RESEARCH ACTIVITIES

CLOUD PHYSICS DIVISION CSIRO

JUNE 1976

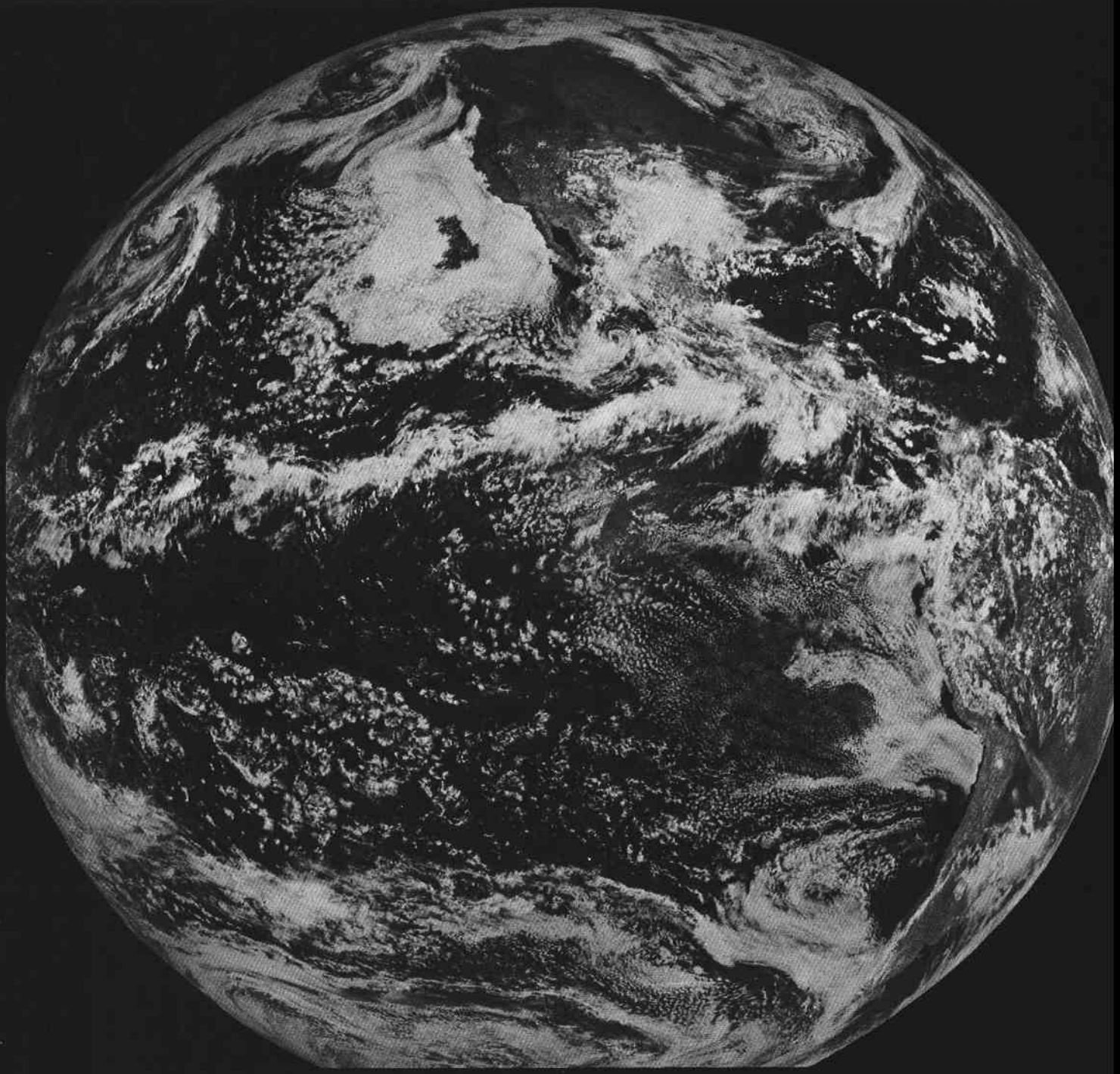
FOREWORD

Weather and climate are everyday concepts which affect the well-being and livelihood of all mankind. Many organizations around the world are engaged in research aimed at understanding, forecasting and, where possible, modifying the weather. Within the Commonwealth Scientific and Industrial Research Organization research into atmospheric problems is largely concentrated within the group of three Divisions constituting the Environmental Physics Research Laboratories — namely the Divisions of Atmospheric Physics, of Cloud Physics and of Environmental Mechanics. The Division of Cloud Physics is located some 16 km north-west from the city centre in the Sydney suburb of Epping, where it shares a building complex and many facilities with the Division of Radiophysics.

The Division of Cloud Physics is concerned primarily with those atmospheric processes which govern cloud formation and the production of rain. Its research therefore ranges from studies of the sub-microscopic particles in the atmosphere upon which cloud droplets form and which stimulate the formation of ice crystals to the investigation of fields of clouds covering an area exceeding ten thousand square kilometres. The concentration, size distribution and chemical nature of small particles, which are present in abundance almost everywhere in the atmosphere, are all factors of major importance. In any given meteorological situation these factors can influence not only the ability of a cloud to produce rain but also the way in which clouds transmit and scatter the sun's radiation, and hence they can affect climate on a global scale. Since man's activities, both agricultural and industrial, often result in the production of large numbers of smoke and dust particles which find their way to all levels of the atmosphere the Division regards it as important to find out whether adverse effects could result from this pollution. Cloud seeding, which is also based upon the effect of special small particles on the behaviour of clouds into which they are injected, is an additional activity of the Division.

Apart from its well-equipped laboratory at Epping, the Division owns a DC3 aircraft which it has instrumented for the measurement of various cloud properties. Other aircraft are hired from time to time and after fitting with special equipment are used for additional cloud research or for cloud seeding. The Division's research into upper atmospheric and stratospheric particles is dependent upon the use of instruments attached to large balloons flown from Mildura by the Balloon Launching Station operated by the Department of Science.

This booklet describes the recent work of the Division and the current status of its research into cloud physics, cloud seeding and the atmospheric aerosol.

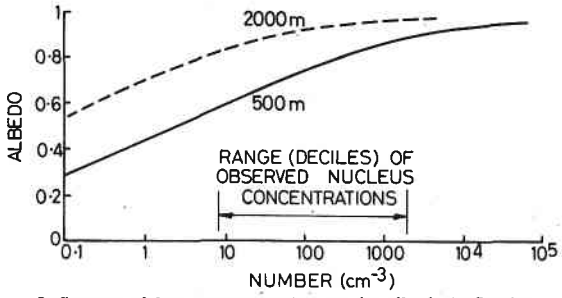


Almost half of the earth is included in this photograph taken by a United States N.O.A.A. satellite on 13 September 1975. The North American continent (upper centre) is largely free of clouds, except in and near the Gulf of Mexico. South America is also visible (lower right). The oceans are almost black, and light reflected from oceanic areas is almost entirely due to clouds. Cyclonic storms are visible in the north-west Pacific (upper left) and off the west coast of Mexico (near the centre of the photograph).

THE ROLE OF CLOUDS IN LARGE-SCALE PROCESSES

The sun delivers radiant energy to the earth at a rate of about 1400 W m^{-2}; this amounts to about 350 W m^{-2} if averaged over the whole of the earth's surface. The proportion of this energy which actually reaches the surface is dictated by a number of factors, the most important of which are the extent and nature of cloud cover. Clouds can reflect 80% or more of the incident solar energy back to space — satellite pictures show very clearly the importance of clouds in this regard. The energy reaching the surface represents the heat input to the "boiler" of the atmospheric heat engine and energy lost to space by longwave infrared radiation represents heat removed in the "condenser" of the engine; this radiation is also influenced greatly by clouds which, being higher and cooler than the surface, radiate less to space than the surface does in the absence of clouds.

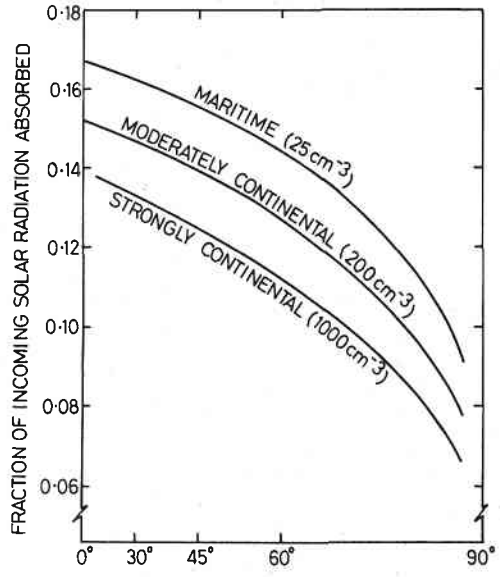
There has been a tendency to discount the role of clouds in discussing radiative effects on weather and climate simply because of these counteracting effects, but the counterbalance is not in general exact. A comparatively thin cloud is just as effective as a thick cloud so far as longwave radiation is concerned, whereas shortwave reflection by a cloud continues to increase with increasing thickness. Furthermore the thickness involved — optical thickness — can be enhanced by increasing the drop concentration and size as well as by increasing geometric thickness. Since drop concentration is determined primarily by the atmospheric particle population and is not greatly affected by larger-scale meteorological parameters, atmospheric particles can have a significant influence on the energetics of the atmosphere, i.e. on weather and climate. The importance of such influences becomes more apparent when one realizes that most of the solar energy input is not available for influencing weather systems because it is evenly distributed. It is only "available potential energy" that provides the source for atmospheric motions, and it has been calculated that the generation and dissipation of this available potential energy occur at rates of the order of 1 W m^{-2}; even the highest rates calculated (around 10 W m^{-2}) still represent only a few percent of the solar input figure of 350 W m^{-2} mentioned earlier. Satellite-



Influence of drop concentration on the albedo (reflecting power) of clouds of differing thickness.

borne instruments have measured systematic differences in radiation between the high- and low-pressure regions of mid-latitude storms. The differences, amounting to 35 W m^{-2}, are quite comparable to the estimates of available potential energy generation rates and support the contention that radiative effects — primarily due to clouds — may be important in weather.

Latent heat, another very important energy component, is released during condensation, but the release of latent heat during cloud formation is not necessarily permanent; the same amount of latent heat is used up during evaporation of a cloud, and it is only from precipitation which falls to the surface without evaporating that a net heating is realized. Two apparently similar clouds can differ widely in their ability to produce precipitation — a question which has been under study by the Division of Cloud Physics for a considerable time. Again particles come into the picture — the cloud nuclei determine the number and size of the cloud drops which are formed and this strongly influences the ability of the cloud to rain; freezing nuclei likewise modify the micro-structure of the clouds and their ability to rain.



Absorption of solar radiation by clouds.

A further influence of particle population on cloud properties and hence on important radiative properties of clouds relates to the absorption of solar radiation. Computations which we have recently completed show that maritime clouds containing relatively few droplets per unit volume absorb about 3% more of the incoming solar radiation than continental clouds — which contain many droplets per unit volume even though they may be otherwise similar. The difference which is systematically found between continental and maritime clouds is known to be caused by the greater number of particles (specifically the greater number of cloud nuclei) present in continental air. The 3% change calculated is small in terms of total energy but is large when viewed in the context of available energy.

From the point of view of climate — which is essentially a long-term average of day-to-day and year-to-year weather — clouds are of undisputed importance because of their influence on radiation. This is especially true for the highest clouds (cirrus), which some recent reports suggest have increased during the 1960s. Contrails produced by high-flying aircraft are a form of cirrus and a very common example of man's influence on cloud, but whether these represent a significant climatic influence is not yet known. Such questions are nevertheless of obvious importance.

FORMATION AND GROWTH OF CLOUDS AND CLOUD FIELDS

The first step in the process of cloud formation is the evaporation by the heat of the sun of water from oceans, lakes or moist ground. As this moist air rises it cools and eventually a cloud forms as condensation occurs on minute contaminant particles, known as cloud condensation nuclei. The droplets so formed are initially less than one-thousandth of a millimetre in diameter and they grow by continued condensation, or by coalescence as they collide with one another; some eventually fall as raindrops, perhaps a millimetre or more in size.

All clouds start life as a small "puff" of condensed moisture, grow and finally disappear by evaporation or by precipitation as rain, hail or snow. Despite the enormous variety of shapes and sizes of clouds certain distinctive cloud types with characteristic lifetimes are recognizable. But clouds can seldom be regarded as isolated individuals: they often occur in aggregates and interact with one another by affecting the motions, temperature and humidity of the clear-air environment in which they have formed. The concept of a "field of clouds" therefore seems appropriate.

We can best appreciate the nature of a cloud field by studying photographs taken from high-flying aircraft or from satellites. It is clear that quite marked changes in cloud grouping can occur on scales of a few hundred kilometres; on a much larger scale individual clouds seem to be insignificant but the patterns of cloud fields suggest a connection with the large-scale motions in the atmosphere such as cyclonic and anti-cyclonic circulations. Our knowledge of the mechanisms responsible for these connections is still very rudimentary; weather forecasting and weather modification activities would benefit greatly from increased knowledge of these factors.

The photograph reproduced above was taken from an aircraft flying at an altitude of 20 km; the horizon is therefore about 500 km distant. In the foreground we see a fairly uniform population of cumulus clouds while in the middle distance two large areas are substantially free from such clouds although some layer cloud at a different level is present.

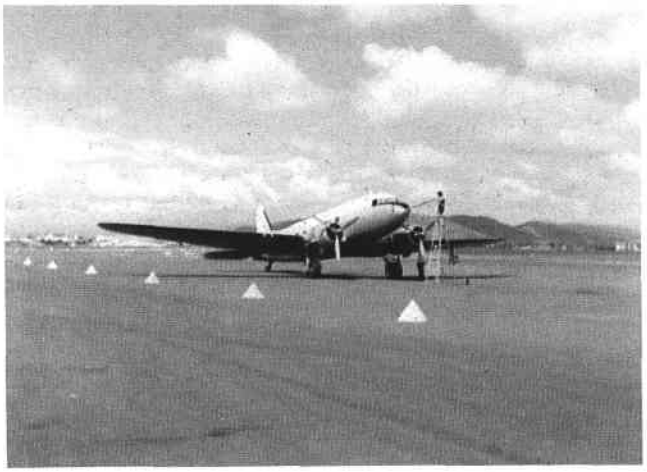
CLOUD FIELDS OVER LAND AND OCEAN

OBSERVATIONS FROM AIRCRAFT

Fields of cumulus clouds frequently form over both continental and oceanic areas of the earth. Besides producing rain they also affect climate and weather on a global scale. Especially in tropical regions cumulus clouds make a major contribution to the transport of energy in the atmosphere from mid-latitudes towards the equator.

The development of cloud fields over land proceeds differently from their development over ocean, mainly because of the difference in heating of land and water surfaces by solar radiation. The land suffers a much greater diurnal variation in surface temperature, and cumulus cloud fields over land usually form, grow and decay within the period from the forenoon until late afternoon. However, over the ocean, and particularly in the tropics, individual clouds are continually growing and decaying whilst nevertheless maintaining a fairly steady-state cloud field throughout the day and night.

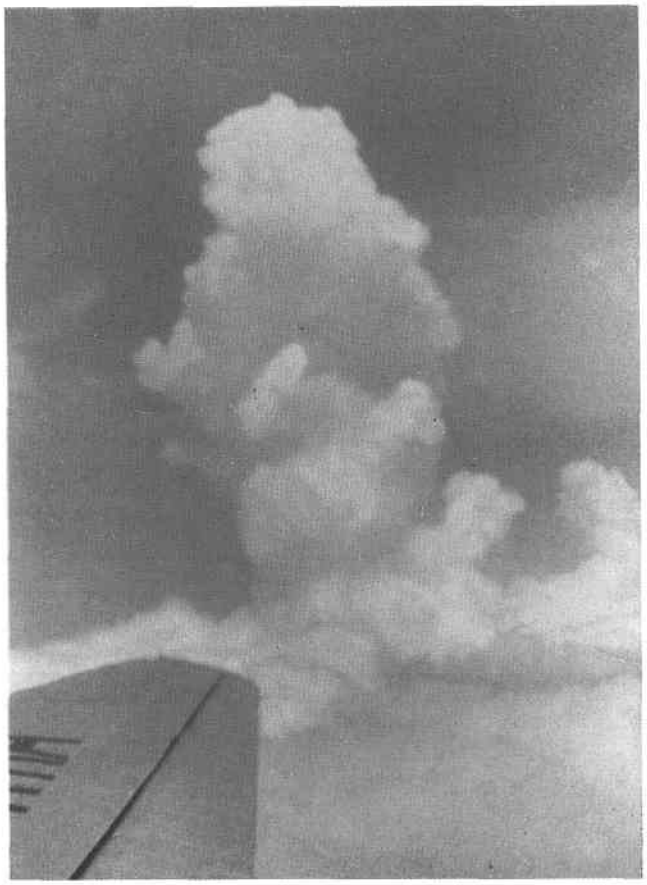
We have made measurements of the transfer of heat and moisture from the surface of the tropical ocean into the air during both fair weather and disturbed conditions. In the former condition rather uniformly populated fields of small, non-raining clouds form; in the latter, towering masses of cloud and heavy rain showers predominate, at least partly in consequence of the enhanced rates of upward transfer of heat and moisture.



