RESEARCH ACTIVITIES CSIRO DIVISION OF CLOUD PHYSICS

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FOREWORD

Nearly all man's activities are influenced to a greater or lesser degree by the weather and on this driest of continents weather and rainfall are vital everyday concerns. A number of Divisions in the Commonwealth Scientific and Industrial Research Organization are engaged in research aimed at elucidating those factors which influence the weather. The Division of Cloud Physics is concerned primarily with small-scale processes the microphysics of clouds — which in a given meteorological situation may determine whether or not clouds will produce rain. It is through deliberate modification of these processes that man has the greatest hope of being able to influence the weather to his advantage; by the same token man's everyday activities, both agricultural and industrial, produce atmospheric pollutants which may have deleterious effects on the weather through their effect on cloud microphysics. If such

adverse effects are to be avoided their cause must be thoroughly understood.

The Division of Cloud Physics was formed in 1972 from a group which, as part of the Division of Radiophysics, had been engaged in a study of rain processes and the means of influencing them for 25 years. The Division now forms part of the Environmental Physics Research Laboratories; its sister Divisions are those of Atmospheric Physics in Melbourne and Environmental Mechanics in Canberra; it is located in the Sydney suburb of Epping some 16 km north-west from the city centre; it shares a building complex and many common services with the Division of Radiophysics. 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.

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CLOUD PHYSICS

Clouds form as the result of the condensation of water vapour upon tiny hygroscopic particles which are always present in the earth's atmosphere in concentrations which vary from place to place and time to time. The cloud droplets are initially small and must increase in mass some millions of times if they are to be large enough to fall to the earth's surface as rain. Their growth may result from the collision and coalescence of many single droplets or it may be accelerated when an ice crystal forms high up in a cloud where it grows rapidly before falling and collecting other ice crystals or water droplets in its path.

In some clouds rain may result solely from the coalescence of water droplets: this is most common in maritime situations, where comparatively small clouds often produce copious rainfall. In other clouds ice crystal growth is a necessary prelude to the production of significant precipitation: such clouds are typical of inland areas. In very large clouds, wherever they are formed, ice crystal growth is important, but the significance of its role can only be determined by detailed observation. Most of the attempts to increase rainfall or to modify cloud behaviour, particularly in this country, depend upon influencing the role of ice crystals in the precipitation process, since ice crystals form the most effective trigger and can be affected more readily than the water droplets which form the bulk of the cloud.

Initial cloud formation is, of course, dependent upon the presence of enough water vapour in the atmosphere and sufficient lifting or cooling of the air mass for condensation to occur; these factors are determined by orography and by the large-scale synoptic processes such as the movements of high and low pressure systems and the presence of fronts. Rainbearing clouds take different forms, ranging from the deep layer clouds which produce persistent steady falls of moderate intensity over wide areas to convective clouds which in the final stages of their growth may become towering thunderstorms and produce precipitation which is very heavy but of relatively brief duration and covering only a limited area. Because they are only of moderate size and are accessible to investigation by aircraft most emphasis has been given to a study of convective clouds. Dynamically these clouds are complicated, the rain process interacting strongly with the updrafts or downdrafts in the cloud and mixing between the cloud and its clear-air environment serving as a brake on cloud growth.

Recent research in cloud physics at the Division has centred on the following topics: the size, nature and origin of the nuclei of condensation; the size distribution of the droplets formed by condensation and its relation to the spectrum of nuclei; the role of the turbulent updraft in a convective cloud in droplet growth and cloud behaviour; the size and origin of the nuclei which stimulate freezing and upon which ice crystals grow; the ice crystal form and concentration in natural clouds and its relationship to the ice nucleus spectrum. Allied to these studies has been an investigation of the buoyant thermals which transport heat and vapour upwards from the earth's surface and result in cloud formation.

While some of this research work can be performed in the laboratory most of it must be done in the free atmosphere in natural clouds and their surroundings; the use of an aircraft carrying a wide variety of specially constructed instruments has proved invaluable in these investigations.

The Macrophysical Properties of Clouds

CLOUD FORMATION

To form and maintain a field of clouds moisture must be supplied at the condensation level; turbulent, upward transport is largely responsible for the formation of cumulus clouds and so considerable effort has been devoted to the study of convection in clear air below cloud base.

We have found that the upward fluxes of heat and moisture are carried by plume-like structures having typical horizontal dimensions of 200 to 500 m and heights up to perhaps 1 km over level land. These structures are recognizable from the temperature fluctuation records obtained by an aircraft in level flight; the upward-moving, turbulent air in the plumes exhibits a rapidly varying temperature which exceeds the steady temperature of the downdraft regions between plumes by as much as 0.5 to 1°C. Although identifiable from their temperature characteristics alone at heights exceeding about 50 m, these convective updrafts nevertheless show evidence of inflow of air at just a few metres above the ground.

The vertical eddy fluxes of heat, moisture and momentum have been calculated from measurements of temperature, humidity and velocity made many times per second during the flight of our instrumented aircraft. Convection develops in cool air streams moving over warm seas, although the detailed structure is not as readily observed as over land. The upward heat flux is usually less, because air-sea temperature differences are normally much smaller than the temperature differences which develop between land surfaces and the air during the daytime.



Eddy fluxes of water vapour measured over tropical sea compared with values calculated from air-sea temperature difference and wind speed.

Flux measurements in convective conditions over the tropical ocean have provided basic data for the study of the persistent cloud field known as trade wind cumulus. Additionally, our measurements confirm that for some applications the simpler, bulk aerodynamic method, based on averaged wind speed and air-sea temperature difference, is sufficiently accurate.

To facilitate the measurement of rapidly varying atmospheric properties, computer-controlled magnetictape recording equipment has recently been installed and successfully used in the Division's research aircraft. The records produced during field operations are suitable for immediate processing by the C.S.I.R.O. computing network.

CLOUD DYNAMICS

Casual observation of a cloud usually suggests a complicated field of motion in three dimensions; our study of cloud dynamics aims to investigate the general characteristics of such motions and to reveal their influence on the internal properties and overall behaviour of clouds.

