RESPONSE OF NATURAL ICE NUCLEI TO DEPOSITED SILVER IODIDE.

ABSTRACT

Aim: To demonstrate that deposition of silver iodide particles on the ground creates secondary ice nuclei and that these interfere with analyses of experiments designed to increase rainfall.

Methods: Measurements of ice nucleus concentrations following the cessation of deposition of silver iodide during cloud seeding show that persistent secondary nuclei had been generated. The information is used to predict the effects on analyses of cloud seeding experiments. Effects on rainfall in large areas surrounding cloud seeding operations are then examined.

Results: The predictions have explained effects such as a decrease in seeding effects with increasing application of silver iodide or with the duration of its application. Widespread downwind increases in rainfall appear to have accompanied all cloud seeding experiments. The explanation offered is that silver iodide increases the airborne concentration of ice nucleating bacteria or their properties. The secondary nuclei appear to be more efficient in stimulating rainfall than the silver iodide nuclei.

Conclusions: Attempts to increase precipitation from clouds by seeding with silver iodide ice nuclei has been motivated by water shortages in many parts of the world. This paper has indicated that the frequent apparent slight or negligible success of such operations may have been due to the influence of the silver on ice nuclei of microbiological origin. An understanding of that influence is urgently required. Much larger and more widespread gains in rainfall than have been found in the past might then be obtained.

Keywords: Ice nuclei, bioaerosols, cloud seeding, weather modification

1. INTRODUCTION

It has been known for a very long time that pure water does not freeze until the temperature falls well below 0^0C. For example it was demonstrated nearly three centuries ago [1] that 1-10ml of boiled rainwater in sealed glass tubes could remain liquid to -9.4^0C. In the absence of particles or contact with a surface, cloud drops can remain liquid to about -40^0C, [2,3]. At this temperature, water freezes without the aid of solid insoluble particles thought to be responsible for freezing cloud drops at warmer temperatures. The sources, properties, temperature of activation and relative importance of those particles to the development of precipitation are still being investigated. Mineral dusts were long thought to be the most likely source although few such particles are active ice nuclei (IN) at temperatures warmer than -15^0C, [4]. An entirely different range of sources was opened up by the discovery that very small particles in leaf litter were capable of freezing water drops at temperatures much closer to 0^0C, [5]. One active ingredient was soon identified as a common bacterium,*Pseudomonas syringae*, [6]. Following that work a very wide range of biological material capable of freezing water at temperatures warmer than most mineral dusts has subsequently been identified [4]. A more recent addition to the list of ice nucleating aerosols is rust spores [7], potentially important because they are relatively common in the atmosphere.

As a result of these discoveries there has been increasing interest in the possible effects of bioaerosols on clouds and climate. An attempt to model their impacts [8]found that if present in sufficiently high concentration, bioaerosols could modify the extent and properties of clouds, precipitation and solar insolation. The important question is how often ice nucleation active (INA) populations reach those concentrations. It is complicated by the demonstration [9] that fragments of ice nucleating bacterial membranes can also be active ice nuclei. Their detection in the atmosphere would be very difficult so that present estimates of atmospheric concentrations of IN of biological origin could be underestimates.Measurements of IN concentrations strictly relevant to natural clouds are usually inexact because of the difficulty of replicating factors such as the time required for particles to contact a water drop or to dissolve a soluble or relatively insoluble coating. Ice crystal multiplication may on occasions be a more important problem in relating IN measurements made with laboratory instruments to ice crystal concentrations in clouds. At temperatures between -3 and -8^{0}C in clouds containing water drops >24µm diameter and also <13µm, [11], a few starter crystals could lead to an avalanche of secondary crystals through riming. This is the temperature range in which a small proportion of ice nucleating biological cells are active. Although mineral dusts have been associated with an observed ice crystal multiplication event [12] an earlier work [13] had shown that biological particles attached to soil dust could raise the nucleation temperature of the mineral component.

The aim of this paper is to describe some observations of large and prolonged increases in IN concentrations associated with releases of silver iodide (AgI) particles nearby. The increases cannot readily be attributed to an effect of mineral dusts, leaving an effect on biological IN as the most probable explanation. Possible causes and consequences of these effects will then be discussed.

2. Protracted increases in IN concentrations or precipitation following the release of AgI

A smoke consisting of very small silver iodide (AgI) particles has been found [14] to be capable of transforming water clouds to ice crystals at temperatures where most natural clouds contained no ice crystals. This discovery opened up the way to practical cloud seeding aimed at increasing precipitation. Before the development of suitable methods for detecting the presence of ice crystals in clouds (about 1970) it was usually assumed that in the absence of obvious glaciation a cloud contained insufficient ice crystals for optimum precipitation efficiency. Randomized cloud seeding experiments began in Australia in 1955 using “seed/no seed” periods for estimating effects relative to an unseeded control. Analysis of these experiments assumed that the AgI particles would be deposited on the ground, de-activated by exposure to UV, or leave the area of interest within 24 hours.

The first regular daily measurements of IN concentrations (at -20^{0}C) in conjunction with upwind AgI smoke generators were made[15]at Climax, Colorado, beginning in November 1954. A continued increase was found during the seeding project and a slow decline after the seeding had ended. More specific effects from a continuation of this work [16] revealed IN counts ten times higher on unseeded days in seeded intervals than on pre-seeding days. Even more remarkable was that the enhanced concentrations took at least six months to disappear after AgI releases were stopped. During that six-month period average concentrations were a factor of five higher than before seeding began. This should have warned experimenters that the assumption that AgI would only affect an area for 24 hours was seriously wrong and that seeded areas could eventually remain effectively seeded in no-seed periods.

By 1960 a suspicion was growing that some unknown factor was contributing to a downward trend in the estimated success rates in Australian cloud seeding experiments. This was attributed [17] to an influence such as that shown by the Colorado work that caused unseeded periods to have become effectively seeded. Measurements of IN concentrations were therefore commenced in each of the two study areas in the cloud seeding experiments in New England and Warragamba. Their locations are shown in fig.1 together with those of other experiments later mentioned in the text.

