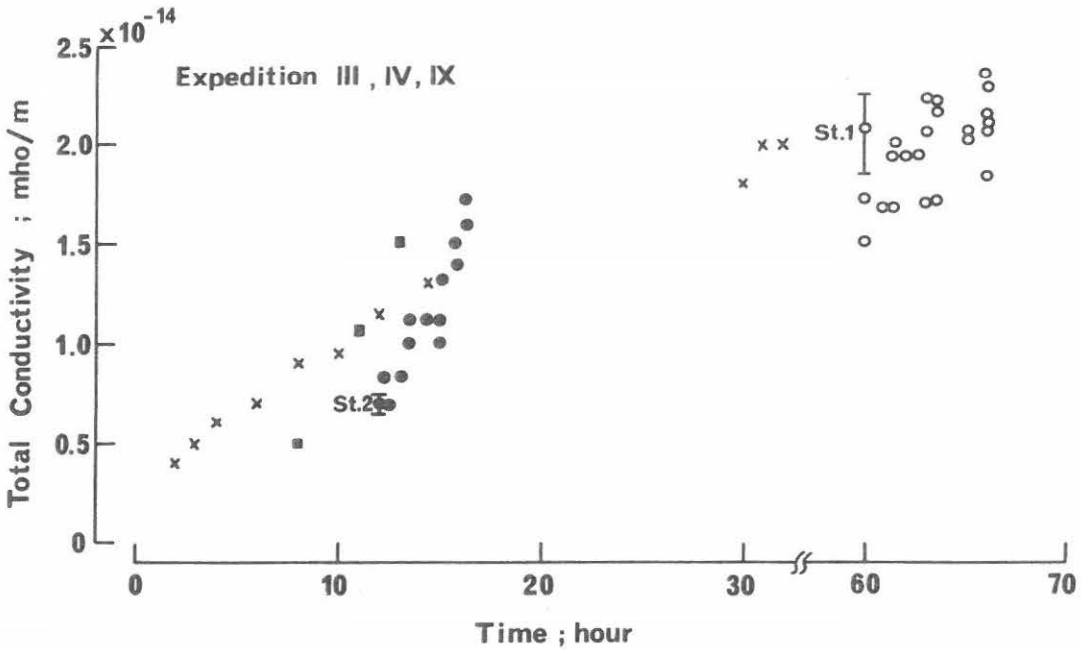

Figure 56. Showing the dependence of total conductivity on the age of air masses. Crosses and filled squares refer to the results of Expeditions III and IX respectively. Other symbols have the same meaning as in Figure 45.