One of the most important and well-established observations made in cumulus clouds by means of instrumented aircraft concerns their liquid water content. We find that almost everywhere in the cloud there is appreciably less liquid water than would result from the lifting of saturated air from cloud base under adiabatic conditions (i.e. without loss or gain in heat content). Whilst this is now accepted as evidence of mixing between the cloud and the drier, surrounding clear air the details of the process are still not fully understood.

However, some progress has been made through aircraft measurements of the rapidly fluctuating speed and direction of the air flow in cumulus clouds which are entirely warmer than the freezing point of water. In the vicinity of cloud base we generally observe a chaotic velocity structure, whilst in the main body of the cloud a general updraft region exists. Nearer to cloud top we usually find a strongly turbulent zone. There is also some evidence for a more or less well-organized downdraft around the cloud and close to the cloud edge.

The scales of motion in this type of cloud are indicated by the form of the spectrum of vertical velocity computed from aircraft measurements. Our observations suggest that near to cloud top and cloud base there is rather more energy in scales of the order of 100 to 200 m wavelength than in the body of cloud, where larger scales account for proportionately more energy. The severity of the in-cloud turbulence, as specified by root-mean-square vertical velocity, is found to depend quite strongly on the static stability of the cloud environment: a strongly stable environmental temperature gradient is associated with less turbulent conditions in cloud.



Comparatively shallow layers of air in which the upward increase in temperature is sufficiently steep to ensure static stability are frequently observed in the atmosphere. Such inversion layers often limit the growth of cumulus cloud tops and define the upper surfaces of



Air velocity measurements in cumulus clouds. The arrows show the magnitude and direction of air motion to the same scale in each diagram. stratiform clouds. Whether such clouds continue to grow, precipitate or disperse is probably determined to some degree by the extent of mixing with drier air in and above such inversion layers. A study of the mechanisms by which such stable layers are penetrated or eroded by the effects of cloud and clear air convection has recently been commenced with measurements of the velocity fields in the vicinity of temperature inversions.

CLOUD MODELS

Computer simulations of cloud behaviour, or cloud models, are intended to provide numerical predictions of the development of cloud properties with time, given certain initial data. Models of various degrees of complexity are currently being developed at institutions around the world; their success is judged by the extent to which the calculated results agree with observations.

Numerical models appear to be the best means at present available of including cloud effects in global atmospheric forecasting systems. Furthermore, comparison of the predictions of a reliable cloud model with observations on clouds which have been deliberately modified by cloud seeding, or unintentionally modified by pollutants, will ultimately provide a means of assessing the significance of such intervention with natural processes.

The simplest models deal with the variation of atmospheric properties in one dimension — the vertical. We have found that the predictions from such models of the growth and internal properties (such as liquid water content) of a cloud are not in agreement with our aircraft observations. More elaborate, three-dimensional models are also being developed by workers overseas and carefully planned observations are needed for critically testing them. Our own observational program is expected to play an important part in providing the data required.

Condensation Nuclei and Cloud Droplet Formation

Since the work of John Aitken in the 1880s it has been known that condensation in the atmosphere always takes place upon small particles that are to be found floating freely at all levels. However, not all particles are equally active as condensation nuclei, and it is only a very small fraction of the total aerosol mass which normally plays any role in cloud formation. The particles upon which condensation first takes place as the air cools are the soluble ones and the number activated depends upon their size spectrum, their chemical composition and the cooling rate.

Attention was drawn to the importance of the condensation nuclei in rain formation some 15 years ago by a member of this group, who suggested that the main difference between maritime clouds, which readily produce rain, and continental clouds, which do not, lies in differences in their cloud droplet concentrations resulting from differences in the condensation nucleus spectra in the two air masses. The physical reasoning behind the suggestion lies in the fact that if there are large numbers of active condensation nuclei sharing the available water vapour much smaller cloud drops result than when the concentration of nuclei is small; very small drops do not collide with their neighbours as readily as do larger ones and hence do not grow to



A comparison between the average number of droplets observed in cloud and that calculated from measurements of the cloud nucleus spectrum. The circles and crosses show respectively conditions upwind and downwind of a strong source of pollution.

produce rain. When this suggestion was first made means were not available to measure the spectrum of condensation nuclei, since those of interest become active at relative humidities only just above 100% and these humidities are difficult to produce in a controlled manner in the laboratory. However, when we were able to develop equipment it was adequately demonstrated that the cloud droplet spectrum depended directly upon the nucleus spectrum: clouds formed in air masses rich in nuclei consist of higher concentrations of smaller drops than those formed in cleaner air. Indeed, smokepolluted air contains tremendous numbers of active



Equipment used for detecting cloud nuclei. The bright streaks of light are from small droplets falling through an intense beam of light which passes between two wet surfaces maintained at a small temperature difference.

nuclei and the clouds which form in this air are unlikely to produce rain. Nevertheless, while smoke from bush or agricultural fires or from industry produces high concentrations of nuclei in nearby areas, on a global scale man's activities appear to add comparatively little in a direct way to the general background of active condensation nuclei. This is a question however which needs a great deal more attention because of its potential importance.

Cloud nuclei are detected in the laboratory by subjecting a sample of air to a small controlled supersaturation produced in the space between two wet surfaces which are maintained at different temperatures. When the bottom surface is cooled below the temperature of the upper surface heat and vapour diffuse downwards and, owing to the non-linear way in which the saturation vapour pressure of water varies with temperature, the air in the centre of the chamber becomes supersaturated, with droplets forming on any nuclei present. The degree of supersaturation is a function of the temperature difference; this is varied to measure the relation between the number of nuclei that are activated and the supersaturation. This relationship enables us to calculate the number of droplets which would form when air containing this population of nuclei is cooled at a given rate. We can then compare the prediction with the number of drops which are observed in a cloud formed in a known updraft which produces a calculable cooling rate.



Schematic diagram of equipment used for evaporating cloud nuclei to obtain a guide to their chemical composition.

In a laboratory experiment we passed air containing these nuclei through a strongly heated silica tube, cooled it and then tested its nucleus content: we found that when the tube was heated above about 200°C very few nuclei survived the passage—they had been evaporated. From these and other tests we are now confident that most of the active cloud nuclei are small particles of ammonium sulphate, not sodium chloride originating from the sea surface as was originally thought.