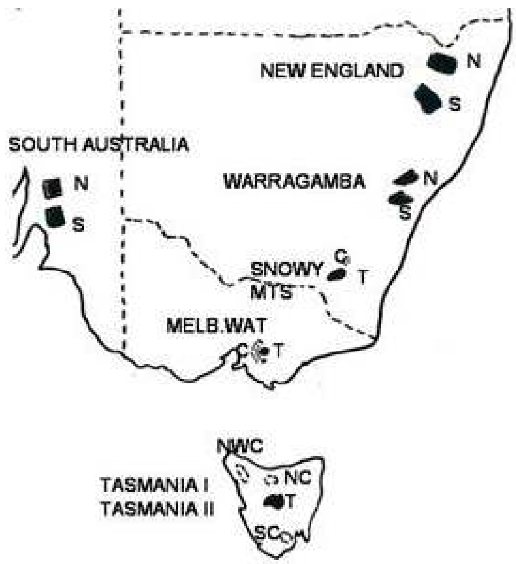

Fig.1: Location of area cloud seeding experiments in south-east Australia, 1955-1992.Target areas are labelled T for single target experiments or N and S for crossover experiments. Unseeded control areas are labelled C.

Of the experiments shown in figure 1,the Snowy Mountains [18] operated from 1955-1959, South Australia 1957-1959, New England 1958-1963[19] and Tasmania 1964-1971, [20, 21]. The results of the Warragamba experiment (1958-1963)have not been formally published. The details used here have been drawn from internal reports of the Radiophysics Laboratory, CSIRO, Australia.

The IN measurements used the filter method [22] in which air totalling 300l was drawn through a membrane filterat a slow rate for 24 hours. The filter was then cooled to -15^0C, the humidity raised to water saturation and the ice crystals that grew were counted after 20 minutes. As an absolute measure of IN concentrations the method has some shortcomings. It detects only nucleation by deposition from the vapour correctly, the more efficient nucleation in the immersion and contact modes being at best partly detected. It can also be affected by large concentrations of hygroscopic particles that reduce the relative humidity in the vicinity of a potential IN. However, that was not a problem at the four elevated and sparsely populated regions under consideration here.Its advantages are that it does not involve skilled operators in the field, provides complete daily average concentrations, is very cheap and gives reliable relative concentrations.

All cloud seeding on the mainland stopped in December 1963 but the IN measurements continued for another 16 months. In 1964 the experiment in Tasmania began, using a year-on, year-off seeding plan designed to avoid any cumulative persistent after-effects of seeding. Interest in persistent after-effects waned and it was not until 20 years later that the results of the IN measurements were analysed for them. Monthly means of IN concentrations as a function of time from the cessation of seedingthen showed the trends seen in figure 2.

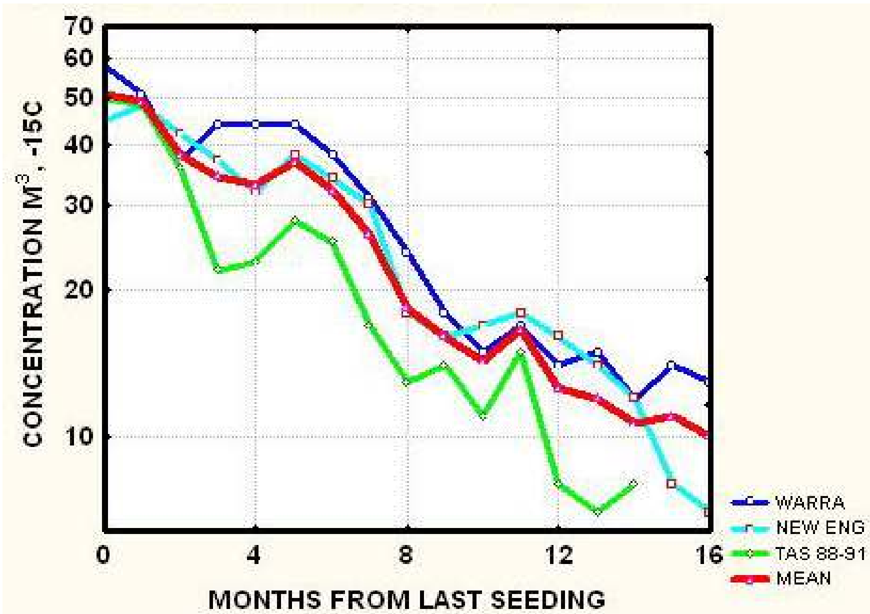

Fig.2: Mean monthly concentrations of IN at -15^{0}C in two areas of each of the New England and Warragamba experiments and in a Tasmanian non-randomised cloud seeding operation.

The slow decrease in concentrations following the end of seeding[23] were sufficiently similar to those earlier reported in Colorado that a measurement campaign was undertaken during and after a three-year cloud seeding operation by the Hydro-Electric Commission of Tasmania. The decreased concentrations again continued for a year after the end of seeding (unpublished). A logarithmic fit to the mean of the three series of measurements shows an exponential decrease with a time constant of 250 days.

3. Effects of enhanced IN concentrations due to AgI on conventional analyses of cloud seeding experiments

The Snowy Mountains and Tasmanian experiments used a target area (precipitation T) that was seeded or unseeded on a limited randomization basis with an average period length of about 11 days. T was compared with that in an unseeded control area (precipitation C). The effect E of seeding for those twotype1 experimentswas estimated from the double ratio:

$$E=\sum(T/C)_{seeded}/\sum(T/C)_{unseeded}\ldots\ldots\ldots\ldots\ldots(1)$$

The remaining experiments used two target areas only one of which was seeded in any period, the other acting as a control. Limited randomization decided whether the north area (precipitation N) or the south (precipitation S) was to be seeded. Period lengths were about the same as in type 1except for Warragamba where it was daily. The effect of seeding for thesetype 2 "crossover"experiments was calculated from:

$$E=\sqrt{\{\sum(N/S)_{north\ seeded}/\sum(N/S)_{south\ seeded}\}}..(2)$$

If it is assumed that secondary IN generation from each seeding event was directly related to the amount of AgI but decayed exponentially with a time constant of 250 days and was cumulative, the increase in $T_{unseeded}$due to the secondary IN will have caused a decrease in E for type 1 experiments. For type 2 experiments the decreasein E would be larger because both areas would have been affected during their unseeded periods.

