A Characteristic of Dry Ice as a Glaciogenic Seeding Agent

by
Brian Jay Morrison

Department of Atmospheric Science
Colorado State University
Fort Collins, Colorado

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ABSTRACT

A CHARACTERIZATION OF DRY ICE AS A GLACIOGENIC SEEDING AGENT

Special apparatus was constructed and a series of experiments were designed utilizing the Colorado State University Cloud Simulation and Aerosol Laboratory 960 liter Isothermal Cloud Chamber to characterize dry ice as a glaciogenic cloud seeding agent under conditions that realistically simulate dry ice seeding. The primary objectives of the laboratory study were to establish an accurate effectiveness spectrum, determine the rates of ice crystal production and identify the primary mechanism of nucleation through the methodology of chemical kinetics.

In a secondary study, a simple numerical model simulating the fall and sublimation of cylindrical dry ice pellets through convective clouds was developed in an attempt to provide an independent estimate of dry ice effectiveness. The model was based on microphysical measurements of dry ice seeding curtains in cumulus congestus turrets over the eastern Transvaal of the Republic of South Africa using an instrumented Learjet

aircraft and on studies of the sublimation and fall characteristics of cylindrical dry ice pellets.

A complete effectiveness spectrum was obtained from the cloud chamber data for 5 mm diameter spherical dry ice pellets at a cloud liquid water content of 1.5 g m^{-3}. The results show a moderate temperature dependence from 2.2 x 10^{11} ice crystals produced per gram CO_2 sublimed at -2 ^oC to 8.9 x 10^{12} at -20 ^oC using one sublimation rate formula, and from 5.0 x 10^{11} to 1.9 x 10^{13} using another formula. There are indications that some overseeding may have occurred at temperatures as warm as -6 ^oC which could result in an underestimation of effectiveness at the colder temperatures.

The effectiveness values obtained from the numerical model using data from 17 cumulus clouds also imply a temperature dependence and are generally within an order of magnitude of the laboratory values. This lends a certain degree of confidence to the laboratory results. Model-calculated ice crystal concentrations of ice crystals in the dry ice seeding plumes suggest that the high concentrations frequently observed can be explained solely by dry ice nucleation.

The rates of ice crystal production from dry ice seeding are very fast. For 5 mm diameter pellets at a cloud liquid water content of 1.5 g m^{-3} they range from 90% of the counted ice crystals produced in 2.4 minutes at -2 ^oC to 1.3 minutes at -15 ^oC. This is much faster than most conventional glaciogenic aerosols at water saturation. The actual nucleation process occurs so quickly, that the rates of ice crystal production from dry ice seeding are determined by other factors such as particle growth and fallout.

Given that the rates of ice crystal production from dry ice seeding are not governed by the process of nucleation, the methodology of chemical kinetics to study nucleation and identify dominant nucleation mechanisms could not be used in this instance. The ice crystal yield data, however, clearly show that the predominant nucleation mechanism cannot be due to droplet freezing. It must rather be vapor dependent, most likely homogeneous condensation-freezing.

Brian Jay Morrison
Atmospheric Science Department
Colorado State University
Fort Collins, Colorado 80523
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I would like to thank Graeme Mather and Vic Whiteman of Cansas International Corporation for many constructive comments and encouragement. And, finally, I would like to thank Dave Emmitt and Michael Garstang of Simpson Weather Associates for providing the financial support to make this work possible.

DEDICATION

This thesis is dedicated

With love, to my wife, Margaret,
for her caring, friendship, and strength which has
nourished this work;

With joy, to my children, Gregory and Camilla,
who have shown me the new life and excitement in
each fresh day;

With gratitude, to my parents, Donald and Ellen Morrison,
for their support, wisdom, and sacrifices
that have made it all possible.

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CHAPTER I INTRODUCTION

Fundamental to ice-phase cloud seeding is the choice of glaciogenic seeding agent. The degree to which the functioning of a seeding agent is known and understood is crucial to the validity of any seeding hypothesis. In general the utility of glaciogenic agents has long been based primarily upon "effectiveness" defined in terms of ice crystals produced per unit mass of seeding material consumed. Recent research based on the application of chemical kinetics methodology to ice nucleation, however, has suggested that nucleation mechanisms and rates of ice crystal production are also of integral importance in the evaluation of a seeding agent.