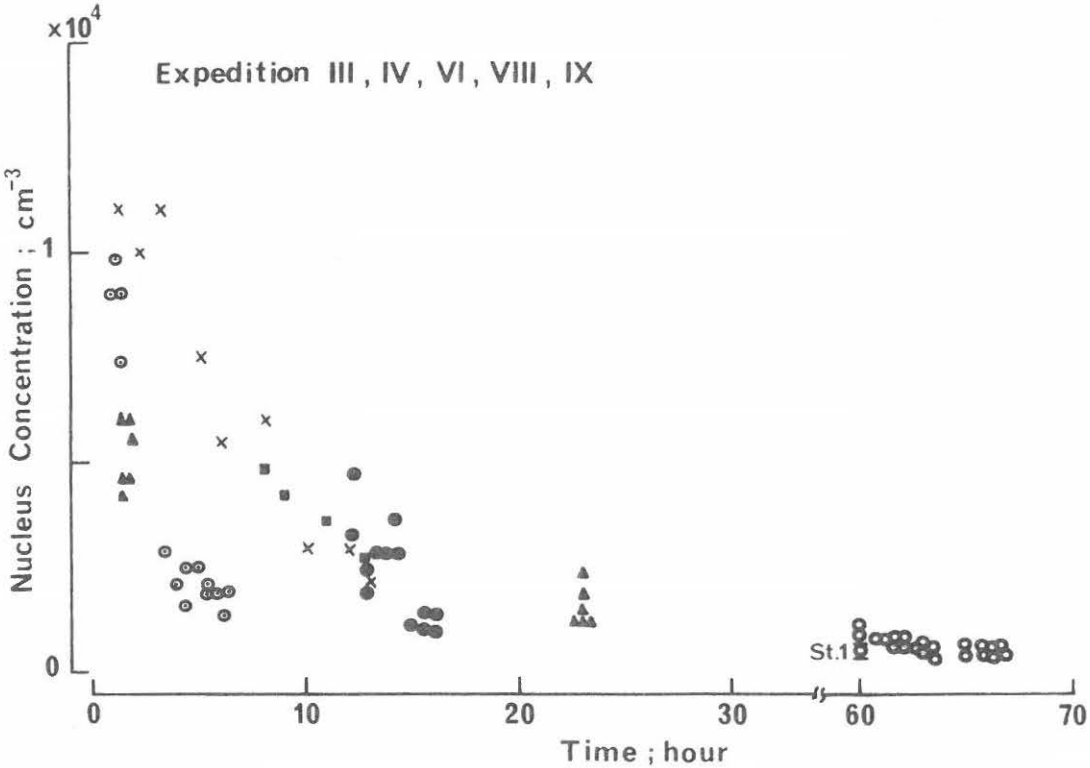

Figure 57. Showing the dependence of nucleus concentration on the age of air masses. Data from Expeditions III, IV, VI, VIII and IX have been superposed.

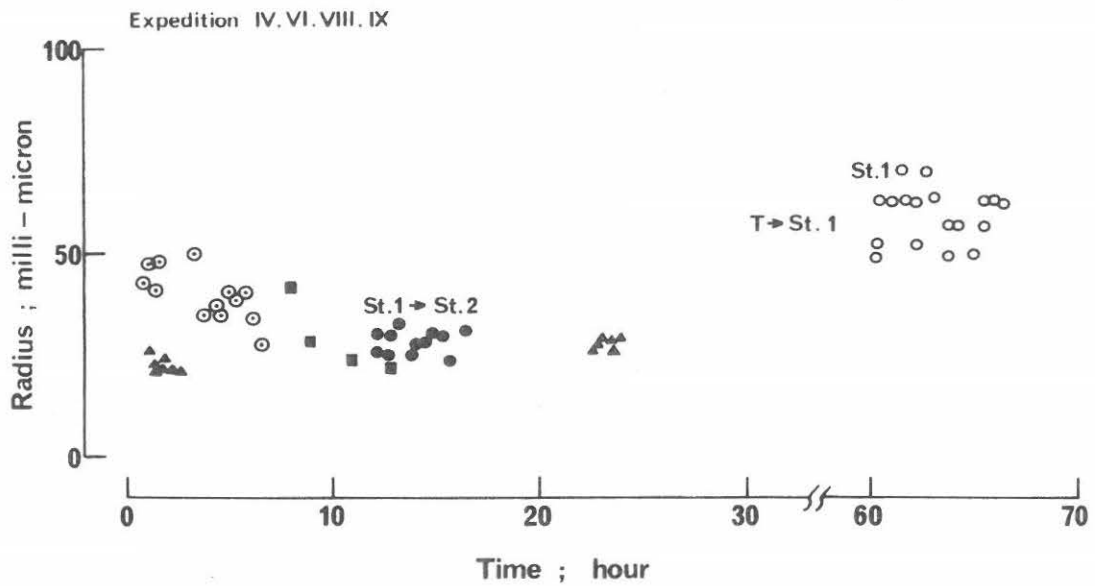

Figure 58. Showing the variation of equivalent radius with time. Data from Expeditions IV, VI, VIII and IX.

important. Another important conclusion can be drawn from Figure 57, viz., that the distribution of nucleus concentration which over the land is highly irregular, becomes gradually uniform with time as the polluted air extends over the ocean. Further, we note that the variations of conductivity and nucleus concentration tend to level off for times greater than about 60 hours.

Even in the case of the measurements on the equivalent radius, the time histories of air masses are of equal importance. The dependence of this parameter on time is shown in Figure 58 in which the symbols have the same meaning as before. It is evident that the data as plotted in the figure show more systematic than those plotted in Figures 47 and 50. A study of the variation of equivalent radius with time is of great physical importance in understanding the corresponding variation of the nucleus concentration. For example, an equivalent radius decreasing with time might cause a corresponding decrease in nucleus concentration via the difference in residence times of the nuclei, and an increase in it with time could presumably results from coagulation.

3.2.3. The inter-relationship between the small ions and condensation nuclei in the oceanic atmosphere :

In our attempts to gain a physical insight into the various processes controlling the electrical state of the atmosphere, whether it be over land or oceans, the importance of the relationship between the ion-pair production (q), small ion density (n) and nucleus concentration (Z) cannot be overemphasized.