The simplest demonstration of the effects of secondary IN generation is to compare the observed E with the number of hours of seeding for each year of operation. This is shown in figure 3 for the

Snowy Mountains experiment. The apparent success of the early years led to the hope that increased seeding would lead to an even better result, culminating in the very poor result in the last year. In the Tasmanian experiment, the double ratio in the first year was 1.24 with only 23 h seeding, in the third year 1.05 with 181h. In years 4 and 5, seeding hours were reduced to 128h and then 76h and the double ratios in both were 1.11.

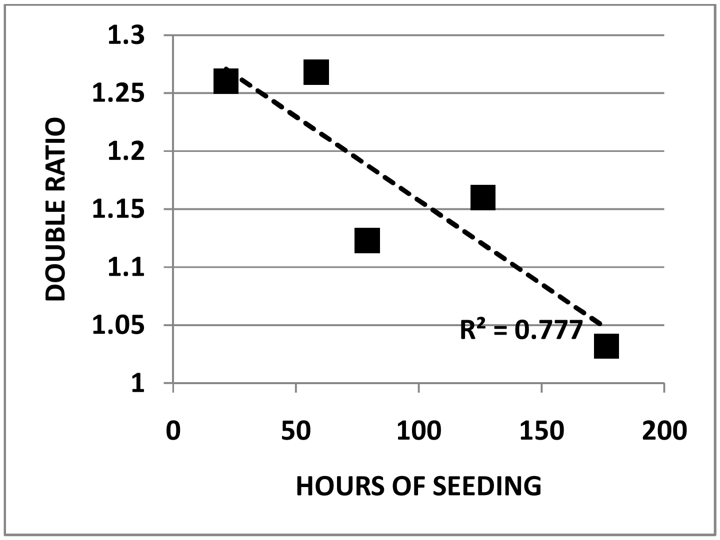

Fig.3: In the Snowy Mountains experiment, E measured by the double ratio decreased substantially as the number of hours of seeding per year increased. E used period totals, not just seeded days.

Similar trends in crossover experiments have been found by treating the two seeded areas as separate experiments comparing their rainfall with that of usually upwind areas.

In the Tasmanian experiment T/C double ratios for autumn, winter, spring and summer averaged over the five seeded years were respectively: 1.29, 1.23, 1.05 and 0.97, [20, 21]. Seeding began in autumn in each seeded year and the decrease through the year was attributed to an unexplained seasonal effect. The alternative proposition that the decrease was due to an increase in secondary IN concentrations as a result of seeding was not considered. It can be tested by modelling cumulative effects of AgI deposition using the IN measurements of fig.2.

A logarithmic decrease provides a close fit to the red curve in fig.2 which is the mean IN concentration for the three experiments as a function of time from the last seeding.If IN concentrations resulting from one day's seeded mass m_0 gm of AgI is $\mathbf{N_0}$, then $\mathbf{d_i}$ days after seeding, the residual effect could be expressed in the form:

$$\mathbf{N_i} = \mathbf{N_0} m_0 \exp(-\alpha(\mathbf{d_i}-\mathbf{d_0}))$$

where α is the mean slope per day (0.004) of a logarithmic fit to the curves of fig. 4. If the effects of each seeding are linearly cumulative, the net effect of all previous seeding on IN concentrations on day $\mathbf{d_i}$will be the "cumulative seeding index":

$$\mathbf{CSI} = \mathbf{N_0} \mathbf{\Sigma} m_i \exp(-\alpha(\mathbf{d_i}-\mathbf{d_0})).$$

The formula was first applied to the period T/Cdouble ratios for each declared period, whether seeded or unseeded, for the Snowy Mountains experiment of 1955-1959 with the result shown in Figure 4.

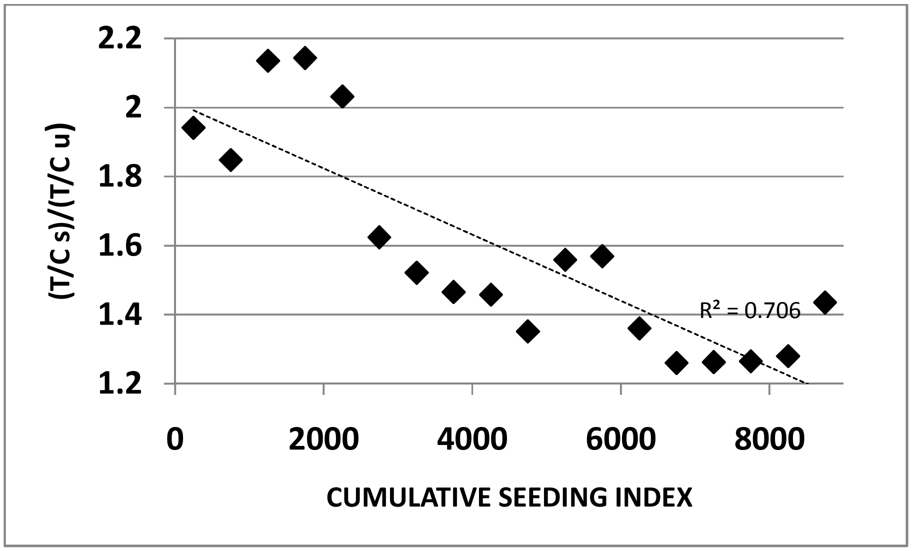

Fig.4: Comparison of mean ($T/C_{seeded}/T/C_{unseeded}$) as a function of CSI at intervals of 500 for the Snowy Mountains experiment of 1955-1959.

It is obvious from fig. 4 that E decreased considerably at high values of CSI. An increase in T during unseeded periods due to persistent AgI-induced INwould explain the result..

