Haze Distributions in the Troposphere

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Project director: Dr. William E. Marlatt
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ABSTRACT

HAZE DISTRIBUTIONS IN THE TROPOSPHERE

This study examined horizontal and vertical haze distributions in the troposphere and their possible relationships with the air mass present, surface features present, precipitation, convection, time, and temperature. Inherent in this approach is the investigation of transport properties of haze. Haze data taken from aircraft were classified and coded using the above parameters. Average values for each class were obtained.

From results of this study it was concluded that precipitation, convection and subsidence played a major role in describing haze distributions and transport processes. Temperature was found to be of lesser importance in the troposphere.

ACKNOWLEDGEMENTS

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Chapter I

INTRODUCTION

Aerosols or particulate matter in the atmosphere are significant factors in understanding many physical and chemical processes that occur at or above the earth's surface. Problems in understanding the circulation, turbulence, cloud physics, air pollution and radiation could be simplified if the whole concept of aerosols in the atmosphere was better known. The problems are related to both large and small scale features. Unfortunately, little work has been done on aerosol development and distribution in the atmosphere. This is primarily due to the diversity in the nature of aerosols. Moreover, aerosol activity in the troposphere has had very meager exploration, particularly in the larger ranges.

In this paper, several features of natural aerosol (haze) distribution in the troposphere are investigated. The long range study under which the investigation reported here was one facet, was to examine the effects of haze on long and short wave radiation transfer. When this is known, questions on radiation and heat budget of the earth may be then answered. By using an empirical approach certain relationships shall be examined to evaluate basic transport tendencies of haze. The primary objectives are to answer the following questions:

1) Is there a difference between aerosol profiles in maritime and continental air masses?

2) What is the urban versus rural influence on particle count in the vertical?

3) Does precipitation influence particle count in the vertical?

4) What is the convection influence on particle count in the vertical?

5) How much horizontal variation exists in aerosol distributions?

6) What are the time considerations on aerosol distributions ?

7) How much does temperature influence the vertical structure of haze ?

2.2 Sources and composition of haze.

To appreciate the importance of aerosols in the atmosphere as to different physical and chemical characteristics, one must look at source regions, the composition and the processes whereby the material is transported throughout the atmosphere. The sources for particles greater than 0.2 μ are:

a) Sea salt nuclei. Over the ocean surface sea salt nuclei are formed by the bubble bursting mechanism, Kientzler, et al. (1954) and are transferred directly into the atmosphere.

b) Natural fires. Though not a common occurance, natural fires contributed significantly to the aerosol population (Cadle, 1966). These natural fires are a very non-steady state phenomenon.

c) Wind blown dust or soil. Surface winds are very important tools by which soil particles are transported from the continent to the atmosphere.

d) Terpene-like and other hydrocarbons. Went (1960) estimated that 17×10^7 tons per year of terpene-like and other hydrocarbons are released to the atmosphere by forests and decomposition of organic material.

e) Volcanoes. Throughout history volcanoes have been known to have caused large decreases in visibility due to the number of particles transported to the atmosphere. Volcanic eruption is not a common happening, but does contribute to the atmospheric membership.

f) Spores and bacteria. These organisms are also important constituents in the atmosphere. They are formed on a continental surface and are transported upward by wind action.

g) Chemical reactions. Many chemical reactions between gases take place in the atmosphere to produce aerosols. This is then a source for aerosols that originates in the atmosphere itself.

h) Activity of man. Man contributes significantly to the

aerosol content of the atmosphere. Combustion of natural materials is largely responsible for reduction in visibility in urban areas.

The major source of large nuclei to the atmosphere is the continents. There is a much smaller contribution from the ocean surface. Giant nuclei also have a general component from the continent, but they have a larger contribution for the ocean in the form of sea salt nuclei.

The main sources of aerosols can be divided subsequently into three classes according to Kuroiwa (1953):

1) sea salt
2) combustion
3) soil materials

Again the important role of the continental surface can be recognized.

From the source regions one can directly infer the composition of the particles. Sea salt, as the term indicates, is assumed as a first approximation to contain the same composition as the sea. The major constituent is NaCl which gives large quantities of the chloride ion for chemical analysis. It is a mistake, though, to use Cl^- as the single criteria for identification of maritime air. As documented by Junge (1956) there are a greater number of large Cl^- particles over the continents than over a maritime situation.

On continents natural fires put forth carbon and carbon compounds into the atmosphere. Soil products dominated by silicates and carbonates, are injected into the atmosphere by the wind mechanism. Organic substances are part of the composition of the atmosphere due to terpenes and bacteria type products. The organic nature of the atmosphere is a complex problem. The exact composition of the atmosphere due to volcanoes is still not known, but there is sufficient evidence to identify the presence of surfur and sulfur compounds. Compounds of nitrogen, carbon, and sulfur result from the activities of man. Many of these compounds interact in the atmosphere to form aerosols. Thus, it may be concluded that the atmosphere is a very

"mixed suspension" containing sea salt to organic compounds to quartz.

From the above a natural classification of particulate substances in the atmosphere would be:

1) Inorganic, water soluble (25%)
2) Organic, water insoluble (15%)
3) Inorganic, water insoluble (60%)

The percentages in parenthesis are very crude estimates given by Junge of the composition of aerosols (1967). From this we can see the number of the siliceous and carbon compounds in the atmosphere. In addition, the danger of depicting the atmospheric compositional material by means of rain water analysis is evidenced. The presence of water both in vapor and liquid from in the atmosphere affects the above percentages substantially.

Fig. 2.2.1 shows the analysis for four different chemical species SO_4^{--}, NH_4^{+}, NO_3^{-}, and Cl^{-} in the size range 0.08 μ to 0.8 μ. It may be seen that all quantities decrease with increasing maritime influence. Even the value for chloride follows this general rule. Fig. 2.2.2 given the same comparisons as Fig. 2.2.1 except particles were collected from a cascade impactor in the size range 0.8 to 8.0 μ. Here SO_4^{--}, NH_4^{+}, and NO_3^{-} again decrease with increase maritime influence, but the chloride ion increases. The sulfate ion seems to be the most dominant over the continent for both size ranges.

Another aspect examined is the chemical composition contrast of urban and rural areas. Chambers et al. (1955) made a study of urban-rural chemical composition for aerosols greater than 0.3 μ. The results, Tables 2.2.1 and 2.2.2 show total values of the urban areas to be about 2 to 3 times that of the rural areas. Many insoluble constituents were noted in this analysis. Soluble NO_3^{-1} and SO_4^{-2} were found in only small quantities. It is interesting to note the artificial Pb and F^{-1} values are higher for urban areas, but the difference is not as great as one might expect.

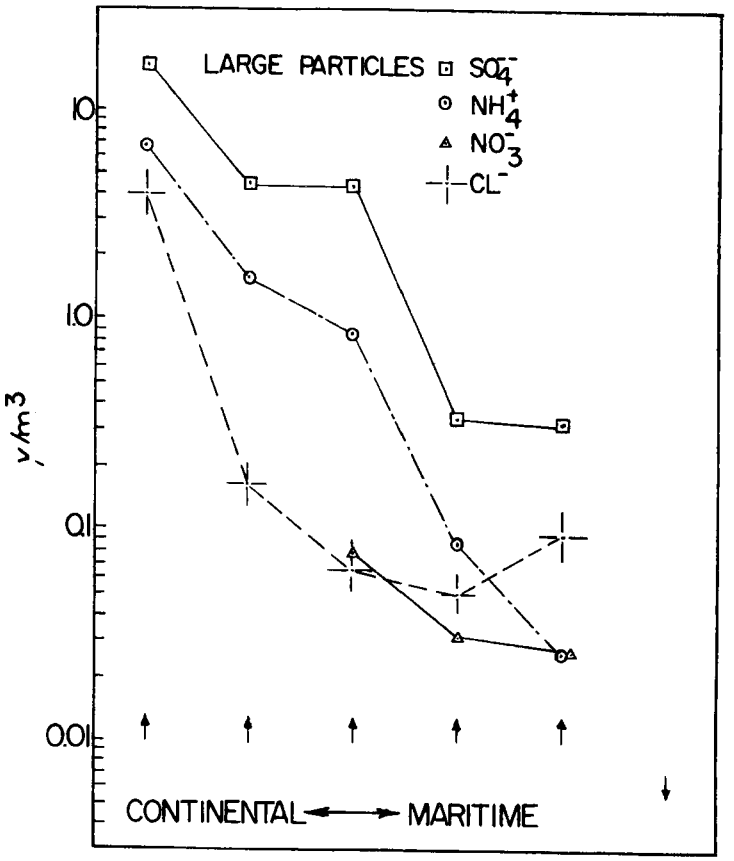

Fig. 2.2.1

Survey of he average chemical composition of large particles arranged according to air mass (after Junge, 1963).

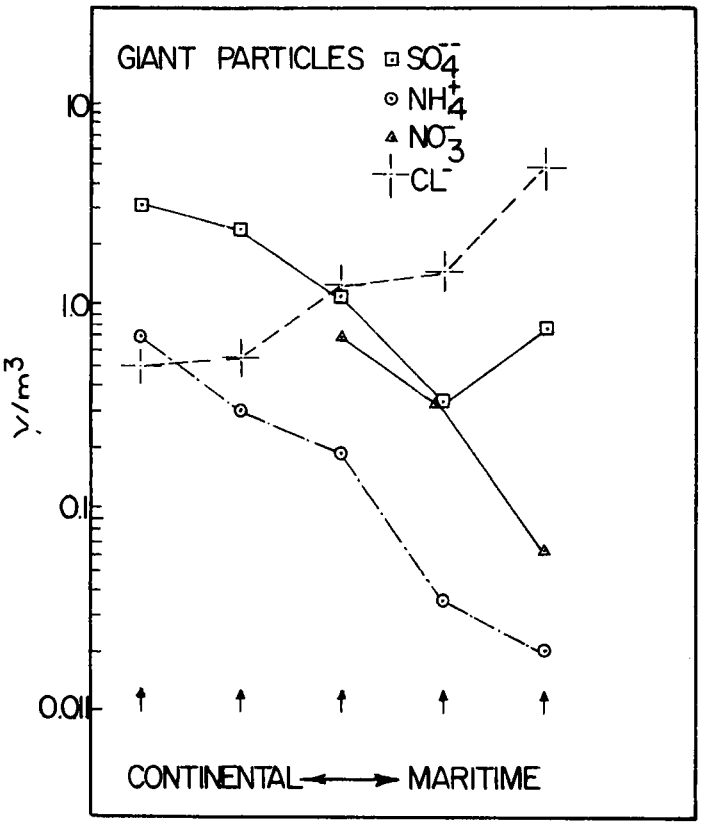

Fig. 2.2.2

Survey of the average chemical composition of giant particles arranged according to air mass (after Junge, 1963).

2.3 Aerosol Transport Processes

The mechanisms by which aerosols are moved through the atmosphere from one place to another are very complex and not thoroughly understood. The basic troposphereic aerosol cycle can be evaluated from a knowledge of sources using Mason's classification of aerosol producing mechanisms (1962). This classification is based on (1) condensation and deposition of vapor; (2) chemical reactions; (3) mechanical disruption and dispersal; (4) coagulation. Aerosols are injected into the atmosphere from the surface by a number of different processes. The most important of these are:

a) Bursting bubbles. Salt particles from the ocean are formed by bubbles that burst at the surface. There results from this action a jet-like effect with the smaller particles tossed upward to mix with the atmosphere.

b) Wind. It is obvious to anyone caught in one that a dust storm results in the accumulation of many soil particles in the air. Wind mechanically disrupts soil material and carries it above the surface. In the mid-latitudes wind general increases with height in the troposphere. This provides a means by which particles can be sustained in a mixed state for great periods of time. Many authors have examined the influence of wind on sea-salt concentration over the ocean and have shown the greater the wind forces the greater the production of sea-salt concentrations in the atmosphere.

c) Convection. Convective action of the atmosphere can produce strong vertical currents which will transfer particles from the lower levels to higher levels. The vertical extent of these updrafts is limited by heating and terrain effects and is usually signified by the cloud tops. Convection has a major effect on particles already existing in the atmosphere and tends to transfer them upward to higher levels.

d) Condensation and deposition of vapor. Warm combustion

products and lighter-than-air gases are readily distributed upward in the atmosphere. Once placed in the atmosphere they cool and combine with condensation nuclei to form aerosols.

It is necessary that there be processes where aerosols are removed from the atmosphere if the atmosphere is not to become a solid 'aerosol'. The primary mechanisms by which aerosol particles are returned to the surface are:

a) Sedimentation. The force of gravity acts upon particles and removes them from the atmosphere at essentially their terminal velocity. The terminal velocity of particles falling freely through still air can be approximated for drops less than 20 μ by:

$$u = 1.2 \times 10^6 \, r^2 \ \text{cm/sec}$$

when u is the terminal velocity and r is the radius of the particle. It may be seen that, for particles less than 1μ diameter, $u = 3 \times 10^{-3}$ cm/sec. This would indicate that gravity is not an important factor for particles less than 1μ . The earth is, therefore, a sink only for giant particles. This can be illustrated also by considering the residence time (τ) of a tropospheric aerosl. By eliminating eddy diffusion considerations and assuming a vertical constant aerosol layer up to a given height ($\overline{H}$), τ can be determined by:

$$\tau = \overline{H}/u$$

where u is the terminal velocity. For $\overline{H}$ = 5 km (≈ 15, 000') and using Stokes Law the Table 2.3.1. can be designed. From this table, the fact is evidenced that particles 1μ diameter and smaller are not influenced greatly by sedimentation.

b) Precipitation influences. These consist of rainout and washout processes, whereby aerosols are taken up by rain water in the cloud and below the cloud, respectively. Rainout can be characterized by:

$$K_1 = c \cdot e/L$$

where c is the concentration of a particular constituent in air, K_1 its

TABLE 2.3.1.

Fall Velocity and Residence Time of Aerosol Particles

τ for Height $\overline{H}$ = 5 km

Particle radius, r	.1	1.0	2.0	4.0	6.0	8.0	10.0	15.0	20.0
Fall velocity, cm/sec	1.2×10^{-4}	1.2×10^{-2}	4.8×10^{-2}	.19	.53	.77	1.2	2.7	4.8
τ, days	47000	470	118	30	10	6.5	4.7	2.1	1.2

concentration in cloud water, e the rainout efficiency, and L liquid water content of cloud. It may be shown that rainout efficiencies are highest for particles larger than 0.4 μ diameter. The quantity c appears to be dependent on the amount (type) and the history of the rainfall. c is higher for situations of frequent showers. Also c is lower during stratus shower activity rather than thunderstorms. Georgii and Weber (1960) showed that concentrations were approximately doubled in rainfalls that followed dry spells than those of wet periods.

Washout is a function of cloud base level, evaporation, size of falling drops and frequency of falling rain drops. Junge (1963) calculated that the fraction of aerosol removed by washout from the atmosphere was only 4%. (Using average conditions on a per day basis). This value is for aerosol masses with sizes larger than 4μ diameter. For particles of less than 4μ diameter the fraction is smaller because washout is less effective on smaller particles. Junge's calculations are based on only average conditions which may not reflect large accumulations of rainfall over time. Even considering rainout and washout together, it would appear that any appreciable removal of aerosols must be a result of long term effects. Junge (1963) found that the residence time of aerosols due to the combined effects of rainout and washout ranged from two to twenty four days for average conditions and for aerosols greater than 4μ diameter. This range in accordance with present data is acceptable in the lower troposphere.

c) Impaction with the earth. Particles are removed from the atmosphere after they come in contact with the surface of the earth or large structures stationary on the surface. There is a serious lack of quantitative information about this removal process. The size of particles, terrain and vegetation are factors which must be considered. The more irregular the surface the greater the impaction effect will be.

d) Subsidence. Just as vertical motions carry aerosols away from the surface (convection) vertical movements bring aerosols

toward the surface by subsidence. Subsidence is not a direct removal process, but is a mechanism for redistributing aerosols in the atmosphere. Subsidence prevents vertical mixing and limits the vertical extent of haze layers. An example of this process is the subsidence aloft above cloud base in the tropics which is a result of the general circulation of the region. This effect tends to keep most aerosols below the trade wind inversion.

When discussing aerosol production and removal processes two important considerations, coagulation and diffusion must be included. Coagulation is the process whereby small particles agglomerate together to form larger particles. Coagulation is, therefore, dependent upon the number of 'efficient' collisions between particles to produce the agglomeration. The number of collisions is in turn related to the energy of the system.

In the atmosphere, coagulation is important because of its' role in the 'aerosol cycle'. As small particles coagulate they become larger particles. Larger particles are more susceptible to the removal processes. This places a limit on the lower end of the aerosol size spectrum. The effect of coagulation on aerosols is seen dramatically when observing volume changes from Aitkin nuclei to large particles in which there is an influx or movement of particles from Aitkin to large particles.

The process of diffusion is used to explain small scale particulate distribution in the atmosphere. Basically diffusion is a function of temperature and is manifested by eddy currents. As particles diffuse through the atmosphere the controlling factors must be the number density of particles and the character of the small eddies.

Diffusion plays a very important role in coagulation. The diffusive elements of a particulate cloud essentially determines the extent of coagulation. The rate of coagulation must, therefore, be considered in discussing the production-removal processes.

The productive processes are balanced against the removal processes to obtain a steady state approximation. A description of the vertical distribution of aerosols may be attempted by considering this first approximation. The diurnal variation of Aitken and large particles may be explained partially on this basis, with the change in temperature being the focal point. The steady state can also be used, again as a first approximation, to explain the decrease in aerosol concentration with height for particles greater than 2μ. The eddy-diffusion-sedimentation equilibrium is the tool used here. For smaller particles the steady state is not a valid assumption; rather, coagulation and washout seem to dominate the system.

A basic picture of how aerosols move in the vertical dimension has been presented. It is assumed here that aerosols tend to follow the general circulation patterns on the large scale horizontal plane. In the horizontal, diffusion and coagulation again play dominant roles. Many questions are yet to be answered concerning the nature and movements of aerosols in the atmosphere.

Chapter III

METHOD OF AEROSOL MEASUREMENT

3.1 Aerosol Counting and Sampling

Over the past years there has been an increased amount of interest in measuring and describing the aerosol content of the atmosphere. This is the result of the fact that aerosol parameters have a large contribution in problems in radiation transfer, air pollution and other aspects of atmospheric physics and chemistry. As a result, there has been a number of different aerosol counters and samplers designed.

Although the literature contains references to many such instruments, the lack of good representative data for the total spectrum of aerosol particles from the surface and troposphere persists. One of the main problems here arises from the wide variability of aerosol sizes and composition. In addition, most counters are capable of registering only in a limited interval of the aerosol spectrum. The techniques of collection and counting of a given sample have not been fully developed. The adaptability of the instrument to take airborne data and to efficiently and quickly count the particles are serious problems.

Aerosol counters are of four different types:

a) Sedimentation collection. Sedimentation methods are accomplished by either condensation of particles in a supersaturated atmosphere (Aitken, 1923), or by deposition of particles on a surface by means of an electrical, thermal or gravitational field (Fletcher, 1960). These methods are appropriate only if there is a high concentration of particles in the volume or if the particles have high terminal velocities. The technique used in these methods is primarily accomplished by counting by eye from a photograph made of a slide (May, 1945) or from an ultramicroscope (Whytlaw-Gray, 1936).

b) Filtration collection. Filtration methods have become more sophisticated with recent advances in technology. In this method aerosols are collected by forcing air through a filter or a series of filters; and the counting done by the autoradiographic method (Leary, 1951) or a microscope (Silverman, 1941). This method is, of course, not useful for collection of volitile particles, liquid particles, large concentrations or size analysis of particles.

c) Impaction or Impingment collection. By forcing air toward a perpendicular surface one can size discriminate between particles and can collect them on this surface. The particles can also be directed through a curved path producing the same effect. There are a number of these impactors. A critical review of these was made by Green and Lane (1964). Counting is usually done by photographic or microscopic analysis.

d) Photoelectric counting. Pollack and Nolan (1946) and others have developed an instantaneous photoelectric method of counting particles which is based on measuring the light attenuation of the supersaturation fog produced in the Aitken counter described earlier. Optical techniques have been developed for counters such as the Bausch and Lomb (Martens, 1966) where the attenuation of light through an isokinetic flow of aerosols is measured. Counting is dependent upon the scattering properties and the size distribution of the particles.

A more detailed description of the above instruments can be found in references Green (1964), Fletcher (1960), Junge (1963) and Walton (1952).

3.2 The Bausch and Lomb Dust Counter

For the purposes of this study an instrument was required which could:

a) Count aerosols in the size range 0.3 μ and greater (more specifically, 0.3 μ -10.0 μ). A collection of aerosols for chemical analysis or a sensitive size distribution analysis was not important

for the objectives of our studies.

b) Take airborne data at different altitudes.

c) Provide instantaneous results for in-flight interpretation.

The instrument must present the data in a form for quick and efficient analysis.

To satisfy these requirements the Bausch and Lomb Dust Counter, model 40-1 was selected.

The Bausch and Lomb Dust Counter 40-1 as stated above, operates on the principle of counting particles photoelectrically. Perhaps a better description of the B & L (Bausch and Lomb Dust Counter 40-1) is that it is an electro-optical aerosol counter. The effects of light scattering or light adsorption on atmospheric particles are measured electrically. This instrument is capable of measuring particles in different size ranges of: $\geq 0.3\mu$, $\geq 0.5\mu$, $\geq 1.0\mu$, $\geq 2.0\mu$, $\geq 3.0\mu$, $\geq 5.0\mu$ and $\geq 10.0\mu$ diameter. Concentrations from 0 to 10^6 particles per cubic foot are detectable with this instrument. The size of the instrument is approximately 12" x 12" x 19" and weighs 23 pounds. It requires an operating power of about 250 watts. This instrument is, therefore, very satisfactory for use in a light airplane.

The Bausch and Lomb Dust Counter is based on the principle of near-forward scattering of light through a flow of aerosol particles. Forward scattering gives the more intense signal than right angle or back scattering and the intensity if less dependent on the refractive index of the particles than in the other directions. Fig. 3.2.1 illustrates the optics of the system. Direct forward light is trapped in the cone. All light that is scattered in a solid angle 15° to 60° in the forward direction is detected. The shape of the particles has less influence on the light collected scattered across this range of direction than from collections made uni-directionally. The volume in which the air sample is collected and analyzed is only 0.5 mm^3. Thus, the probability is very high (99.97%) that not more than one particle

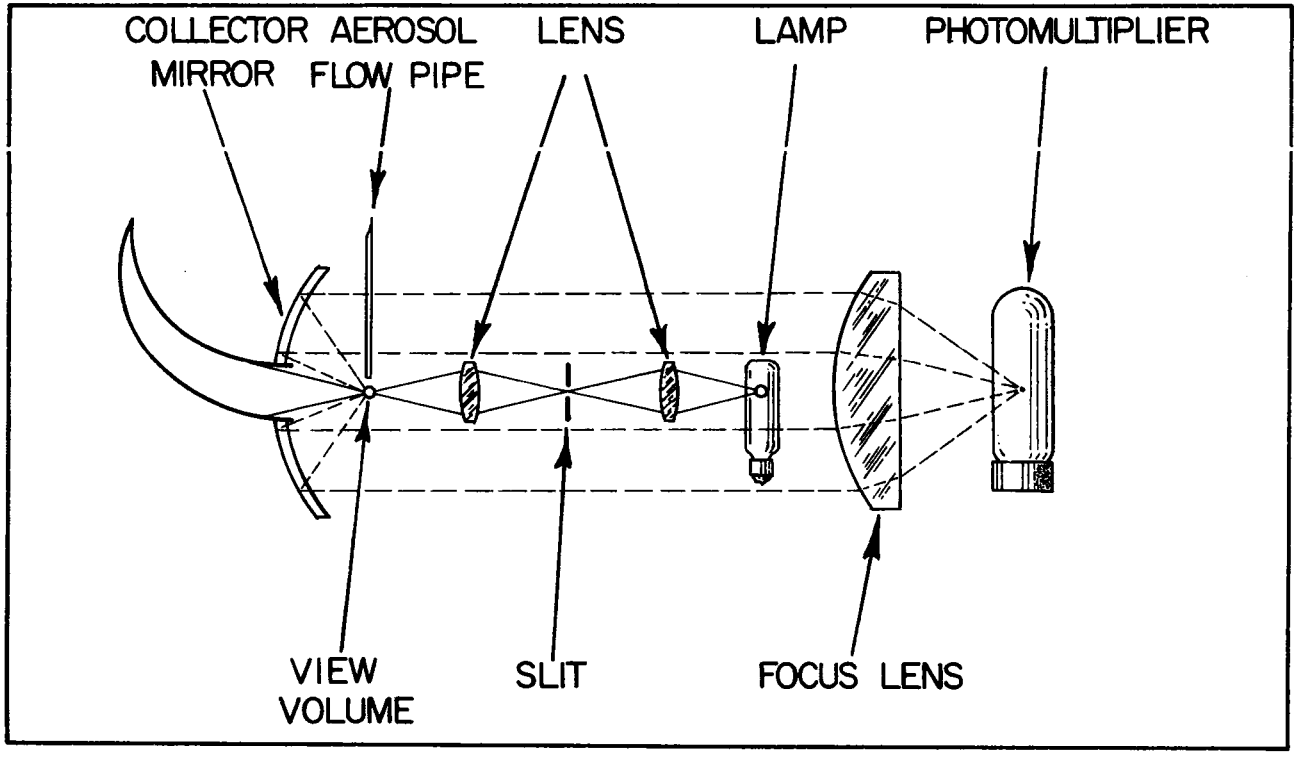

Fig. 3.2.1.

Optical system of the Bausch and Lomb Dust Counter, 40-1 (after Martens, 1966)

will be measured at any given time. This volume (view volume) is constructed at the point where the flow of the aerosol sample and the beam of light intersect.

The electronics of the Dust Counter are based on the fact that different size particles in the view volume will scatter a constant light beam source at different intensities. These intensities are detected by a photomultiplier (Fig. 3.2.2) and converted to pulses of electrical current. The pulses are passed through one of eight selected load resistors to get the desired particle size range. After the pulse is amplified it is passed to a threshold detector (pulse height discriminator) and then is directed to a calibrated meter or to an electrical output to an external recorder.

The air sampling design of the B&L Counter is another unique feature of this instrument. A diagram of the sampling system is shown in Fig. 3.2.3. The small volume of air to be sampled is drawn through a tube into the view volume by a positive displacement air pump with the flow produced regulated by a by-pass valve. Mufflers are installed to minimize noise. In the closed loop a large volume of clean air is circulated to flush out the system and create laminar flow. This operation prevents aerosol build-up in the instrument and eliminates turbulence in the view volume.

To insure accurate operation the Dust Counter must be calibrated periodically. It is calibrated by passing a sample of microscopic polystyrene latex spheres of a known concentration and size by the view volume.

