Stratospheric ice nucleus measurements from balloons

By E. K. BIGG and G. T. MILES, *Radiophysics Laboratory, CSIRO, Sydney, Australia*

(Manuscript received January 14, 1963)

ABSTRACT

The ice nucleus content of stratospheric air was measured at one height in the range 13 to 27 km, on each of 13 experiments, by means of a vacuum pump and filters carried on large balloons.

The concentrations were in close agreement with those found by TELFORD (1960) using an expansion cold chamber in an aeroplane and, per unit mass of air, were comparable with the extreme maxima recorded at the ground in long sequences of measurements in Australia. It is suggested that localised rapid transfer of air from stratosphere to lower troposphere is a probable cause of some ice nucleus storms at the ground. The source of the stratospheric ice nuclei is uncertain but reasons are given for supposing the nuclei to be of extra-terrestrial origin.

Introduction

TELFORD (1960) has described measurements of ice nucleus concentrations above and below the tropopause using an expansion cold chamber which showed the existence of a surprisingly large concentration of ice nuclei in the stratosphere.

To appreciate fully the significance of his results it is necessary to present them in a slightly different form. If the curves of best fit of number of ice crystals per N.T.P. litre against temperature are constructed from his table 1, and averaged into two height groups, Fig. 1 is obtained. In order to compare the results with those to be described, they are multiplied by the ratio of mixing cold chamber counts to expansion cold chamber counts given by KLINE (1961). (BIGG, MOSSOP, MEADE and THORNDIKE (1962) have shown that ice nucleus concentrations derived from sampling with the millipore filters used in the present experiment are closely comparable to those found with mixing cold chambers.)

On the diagram also are shown both the mean and maximum concentrations of ice nuclei measured during lengthy sequences of observations with mixing chambers at various times and places. This figure shows that all the nuclei measured at the ground could have been derived from the stratosphere if suitable mixing conditions prevailed. It is surprising that ground values exceeding these are not more frequent since nucleus concentrating mechanisms exist in the lower troposphere BIGG, (1961).

The implications of Telford's experiment are so important that it is essential to test them by an independent method. BIGG, MILES and HEFFERNAN (1961) have described a preliminary attempt at this which unfortunately was not quantitative owing to lack of information on the effective volumes of air sampled.

This paper describes a new experiment using balloons as the vehicle, which has provided quantitative information on stratospheric concentrations, and also allowed us to determine a vertical profile which gives some indication of the source of the nuclei.

The Equipment

Our equipment formed part of the payload of the "Project Hibal" balloons released at lat. 34° S long. 142° E by the Australian Department of Supply for the U.S. Atomic Energy Commission, and used their 28 volt power supply. The balloons rose at about 5 m/sec to nominal heights of 15, 18, 21, 24, and 27 km in successive flights, where they floated at constant altitude for a period until "cut down", when the payload was parachuted to the ground. A spotting air-