For the Tasmanian experiment,1964-1971,the northwest control (precipitation **NWC**) is the one least often to have been downwind from the target area. The double ratio (target east and west combined) $\mathbf{(T/NWC)_{seeded/}(T/NWC)_{unseeded}}$ was 1.17 for CSI<500 and 0.96 for CSI>500 (maximum CSI=5600). This ~20% difference seems unlikely to have occurred by chance, but could have been affected by seasonal differences in IN for seeded and unseeded periods since the latter included more summer periods than the former. However, in that experiment, days within the operational months were divided into two categories: "suitable for seeding and seeded (***ss***), suitable for seeding but unseeded (***su***)". Comparison of (**T/NWC**) ratios for the two categories should have been free from bias and showed the decreasing trend with CSI seen in fig.5. The decrease is so large that it is unlikely to have arisen from chance. It would again be explained by an increase in Ton unseeded days caused by the increasing concentrations of secondary IN.

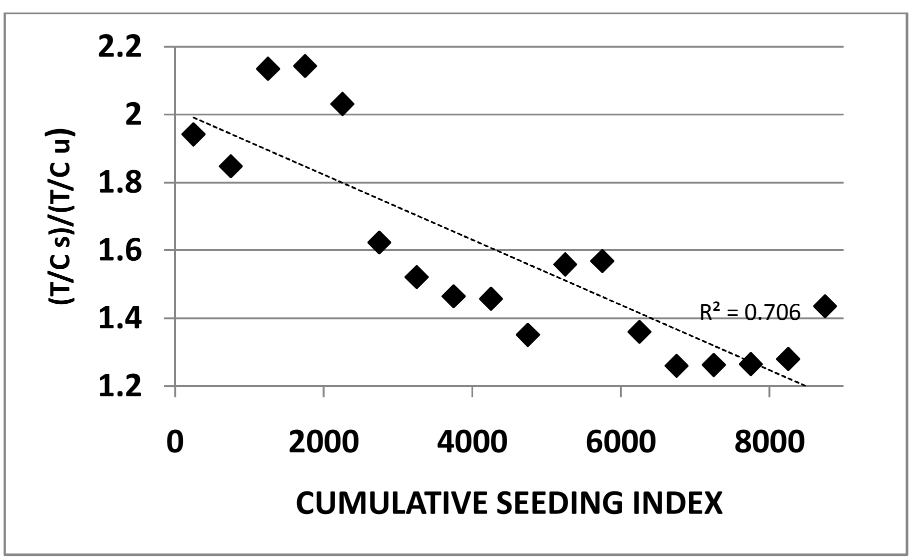

Fig. 5: Target/Northwest control precipitation on days suitable for seeding (**ss**), divided by the same ratio for days suitable for seeding but unseeded (**su**), compared with the cumulative seeding index. This measure of E refers to individual days only, not to period totals.

Days suitable for seeding but unseeded were not recorded for the other experiments but for the crossover experiments calculations were made by treating each seeded area as a separate experiment and creating “control”areas to the west. All showed decreases in E with increasing CSI. It is sufficient here to note that the New England experiment in which one area remained unseeded for as long as 30 days on three occasions in the first year showed an apparent 31% increase in rainfall. After 6 years, the overall increase was only 4%. In the Warragamba experiment with its daily randomisation,even in the first year there was not a significant apparent change in rainfall. In the presence of secondary IN generation due to repeated seeding of both areas with AgI,the short time between successive seedings would have led to much more rapidly increasing CSI in each area and correspondingly rapid decreases in double ratios.

It is of some interest to know how rapidly a response to AgI can be detected. The mean effect of secondary IN on target rainfall in the 30 days following seeding was investigated [24] for the Tasmanian experiment only, using a superposed epoch analysis. It was then extended to include all previous experiments [25] by setting up artificial control areas for each area of the crossover experiments.A similar analysis for a ten year hail mitigation experiment at Nelspruit in South Africa’s Transvaal [26] produced the strikingly similar result shown in fig. 6.

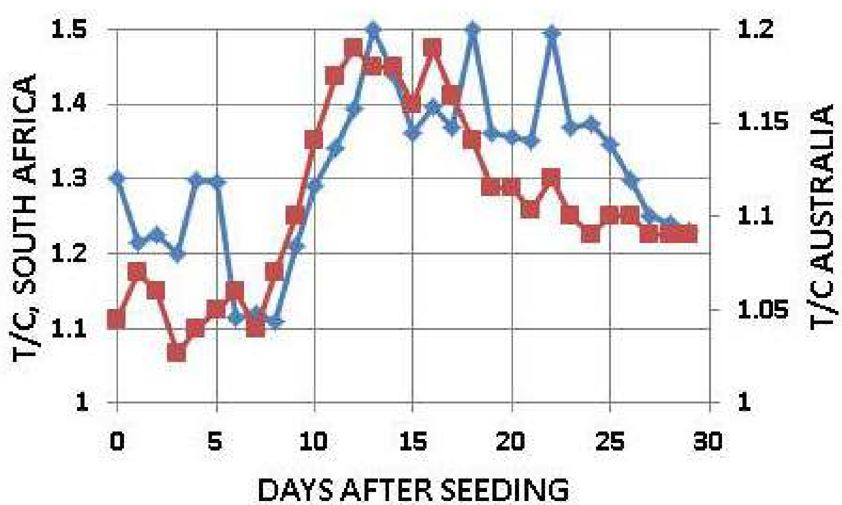

Fig.6. Mean (Target/Control) on each day 1 to 30 following seeding divided by the mean T/C for the preceding 30 days. South African experiment: blue,Australian experiments: red. Note the different scales.

The South African and Australian T/C changes with time from a seeded day were obviously correlated although differing by a factor of 2. The much larger amount of AgI used in South Africa and the greater number of years of operation could be responsible for the difference. T/C here is a single instead of a double ratio,so that it measures only the mean changes withtime in target rainfall following individual seeding episodes, whether due to AgI or secondary IN. The most significant increase in target rainfall and therefore presumably in secondary IN began about 10 days after a seeding event and had a peak effect on rainfall double that of the increased T/Con the day of seeding.