And yet, there are only a few experimental investigations to date which seek to study this fundamental relationship (Ikebe, 1970, Ikebe and Kawano,1970). And especially in oceanic atmosphere, such experiments have been very scarce (Sagalyn, 1958, Morita et al., 1971 and Shimo et al., 1972). It is in this context that the author has undertaken a study of the inter-relationship between the small ions and condensation nuclei in the oceanic atmosphere with the help of the data obtained by means of the ship-borne measurements.

Under equilibrium conditions, the relationship between q, n and Z may assume a simple form as given in equations (11) and (12) of Chapter I. Of course, these parameters are related through loss coefficients such as α (the recombination) and β (the attachment) and hence measurements of q, n and Z might, under suitable conditions, lead to estimates of α and/or β . Certain complications, however, arise when one attempts to estimate α and β in the atmosphere over land. For example, q, the production depends not only on cosmic ray ionization but on various radioactive sources in the earth's crust and in the atmosphere. Further, the measurements on Z are much complicated by the presence of a great variaty of sizes in the aerosols. The recent experimental investigations by Ikebe and Kawano (1970) and Shimo et al. (1972) clearly show that, in some cases, β as determined from equation (12) is well correlated to the diffusion coefficient (or equivalent radius) of nuclei and hence cannot be considered as a constant. Similarly, the theoretical investigations by several workers (Junge, 1955, Keefe and Nolan,1962, Bricard,1965 and others) show that the ratio n/n_0 depends on

the particle size distribution. This means that β is a function of the particle size under the large ion equilibrium conditions.

On the other hand, the measurements in the oceanic atmosphere have the advantage of being amenable to a simpler interpretation primarily because only the ion-pair produced by cosmic rays need to be taken into account, thus eliminating the necessity of considering the space and time variation of ionization (Ikebe, 1970). In addition, the size and number of aerosols could be considered as rather uniform in aged air masses as shown in the previous section, and we thus could hope to determine β uniquely from equations (11) and (12) depending on the nature of air masses When the nucleus concentration Z is relatively high, we expect a linear relationship between q/n and Z with the slope representing β (equation 12). When the condition $Z >> (4\alpha q)^{1/2}/\beta$ is not valid, equation (11) needs to be completely satisfied, and we can estimate both α and β from the linear relationship between q/n^2 and Z/n for the aged air masses having the rather uniform equivalent radius of nuclei. Following Parkinson and Weller (1953), Ikebe(1970) and Shimo et al.,(1972), we can take a value of 1.9 ion-pairs per cm^3 per sec. for q in equations (11) and (12). The scatter plot of q/n versus Z for all measurements of Z larger than 4000 particles per cm^3 is shown in Figure 59. The straight line shown in Figure 59 is the linear least-square fit between the two parameters. The slope of straight line is 1.16 x 10^{-6} cm^3/sec. Kawano et al.(1970) have reported that, over land, their measurements of q, n and Z could be interpreted by the simple relation (12). In the present case, it seems probable that for distance of about 100 km from shore, the equilibrium

conditions governing the relation between n and Z, are the same as those over land. In other words, for times within about 10 hours of the aged air masses, the correlation may be well expressed by the simple equation (12).

For the aged air masses having relatively low nucleus concentration, expression (11) would be satisfied. Two examples of the linear relation between q/n^2 and Z/n are shown in Figures 60 and 61 which depict the data from Expedition IV. Now values of n can be derived from measurements of conductivity and by using the measured average ionic mobility of $1.3 \times 10^{-4} m^2$/volt. sec. obtained from Expeditions II and III. Again, q is taken as constant at 1.9 ion-pairs per cm^3 per sec. The data points in these figures the half-hourly averages of the parameters.

It may be mentioned here that the data obtained from the air masses having the age greater than about 10 hours and having the rather uniform equivalent radius of nuclei were used in the present analysis. For example, the value of equivalent radius corresponding to the data shown in Figures 60 and 61 lies in the range of 50 to 70 mμ (ave. 60 mμ) and 25 to 35 mμ (ave. 30 mμ), respectively.