When air containing the nuclei passes slowly through small tubes of length many times their diameter some of the nuclei will diffuse to the walls. The fraction which diffuse depends upon their size, the flow rate and the tube dimensions. We have deduced from these measurements that cloud nuclei are typically only a few millionths of a centimetre in radius and thus are only 10^{-17} to 10^{-16} g in mass. It is estimated that particles



Fraction of nuclei that survive passage through very finebored tubes as a function of air flow and nucleus size.

of ammonium sulphate as small as this would have a lifetime of about a day in the atmosphere, since they tend to evaporate. Since it is also known that these cloud nuclei are found over the open sea thousands of kilometres from land, and since it seems unlikely that the sea could provide a source of ammonium sulphate, it is concluded that the nuclei are formed in situ from gases widely dispersed and commonly present in the atmosphere. The gases involved include SO_2 , H_2S and NH_3 ; these are likely to originate primarily from the land and are mainly of natural origin from volcanic activity or from decaying vegetable matter; however, significant quantities of SO_2 do result from industrial processes.

There is an important aspect of the condensation nucleus population which has only recently received attention. In addition to producing changes in the rain process, increases in droplet concentration arising from increases in nuclei result in a cloud which scatters more sunlight back into space. It is well known that a cloud which appears brilliantly white contains greater concentrations of smaller droplets than one which appears dark in colour. Thus on a global scale increasing pollution



The albedo of clouds of different thickness as a function of their cloud droplet concentration.

can, through its effect on cloud droplet concentration, result in less of the sun's energy reaching the earth. Calculations we have made indicate that this effect is particularly important for relatively thin clouds. For a cloud 500 m thick the albedo (i.e. the fraction of incident energy that is reflected) increases from 0.6 to nearly 0.9 if the droplet concentration increases from 10 to 1000 cm⁻³, a change which can well result from severe pollution.

Because of the potential importance of these nuclei in affecting climate, and a desire to know whether man's activities were producing marked changes, we initiated a long-term study in 1969 at Robertson in southern New South Wales. Hourly observations have now been made on most days for over four years of the concentration of nuclei active at a supersaturation of 0.75%. Large



Typical diurnal variation of cloud nucleus concentration at an isolated field station.

changes in concentration of nuclei have been observed throughout the year and from year to year but no consistent pattern has yet emerged, except in one regard: there is a strong diurnal variation with a minimum concentration in the early morning and a maximum in the late evening; its cause is, as yet, unknown.

The Growth of Cloud Droplets by Condensation and Coalescence

To date most measurements of cloud droplet size distribution have depended upon collecting samples of the droplets in some way and subsequently sizing and counting them under a microscope. Our technique has been to impact the drops on to small sooted glass slides exposed for a brief period to the cloud through which the aircraft passes. The drops spread on impact and then evaporate leaving clear impressions in the thin carbon film. The size of the impression depends upon the droplet size and the speed with which it hits, and for our conditions it is typically three or four times as large as the drop. Each slide samples only a small volume of cloudy air (about 10 cm³) and a large number of samples taken at different levels in a cloud are necessary to build up a picture of the importance of different factors in affecting cloud droplet growth. The device used employs magazines, each containing 18 slides; these can be exposed at intervals of a second or more as required, by repeated setting and releasing of a springloaded striking pin which projects the slide round a race-track past an opening through which the cloud droplets pass.

Our interests have centred on a study of convective clouds of moderate size and usually those free from rain. These restrictions were imposed partly because of instrumental limitations and partly because of the simpler physical processes involved and the greater likelihood of comparing the observations with theoretical predictions. A fairly clear picture emerges from this study. Once a cloud has grown to reasonable dimensions the droplet size distribution at a given level changes comparatively little, either with position or during the subsequent growth and decay of the cloud. The major factors determining the droplet size distribution are the nucleus spectrum and the height above cloud base at which the observation is made.



The cloud droplet sampler in its operating position outside the co-pilot's window.



The sampler showing a magazine, a slide and a photomicrograph of the impressions left by cloud droplets in the soot coating of a slide.



The droplet size distribution is seen to change little with time in these three different clouds.

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Theoretical calculations of the way in which droplets grow by collision and coalescence with their neighbours — which may result in rain — have indicated that initially it is only the "tail" of the size distribution i.e. the largest drops — which is affected. The main part of the spectrum is unchanged. Observations of the droplet spectrum at a fixed level as rain developed in a cloud substantiate these conclusions.

It is possible by numerical techniques to model the effect on the droplet spectrum of mixing between a cloud and its clear-air environment. These calculations suggest that while the mixing reduces the liquid water content at any given level in the cloud it plays no major role in affecting the droplet size distribution. Again our observations support the predictions. However, near cloud tops there are strong indications from the observations that the vigorous mixing taking place is the cause of the development of a bimodal size distribution.

Convective clouds are obviously turbulent, with marked fluctuations in vertical air motion. According to our calculations this turbulence results in continual changes in the droplet size distribution with time as a cloudy parcel of air rises and falls. However, at any particular level the past history of the parcel in its turbulent motion is of little importance: the size distribution is determined almost completely by the nucleus spectrum and the amount of water condensed on the nuclei — i.e. on the height of the parcel above cloud base.



Bimodal droplet size distributions observed near cloud top at successive sampling positions during a traverse.



The change in height of a parcel of air undergoing an assumed random upward and downward motion is shown on the left. The calculated droplet size distribution at each passage of the parcel through the 100 m levels is shown on the right.

We are now at a point where we understand fairly well the early stages of droplet growth, the stages where condensation dominates: our observations are in accord with our expectations on theoretical grounds. The subsequent growth to raindrop size by collision and coalescence is a process which is not as well understood. Much has been done theoretically and in the laboratory, both here and overseas, but few observations have been made in natural clouds which would allow comparisons to be made. Indeed equipment is only now becoming available which permits the appropriate observations to be made; such a study is a major aim in our future work.

Ice Nuclei

Small water droplets falling freely in cold, filtered air do not freeze in appreciable numbers until the temperature falls below about -35° C, yet in the atmosphere clouds often contain substantial numbers of ice crystals at much warmer temperatures. We assume that in the free atmosphere there are some special particles that aid the formation of ice crystals, and these particles are called ice nuclei.