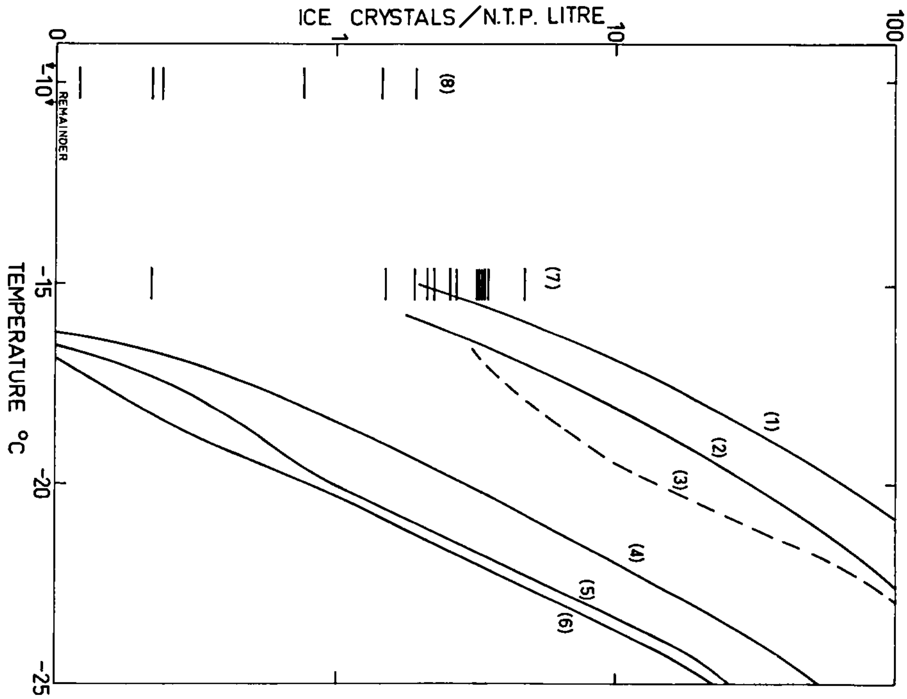

FIG. 1. Comparison of Stratospheric and ground measurements of ice nuclei. (1) Telford, 13 km (aircraft pressure 7.9 km) (2) Telford, 7.6 km (aircraft pressure 4.6 km) (3) Highest ground concentrations recorded in Australia and Antarctic, 8 series. (4) Ground concentrations lat 78° S 1 Dec. 1961–10 Feb. 1962 (5) Ground concentrations lat 34° S 1 Nov. 1956–1 Feb. 1957 (6) Ground concentrations lat 27° S 5 Nov. 1956–1 Feb. 1957 (7) Balloon experiments 13–27 km, −15°C (8) Balloon experiments 13–27 km, −10°C.

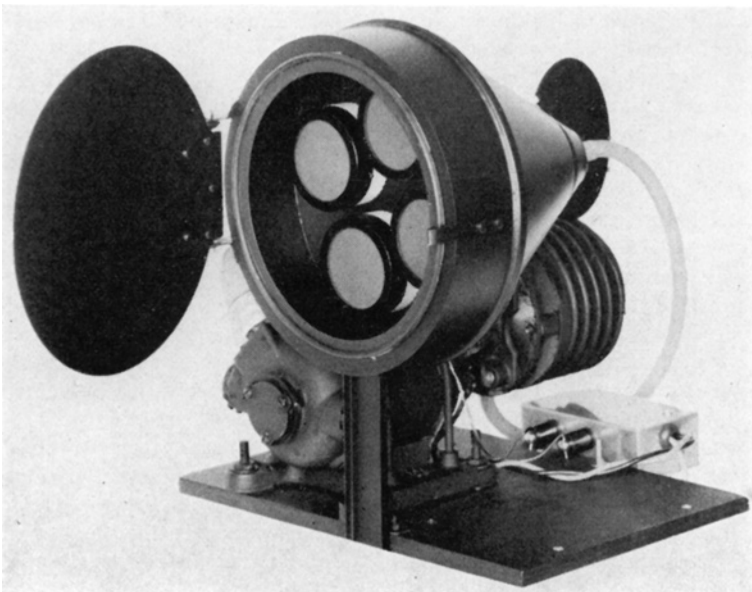

FIG. 2. Balloon-borne air sampler.

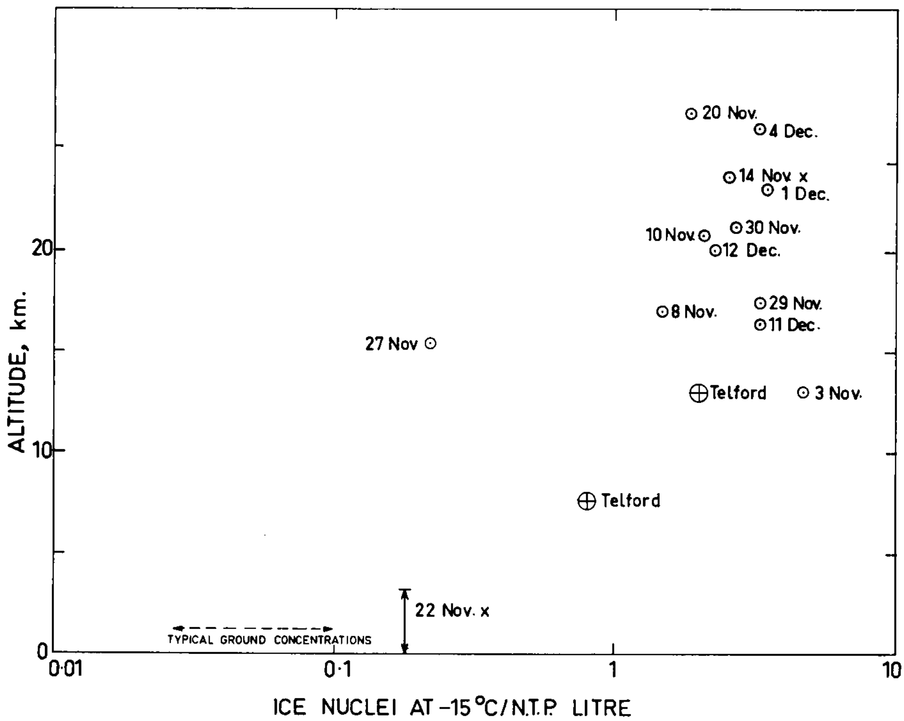

FIG. 3. The dependence of ice nucleus concentrations at $-15°C$ on altitude.

craft directed a ground team to retrieve the apparatus and recovery was usually very rapid. The filters, in sealed containers, were mailed to the Radiophysics Laboratory for processing.