Preparation for research into a field of tropical cumulus clouds.

Over the land the lowest layer of the atmosphere cools during clear nights as heat is lost to space by radiation from the earth. A statically stable, or temperature-inversion, region is formed in which the cooler, denser air is at the bottom of the layer. Convection, which commences soon after sunrise, lifts and finally disperses the overnight inversion, usually before noon in fair weather conditions. This process is a prerequisite for the formation of a field of cumulus clouds.

We have found that the rising convective elements (or "thermals", as glider pilots and others call them) are typically a few hundred metres in horizontal extent and 0.5 to 1 C warmer than the neighbouring air below the inversion. As these elements impinge on and mingle with



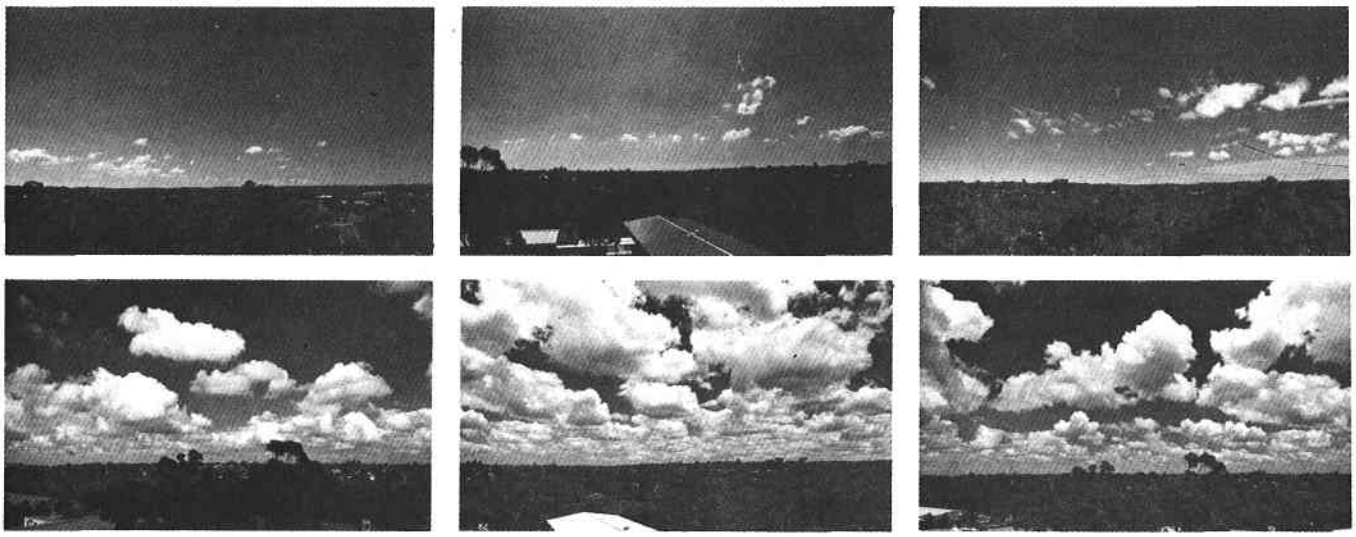
Vigorous growth of a cloud tower over the ocean.

warmer, stable air above they generate turbulent motion which mixes and disperses the inversion layer. This process has been studied quantitatively by the use of the Division's specially instrumented aircraft.

As the overnight inversion weakens and disperses, some moist air parcels rise buoyantly from near the ground to a height at which condensation takes place and the first small clouds begin to form. The typical sizes of air parcels rising up through the layer between the ground and cloud base (the "sub-cloud layer") and the intensity of associated humidity and temperature fluctuations are revealed by measurements made in horizontal flights at various levels in the sub-cloud layer. Such information is needed to create realistic numerical and theoretical models of the lower atmosphere.

CLOUD MODELS

The aim of any scientific endeavour is to gain an understanding of a phenomenon so that the future behaviour of that phenomenon can be predicted. Thus the observations made from our research aircraft can be used first to gain an understanding of clouds and then to help develop models of cloud behaviour. Subsequently aircraft measurements can be used to test the accuracy of any predictions given by cloud models. Particularly important uses for cloud models would be to test the efficacy of



The development of a typical field of fair-weather cumulus cloud over land is traced from mid-morning (top left), through noon, to a maximum in the early afternoon and to the onset of decay in mid-afternoon (bottom right).

cloud seeding on different cloud types and the effects of clouds on the global circulation.

A cloud model is a system of equations whose solution approximates the temporal and spatial development of parameters such as the velocity and liquid water content fields. Buoyancy-driven flows such as clouds are strongly turbulent, and turbulence is perhaps the outstanding unsolved problem of fluid mechanics. Cloud models therefore must reflect the complexity of the physical phenomena, and a computer must be used in general to obtain solutions.

Our earlier work has shown that one-dimensional models of individual clouds give an inadequate representation of observed cumulus clouds, and although three-dimensional, time-dependent cloud models are being developed overseas they require enormous amounts of computer storage and computation time. On the other hand, there are realizable cloud fields, such as trade-wind cumulus rolls or stratus layers, in which the average motion is at most two-dimensional to a good approximation. Models of such situations are much less complex than fully three-dimensional ones and consequently the implications of the model are more readily understood and compared with observations.

Cumulus clouds are initiated by the condensation of water in buoyant thermals which propagate upward from the ground and hence an understanding of the sub-cloud layer would seem to be necessary in obtaining an understanding of the cloud layers itself. The sub-cloud layer has been modelled simply and analytically as a field of interacting thermals. Although the predictions of the model are consistent with some observations, it is clear that further measurements of the detailed structure of this layer are required to develop more complete models.

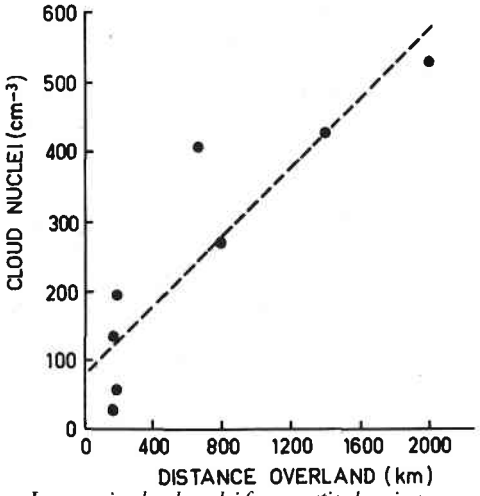
Below: A typical steady-state fair-weather field of small cumulus clouds over the tropical ocean.

THE ROLE OF PARTICLES IN CLOUD FORMATION

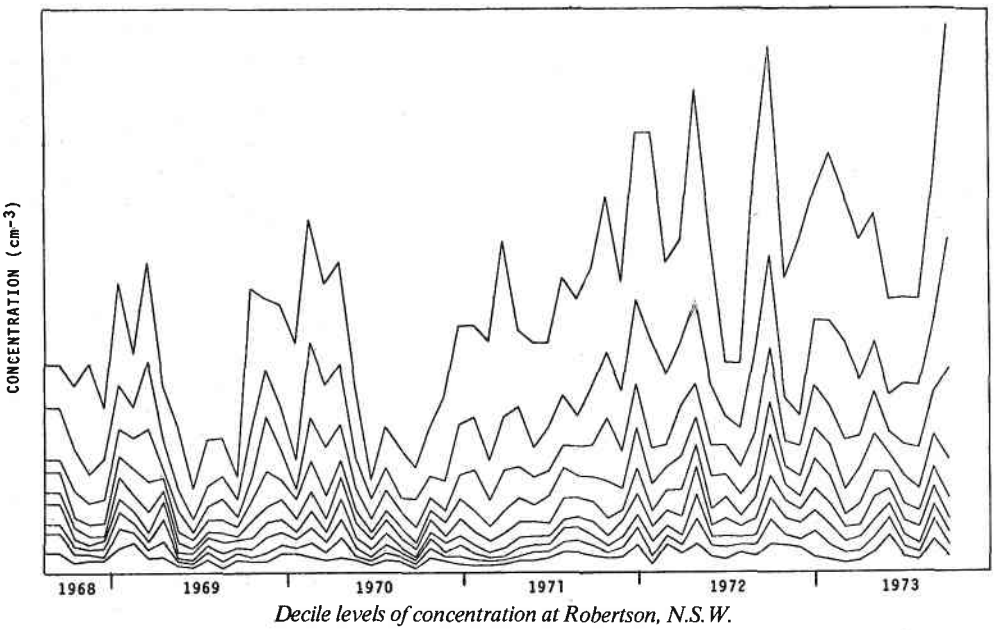
CLOUD CONDENSATION NUCLEI

The concentration and sizes of droplets in a cloud not only affect such gross properties as the ability of the cloud to reflect and absorb radiation but also some of its basic microphysical parameters: most importantly, its ability to produce drops large enough to fall out as rain. In turn, the concentration of droplets is strongly influenced by the particle content of the air below cloud base. However, not all of these particles are of importance in cloud formation, but only those which allow water vapour to condense on them at the low supersaturation of <1% commonly found in clouds; such particles are called cloud nuclei or cloud condensation nuclei.

The measurement of the concentrations of cloud nuclei is achieved in the laboratory by producing a small supersaturation region in a closed vessel and then counting by photographic, television or similar techniques the droplets contained in an intensely illuminated and well-defined volume. Measurements of this type have shown that often only a small proportion of the total particle population act as centres for condensation in natural clouds. It has also been shown that this proportion can vary widely and that there is a strong dependence of cloud drop concentration on the "dirtiness" of the air, with the drop concentration increasing as the air mass changes from clean maritime air to clean continental air to polluted urban air. We have found, for example, a threefold or more increase in drop concentrations in clouds downwind of cane-burning activities along the Queensland coast which was paralleled, in fact caused, by an increase in the concentration of cloud nuclei produced by the cane fires. A series of experiments in Victoria (where the passage of cold fronts produces abrupt changes in the wind direction — from continental northerlies to clean maritime south-westerlies) has shown that there is a correlation between the numbers of cloud nuclei and the time (or distance) the air mass has spent over land. However, this observation does not necessarily imply a land-based production mechanism, since there are many other meteorological variables which change in unison with the continental-to-maritime change in air flow. Such increases in cloud nucleus concentrations lead to clouds containing large numbers of very small drops which are more stable, rain less readily and reflect more solar energy back to space than clouds formed in cleaner air with lower nucleus concentrations.



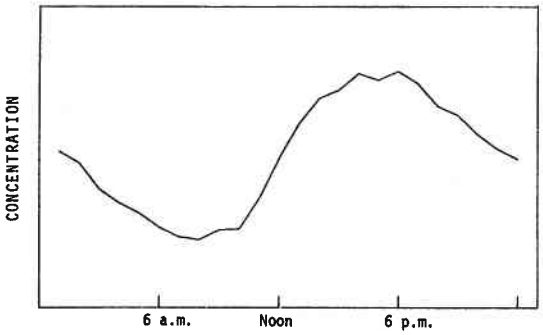
Increase in cloud nuclei for a particular air mass moving inland over southern Australia.



Decile levels of concentration at Robertson, N.S.W.

The resulting importance of cloud nuclei to the radiation budget and climate as well as to rain formation has led us to study in some detail the composition, characteristics and distribution of cloud nuclei. The particles are very small (10^{-17} to 10^{-16} g in mass), and most of them are probably produced from trace gas pollutants in the atmosphere. Most are relatively volatile and in that

respect behave in the same way as laboratory-produced particles of ammonium sulphate. Ammonium sulphate is a common pollutant in the troposphere and stratosphere and the cloud nuclei are probably made up of this compound, almost certainly mixed with other materials. The aerosol as a whole is a dynamic population in which the particles continually interact with each other, experiencing coagulation, condensation and evaporation. While the specific source or sources of all small particles are still obscure, we have been able to produce particles, some of which are effective as cloud nuclei, by irradiating with ultraviolet light samples of clean atmospheric air from which the original particles were removed by filtration. The composition of these artificial nuclei appears to be similar to that of natural nuclei; on the other hand, the wavelength required is not present in the atmosphere in appreciable amounts so that we cannot be certain of the relevance of these results to problems about the source of the natural nuclei.

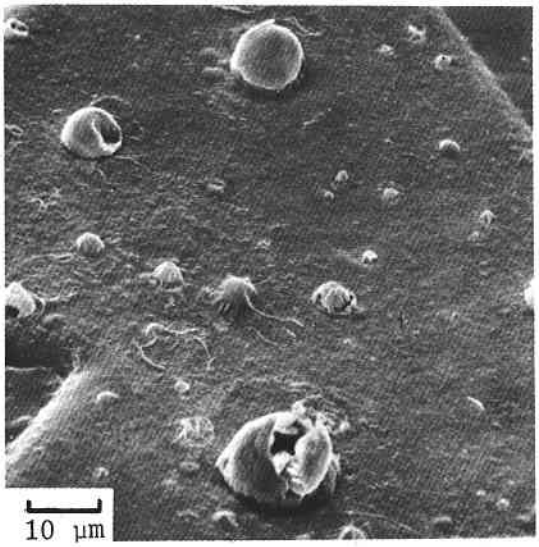


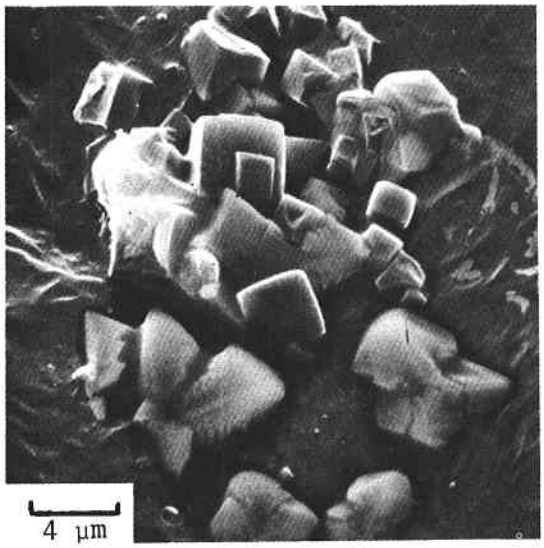
Diurnal pattern of cloud nuclei at Robertson, N.S.W., averaged over days on which the wind was steady.

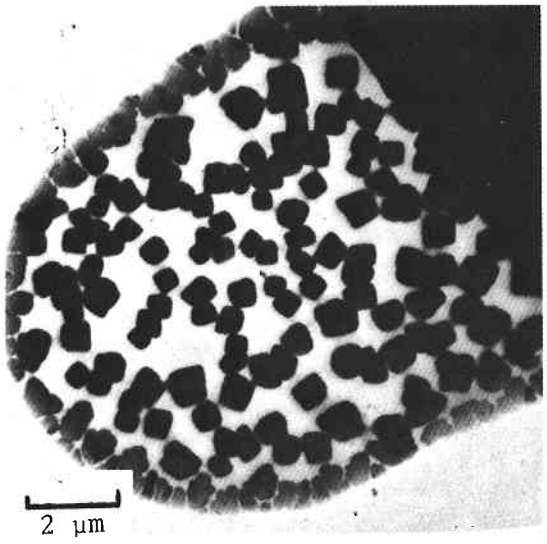
Long-term measurements of cloud nucleus concentrations have been under way since 1968 at a field site at Robertson, N.S.W. Originally planned to be of five years' duration these measurements were completed in 1973 but have been continued at a lesser sampling frequency since then. The figure illustrates how the concentrations varied month by month during the five-year period. An increase in the "dirtier" samples (higher decile curves) is not surprising, but the cleaner samples (e.g. the first and second deciles, representing the values exceeded by 90% and 80% of all measurements) also show a distinct increase with time. In view of the high seasonal variability and the possibility of systematic changes in circulation during the period one must be careful about the interpretation of such a trend.

The averaged diurnal variation of cloud nucleus concentration at Robertson for the 1968-1973 period is shown above. This figure refers to days on which the wind was steady and from a "clean" direction, and shows a very distinct pattern, with a minimum around 6 a.m. and an evening maximum. Since this pattern was first reported here it has also been found by other workers in data from upstate New York; it is probably a general feature associated with a photochemically-induced diurnal cycle in trace gas composition or in the particle formation process itself.