The value of α in the present results lies between 1.6 and 1.8×10^{-6} cm^3/sec., and that of β between 1.0 and 4.2×10^{-6} cm^3/sec. These estimates of α and β can be compared favorably with theory and with the results obtained by other workers. For example, the Thomson theory predicts a value of 1.7×10^{-6} cm^3/sec. for α at N.T.P. Similarly, from their measurements of ionization, nucleus concentration and conductivity over the North Atlantic Ocean, Parkinson and Weller (1953) reported a value

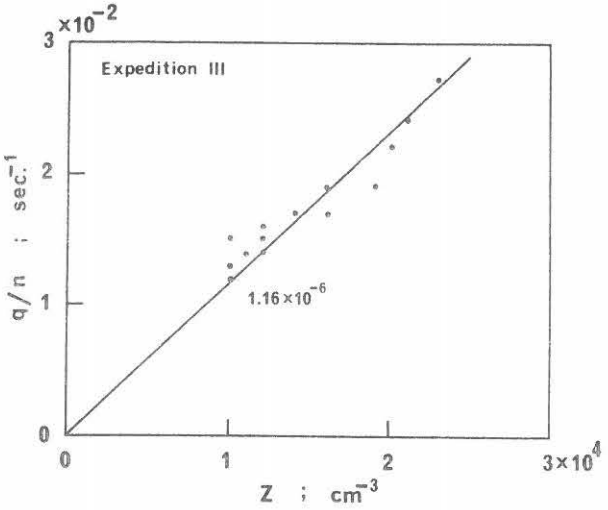

Figure 59. Scatter plot of q/n versus Z. Data from Expedition III. The straight line fit yields a value of $\beta = 1.16 \times 10^{-6}$ $cm^3/sec.$

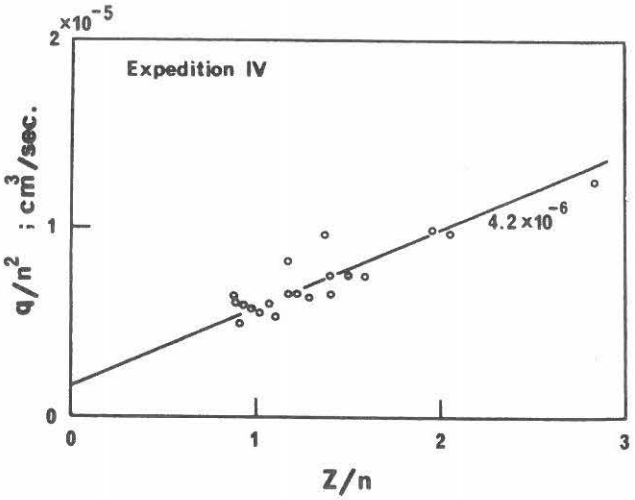

Figure 60. Scatter plot of q/n^2 versus Z/n. Data from Expedition IV. ('Forward' journey from Tokyo to St.1. See also Figure 45). The estimated values of loss coefficients are $\alpha = 1.8 \times 10^{-6}$ $cm^3/sec.$; $\beta = 4.2 \times 10^{-6}$ $cm^3/sec.$

of 2.4×10^{-6} $cm^3/sec.$ for the attachment coefficient. It thus follows that equilibrium conditions in the oceanic atmosphere of low nucleus concentration, are well represented by equation (11).

The present results also show a dependence of β on the equivalent radius. The dependence is clearly brought out in Figure 62. Ikebe and Kawano (1970) and Shimo et al. (1972) have also reported a similar dependence of β on the equivalent radius of nuclei in the range of 10 to 50 $m\mu$ in the atmosphere over land and sea. They found the slight differences of the dependence of β on the equivalent radius over the land and sea, and they attributed their finding to the possible difference of chaged state of nuclei over the land and sea. Keefe and Nolan (1962) have calculated theoretically the values of η/η_0 as a function of the radius. Assuming the large ion equilibrium in the atmosphere over oceans, we can estimate the values of β as a function of the radius. For comparison, both the experimental and theoretical results are also shown in Figure 62. The full and broken lines show the results obtained by Ikebe and Kawano (1970) and Shimo et al. (1972), while the dotted line indicates the theoretical values. It is at once clear from the figure that the agreement between the theoretical estimates and the present experimental results is reasonably good. On the other hand, the results of Ikebe and Kawano (1970) and Shimo et al. (1972) deviate significantly from both the theoretical and present experimental estimates. One of the causes of this discrepancy may be attributed to the difference of degree of pollution of the relevant air masses. The measurements of Ikebe and Kawano