3.3 Other Aerosol Sampling Devices

The Bausch and Lomb Dust Counter 40-1 satisfies the requirement of this study. For reader information a brief description of other popular samples and counters is included.

a) Goetz Aerosol Spectrometer ultracentrifuge. The Goetz Spectrometer (Goetz, 1960) collects particles in the size range of

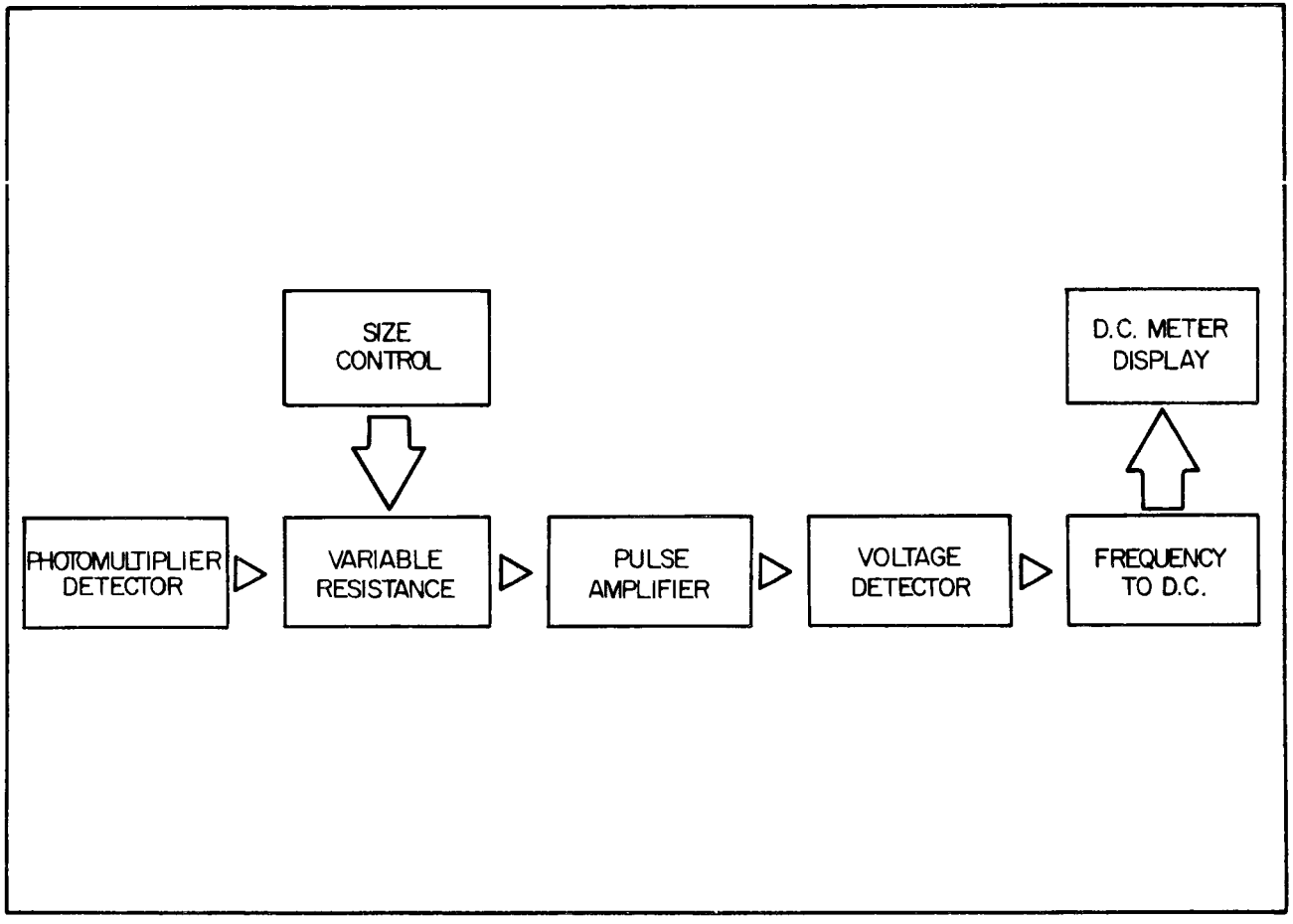

Fig. 3.2.2

Electronics flow diagram of the Bausch and Lomb Dust Counter, 40-1 (after Martens, 1966)

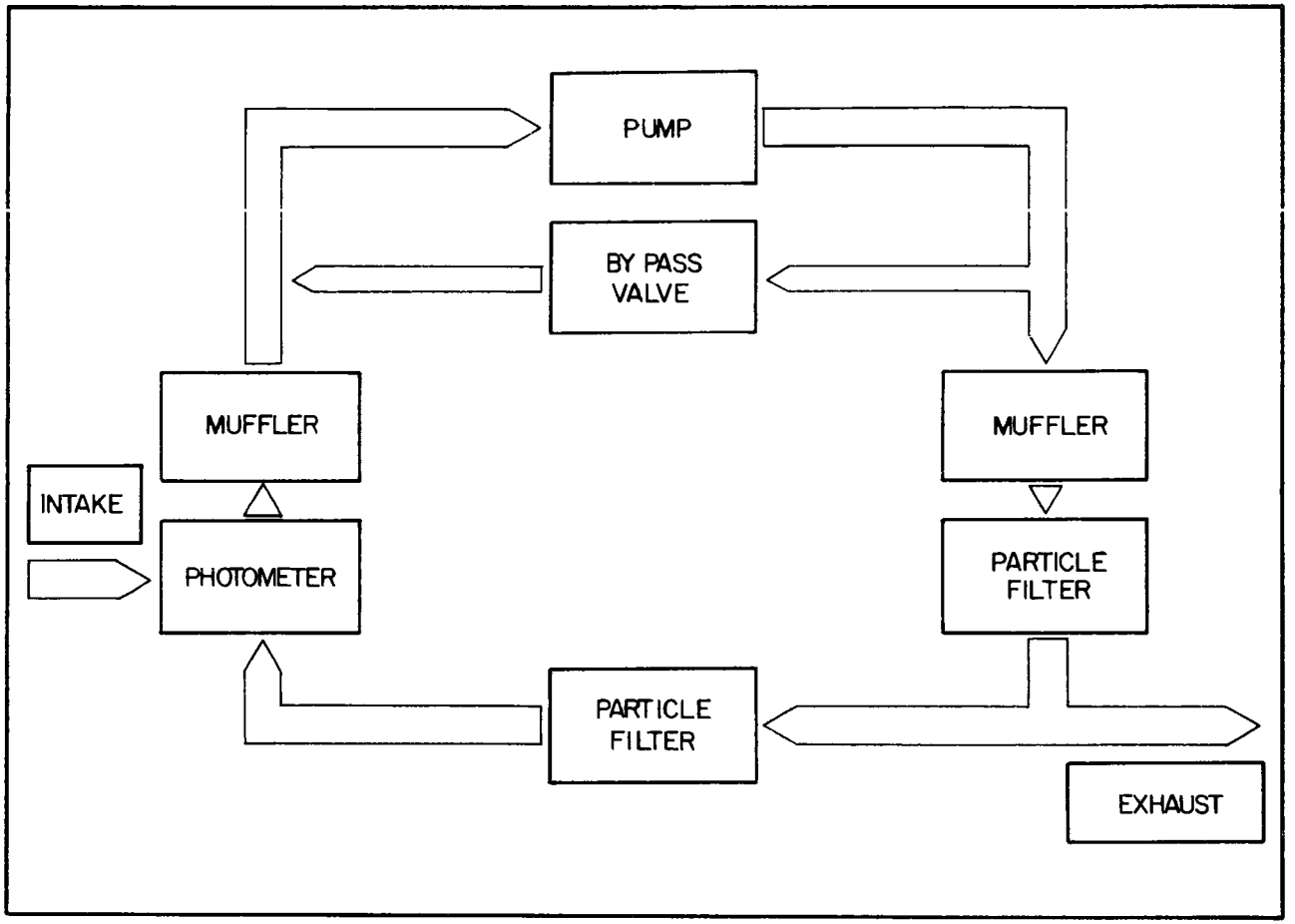

Fig. 3.2.3

Air sampling system of the Bausch and Lomb Dust Counter, 40-1 (after Martens, 1966)

0.03μ to 3.0μ diameter. The instrument operates on the principle of centrifuging a small sample of air and collecting the particles which have deposited on a chrome-plated foil. The particles are depositied according to their sizes due to the relationship between a particle's size and its terminal velocity. A photograph is taken over portions of the foil and can be analyzed by means of a planar-scanner (Gerber, 1967). The planar-scanner optically and automatically counts particles in a photograph. The Goetz ultracentrifuge is an adequate method of collecting particles; although, it is not very efficient for low concentration counts and errors due to orifice loss and the non-spherical nature of some aerosols do occur. The instrument and the counting device are not suitable for airborne measurements because of its size.

b) May Cascade Impactor. The May Cascade Impactor (May, 1945) is a series of force impactors which have different size jets in front of them to change the flow of a given air sample that is drawn into the instrument (Fig. 3.3.1). The smaller and lighter particles follow the flow more readily. This produced a spectrum of particles on the different impactors (B_1, B_2, B_3 and B_4) that can be analyzed by optical methods. This instrument does not collect particle by size but by mass. (This statement is true for most impaction instruments). The aerosol size range claimed for the May system is 0.7μ to 15μ. diameter. The efficiency of this interval, however, is somewhat variable due to the inability of the jets to provide for the whole range. This system was intended primarily for liquid particles and some errors are inherent for solid aerosols. Deviations from isokinectic flow can also produce errors.

c) Torgeson-Stern Impactor-fliter system. A schematic diagram of the Torgeson-Stern Impactor-filter is shown in Fig. 3.3.2. This device consists of a two-stage impactor and a back-up filter for collection and fractionation of particles. The range of aerosol sizes is 0.1μ to 4.0μ. The efficiency in this range is good with some

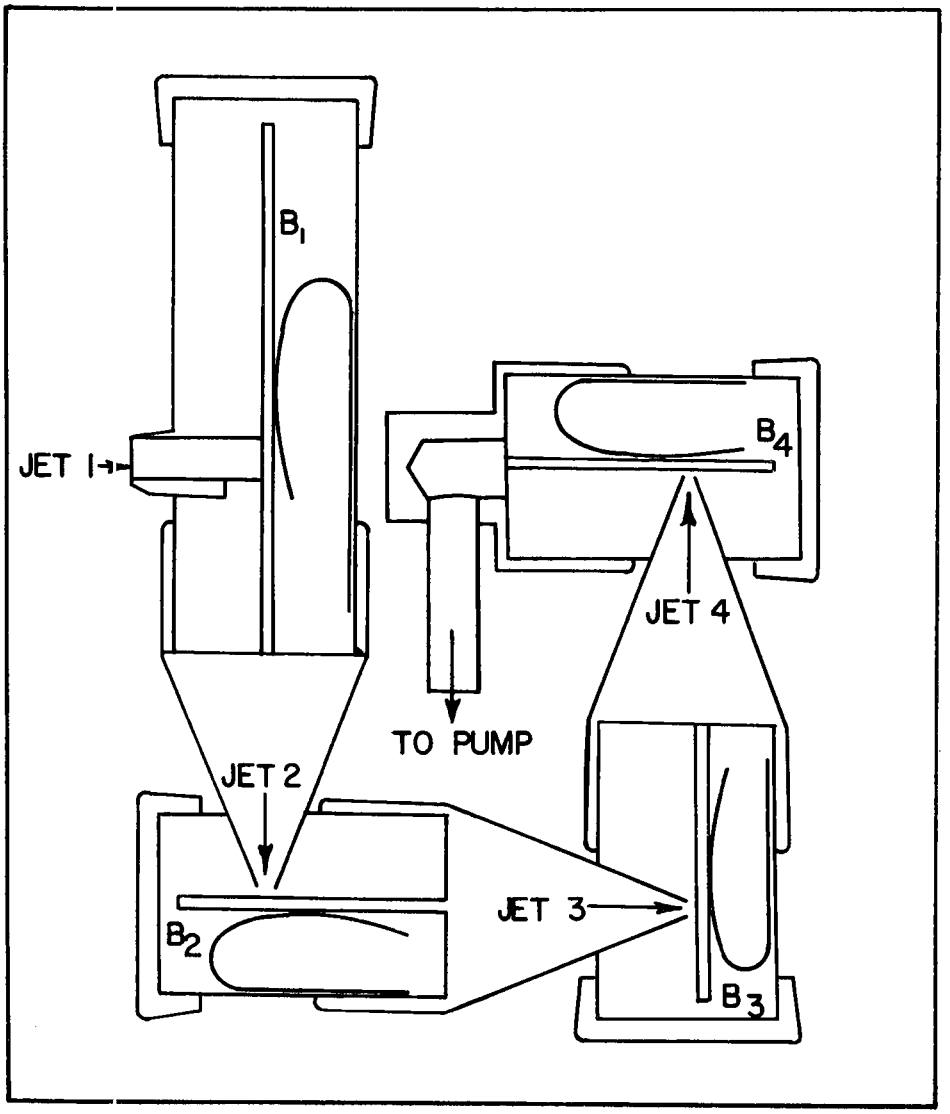

Fig. 3.3.1

Diagram of the May Cascade Impactor (after Green, 1964)

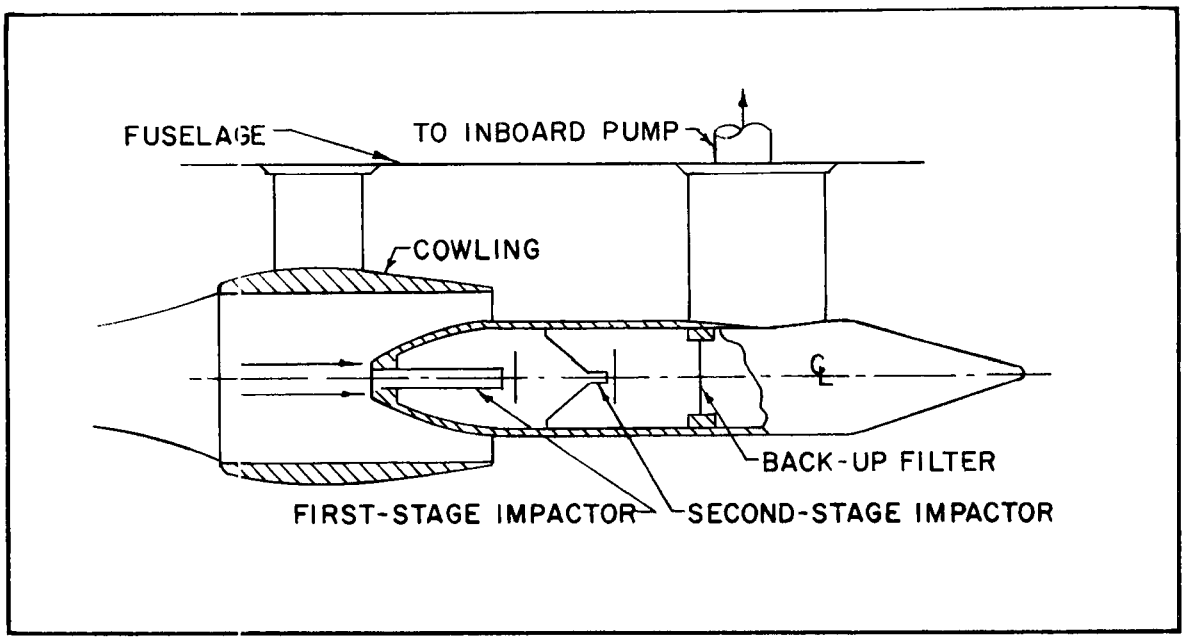

Fig. 3.3.2

Diagram of the Torgeson-Stern Impactor-filter system
(after Torgenson, 1966)

limitations at 0.1μ and in the 1.0μ to 3.0 μ band where there are gaps in the total spectrum of sizes. The counting or other analysis of the particles collected can be accomplished by optical techniques. These can be most tedious and time consumming since the measurements of the impactors and the filter both must be made. This instrument is well adapted to airborne operation.

The system employs the use of a pump to produce isokinetic flow and a IPC-1478 filter. The impactor stage construction was based on two assumptions: adiabatic flow in impactor nozzles, and isothermal flow between stages.

d) Pollack-Nolan Photoelectric Counter. As indicated above, the photoelectric counter is based on the concept of light attenuation of a supersaturated fog produced from particles. This method, developed by Pollack and Nolan (1946), differs from the Bausch and Lomb system in that a larger volume of air is analyzed and the attenuation is a function of all particles acting together; while the Bausch and Lomb Dust Counter concentrates on the optical properties of single particles in a small volume. Fig. 3.3.3 illustrates the photoelectric counter. The nature of the Pollack-Nolan counter is related to the ability of the particles to act as condensation nuclei, and therefore can measure primarily condensation nuclei.

It can be seen that each of these instruments could produce different answers to the same questions depending on the use and interpretation of each.

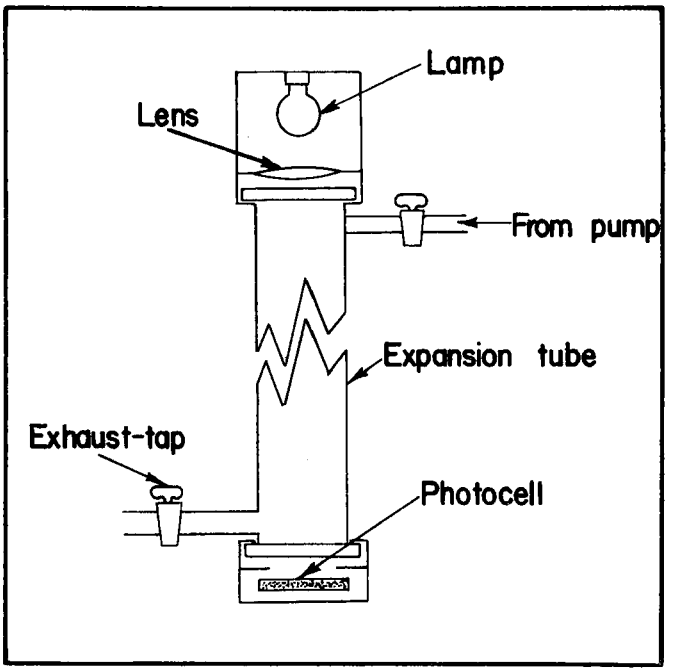

Fig. 3.3.3

Diagram of the Pollack-Nolan Photoelectric Counter (after Fletcher, 1960)

Chapter IV

DATA COLLECTION & PROCESSING

4.1 Data Source

The aerosol measurements were obtained using a Bausch & Lomb Dust Counter mounted on a Piper Twin Comanche, a Convair 990 and a Douglas C 54.

In 1966 flights were associated with the NASA Convair 990 Meteorological Flight I program. In the time period May 27 to June 17, 1966 Colorado State University flew 19 support missions for the NASA flights in a Twin Comanche aircraft. In 13 of these flights aerosol data were taken by the Bausch and Lomb Dust Counter. Almost all the data (one exception) were recorded in the late morning to late afternoon portion of the day. These flights were made primarily in the general San Francisco oceanic, coastal, and inland area with few flights in the South Atlantic coastal region in the vicinity of Charleston and Atlanta.

In 1967 flights were associated with the Line Islands Project. From April 6 to April 16, 1967 Colorado State University flew 6 flights in the vicinity of Christmas Island in the tropical Pacific. Airborne aerosol measurements were taken in the Wood's Hole Oceanographic Institution Douglas C-54 aircraft. Temperature data for times corresponding to the aerosol data were a result of the radiosonde network on Christmas Island. These data were taken during all hours of the daylight.

In 1967 flights were associated with NASA Convair 990 Meteorological Flight II program. From May 5 to June 8, 1967 Colorado State University participated in the NASA 990 program and collected aerosol and temperature data on the Convair 990 for 39 flights (26 vertical and 13 level flights). These flights concentrated in the regions of San Francisco and Dallas, Texas. Most of these flights were taken in the after noon hours.

In 1967 flights were made through western United States, central United States and western Canada. Aerosol and temperature data were collected aboard a Piper Twin Comanche aircraft for vertical profiles for 23 flights. Flights were flown over urban and rural areas covering western and central United States and Canada. These flights took place in the late morning or afternoon. These flights will be refered to as the "Comanche Flights".

In 1968 flights were associated with NASA Convair 990 Meteorological Flight III program. During the time period June 5 to June 21, 1968 Colorado State University participated in NASA Convair 990 program (Meteorological Flight III). Aerosol and temperature measurements were taken on board in the Convair 990 for 20 vertical profile flights. All flights were flown in the late mornings and afternoons. These flights were made generally over the state of California and the lower Mississippi River Valley.

4.2 How data was taken for each flight

a) 1966 CV990 Flights (Twin Comanche). Data for these flights were taken at prescribed levels in the atmosphere from 500 to 2000 foot intervals. The data were recorded by means of the meter installed on the Bausch and Lomb instrument.

b) 1967 Line Islands Flights. The data for these flights were collected in the same way as the 1966 CV990 Flights. 1000 foot intervals were used exclusively.

c) 1967 CV990 Flights. Data for these flights were taken according to time but not any specified altitude. Again, the recording device was the meter on the Dust Counter. Each reading of the Bausch and Lomb was accompanied by a specific time. Also records were kept on board the plane of altitude versus time.

d) Comanche Flights. Data for these flights were recorded on magnetic tape which was analysed by computer. The vertical profiles were obtained by flying at specified levels in the atmosphere (every

500 foot intervals) for approximately 2 minutes per level. This permitted a number of different readings for each level.

4.3 Processing of data

For the purpose of classification of different divisions (or sets) based on various criteria, all data within a set was summarized by averaging. Since the data were not homogeneous with height, a method was developed to summarize all data into standard height levels. This was done by constructing 1000 foot intervals throughout the troposphere and assigning the haze measurements in that interval to the mid-point of the interval. All data were analysed in this manner, except the Comanche data which separated it by 500 foot intervals.

To further describe the nature of these sets, the range of values and the standard deviation, $\sigma = \frac{\Sigma (y - \bar{y})^2}{N}$, was computed for each level. Both averages and standard deviations were obtained by means of a simple computer program.

Since all levels do not have the same number of observations averaged, tables are included indicating the number of observations used for each average.

Accumulative values of each average profile were calculated by summing up the 1000 foot intervals every 5000 feet to the top of the profile to obtain total amount of haze in the atmosphere.

a) 1966 CV990 and Line Island Flights. These data provided vertical profiles of aerosols and temperature at levels in the atmosphere up to 18,000' msl. The temperature data for the Line Islands area was analysed by interpolating the Christmas Island radiosonde data.

b) 1967 CV990 Flights. Aerosol and temperature data produced profiles of vertical parameters as a function of time which can be height interpreted up to 35,000' msl.

c) Comanche Flights. The average of the points for each level was obtained and plotted as a function of the fixed levels. This

permitted graphing of vertical profiles up to 15,000' msl.

d) 1938 CV990 Flights. Data for these flights were taken at three second intervals. Vertical profiles were obtained by averaging data over thirty seconds and plotted with respect to height for the mean time over that 30 second time interval. The maximum height for these profiles is 35,000' msl.

e) 1938 CV990 Flights. These data were also recorded on magnetic tape. The altitudes were related to time as for CV990 Flights in 1937.

Chapter V

METHOD OF ANALYSIS

5.1 Purpose of analysis

By evaluating the data through the process of division and classification discrimination of the main principles of aerosol distribution and transportation in the troposphere is possible. This approach is the one use in solving the questions asked in Chapter I.

Junge (1963) found a substantial difference between the particle count for a maritime (oceanic) air mass and a continental air mass (at least 10%). Then, will the air mass source (maritime or continental) contribute significantly to the number of haze particles over a given location?

Another contributing factor could be the surface properties of the location in question. The surface properties can be related to either human activity (populated and non-populated regions) or geography (land and ocean areas). Then the question asked would be: Does the surface contribution to the atmosphere significantly influence the haze count in the troposphere directly above a region?

The effects of precipitation rain-out and wash-out on aerosol (haze) amount is a problem that has not been adequately surveyed. Do unusual instances of precipitation (or no precipitation) alter haze counts below cloud layer?

Another synoptic parameter is the effect of convective activity on haze where vertical wind and temperature variables change quickly. Can convective motion appreciably change the vertical structure and numerical count of haze particles?

The above questions dwell in the dimension of the vertical, but variations in the horizontal plane are also important. Questions of aerosol transport would be easier to answer if these variations could be found and understood. Then by deductive elaboration, one asks:

profiles were classified according to what surface area the majority of the flight covered. All borderline cases were classified urban to retain as much homogeneity as possible.

c) Classification of data with respect to precipitation influences. The nature of the precipitation relationship is discussed only in general and extreme value terms for aerosol situations resulting from recent periods of no precipitation versus recent periods of sizeable amounts of precipitation. The precipitation climatology of each of five sets of aerosol data were examined. Flights from the ends of the "precipitation spectra" were compared.

Data were obtained on flights where there had been no precipitation reported in that area (nearest station) at least six days prior to flight date. These data were compared to where there was at least 0.2 inch of precipitation in the last 48 hours and at least 0.4 inch rain in the last seven days.

d) Classification of data with respect to influence of convection. Weather associated with frontal systems is normally convective in nature. Therefore, if there has been a frontal system in the area in the last 24 hours as observed by the daily weather maps or by a local observer then aerosol data obtained were designated as a set for comparison with the rest of the data.

e) Classification of data with respect to horizontal variability. The classification of horizontal data was not as rigorous and was organized according to altitude. Four basic levels (surface, 4000' msl, 1000' msl, and 3500' msl) were selected for study. Representative cases were used for these levels.

f) Classification of data with respect to time and place. As indicated earlier primary data separation was: (1) 1966 CV990 Flights, (2) 1967 Line Island Flights, (3) 1967 CV990 Flights, (4) 1967 Comanche Flights, and (5) 1968 CV990 Flights. The synoptic situations varied with each.

Temperature data were not classified but individual case studies were examined.

Chapter VI

RESULTS

6.1 Air mass source comparison

Fig. 6.1.1 illustrates average haze profiles from all data from mairtime air and all data from continental air. Table 6.1.1 shows the average number of values used in Fig. 6.1.1 for each point in the interval sfc to 15,000' msl and in the interval 15,500' msl to 35,000' msl, and the average standard deviation for each of these intervals. The continental profile does not decrease with altitude up to 17,000' msl. The maritime data started at a higher value at the surface but decreased with height. There is for both maritime and continental data, at the upper levels, more variation for the thousand foot intervals (this is especially noticeable in the continental profile above 15,000' msl). Fig. 6.1.2 depicts the corresponding temperature profiles for the total maritime and continental data. It is interesting to notice the similarity in the two profiles. Noticeable features are: (1) the small inversion at the surface for maritime air, (2) the inversion situation starting at 14,000 feet for both sets (the inversion is particularly strong for continental air) (3) the inversion at 21,000 feet for both cases (4) the strong inversion at 24,000 feet for the maritime data. In Fig. 6.1.3 total aerosol accumulation is plotted. The most striking feature is the intersection of the two curves at approximately 20,000' msl. There are more haze particles in the whole troposphere for a continental air mass than for a maritime air mass, however, in the lower half of the troposphere there are more particles in a maritime source.