The concentrations of the cloud nuclei, typically of the order of hundreds per cubic centimetre, are important in determining the total number and the average size of cloud droplets, but they only partly control the stability of





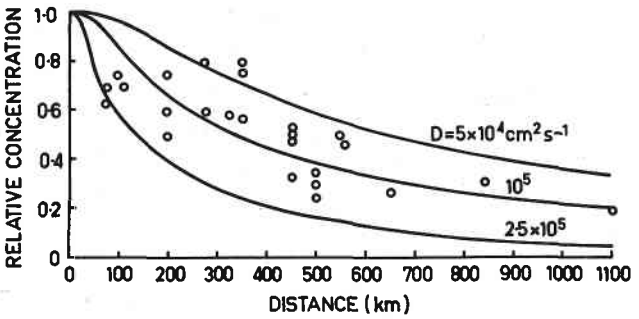


Large salt particles observed under the electron microscope.

the cloud. Precipitation-size elements usually form as the result of collisions between a few larger drops and the background sea of much smaller drops. Larger particles above about 0.1 μm are known to result from salt spray and ocean foam. By virtue of their chemical affinity for water, these salt particles commence to take up water at relative humidities above 75% and can therefore form droplets well before they enter cloud base, obtaining a head start in the race to form larger droplets. Although much rarer than the smaller condensation nuclei (typically attaining concentrations about 0.1 cm^{-3}), "giant" salt particles may therefore influence rain formation.

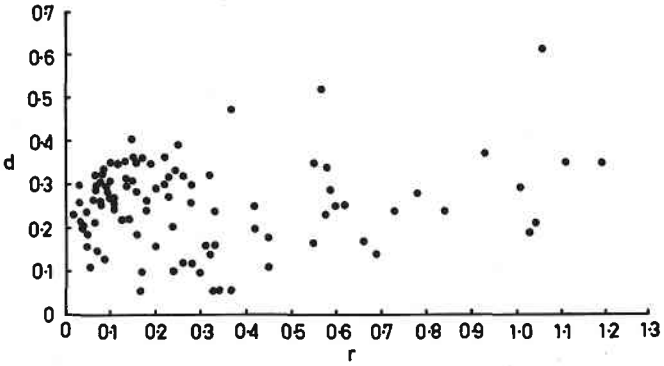
Recent laboratory and field evidence accumulated by this Division suggests that the "tail" of the droplet spectrum is also important in some of the ice-related processes of the colder clouds. The collision between an ice crystal and a large cloud droplet can generate splinters of ice which subsequently grow into larger ice crystals and affect the delicate ice/water balance of these clouds. This has given further impetus to the study of large drops and hygroscopic particles.

It is very difficult to measure particles in the size range of 0.1 to 0.5 μm and at the same time determine whether they are hygroscopic. Particles of this size are too small to be collected by impaction and are at the lower limit of detectability of optical sizing techniques. For particles above about 0.5 μm diameter the Division has used three techniques. In the simplest of these a glass slide coated with a hydrophobic substance is exposed to the airstream from an aircraft. The particles, which often exist as solution droplets at the relative humidities at which they are collected, impact on the slide and adhere to it. When



Relative concentration of salt particles of mass greater than 10^{-12} g as a function of penetration. The curves are theoretical for various eddy diffusivities; the circles represent the results of field observations.

the slide is subjected to a controlled humid environment back in the laboratory, the salt particles again grow into visible droplets and can be sized and counted using a microscope. A more recent but similar technique impacts the particles on fine copper grids coated with nitrocellulose, suitable for study in an electron microscope. When collected the solution droplets spread and form a slight depression in the thin film, and the "spread diameter" gives a measure of the original nucleus mass. Slow drying allows the salts to crystallize and the general cubic shape is easily recognized. Care is needed however in interpreting the shapes of the crystals, since rapid evaporation caused by the high vacuum of the electron-microscope sometimes produces unusual shapes. The third method relies on the well-known yellow flash that accompanies the vaporization of any salt containing sodium. In an instrument which is small enough to be carried in a light aircraft the sampled air is drawn into a flame and the magnitude of the light flash emanating from each particle is measured with a photomultiplier and associated electronics. A major advantage of this technique is that the concentration and sizes can be displayed as the air is sampled, and the data reduction inherent in the previous two methods can be eliminated.



The dispersion (d) near cloud base as a function of the ratio (r) of the observed liquid water to that pertaining to an unmixed parcel.

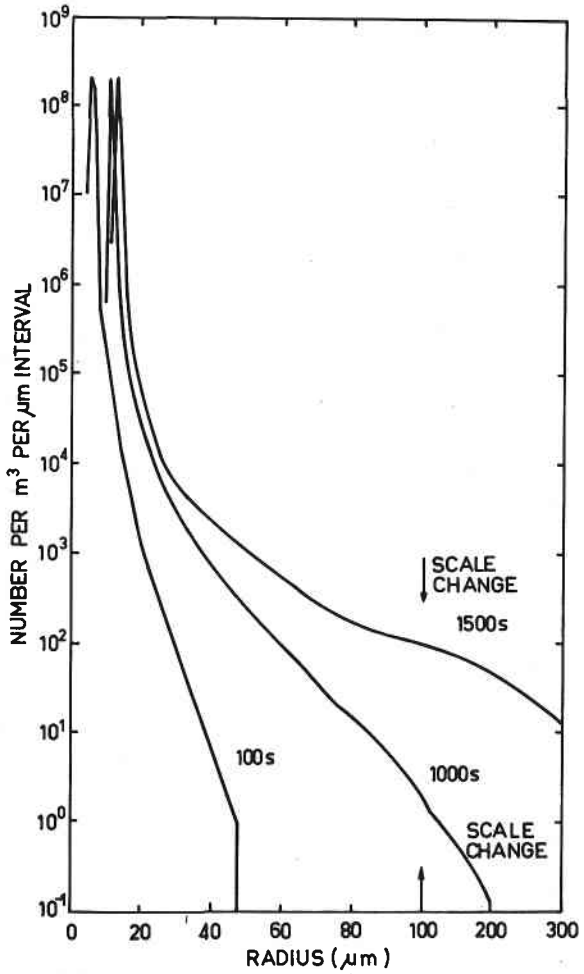
Measurements carried out in eastern Australia indicate that under onshore wind conditions the concentration of giant hygroscopic particles near the coast is about 0.1 cm^{-3} (10^5 m^{-3}). At cloud levels the concentration decreases slowly as the air mass moves inland, being about one-fifth of the initial value after a penetration of 1000 km. Since the smaller cloud condensation nuclei are increasing in numbers as the air spends more time over the land, the implication is that the transition from a maritime to a continental aerosol involves two quite separate modifications, one producing a decrease in the large hygroscopic particles and the other an increase in the smaller cloud nuclei. It is the interplay between the droplet populations associated with these two effects that determines the stability of clouds which form.

FORMATION AND GROWTH OF CLOUD DROPLETS

Measurements of the supersaturation spectrum of cloud condensation nuclei and large hygroscopic particles represent only the first step in providing a description of cloud microphysics. An instantaneous measurement of the cloud droplet spectrum, itself an important cloud variable, also allows us to check our ideas of condensation growth. For this purpose the Division previously used small soot-coated slides on which the cloud droplets impacted, leaving clear impressions related to their size. Unfortunately each slide could sample only about 10 cm^3 of the cloud and considerable effort was involved in data reduction to obtain the desired information about the droplets in this small volume. With a view to reducing this labour we have recently acquired a laser light-scattering probe, which is mounted externally to the aircraft. During cloud penetration the droplets pass through a well-defined laser beam and the pulse of light scattered by each drop (which is proportional to the size of the drop) is detected by a sensitive photo-diode, amplified, and electronically sorted according to its amplitude. In this way, the concentrations of droplets in 15 size categories from 3 to

45 μm in diameter are available in real time in flight.

Analysis of a large number of droplet samples collected on sooted slides in cumulus cloud has allowed us to describe in some detail the general features of the droplet spectrum: many droplet distributions are found to have more than one peak, the bimodal distributions increase in frequency with height above cloud base and with decreasing stability of the environment, the droplet distributions are similar at all positions across the cloud at any one height, and the bimodal size distributions are not confined to cloud edges.



Calculated droplet distribution at heights of 100, 1000 and 1500 m above cloud base.

Measurements of the liquid water content in cumulus clouds have shown it to be much less than it would have been in the absence of mixing between the cloud and its clean-air environment. However, as can be seen from the diagram, the dispersion of the cloud droplet size distribution (i.e. its shape) at levels near cloud base seems to be unrelated to the amount of mixing between the cloud and its surroundings.

Observations of the droplet distribution have also been made at fixed levels during the middle lifetime of cumulus clouds. An analysis of the results suggests that there is little change with time in the shape of the distribution or in the mean droplet diameter. However, the number concentration of droplets initially increases and then decreases, with the greatest relative changes occurring in the upper levels of the cloud.

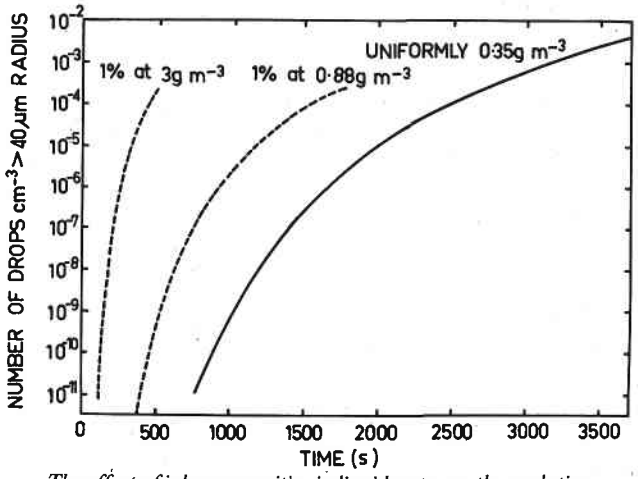
NUMERICAL SIMULATION OF THE CLOUD MICROSTRUCTURE

Many of the features of the droplet distribution found a few hundred metres above cloud base are theoretically understood and have been modelled numerically. It has been shown theoretically that the two most important parameters are the cloud nucleus spectrum and the updraught velocity. A feature that we cannot yet explain satisfactorily is the observed width or dispersion of the droplet distribution. Numerical calculations predict fewer small droplets than are generally observed. While observations suggest that this feature is caused by mixing between the cloud and the environment, no satisfactory theoretical model is yet available.

Our interest in the droplet size distribution well above cloud base centres on the rare, large droplets which eventually become raindrops. In this region of the size spectrum there have been few observations. However, our numerical calculations have shown that growth by collision and coalescence can lead to the formation of a "tail" on the distribution. In warm clouds it has been shown that the favoured few drops in the tail of the distribution are the source of precipitation particles and that the growth rate of the tail depends markedly on the total droplet concentration and on the liquid water content of the cloud. The liquid water itself is, of course, a function of the height of the air parcel above cloud base and of the amount of environmental air mixed into the cloud. In those clouds where the likelihood of precipitation is marginal, numerical calculations show that high concentrations of giant nuclei may influence the time taken for the onset of precipitation.

Our cloud observations have shown that there are often significant fluctuations in the horizontal liquid water profiles on a large scale. Calculations show that clouds containing very small pockets of high liquid water can produce rain much more rapidly than identical clouds having a more uniform liquid water profile but the same average liquid water content.

All the calculations described above have been performed with cloud models in which the updraught velocity is an imposed parameter. The future work of the Division will be concerned with the development of dynamic models, i.e. ones which allow for the interaction between the microphysics and the air motions in the cloud and environment.

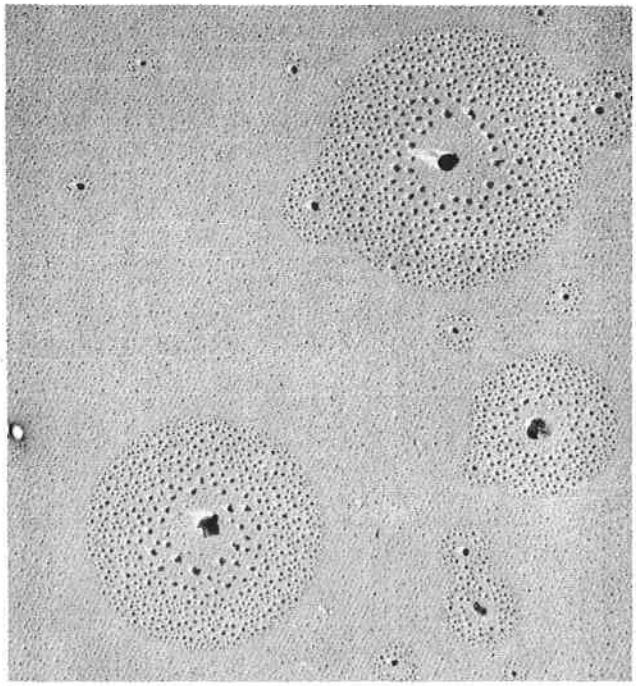


The effect of inhomogeneities in liquid water on the evolution of the droplet size distribution.

PARTICLES IN THE STRATOSPHERE

Proceeding upwards in the atmosphere, well above the regions normally affected by clouds and weather processes, one comes across a rather surprising layer. This layer, centred at a height of about 20 km, that is in the lower stratosphere, is found world wide. It consists of submicroscopic particles of sulphuric acid and ammonium sulphate, typically a few tenths of a micrometre in diameter, suspended in a stable layer of air. Too small to be seen directly, these minute particles can however scatter and absorb solar radiation. A spectacular demonstration of this is a vivid twilight glow which appears when their concentration is high. A far more serious but less obvious consequence of the interception of solar radiation by small particles is the cooling at the ground and warming of the upper atmosphere that can ensue. While such effects on temperature are usually very small significant temperature variations have been noted following massive volcanic eruptions.

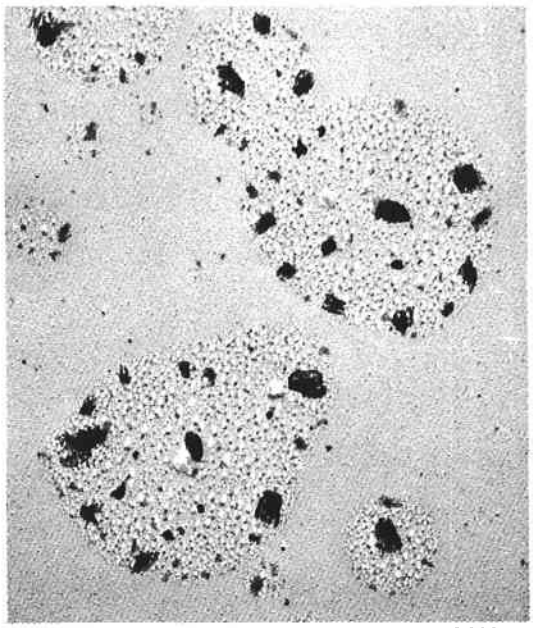
Once in the stratosphere, particles may stay there for up to one or two years, depending on their altitude. This



Transmission electron microscope photograph of typical stratospheric particles containing sulphuric acid.

is in marked contrast to the residence time of particles in the lower atmosphere, where clouds and rain ensure their removal within a period ranging from days to weeks. As a result, any particles introduced in the stratosphere — for example by supersonic aircraft or the proposed space shuttle — can be expected to accumulate with time. Exhaust products from the aircraft engines may also interfere with the normal processes of particle production, in which ultraviolet light from the sun produces chemical reactions between certain atmospheric trace gases; for example, the exhaust products may act as catalysts to enhance the natural concentration of particles.