Ice nuclei are important in the initiation of precipitation in cold clouds and are in direct competition with the artificial ice nuclei injected in cloud-seeding experiments. We therefore need to know how many are present at various temperatures and also something about their origin and properties.

In principle all that is necessary to count ice nuclei is to cool an air sample in a suitable enclosure to the temperature of interest and count the ice crystals that fall out. In practice there are difficulties in forming a cloud under conditions resembling those in the atmosphere, in avoiding frost formation on the walls, and in counting the very tiny crystals.

An early version of ice nucleus counter produced here and still popular pre-cooled the air sample to -10° C in a chamber with glycerol solution on the walls to prevent frost, then produced the cloud by means of a sudden reduction of pressure. The ice crystals were made visible by growing them in a supercooled sugar solution.



An early ice nucleus counter developed in this laboratory and still in use. The tray at the top has been raised to show the ice crystals collected.

A method also developed here gives comparable results and is slowly becoming the most common way of counting ice nuclei. The air sample is drawn through a filter with very fine pores so that the particles are left on or near the surface. The filter and its load of particles are cooled down to the temperature of interest in a very shallow metal container. The humidity is raised by increasing the temperature of the ice-covered lid of this container and the ice crystals grow in about 20 minutes to a size where they are readily counted with the naked eye.



Ice crystals growing at $-20^{\circ}C$ on ice nuclei collected on one 5 mm square of a filter.

An attempt was made some years ago to compare ice nucleus concentrations simultaneously around the world. It was a massive undertaking involving about 40 voluntary helpers who exposed four filters daily in different parts of the world for up to three months. Ninety per cent of the filters were returned to this laboratory for processing while the remainder went to the British Meteorological Office.

There was a remarkable uniformity in average ice nucleus concentrations around the world. Values of 30 m⁻³ at -15° C and 300 m⁻³ at -20° C were typical. Since 1000 m⁻³ is barely sufficient to provide a reasonable rate of rainfall in deep supercooled cloud, it is clear that the atmosphere tends to be deficient in natural ice nuclei. However, mechanisms for producing secondary ice crystals apparently exist in some clouds so that this statement may not always apply.



Results of simultaneous measurements of ice nuclei in different parts of the world.

We do not know the size of ice nuclei with certainty. The methods involving diffusion, sedimentation and selective filtration used for cloud nuclei are applicable but are more difficult to apply because of the great rarity of ice nuclei — about 30 m⁻³ (at -15° C) compared with 3x10⁸ m⁻³ for the cloud nuclei. The majority appear to be very small (less than 0.1 μ m diameter) but may become attached to larger inert particles. Many attempts have been made to isolate the nuclei in natural snow crystals; usually one or two large particles are found, together with some hundreds of extremely small particles. The large particles are usually assumed to have been the ice nuclei, probably because they are often clays — clays are known to have ice-nucleating properties when freshly crushed, and are common in the atmosphere. There is little evidence in Australia that clay particles contribute significantly to the population of ice nuclei, probably because in the weathered state in which they occur in particle form in the atmosphere their surfaces are unsuitable for nucleation. For the past three years four filters per day have been exposed for us on the top of the forward mast of an Antarctic research vessel (the USNS Eltanin) by a Bureau of Meteorology observer attached to the ship. At the same time measurements have been made daily at many stations on the ground. Average measurements in each 5° latitude x 10° longitude during the ship's travels are compared in the accompanying figure with the ground-based measurements (shown by crosses). Concentrations higher than 30 m⁻³ at -15°C are shaded. Obviously the highest concentrations of nuclei bear little relation to the dustproducing centre of Australia.

Results of ship-board measurements of ice nucleus concentration at $-15^{\circ}C$.

One of the features of ice nucleus experiments in Australia is the relation between daily rainfall and daily mean ice nucleus concentrations. For example, during nine years of continuous measurements just concluded at Bronte Park in the centre of Tasmania, the mean concentrations on days with different amounts of precipitation varied as shown below.

Relation between rainfall and ice nucleus concentration at Bronte Park, Tasmania 1964-1972.

A simple-minded interpretation might be that more nuclei lead to more rain. Other considerations in this experiment suggest that the situation is much more complex than that.

The future of this work requires improvements in technique and a greater emphasis on airborne measurements. Identification of the nature and origin of the nuclei are ultimate goals the attainment of which will be difficult.

Studies of the Properties of Mixed Clouds

A large proportion of the rain-bearing clouds that are found over Australia contain both ice crystals and supercooled water drops. The rain-making techniques used in this country depend in principle upon introducing ice crystals into supercooled clouds that contain very few ice crystals. In recent years this Division has undertaken a detailed study of the concentration and growth modes of ice crystals in natural clouds. These measurements have been supported by complementary experiments in the Division's cold rooms and by numerical studies using the computing facilities provided by C.S.I.R.O.

CLOUD-SAMPLING TECHNIQUES

The ice crystals and water drops are sampled from an instrumented DC-3 aircraft as it flies through a cloud. The cloud particles enter a divergent tube designed to decelerate the air stream before the particles impact on a moving film coated with a viscous solution of formvar in chloroform. The chloroform subsequently evaporates, leaving replicas of the collected particles in the dried formvar, thus providing a permanent record of the water drops and ice crystals present in the cloud. One problem with this instrument is that while it replicates columnar and small platelike ice crystals extremely well the more fragile stellar crystals frequently break up either in the air stream or on impact. An aluminium foil impactor is used to record the concentrations of water drops and ice crystals greater than 1 mm diameter. With this instrument large particles make an imprint in the aluminium foil and water drops can be distinguished from ice particles by the shape of their imprint. The number of ice nuclei in the cloud is measured by sucking air from outside the aircraft through millipore filters which are processed later in the laboratory.

Replicas of ice crystals collected in supercooled clouds.

FIELD STUDIES

An example of our field studies is seen in an investigation in May 1968 of small maritime cumuli off the coast of Tasmania. It was found that ice crystals and water drops in these clouds were not uniformly dispersed, but rather that there were distinctive regions of water drops and ice crystals. A particularly significant observation was that in some clouds no colder than -10° C there were regions consisting almost entirely of ice crystals in concentrations 10,000 times greater than the concentration of ice nuclei. These excessively high concentrations were not a function of cloud-top temperature but rather a function of the width of the cloud.