To examine maritime air directly over an ocean surface the Line Island profiles were examined in detail (Fig. 6.1.4 and Table 6.1.2). This extreme maritime situation had a high surface aerosol count which decreased rapidly up to 14,000' msl. During the flights

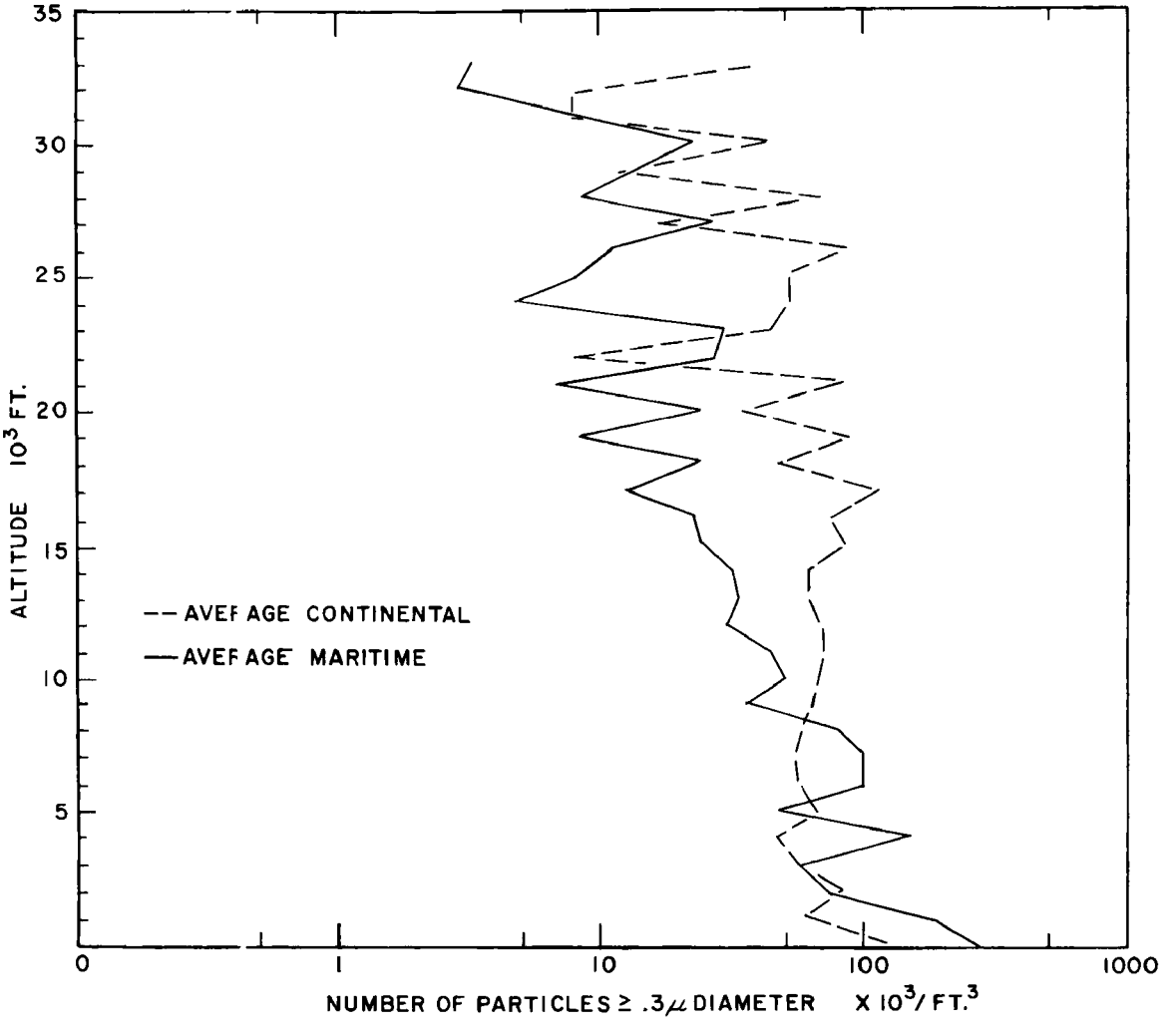

Fig. 6.1.1

Average haze profiles for maritime and continental air masses.

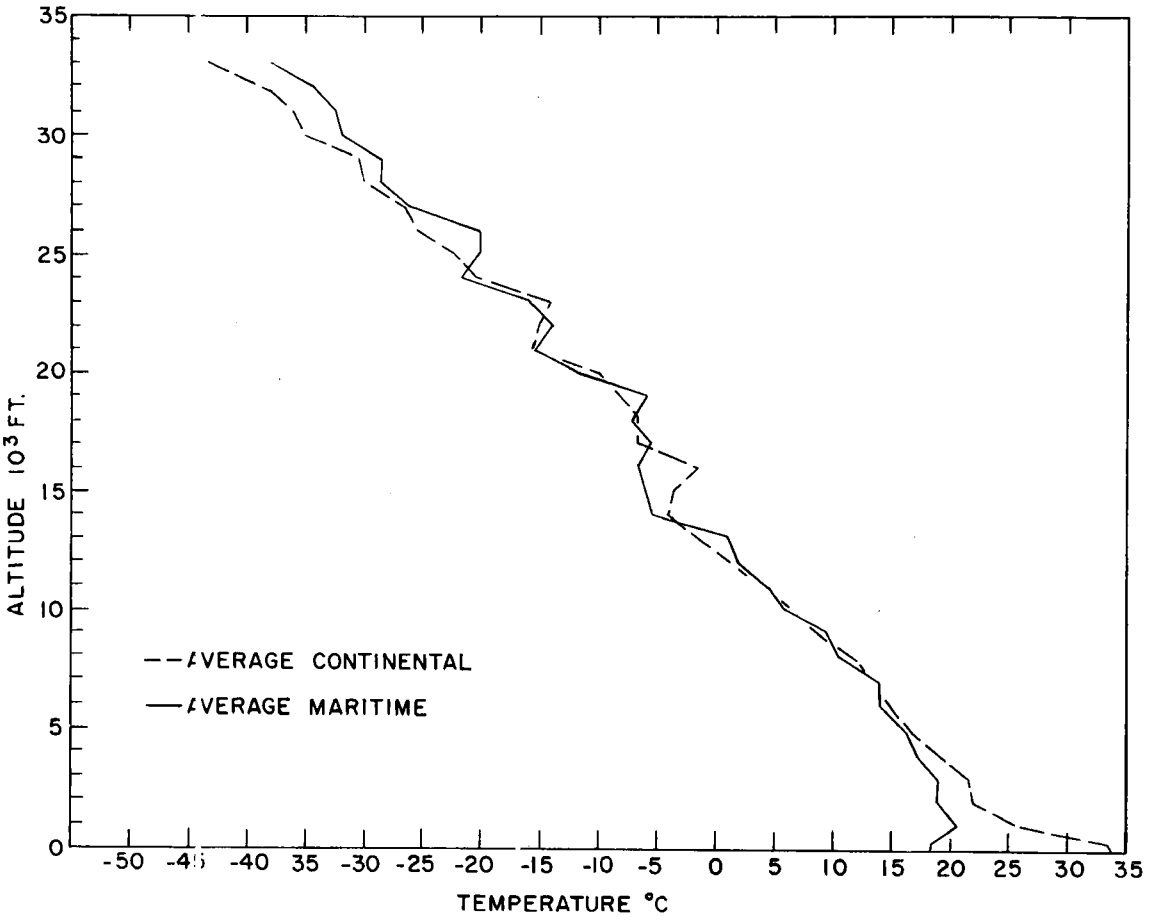

Fig. 6.1.2

Average temperature profiles for maritime and continental air masses.

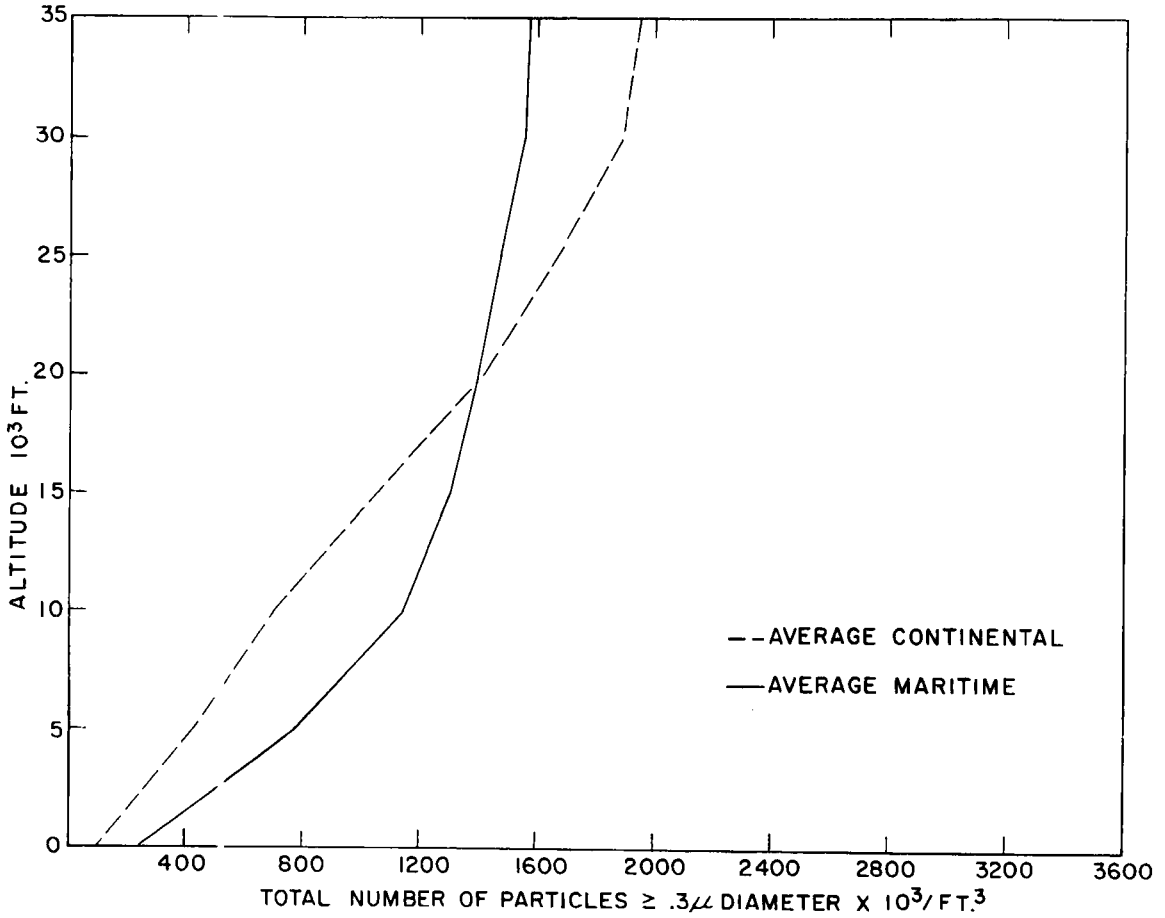

Fig. 6.1.3

Average total accumulations for maritime and continental air masses.

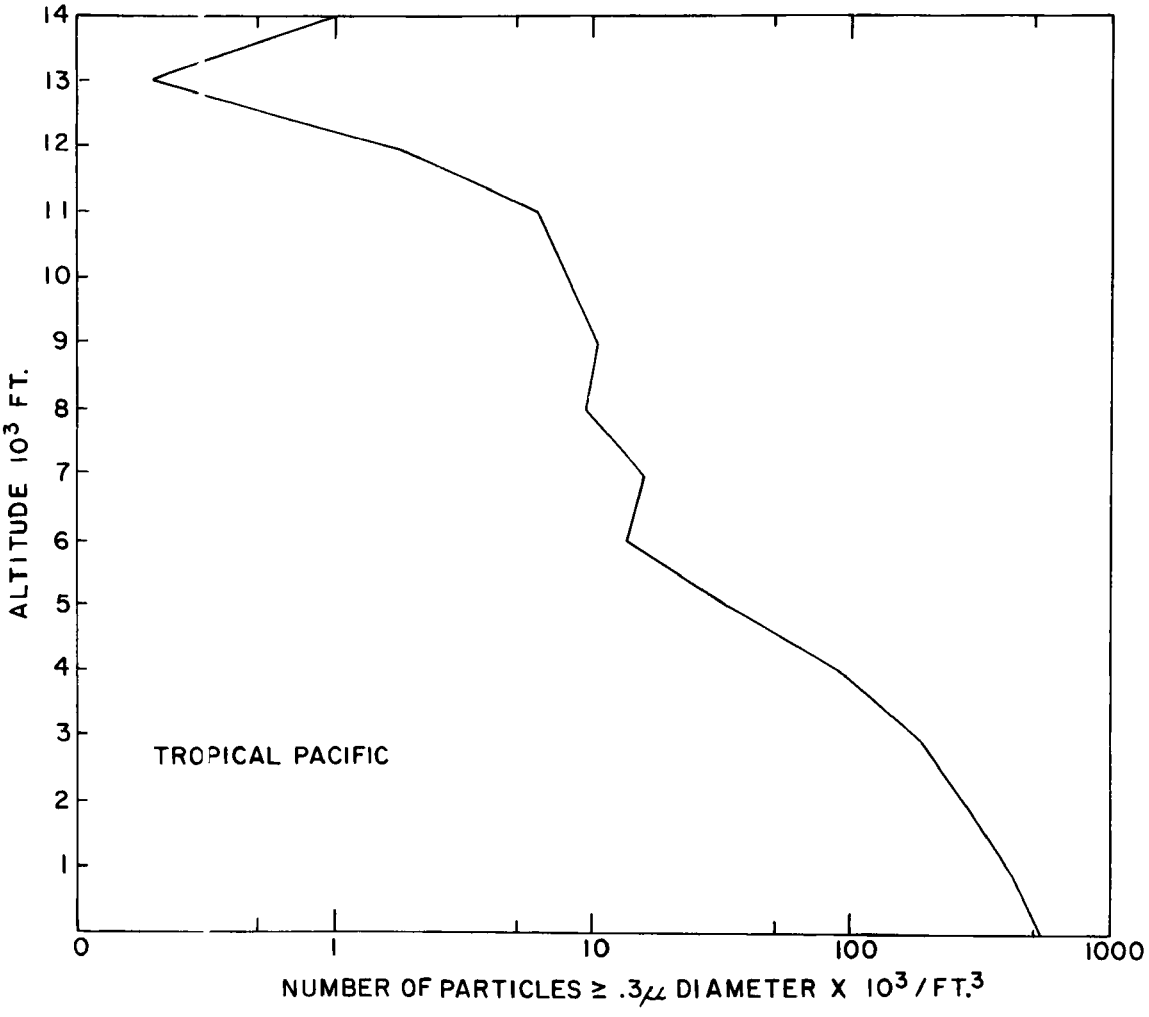

Fig. 6.1.4

Average haze profile for tropical Pacific air mass.

Table 6.1.2

Same as Table 6.1.6 excpet for points plotted in Fig. 6.1.4

		Line Islands
Average no. of points		
sfc	0-15,000'	4
	15-35,000'	0
Average standard deviation		
sfc	0-15,000'	59
	15-35,000'	0

an observer noted that, in many of the instances, there was an apparent trade wind inversion between 4,000 and 5,000' msl. This feature does not appear at least not sharply, though, on the temperature profile (Fig. 6. .. 5).

The arctic region is another area where clearly distinguishable air masses are present. The average data profiles for several flights taken in 1967 over Alaska are shown in Figs. 6.1.6 and 6.1.7 and Table 6.1.3. The arctic profile (Fig. 6.1.6) is characterized by very low particle counts and quite a decrease in count with altitude. The temperature profile (Fig. 6.1.7) indicates cold air and is marked by a strong inversion at 9,000 feet and several smaller inversions. Fig. 6.1.8 permits a comparison of aerosol accumulations profiles for Alaska and the tropical ocean.

6.2 Surface influences

a) Land versus ocean. To investigate the effect of surface influence on total haze amount and its distribution, a comparison between land and ocean profiles was done. This comparison was not based on the air mass present but rather on the nature of the underlying surface. Figs. 6.2.1, 6.2.2 and 6.2.3 shows the results of this analysis. Table 6.2.1 gives the standard deviation and average number of observations that were used in Fig. 6.2.1. The total land aerosol profile has many more data points that does the oceanic. The number of particles is greater for oceanic areas below 16,000' msl than for land areas. The average land profile gives a smoother profile and does not decrease with altitude as rapidly. The average temperature profile shows a strong inversion at the surface over the ocean. The profiles are both irregular except for the land profile section below 14,000' msl. The total accumulation curve (Fig. 6.2.3) shows a substantial difference between the two cases; the total ocean curve has the same shape as the maritime curve but is almost double the total haze amount at 35,000 feet.

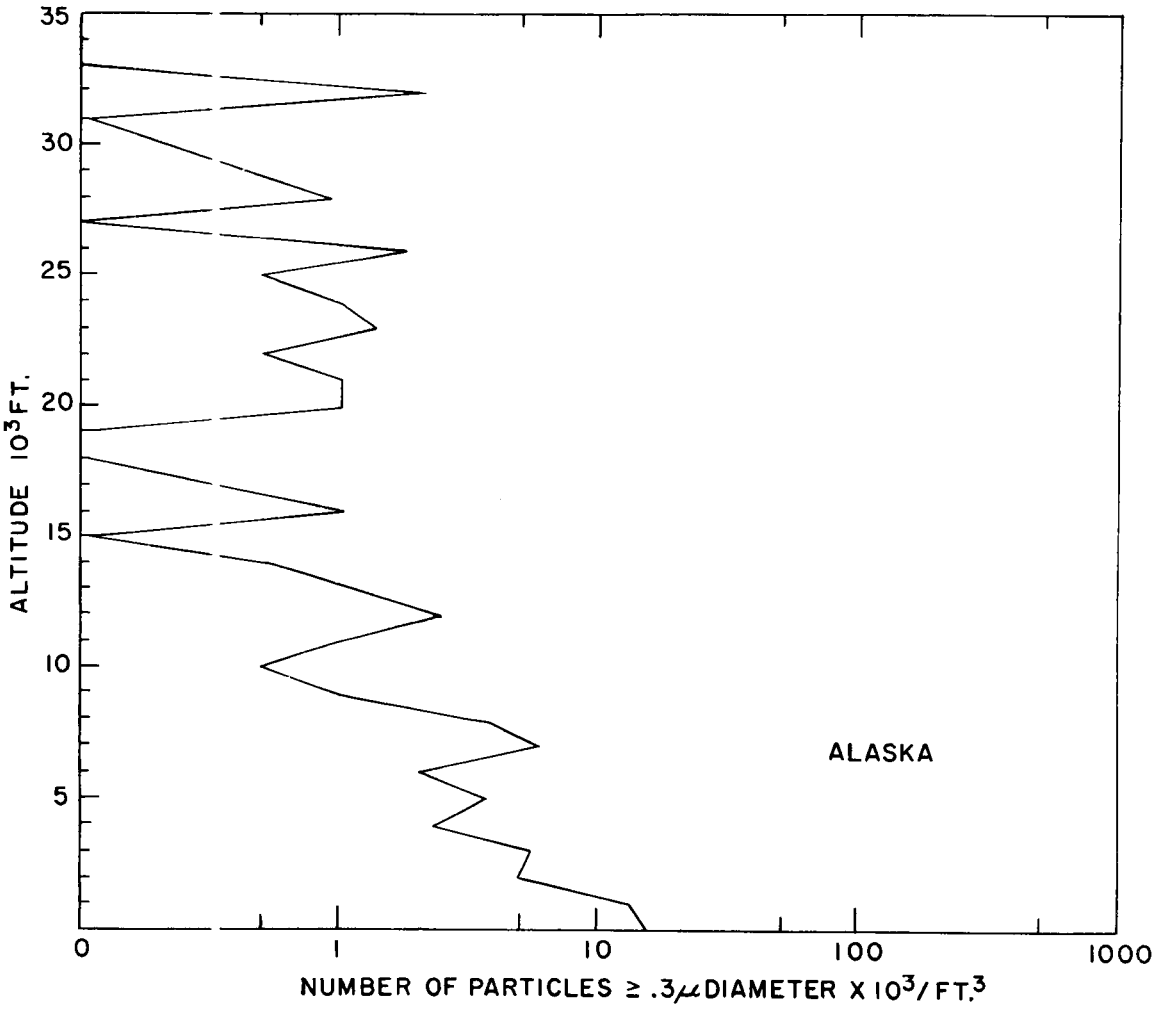

Fig. 6.1.6

Average haze profile for Alaskan air mass.

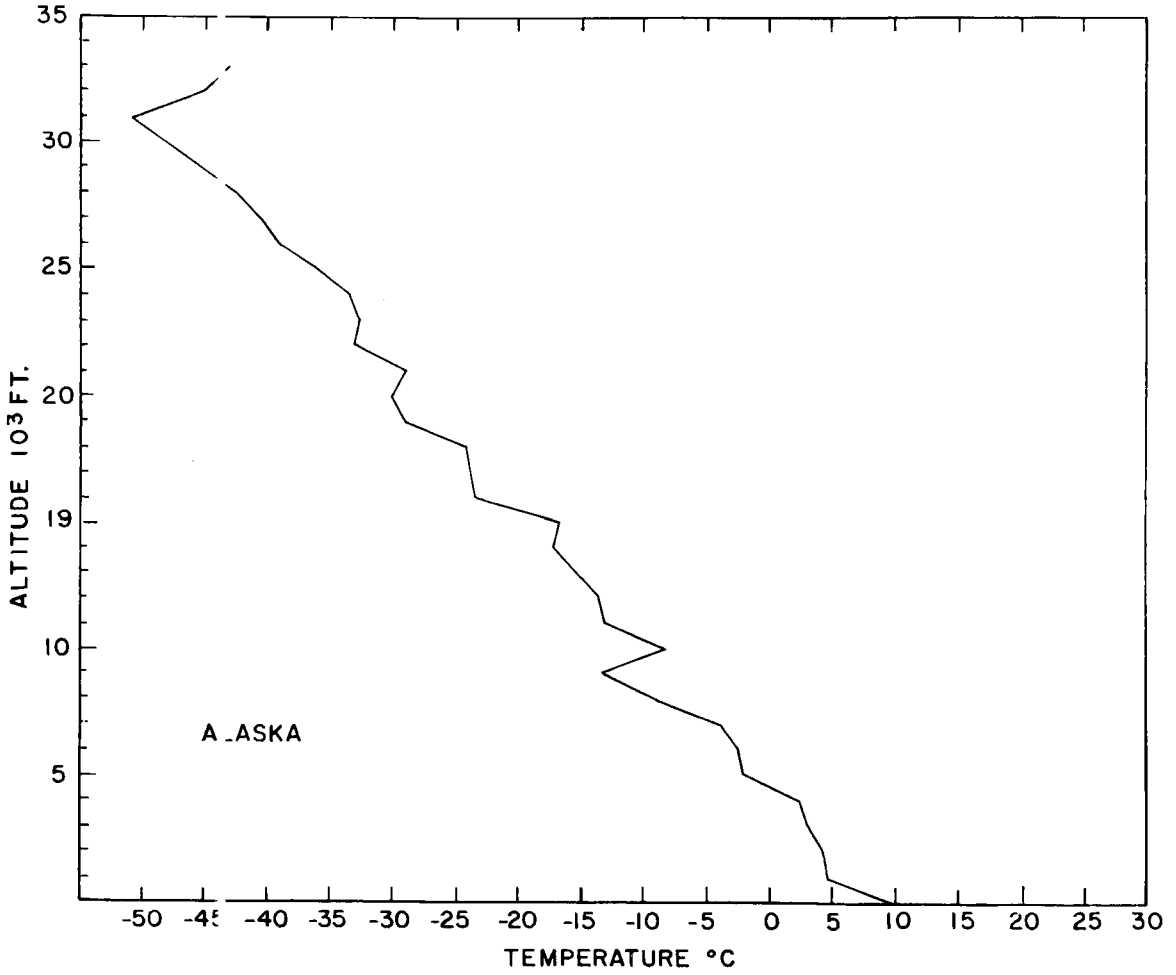

Fig. 6.1.7

Average temperature profile for
Alaskan air mass.

Table 6.1.3

Same as Table 6.1.1 except for points plotted in Fig. 6.1.6

		Alaska
Average no of points	sfc 0-7500'	8
	75-35,000'	2
Average standard deviation	sfc 0-7500'	6
	75-35,000'	.5

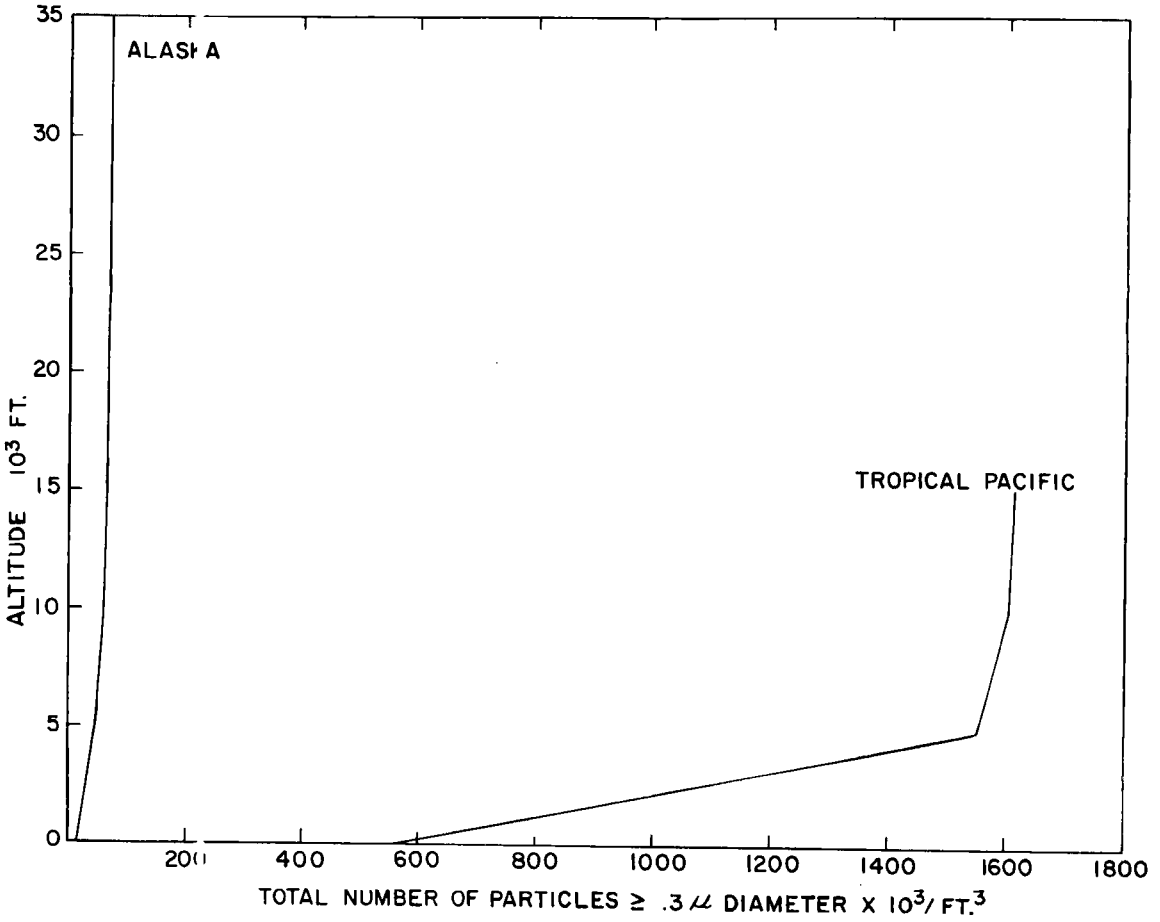

Fig. 6.1.8

Average total accumulations for tropical Pacific and Alaskan air masses.