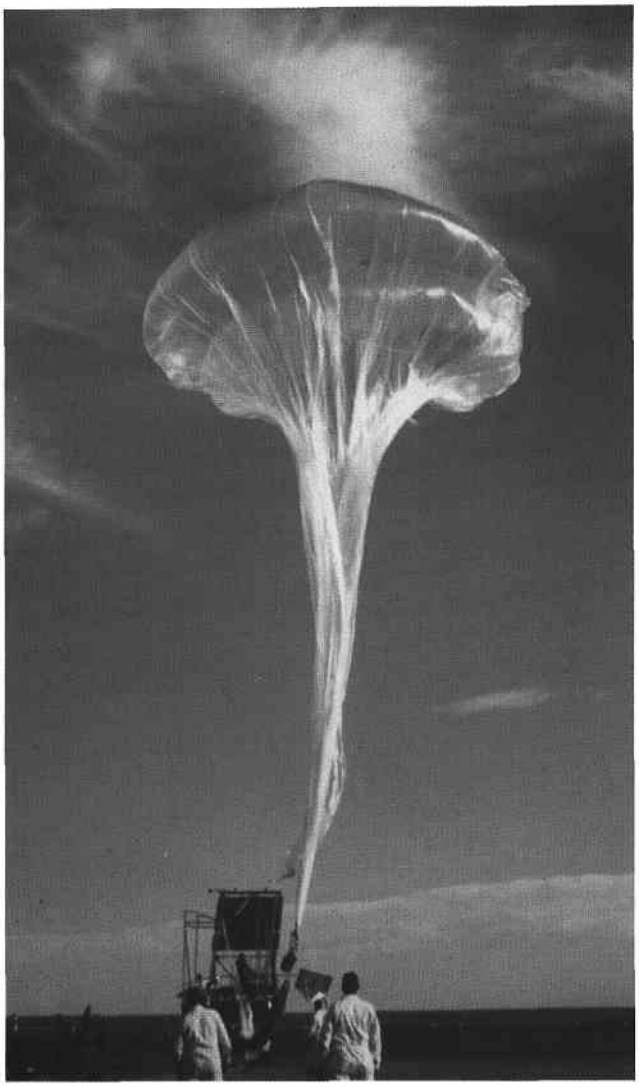
Before any significant changes in the stratosphere occur as a result of man's activities, it is essential that we study the size, composition and concentration of the naturally occurring particles and fully understand their production and transport mechanisms. Since most pollutant material is likely to be injected in the northern hemisphere, southern hemisphere levels should provide a reference level for determination of any trend as it occurs. It is clearly important that studies be made in both hemispheres.



Stratospheric particles collected in mid 1975 from Mildura, after the eruption of Volcan de Fuego in late 1974.

Direct measurements of the stratospheric layer began in the Division in 1962 with sampling from U-2 aircraft. The identification of particles of volcanic origin from the 1963 Agung eruption were a direct result of this work. From late 1967 to 1974 the Division sampled stratospheric particles with an impactor designed in the Division and flown on balloons by the U.S. Atomic Energy Commission along with their instruments for measuring radioactivity.

The impactor used for particle sampling utilizes a small vacuum pump to create a high-velocity air jet through a small nozzle. This air jet impinges on a slowly rotating disk carrying specially prepared electron microscope screens. As a result of their inertia, the small particles are unable to follow the air; they impact on the prepared surface and, partly because they are semi-liquid, they stick. This method of collection allows both the size and concentration of particles to be determined as a function of altitude. It also enables a limited number of chemical tests to be performed on the particles by evaporation of a thin film of a suitable reagent on to the grid surface, either before or after collection. An automatic camera viewing the horizon and taking photographs approximately every 600 m is carried with the impactor and provides further evidence of dust-rich layers which are revealed as bright bands in the darker background sky. Additional information, such as air temperature and the performance of the instruments on board, is telemetered to the ground station and recorded for later inclusion in the analysis.



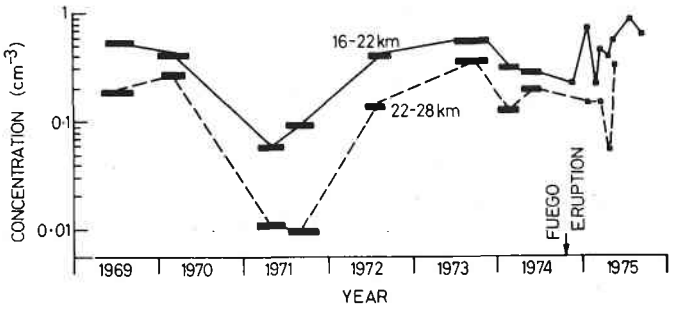
A large helium-filled balloon commencing to lift its payload from the launch truck at the Balloon Launching Station at Mildura.

The instrument package is carried aloft by large plastic balloons launched from the Balloon Launching Station at Mildura. To reduce any risk of contamination, sampling is carried out from a position more than 100 m below the balloon on ascent and during the float period. After the predetermined float period the instruments are released from the balloon and parachuted back to earth. During the descent, sampling takes place 100 m below the parachute. After the parachute descent, the instruments, which are tracked through their flight by radar, are located by a search aircraft which directs the ground crew to make a recovery.

In 1974 the program was reorganized and expanded. It now operates independently with regular bi-monthly sampling from the Mildura site, providing the only continuous southern hemisphere data of its type. This program also includes co-operative ventures with the University of Melbourne, the Max Planck Institute and the CSIRO Division of Atmospheric Physics to extend the type of information gathered and to obtain additional data such as water vapour concentration.

Aerosol layers appearing as bright bands near the horizon with the camera pointed towards the sun. The photograph was taken in mid 1975 from an altitude of 27 km at a time when increased concentrations of particles were observed.

In late 1974, the eruption of Volcan de Fuego in Guatemala (at 14°N.) provided a chance to obtain data on the build-up and dispersion of material injected into the stratosphere. Effects of this eruption were noted widely in the northern hemisphere about a month after the eruption. At the Mildura site however, at 34°S., the main influx of this material took 7½ months to arrive. Analysis of data obtained up to this period and subsequently is providing unique information on the behaviour of the stratospheric layer.

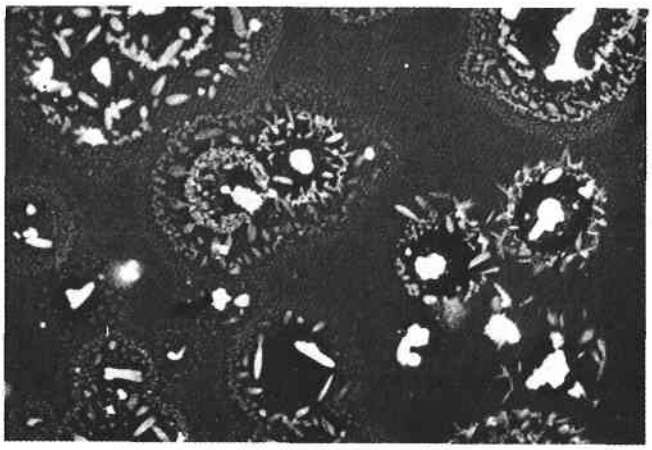


Altitude-averaged concentrations of particles per cubic centimetre with diameters greater than 0.23 μm — obtained with the impactor flown from Mildura.

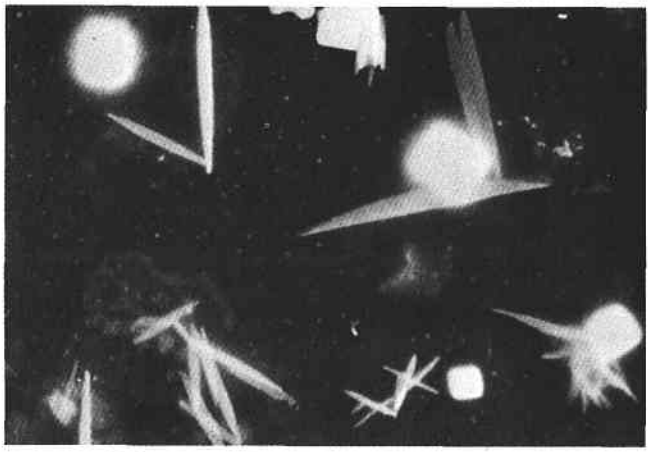
Results on the seasonal and longer-term trends in particle concentration are only now becoming available. Many of the questions basic to our complete understanding of the behaviour of these particles however still remain unanswered.

CHEMICAL ANALYSIS OF PARTICLES

For many years some excellent techniques have existed for determining the elemental composition of quite small masses of material. However, these techniques are unsuitable for detecting the most common atmospheric ions (sulphate and ammonium), cannot detect nitrate, and have marginal performance with some other common ions. Neither can they provide the most important information needed in interpreting the origins and effects of atmospheric particles — i.e. the size distribution of particles containing a given ion or substance and the proportion of that substance in each particle.



Sulphuric acid particles (captured downwind of the Mt. Isa lead and copper smelters) on a thin film of calcium fluoride.



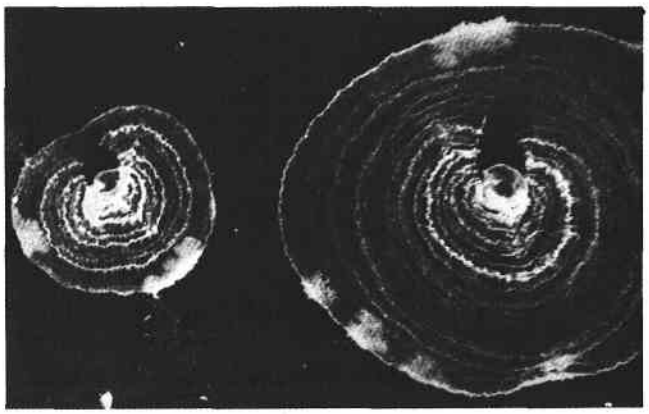
Sea-salt particles recrystallized on calcium fluoride.

The Division of Cloud Physics has now developed a technique for detecting the common atmospheric ions in particles down to a very small size and has used it in conjunction with a simple standard method for detecting the overall amount of the same ions in larger aerosol samples. The single-particle analysis technique uses conventional chemical reagents but applies them over the particles in a thin film by vacuum deposition. Exposure to water vapour promotes a reaction, the products of which are viewed with an electron microscope. Some examples are discussed below.

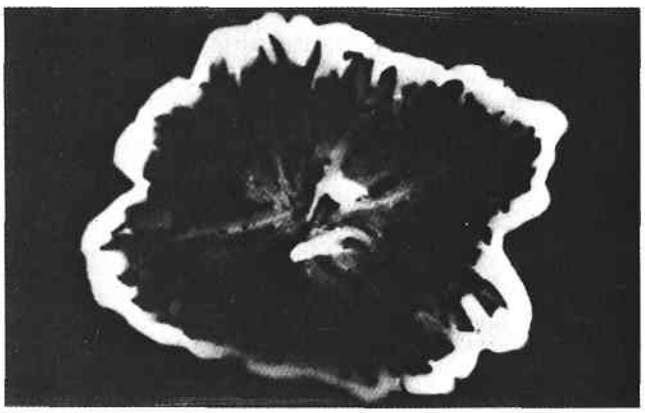
Droplets of sulphuric acid are by far the most common stratospheric particles and are also common in industrial effluents. If they are captured on a thin film of calcium fluoride, or if this compound is used to coat them after capture, etch marks and characteristic crystals are formed. Ammonium sulphate particles — quite the most numerous particles in the atmosphere — also slowly etch calcium fluoride in a humid atmosphere, leaving a single central black crystal if they are large, or a small dark flag on the edge of the hole if they are small. Other ammonium salts behave in a similar fashion. This coating also allows soluble particles such as sea salt to spread out and recrystallize, showing the individual components.

Sulphate is probably the most common atmospheric ion and a thin film of a soluble barium salt (chloride or iodide) is used to detect it. A thin film of inert silicon monoxide placed over the reagent before supplying water vapour improves the reliability of the reaction and leaves a "shadow" which shows the size and shape of the unreacted particle.

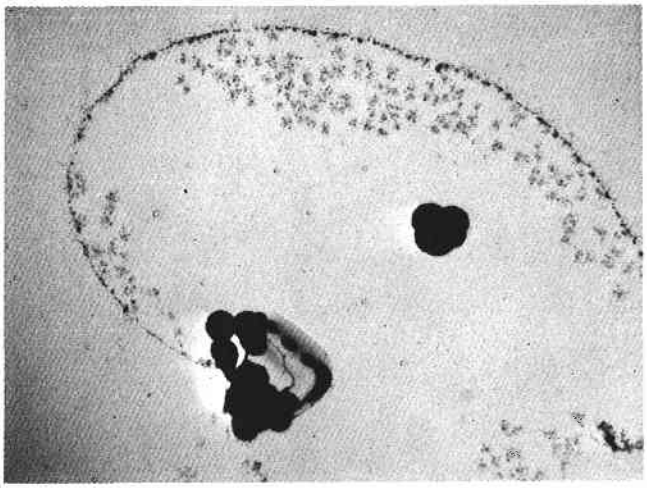
Nitrate is another common ion in continental atmospheres and it can be detected using the organic reagent "nitron". When silicon monoxide is used as a shadow the reaction looks rather like the sulphate reaction. Without the silicon monoxide needle-shaped crystals are formed which may be confused with some of the recrystallized sea-salt components.



Ammonium sulphate particles after coating with barium chloride and silicon monoxide and exposure to water vapour.



Ammonium nitrate particle after coating with nitron and silicon monoxide and exposure to water vapour.

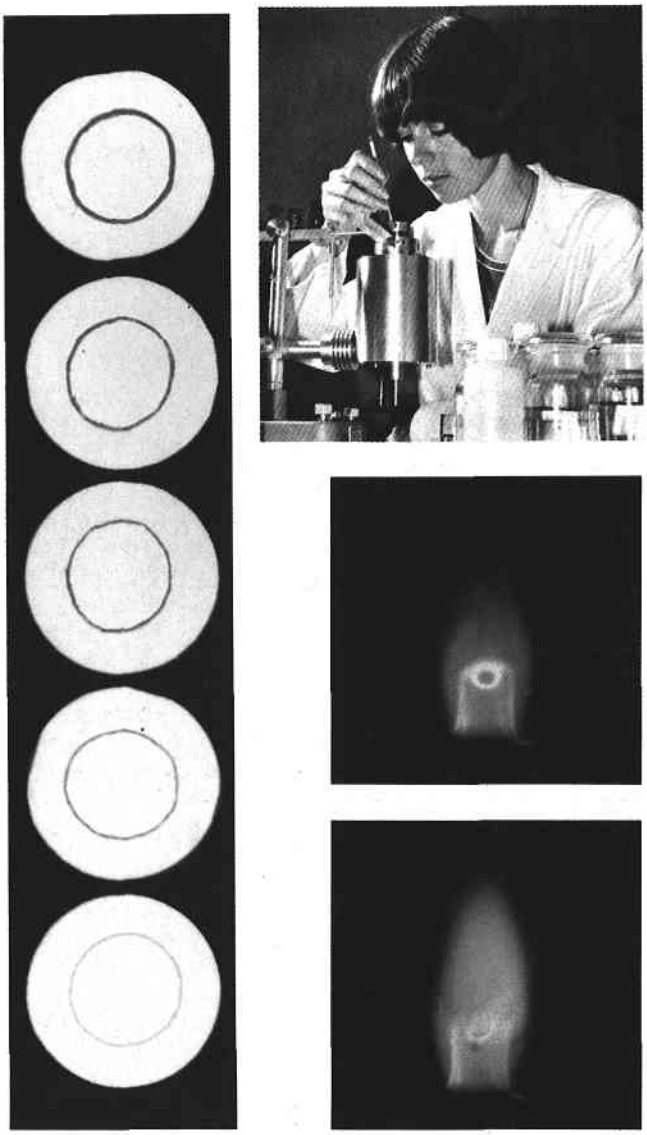


10% of ferric sulphate in sodium chloride after coating with 8-hydroxyquinoline and exposure to water vapour.

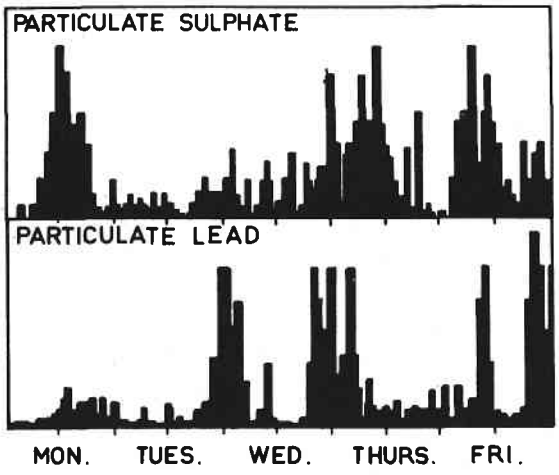
Salts of calcium, magnesium, iron, and other metals are also often found in aerosols. A useful "catch-all" reagent is 8-hydroxyquinoline, which gives precipitates with most metallic ions other than sodium, potassium and ammonium over a wide range of acidities.

When the technique described above was first used it was difficult to make it fully reliable but the use of permeable films such as silicon monoxide over the particles before exposure to water vapour appears to have solved the early problems.

For determining the overall proportion of the common ions, a large aerosol sample is collected on filter paper and clamped in place over a heated ring. A solvent, such as dilute hydrochloric acid, is dripped on to the centre of the paper leaching out the soluble material and carrying it outwards. As the solution encounters the hot ring, the solvent evaporates, concentrating the salts in a very thin ring. Normal colorimetric analytical techniques are then applied to this ring. This procedure is known as the "ring oven" technique. It requires a minimum of equipment and is sufficiently sensitive to be very useful at normal atmospheric concentrations of the more common ions. It has been applied to the aerosols collected on survey flights over Australia (described later in this booklet) and also to



Above left: Rings produced by 5, 2, 1, 0.5 and 0.2 μg of lead.
Above right: Undisturbed flame from photometer (top) and with sodium particle in the flame (bottom).
Top right: The ring oven.

