

1 **RESPONSE OF NATURAL ICE NUCLEI TO DEPOSITED SILVER IODIDE.**

2

3

4 **ABSTRACT**

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

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

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

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

23

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

25

26 **1. INTRODUCTION**

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

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

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

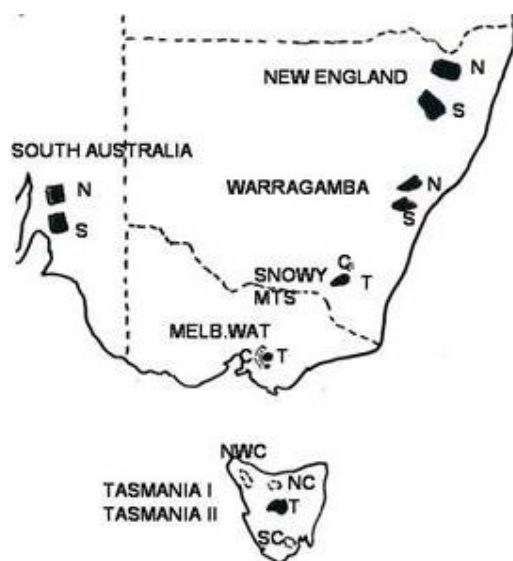
63 **2. Protracted increases in IN concentrations or precipitation following the release of AgI**

64 A smoke consisting of very small silver iodide (AgI) particles has been found [14] to be capable of
65 transforming water clouds to ice crystals at temperatures where most natural clouds contained no ice
66 crystals. This discovery opened up the way to practical cloud seeding aimed at increasing
67 precipitation. Before the development of suitable methods for detecting the presence of ice crystals in
68 clouds (about 1970) it was usually assumed that in the absence of obvious glaciation a cloud
69 contained insufficient ice crystals for optimum precipitation efficiency. Randomized cloud seeding
70 experiments began in Australia in 1955 using "seed/no seed" periods for estimating effects relative to
71 an unseeded control. Analysis of these experiments assumed that the AgI particles would be

72 deposited on the ground, de-activated by exposure to UV, or leave the area of interest within 24
73 hours.

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

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



90

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

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

98 The IN measurements used the filter method [22] in which air totalling 300l was drawn through a
99 membrane filter at a slow rate for 24 hours. The filter was then cooled to -15°C , the humidity raised to
100 water saturation and the ice crystals that grew were counted after 20 minutes. As an absolute
101 measure of IN concentrations the method has some shortcomings. It detects only nucleation by
102 deposition from the vapour correctly, the more efficient nucleation in the immersion and contact
103 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 seeding then showed the trends seen in figure 2.

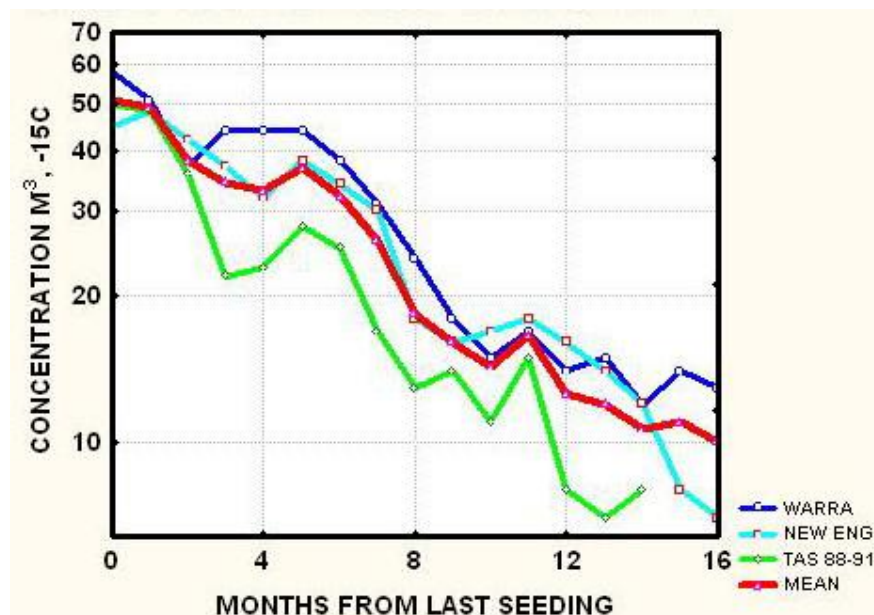


Fig.2: Mean monthly concentrations of IN at -15°C in two areas of each of the New England and Warragamba experiments and in a Tasmanian non-randomised cloud seeding operation.