In subsequent field trips near Wagga Wagga in New South Wales and near Adelaide in South Australia cumuli and shallow stratocumulus layers formed in air with a continental origin were studied. It was found that the more continental cumuli contained large numbers of ice crystals, comparable to those found in the maritime cumuli of the earlier field trip. However, extensive traverses through the stratocumulus layers revealed concentrations of ice crystals similar to the ice nucleus concentrations. It was also found that the cumulus clouds contained measurable concentrations of large drops and large graupel particles whereas these were absent in the stratocumulus layers.

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

There is no satisfactory explanation so far to account for the discrepancy between the numbers of ice crystals and ice nuclei in the cumulus clouds, but it seems likely that large drops or large graupel particles are necessary before this ice "multiplication" process occurs.

LABORATORY EXPERIMENTS

Many of the microphysical processes taking place in a mixed cloud can be simulated in laboratory experiments carried out in a cold room.

A considerable number of laboratory experiments have been made to test hypotheses put forward to explain ice crystal multiplication in clouds. Two of the most plausible explanations involve the production of ice splinters by the shattering of frozen drops or by riming, i.e. by the collection of small water drops which subsequently freeze. However, experiments here and elsewhere have not yielded any evidence to suggest that either of these explanations is correct.

On the other hand laboratory experiments have been very successful in simulating the growth modes and growth rates of ice crystals in natural clouds. Inside one of our two laboratory cold rooms a chamber has been constructed in which a cloud can be formed, and by seeding this supercooled cloud it is possible to produce ice crystals similar to those observed in natural clouds. After the cloud is seeded ice crystals are collected on an oil-coated slide; these are photographed and then melted to obtain the mass and density of the ice crystals. In this way the densities and growth rates of various types of ice crystals have been studied. These experimental measurements agree moderately well with our theoretical calculations of the growth rate.

Ice crystals being collected and examined in the cold room.

NUMERICAL SIMULATION STUDIES

Once an ice crystal forms on a nucleus it grows by vapour diffusion until it is large enough for the riming process to become important. The crystal habit and the growth rate vary with temperature; the mass growth rate shows two peaks at about -5° C and -15° C differing by one to two orders of magnitude from the minimum growth rate at -10° C. By numerical integration of the growth equations we were able to reproduce

The computed (crosses) and measured (dots) masses of ice crystals as a function of temperature. The dashed line refers to the mass computed on the basis of a spherical ice crystal.

the main features of the observed growth rate. In the early stages of growth the controlling factor is the vapour pressure difference between the environment and the ice crystal; the effect of its shape is small enough to allow the use of a simpler spherical ice crystal model in the calculation. At later stages the crystal geometry plays a major role in influencing the growth rate. The peaks in the growth rate at -5° C and -15° C are found to be associated with the high axial ratios of the crystal form.

Numerical simulation experiments have also been used to study the rate of glaciation of clouds, that is the rate at which ice forms in mixed clouds. It was found that when the cloud of the numerical model, which was assumed to be at a temperature of -10° C, was seeded with a number of ice crystals similar to the number of ice nuclei in natural clouds the calculated rate of glaciation was very slow. When the cloud was seeded with 20 times more ice crystals than ice nuclei the glaciation proceeded more rapidly and most of the ice particles grew large enough to precipitate from the cloud. However, when the cloud was seeded with 1,000 times more ice crystals than ice nuclei it was found that while the cloud glaciated even more rapidly most of the ice crystals were too small to fall out as precipitation. The calculations demonstrate that there is an optimum number of ice crystals for efficient cloud seeding.

It has been proposed that precipitation can also be enhanced by what is called "dynamic seeding". In this form of seeding the latent heat released by the freezing process is used to make the cloud grow in size and hence to be capable of producing more rain. Calculations show that the number of ice crystals in the cloud must be increased many thousands of times before sufficient latent heat is released to make the method effective.

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CLOUD SEEDING

Rain falls from clouds which contain enormous quantities of water, and nearly all of this water originally comes from the sea. The clouds form as a result of cooling, which generally is caused by lifting of the air mass; this requires a tremendous amount of energy, which has its origin in radiation from the sun passing through the atmosphere and warming the surface of the earth. If either water or energy is missing we cannot hope to make up the deficit by artificial means. Hence if we wish to increase the rainfall we must find some trigger mechanism to increase the efficiency of the natural processes of rain formation. Two potentially useful triggers can be found in the cloud microphysical processes involving nuclei.

Rain, as has been mentioned earlier in this booklet, can result from two processes involving either simple coalescence of small cloud droplets or the presence of ice crystals in a cloud. If there is an abundance of active cloud nuclei there will be many very small droplets produced; these all tend to grow to about the same size, and hence to fall at the same rate, thus reducing the likelihood of collision and coalescence. It might be possible to stimulate coalescence by the introduction into the cloud of a relatively small number of droplets somewhat larger than the others, or of hygroscopic particles upon which these larger droplets could grow. If, on the other hand, the clouds grow so large that their tops become sufficiently cold and ice nuclei are present ice crystals will form which may grow to produce useful rain. If ice nuclei are absent or if there are not enough of them, no rain (or little rain) will fall; however, with clouds suitable in all other respects the rain process can be started if the clouds are "seeded" with ice crystals. This can be done by the introduction of ice nuclei such as silver iodide or of 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. However, stimulation of the ice crystal process appears to be even more promising in principle and much easier to apply in practice: it has been the subject of a comprehensive program of research by this group since 1947.

Cumulus cloud before seeding, same cloud 8 minutes after seeding, and rain falling from the same cloud.

Seeding Single Clouds

Seeding clouds may modify them in many ways, but in Australia we are mostly concerned with the effects that seeding has on rainfall.

Ideas for modifying clouds by seeding in order to induce rainfall may be developed in the laboratory, but sooner or later these ideas must be tested by experiment in the natural environment — that is on real clouds. The traditional choice for this purpose has been the isolated cumulus. This forms a good experimental unit because it is a discrete entity whose life history can be studied, and because the stimuli applied can be clearly related to subsequent developments. However, comparatively little of the total rain may fall from this type of cloud and it is not possible to deduce from these experiments what would happen when other types of cloud were seeded. Nevertheless these experiments are an indispensable link between laboratory investigations and attempts at large-scale seeding of real clouds.