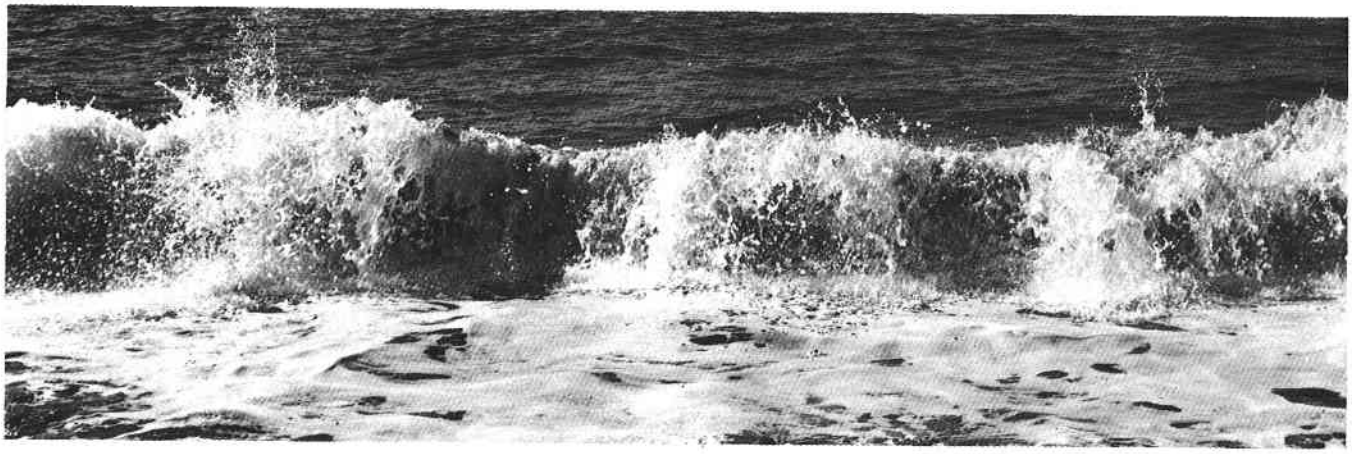


Variations in concentrations of sulphate and lead with time as shown by the ring oven.

a study of ions present in city air and sampled at hourly intervals for one week.

Both the ring oven technique and that of individual particle analysis are tedious and cannot be used during aerosol collection. It would often be a great advantage to be able to read out continuously the concentration of particles containing specified amounts of a given element. For this reason a "flame photometer" has been developed for use in an aircraft. The particles are entrained into a small stable flame and emit characteristic radiation as they become heated. A narrow-band optical filter selects the radiation peculiar to a particular element, the light is detected by a photomultiplier, and the pulses are counted. So far only a sodium detector has been flown. It showed (as particle collectors do) that sodium-containing particles are mostly confined below cloud tops or inversions and that there are considerable variations in their concentrations.

ORIGIN OF AEROSOLS

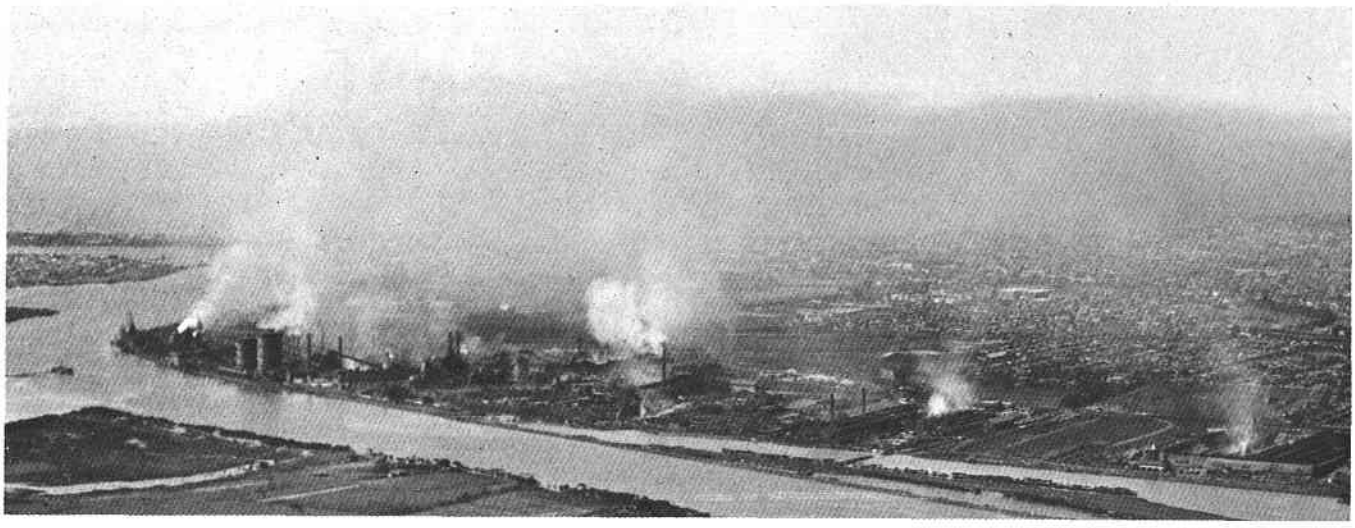


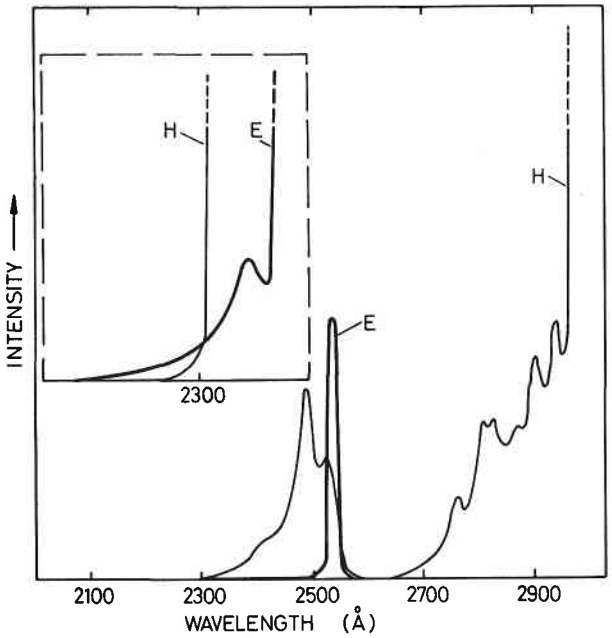
The dust blown from a dirt road or a dry sandhill is a familiar form of atmospheric aerosol, as is the saline spray blown from breaking surf. Such particles are huge compared to the majority of the particles found in the atmosphere, where for every particle greater than 1 μm diameter there may be a thousand between 0.1 μm and 1 μm and a million between 0.01 μm and 0.1 μm. It is difficult even in the laboratory and in industry to produce particles below 1 μm by grinding, pulverizing or spraying, and it seems unlikely that many such particles are present in soil, sand or rocks at the earth's surface; even if they are there their removal by air is likely to be difficult, since such fine materials adhere tenaciously and even when detached have so little inertia that they are almost immediately stopped by the viscous drag of the air.

Particles below 1 μm are found in copious numbers in smoke and in natural "blue hazes", and in both situations some, at least, of these particles originate from materials which were originally gaseous. It is known from laboratory work that sulphuric acid droplets can be formed under ultraviolet irradiation from a mixture of particle-free water vapour and sulphur dioxide, and that ammonium sulphate particles can be formed from water vapour/sulphur dioxide/ammonia mixtures. Ammonium sulphate is commonly found in bulk aerosol samples and in rain, while sulphur dioxide and ammonia are ubiquitous trace pollutants in the atmosphere even in nominally "clean" air.

As time progresses other gaseous reactions (photochemical or otherwise) may be found which also can cause particles to form. What is required for particle formation (in the laboratory or in the atmosphere) is a vapour pressure which exceeds perhaps a hundredfold or more the equilibrium vapour pressure of the compound concerned in solid or liquid state. The amounts of material involved can be exceedingly small. Measurements in this laboratory indicate the vapour pressure of ammonia in equilibrium with solid ammonium sulphate to be around 10^{-10} atmospheres at 20 C. This is equivalent to about 0.1 μg per cubic metre of air, and similarly low levels of concentration are likely to be involved for other gas traces.

The mass concentration of 1000 particles per cubic centimetre with radii around 0.01 μm is itself only about 0.01 μg per cubic metre so very low concentration levels are a necessary feature of the problem. At such low levels molecular encounters are infrequent and molecules are likely to strike the walls of any containing vessel as often as they encounter molecules with which they may react. For these reasons it is difficult to extrapolate to the atmosphere results obtained in the laboratory, and the difficulty is compounded by disagreements, often serious,

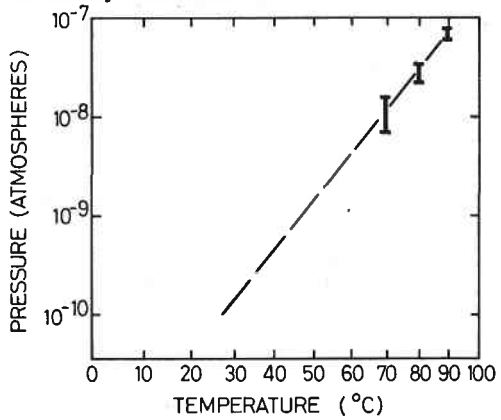




UV spectra of two lamps. Illumination of filtered air by lamp E produced particles.

between apparently similar measurements. The figure shows the UV spectra of two lamps one of which (H) produced no particles whatever in filtered outside air, whereas radiation from the other (E) was a copious source of small particles (producing 10,000 to 100,000 particles per cubic centimetre where lamp H was totally ineffective). The difference was attributed to relatively slight amounts of radiation emitted by lamp E at wavelengths shorter than 2250 Å. With a very much higher detection sensitivity this radiation was observed (inset to figure) in the output from lamp E.

While results such as these give some insight into particle production they do not necessarily represent the process whereby particles are formed in the free air. The wavelengths required in the laboratory experiments are almost entirely absent in the atmosphere; on the other hand no container walls are present in the atmosphere to adsorb or modify molecules.

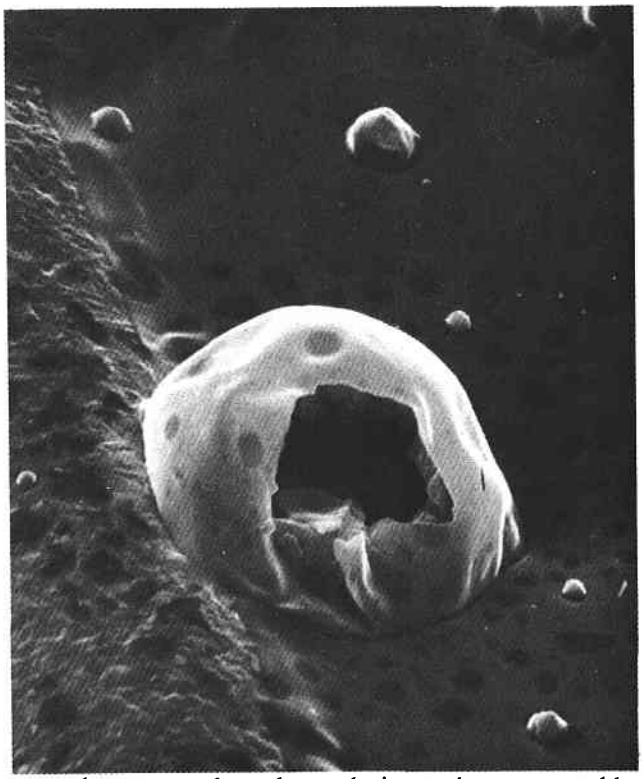


Vapour pressure of ammonium sulphate as a function of temperature (the vapour pressure of gaseous ammonia ranges from 10^{-9} to 10^{-8} atmospheres).

The oceans are an obvious natural source of hygroscopic particles, NaCl in particular. Minute droplets of sea water are ejected into the atmosphere when bubbles of air submerged by breaking waves burst as they rise back to the surface. Subsequent evaporation of the water leaves the salt particle in the atmosphere, whence it can be carried upwards and inland to act as a possible condensation nucleus. These particles are rarely carried above cloud top or inversions, since at these levels they have usually accumulated sufficient water to fall against any weak updraughts.

Under conditions of fine weather and on-shore winds, the particles sediment out with terminal velocities of the order of 0.01 cm s^{-1} (10^{-4} m s^{-1}). They can be carried many hundreds of kilometres inland and are consequently not restricted to maritime regions only. They are really only effectively removed from the atmosphere by rain — either by acting as precipitation centres as already described or by falling victim to scavenging rain drops.

Such hygroscopic particles from the ocean constitute an important component of the atmospheric aerosol but their importance is mainly confined to the "giant" range of particle size (>1 μm). The ocean does not seem to be as important a source for smaller particles, especially below 0.1 μm, where the ubiquitous sulphate particles predominate. At one time it was thought that when giant particles dried out (in the air or on the earth's surface) they produced smaller fragments as a result of the stresses involved. It can certainly happen (as an accompanying electron microscope photograph shows very clearly), but it does not now appear a likely major source of smaller atmospheric aerosol particles. This is probably because the drying-out process is rarely vigorous enough in the

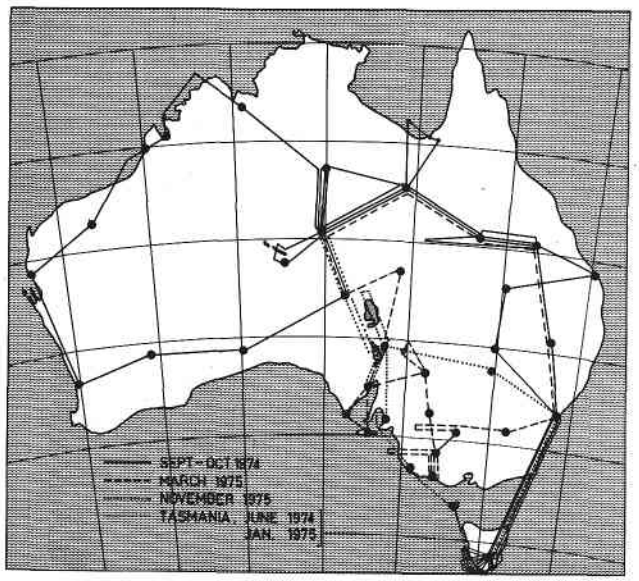


atmosphere to produce the explosive action suggested by the appearance of the micron-sized salt particle shown in the photomicrograph (that particular particle may well have exploded in the electron microscope because of the vacuum or the irradiation by the electron beam). The progressive dominance of ammonium sulphate or related compounds in the submicron size range is in accord with the idea that breakdown of bulk material becomes progressively more difficult with decrease of particle size whereas production from the vapour becomes progressively more effective.

MONITORING THE ATMOSPHERE

The importance of understanding the causes of climatic change is now fully recognized, and attempts are being made under the auspices of the World Meteorological Organization to establish a world network of monitoring stations, called baseline stations, at sites free from local pollution. The aim is to provide continuing, long-term, high-quality records of those quantities that may influence climatic change on a global scale. Solar radiation, the distribution of aerosol particles, and the concentrations of various gases, notably carbon dioxide, are all given high priority in this monitoring program.

Four monitoring stations, in Alaska, Hawaii, Samoa and at the South Pole, have already been established by the United States. The Australian Government has agreed in principle to fund one such station, and the Division of Atmospheric Physics through its interests in radiation and gases and the Division of Cloud Physics through its interests in aerosols have assisted other government agencies in the site selection. Partly because of the distribution of existing stations the choice has been narrowed to southern Tasmania, with a likely site located on South-East Cape. While CSIRO is likely to maintain a continuing research interest in any monitoring station which is set up the main responsibility for the routine observations will be borne by the Department of Science.

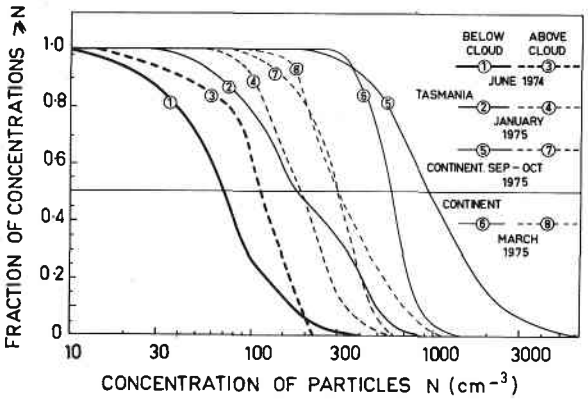


Routes flown while collecting particles at altitudes from 0 to 6000 m.