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 two type 1 experiments was estimated from the double ratio:

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

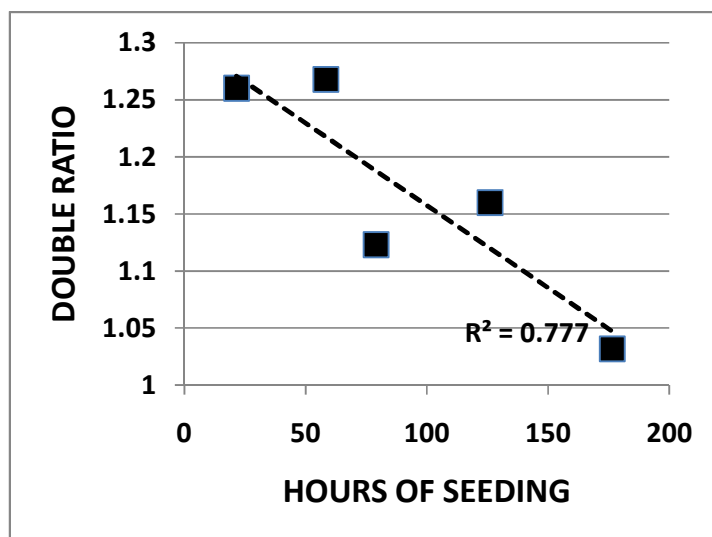
The remaining experiments used two target areas only one of which was seeded in any period, the other acting as a control. Limited randomization decided whether the north area (precipitation N) or the south (precipitation S) was to be seeded. Period lengths were about the same as in type 1 except

133 for Warragamba where it was daily. The effect of seeding for these type 2 “crossover” experiments
134 was calculated from:

135
$$E = \sqrt{\{\sum(N/S)_{\text{north seeded}} / \sum(N/S)_{\text{south seeded}}\}} \dots (2)$$

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

141 The simplest demonstration of the effects of secondary IN generation is to compare the observed E
142 with the number of hours of seeding for each year of operation. This is shown in figure 3 for the
143 Snowy Mountains experiment. The apparent success of the early years led to the hope that increased
144 seeding would lead to an even better result, culminating in the very poor result in the last year. In the
145 Tasmanian experiment, the double ratio in the first year was 1.24 with only 23 h seeding, in the third
146 year 1.05 with 181h. In years 4 and 5, seeding hours were reduced to 128h and then 76h and the
147 double ratios in both were 1.11.



148
149 Fig.3: In the Snowy Mountains experiment, E measured by the double ratio decreased substantially
150 as the number of hours of seeding per year increased. E used period totals, not just seeded days.

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

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

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

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

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 d_i will be the “cumulative seeding index”:

$$CSI = N_0 \sum m_i \exp(-\alpha(d_i - d_0)).$$

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

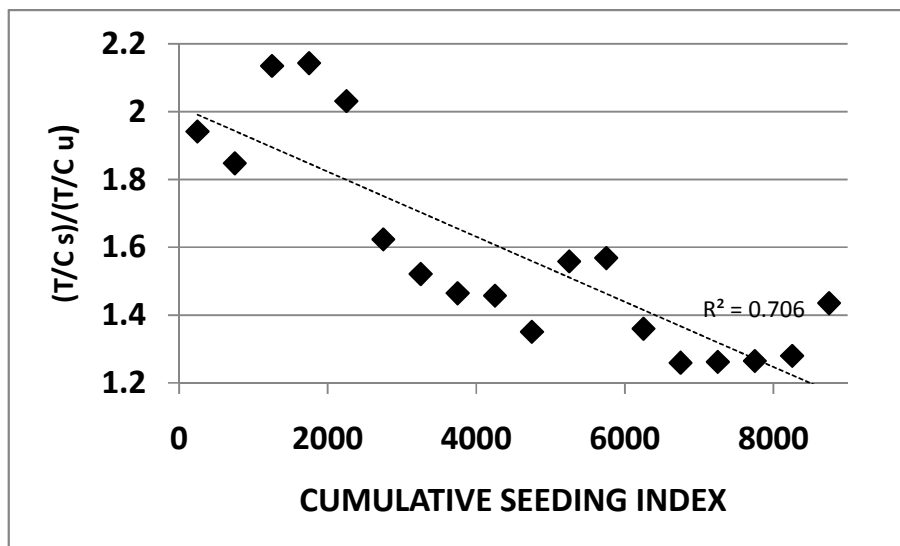
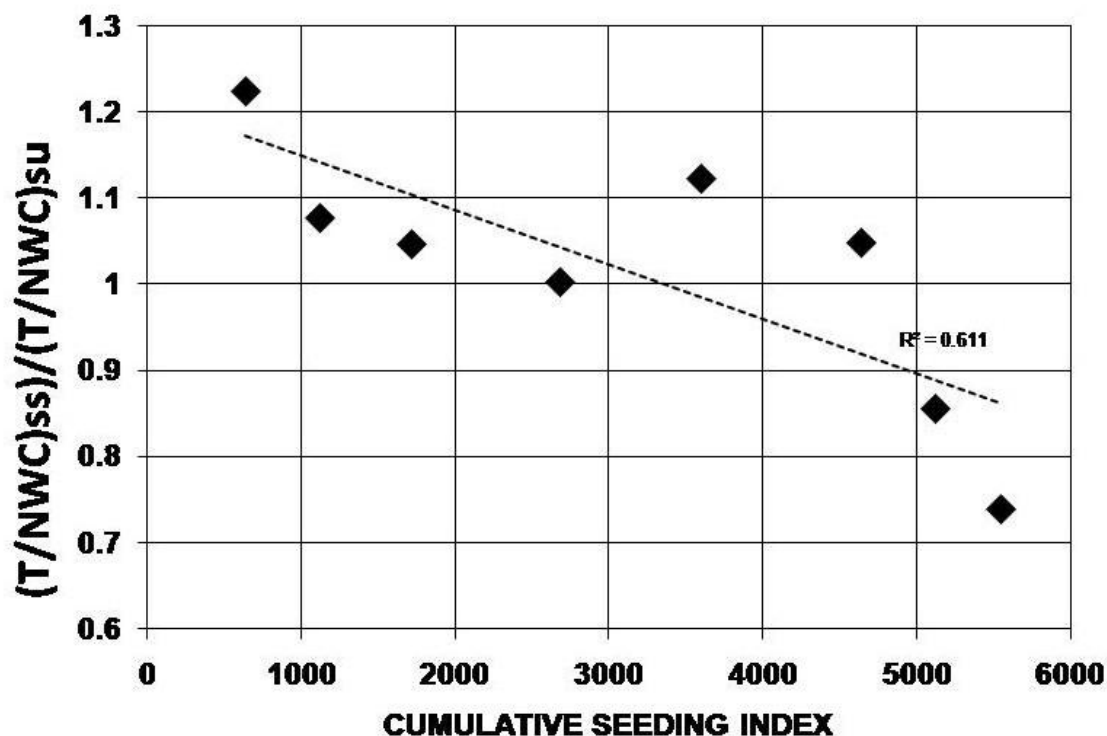


Fig.4: Comparison of mean $(T/C_{\text{seeded}} / T/C_{\text{unseeded}})$ as a function of CSI at intervals of 500 for the Snowy Mountains experiment of 1955-1959.

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 IN would 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) $(T/NWC)_{\text{seeded}} / (T/NWC)_{\text{unseeded}}$ 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.