The serendipitous discovery in 1946 by Vincent Schaeffer that a small amount of dry ice could produce massive numbers of ice crystals in supercooled water clouds is commonly recognized as the birth of modern weather modification. Since then, dry ice has been used as a seeding agent in weather modification operations throughout the world. Several laboratory and field studies have been conducted over the years to determine the effectiveness of dry ice. The various results have ranged from 10^9 to 10^{13} ice crystals per gram of dry ice sublimed spanning four orders of magnitude. Dry ice effectiveness is considered to be temperature independent, although only one study has seriously investigated this aspect. The wide range of results in characterizing dry ice is indicative of the difficulty in realistically simulating dry ice seeding in the laboratory and of the lack of controls and adequate description and measurement of cloud physical phenomena in the field.

The purpose of the study presented in this thesis is to experimentally characterize dry ice as a glaciogenic seeding agent for weather modification. This is done through laboratory investigations using the Colorado State University (CSU) Cloud Simulation and Aerosol Laboratory 960 liter Isothermal Cloud Chamber (ICC) and through numerical modeling based on aircraft observations of dry ice seeding curtains in supercooled convective clouds. This research is unique in three important respects: It is the first attempt to establish an accurate dry ice effectiveness spectrum utilizing the CSU ICC. Secondly, it is the first study to document the rates of ice crystal production by dry ice seeding and to assess their significance. And, thirdly, it is the first attempt to gain insight into the mechanism of nucleation from dry ice seeding using a chemical kinetics approach. The results from this study are intended to contribute to the scientific basis for the selection and utilization of dry ice in weather modification programs.

During the course of this research, portions of the results have been presented in Morrison et. al. (1984a and 1984b).

1.1 Primary Objectives

Three large cloud chamber facilities have been used routinely for the determination of glaciogenic aerosol effectivities. These are at the CSU Cloud Simulation and Aerosol Laboratory (Grant and Steele, 1966; Garvey, 1975), the Naval Weapons Center (Odencrantz, 1969), and the South Dakota School of Mines and Technology (Donnan et. al., 1971), of which only the CSU facility is still in operation. Large cloud chambers have been unique tools for the study of glaciogenic aerosols. This is because cloud properties can be accurately controlled while the large size minimizes wall losses and permits use of relatively large amounts of aerosol samples from actual field generators. Additionally, the CSU ICC has been shown to have an ice nuclei residence time scale longer than that required for most nucleation processes (DeMott et. al., 1983). This makes it possible to use the ICC as a chemical reactor for ice nucleation investigations using chemical kinetics methodology.

Modifications were made to the CSU ICC and special apparatus constructed to permit realistic, controlled seeding by spherical dry ice pellets. Experiments were designed to meet the following objectives:

1. Establish an accurate effectiveness spectrum for dry ice. Investigate the effects of differing cloud water contents, pellet sizes and velocities.
2. Determine the rates of ice crystal production.
3. Using the information on the rates of ice crystal production, apply the methodologies of chemical kinetics to gain insight into the mechanism (or mechanisms) of dry ice nucleation.

1.2 Secondary Objective

The usefulness of cloud chamber studies is limited without some indication of transferability to the "real world". Since 1981, a large and comprehensive database on dry ice seeding curtains in cumulus congestus clouds has been obtained in the course of the weather modification research program conducted in the eastern Transvaal of the Republic of South Africa. Additionally, the characteristics of the dry ice pellets used have been well studied and documented. Based on this data set, a simple numerical model was developed to simulate dry ice seeding for the following objective: Using the simulation based on actual field data, develop an independent estimate of dry ice effectiveness for comparison with the laboratory results.

CHAPTER II LITERATURE SURVEY

2.1 A Historical Note

The first use of dry ice in an attempt to modify the weather is often attributed to the work done by Schaeffer and Langmuir in 1946. Although they were the first to discover the glaciogenic property of dry ice (reported in Schaeffer, 1946), experiments in cloud modification using dry ice were apparently conducted in the early 1930's in Holland by August W. Veraart (Byers, 1974). Veraart made airborne releases of large quantities of dry ice with the idea of inducing precipitation by cooling air to form clouds or by increasing the amount of condensed water in existing clouds. Although Veraart claimed positive results, the scientific community was skeptical and nothing further came of his work.

2.2 Dry Ice Effectiveness

Despite optimistic early estimates that dry ice effectiveness could exceed 10^{16} or 10^{17} ice crystals per gram of dry ice sublimed (Schaeffer, 1946; Langmuir, 1948), laboratory and field studies have yielded lower figures. The results from the laboratory measurements of dry ice effectiveness are summarized in table 1 and the results of field measurements are summarized in table 2.