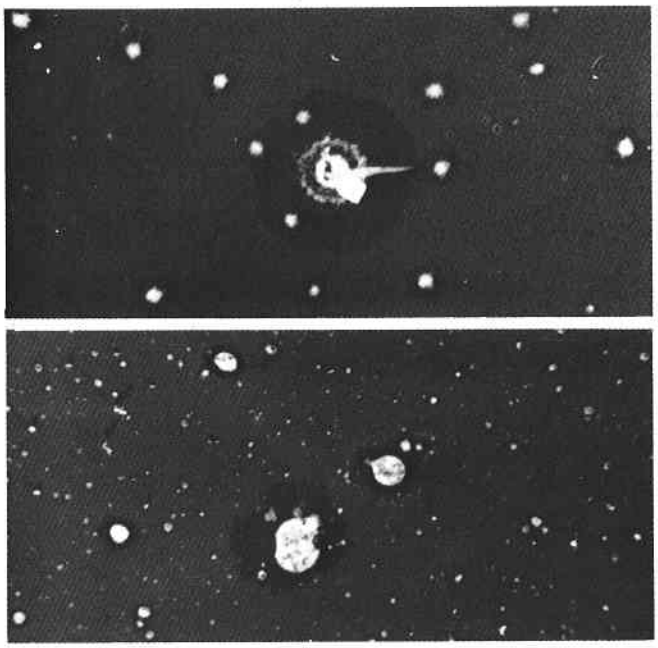
The role of this Division was to find whether atmospheric particles at proposed sites in Tasmania are influenced by local sources or whether they are representative of a wider area of the globe, and then to commence measurements at the sites. No previous comprehensive studies of particle types, size distributions and concentrations as a simultaneous function of altitude and position over a wide area had been made anywhere in the world, and these are vital if we are to interpret the results found at ground sites in their context of global values. The Division therefore carried out over a wide area of Australia a series of measurements in instrumented aircraft; these measurements were made in conjunction with ground measurements with the aim of comparing particle types and concentrations found at any particular ground site with those found in the air over the site, in the air upwind of Tasmania, or in the air anywhere else over Australia.

Flights of two to three weeks' duration were made in June, September and October 1974, and in January, March and November 1975 and ground expeditions were made simultaneously on two of these occasions. Altogether about 100,000 km were flown on tracks shown on the accompanying map. Investigations included measurements of the concentration of the total aerosol, of cloud nuclei and larger particles, and of the chemistry of individual particles and of the aerosol as a whole.

Local pollution must be absent from any baseline station for much of the time or the measurements will have a local rather than a global significance. Now a sensitive measure of pollution or "cleanliness" is the concentration of all particles down to diameters of about 0.001 μm. Such particles are usually detected indirectly: e.g. by humidifying the air sample, cooling it rapidly by a large expansion and counting the water drops that form on the particles. For our measurements we used Pollak's version of such a counter, with slight modification for stable operation in turbulent flight conditions. The measurements of small-particle concentrations show that, away from towns, and in the absence of grass or forest fires, the air over inland Australia is relatively clean by world standards: it is extremely clean in the region of Tasmania proposed for the baseline station. A count of 300 cm^{-3} has been considered a typical minimum value, but in winter in southern Tasmania the count at the ground site may be consistently less than 100 cm^{-3}, "cleaner" than the air above cloud tops. This shows an absence of local surface sources that will greatly aid interpretation of results when the monitoring station is finally set up. In addition we found that the air at ground level usually has almost the same particle concentration as the sub-cloud air upwind



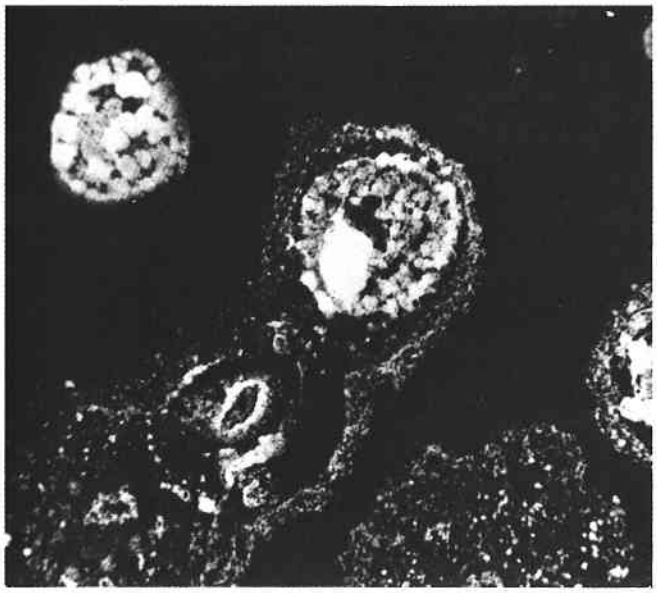
Fraction of occasions on which particle concentrations exceed stated values.



Particles collected over southern Tasmania (top) and continental Australia (bottom), on a thin film of copper.

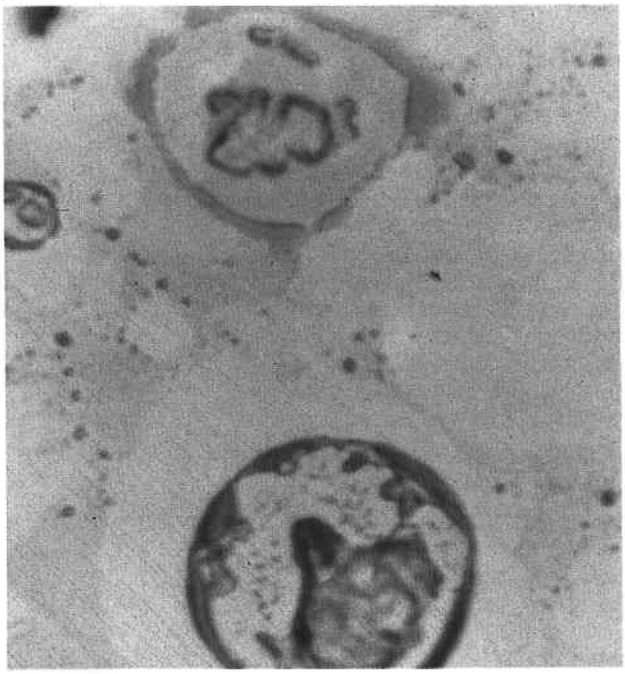
of Tasmania, and from this point of view southern Tasmania will be an outstanding site. The main difficulty in interpretation will be the extent to which local cloud and rain deplete particle concentrations.

Those particles most important to cloud evolution are the so-called cloud nuclei — the particles on which cloud droplets form as the air cools. Usually they are only a small proportion of the total number of particles present, since even hygroscopic particles have to exceed about 0.01 μm diameter in order to promote droplet formation under typical conditions. However, in southern Tasmania the number of cloud nuclei is almost equal to the total number of particles. The reason for this was made clear when the particles were examined: they were all hygroscopic and relatively large, typically being about 0.1 μm diameter. (In continental regions many particles are smaller than this and only some are hygroscopic.) These small particles were collected by electrically charging them and driving them with an electric field on to a special surface pre-coated with a thin film of copper which hygroscopic particles etched when they took up water.



Particles containing sulphate give lacy rings with barium chloride while sodium chloride remains unchanged.

Larger particles, which may also affect cloud lifetime and the formation of precipitation by broadening the cloud droplet spectrum, were also collected. Those with diameters 0.5 to 5 μm were collected by impaction on electron microscope grids, while larger ones were collected on glass slides and examined with an optical microscope. Chemical tests were carried out by the "thin film" method which is described earlier in this booklet. An example of the sulphate test applied to a mixed sea-salt and sulphate aerosol is shown left. The sodium chloride particles are unchanged and appear to be groups of small squares. Those containing sulphate spread out in concentric rings of barium sulphate. Aged sea-salt aerosols appear to have less chloride and more nitrate and sulphate. Sulphate dominates above cloud tops and in particles smaller than about 0.5 μm diameter.



Particles containing sodium give a yellow colour with dipicrylamine; potassium gives a deep red.

When the particles are large enough the same sort of chemical tests can be used with the optical microscope, with the added advantage that colours can be observed in the reaction products. The example shown is important because it shows that sodium and potassium may become separated during particle generation.

As a result of the surveys we now have some idea of where the particles over Australia come from, how long they survive in the atmosphere, how they are distributed with height and position and how their concentration varies. Further measurements are planned to ensure that those already obtained are representative. This work will then form a useful reference in the future for detecting atmospheric changes induced by increasing population and exploitation of resources, as well as fulfilling its primary purpose in helping to interpret results from a monitoring station.

STUDIES OF ICE CRYSTALS IN CLOUDS

Ice crystals form in clouds which grow high enough for their tops to be cooled well below 0 C. The ice crystals then grow at the expense of water drops in the cloud and if conditions are cold enough they fall to the ground as snow or hail. In Australia it is more usual for them to melt on the way down and fall as rain drops. Much of our rain, particularly in winter, starts in this way. This accounts for our interest in ice crystals, their origin, numbers, rate of growth, shape, ability to sweep up water drops, and so on.

When an ice crystal forms inside a cloud it is very small. The ice crystal grows by vapour diffusion until it reaches a size where it can fall and sweep up cloud droplets. With the onset of riming the ice crystal grows much more rapidly. Therefore in order to be able to interpret the shape and sizes of rimed and unrimed ice crystals found in clouds it is important to be able to describe mathematically the vapour diffusion process. In parallel with theoretical studies we have in recent years made measurements of ice crystal growth in artificial clouds in the laboratory.

The Division has also put a great deal of effort into studying the concentration of ice crystals which occur in various types of cloud, since this influences the amount of rain that falls. For efficient rain production we need about one crystal per litre. This can be deduced from the size of raindrops reaching the ground and the total weight of condensed water in a given volume of cloud, on the assumption that each drop starts as an ice crystal. If the crystal concentration is much less than one per litre, then little, if any, rain will result from the ice crystal process (although other mechanisms may prevail). If on the other hand the concentration is much greater, then each crystal will be too small to fall from the cloud and we will get a stable non-precipitating cloud of ice crystals.

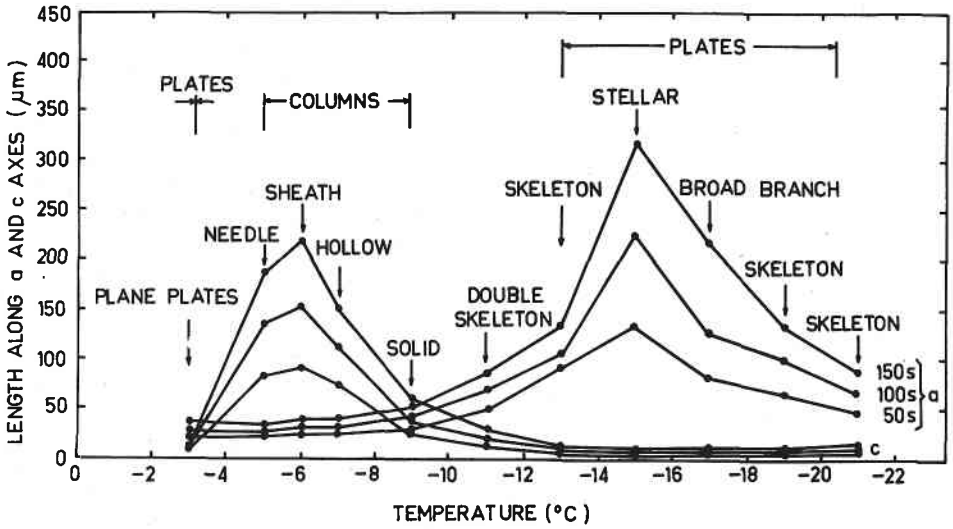
In measuring the concentration of ice crystals in clouds we have sometimes encountered much higher values than current theories would have led us to expect. To explain these anomalous results we have had to set up laboratory experiments to study what happens when ice particles grow by sweeping up supercooled cloud droplets. In this way there has been a constant interplay between laboratory experiments and field studies.

LABORATORY MEASUREMENTS OF ICE CRYSTAL GROWTH

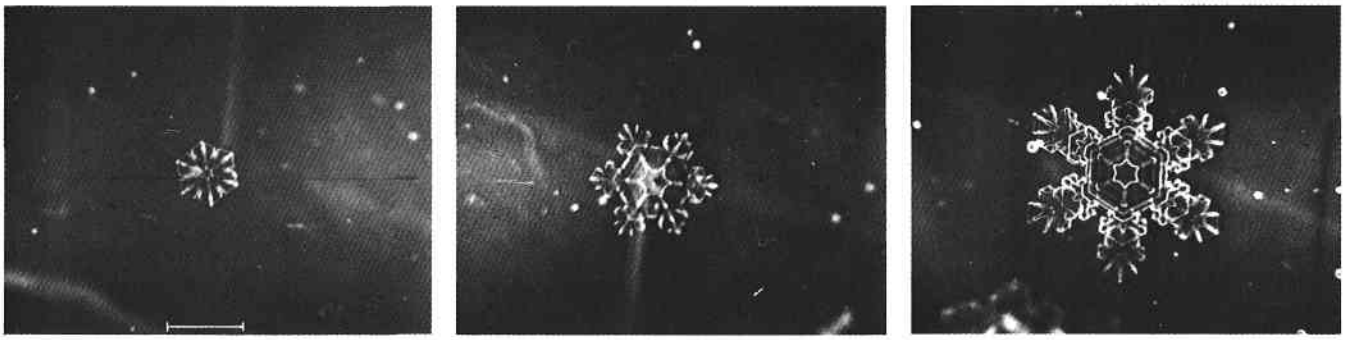
The rate at which ice crystals grow, the shapes they assume and their densities have been simultaneously studied in the laboratory. An artificial cloud of water drops is made by introducing water vapour into a large plastic-walled chamber in a cold room. Because it is desirable to make the parameters controlling the ice crystal growth as realistic as possible the properties of the cloud are carefully monitored. The liquid water content and the number of drops in the cloud are measured to ensure that at all times the water vapour pressure inside the chamber is at least sufficient to produce saturation. The temperature of the chamber is set between -3 and -21 C and is measured at more than one place to ensure that the chamber is very nearly isothermal.

The ice crystals are introduced into the top of the cloud by a suitable seeding technique and grow as they fall through the cloud. In a large cloud chamber, such as the one used, nearly all ice crystals have fallen out at the end of three minutes. At appropriate intervals of time crystals are collected on oil-coated slides, photographed, and then melted to obtain their mass and density.

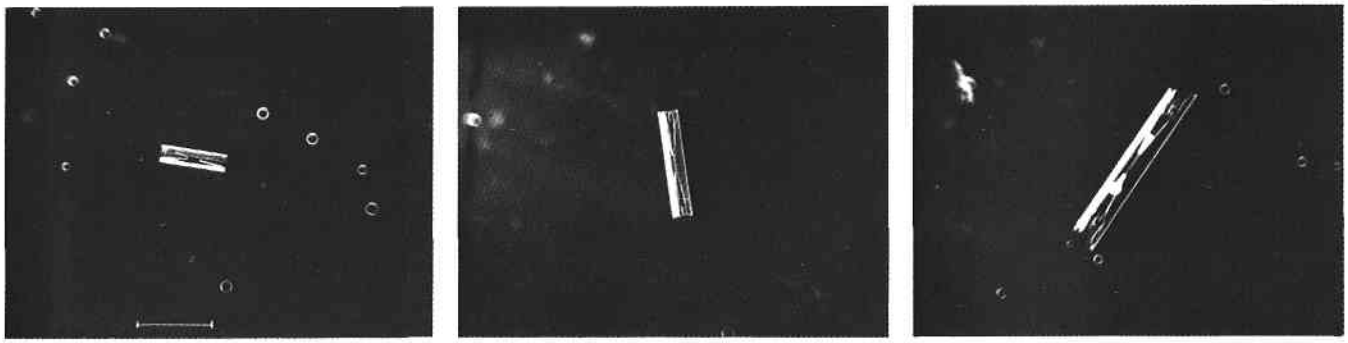
In these early stages of ice crystal growth we find that



Measured values of the length of the main crystal axes of ice crystals growing for 50, 100 and 150 s in a supercooled water cloud.