187

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

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

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

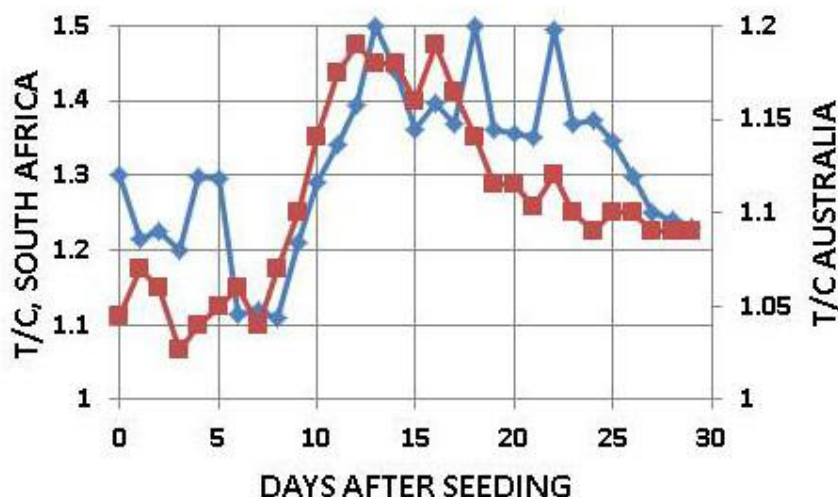


Fig.6. Mean (Target/Control) on each day 1 to 30 following seeding divided by the mean T/C for the preceding 30 days. South African experiment: red, Australian experiments: blue. Note the different scales.

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 with time 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/C on 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 3000km² 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 measurements of 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 2000km² and if the AgI was spread uniformly over it, the accumulated surface concentration would be only about

239 200 $\mu\text{g}\cdot\text{m}^{-2}$. In fact, much of the AgI would often have escaped precipitation within the targeted area
240 and been deposited further downwind.

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

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

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

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

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

279 **5. Assessing cloud seeding experiments when there are persistent effects of** 280 **seeding.**

281 Figs. 3-6 have shown how seriously misleading the statistically derived changes in rainfall due to
282 seeding with AgI can be because of secondary IN generation. How can the real effects be assessed?
283 There is one simple method that has been used in the operational seeding in Tasmania, described in
284 annual reports of the Water Resources Department of the Hydro-Electric Commission, e.g. [33]. It
285 involves expressing the total precipitation of all rainfall reporting sites in a large area around the target
286 for the whole seeding season (not just the seeded days) as a proportion of their average in common

unseeded years. In fact, one or more of 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 operated throughout 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 unpublished except 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.