Portion of the sampling apparatus is shown in Fig. 2 and consists of a timer, a 28 volt electric motor driving a vane-type vacuum pump which drew air through the filters, and a device for sealing the filters after sampling had ceased. Sampling was carried out only when the balloon was at constant altitude and the exact volume of air filtered was measured by a revolution counter attached to the shaft of the pump. Volumes sampled varied from about 400 l per filter in the lower to 2000 l in the higher flights and the times of operation from 20 to 130 min.

On several flights the door failed to seal completely and a certain amount of ground dust entered on landing. This appeared to make no difference to the ice nucleus count at $-15°C$, and while the results could have been eliminated from the analysis it seems pointless since they agree with the remainder. They are marked with an X in Fig. 3.

The Measurements

The measurements made at $-15°C$ have been marked on Fig. 1 and are a striking confirmation of Telford's results. One might suppose from the agreement that the ice nucleus concentration of the stratosphere is relatively constant, but we must remember that the earlier experiments although only qualitative, showed some occasions with very low concentration of ice nuclei. More recent measurements by Mossop and Heffernan (so far unpublished) from high-flying aircraft have often given considerably lower concentrations. The stratospheric concentrations of ice nuclei are therefore probably very variable and the present series may have been exceptional.

In Fig. 3 the concentrations measured at $-15°C$ are plotted as a function of height, and it suggests that a lower boundary of the high ice nucleus concentrations may have existed, which on one occasion was above 15 km and on another below 13 km. In Telford's winter measurements it was clearly below 13 km. The constancy with height shown by this figure is interesting, but the stratification of other types of aerosols, which will be discussed in a separate paper, suggests that the ice nucleus concentrations should vary very greatly with height on some occasions, and constant level sampling may then give quite unrepresentative results. It is probable therefore that Fig. 3 represents a particular rather than a general distribution of nuclei with height.

At $-10°C$ the fluctuations in concentration in the present experiment were more conspicuous than at $-15°C$. The mean concentration was 0.4/N.T.P litre, the value 1/l being exceeded twice while six of the flights returned less than 0.1/l.

Filters from a flight which failed to ascend due to icing in cloud were exposed at heights between 2 km and the ground and showed only 0.02/l at $-10°C$ and 0.2/l at $-15°C$. As the protecting door also failed to seal, this flight illustrated that contamination from ground sources may be negligible at the temperatures of measurement.

Discussion

We have seen that stratospheric concentrations of ice nuclei can be comparable with the maximum values encountered in ground measurements in Australia, or about 10 times as high as the mean values. It has been demonstrated by Danielsen (1959), Staley (1960) and Kroening and Ney (1962) that rapid transfer of stratospheric air to the lower troposphere may take place in localised regions. When that air contains high concentrations of ice nuclei and leakage into the troposphere occurs, tropospheric ice nucleus storms would be expected. Many of the phenomena of ice nucleus storms discovered by Bigg (1961), appear to be consistent with this mechanism of their generation.

The problem of the origin of ice nuclei is not thereby solved, and this information shows only that stratospheric residence may be one stage in their history.

The main possibilities are that the stratospheric nuclei (1) originate as terrestrial dusts, (or possibly industrial pollution) which are carried up to the stratosphere by convection or entrainment into streams of air ascending into the stratosphere, and become concentrated there due to the infrequency of effective removal mechanisms; (2) have been generated in the stratosphere e.g. they may be the sulphate particle discovered by Junge (1960); and (3) are of extra-terrestrial origin.

In the northern hemisphere evidence has accumulated that there are a number of terrestrial sources producing ice nuclei operative above $-20°C$ but in the southern hemisphere no significant source has been discovered in spite of very widespread measurements. To explain the observed stratospheric concentration of ice nuclei in the southern hemisphere it would be necessary on the first hypothesis to demonstrate that either local dusts become activated in the stratosphere or that there is a transfer of air across the equator.