In the first experiments of this type dry ice was dropped into the tops of supercooled clouds from RAAF aircraft. This proved to be effective. However, a large quantity of dry ice was believed to be necessary and a large, expensive aircraft was needed to carry it to a high enough altitude, so the method was very costly. It was cheaper to use artificial nuclei in the form of silver iodide smoke: a much smaller quantity was needed and it could be released from a light aircraft into updrafts at the cloud base. This technique was used for many years. It too was effective in many conditions but sometimes it was doubtful if the seeding material reached the best clouds because it was difficult to see their tops from underneath.

We have returned to seeding clouds at the top, both because it should be better and because there have recently been two technical developments which make it easier. Pyrotechnics impregnated with silver iodide which can be dropped into cloud tops are now available and light aircraft can now fly high enough. We are also trying dry ice again, for several reasons. It now seems possible that much less is needed than was previously supposed, it is now much easier to prepare and store than it used to be, and it is safer than pyrotechnics.

Early experiments involved simple observations of events following seeding. The crew of the seeding aircraft observed visually, or by photographs where possible, and reported on the appearance of the rain.

More sophisticated methods of observation have recently been introduced. These include measurements of rain made by flying through it and exposing a variety of sensing elements, and the use of radar.

Silver iodide burner on aircraft wing.

But some of the rain which follows seeding might have occurred naturally. To find out how much is due to the seeding we select a series of clouds, all complying with the same specification, seed half of them and leave the other half unseeded. Then the subsequent behaviour of both halves is observed and compared. If statistical controls are adequate the differences between the behaviour of the seeded and unseeded samples can be attributed to the seeding. In order to avoid any possibility of bias (conscious or subconscious) on the part of those responsible for selecting and observing the clouds, it is important that they do not know which are seeded until after all the observations are complete.

Raindrop impactor on nose of aircraft.

A series of experiments of this type was performed in which clouds were seeded or not on a random basis using silver iodide smoke released into the bases of carefully selected clouds. The total rain falling from each was measured by flying through it repeatedly with measuring instruments on the aircraft. Rain fell from a much higher proportion of the seeded clouds than the unseeded, and on the average several times more rain fell from the seeded clouds.

Total rainfall from isolated cumulus clouds.

EXPERIMENTS AT EMERALD

Recent experiments in seeding single clouds have been concentrated around Emerald, Queensland, where extra rain would be very valuable. A program, expected to have three phases, is in progress.

In the first stage, which is now complete, the clouds were examined to see if they were suitable for seeding and to develop the most promising seeding techniques.

Our tentative conclusions are as follows:

(a) Clouds are frequently considered suitable for seeding but their behaviour varies markedly from day to day. For example, sometimes they all rain naturally; at other times most of them apparently do not rain unless seeded. Efforts are being made to find out what features of the synoptic situation or atmospheric nucleus content account for these differences.

(b) Most of the rain in this area falls from large, complex cloud masses — the traditional isolated cumulus accounts only for isolated, light showers. New turrets of towering cumulus growing on the edges of large storms are frequently suitable for seeding but the effects on the whole system of seeding these turrets is very difficult to observe. Radar provides the only means of studying the cloud group as a whole. (c) Radar was initially fitted to the seeding aircraft and it has proved to be very useful, showing rain echoes which often grow and strengthen after seeding. However, in this form it suffers from the limitation that it can only be used to study one cloud system at a time, so that seeded and unseeded systems cannot, in general, be studied simultaneously. A ground-based radar is being provided for future experiments which should enable us to study clouds over a much larger area.

(d) Seeding these large clouds from the bottom is difficult, because the air crew cannot see where the best clouds are; it is also hazardous, because the clouds often form over hills and sometimes produce hail. Seeding from the top is more practical. The relative merits of pyrotechnics and dry ice for this purpose are being evaluated.

(e) When clouds are seeded with dry ice at about the -10° C level the cloud tops grow and glaciate almost immediately and sometimes dramatically. Modifications following seeding with pyrotechnics have usually been more difficult to distinguish from natural development.

FUTURE WORK

The first stage of investigation in the Emerald area has been satisfactorily concluded: suitable clouds appeared reasonably often and seemed to react favourably to seeding. The second stage is now starting: this will consist of a randomized series of trials as described in the previous page. Finally, if the second stage shows that the rain from individual clouds can be increased, attempts will be made to measure the average rainfall increase which seeding can in practice produce over the area as a whole.

Cessna 402 aircraft used for cloud-seeding experiments.

Cloud Seeding over Large Areas

Convincing evidence that rain can be stimulated from isolated clouds leads us to hope that cloud seeding can cause economically useful rainfall increases. Unfortunately, isolated clouds are relatively rare and only a small percentage of the total rainfall of a given area falls from them. Moreover the methods used for observing the results of seeding isolated clouds cannot be applied in the more complex situations where some clouds are already precipitating. Therefore we must attempt to measure directly the changes cloud seeding produces in average rainfall over a large area.

The basic principle of an appropriate experiment is as follows. Clouds are seeded over a defined area of country, called the "target" area, and the rain that falls is compared with an estimate of how much rain would have fallen if seeding had not occurred. Eventually it may be possible to use physical measurements of the state of the atmosphere to make a sufficiently accurate prediction of rainfall in the absence of seeding, but for the present we have to derive our estimate from rainfall measurements in the target area and in control areas where no seeding takes place.

The simplest rainfall estimate for an area is its "normal rainfall", based on historical records, but of course rainfall on a given occasion may be very different from the normal. Sometimes an area (or combination of areas) can be found where the rainfall is well correlated with that in the target area and so can be used to improve the estimate. However, no estimate based solely on historical records is trustworthy because it may be upset by, say, a change in the habits of synoptic systems. But if time is divided into periods and seeding takes place in only about half of them, then rainfall relationships in target and control areas during unseeded periods form a good basis of comparison with similar relationships in seeded periods. The seeded and unseeded periods should not alternate, because they might synchronize with some weather cycle, but should be allocated by a random process, such as spinning a coin.