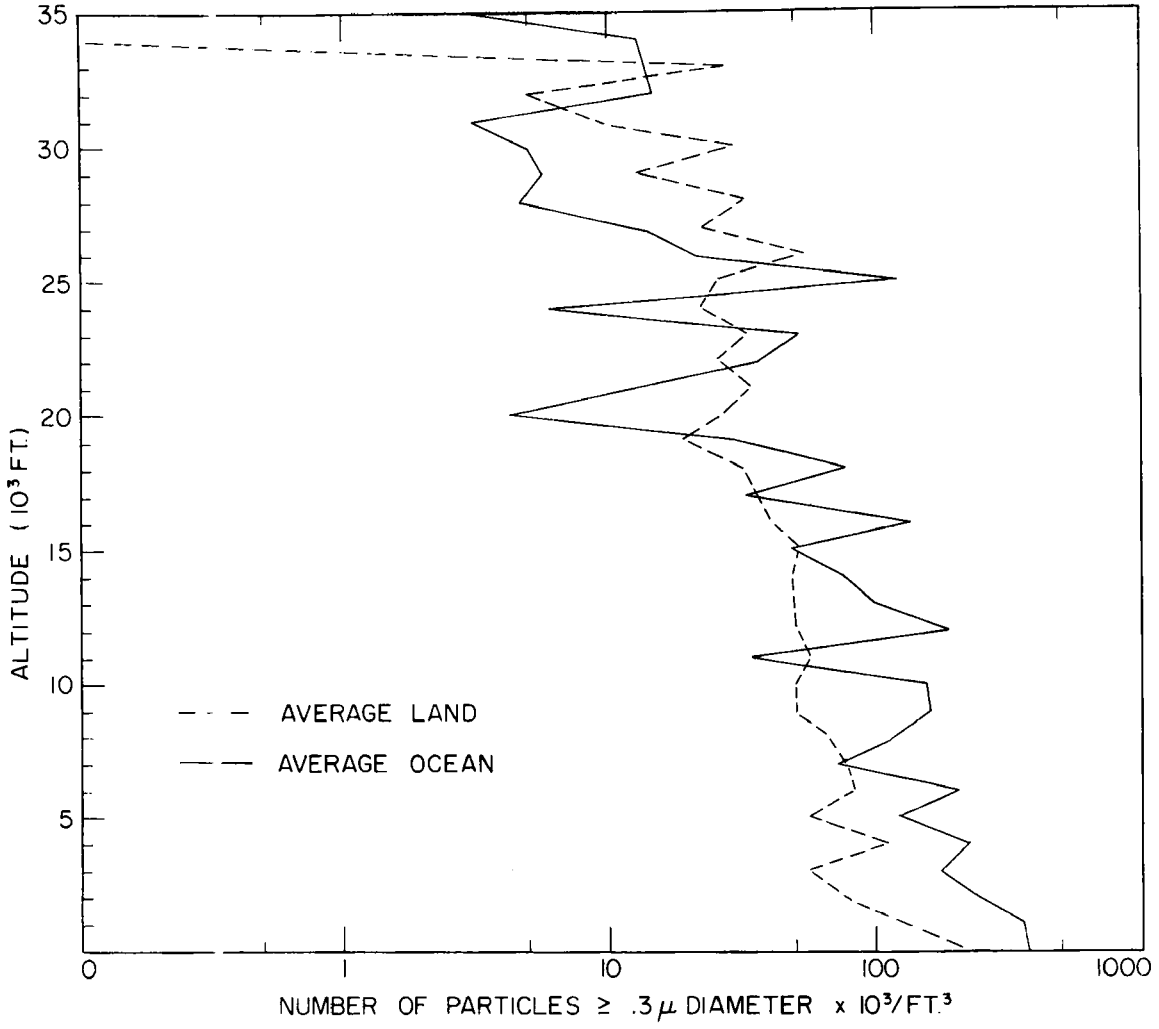

Fig. 6.2.1

Average haze profiles over ocean and land surfaces.

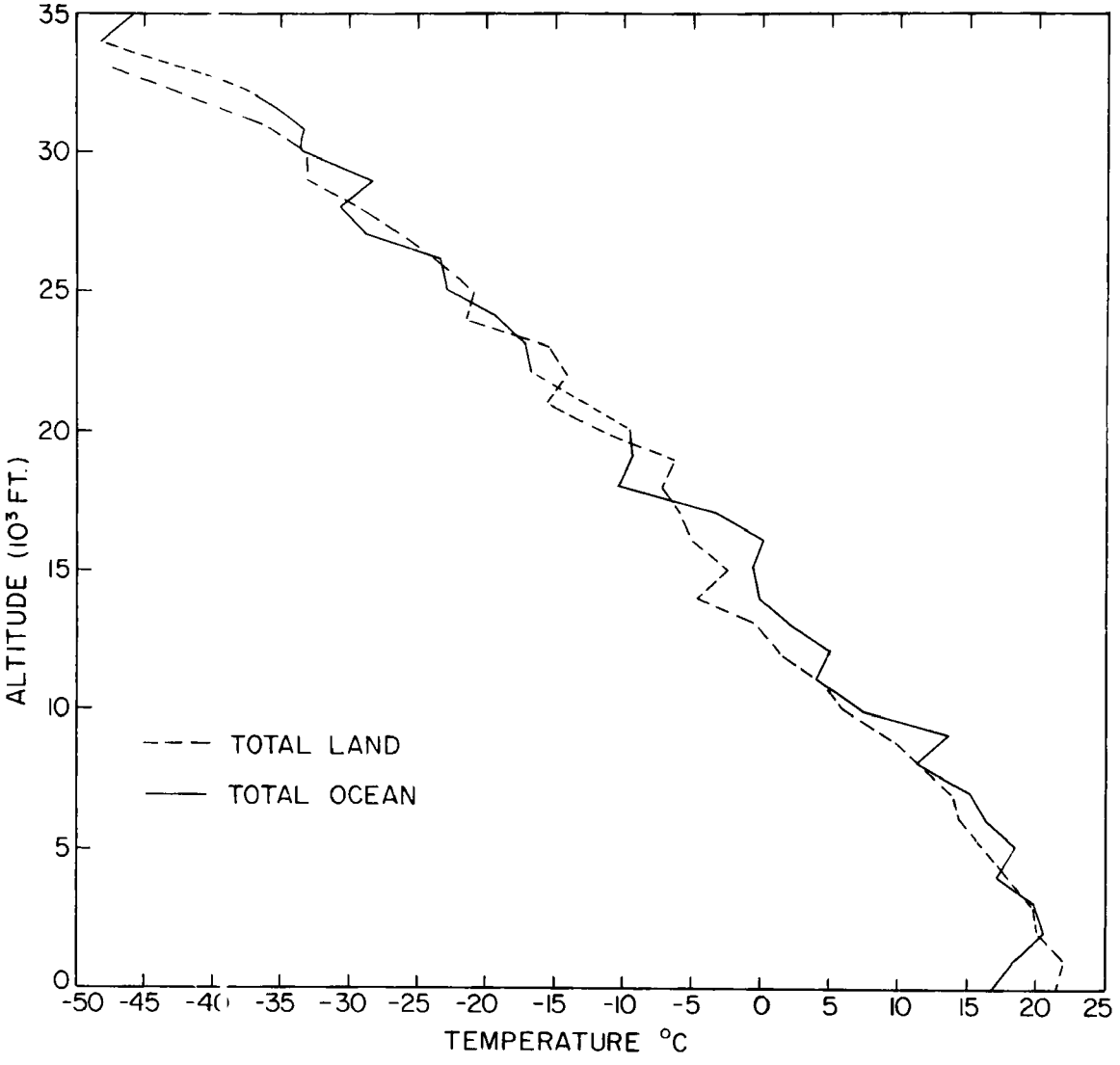

Fig. 6.2.2

Average temperature profiles over ocean and land surfaces.

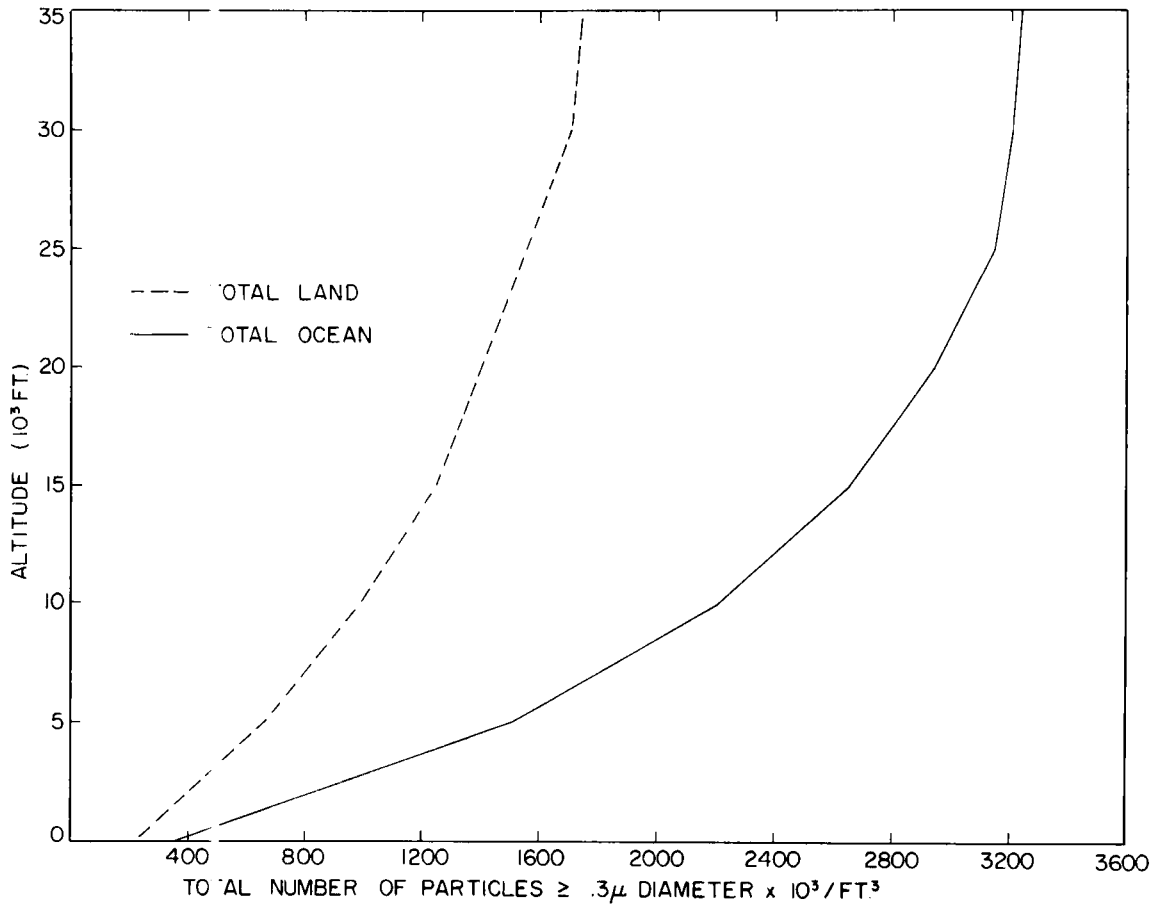

Fig. 6.2.3

Average total accumulations over ocean and land surfaces.

b) Urban versus Rural. The surface contribution to aerosol populations from average rural and urban profiles is shown in Figs. 6.2.4, 6.2.5 and 6.2.6. Table 6.2.2 gives the appropriate values for Fig. 6.2.4. An interesting feature of average haze particles of both groups is shown in Fig. 6.2.4 where both profiles do not show any significant differences below 20,000 feet. In this region they both decrease with height. Above 20,000' msl the rural area haze decreases with increasing height, while the urban area haze does not show this trend.

The temperature profile of these two sets is shown in Fig. 6.2.5. Extremely interesting is the similarity of the two from 3,000' msl to 16,000'msl. Above 15,000'msl the average temperatures are more irregular with the urban profile being somewhat warmer. The large temperature inversions from the surface to 3,000' msl and at 14,000' msl of the urban average data is also noted.

The total aerosol accumulations over the urban and rural areas is illustrated in Fig. 6.2.6. At the lower levels the urban areas appears to have a few more particles than the rural area. This difference increases at upper levels. The increase of particles with height appears more constant over the average urban area.

6.3 Effect of precipitation of haze

The precipitation classes mentioned previously were analysed and are depicted in Figs. 6.3.1, 6.3.2 and 6.3.3. Table 6.3.1 gives the average values for each level for this comparison. In Fig. 6.3.1 there is a marked difference noted between these two profiles in the region from the surface to 15,000' msl; the recent precipitation haze profile increases with height and the no recent precipitation haze profile decreases with height. Above 15,000' msl the recent precipitation data set is erratic. Although based on only a small sample, it does seem to decrease slightly and to be close to those average values of the no recent rain set. The difference in the two profiles below 15,000' msl is less at 15,000' msl than in the lower levels.

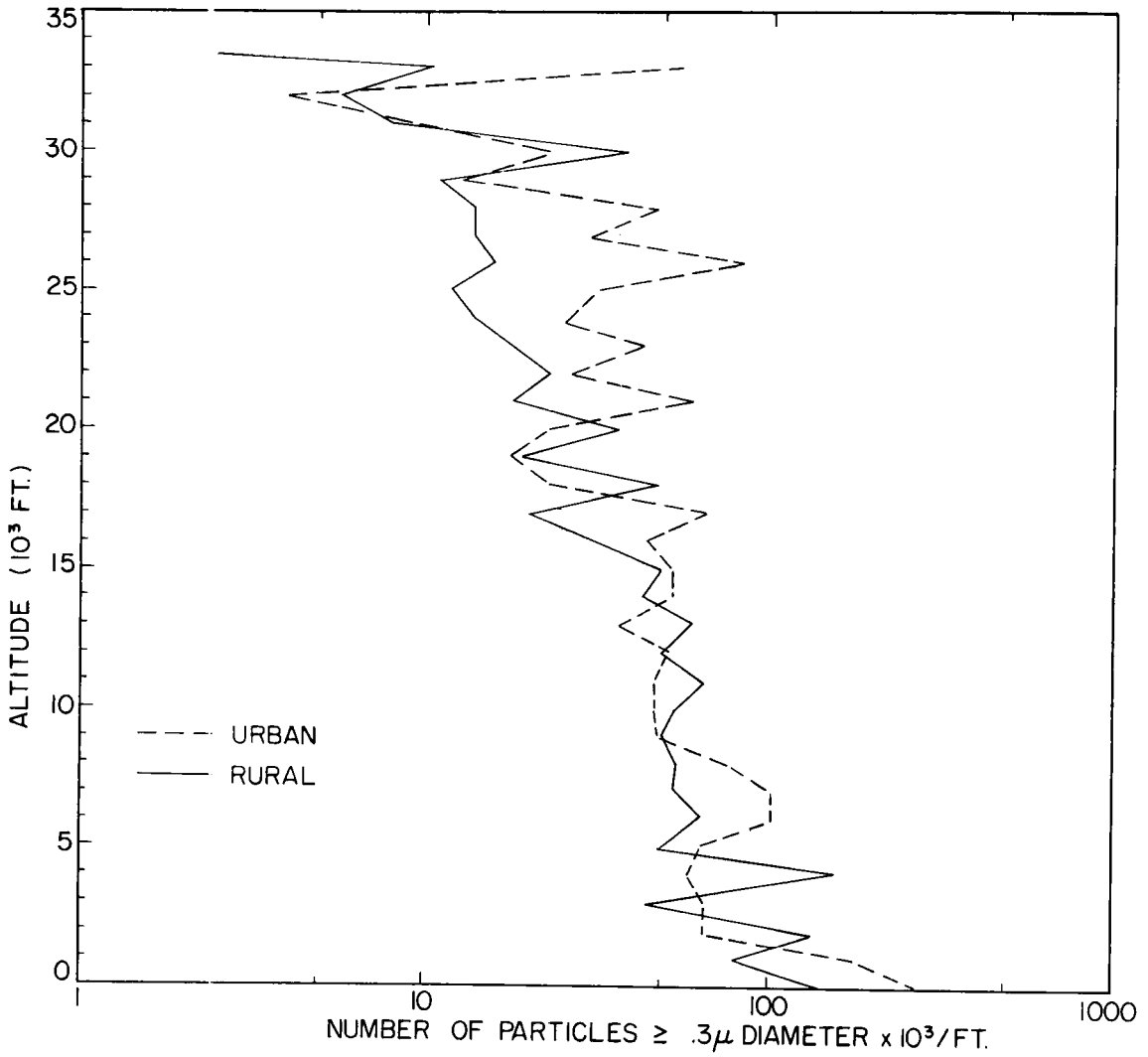

Fig. 6.2.4

Average haze profiles over urban and rural areas.

Table 6.2.2

Same as Table 6.1.1 except for points plotted in Fig. 6.2.5.

		Urban	Rural
Average no. of points			
sfc	-15,000'	21	29
	15-35,000'	6	4
Average standard deviation			
sfc	-15,000'	115	86
	15-35,000'	58	17

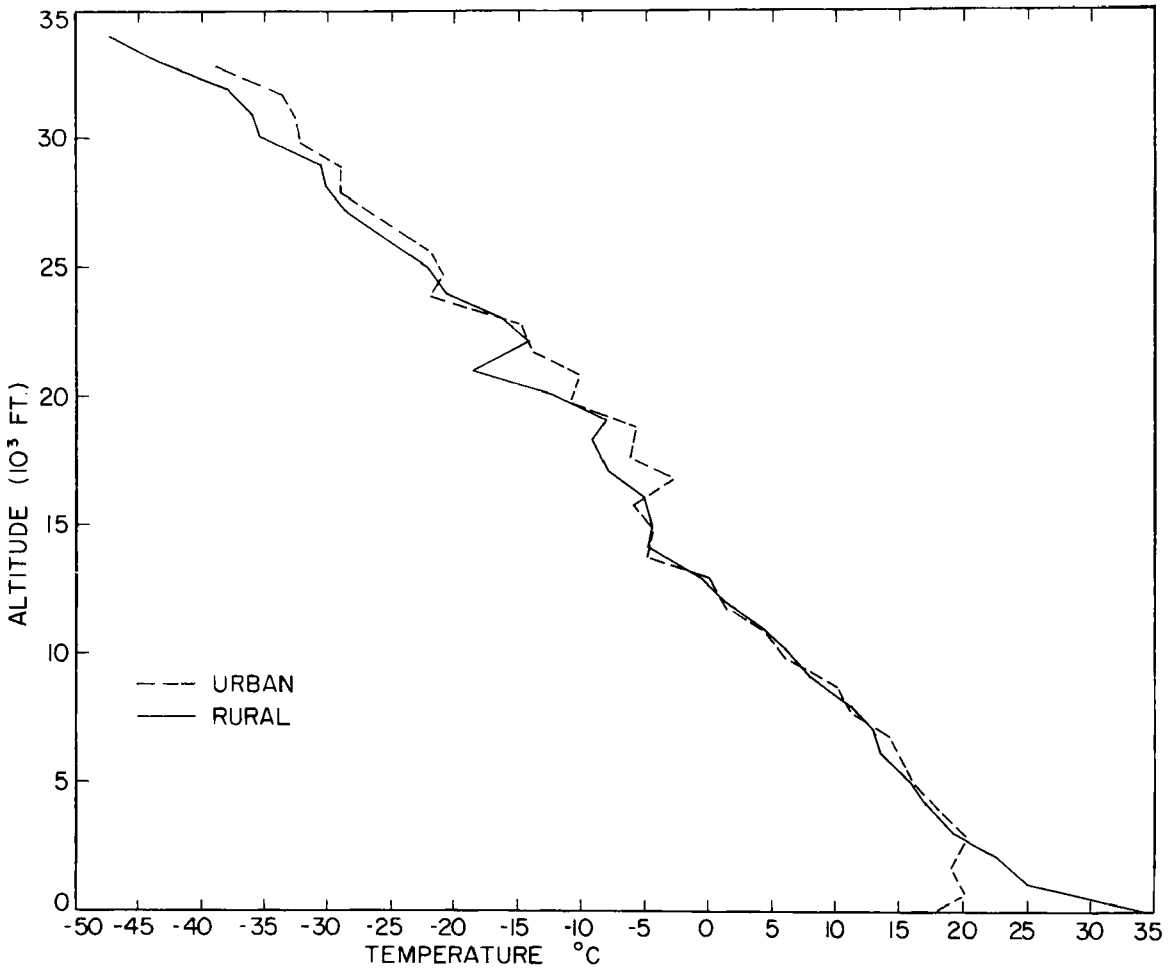

Fig. 6.2.5

Average temperature profiles over urban and rural areas.

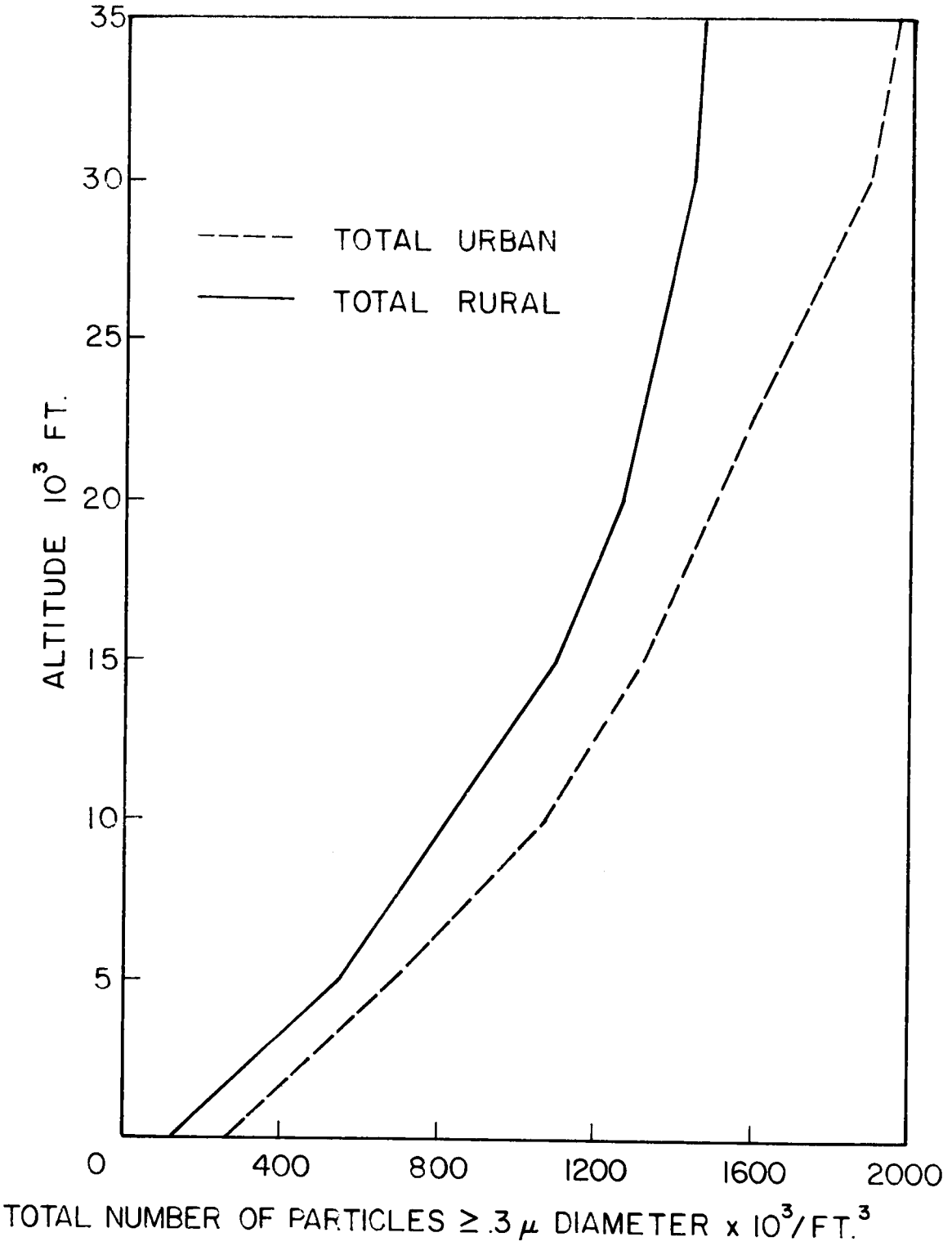

Fig. 6.2.6

Average total accumulations over urban and rural areas.