A cloud seeding experiment was conducted by Melbourne Water from 1987-1991 [27, 28] aimed at increasing water in one of the city's reservoirs. Although the organizers were sceptical of the reality of persistence effects, it was agreed that a network of IN measuring sites should be set up around the target and control area. In setting up this network it was specified that correlations between the Australian contribution to fig.6 and both rainfall and IN measurements would be used to show the presence or absence of persistent effects of seeding. The ratio of IN concentrations in the targeted area to those at an upwind site showed a significant correlation, corroborated by corresponding comparisons with rainfall from the dense rainfall gauge network installed for the experiment. Correlation with the Australian component of fig. 6 significant at the 0.1% to 1% levels occurred in an area of about 3000km^2 directly downwind from where seeding took place. The implications of this result were ignored by those conducting the experiment.

4. Suggested cause of the cumulative effects of seeding.

The measurementsof IN following the cessation of seeding in Colorado and Australia, showed that AgI deposited on the ground led to a production of persistent secondary IN. Application of this information to the results of cloud seeding experiments has explained many puzzling features of their results, such as the decreased apparent success with seeding hours per year or with the duration of the experiments. The immediate problem in explaining the secondary IN production is the extremely small amounts of AgI deposited on the surface. For example in the Snowy Mountains experiment, the five year total was 460kg. This may seem a lot but the target area was of the order of 2000km^2 and if the AgI was spread uniformly over it, the accumulated surface concentration would be only about 200µg.m^{-2}. In fact, muchof the AgI would often have escaped precipitation within the targeted area and been deposited further downwind.

AgI particles deposited on the ground will dissolve in the rain water if very small and the AgI will be left as a thin film on surfaces when the water evaporates or will sink into the ground. Films left on surfaces will then decompose under the influence of UV into extremely small Ag particles, and the iodine will escape. Undissolved AgI particles will also decompose on the surface into Ag nanoparticles. Re-entrainment of nanoparticles is not a reasonable explanation for persistent increases in IN because of the strong electrostatic forces binding them to the surface as well as their loss of nucleating ability by exposure to light.

A suggestion has been made[29]that the iodine liberated by UV might create secondary IN through reaction with plant oils. The relatively high vapour pressure of iodine and the tiny amounts involved make this a very improbable explanation. Changes to ice-nucleating properties of mineral dust by deposited AgI is also unlikely because the amounts left on the surface will be so thinly spread and will have probably decomposed before becoming airborne. The only plausible mechanism involves a modification of the ice-nucleating properties or airborne concentrations of existing potential IN such as plant-living INA bacteria.

The silver ion is highly reactive, binding readily to proteins. AgI in solution in rainwater impacting on leaves will therefore potentially affect plant-living bacteria as well as their habitat. Bacteria will accumulate silver directly from rain containing a very weak solution of AgIand subsequently from the surfaces on which they live when dew or further rain allows the Ag^+ ion to form from the Ag residues.

The toxicity of silver nanoparticles for both gram negative and gram positive bacteria has been well established. In areas where the AgI was heavily deposited, a decrease in the natural IN could result from this toxicity. However an increase in IN, not a decrease, is needed to explain the cumulative increases of IN concentrations described here. One possible way in which this could happen is suggested by observations [31] that concentrations of the silver ion about equal to that in a saturated solution of AgI (3µg.l^{-1})actually stimulated growth in *E.coli*- a "hormetic" response- whereas stronger concentrations killed them. Even at much higher concentrations of silver nanoparticles it has been foundthat a different bacterium *Cupriavidusnecator*experienced growth at certain stages of development [32].Suppose that the stimulus to growth of INA bacteria resulting from exposure to extremely small amounts of Ag^+ results in a significant effect on bacterial populations. Then assumingthat increased populations in the phyllosphere lead to increased airborne populations, increased IN concentrations following seeding episodes would persist until the supply of silver ran out. Before such an explanation can be accepted, a great deal of experimental work will be needed.

Another suggestion [24] is that accumulation of silver by bacteria puts them under stress making it more likely that they would be dislodged from leaves by wind. There is at present no evidence for such a process.Another possibility for which there is also no observational evidence is based on the fact that AgI and INA bacteria have one thing in common – surface structures capable of binding water into an ice lattice. It could be speculated that the influence of the AgI is to increase the rafting of INA proteins on bacterial membranes, making a greater proportion of bacteria such as *P. syringae*active IN.

5. Assessing cloud seeding experiments when there are persistent effects of seeding.

Figs. 3-6 have shown how seriously misleading the statistically derived changes in rainfall due to seeding with AgI can be because of secondary IN generation. How can the real effects be assessed? There is one simple method that has been used in the operational seeding in Tasmania, described in annual reports of the Water Resources Department of the Hydro-Electric Commission, e.g.[33]. It involves expressing the total precipitation of all rainfall reporting sites in a large area around the target for the whole seeding season (not just the seeded days) as a proportion of their average in common unseeded years. In fact, one or moreof the following months as well could be included in the analysis because of the persistence of secondary IN. If the pattern of high rainfall ratios appears to have been consistently related to the target area, then an effect of seeding could be provisionally assumed. The main problem with the method is that it cannot readily be disproved that the patterns of rainfall highs and lows were due to large scale influences on rainfall,e.g. the El Nino-Southern Oscillation. Extreme localised events such as a strong cyclone can also modify rainfall patterns.The method is also unpopular because it is very difficult to ascribe a significance level to the result. However, a method that shows the real effect with some added randomness may be preferable to one that inevitably grossly underestimates the overall effects of seeding and does not show the geographical limits of any effects of seeding.

Comparing pairs of seven year unseeded groups in New England and in Tasmania suggested that the range of ratios from random causes is usually 90 to 110%, with random patterns of high and low ratios. Much larger variations during cloud seeding experiments would make it less likely that they were unrelated to AgI deposition but would not be proof. The other feature that would indicate a real effect is if the pattern of highest rainfall ratios should occur in the predominant downwind direction of seeded days, because not all the AgI will usually be deposited by precipitation in the targeted area. Due to the persistence of the secondary IN, there will potentially be a widened area of influence due to winds from other directions on non-seeded days when precipitation is possible.