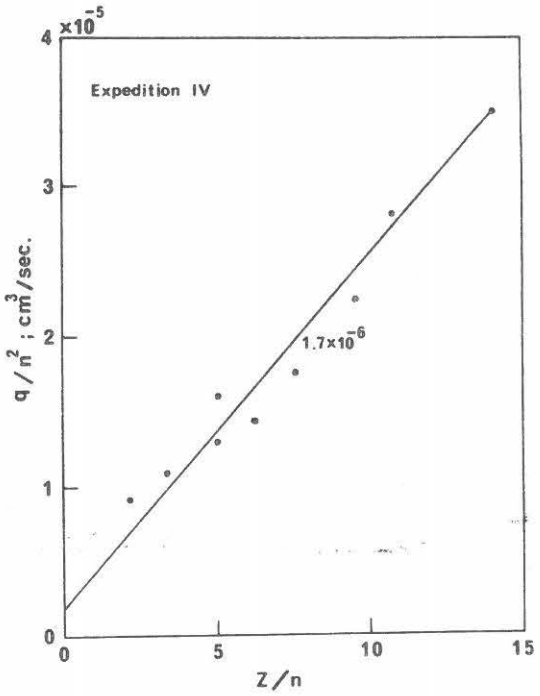

Figure 61. Same as Figure 61 (Data refer to 'return' journey from St.1 to St.2). α = 1.7 x 10^{-6} cm^3/sec. ; β = 1.7 x 10^{-6} cm^3/sec.

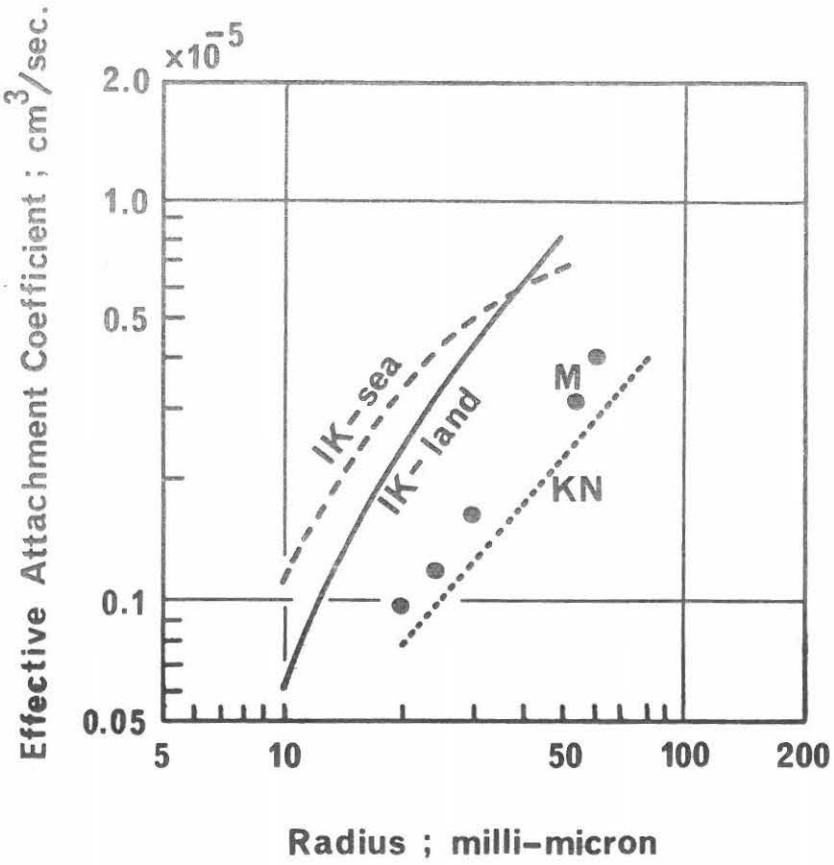

Figure 62. Showing the correlation between the effective attachment (β) and the equivalent radius. For comparison, both the experimental and theoretical results are also shown. M : the present author; IK : Ikebe and Kawano (1970) and Shimo et al. (1972); KN : Keefe and Nolan (1962).

(1970) and Shimo et al. (1972) were carried out in the regions of the relatively high nucleus concentration and their measurements seem to be much complicated by the presence of a great variaty of sizes in the aerosols. On the other hand, the sizes and number of aerosols could be considered as rather uniform in the atmosphere far from land and especially in aged air masses. One thus could expect good agreement between theory and experiment.

Even though atmospheric pollution caused by the industrial activity makes it almost impossible for large ions to acquire equilibrium over land (Sekikawa and Kojima, 1969), one does expect that large ion equilibrium conditions easily prevail in the atmosphere over ocean and especially in aged air masses. Indeed, one could view the results depicted in Figure 62 as lending support to the contention that the large ion equilibrium conditions prevail in the atmosphere over oceans (far from shore).