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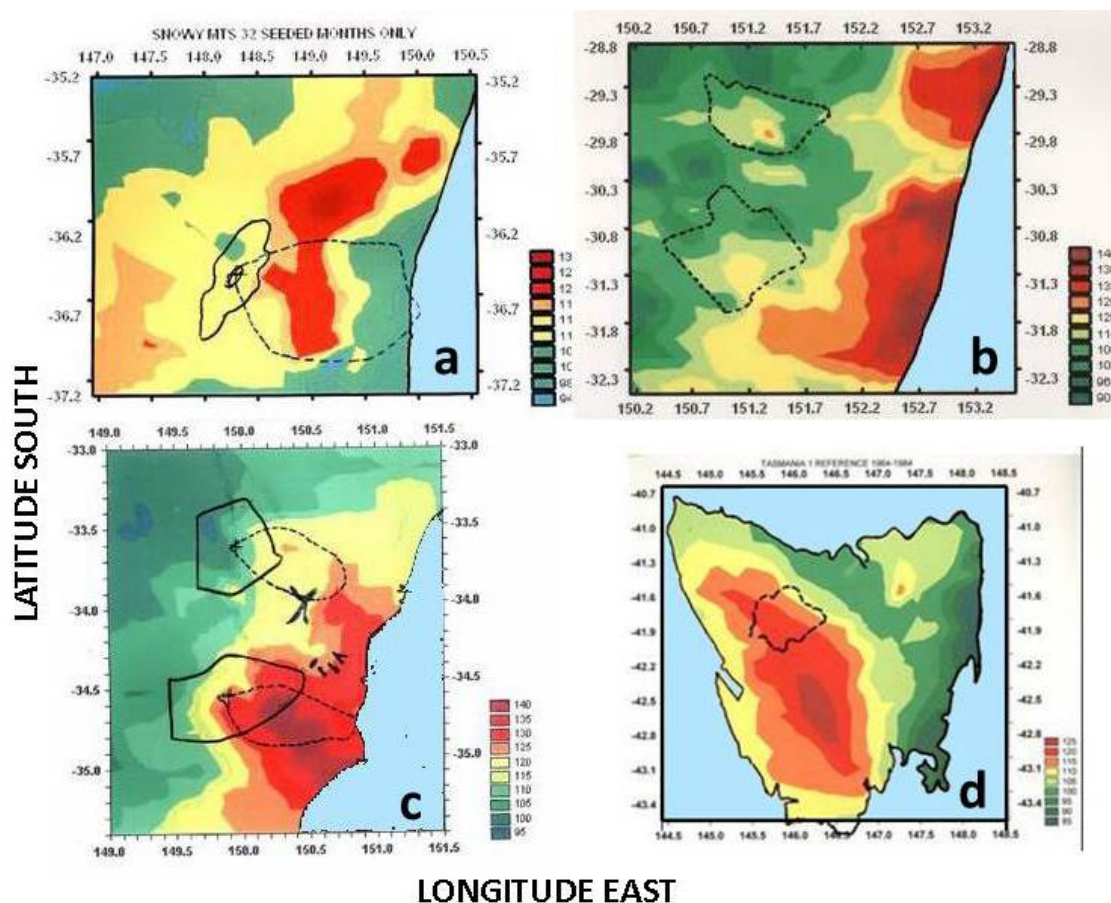


Fig.7: Target areas of four early cloud seeding experiments shown by solid lines in a and c, dashed lines in b and d. The diagrams show contours of the ratio of mean rainfalls in seeded months to the means in corresponding months of seven unseeded years. The distribution of wind directions on seeded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

The high ratios relatively far downwind from the AgI delivery areas in each figure suggest that rainfall was increased more effectively where deposition of AgI was relatively slight. The pattern of ratios is consistent with the hypothesis that toxic effects of AgI sometimes 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° rectangles 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 involved is 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 1m³ of air daily from each chamber were used to count IN active at -10°C 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 be either fungal spores, bacteria or parts of their membranes, or brochosomes (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 experiment but with an emphasis on microbiology should be considered.

Confirmation of the apparent far-downwind effects of AgI on rainfall seen in figure 7 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, although a 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 AgI will 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 areas through 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.

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478 **Figures:**

479 1: Location of area cloud seeding experiments in south-east Australia, 1955-1992. Target areas are
480 labelled T for single target experiments or N and S for crossover experiments. Unseeded control
481 areas are labelled C.

482 2: Mean monthly concentrations of IN at -15°C in two areas of each of the New England and
483 Warragamba experiments and in a Tasmanian non-randomised cloud seeding operation.

484 3: Fig.3: In the Snowy Mountains experiment, E measured by the double ratio decreased substantially
485 as the number of hours of seeding per year increased. E used period totals, not just seeded days.

486 4: Comparison of mean $(T/C_{\text{seeded}} / T/C_{\text{unseeded}})$ as a function of CSI at intervals of 500 for the Snowy
487 Mountains experiment of 1955-1959.

488 5: Target/Northwest control precipitation on days suitable for seeding (**ss**), divided by the same ratio
489 for days suitable for seeding but unseeded (**su**), compared with the cumulative seeding index. This
490 measure of E refers to individual days only, not to period totals.

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

494 7: Target areas of four early cloud seeding experiments shown by solid lines in a and c, dashed lines
495 in b and d. The diagrams show contours of the ratio of mean rainfalls in seeded months to the means
496 in corresponding months of seven unseeded years. The distribution of wind directions on seeded
497 days are shown as dashed lines in **a** and **c**. Red areas have ratios >1.2 .

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