One of the earliest attempts to measure dry ice effectiveness was by Weickmann, Aufm Kampe, and Kelly (Weickmann, 1957). They dropped a dry ice pellet from the ceiling to the floor of a room-sized cold chamber containing a supercooled cloud at $-10\ ^{o}C$. The technique for counting ice crystals and determining mass of dry ice sublimed was not specified. An effectiveness of 10^9 at $-10\ ^{o}C$ was reported and they observed a large decrease in ice crystal concentration at warmer temperatures (-6 and -2 ^{o}C).

Eadie and Mee (1963) conducted tests to study the effect of dry ice pellet fall velocity on ice crystal production (described in section 2.4). From their experiments they computed an effectiveness of 10^{10} from -10 to -24 ^{o}C. However, they note that the concentration of ice crystals in their 2 liter cold box exceeded 2×10^4 cm^{-3} and they suggest this may represent an upper limit to the number of ice crystals that could be present due to competition for water vapor. This would result in an underestimation of effectiveness.

In an investigation of homogeneous nucleation due to the evaporative cooling of volatile liquids sprayed into a supercooled cloud in a cold box, Fukuta (1965) also made tests with dry ice for comparison purposes. He reports an

Table 1

LABORATORY MEASUREMENTS OF DRY ICE EFFECTIVENESS
BY OTHER INVESTIGATORS

Source	Apparatus	Num Trials	Effect	Temp (^oC)
Weickmann, 1957	room-size cold chamber	1	10^9	-10
Eadie and Mee, 1963	2 liter cold box	60	10^{10}	-10 to -24
Fukuta, 1965	cold box	--	10^{10} to 10^{11}	-10
Fukuta *et. al.*, 1971	795 liter out-door cold chamber	23	8×10^{11}	-2 to -12
Horn *et. al.*, 1982	re-analysis of Fukuta *et. al.* (1971)	23	8×10^{12}	-2 to -12

Table 2

FIELD ESTIMATES OF DRY ICE EFFECTIVENESS
BY OTHER INVESTIGATORS

Source	Num Seed Events	Effect	Temp (^oC)
Allee *et. al.*, 1972	2	7.0×10^{10} 7.5×10^{10}	-6.0 to -7.5 -8.5 to -10.8
Holroyd *et. al.*, 1978	17	2.0×10^{11} to 5.0×10^{11}	not given
Hobbs *et. al.*, 1978	*	3.0×10^{10} to 1.0×10^{11}	not given
Lawson, 1978	3	1.0×10^{12}	not given
Stith, 1984	6	9.8×10^{11} to 1.5×10^{13}	not given

* re-analysis of Holroyd *et. al.* (1978)

effectiveness of between 10^{10} and 10^{11} at -10 °C. No details are provided on the experimental approach used.

The most recent laboratory measurements of dry ice effectiveness are by Fukuta _et_. _el_. (1971). They dropped spherical dry ice pellets down a 40 m tall cylindrical football stadium steel chimney with a 795 liter observation chamber at the bottom. The experiments were done in the wintertime at ambient outdoor temperatures. Prior to the pellet drop a supercooled fog was injected into the observation chamber. After the pellet had fallen through it, the observation chamber was isolated from the chimney and fog reservoir and a small fan was activated to stir the contents of the chamber. Formvar replicator slides on the floor of the observation chamber were used to measure ice crystal production. Pellet velocities through the chamber were determined from stroboscopic photos. Sublimation rates were determined theoretically except for 0.9 cm diameter pellets which were measured by observing the mass loss after being swung on a string at a speed of about 10 m s^{-1} for a known length of time.

The experiments by Fukuta _et_. _al_. used spherical pellets ranging in size from 0.32 to 1.91 cm. The fall velocities of the pellets were from 52% of theoretical fall speed for the largest pellets to 92% for the smallest. They concluded that dry ice effectiveness is about 8 x 10^{11} ice crystals per gram of dry ice sublimed and is independent of temperature (-2 to -12 °C) and pellet size.

The results of Fukuta et. al. were questioned by Horn et. al. (1982) who found a mathematical error which increases the effectiveness value one order of magnitude to 8 x 10^{12}. Furthermore, Horn et. al. note that the supercooled cloud in the observation chamber was not replenished once the chamber was isolated after pellet transit. The average ice crystal concentration in the chamber was around 590 cm^{-3} and they suggest that additional ice embryos might have evaporated as a result of competition for the available water vapor. As a result, Horn et. al. conclude that dry ice effectiveness would be likely to exceed 10^{13}.