The advantages of "randomization" include:

- (a) less possibility of confusion due to unexpected variations in the weather;
- (b) the availability of valid statistical techniques which can assess the results and the precision of the measurements and can reduce the necessary duration of experiment;
- (c) the possibility of installing additional rain gauges for the experiments since historical records are no longer necessary.

EARLY EXPERIMENTS

Our early "area" experiments were designed to answer the simple question, "By how much can cloud seeding increase rainfall over a designated area?". It has gradually become clear that the effects of seeding may be quite complicated and may vary with certain conditions, such as cloud type; it takes an experiment of sophisticated design to give a clear picture of the effects, and the design must provide for the possibility that some of the results may be different from those expected.

Four experiments, in which silver iodide smoke was released from aircraft directly into clouds, were performed between 1955 and 1963 in parts of south-east Australia where: (a) clouds suitable for seeding were thought to occur reasonably frequently; (b) it was possible to measure the rain; (c) two areas of 2500 to 8000 km^2 could be selected with good rainfall correlations; and (d) agriculture or industry would benefit if a rainfall increase could be provided.

The design of these experiments was, in retrospect, overly simple and their results are not as clear-cut as they might have been. However, they suggested that:

(i) On the western slopes of the Great Dividing Range it should be possible to increase rainfall by seeding.

(ii) The seeding appeared sometimes to increase and sometimes to decrease the rainfall. The experiments were not sensitive enough to define the conditions for success, but there was some indication that seeding was more effective with cumulus clouds forming in continental air masses than with those forming in air of recent maritime origin.

(iii) The results in the first year's operation differed from those in subsequent years in all four experiments. This was unexpected and, if it was not fortuitous, has not been explained, unless in some way the results of seeding can persist long after the seeding stops. If rainfall increases which are due to seeding progressively spread into the unseeded periods, or into the control areas,

Observed result in first, second and subsequent years for four Australian experiments combined. Short horizontal lines represent mean result for year.

they would detract from the experimenter's ability to distinguish the increases even if they were the same as they were at the beginning.

The Twin-Otter aircraft used in the Tasmanian experiment.

AN EXPERIMENT IN TASMANIA

An experiment was conducted in Tasmania from 1964 to 1970. It was based on the experience of the earlier series, and its design, as well as being improved in respect of details such as seeding technique, was also improved in various major respects. The intention was to make it possible to:

- (a) detect the results unambiguously even if they varied with conditions;
- (b) define the conditions in which successful results were achieved;
- (c) study any apparent deterioration of results with time which might occur.

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 control areas. The seeding occurred in alternate years, so that any possible build-up of seeding effects during the "on" years and its dying away in the intervening "off" years could be studied.

Clouds and other meteorological quantities were observed during both the seeded and unseeded periods: thus rainfalls could be compared on seeded days of a given type and on unseeded days of the same type, enabling us to find out the conditions in which seeding is most — and least — effective.

The target area of about 2500 km^2 was the catchment of a hydroelectric power scheme which is on a plateau about 1 km above the sea. Rain is mostly associated with cold fronts and low-pressure systems passing over or to the south of Tasmania and it falls in all seasons with a small maximum in winter.

The experiment provided a formidable collection of

data which is still being analysed with the co-operation of C.S.I.R.O.'s Division of Mathematical Statistics.

Preliminary results are as follows. If we look at the results for each season separately a clear and optimistic picture emerges. In spring and summer there is no convincing evidence that seeding had any effect, but in autumn and winter there was a rainfall increase in the target area estimated as 15% to 20% at a very satisfactory statistical significance level. Many aspects of the data provide confirming evidence so that we can confidently accept this increase as being the result of seeding; its value greatly exceeds the cost of the operation.

Detailed evaluations are still being made but initial indications are that the best conditions for seeding involve stratiform clouds and days with moderate natural rain.

The picture concerning any persistent effects of seeding is not clear and it does not appear likely that this experiment will prove whether they do or do not exist.

PRACTICAL APPLICATIONS

When there is a drought or a period of bushfire danger the question naturally arises as to whether cloud seeding could afford any relief. It is not possible to say whether seeding would be worth trying on a given occasion because it would depend on whether suitable clouds occurred. Nevertheless it has sometimes seemed desirable to make the attempt. The cost of the seeding operation can be regarded as a sort of insurance premium which will be repaid (perhaps many times over) if suitable clouds appear and can be stimulated to produce rain.

Practical applications of this type have been the responsibility of State Governments, and for some years we conducted courses of instruction in which officers of the States were taught the techniques of cloud seeding, for use in their home State when the need arose. Several drought-relief projects have been carried out in this way, but in the absence of controls, such as are found in the Tasmanian experiment, we cannot tell how successful they have been.

Tasmania — the experimental areas.

FUTURE EXPERIMENTS

Improvements in experimental design can be expected to follow from theoretical investigations now being conducted using high-speed computers. We can now perform simulated experiments: for example, the rainfall in a given area can be assumed to be altered in a given way by seeding. Several experimental designs can be checked to see which does the best job of detecting the assumed changes due to seeding. In this way, experience can be obtained using a few hours of computer time at reasonable cost which would take decades of field trials (at very much higher cost) to duplicate. These studies, together with the experience of the Tasmanian experiment, should enable us to design better "area" experiments in the future.

Refilling with silver iodide solution.

A power station in the target area.

PARTICLES IN THE STRATOSPHERE

About 20 to 30 minutes before sunrise or after sunset on a cloudless day an acute observer looking towards the sun will notice a bright but brief-lived patch of colour in the sky. When the atmosphere is moist to a considerable depth this may be purple or rose in colour but in very dry continental air it is more usually a mustard colour when high in the sky and yellow or orange near the horizon. More than 50 years ago it was realized that this was due to scattering of sunlight from a layer of particles some 20 km above the surface, but it was not until 10 years ago that the first samples yielded the surprising information that the particles were composed mainly of ammonium sulphate. Shortly afterwards an American contingent of U-2 aircraft visited Australia and we were able to arrange for them to sample particles in the stratosphere for us.

Quite by chance during this time a massive volcanic eruption occurred on the island of Bali in Indonesia. The dust spread rapidly south, giving at first spectacular fiery red sunsets and later extinguishing the twilight colours altogether. The particles collected at first contained relatively large, angular, insoluble fragments of volcanic ash but within a year these were replaced with entirely liquid particles of sulphuric acid.