Fig.7 shows the patterns of rainfall at all available Australian Bureau of Meteorology rainfall gauge sites during seeded months plus one, divided by mean rainfalls for corresponding periods in seven unseeded years for each of four experiments. Westerly winds on the day of seeding were predominant in a, b and c and north-westerly winds in d. The distribution of winds on seeded days are shown by dashed lines in 7a and 7c. The highest ratios of seeded/unseeded are coloured red, while green means little change. Maximum values were: 7a: 1.3, 7b and 7c:1.4 and 7d: 1.25. The anomalies are all much larger and more widespread than in the trial unseeded comparisons and are consistently in the downwind direction. Because this type of analysis considers months on which any seeded days occurred, the great majority of days were unseeded. In fig.7d, the apparent increases were about double those deduced from target/control double ratios.

The extension of elevated rainfall ratios to the southwest of the seeded area of fig.7a might seem to cast some doubt on the reality of effects downwind from the seeded area. However, from September 1954 to November 1956 a ground-based AgI generator was operatedthroughout every night with southwest or west winds at a mountain site (1000m) in the extreme left lower corner of figure 7a with the hope of increasing precipitation in a valley to the east and northeast. No obvious effects were found in the targeted area that was close to the delivery site and the work remains unpublishedexcept for an internal CSIRO report by E.E.Adderley. It was thought that the site was sufficiently removed from the randomized experiment that began in 1955 to have no influence on it but the enhanced rainfall ratio pattern downwind from it resembles that of the main experiment. No records exist of the amount of AgI used but because of the long operating hours and daily use, it must have been large.

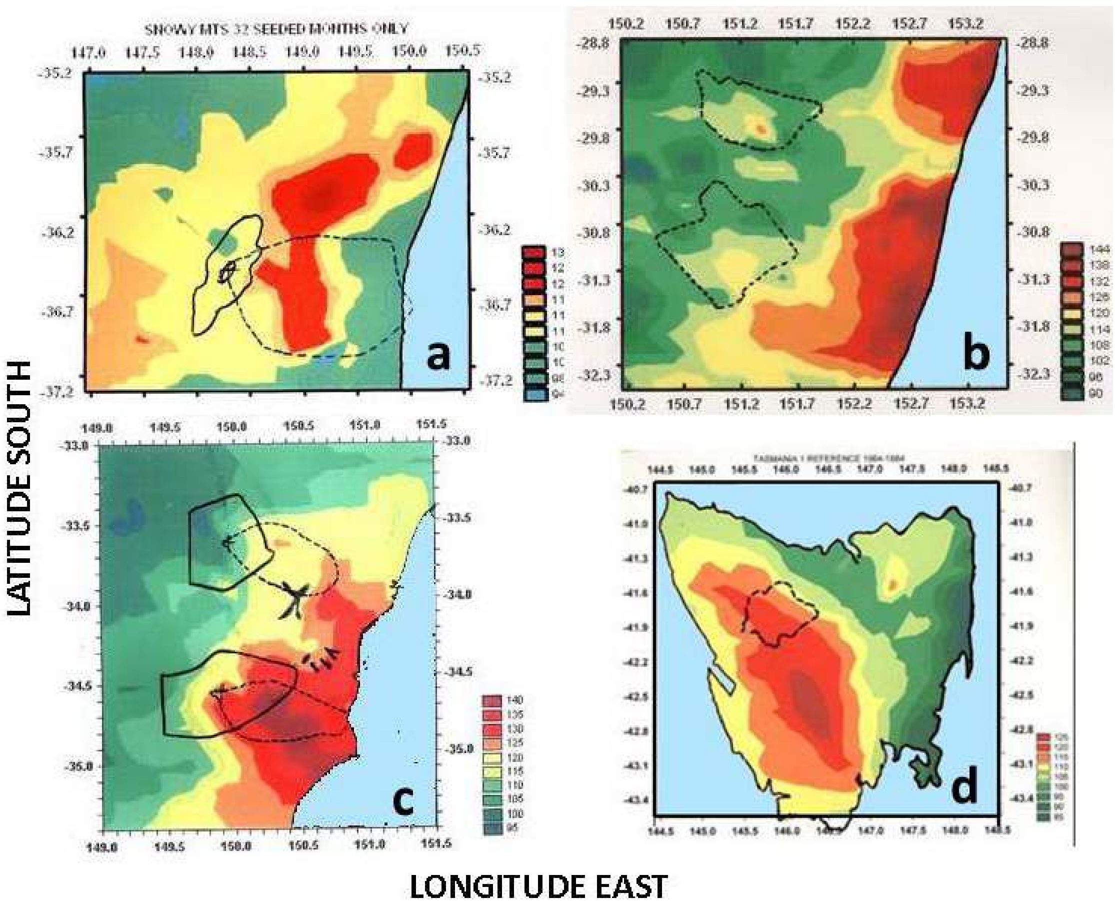

Fig.7: Target areas of four early cloud seeding experiments shown by solid lines in aand c, dashed lines in b and d. The diagrams show contours of the ratio of mean rainfalls in seeded months to the means in corresponding months of seven unseeded years. The distribution of wind directions on seeded days are shown as dashed lines in **a** and**c**. Red areas have ratios>1.2.

The high ratios relatively far downwind from the AgI delivery areas in each figuresuggest that rainfall was increased more effectively where deposition ofAgI was relatively slight. The pattern of ratios is consistent with the hypothesis that toxic effects of AgIsometimes reduced natural IN concentrations near the area of delivery of the AgI, but enhanced them in areas where deposits were slight. Some support for this comes from a diagram [23] showing rainfall ratios in 1^0rectangles of latitude and longitude in the area covered by figs.7b and 7c for the year (1964) following the cessation of seeding in New England and Warragamba. Mean precipitation in 27 contiguous squares adjoining the former seeded areas was 107% of average, but in the remaining 37 squares was only 80% of average. It could be supposed that by 1964 the Ag ion availability in the former seeded areas now fell within the range where bacterial growth more often led to secondary IN production than to toxicity and resulted in increased rainfall.

6. **Discussion**

Perhaps the most important conclusion from figs. 6 and 7 is that the secondary IN were far more effective in increasing rainfall than the AgI particles. The fact that their increase was not obvious until more than a week from seeding and that the greatest accumulated effects apparently occurred well downwind from the targeted areas explains why their influence has not been generally recognised. Another factor is that those conducting cloud seeding have in the past been mainly trained as engineers and atmospheric physicists and effects of AgI particles alone lasting more than a day or two appeared impossible. That microbiological entities may have been involvedis even now only slowly becoming recognised.