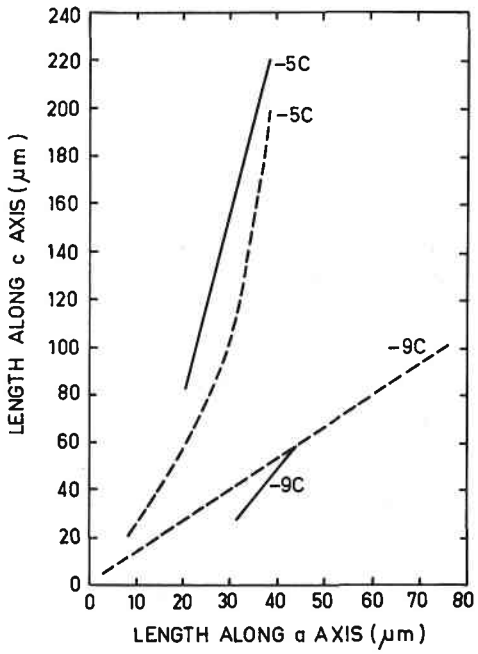
Broad-branched ice crystals grown at -17 C — becoming more elaborate as they grow. The growth times are 50, 100 and 150 s and the scale is 100 μm.



Sheath crystals grown at -6 C. Growth times and scale are as above.

the lengths of the ice crystal axes increase linearly with time. There are wide variations of axial growth rate with temperature, as shown in the figure. The two well-known peaks in growth rate in the respective axial directions lead to the growth of needle-like crystals at about -6 C and very thin plate-like crystals at almost -15 C. Near the latter temperature we find the most decorative shapes and these become more elaborate as the ice crystal grows.

The peaks in the axial growth rates at -6 and -15 C are reflected in the relative masses of the ice crystals at the various temperatures. However, there is a minimum in the density of the ice crystals at -6 C and this is consistent with the mass of ice crystals at -15 C being twice that at -6 C for the same growth time.



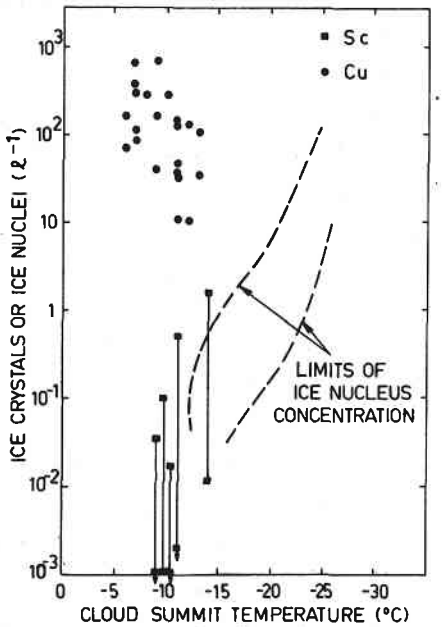
Comparison of estimated crystal axial dimensions (full line) with field measurements at the temperatures indicated for the columnar regime.

In laboratory studies there is a need to ensure that measurements are relevant to atmospheric conditions. Apart from ensuring that the laboratory environment models the atmosphere as closely as is practicable it is necessary to compare the laboratory measurements with similar measurements made in the field. Measurements of ice crystals sampled from our aircraft show that indeed the characteristic ice crystal shapes occur at approximately the same temperature as observed in the laboratory. While there are few reliable growth rates available from field measurements, the axial ratios of ice crystals sampled in the field can be compared with those formed in the laboratory. In the columnar region of the ice crystal growth the agreement is very good, as can be seen from the accompanying figure. At low temperatures, where plate-like crystals form, the crystals formed under laboratory conditions are somewhat thinner than those observed in the field.

The present experimental results make it possible to compare various diffusional models of ice crystal growth. Our observation that the axial growth rates are linear during the first three minutes of growth allows the formulation of a simple parametric model of ice crystal growth that is ideal for inclusion into more complex numerical cloud models. There is a need, however, to extend these studies to times longer than three minutes, but this is not possible with our present techniques. Nevertheless, the time period over which measurements have been made is significant, since the ice crystals have grown to sizes where in a real cloud riming is expected to become important. Once the riming process occurs it will dominate over vapour deposition in determining growth rate.

CONCENTRATION OF ICE CRYSTALS IN CLOUDS

The tiny drops of which clouds are composed can be cooled well below 0 C without freezing. In fact, ice crystals only appear if there are suitable dust particles present in the cloud to assist the freezing process (the "ice nuclei"). The ice nuclei can be counted by making an artificial cloud in a cloud chamber at a low temperature and seeing how many ice crystals fall out. Such measurements have led to the belief that natural ice nuclei are rather rare in clouds at temperatures warmer than -20 C. This apparent rarity of ice nuclei, and consequently of ice crystals, has supported the idea that rainfall can be increased by artificially introducing more ice crystals. Upon this simple concept are based most of the rain-making efforts in Australia and many other countries, where clouds have been "seeded" with ice nuclei such as silver iodide particles.

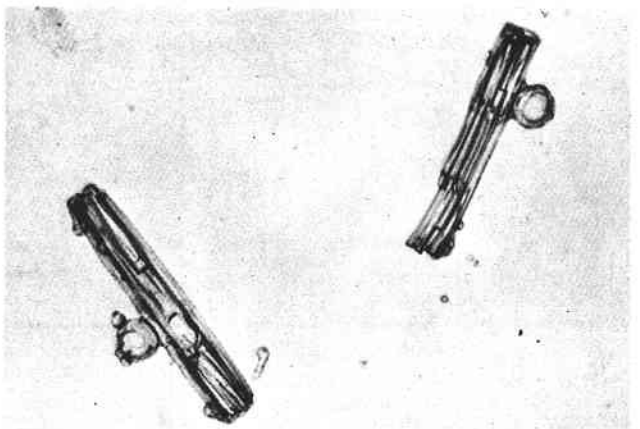


Concentrations of ice crystals and ice nuclei in cumulus and stratocumulus clouds as a function of temperature.

However, over the past 10 years our studies of natural clouds have shown that this simple picture is not always true. An instrument mounted on our DC3 aircraft enables us to make replicas of the water drops and ice crystals in a known volume of cloud. We can then deduce the concentration of ice crystals. In some cumulus clouds, particularly those forming in air that has recently spent a long time over the sea, there are often high concentrations of ice crystals at temperatures of -10 C or warmer. These concentrations can be as much as 10,000 times the ice nucleus concentration. By contrast measurements in shallow layer clouds (strato-cumulus) give concentrations of ice crystals that agree reasonably well with those expected from the ice nucleus measurements.

The cumulus clouds in which these high ice crystal counts are observed also contain appreciable numbers of large rimed ice particles, which are crystals that have grown by colliding with supercooled cloud drops. These "graupel" are absent from the shallow layer clouds and could therefore be thought to play some part in the apparent "multiplication" of ice crystals.

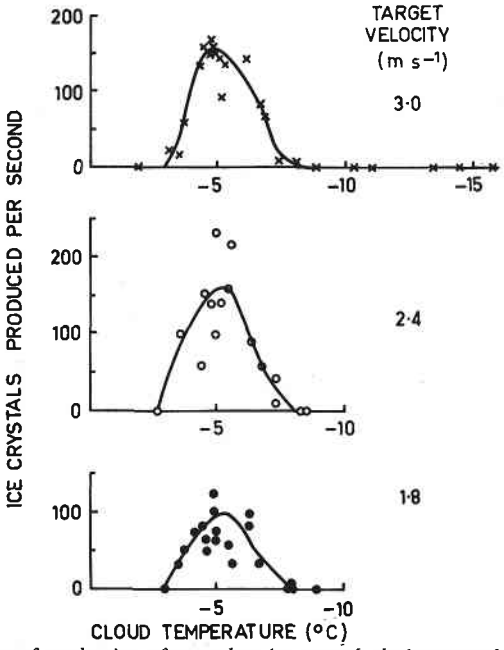
Sampling flights through such clouds near their tops showed that they contained several distinct regions or



Replicas of two hexagonal columnar ice crystals sampled at the -7 C level in a cumulus cloud off Tasmania. They are about 0.4 mm long. They have started to grow by riming, each having captured a large water drop.

"cells". Cloud towers that were still rising consisted almost entirely of water drops. Some of these had grown large enough to be classed as "drizzle drops". In another part of the cloud there would be fewer drops but ice crystals and graupel would be present. A third type of cell was also encountered in some clouds. Here the water drops had almost entirely disappeared and the graupel were fewer (presumably because of fall-out) but the ice crystals had reached very high concentrations, say 100 per litre. These crystals were columnar in shape, indicating that they had formed between temperatures of -3 and -8 C.

These three types of cell are thought to represent a progression in time from the youngest, newly risen water drop region, to the oldest cell, where almost all the drops have evaporated as the ice crystals have grown. There seems to be a process at work by which, over a space of time, the original ice crystals can be multiplied. If the cloud lasts long enough it eventually reaches the stable condition mentioned earlier in which the numerous ice crystals are all too small to fall out.



The rate of production of secondary ice crystals during growth of rime upon a rod 30 cm long and 1.8 mm in diameter moving through a cloud at 3, 2.4 and 1.8 m s⁻¹.

Wind tunnel used in riming experiments.

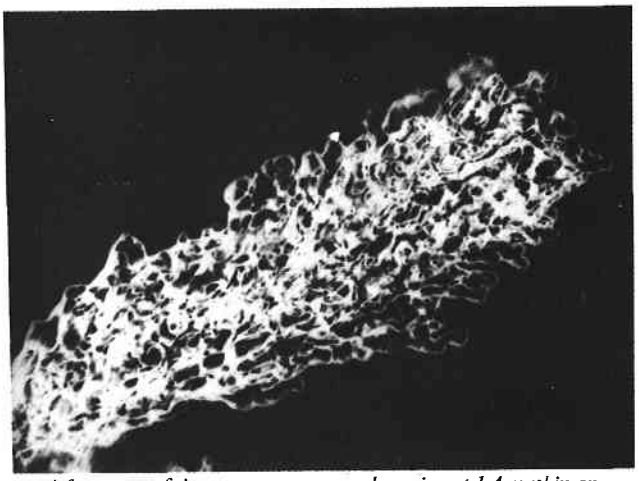
LABORATORY EXPERIMENTS ON ICE CRYSTAL MULTIPLICATION

The evidence that graupel particles appear to be connected in some way with the multiplication process supports the suggestion, first made about 30 years ago, that ice "splinters" may be thrown off when a graupel particle sweeps up supercooled drops which then freeze. Until recently however laboratory experiments failed to confirm this theory.

The cloud conditions under which splintering can take place were eventually established by experiments in this laboratory over the last few years. In these experiments the growth of ice crystals by sweeping up supercooled drops was imitated. This riming growth took place upon a metal rod moving through an artificial cloud of supercooled drops in a laboratory cold-room. If the cloud temperature was about -5 C it was found that copious ice splinters were thrown off during the riming process. As well as being dependent on cloud temperature it seems that this process needs the presence of moderately large drops: splinter production was found to be related to the number of drops of diameter about 25 μm and larger that were swept up.

In clouds where the necessary large drops exist, each ice crystal that grows by sweeping up such drops will produce a certain number of secondary ice particles. Each of these can then grow, rime and multiply in its turn. In this way it seems possible for the original crystals formed upon ice nuclei to increase in number by several orders of magnitude within a reasonable cloud lifetime.

Studies are still in progress to determine the exact physical mechanism by which the secondary ice particles are produced. The splinters themselves give little indication as to their origin because by the time they can be collected for examination they have already grown into regular symmetrical ice crystals.



A fragment of rime grown upon a rod moving at 1.4 m s^{-1} in an artificial cloud at -5 C. The latest growth is at the right.

The structure of the rime is being examined in case there are peculiarities that are unique to the -5 C temperature region. So far no evidence has been found of any frail needle-like growth that could perhaps be broken off by the impact of the larger cloud drops. As shown in the photograph, the rime structure consists of frozen drops that have partially merged into one another.

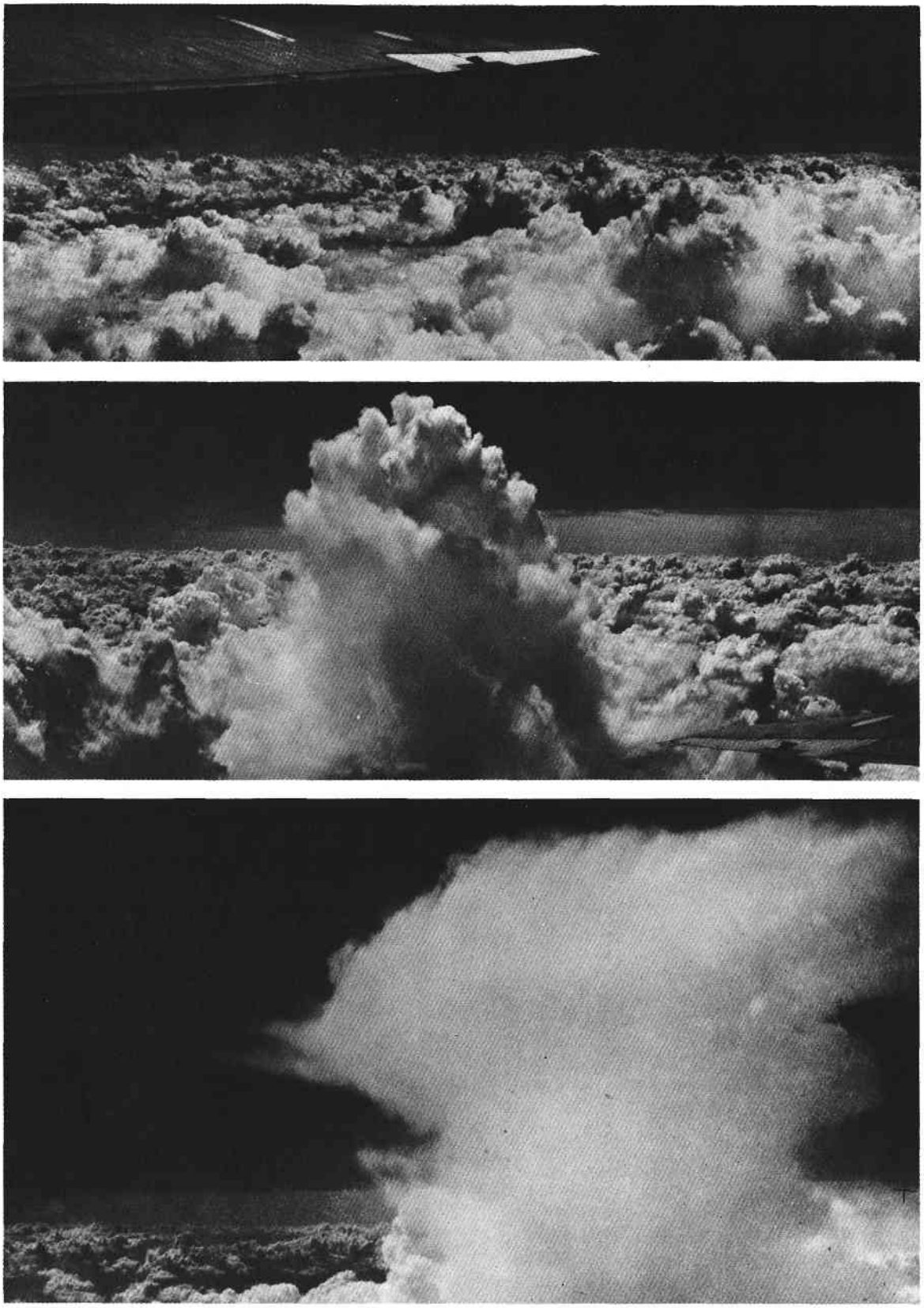
IMPLICATIONS FOR CLOUD SEEDING

While the physical mechanism of splintering is not yet understood, its possible implications for artificial rain-making are being explored. Clouds in which abundant ice crystals are present because of splintering are unlikely to yield more rain when seeded with artificial ice nuclei. This situation is likely to prevail in cumulus clouds near the coast, where the clouds usually contain suitably large drops at the -5 C level. Fortunately such geographic locations seldom require more rain.

Conversely, artificial stimulation is likely to be more profitable in clouds where drops larger than 25 μm only form well above the critical -5 C level. For this to be so, the clouds must contain high concentrations of drops so that very few can grow to large size. Such clouds form in "continental" air that has spent a long time over land and contains many particles capable of acting as cloud condensation nuclei. Continental cumuli which contain about 1000 drops per cubic centimetre and have cloud bases above the +10 C level appear to offer good chances of success in attempts to increase rain by seeding with ice nuclei.

In the Tasmanian cloud-seeding experiment (described later) the rainfall increase appeared to be greater when the seeded clouds were stratiform rather than cumuliform. This may indicate that in stratiform clouds in which there is a lack of big drops, the multiplication process is comparatively unimportant.

It is interesting that the number of cloud condensation nuclei should play such an important part in precipitation development. It obviously governs the growth of the drops themselves, but we now believe that it also has great influence on the number of ice crystals that form in supercooled clouds.