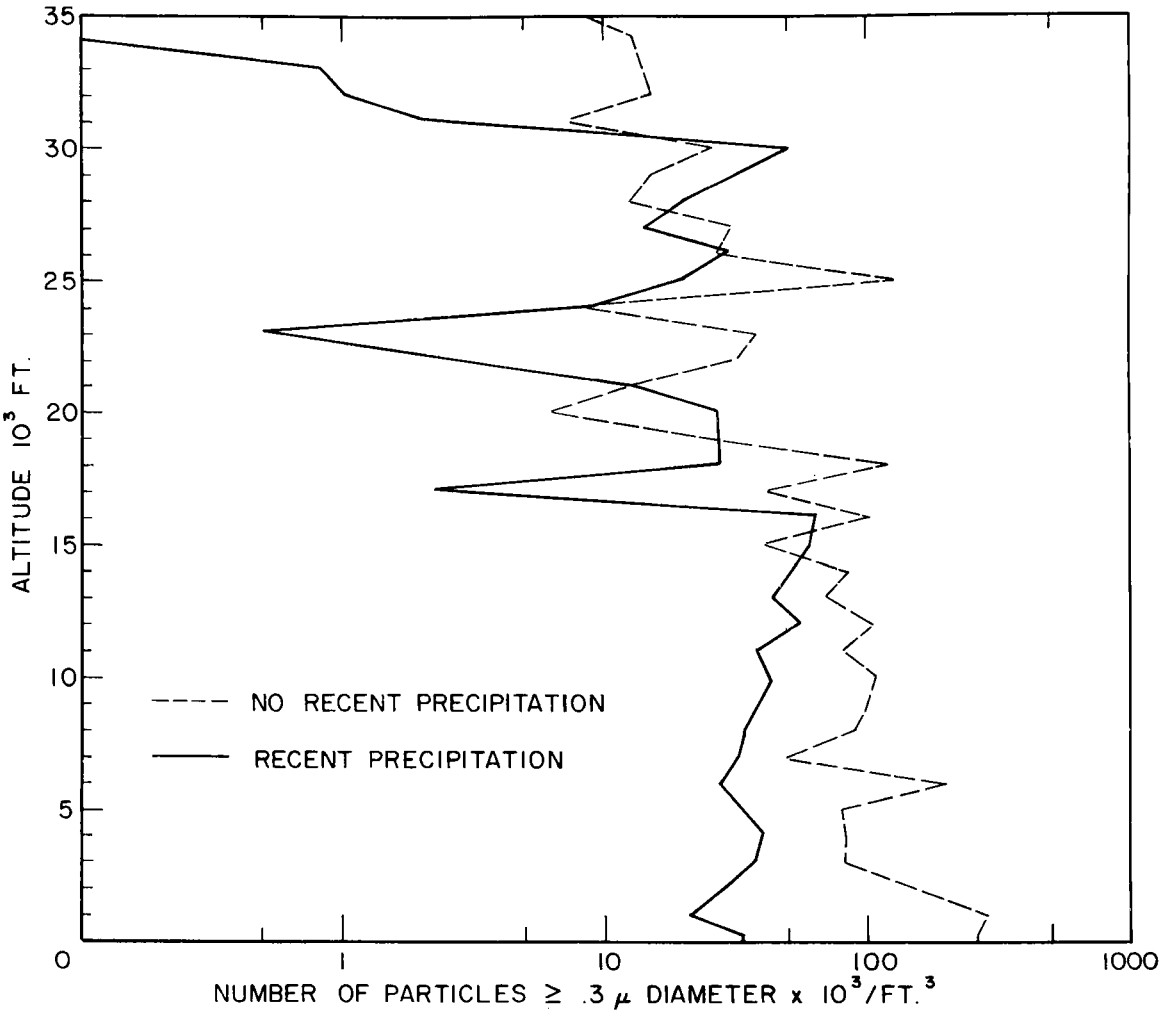

Fig. 6.3.1

Average haze profiles associated with recent precipitation and no recent precipitation.

Table 6.3.1

Same as Table 6.1.1 except for points plotted in Fig. 6.3.1

		Recent Ppt	No Recent Ppt
Average no. of points			
sfc	-15,000'	15	14
	15-35,000'	2	5
Average standard deviation			
sfc	-15,000'	40	135
	15-35,000'	19	52

The temperature profiles in Fig. 6.3.2 further indicate the contrast of these two situations. The recent precipitation set was significantly cooler than the set in which it had not rained for at least six days. The latter situation is characterized by two strong inversions, one in the lower levels and one at 14,000' msl. Around 13,000' msl the recent precipitation temperature profile decreases very substantially with height. This is the same height region where the particle counts were the same.

The total accumulation in Fig. 6.3.3 shows the irregular shape of the recent precipitation curve and the great difference in total amount of haze particles for the troposphere.

Further examination of the two cases revealed that almost all of the data used for the recent precipitation class were data taken on 1967 CV990 Flights and almost all the data classified as no recent precipitation resulted from the 1968 CV990 Flights. Assuming no instrumental or sampling errors, the likely comparison to make is between the average profiles for each set of data and the precipitation climatology for each set of data. Fig. 6.3.4, 6.3.5, 6.3.6, 6.3.7 accompanied by Table 6.3.2 explore this possible connection. The Line Island and Alaskan data were eliminated from consideration due to poor precipitation data. Also, it must be remembered that the CV990 data for 1968 used $\geq 0.5\,\mu$ rather than $\geq 0.3\,\mu$ sized particles. This will of course tend to make the values smaller than they actually are when compared to $\geq .3\,\mu$ range. Fig. 6.3.4 shows the average profiles for each particular set of data. There is a large variation between these profiles sometimes exceeding two orders of magnitude. The data decrease with altitude with the 1967 Comanche and 1967 CV990 values decreasing less than the others. The 1967 Comanche and 1966 CV990 data seem to have less variation in the lower levels than the other two.

Fig. 6.3.5 is the total accumulation for land values during the

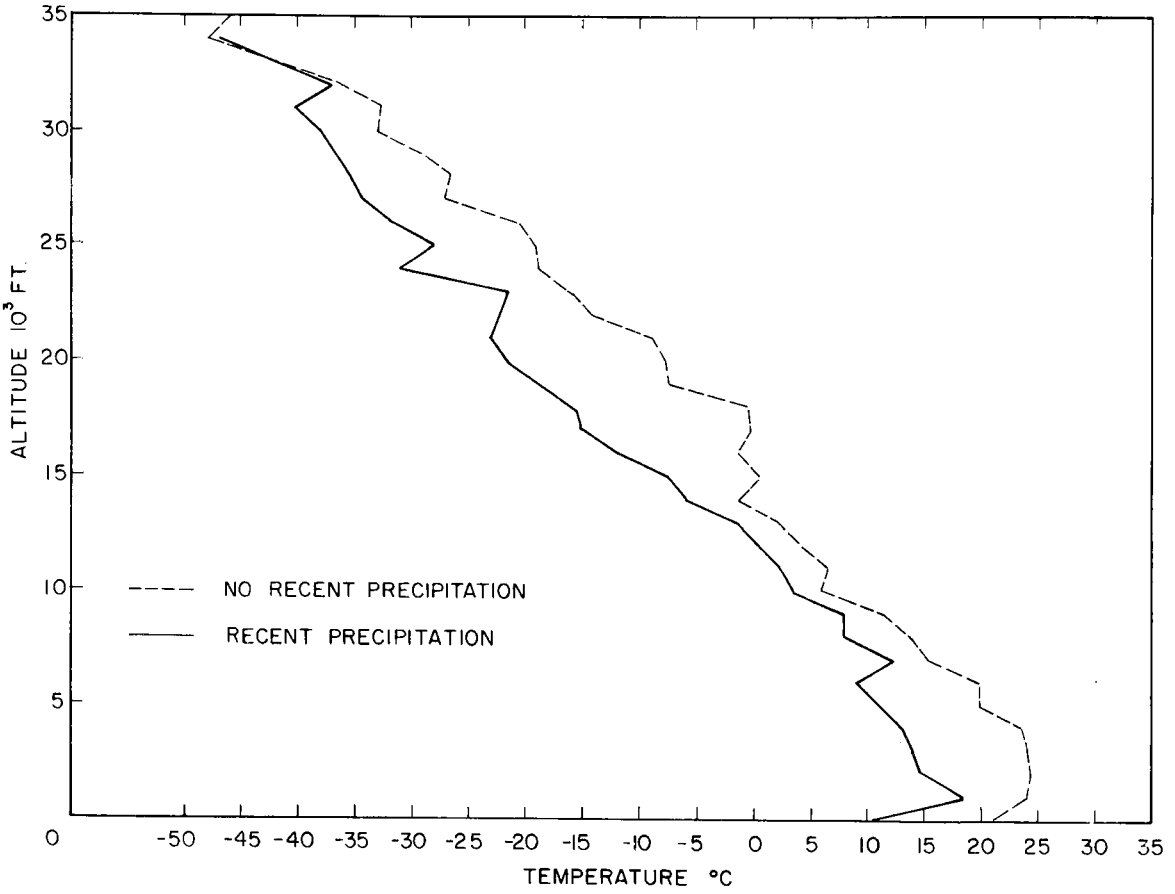

Fig. 6.3.2

Average temperature profiles associated with recent precipitation and no recent precipitation.

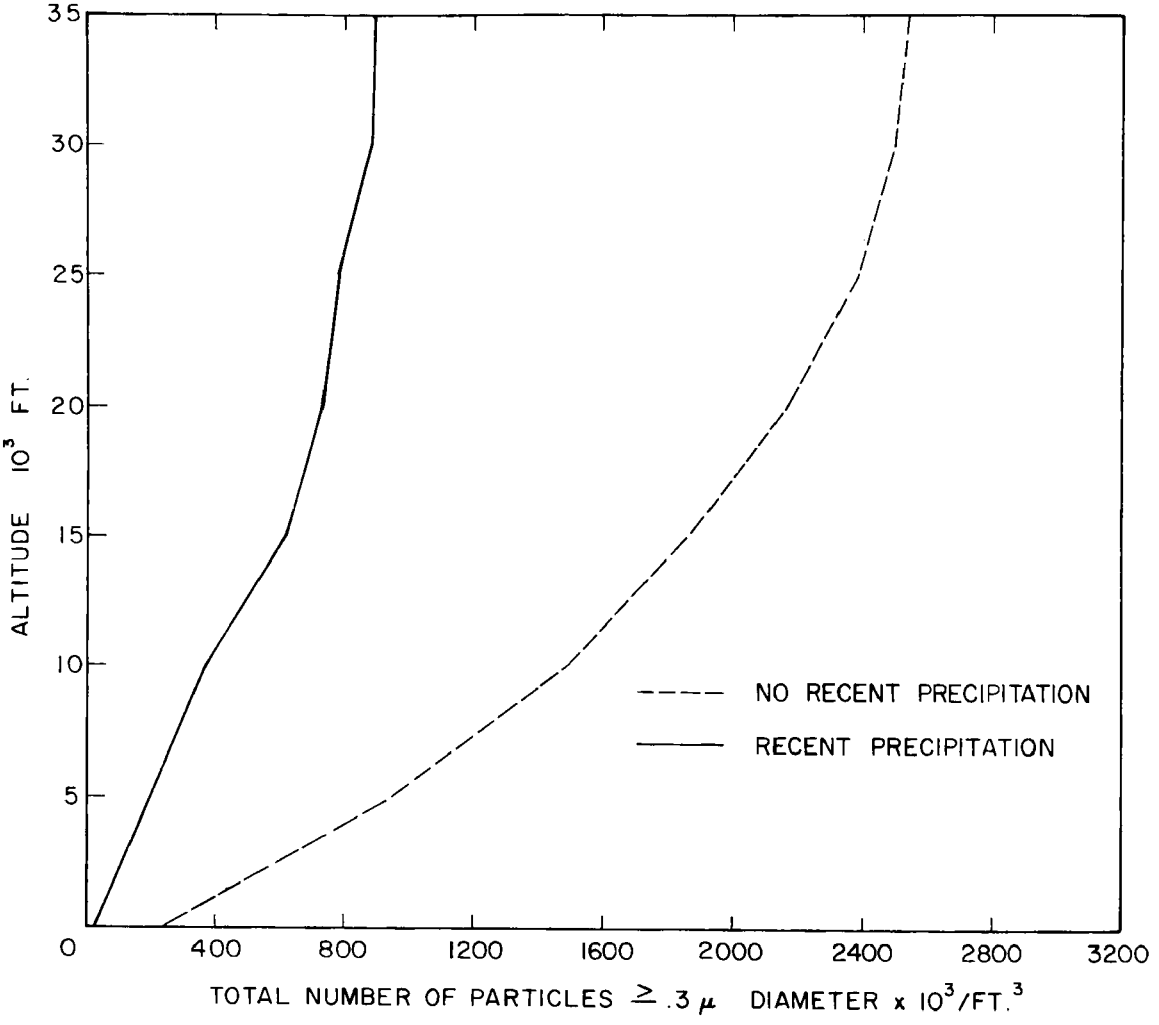

Fig. 6.3.3

Average total accumulations associated with recent precipitation and no recent precipitation.

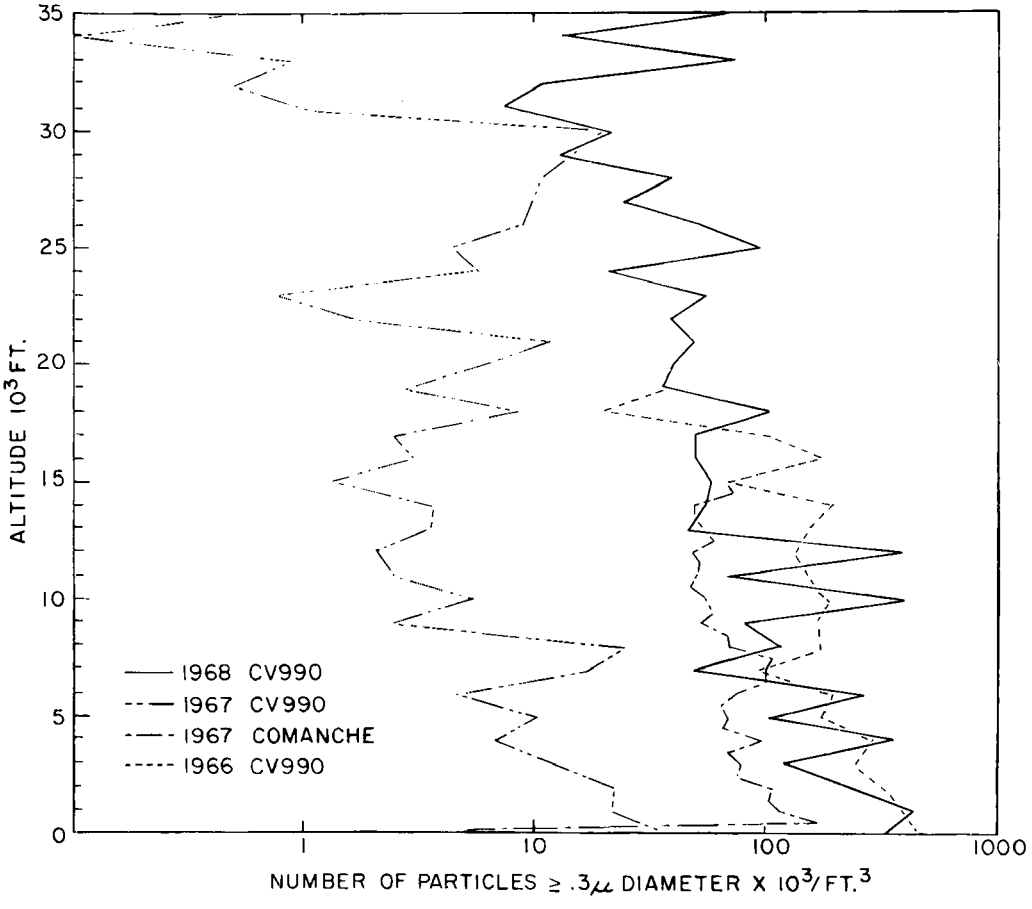

Fig. 6.3.4

Average haze profiles analysed according to year.

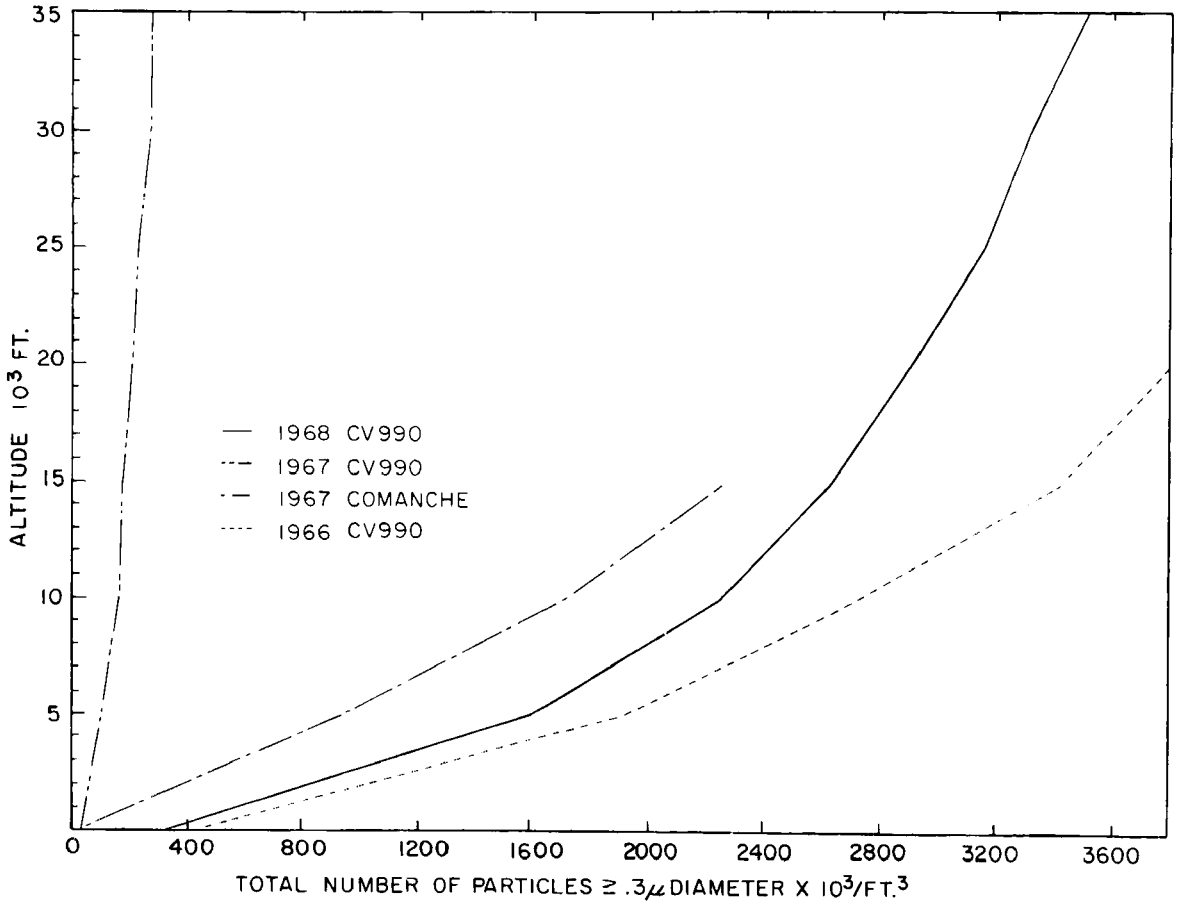

Fig. 6.3.5

Average land accumulations for 1966, 1967 and 1968 taken from CV990 observations.

three different CV990 flights. There is quite a difference between these values from the surface to the top.

The temperatures shown in Fig. 6.3.6 indicate that the average air mass in the 1967 data was generally cooler than the other flights. Also, the 1967 data were more erratic than the other data with many small inversion steps. The inversion at the surface is characteristic in all the profiles.

The total accumulations (Fig. 6.3.7) again illustrate how much the haze counts can differ from time to time. The order of increasing accumulation should be noted. The total amount of haze in a vertical column extending the length of the troposphere would vary enormously from 1967 to 1968 during the times the flights were taken.

Before proceeding further, we should discuss the significance of the standard deviation. In Table 6.3.2 a simple test was made to ascertain whether $\sigma_1^2 = \sigma_2^2$ (using a F-test). It was found that only the 1967 CV990 data did not conform to any significance level. But in examining the situation further it was found that the standard deviation mirrors the magnitude of the haze count. Due to the fact that the lower end is limited to zero the lower counts will probably have lower σ and vice versa. Then the importance of σ has been minimized.

For a comparison the average number of days since rain prior to flight, the average precipitation for this date, and the average amount of precipitation for the week preceding the flight were calculated for four cases. The results are listed in Table 6.3.3. It is seen that 1967 was the wettest period followed by 1967 Comanche, 1966 and 1968 in order. For example at San Francisco, May and June 1967 was characterized by below normal precipitation and average temperatures (almost drought conditions prevailed). At San Francisco in June 1968 0.86 inch of rain was recorded, 0.75 inch above normal and Dallas recorded 1.76 inches, 1.48 inches below normal. July and August months in 1967 were somewhat below normal precipitation for

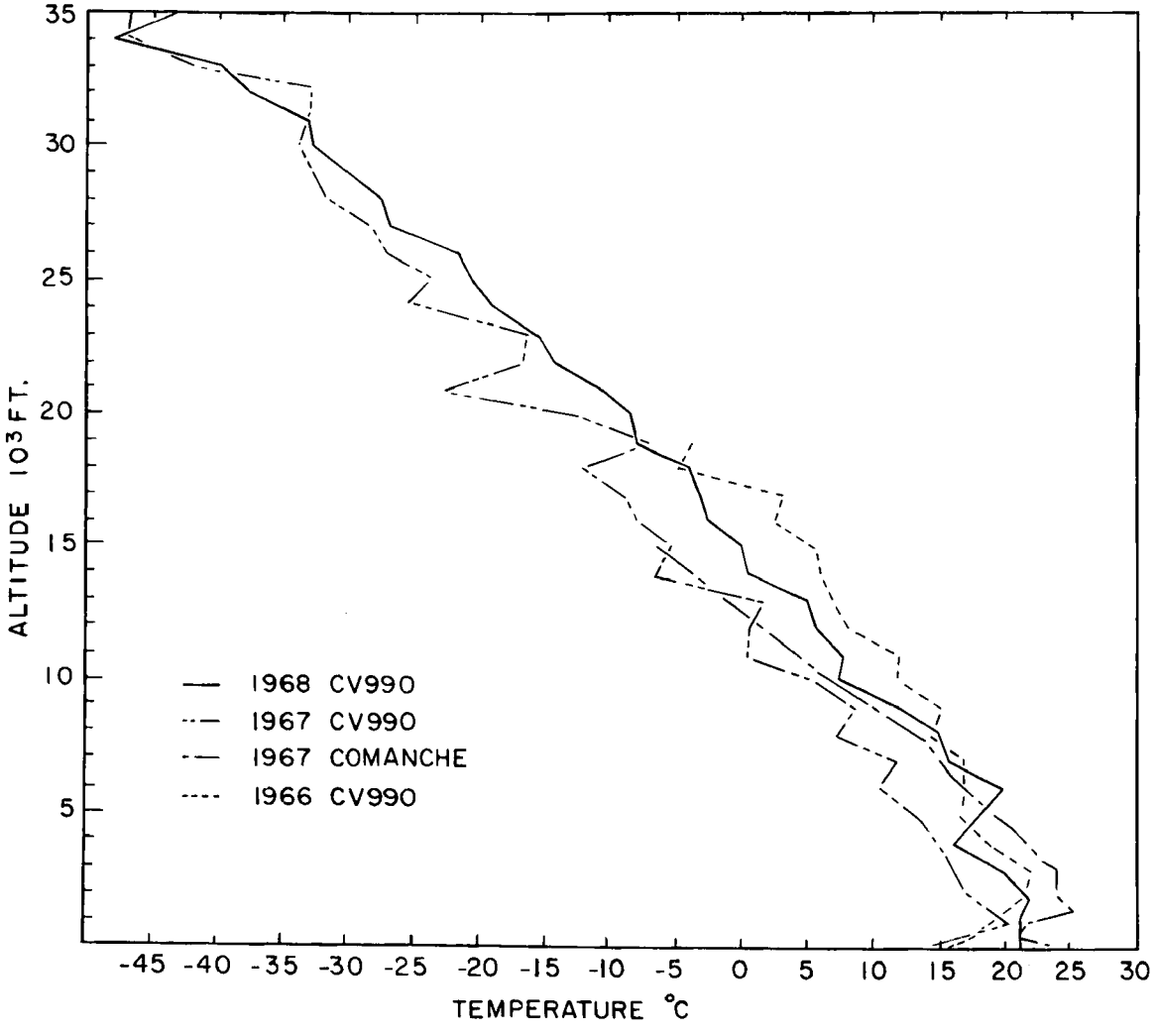

Fig. 6.3.6

Average temperature profiles analysed according to year.

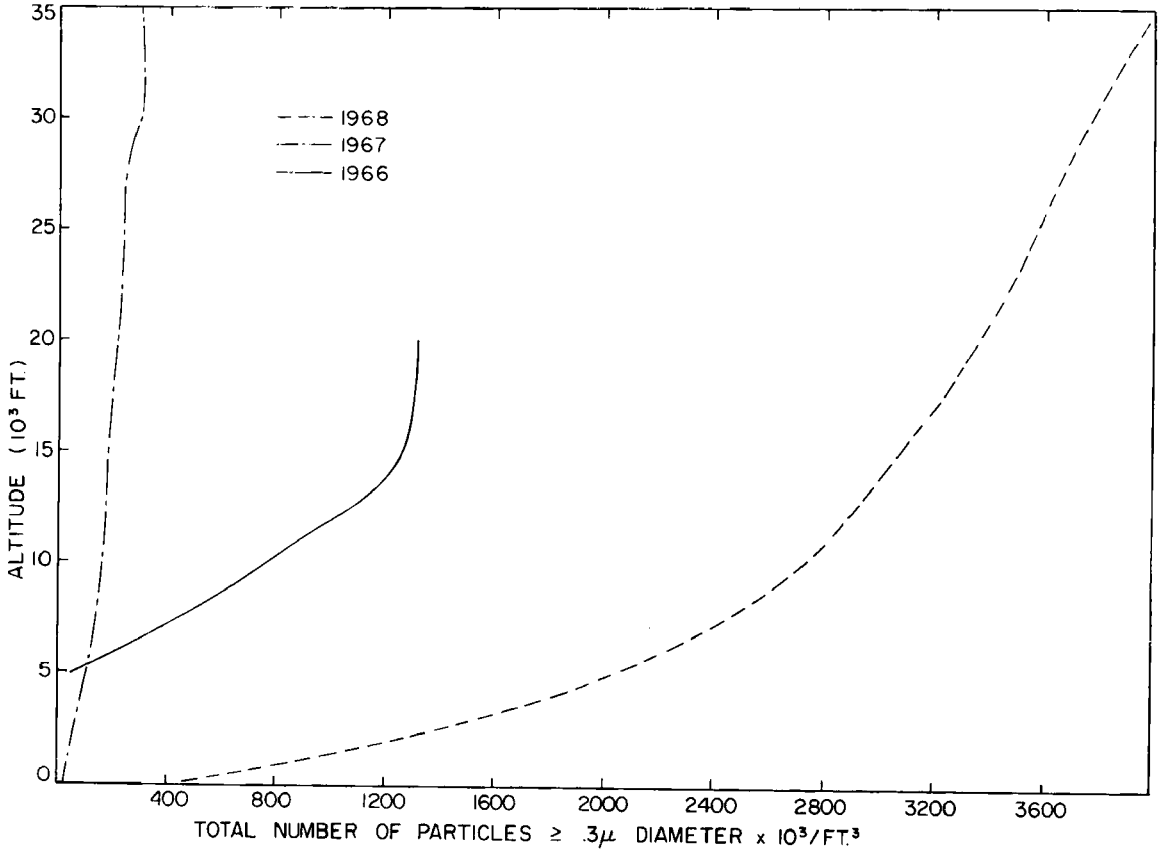

Fig. 6.3.7

Average total accumulations analysed according to year.