Electron microscope photograph of large volcanic ash fragments encrusted with soluble material. Typical preeruption particles are also present.

With the departure of the USAF in 1964 this work was dropped but from late 1967 to the present time sampling equipment has been placed on balloons launched by the Department of Supply from Mildura, Victoria (or other sites). Conspicuous changes have taken place in the particles present at 20 km as compared with those typical before the Bali eruption. Intricate rings of tiny droplets surrounding central particles now dominate and these particles can be shown to be largely composed of sulphuric acid.

Stratospheric particles containing sulphuric acid.

In addition to the particle samplers on the balloons a small automatic camera is carried taking photographs of the horizon at intervals of two minutes, or about 500 m in height. It has provided a spectacular record of the course of twilight as seen from various altitudes.

The familiar "purple glow" near sea-level gives way to a vivid fiery arc as one approaches the stratosphere, while above the bulk of particles the twilight is virtually colourless.

Colourful twilight glow at the base of the stratosphere, 10 km altitude.

Relatively colourless twilight glow seen from an altitude of 30 km.

Interesting as such pictures are the main value of the camera is to show the detailed layer structure of the stratosphere revealed by the light scattered from particles.

Marked layering of the light-scattering particles is usual at altitudes near 20 km.

The presence of these layers indicates the dangers of assuming that measurements at one level in the stratosphere are typical of all of it. To overcome this we have designed a particle-sampling system in which a vacuum pump pulls air at high speed through a small nozzle. Underneath is a slowly rotating disc bearing electron microscope screens. The air turns the corner but the inertia of the particles carries them on to hit the surface, where their semi-liquid nature ensures that they stick. This system provides a complete cross-section of particle numbers and size distributions as a function of altitude, as in the diagram below.

Size distribution of particles with altitude on one flight.

The particles are predominantly small, the most common diameter of the equivalent spheres being somewhat less than 0.1 μ m.

Typical size distribution of particles near 18 km altitude on the above flight.

CHEMICAL TESTING OF PARTICLES

In searching for the origin of the stratospheric particles it is important to attempt to determine their chemical nature. With typical diameters of less than 0.1 μ m typical masses will be less than 10⁻¹⁵ g. This is far below the limits of detection of usual analytical techniques. We have therefore devised a method that appears to be applicable not only to particles in the stratosphere but also to the more numerous and possibly more important particles of the lower atmosphere.

In any ordinary "wet" chemical test it is extremely difficult not to lose the original particle or to add unwanted ones. We avoid this by catching the particles on a thin layer of reagent and causing the reaction to proceed by exposing them to the vapour of a suitable solvent. As the solvent molecules become adsorbed on the surface, solution of the particle or reagent occurs and reactions may proceed in the moist surface layer which can subsequently be examined under the electron microscope. Suppose for example we wish to determine the proportion of stratospheric particles that contain sulphate. To do this we catch the particles on a thin layer of barium chloride and then expose them to alcohol vapour for 24 hours. Those containing sulphate change completely in appearance because the very insoluble barium sulphate is precipitated in the presence of the solvent vapour. Typical examples of stratospheric particles on carbon films are shown below and may be compared with similar particles captured and caused to react on a barium chloride surface.

A group of stratospheric particles caught on a carbon surface.

A group of stratospheric particles caught on a barium chloride surface.

There does not seem to be any great limitation to chemical testing by this technique. Tests for nitrates, chlorides, persulphates, oxy-acids and so on are relatively simple, requiring only a careful standardization of methods. Reagent films may be added after particle capture, so that multiple tests are possible on the one

Particles caught at ground level on a nitron surface form bundles of needle crystals when exposed to water vapour if they contain nitrate.

Reactions of chloride particles with a silver nitrate surface kept at 96% humidity for 24 hours.

group of particles. The method looks most promising in dealing with urban pollution problems. For example, a common pollutant consists of droplets of sulphuric acid which react readily with a thin copper layer in the absence of extra moisture. If we compare the reactions on copper (on the left) of tiny droplets of sulphuric acid manufactured in the laboratory, with the reactions on copper of an urban aerosol (right) we see that many of the larger particles contain free sulphuric acid.

Reaction ring left on copper by droplet of sulphuric acid.

Large particles containing chloride collected in Queensland in maritime air.

The virtue of this method of testing is that the *size* range of a given constituent can be examined. The effect of a particle on the environment depends critically on its size — those that are large may be a nuisance to household washing while those that are small and caught by the lungs may be a health hazard. The usual form of analysis, which is dependent upon catching the particles in a large sample of air, cannot discriminate in this way. An apparently harmful major constituent identified in such a sample may in fact be harmless because it occurs in a few large particles that will fall out rapidly, while a minor constituent may be of great significance because it is distributed in vast numbers of very small particles.

For a cloud physics group one of the most interesting possibilities is that we may be able to determine the composition of cloud nuclei, which was originally thought to be sea salt. Indirect evidence based on the stability of the nuclei to heating now favours compounds such as ammonium salts. Our preliminary investigations on collection of typical particles in clear (non-urban) atmospheres show them to be rounded hygroscopic particles that decompose in the electron beam and react completely with barium chloride. Their behaviour is identical with that of ammonium sulphate particles produced in the laboratory. However, giant particles (>1 μ m diameter) which appear to be predominantly sodium chloride have been found in quite large numbers on some occasions.

Reaction ring left on copper by a particle collected from urban air.

Reaction of large particles with barium chloride showing presence of sulphate.

In the past, aerosol work has tended to be divided into the separate sections of stratospheric, tropospheric and urban aerosols. The atmosphere does not recognize these distinctions. Air interchanges between stratosphere and troposphere are common, and urban aerosols in places like Europe and North America are making their presence felt over ever-widening areas. Our aim is to attempt to study the particles of the atmosphere as a whole. An important part of the work is to compare particles in the atmospheres of the northern and southern hemispheres. Measurements following nuclear explosions have shown that particles liberated in one hemisphere tend to stay there. Thus the effects of industrialization and other human activities on atmospheric particulates should be far greater in the populous northern hemisphere than in the southern. Already we have co-operative stratospheric sampling programs with the University of Wyoming and the National Center for Atmospheric Research in Colorado, and we hope that this work can be extended.

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