One of the first successful cloud-seeding experiments in which rain was induced to fall to the ground was performed near Bathurst, N.S.W. on 5 February 1947. The photographs show, from the top down, the appearance of the clouds before seeding and after 13 and 45 min.

CLOUD SEEDING

INTRODUCTION

Clouds form if two conditions are fulfilled: the air must contain sufficient water vapour and this moist air must be cooled to the point where the water vapour condenses. Cooling usually occurs when the air is lifted, which requires a supply of energy, coming originally from the sun. The water, which primarily comes from the sea, and the energy from the sun are both in quantities so large that we cannot influence them to any appreciable extent. Hence we cannot exert much influence on the formation of clouds. We can however attempt to influence clouds already formed.

Even if clouds are present, rain only forms if various physical conditions are met; sometimes one of these requirements may be absent, and hence if we can supply this missing quantity we may be able to influence the rainfall. This concept (in the dry Australian environment) provides the stimulus for a continual programme of research by the Division of Cloud Physics.

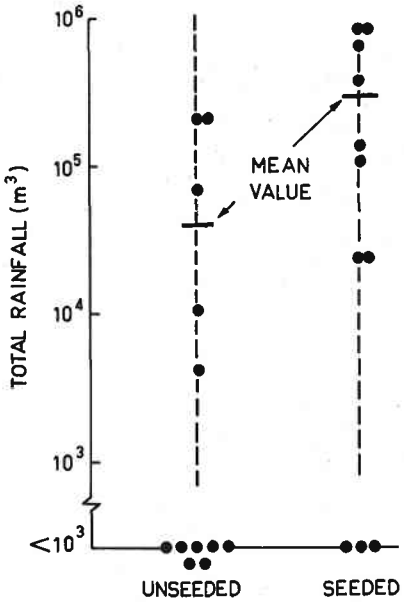
Rain can form by two processes, each depending on the presence of special aerosol particles, or nuclei, and it is through the supply of these nuclei that we generally attempt to influence rainfall.

The first rain process we will consider is that which occurs in warm clouds. If there are plenty of active cloud condensation nuclei, clouds contain large numbers of very small droplets, a million or more of which would be needed to form one typical raindrop. These droplets all tend to grow to the same size, and hence to fall at the same rate, thus reducing the chance that raindrops will grow by collision and coalescence. It might be possible to stimulate coalescence by introducing into such a cloud a relatively small number either of droplets somewhat larger than the others or of hygroscopic particles on which large droplets could form.

When clouds rise to levels where the water droplets are supercooled a second process of rain formation can operate. If active ice nuclei are present ice crystals will form which grow rapidly by sublimation and subsequently by collision and coalescence with other ice crystals or water droplets in their path as they fall. They may subsequently melt to produce useful rain. If ice nuclei are absent, or if there are not enough of them, it is possible that no rain will fall and the clouds will just evaporate. However, if the clouds are suitable in all other respects the rain process can often be started by "seeding" them with ice crystals. This can be done by the introduction of either substances which can act as ice nuclei, such as silver iodide, or very cold substances, such as dry ice.

Stimulation of the coalescence process is promising in principle, but so far it has received comparatively little attention either here or overseas because it presents many practical difficulties. Stimulation of the ice crystal process appears to be even more promising in principle, and much easier to apply and evaluate in practice. Cloud-seeding research in Australia has therefore been concentrated on the latter process.

The first cloud-seeding experiments in Australia were performed on isolated cumulus clouds. Such clouds were chosen for experiment because they are of moderate size and have a reasonable lifetime so that any stimulus can be conveniently related to subsequent developments. Dry ice was dropped into the tops of supercooled cumuli from RAAF aircraft, a procedure which resulted in extra rain when conditions were suitable. However, this technique involved carrying large quantities of dry ice to high altitudes and this necessitated the use of large, expensive aircraft. It was found to be much cheaper to use other artificial ice nuclei, such as those resulting from the production of silver iodide smoke. With this material a much smaller aircraft was satisfactory and various



Total rainfall from isolated cumulus clouds.

delivery techniques were available, for example releasing the smoke into updraughts at cloud base, or dropping pyrotechnics containing silver-iodide into cloud tops. In Australia these techniques have been successfully used in many well-designed experiments where a series of clouds have been selected as complying with a specification, and only half of them, chosen by a random process, have been seeded, the remainder serving as controls. On the average, several times more rain fell from the seeded clouds, and the random selection gives confidence that the rainfall increase was due to the seeding.

EXPERIMENTS OVER LARGE AREAS

Not all clouds which are capable of producing rain are suitable for seeding. Further, there is always the possibility that seeding one cloud of a group and causing it to rain more heavily may affect the rain from its neighbours. Thus, even if cloud seeding can increase the rain from a single cloud, it does not necessarily follow that an economically useful increase can be stimulated in the rain over an area of country: this is something that must be demonstrated separately.

A simple experiment to provide such a demonstration could take the following form. Suppose that time is divided into periods (say a week) and two areas of country

(say a few thousand square kilometres) are chosen, in both of which the rainfall is measured. Suppose further that clouds over one of the two areas are seeded in any one period while the other area is used as a control. Then if the seeding is successful we would expect to find that over a long enough period the rainfall balance between the two areas should be altered in favour of the one which was seeded.

Four experiments of this general type were carried out in Australia between 1955 and 1963. In retrospect we now realize that their design was too simple to enable reliable results to be obtained. Indeed the extreme variability of natural rainfall makes it difficult even in a very sophisticated experiment to detect changes unless they are very large or protracted. However, these early experiments suggested that on the western slopes of the Great Dividing Range it should be possible to increase rainfall by cloud seeding. Rather unexpectedly it appeared that the result of seeding was to increase rain on some occasions but to decrease it on others. While these early experiments were not sensitive enough to define the conditions required for success it appeared that seeding was more effective with cumulus clouds forming in continental air masses than with those forming in air of recent maritime origin.

TASMANIAN EXPERIMENT

The lessons learned in these early cloud-seeding experiments over large areas were incorporated in an experiment in Tasmania, where it is relatively wet but where still more rain is required for a hydro-electric catchment scheme. Improvements in the design were intended to make it possible to detect unambiguously the results of seeding even if they varied with meteorological conditions, and to determine the conditions in which the best results were achieved. Time was divided into periods of about two weeks' duration and seeding took place in half of them, selected at random. Rainfall was measured in the target and in three (unseeded) control areas. Observations of clouds and other meteorological quantities were made during both the seeded and unseeded periods so that rainfalls on seeded days of a given type could be compared with falls on unseeded days of the same type.

The first stage of this experiment was conducted between 1964 and 1970 and yielded very satisfactory results. In spring and summer there is no clear evidence that seeding affected the mean rainfall, but in autumn and winter there was a mean rainfall increase, estimated as about 15% in the target area, at a satisfactory significance level. Many aspects of the data provide confirmatory evidence so that we can confidently accept this increase as being the result of the seeding.

A second stage of this experiment, intended to be of four years' duration, was started in 1971, with the aim of confirming that results similar to those of the first experimental stage could be achieved. The cloud seeding was then carried out by the Hydro-electric Commission of Tasmania with the Division supplying advice as required. In 1971 the results again showed substantial rainfall increases in autumn and winter, just as they had in the first experiment. However, seeding was suspended (by the HEC) when record rainfalls caused the reservoirs to overflow and further rain would have been detrimental. The experiment has not been resumed.

This experiment has provided a mass of data from which it should be possible to extract many valuable conclusions about the detailed effects of seeding. In order to make the best use of this data and derive reliable conclusions it has been necessary to develop new statistical methods; the Division has been fortunate to obtain co-operation in this matter from CSIRO's Division of Mathematics and Statistics.

The detailed analysis is not yet complete but results obtained so far suggest that:

(a) There was an increase in mean rainfall in autumn and winter, as concluded from the earlier, simpler analysis.

(b) In these seasons seeding was generally most successful in dry or moderately wet periods. In the wettest periods there is no evidence that rain was increased; indeed there is a suspicion that in some circumstances it was decreased.

(c) In Tasmania's predominantly maritime air masses, seeding stratiform clouds increased rainfall: seeding cumuliform clouds did not.

(d) There is some indication that in springtime, although seeding caused no change in mean rainfall, it may have caused increases in dry periods and decreases in wet ones.

These results, and others which will probably emerge shortly, should enable adjustments to be made to the seeding technique which will make it even more effective.

PLANNING OF AREA EXPERIMENTS

From overseas as well as Australian experience it is becoming increasingly clear that before an area cloud-seeding experiment is started there must be much more preparation than there has been in the past. Such preliminary work must aim at establishing that:

(a) There is reason to believe that the rain in the area can be increased — i.e. that clouds suitable for seeding are of common occurrence.

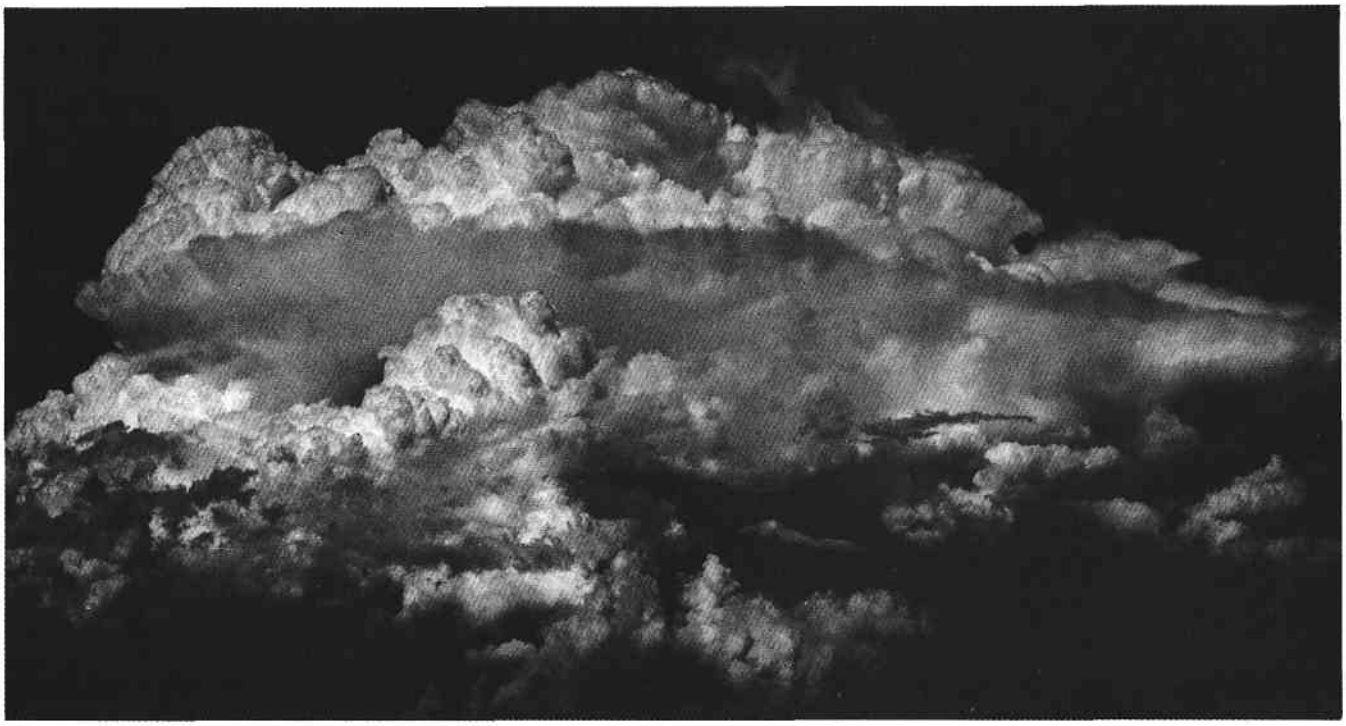
(b) There is reason to believe that if rain can be increased, the change can be detected with acceptable certainty by known physical and statistical techniques in an experiment of reasonable duration.

It is now considered that this preparatory work should occupy two or more years of research effort, most of which would be into the cloud processes whereby natural rainfalls are produced. Thus, while some exploratory cloud seeding may occur, a great deal of research would be aimed at invesigating the microphysics of the clouds in the area. Such microphysical investigations would include studies of the cloud droplet size distribution and how much of the cloud contained supercooled water, as well as studies of natural ice nuclei and ice crystals within the clouds. Further studies would be made of the variability of the natural rainfall and of the appropriate statistical designs for detecting increases which might occur should a cloud-seeding experiment be started.

Two areas, in Queensland and Victoria, have been studied along these lines to see if they are suitable for cloud-seeding experiments.

QUEENSLAND

Large cumulus clouds, apparently suitable for seeding, have frequently been observed in Queensland in summer. These were studied in 1972-1975 in an area around



A large cumulo-nimbus cloud in Queensland.

Emerald, where extra rain would be valuable for irrigation and mining.

In a series of experiments isolated cumulus clouds were seeded, producing almost immediate conversion of the cloud tops from water to ice with an obvious change in appearance. The rain which followed seeding was sometimes greater and sometimes less than expected on the basis of that which fell from neighbouring unseeeded clouds. A substantial research effort would be required to isolate the meteorological conditions associated with these apparent changes in rainfall. It was also found during the cloud study that only a small proportion of the rain of this area falls from single isolated clouds whereas heavy rain falls when several clouds merge to form a huge cloud mass. Overseas experiments suggest that there is some reason to hope that seeding can influence the chance of clouds merging, but to get definite evidence on this point would require many years' research. In spite of these problems it seems possible that with sufficient effort an effective means of cloud seeding could be developed to produce extra rain in this area. However, the problem of detecting extra rain on the ground is complicated by the extreme variability of rainfall in the area studied. Intense, widely separated showers fall from isolated cumulus clouds or cloud masses, a situation which makes for difficulties in the measurement of mean area rainfall and necessitates the use of large numbers of rain gauges. A simple ground radar was installed to see whether it could provide sufficient information to provide an adequate supplement to an existing sparse network of rain gauges. While there are still possibilities that a more sophisticated radar might prove successful the results obtained with the simple equipment were not promising.

As a result of these preliminary experiments it was concluded that while opportunities for cloud seeding undoubtedly exist in the area a controlled experiment would need to be of very many years' duration if it were to yield a reliable answer. Such an experiment is beyond the resources of the Division.

VICTORIA

In the wheat-growing areas of western Victoria the winter rains are usually adequate but the crop yield is influenced by spring rain, which is often insufficient. Therefore, the Division is investigating this area to see if it is suitable for a cloud-seeding experiment.

Measurements are being made in the clouds to see if they fulfil the physical conditions required for a successful seeding experiment. Results obtained so far suggest that on many days in the spring of 1975 clouds already contained adequate numbers of ice crystals for rain formation and were therefore unsuitable for seeding. On other days however stratiform clouds in the area contained abundant supercooled water and comparatively few natural ice crystals and appeared to be in all respects suitable for seeding. These stratiform clouds were similar to those successfully seeded in Tasmania except that they were less turbulent. Further study is needed before we will know how often these suitable conditions occur but at this stage the area appears to be a very suitable one.

A statistical study of the variability of the natural rainfall, as shown in the historical rainfall records of the area, is also proceeding to see if any extra rain which might be stimulated could be detected in an experiment of reasonable length. Initial indications are again favourable: the spatial variability of rainfall is acceptable, the facilities for measuring it are good, and assessment of results could be assisted by the use of other meteorological measurements (e.g. by radiosondes) which are available. The relative merits of various experimental designs for this area are being investigated.

Further study should determine whether a major cloud-seeding experiment in this area would be justified.

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- G. K. Fowler
- Elizabeth A. Monticone

Front row, left to right: *Elizabeth A. Monticone, Judith A. Williams, Mary E. Brauer, Diana du Cros, Enid F. Turton, Judith P. Hawkes.* Second row, left to right: *S. C. Mossop, D. B. Drewitt-Smith, M. A. Parker, J. Warner, E. J. Smith, A. A. Tapp, Lorraine E. Wakefield.* Third row, left to right: *R. T. Meade, Jenny L. Clarke, C. T. Maher, D. E. Turvey, E. R. Wishart, W. D. Scott, C. G. Michael, Barbara A. Georgeson.* Fourth row, left to right: *B. F. Ryan, J. Meadows, R. E. Cottis, J. L. Gras, G. P. Ayers, P. J. Bosci, C. E. Coulman, E. K. Bigg.* Fifth row, left to right: *W. D. King, R. L. Hobbs, M. D. Thompson, R. N. Phillips, D. C. Lain, B. M. Bartlett, M. J. Manton, J. C. Carstens.* Back row, left to right: *G. K. Fowler, K. J. Seton.*
Absent: *S. Twomey, R. J. Handsworth, N. S. C. Thorndike, Ruth M. Hodge.*