Table 6.3.2

Same as Table 6.1.1 except for points plotted in Fig. 6.3.4.

		Canadian	1966	1967	1968
Average no. of points					
	sfc -15,000'	19	6	18	10
	15-35,000'	0	2	5	8
Average standard deviation					
	sfc -15,000'	82	200	15	136
	15-35,000'	0	26	9	62

Table 6.3.3

Precipitation summary for yearly haze analysis.

	average days since last ppt	average amount on last ppt day (in.)	average ppt in last week (in.)
1966-CV 990	3.8	T	.10
1967-CV 990	2.1	.38	.90
1967-Comanche	3.0	.10	.24
1968-CV 990	5.4	T	.06

the general western United States. And June 1968 for San Francisco had a total rainfall of a trace (0.11 inch below normal). Thus it is seen that the 1967 data was obtained during a period of unusual rainfall conditions (record setting precipitation amounts) and the 1968 data was obtained during unusually dry conditions.

A study of the haze pattern over Dallas, Texas during seven consecutative days permits further evaluation of the effect of precipitation on haze. Figs. 6.3.8 and 6.3.9 illustrate Dallas profiles. The first day, June 1, 1967, followed three days for which the total amount of precipitation recorded was 2.15 inches. On June 1, there was a trace of rain with high humidity (75-85% RH). On the following days a warming trend occurred with no precipitation recorded. The profiles in Fig. 6.3.8 exhibit a confusing situation except in the interval 5,000 to 10,000 feet where there is an order corresponding to the order of days (except for June 1 where there was high humidity and a frontal passage with convection). On June 6 the upper troposphere had much larger aerosol concentrations than the previous days. In these profiles the shift of almost an order of magnitude may be noticed over time span of one week. The temperatures (Fig. 6.3.9) also show a warming trend.

During the 1968 CV990 flights there was one situation showing a precipitation effect. An aerosol profile was obtained over Tulsa, Oklahoma on June 12, one day after it had rained (0.2 inch). Two days later, with no recorded precipitation in the area, another profile was taken over Tulsa. Figs. 6.3.10, 6.3.11 show that the recent precipitation situation has less haze and a corresponding cooler temperature profile.

6.4 Influence of a Frontal Passage on haze

If it is assumed that frontal systems produce the greatest convective situation in summer, then by examination of the haze before and after a frontal system passage, it is possible to determine the

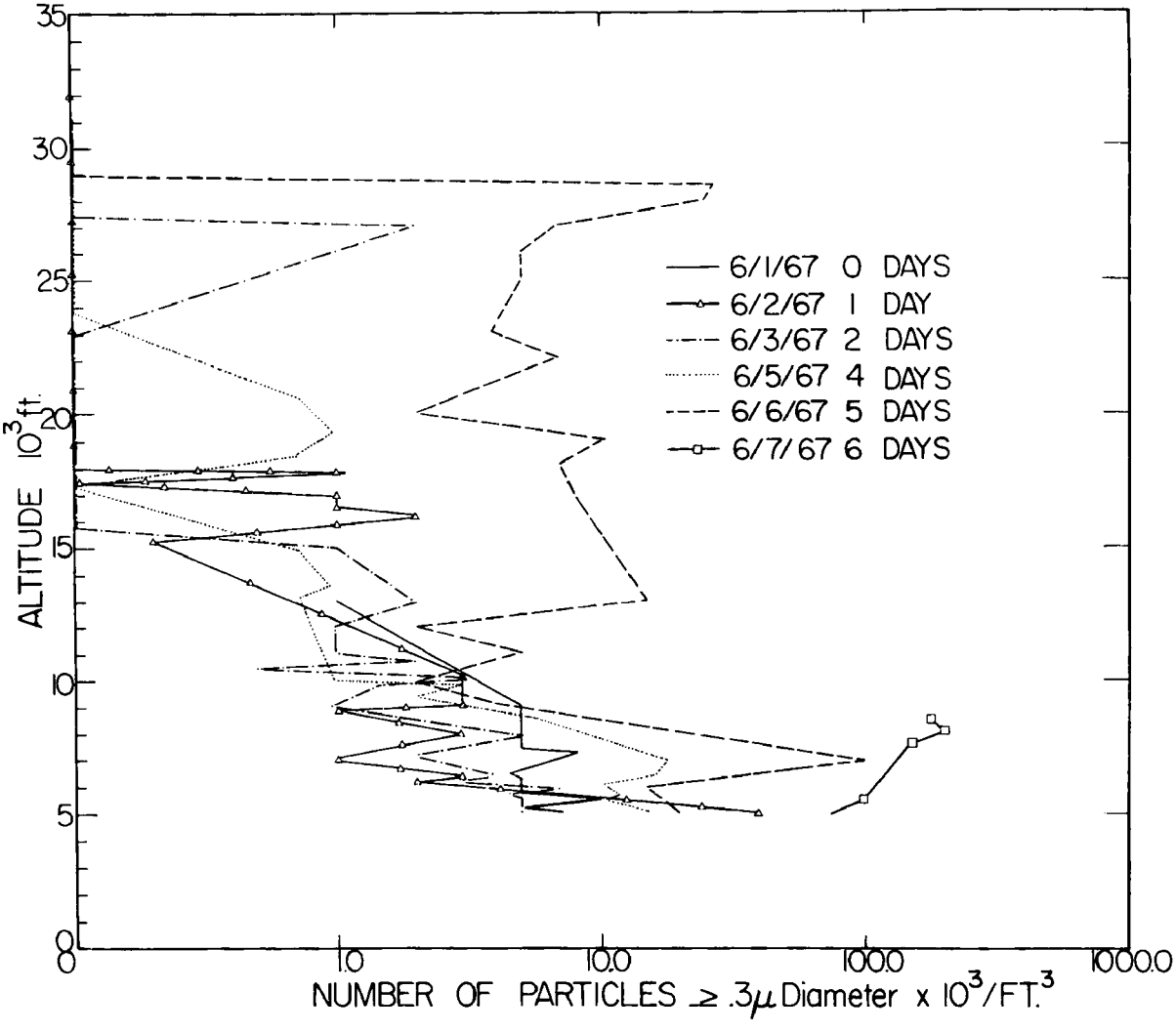

Fig. 6.3.8

Haze profiles taken over Dallas, Texas associated with varying time periods since last precipitation.

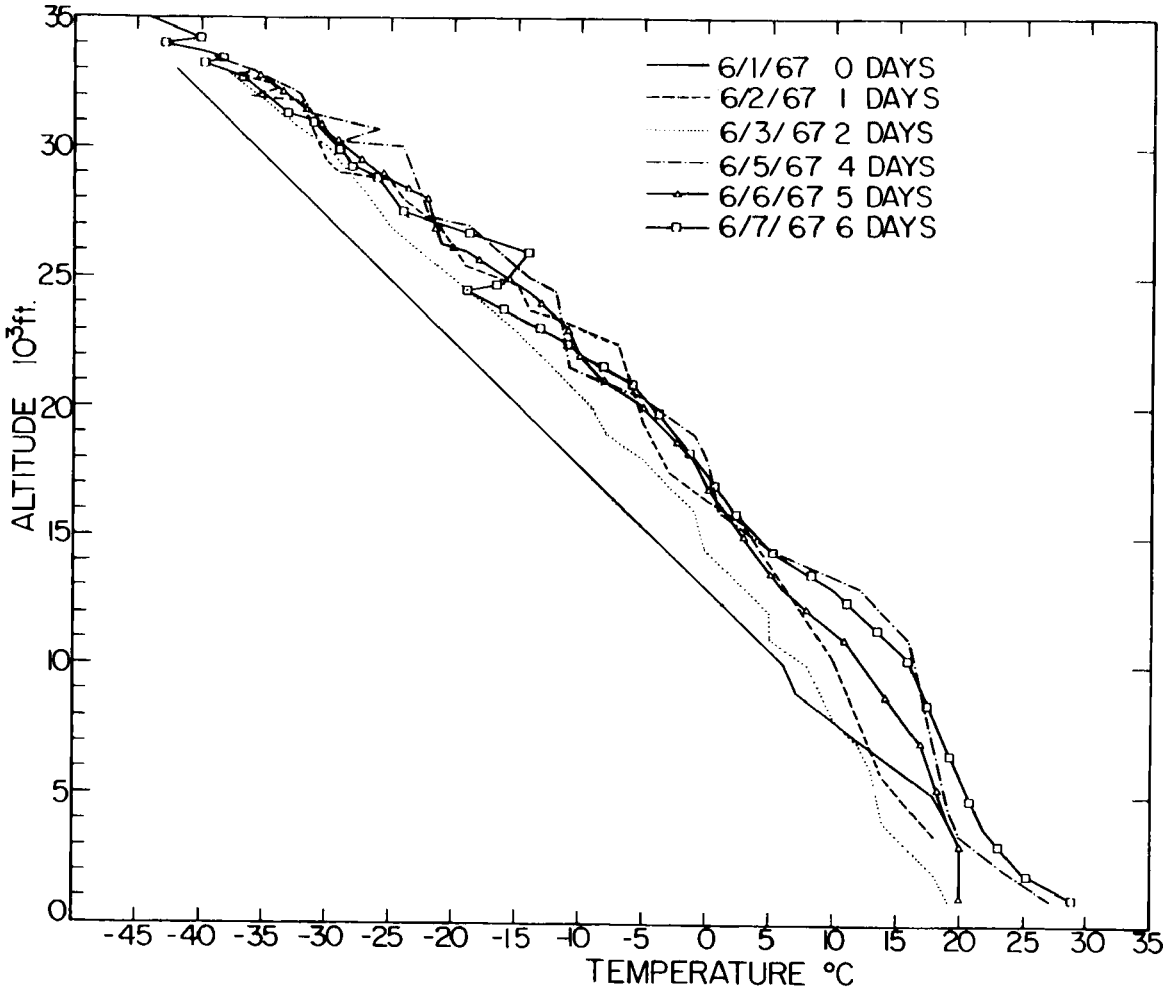

Fig. 6.3.9

Corresponding temperature profiles for Fig. 6.3.7.

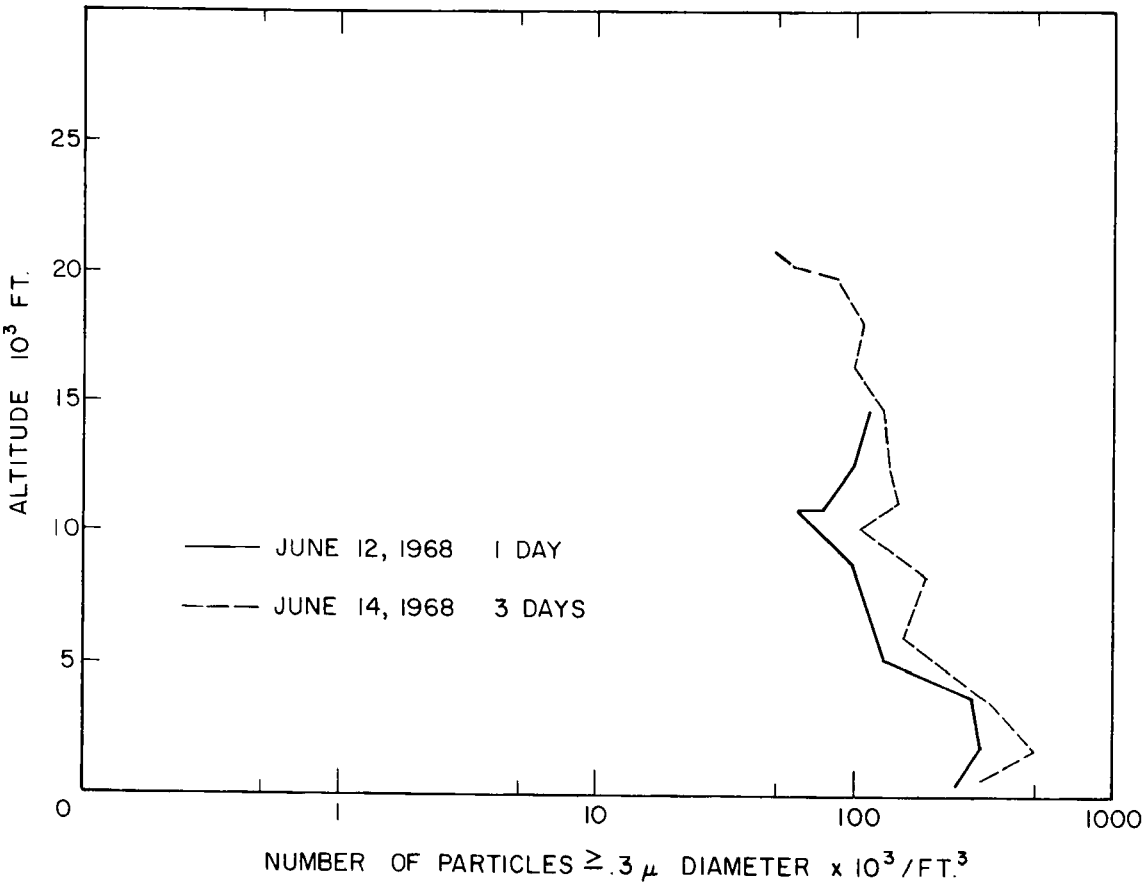

Fig. 6.3.10

Haze profiles taken over Tulsa, Oklahoma during the 1968 CV990 Flights.

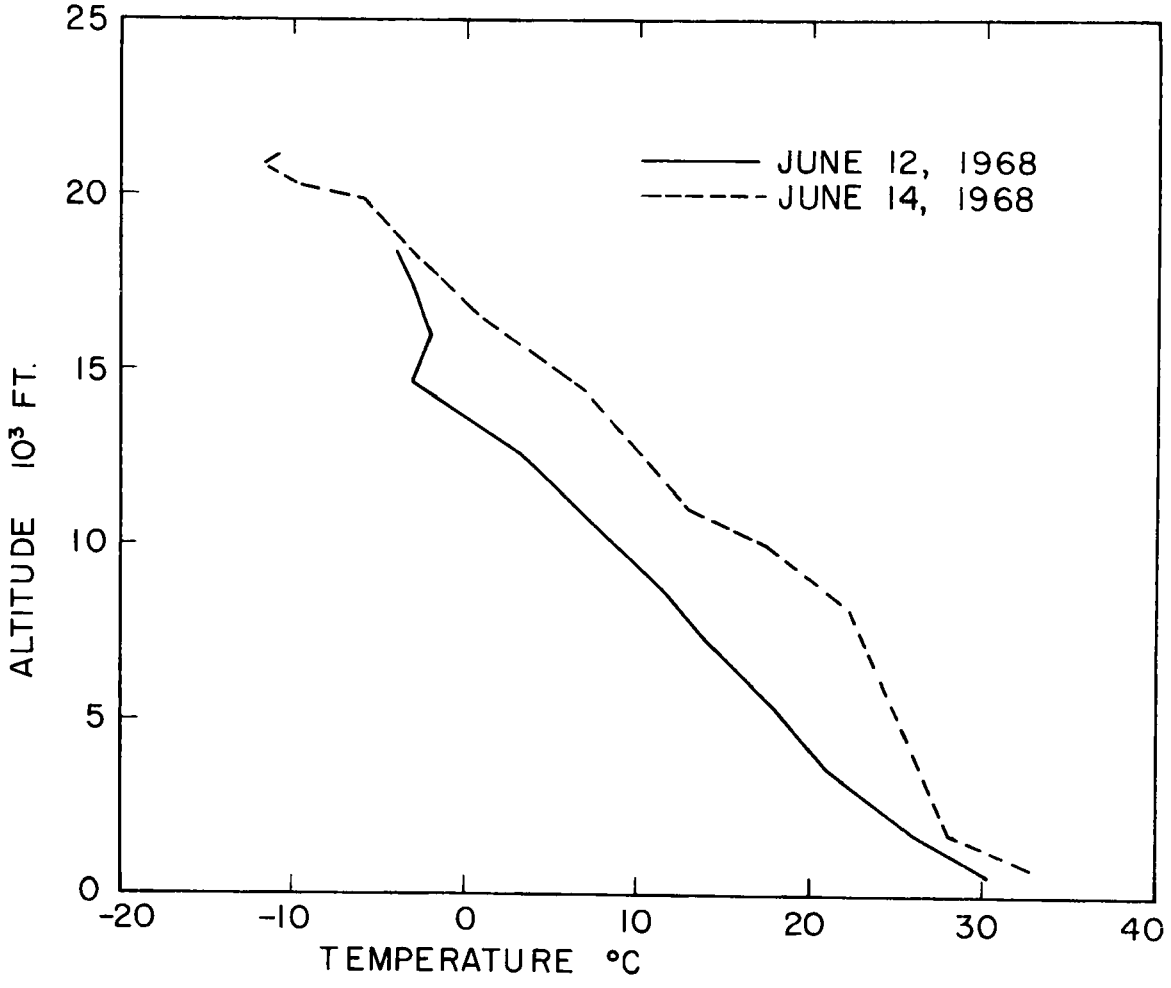

Fig. 6.3.11

Corresponding temperature profiles for Fig. 6.3.9.

extent of the vertical updrafts on haze populations. The Comanche flights offer the best example of this since they were flown during July and August over the Continental region of the United States. Six flights out of the twenty three were found to be made immediately preceding a front or after the front had passed (but not longer than 24 hours). The results are depicted in Figs. 6.4.1 and 6.4.2 and Table 6.4.1 for comparison with the total Comanche data. It is seen that the situation of more convective activity has produced a profile quite vertical in structure and with substantially less than the average aerosols.

6.5 Horizontal variability of haze

Fig. 6.5.1 shows horizontal haze distribution at selected heights. The aerosol concentrations match those of the vertical profiles. The time scale for the data in a, b and c is much shorter than for the data in d (11 minutes for 1 hour). The large variability of aerosol concentration in Fig. 6.5.1 indicates many different parameters at work in the lower levels. For the flight at 32,000 feet the counts are smaller. The values at the surface range from 50 to 200 $x10^3$ particles per cubic feet, the values at 4000' msl range from 8.0-25, the values at 10,000' msl range from 1.5-9.0 and the values at 32,000' msl range from 0.7 - 2.3. A profile taken through the minimum (or maximum) values could give a misleading picture of total haze concentration.

6.6 Influence of time considerations on haze

Figs. 6.3.4, 6.3.5 and 6.3.6 indicted the wide range of values observed between average annual haze amounts. There seems to be no real time relationship, though, between the different years. There was no short term increase in the haze counts due to an assumed increase in man-made pollutants into the atmosphere. Precipitation timing and amount may explain much of this variability. The synoptic situations seems to be the major influence with time related variables being of lesser importance.

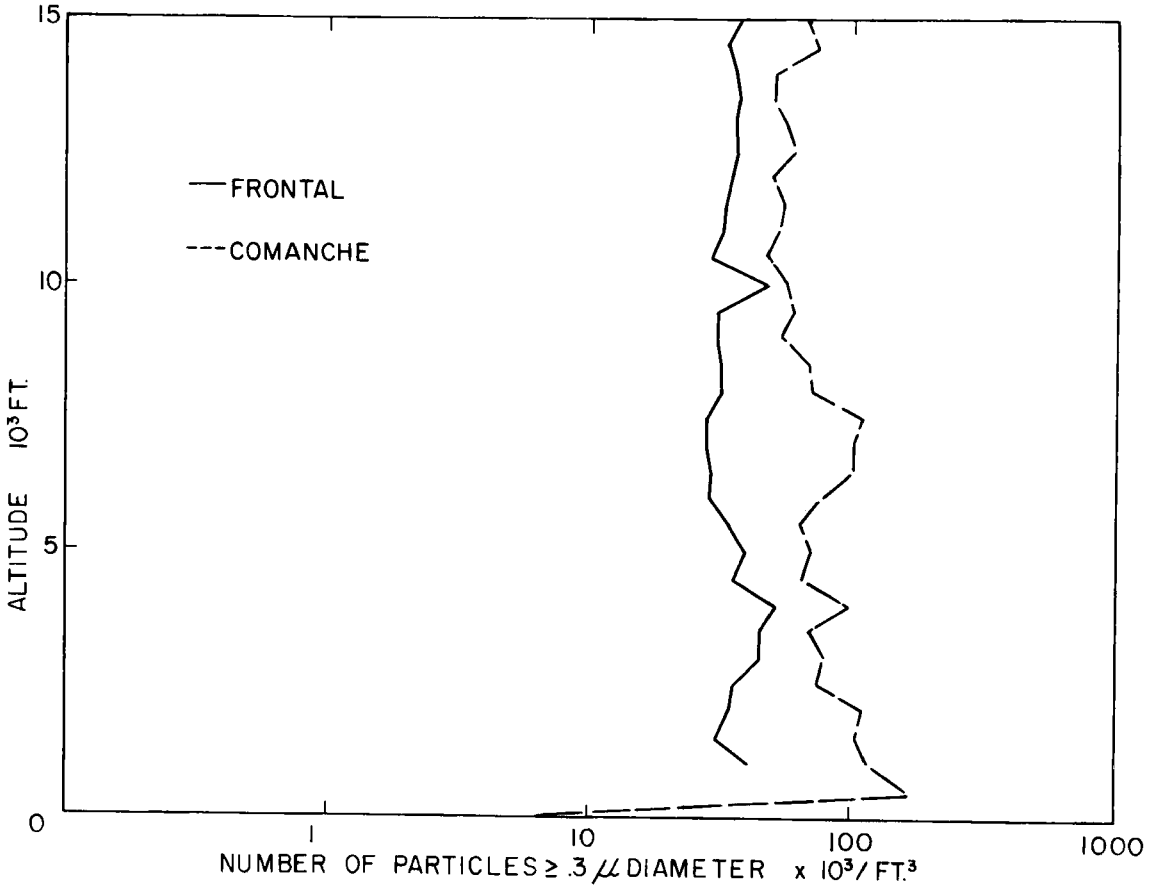

Fig. 6.4.1

Average haze profiles for Comanche data associated with convective activity and total Comanche data.

Table 6.4.1

Same as Table 6.1.1 except for points plotted in Fig. 6.4.1.

	Canadian	Frontal
Average no. of points	19	5
Average standard deviation	82	19

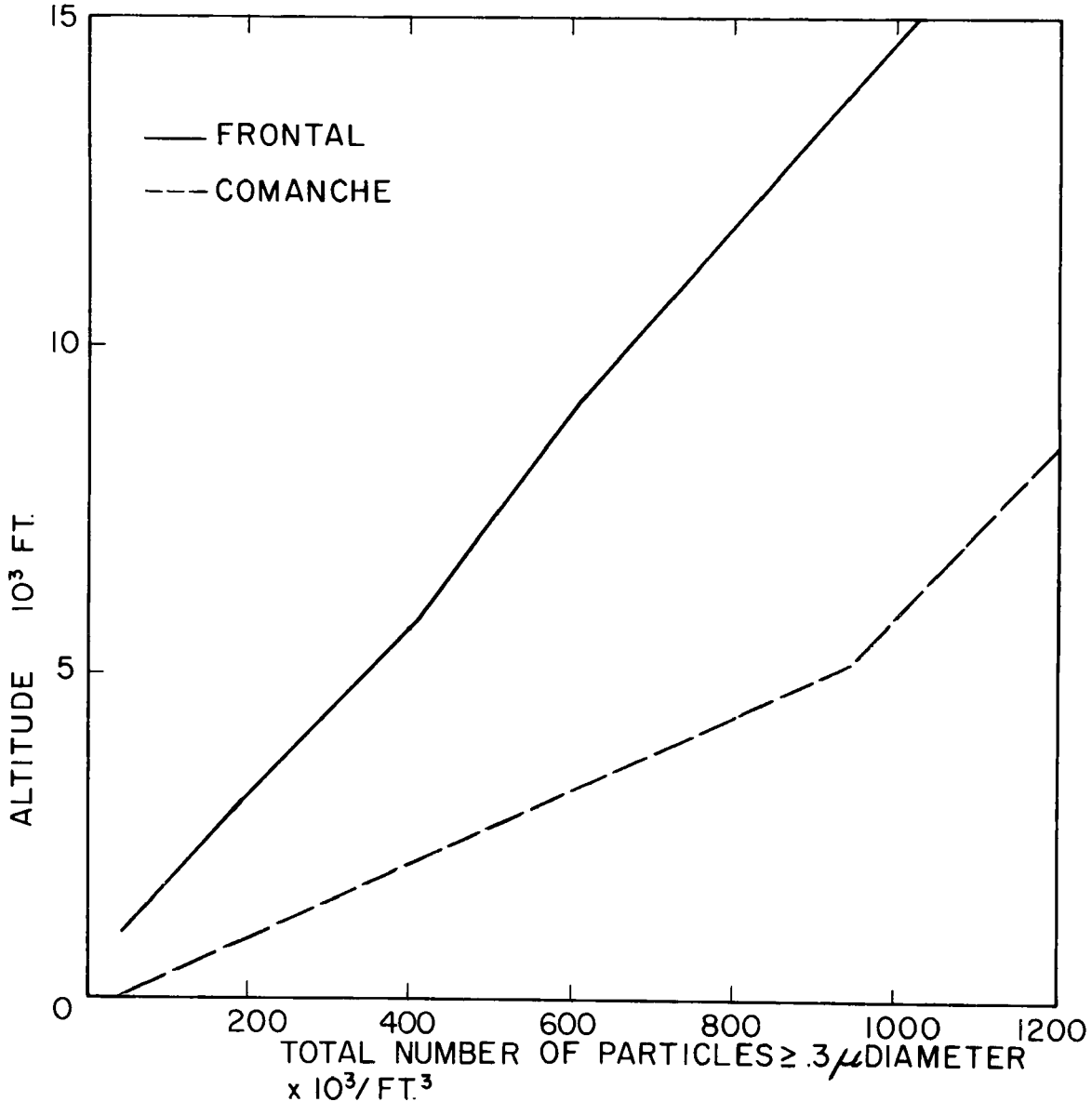

Fig. 6.4.2

Corresponding average total accumulations for Fig. 6.4.1

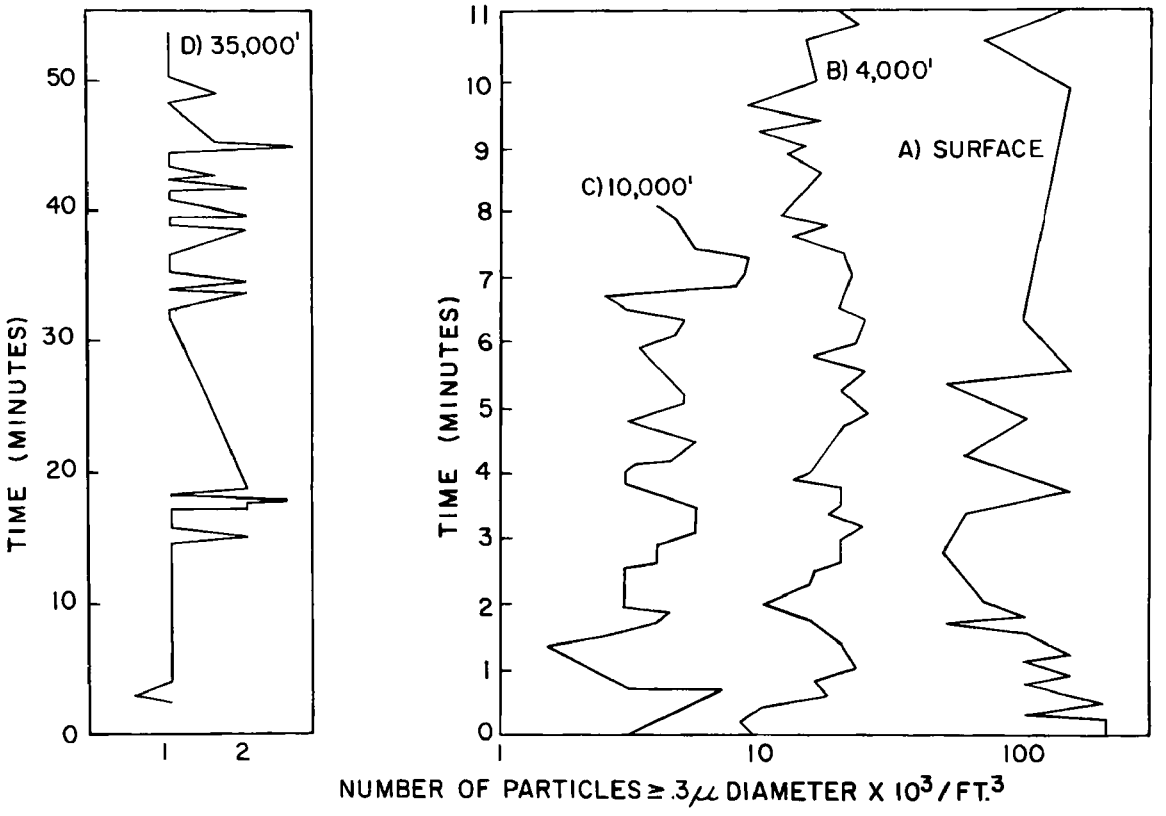

Fig. 6.5.1

Horizontal variation of haze: a) surface (taxiing), b) 4000 feet, c) 10000 feet, d) 35000 feet (note the different time scale). This data was taken June 1-3, 1967 over Texas.