The second possibility has been ruled out by the observation that the hygroscopic material caught in the stratosphere is apparently inactive as an ice nucleus at temperatures above $-20°C$.

The third suggestion originated from Bowen's theory based on rainfall statistics, that meteor dusts are active ice nuclei and arrive at the ground about 30 days after a meteor shower. A major controversy arose, criticism being directed at both the statistics and the physics of the theory, but it is now becoming clear that it is sound in principle although some modifications to the "free-fall" assumption of the transfer of the dust through the atmosphere may be required. As far as ice nuclei are concerned measurements have supported Bowen's theory without providing a conclusive demonstration of its correctness. However, a new development—the rediscovery, in a most convincing fashion by Bradley, Woodbury and Brier (1962) and Adderley and Bowen (1962) that rainfall is closely linked with the phase of the Moon—has at last provided strong evidence in its favour. The following discovery by Bigg (1962) that ice nuclei in the southern hemisphere summer show a similar lunar modulation with the remarkable mean range of about three to one now makes it almost certain that they are of extra-terrestrial origin. While the mechanism of the lunar modulation of incoming meteors or micrometeors is not yet known, Bowen (1962) has shown that it is certainly present even in the relatively large meteors detected by a radar set. Combined with the implausibility that the demonstrably negligible lunar influences on the troposphere and stratosphere could be the cause of the changes in concentration of ice nuclei of ground origin, it now seems certain that the nuclei are predominantly extra-terrestrial.

Conclusions

The most important result of the experiments has been to give a quantitative measurement of some of the ice nucleus concentrations encountered in the stratosphere. These have on some occasions been far higher than one normally finds at the ground leading to the supposition that ice nucleus storms in the troposphere could originate from subsidence and mixing of stratospheric air. The interesting possibility arises that we could use ice nuclei as tracers of stratospheric air. Unfortunately this would be confined to regions where ground sources of nuclei are unimportant, and this might rule out many areas of the northern hemisphere.

The probable extra-terrestrial origin of the stratospheric nuclei of the southern hemisphere by its unexpectedness emphasises our lack of knowledge of mixing and transfer processes in the upper atmosphere. A solution of such problems as the requirement of a fixed time delay between the arrival of particles in the mesosphere and in the troposphere is a stimulating experimental and theoretical challenge.

Acknowledgements

We should like to thank Mr. E. Lawton of the CSIRO Division of Irrigation at Merbein who generously volunteered to supervise the installation and operation of our equipment on each balloon flight, the Department of Supply for their permission to include our equipment in their payload and Mr. E. Curwood (site project leader) for his assistance.

REFERENCES

Adderley, E. E., and Bowen, E. G., 1962, Lunar component in precipitation data. *Science* **137**, p. 749.

Bigg, E. K., 1961, Natural Atmospheric ice nuclei. *Science Progress* **49**, p. 458.

Bigg, E. K., Miles, G. T., and Heffernan, K. J., 1961, Stratospheric ice nuclei. *J. Meteor.* **18**, p. 804.

Bigg, E. K., Mossop, S. C., Meade, R. T., and Thorndike, N. S. C., 1963, The measurement of ice nucleus concentrations using millipore filters. Submitted to the *J. App. Meteor.*

Bigg, E. K., 1963, A lunar influence on ice nucleus concentrations. *Nature*. Vol. 197, No 4863, p. 172.

Bowen, E. G., 1963, A Lunar Effect on the Incoming Meteor Rate. Submitted to *J. Geophys. Res.* **68**.

Bradley, D. A., Woodbury, M. A., and Brier, G. W., 1962, Lunar synodical period and widespread precipitation. *Science* **137**, p. 748.

Danielsen, E. F., 1959, The laminar structure of the atmosphere and its relation to the concept of a tropopause. *Arch. Met. Geophys. Biokl. A* **11**, pp. 293–332.

Junge, C. E., 1960, Sulfur in the Atmosphere. *J. Geophys. Res.* **65**, p. 227.

Kline, D. B., 1961, Some experiments on the measurement of natural freezing nuclei. *Monthly Weather Rev.* **89**, p. 263.

Kroening, J. L., and Ney, E. P., 1962, Atmospheric Ozone. *J. Geophys. Res.* **67**, p. 1867.

Staley, D. O., 1960, Evaluation of potential vorticity changes near the tropopause and the related vertical motions, vertical advection of vorticity and transfer of radioactive debris from stratosphere to troposphere. *J. Meteor.* **17**, p. 591.

Telford, J. W., 1960, Freezing nuclei above the tropopause. *J. Meteor.* **17**, p. 86.