To carry this work further, the hypotheses of section 4 (and any others that are offered) need to be subjected to experimental observations.An early attempt to do this [24] involved two natural grass plots 3m x 2m and 1m apart, each totally enclosed by transparent polythene sheets stretched on a wooden frame 2m high. A lightweight aluminium roof prevented rain from collapsing the plastic. A slight overpressure was maintained in each chamber by pumping in filtered air at $10l.min^{-1}$ to prevent entry of particles from outside. One chamber was watered with a misting spray of saturated AgI solution, the other with plain water. Filters sampling $1m^3$ of air daily from each chamber were used to count IN active at -10^0C and the difference between IN counts in the two chambers plotted as a cumulative curve. The difference first became clear after 10 days and increased most rapidly between days 65 and 180. Electron microscope grids collecting particles by sedimentation in the treated chamber showed nearly all to beeitherfungal spores, bacteria or parts of their membranes, orbrochosomes (300nm particles produced by leaf hoppers). While the experiment showed that more IN were produced by the chamber exposed to AgI, it did not show why.A repetition of that experimentbut with an emphasis on microbiology should be considered.

Confirmation of the apparent far-downwind effects of AgI on rainfall seen in figure7 from experiments and operations in other parts of the world also would be desirable. Huge amounts of seeding have taken place in many parts of the world, for example commercial cloud seeders in USA, and hail suppression programs in USSR and France. If toxic effects of Ag on INA bacteria are important, reductions in rainfall might be found in the areas of heaviest deposition of AgI and increases at a considerable distance.

The earliest and most controversial report of such far downwind effects as those in fig.7b [34, 35] claimed to have instigated a periodicity in rainfall over much of the eastern part of USA by releasing 1kg of AgI on the same day each week for several years in New Mexico. A weekly periodicity became apparent after a few months and even changed its phase corresponding to a changed seeding day. Few scientists could believe that it was caused by the AgI because a similar, althougha less pronounced and prolonged periodicity, was found to have occurred in the previous 50 years. It was also thought that such a large-scale effect could not possibly have arisen from such a tiny amount of AgI. The same investigator (Irving Langmuir) responded to similar criticism of his plan to modify hurricanes:"… it is like assuming that a very large forest could not be set on fire by such a small thing as a single match"[36].The controversy was so bitter that the experiment has not been repeated. Perhaps it should be!

7. **Conclusions**.

Daily measurements of IN concentrations on a scale of years made in conjunction with the release of AgI smokes showed unequivocally that generation of secondary IN persisting for months resulted. When this information was applied to the results of cloud seeding experiments, many of their puzzling features appeared to be explained. It seems likely that past attempts worldwide to increase rainfall or snowpack by seeding with AgIwill have been influenced by the effects of secondary IN. Too much AgI may often have been used in the target area causing a toxic effect on biological IN, while beneficial effects far downwind have been overlooked. If the effects of AgI on INA bacteria can be thoroughly understood and quantified, it is possible that increasing rainfall in specified areasthrough seeding with AgI could become vastly more successful than in the past. The importance of water availability in a world with rapidly increasing population suggests that a continuation of this work should receive a high priority.

References

[1] Fahrenheit, D.G., (1724). Experimentaet observations de congelationeaquae in vacuo factae. Phil. Trans. Roy. Soc., 33, 381-391.

[2] Schaefer, V.J., (1946). The production of ice crystals in a cloud of supercooled water droplets. Science, 104, 457-459.

[3] Schaefer, V.J., (1948). The production of clouds containing supercooled water droplets or ice crystals under laboratory conditions. Bull. Amer. Meteorol. Soc., 29, 175-182.

[4] Després, V.R., Huffman, J.A., Burrows, S.M.,Hoose, C., Safatov, A.S., Buryak, G., Fröhlich-Nowoisky, J., Elbert, W.,Andreae, M.O., Pöschl, U. andJaenicke, R., (2012). Primary biological particles in the atmosphere.TellusB, 64,15598, (40pp.)

[5] Schnell R.C and Vali, G.,(1972). Atmospheric ice nuclei from decomposing vegetation. Nature, 236, 163-165.

[6] Maki, L.R.,Galyan, E.L., Chang-Chien, M.M. and Caldwell, D.R., (1974). Ice nucleation induced by *Pseudomonas syringae*. Appl. Microbiol., 28,456-459.

[7] Morris, C.E., Sands, D.C., Glaux, C., Samsatly,J., Asaad, S., Moukahel,A.R., Gonçalves, F.L.T., and Bigg, E.K., (2013). Urediospores of rust fungi are ice nucleation active at -10^{0}C and harbor ice nucleation active bacteria. Atmos. Chem. Phys. 13, 4223-4233.

[8] Phillips, V.T.J., Andronache, C., Christner, B., Morris, C.E., Sands, D.C., Bansemer, A., Lauer, A., McNaughton C., and Seman, C. (2009). Potential impacts from biological aerosols on ensembles of continental clouds simulated numerically. Biogeosciences, 6, 987-1014.

[9] Lindow, S.E., Lahue, E.A.,Govindarajan, G.,Panopoulos,N.J. and Gies,D., (1989). Localization of ice nucleation activity and *ice-C* gene produced in *Pseudomonas syringae*and *Escherichia coli*. Molecular Plant-Microbe Interactions, 2, 262-272.

[10] Hallett, J. and Mossop, S.C., (1974). Production of secondary ice particles during the riming process. Nature 249, 26-28.

[11] Mossop, S.C., (1978). Some factors governing ice particle multiplication in cumulus clouds. J.Atmos. Sci., 35, 2033-2037.

[12] Crawford, I., Bower, K.N., Choularton,T.W. and 13 others, (2012). Ice formation and development in aged, wintertime cumulus over the UK: observations and modelling. Atmos. Chem. Phys. 12, 4963-4985, 2012.