6.7 Influence of temperature on vertical structure

The question of whether haze particles decrease with height can be answered simply by observing the profiles. In the general case, concentrations do decrease with height, but the decrease is not great and there are instances where there is no decrease (for example Figs. 6.3.4, 6.4.1). This decrease generally extends throughout the entire troposphere.

The influence of temperature has been briefly touched on previously. In many of the profiles a significant haze layer can be associated with a strong temperature inversion (for example in the no-recent-precipitation case at 15,000' msl, Fig. 6.3.4). But also there are cases where this is not the cause.

To further examine the temperature-haze correlation two consecutive days of midday and late afternoon flights were analysed. These are shown in Figs. 6.7.1, 6.7.2, 6.7.3 & 6.7.4. Fig. 6.7.1 shows the haze variability along the profile taken at noon. It is quite dissimilar to one taken five hours later. The temperature curves (Fig. 6.7.2) also show differences in structure. The relationship of temperature change to particle count change is not found. Only that the two haze profiles differ in structure can be ascertained. Fig. 6.7.3 shows how the profile has changed nineteen hours later. No precipitation was recorded during this period. The shape of this haze profile is less variable than the one for the previous day and a strong layer above 25,000' msl is noted. Also the temperature profile (Fig. 6.7.4) for one o'clock is very similar to the one for noon the previous day. The late afternoon profile for June 21 is very similar to the 1 PM profile. The temperature profile shows somewhat cooler air at all levels and generally follows the earlier profile.

Junge (1963) found that particle counts for Aitken particles above a certain level in the troposphere (the exchange layer where there is a general temperature discontinuity) became constant with

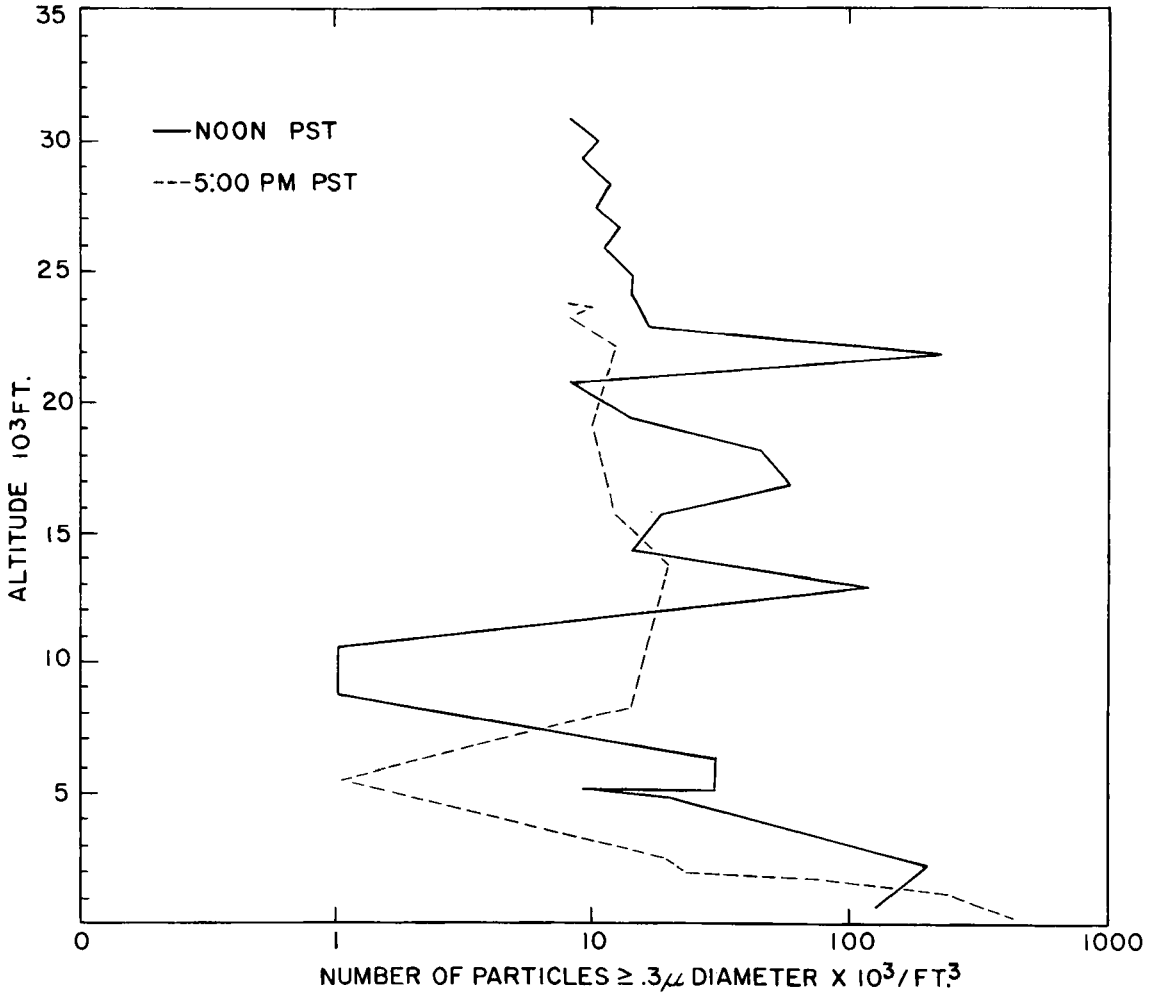

Fig. 6.7.1

Haze profiles measured near San Jose, California at noon and at 5:00 PM on June 20, 1968 in conjunction with the CV990 flights.

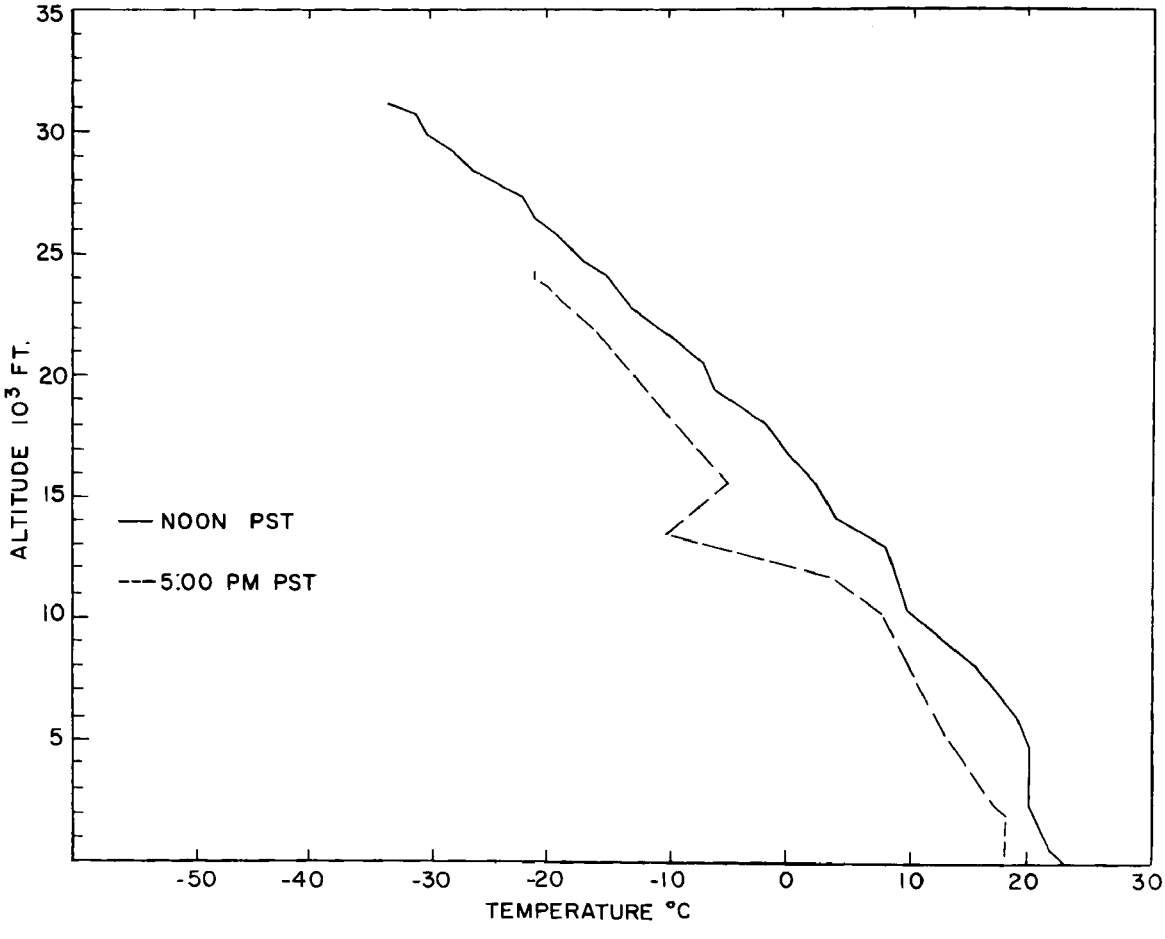

Fig. 6.7.2

Corresponding temperature profiles for Fig. 6.7.1.

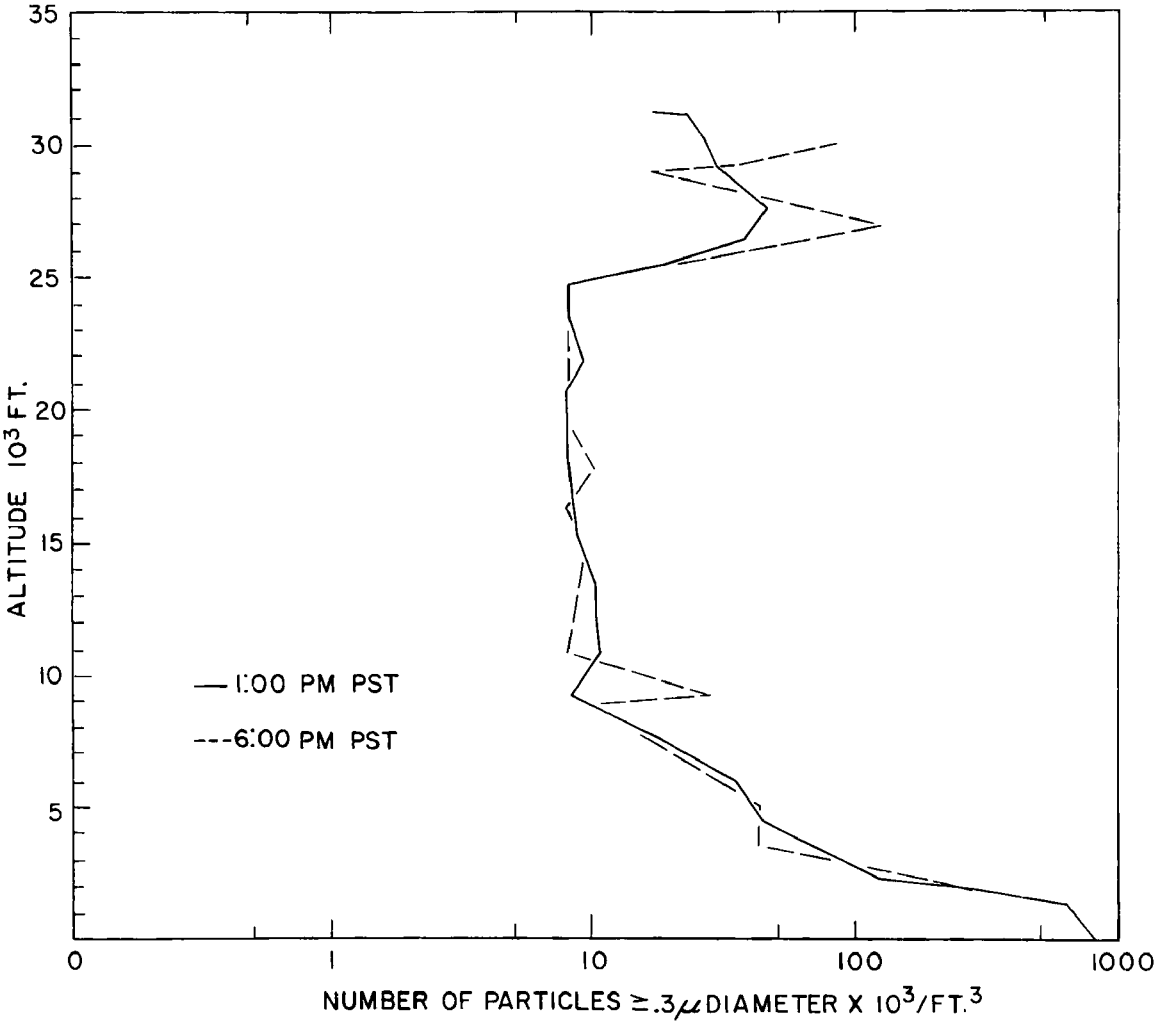

Fig. 6.7.3

Haze profiles measured near San Jose, California at 1:00 PM and at 6:00 PM on June 21, 1968 in conjunction with CV990 flights.

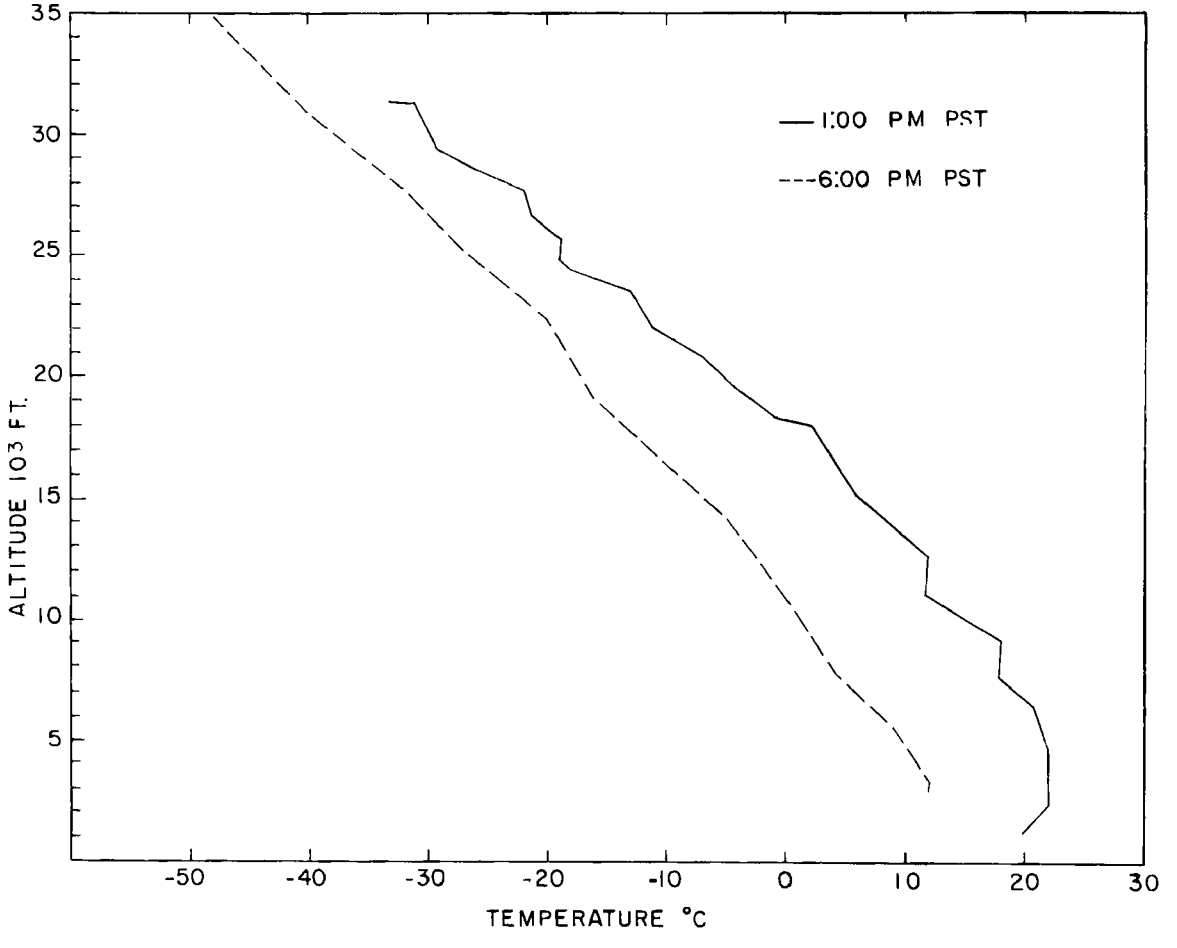

Fig. 6.7.4

Corresponding temperature profiles for Fig. 6.7.3.

height. He reasoned that the distribution of large particles must conform to the same pattern since removal and production processes do not vary appreciably between the two.

This effect, if present, would be very difficult to detect using average profiles. Individual profiles were examined to investigate this hypothesis for large ($\geq$ 0.3 μ) particles. Figs. 6.3.7 & 6.3.8 shows 6 individual profiles over Dallas, Texas. For the warmer days the above effect was observed between 5,000 and 10,000' msl. A haze layer was found associated with the temperature inversion. On the cooler days the same temperature structure was not present nor was there a constant haze layer.

Figures 6.3.9 & 6.3.10 showed individual profiles for several 1968 CV990 flights. The profile for June 14 indicated no definite haze layer between 5,000 to 15, 000 feet but the temperature profile indicated a strong inversion was present. Figs. 6.7.1, 6.7.2, 6.7.3 & 6.7.4 again indicate instances where a temperature correlation cannot be ascertained.

Figs. 6.7.5 & 6.7.6 shows the average of the Alaskan profiles measured in 1967 and one winter time profile taken over South Dakota in November 1968.

The cooler air masses of the Alaskan and winter continental U.S. show a marked decrease in particle concentration (between 10,000 and 15,000 feet for Alaskan average and above 15,000 feet for the winter U.S. case). The temperature profiles reflect this situation to some extent. It is admittedly risky to compare average profiles with one single profile, but is done here to show the effect of the exchange layer on haze distribution.

6.8 Error analysis

Examination of the great variation which exists between the different sets of data leads to the question on error. How accurate is the data? The first source of possible error could be the Bausch

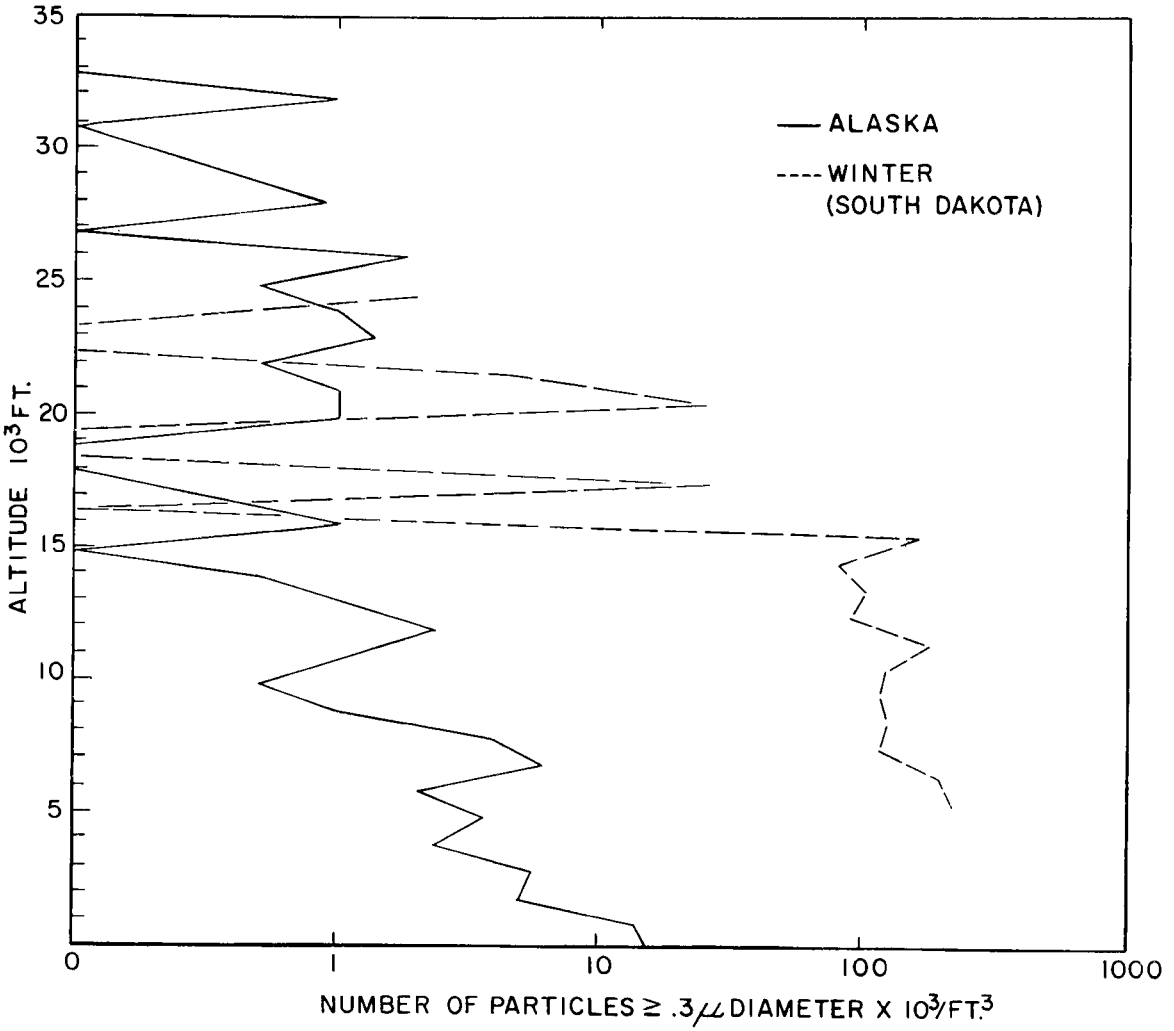

Fig. 6.7.5

Winter and average Alaskan haze profiles.

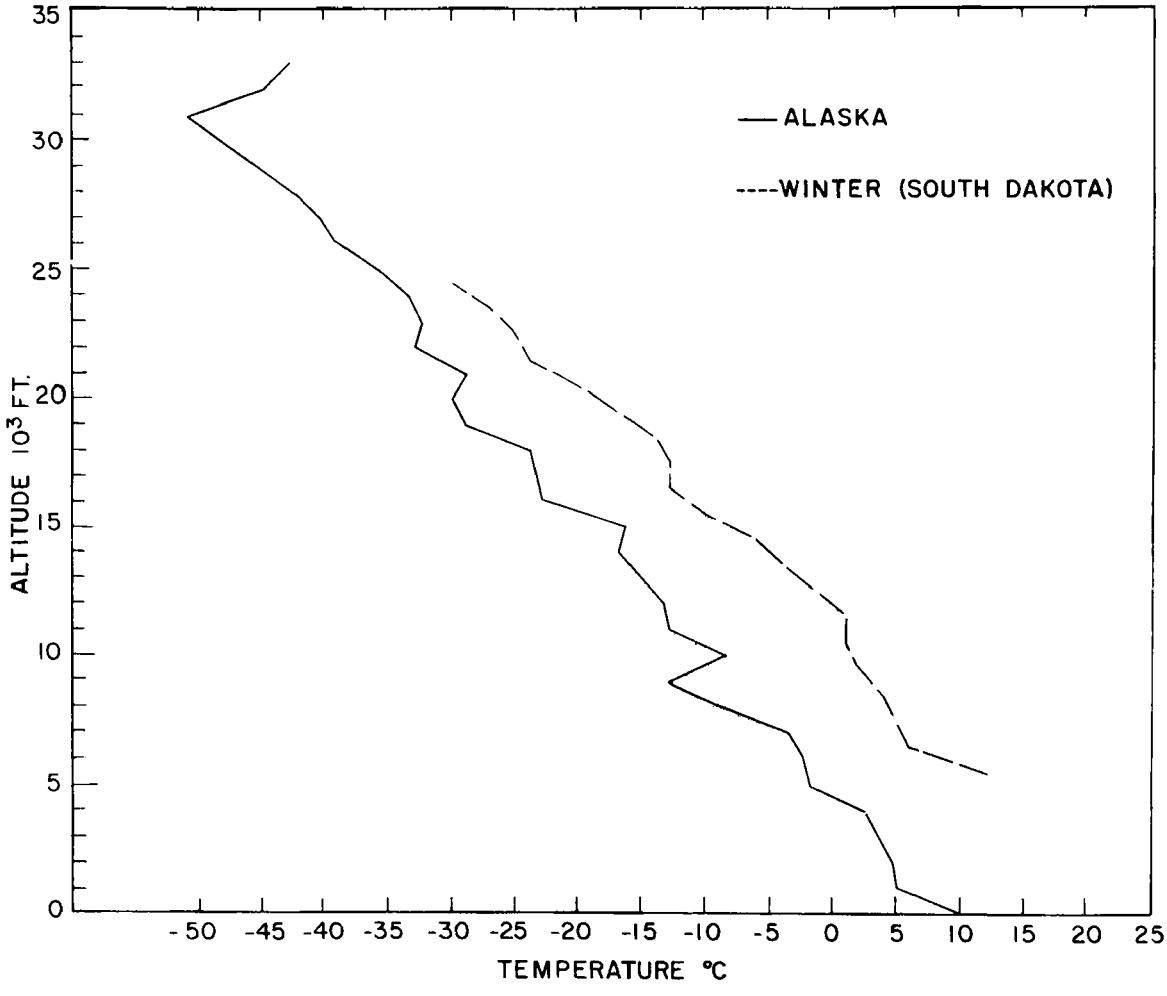

Fig. 6.7.6

Winter and average Alaskan temperature profiles.

and Lomb instrument. The general design of the Bausch and Lomb tends to keep human errors to a minimum, but there are two very critical areas that must be constantly checked: calibration and flow rate. After each set of flights the instrument was calibrated and found to be very sensitive to small calibration changes. Obviously, the instrument must be calibrated exactly the same before each set of flights or serious errors will occur. Attempts were made to do this.