3.2.4. Electric parameters in the atmosphere over mid-ocean :

In this section we present some of the results of the measurements of electric field and conductivity in the atmosphere over the Pacific Ocean, and discuss their relation to global atmospheric electric circuit and to the background level of global pollution at the time of measurements. The measurements of electric conductivity and nucleus concentration described in the previous section definitely show that to obtain reasonable

estimates of the various atmospheric electric parameters, which could be considered as being typical of the undisturbed atmosphere, measurements have to be made at great distances from shore or in the aged air masses. It is for this reason that only the data obtained at distances of the order of 1000 km or more from shore, have been used in the present analysis.

A. The global aspects of the measurements :

Figure 63 shows typical diurnal variations of electric field and conductivity measured. The variation of conduction current derived from these two parameters is also shown in Figure 63. The points show hourly mean values which are determined from simple averages of the measurements spread over ten days of ideally fair weather at a distance of about 1000 km or more from shore. The measurements were carried out during Expedition III whose geographical coverage was shown in Figure 34. For comparison, the world-wide thunderstorm activity, which is taken from Handbook of Geophysics (1961), is also shown in Figure 63. The electric field and conduction current seem to be in phase with the change in the world-wide thunderstorm activity. It is generally believed that the thunderstorm activity integrated all over the world constitutes the principal source in the global atmospheric electric circuit. The present results seem to reflect this global aspect of the atmospheric electric circuit. There is, however, no perceptible diurnal variation in the conductivity shown in Figure 63. This is in accordance with the fact that ionization in the atmosphere over the ocean is due only to the cosmic rays, which show little or no diurnal variation.

Figure 64 shows the latitude variation of electric field.

This figure is obtained by combining the present results with those reported earlier by Takagi and Kanada (1970) for the same season. The values of the parameters in the figure indicate daily averages for ten degree intervals of the geographic latitude The limits of variation of the parameter are also shown at a selection of points in the figure. For comparison, the Carnegie results which are taken from Buis (1968), are also shown in the figure. In contrast with the Carnegie results, the present result do not show a clear latitudinal dependence of the electric field. It is believed that the latitudinal dependence evident from the Carnegie results, is due to the change in the total columnar resistance with latitude. However, the electric field measured at any particular place is extremely sensitive to local changes in the conductivity. It is thus probable that the feeble dependence of electric field on latitude in the present results is due to some local influence. The data, as they stand now, do not allow any further elucidation on the mechanism of such influences.

B. The background level of global pollution as revealed in the conductivity measurements :

The recent run-away growth of the industrial potential of the world has resulted in a sort of "global pollution" and today, there is hardly any place in the world which can boast of a "perfectly clean atmosphere". One can only speak of an atmosphere of varying degrees of pollution. It is being increasingly realized that 'monitoring' of such global pollution is of vital importance. Towards this end, it is worthwhile to pursue the studies of any parameter whose measurement even

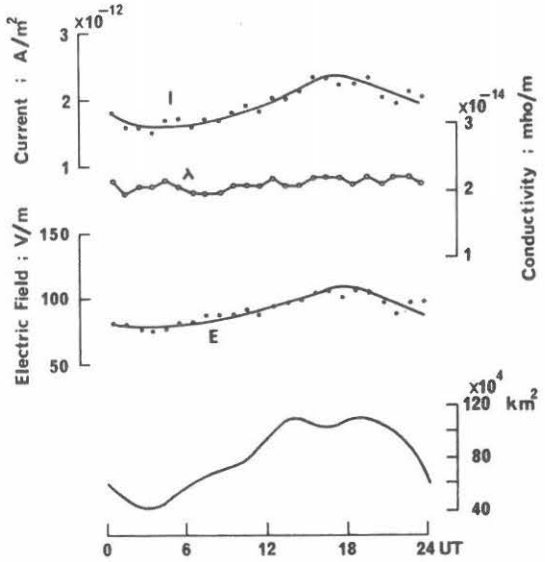

Figure 63. Showing the diurnal variations of electric field (E), electric conductivity (λ) and conduction current (I). The world-wide thunderstorm activity is also shown.