[13] Conen, F., Morris, C.E., Liefeld, J., Yakutin, M.V. and Alewell, C., (2011). Biological residues define the ice nucleation properties of soil dust. Atmos. Chem. Phys., 11, 9643-9648.

[14] Vonnegut, B., (1949). Nucleation of supercooled water clouds by silver iodide smokes. Chem. Revs., 44, 277-289.

[15] Grant, L.O. andTerbutton, V.F., (1957). Ice nuclei in the free atmosphere in the western U.S. Paper presented at 151st national meeting, American Met. Soc. and conference on hydrometeorology, Chicago, Ill., Mar. 21, 1957.

[16] Grant, L.O., (1963). Paper presented at Adirondack Conference on silver iodide phenomena, (Wilmington, N.Y. Nov.1962, reported by V.J.Schaefer, Bull. Amer. Meteorol.Soc., 44, 434.

[17] Bowen, E.G., (1966). The effect of persistence in cloud-seeding experiments. J. Appl. Meteorol., 5, 156-159.

[18] Smith, E.J., Adderley, E.E. and Walsh, D.T., (1963). A cloud-seeding experiment in the Snowy Mountains, Australia.J. Appl. Meteorol., 2, 324-332.

[19] Smith, E.J., Adderley, E.E. and Bethwaite, F.D.,(1965). A cloud-seeding experiment in New England, Australia. J. Appl. Meteorol., 4, 433-441.

[20] Smith, E.J., Veitch, L.G., Shaw, D.E., and Miller, A.J., (1977). A cloud-seeding experiment in Tasmania, 1964-1970.Final report, CP183, Div. Cloud Physics, CSIRO, Australia.

[21] Smith, E.J., Veitch, L.G., Shaw, D.E., and Miller, A.J., (1979). A cloud seeding experiment in Tasmania. J. Appl. Meteorol., 18, 804-815.

[22] Bigg, E.K., Mossop, S.C., Meade, R.T., and Thorndike, N.S.C., (1963). The measurement of ice nucleus concentrations by means of Millipore filters. J. Appl. Meteorol. 2 (2), 266-269.

[23] Bigg, E.K.,(1985). Unexpected effects of cloud seeding with silver iodide. J. Weather Modification, 17, 7-17.

[24] Bigg, E.K., and Turton, E., (1986). Delayed effects of cloud seeding with silver iodide. J. Climate Appl. Meteorol., 25 (10), 1382-1386.

[25] Bigg, E.K., and Turton, E., 1988).Persistent effects of cloud seeding with silver iodide. J. Appl. Meteorol., 27(5), 505-514.

[26] Mather, G.K., Bigg, E.K., and Renton, S., (1990). Apparent persistence effects in the Nelspruit area from silver iodide seeding for hail suppression. J. Appl. Meteorol., 29, 806-811.

[27] Long, A.B., (1995). Physical aspects and design features of the Melbourne Water precipitation augmentation project. Precipitation Enhancement Workshop 24-28 April, 1995, Terrigal, New South Wales, Australia.

[28] Bigg, E.K.,(1995). Test for persistent effects of cloud seeding in a recent Australian experiment. J. Appl. Meteorol.,34,2406-2411.

[29] Rosinski, J. and Parungo, F. (1966). Terpene-iodine compounds as ice nuclei.J. Appl. Meteorol., 5, 119-123.

[30] Rai, M.K., Deshmukh, S.D., Ingle, A.P., and Gade, A.K. (2012). Silver nanoparticles: the powerful nanoweapon against multidrug-resistant bacteria. J. Appl. Microbiol., 112, 841-852

[31] Xiu, Z-m, Zhang, Q-b., Puppala, H.L., Colvin, V.L., Alvarez, P.J., (2012). Negligible particle-specific antibacterial activity of silver nanoparticles.Nano Lett.,12(8), 4271-4275.

[32] Schacht, V.J., Neumann, L.V., Sandhi, S.K., Chen,L., Henning, T., Klar, P.J.,Theophel, K., Schnell, S., and Bunge, M., (2013). Effects of silver nanoparticles on microbial growth dynamics. J. Appl. Microbiol.,114, 25-35.

[33] Searle, I.L. and de Hoog, M.,(1991). Cloud seeding operations in central Tasmania 1990.Hydro-electric Commission, Water Resources Department.

[34] Langmuir, I.,(1953). Analysis of the effects of periodic seeding of the atmosphere with silver iodide.Part II Final Report of Project Cirrus, 340pp.

[35] Langmuir, I., (1955), Widespread control of weather by silver iodide seeding. GE Research Laboratory Report 55-RL-1236.

[36] Byers, H.R., (1974). History of weather modification.In "Weather and climate modification", 3-44.Ed. Hess, W.N., John Wiley & Sons, New York.

Figures:

1: Location of area cloud seeding experiments in south-east Australia, 1955-1992.Target areas are labelled T for single target experiments or N and S for crossover experiments. Unseeded control areas are labelled C.

2: Mean monthly concentrations of IN at -15^{0}C in two areas of each of the New England and Warragamba experiments and in a Tasmanian non-randomised cloud seeding operation.

3:Fig.3: In the Snowy Mountains experiment, E measured by the double ratio decreased substantially as the number of hours of seeding per year increased. E used period totals, not just seeded days.

4: Comparison of mean (T/C_{seeded} /$T/C_{unseeded}$) as a function of CSI at intervals of 500 for the Snowy Mountains experiment of 1955-1959.

5: Target/Northwest control precipitation on days suitable for seeding (**ss**), divided by the same ratio for days suitable for seeding but unseeded (**su**), compared with the cumulative seeding index. This measure of E refers to individual days only, not to period totals.

6. Fig.6. Mean (Target/Control) on each day 1 to 30 following seeding divided by the mean T/C for the preceding 30 days. South African experiment: red, Australian experiments: blue. Note the different scales.

7: Target areas of four early cloud seeding experiments shown by solid lines in a and c, dashed lines in b and d. The diagrams show contours of the ratio of mean rainfalls in seeded months to the means in corresponding months of seven unseeded years. The distribution of wind directions on seeded days are shown as dashed lines in **a** and**c**. Red areas have ratios>1.2.