Allee et. al. (1972) presented one of the first attempts to estimate dry ice effectiveness from airborne observations of clouds after seeding. They seeded homogeneous supercooled stratus cloud decks over the upper Great Lakes region with dry ice at a known seeding rate. The maximum dimensions of glaciation determined from photogrammetry is taken to be the width and length of the seeding plume and its depth is assumed to extend down to cloud base. On the basis of theoretical work by Jiusto (1971) and their own ice crystal concentration measurements from a foil sampler, they assume that a minimum ice crystal concentration of 100 $liter^{-1}$ is required for the ice crystal plume to advance into the liquid water cloud. Therefore, knowing the approximate volume of the ice crystal seeding plume, the mass of dry ice used, and assuming an ice crystal concentration of 100 $liter^{-1}$ in the plume volume, they computed an effectiveness of 7.0×10^{10} for one cloud between -6 and -7.5 °C and an effectiveness of 7.5×10^{10} for another between -8.5 and -10.8 °C.

Holroyd _et_. _al_. (1978) used a similar approach on cumulus clouds in Australia and the USA. The supercooled cloud volume was estimated as a rectilinear solid with horizontal dimensions determined from the aircraft flight track and photography and the depth was taken to be the distance from cloud top to the freezing level. Ice crystal concentrations were measured with several types of airborne electronic particle counters and averaged along the flight path through the cloud over several penetrations. The mass of dry ice sublimed in the supercooled portion of the cloud was estimated in a numerical simulation to be 32%. The sublimation rates in the model were estimated using the empirical mass sublimation time equation from Mee and Eadie (1963) differentiated with respect to time. For comparison they also used the theoretical sublimation rate formula given by Fukuta _et_. _al_. (1971). From their data they computed a "conservative" effectiveness value of from 2 to 5×10^{11} using the Mee and Eadie sublimation formula. Alternative use of the Fukuta et. al. formula increases this result by a factor of 2.4.

Disagreement over the counting efficiency of one of the electronic ice particle counters used led Hobbs et. al. (1978) to suggest that the efficiency estimate of Holroyd et. al. be revised to from 3 x 10^{10} to 10^{11}.

In the study by Holroyd et. al., the ice crystals produced from the dry ice seeding are apparently assumed to be completely dispersed throughout the volume of the supercooled cloud. However, Lawson (1978) notes that the dispersion of a seeding plume can at least initially be described by a Gaussian distribution spreading laterally in time according to basic turbulent diffusion theory. On this basis he computes the expected plume diameter as a function of time based on turbulence measurements in seeded cumulus turrets during the High Plains Experiment (HIPLEX) in 1976 and 1977. In his computations the seeded cloud volume extends from seeding altitude at about $-10^{o}C$ down to the $-2^{o}C$ level. He concludes that dry ice effectiveness is on the order of 10^{12}.

The most recent field estimates of dry ice effectiveness are from Stith (1984). He examined the temporal variation of ice crystals in seven seeding curtains in a stable supercooled stratiform cloud. An average pellet sublimation rate of 3.6 x 10^{-3} g s^{-1} was estimated from pellet drop tests for a fall of 1890 m. Mean efficiencies of from 9.8 x 10^{11} to 1.5 x 10^{13} are reported depending upon the type of electronic probe used to count the ice crystals.

The lack of a temperature dependence for dry ice effectiveness is often mentioned in the literature, although the experiment by Fukuta et. al. (1971) has been the only systematic investigation made of this question. Some further evidence on this subject can be inferred from the experiments by Leonard and Detwiler (1983) which involved dropping cold objects into supercooled water clouds in a 96 liter cold box. They contend that cloud temperature has little effect on ice crystal production for metal spheres with surface temperatures colder than -60 °C (note that the equilibrium surface temperature of dry ice is -78.5 °C). Close scrutiny of their data, however, does in fact show a tendency for an order of magnitude increase in the number of ice crystals produced from the warmer (-5 to -8 °C) to the colder (-20 to -22 °C) cloud temperatures. We might expect to see a similar effect with dry ice pellets.