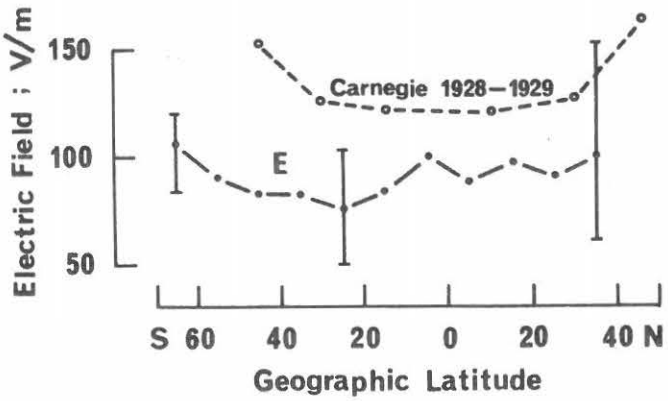

Figure 64. Showing the latitude variation of electric field. The bars indicate the limits of the variation. The broken line shows the Carnegie results.

remotely promises to be of value in monitoring pollution.

The recent developments in atmospheric electricity indicate that the conductivity measurements over the oceans could be converted to yield the concentration of aerosol particles smaller than 0.1 micron in radius since the ionization and mobility of small ions over the oceans can be regarded as nearly constant (Misaki and Kanazawa,1969). Thus the conductivity, measured in the atmosphere over the "mid-ocean", could be a useful index of the background level of global pollution. As described in Chapter I, Cobb and Wells (1970)have found a secular decrease in conductivity over the North Atlantic, the value in 1967 being 1.97×10^{-14} mho/m. On the other hand, the present measurements indicate that the conductivity over the North Pacific was 2.3×10^{-14} mho/m in 1968 and 2.1×10^{-14} mho/m in 1970 respectively. These values are slightly higher than those over the North Atlantic Ocean.

The latitudinal variation of electric conductivity is depicted in Figure 65 which is the result of a judicious reevaluation of the data reported earlier by the author (Morita,1971). The data points in the figure indicate the daily averages along the longitude of 170°W for ideally fair weather conditions. The limits of variation are also shown at a selection of points in the figure. It is seen that the values in the South Pacific Ocean are higher than in the North Pacific Ocean. This shows that pollution in the atmosphere over the South Pacific Ocean is less than that over the North Pacific Ocean.

3.3. A Comment on the Self-Consistency of the Data :

Since data measured at different places and on different expeditions have been used in the previous sections, it would be diserable to check on the consistency of the data. Massing together the measurements of Expeditions II and III, we have obtained the scatter plot of small ion density versus polar positive conductivity as shown in Figure 67 in which all distances are included. The closed circles show the half-hourly averages of the parameters. This small averaging period is necessitated by the rapid changes in the parameters caused by the changing distance from shore. Thus, the closed circles include data presumably affected by land pollution. The single open circle (at conductivity value $\sim 1.0 \times 10^{-14}$ mho/m) refers to a distance of more than 1000 km and hence can be assumed to represent the so-called "undisturbed" or "clean" atmosphere. The encircled dot near a small ion density of about 200×10^{6} m^{-3} shows the values determined at a fixed point in the East China Sea. At two selected points, the experimental errors are also indicated.

It is quite gratifying to note that almost all the points lie reasonably close to a straight line of the form : $\lambda = 1.3 \times 10^{-4}$ e.n. where λ is the polar conductivity, n is the small ion density and e is the electronic charge. This straight line fit can be taken as signifying the consistency of the data.

3.4. Summary and Conclusions :

Nine expeditions have been carried out during the period 1968-71 to undertake measurements of the atmospheric electric

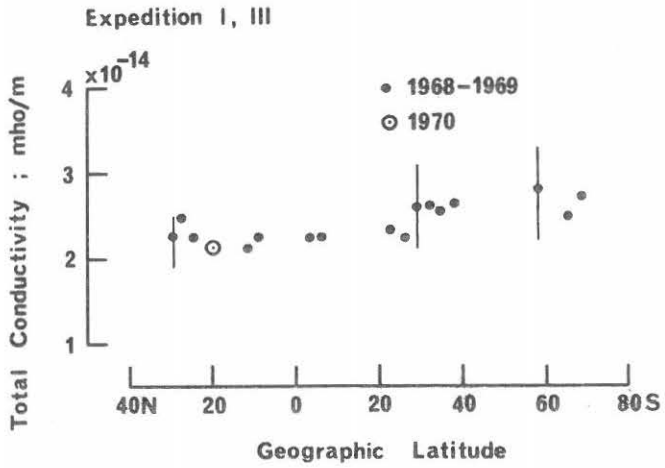

Figure 65. Showing the latitude variation of electric conductivity. The bars indicate the limits of variation.