The rate of flow of particles into the instrument is crucial to the measurement accuracy. On some cases the flow meter was not operating properly and some error may have occurred, but this was a small exception. Overall, this error was nearly eliminated by close control of the flow meter.

Another group of possible errors were sampling errors. The data was taken in two different aircraft which traveled at quite different speeds (100 and 250 knots I. A. S.). The effect of aircraft speed on the sampling is not known exactly, but the data evaluation indicated little difference between 100 and 250 knots for sampling purposes. Another possible error is that the aircraft profiles were not taken vertically but as descents and ascents. This error, however, is small if haze concentrations in the atmosphere can be considered as large horizontal sheets. Indications are that this is an acceptable approximation.

The final source of possible error was the method used to process the data. The averaging process sometimes averages out the essence of a problem. Small structure features were lost by averaging aerosols within 1000 foot intervals. Even by allowing for maximum error here, the total error will still be negligible since it is the overall haze structure which is being analysed and not a small section of it. The classification of data dictates that there be homogeneity among the classes and that each class must be exhaustive and exclusive. We have seen that this is not always the case and some

data must be considered borderline cases. This is a problem that cannot be avoided, however, by using only average values this error will be minimized.

Another problem resulting from the reduction of data is the change in scale for the 1968 990 flights from .3μ and greater to .5μ and greater. This does indeed eliminate a great number of particles from consideration. If the approximate percentage difference of the two sizes is known, however, it is possible to evaluate differences in data sets. During the 1966 CV 990 flights $\geq 0.5\mu$ aerosols were measured. These particle counts averaged 70-90% smaller than the $\geq 0.3\mu$ counts. Fig. 6.8.1 illustrates this difference. During the Line Islands flights the same measurements were taken and the $\geq 0.5\mu$ counts averaged 80% smaller. Thus, profiles containing 1968 data could be smaller than the $\geq 0.3\mu$ particles by at least a factor of five.

In this study 87 flights were analysed. Many of the errors listed above were minimized by the sample size. Errors of significance are the calibration of the Bausch and Lomb and speed of the aircraft. These need more study before they can be evaluated completely.

6.9 Comparisons with other investigators

As indicated earlier, there has been very little research on the vertical distribution of aerosols in the size range 0.2μ - 2.0μ diameter in the troposphere. Junge (1963) inferred that vertical distributions of all size ranges would have many similar properties. Fig. 6.9.1 shows the findings of Wigand (1919) and Weickmann (1955) for vertical distribution of Aitken nuclei. Wigand used data from 15 ballon flights and Weickmann used data from 12 aircraft flights. The most obvious feature of both analyses is the decrease in haze concentration with altitude approximately on the order of $10^3/20,000$ feet. Fig. 6.9.1 illustrates that the removal processes are more dominant than the effect of vertical mixing in the lower levels. Another significant feature of Weickmann's work is the non-linearity of the curve. This

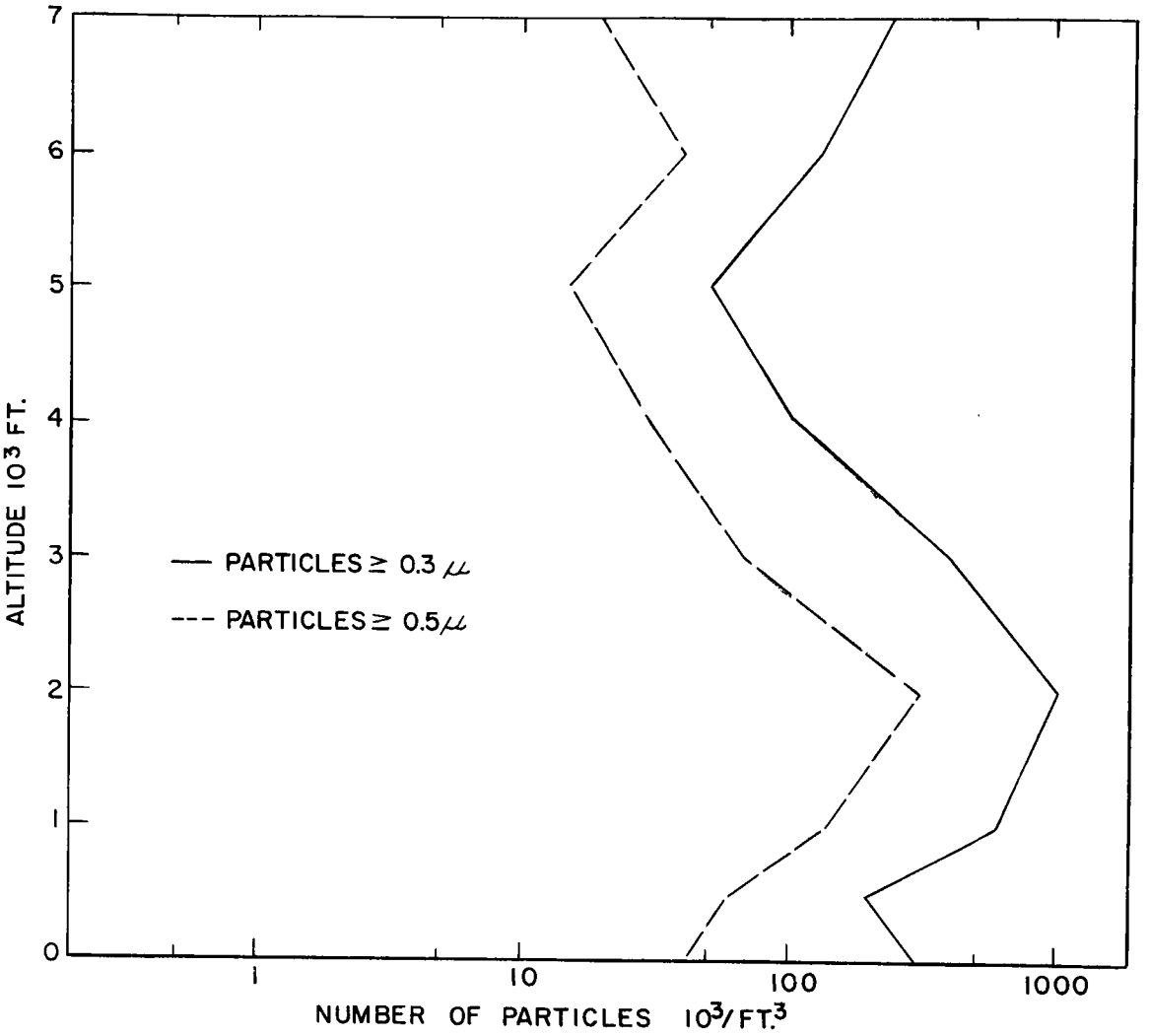

Fig. 6.8.1

Haze profiles for $\geq$ 0.3μ and $\geq$ 0.5 μ particles for Sunset Intersection, California; June 15, 1966.

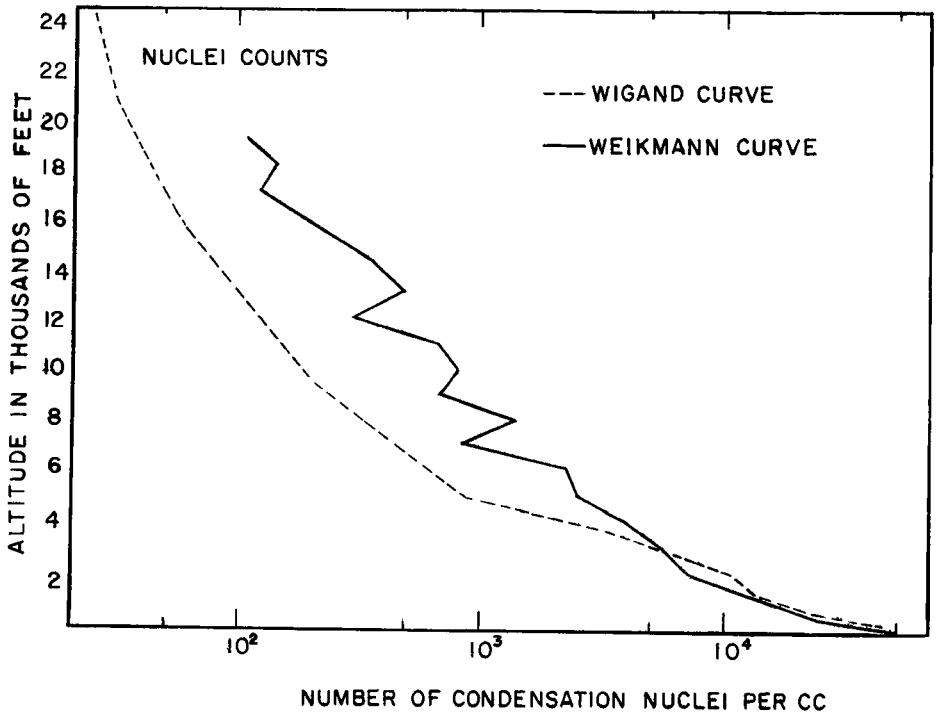

Fig. 6.9.1

Average vertical distribution of Aitken particles (Weickmann and Wigand).

is due to the layering characteristics of aerosols in the atmosphere. Weickmann showed that the tops of the aerosol layer were marked by a sharp decrease in concentration even though the layer itself may not have had a high concentration.

Fig. 6.9.2 shows the vertical distribution of aerosols measured by a number of different investigators. The profiles are for large particles. It may be observed that the amount of decrease versus altitude is in the order of 10^2 for large particles. The wintertime situation, when the removal processes are more dominant and the synoptic situation has less vertical motion upward, shows a stronger decrease than in the summer situation. Penndorf and Sredentopt (Junge, 1963) using attenuation of solar radiation and zenith sky luminance measurement, respectively, found a constant vertical profile above 4-5 km. Junge (1963) described their results as being indicative of the aerosol distribution at this altitude. He considers the 4-5 km value as the upper boundary for the continental exchange layer and noted that the atmosphere is well mixed except for a sharp decrease in the exchange layer. A two-dimensional model indicating non-steady state conditions was used to explain this phenomenon with the interaction of maritime and continental air being the focal point. This method infers that the vertical concentration of particles decreases only slowly as one proceeds inland from a west coast of a mid latitude continent because eddy diffusion increases as one goes inland. In this way, eddy diffusion is shown to be directly related to vertical mixing.

A comparison of the vertical distribution of sea-salt particles in the $\geq 6\mu$ diameter range is appropriate. Fig. 6.9.3 shows the results of Woodcock (1953) measured in the trade winds area and by Byers et al. (1955) for measurements made in Illinois. The profile in the trade winds shows a marked decrease at the trade wind inversion, while the Illinois profile exhibits the effect of convection over

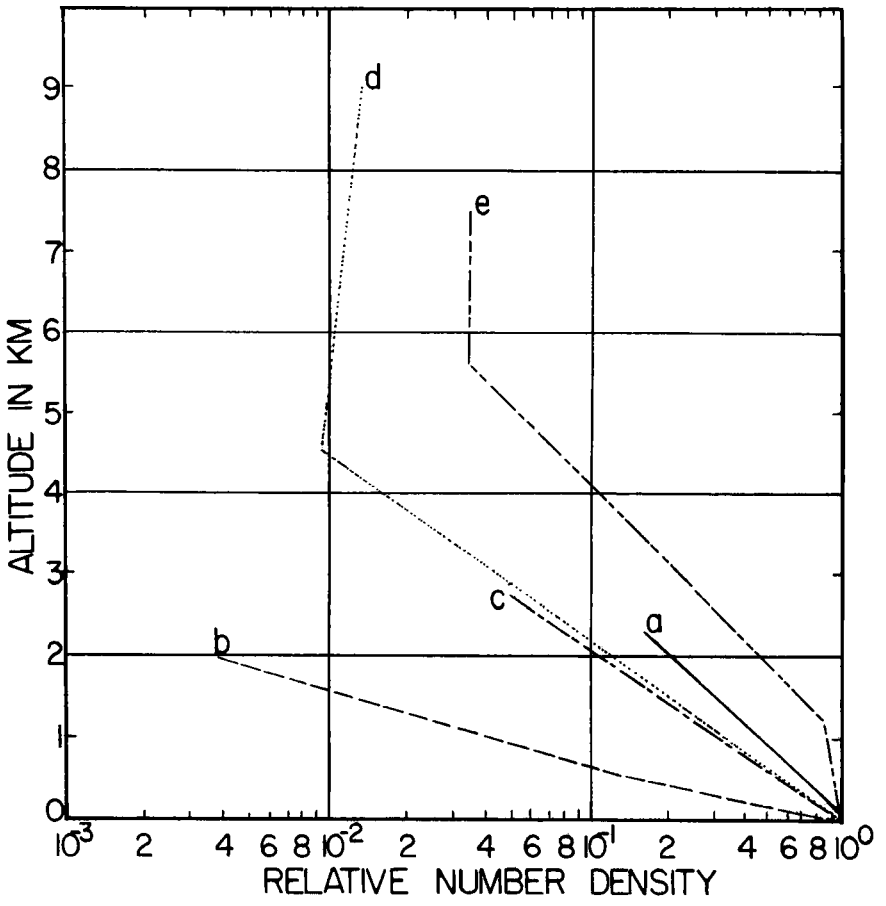

Fig. 6.9.2

Average vertical distribution of natural aerosol particles over Central Europe in relative number concentrations. All curves refer to large particles. Curve a, 12 summer flights. Curve b, 8 winter flights, (Siedentopt). Curve c, 18 flights, impactor (Rossmann). Curve d, calculated curves from observations on the attenuation of solar radiation (Krug, Penndorf). Curve e, calculated curve from zenith sky luminance measurements obtained on 18 flights (Siedentopt). (after Junge).

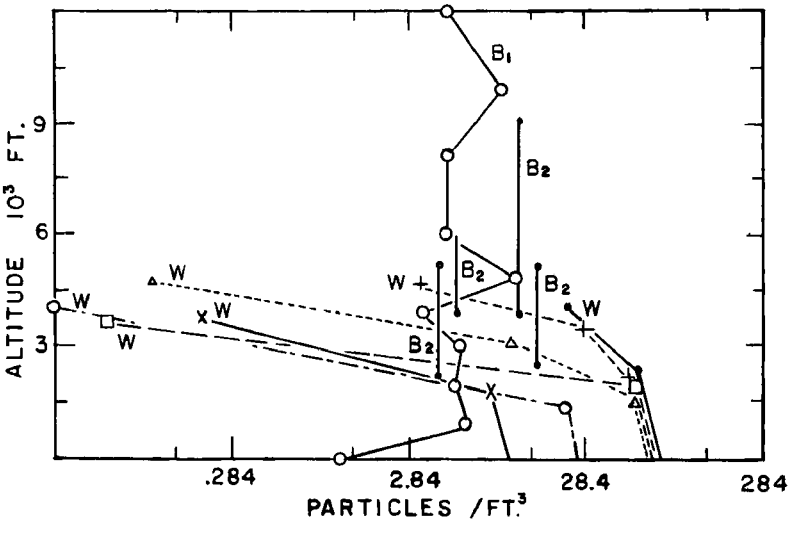

Fig. 6.9.3

Vertical distributionof the number of sea-salt particles having a dry radius $\geq 3\mu$. The curves marked W are measured by Woodcock (1953) in regions of the trade winds; the portion of the curve between the ocean surface and the measured values around 0.5 km is assumed. The curves marked B are given by Byers et al. (1955). Curve B_1 is an average of 3 soundings made in Illinois. Curves B_2 are average concentrations for 4 overland flights southward from Chicago. (after Junge)

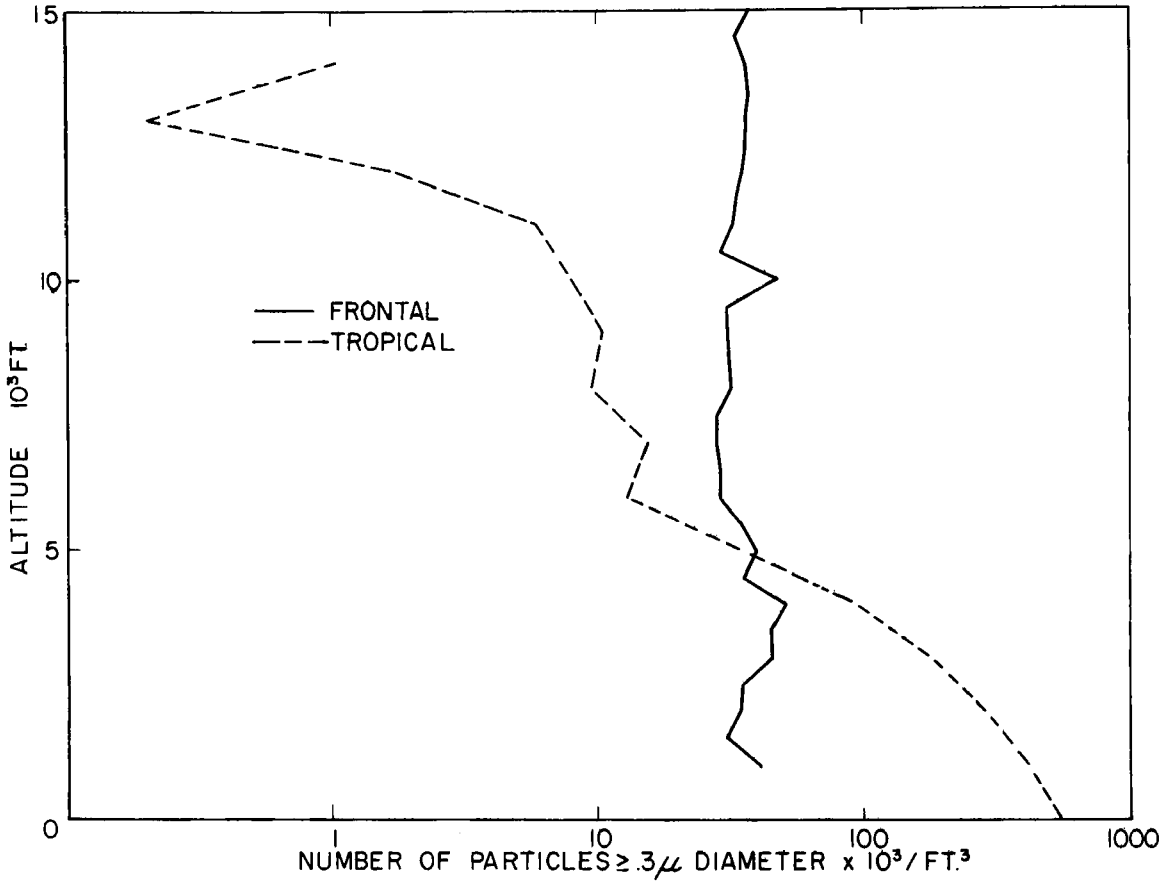

Fig. 6.9.4

Average haze profiles for Comanche data associated with convective activity and tropical Pacific air mass.

land. Fig. 6.9.4 compares the average tropical Pacific sounding with the frontal convective profiles of the Comanche data. It may be noticed here that a great deal of similarity exists between Fig. 6.9.3 and Fig. 6.9.4. This further indicates that error sources for this process are minimal.

Chapter VII

CONCLUSIONS

The results presented in the last chapter can be used to answer the questions proposed in Chapter I.

a) Does the air mass source contribute significantly to the number of haze particles over a given location? The air mass source does not appear to contribute to the haze in the lower levels of the troposphere over the continents. This fact indicates that an air mass is rapidly modified as it enters a particular area. It is feasible that the continent, being the dominant haze source region, controls the haze population even over adjacent coastal waters. Junge (1954) found that a wind blowing onto a coastal region did not effect counts appreciably. There is an overflow from the continental source in the lower levels into maritime source regions. This indicates either horizontal mixing on a large scale without the necessary wind mechanism or that haze is removed quickly in the lower levels from the atmosphere (small τ)

Haze amounts in the upper levels show the influence of the maritime air and vertical mixing as the air mass becomes more continental. Also, for the warm season of year there is generally more subsidence in the upper layers over the ocean than over land;more convection over land than over oceans. This tends to produce a decrease in haze for maritime air and gives a relatively constant concentration for continental air. Since the upper levels are further removed from the main source region (continent), upper level air masses are less susceptible to modification by the source as a function of time and thus reflect the nature of the air mass source region. The total accumulation is greater over the continents because the atmosphere receives a larger contribution from the surface.

Analysis of haze in the air masses of the tropics and the arctic

gave a different perspective to the problem. The tropics are characterized by subsidence above cloud level with the strongest winds in the lower troposphere. The air mass source has characteristics similar to air that has stagnated over the area would have, since the aerosol source region (the ocean) is the same for both cases.

The cool air in Alaska exhibits a similar but more complex effect. As a haze source the arctic is less effective than continental areas at lower latitudes due to snow and ice cover and the absence of human activities.

It is difficult to say, therefore, that a maritime air mass will produce a haze population that is less than that from a continent. From a given lower tropospheric haze profile for the mid-latitudes it is not possible to predict the air mass source. But when the surface haze source region is not as active, the air mass source can contribute in the development of the haze profile. A full tropospheric profile could provide more information on air mass source. This is done by examining the upper levels for maritime or continental characteristics.

b) Does the type of surface significantly influence the haze directly above a region ? It was indicated earlier that the haze source region (continent) was an important feature in haze distribution in the lower levels. Upon examination of the land versus ocean and the urban versus rural distribution, however, no great differences between the vertical components were found. This could be due to; (1) the way in which the data was taken, (2) the large background concentration of haze, or (3) the fact that haze particles mixed rapidly in both the horizontal and vertical. Results presented here favors number three. Either haze moves more in the horizontal than the vertical or removal processes are extremely efficient the lower levels. This is also compatible with the answer to the first question.

c) Do unusual instances of precipitation (or no precipitation)

alter haze counts significantly? In reviewing the above results it must be remembered tha the no-recent-precipitation data contained many 1968 990 Flight data consisting of particles $\geq 0.5\mu$ only. A definite correlation was found between precipitation and haze amounts both on large scale and small scales. Precipitation also affects surface properties of the earth which in turn reduces the effectiveness of the surface as a haze producer. Any prolonged period of drought will produce haze environments that are more dependent on other factors. To discuss precipitation as an important aspect of haze removal in an isolated sense (i. e. without precipitation history) is not possible. From our results precipitation appears to be an efficient means of removal of haze particles from the atmosphere.

d) Does convective activity influence vertical haze counts? This question of the effect of a frontal system on haze distribution revealed some expected results. The strong convective activity ahead of a front lifts many surface particles into the lower troposphere. In addition convection activity results in precipitation. Therefore, the haze concentration in the vicinity of a front may be expected to be nearly constant with the height up to the cloud level due to convection and be less than average due to precipitation.

e) How much horizontal variation exists in haze distributions? From the data presented it may be concluded that haze layers become somewhat more uniform and stable in the upper troposphere although strong evidence for this is lacking due to incompleteness of data. The great vertical variation in the distribution haze below 10, 000'msl showed that the 'layering' effect of haze is not as dominant a feature as may have been expected. While the vertical profiles indicated horizontal layering of aerosols, and while these layers are visible to the human eye (they are certainly homogeneous with respect to haze concentration), the concentration of particles within a layer varied by a factor of four. The non-uniform vertical diffusion mechanism has a definite

role in producing non-uniform horizontal layers. Thus, this vertical mixing property 'erodes' the horizontal layers.

f) What are the time considerations on aerosol distributions? While there are differences in the data between years there is no significant time increase of haze populations to suggest that haze is building up in concentration over the time period of 3 years. Given the 1966 data, 1967 data and 1968 data it would be difficult to predict what an average 1969 haze profile would be.

g) What is the effect of temperature on the vertical structure of haze? Temperature and gradients are the basis for diffusion in the atmosphere and, therefore, temperature gradients have been assumed to control the nature of distribution of haze. In this study temperature was found to play only a minor role compared to the transport processes resulting from precipitation and convection. A large temperature inversion limits vertical motion and acts as an aerosol trap . An aerosol must be present, however, and there must be sufficient vertical motion from below to get particles to the inversion for this to take place. It cannot be assumed that an inversion will always produce a haze layer.

The Aitken nuclei exchange layer discussed by Junge was not found, nor was the temperature discontinuity a stable feature. The exchange layer of the tropopause could produce the same results, but was not explored here. The diurnal fluctuation of temperature within the troposphere did produce diurnal horizontal fluctuations of haze. The role of temperature in haze production and distribution needs further research.

From the works of other authors the expected shape was assumed to be exponential and the decrease very rapid since removal processes are more dominant than production processes in the upper troposphere Results of this study indicated that the decrease was generally small. This may be related to the fact that the measurements were obtained

in late spring and summer with warming at the surface. Particles $\geq 0.3\mu$ diameter are believed to behave differently than Aitken particles.

In conclusion, haze particles were found to have many similar properties to other sized particles, but there are differences that affect the transport of these aerosols. The processes of precipitation, convection, and subsidence play dominant roles in transport of haze. The haze transport 'cycle' is in rapid flux, mainly in the horizontal but with a relatively strong vertical component. The tropics and the arctic represent the simplest models for haze study because all processes are clearly defined. The mid-latitude continents serve as a more complex situation due to the variability of the synoptic situation and the diversity of the continental haze source.

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