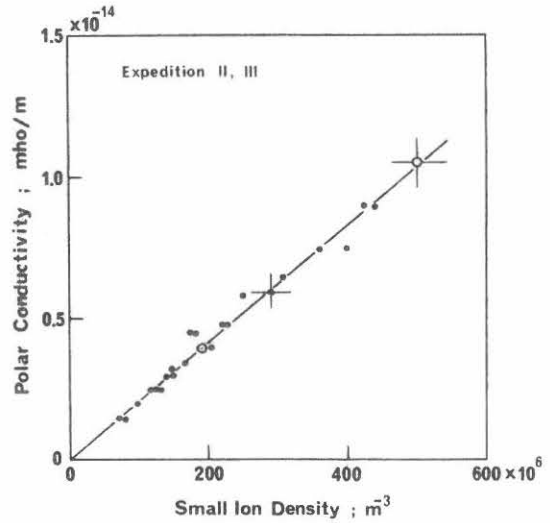

Figure 66. Scatter plot of the positive small ion density and polar positive conductivity. The straight line represents the equation : $\lambda = 1.3 \times 10^{-4}$ e.n.

parameters including nucleus concentration over the Pacific Ocean and the East China Sea. The chief objective of these expeditions has been to understand the behavior of the electrical state of the atmosphere as one moves from land to mid-ocean. The interpretation of most of the data hinges upon the land pollution and its extension to oceanic atmosphere. Some of the important findings are :

(a) Even though, in some cases, the conductivity and nucleus concentration showed systematic dependence on distance from shore, it is concluded that the age of the relevant air masses is the principal factor which controls the electrical state of the atmosphere over the oceans.

(b) The distribution of nucleus concentration which may be highly irregular over land, tends to become uniform with time as the polluted air masses extend gradually over the oceans. The influence of air masses of age greater than about 60 hours on the conductivity and nucleus concentration seems to be very small.

(c) The study of the variation of the equivalent radius with the age of air masses promises to be of importance in understanding the physics of the various processes which seem to control the nucleus concentration and its dependence on the distance from shore.

(d) To obtain values of atmospheric electric parameters which may be considered as being representative of the 'global aspects', measurements have to be carried out at distances of about 1000 km or more from land masses inhabited by civilized population.

(e) Measurements over land or relatively near the shore can be satisfactorily explained by considering only the loss due to attachment of small ions to condensation nuclei. At greater distances, however, both attachment and recombination processes need to be considered. Moreover, the dependence of the effective attachment coefficient on the equivalent radius of nuclei suggests that large ion equilibrium conditions prevail in the atmosphere over the oceans (far from shore).

(f) The conductivity measurements in the atmosphere over the mid-ocean can be used to draw influences about the background level of global atmospheric pollution. The latitudinal variation of conductivity measured indicates that the pollution in the atmosphere over the South Pacific Ocean is less than that over the North Pacific Ocean.

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LIST OF FIGURES AND TABLES

LIST OF PUBLICATIONS CONTAINING

AUTHOR'S CONTRIBUTIONS

(1) Ishikawa, H., T.Takeuti A.Iwata and Y.Morita	1968	Preliminary experiment of atmospheric ion measurements in the transitional region of the atmosphere, Proc. Res. Inst., Atmospherics, Nagoya Univ., 15, 29-41.
(2) Ishikawa,H., Y.Morita and M.Takagi	1969	Balloon observation of atmospheric electrical conductivity up to 24 km in altitude, Planetary Electrodynamics, edited by S.C. Coroniti and J.Huges, 2, 485-495.
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(4) Morita,Y. and H.Ishikawa	1969	Influence of atmospheric aerosol on the ion density profile up to 30 km altitude, Proc. Res. Inst. Atmospherics, Nagoya Univ., 16, 43-52.
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(7) Morita,Y. and H.Ishikawa	1970	Measurements of small ion density in the stratosphere by level flight balloon, J. Geomagn. Geoelectr., 22, 371-372.
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(12) Morita,Y. 1973 Scattering cross-section of freely suspended ice crystals for visible light, Tenki, 20, 23-32 (in Japanese).

(13) Morita,Y., H.Ishikawa, J.Nagasaka and M.Kanada 1973 The land-to-ocean transitional behavior of the atmospheric electric parameters and their relation to atmospheric pollution, J.Meteor.Soc. Japan, 51, 294-306.

(14) Morita,Y. 1973 The inter-relationship between the small ions and condensation nuclei in the oceanic atmosphere, J.Meteor.Soc. Japan, 51,(in press).