2.3 Nucleation Mechanism

Their appears to be some confusion in the literature about the predominant nucleation mechanism of dry ice. Vonnegut (1981) cites sixteen references which imply that freezing of existing cloud droplets, either by contact or homogeneous nucleation, is the primary mode of nucleation despite evidence to the contrary. It is pointed out by

Vonnegut and also in Horn et. al. (1982) that the maximum number of water droplets that could be frozen by dry ice pellets in cloud chamber experiments is much less than the actual number of ice crystals produced. Consequently, the predominant nucleation mechanism must be vapor dependent. Weickmann (1957) and Mason (1981) specifically identify dry ice nucleation as being homogeneous condensation-freezing.

It is interesting to note that in the experiments with cold spheres by Leonard and Detwiler (1983) mentioned in the previous section it was found that ice crystal production was predominantly due to cloud droplet freezing for metal sphere temperatures near and just below -40 ^{o}C. For colder temperatures spontaneous homogeneous nucleation from the vapor becomes predominant.

2.4 The Influence of Pellet Velocity on Effectiveness

Eadie and Mee (1963) investigated the effect of dry ice pellet velocity on ice crystal production by mounting a 1 cm diameter pellet onto a rotating arm connected to the shaft of a small motor which was briefly lowered into a 2 liter cold box. The pellets were tested at speeds of 0.8 m s^{-1} and 16 m s^{-1} moving in a horizontal circle about 1.5 inches in diameter. The supercooled cloud in the cold box was continuously humidified using a small hand atomizer until ice crystal fallout was complete. Ice crystal concentrations in the box were estimated visually through calibrated optics looking into the cold box and from photomicrographs taken of glass slides placed on the bottom of the box.

They found that at temperatures from -7 to -24 °C there was no difference in the ice crystal production between the "fast" and "slow" pellets. However, from -2 to -7 °C the ice crystal production of the slow pellets remained high while that for the fast pellets fell sharply to as much as four orders of magnitude less at -2 °C. They hypothesize that at the warmer temperature ice embryos do not remain in the supersaturated "field of influence" of the fast pellets for enough time to reach critical size. Conversely, at the slow speeds, ice embryos are in the low temperature environment for approximately 20 ms, sufficient for growth to critical size.

Fukuta et. al. (1971) take issue with both the experimental method and theoretical discussion of Eadie and Mee. They identify two possible sources of experimental error: First, the entrainment of warm air into the chamber along the motor shaft could be enough to cause the local temperature to exceed 0 °C at the warmer temperature runs causing some of the ice crystals to melt. Secondly, at the high speed they estimate that the pellet passes through the formed ice cloud about 130 times whereas at slower speeds there is more time for diffusion to enlarge the ice crystal cloud. From a theoretical point of view Fukuta _et_. _al_. contend that Eadie and Mee underestimated the ice embryo growth rate by an order of magnitude and also underestimated the time available for embryo growth in the region of pellet influence by an order of magnitude. Furthermore, Fukuta _et_. _al_. claim that their own experimental results show no velocity effect and they conclude that fall velocity effects are negligible for "normal" purposes.

2.5 The Application of Chemical Kinetics Methodology to Artificial Ice Nucleation Studies

The foundation of chemical kinetic theory is the Law of Mass Action (Guldberg and Waage, 1879) which states that the rate of a chemical reaction is proportional to the masses of the reacting substances. Changes in the rate of reaction to different reactant concentrations and reaction temperatures provide information which may permit deduction of the mechanism of reaction. Chemical kinetics is sometimes confused with non-equilibrium thermodynamics which can neither give information on the time required for a process nor the path the process follows between initial and final energy states.

It was in DeMott _et_. _al_. (1983) and DeMott (1982) that the application of chemical kinetic theory to the study of ice nucleation was first introduced. They demonstrated that the CSU 960 liter Isothermal Cloud Chamber (ICC) could be considered to be a chemical reactor where water vapor, cloud droplets, and ice nuclei are reactants and ice crystals are the products of the nucleation reaction. Changes in the rates of ice crystal production through the systematic variation of concentrations of the reactants can provide information to deduce the mechanisms of nucleation providing that the nucleation process governs the rates of reaction.

CHAPTER III EXPERIMENTAL APPROACH - ISOTHERMAL CLOUD CHAMBER STUDY

3.1 Laboratory Apparatus

The laboratory phase of this research uses the Isothermal Cloud Chamber (ICC) to establish the effectiveness of dry ice and the rates of ice crystal production from dry ice seeding. This is the first time the ICC has been used for the quantitative study of a non-aerosol nucleant. Special apparatus were built to simulate realistic seeding in the cloud chamber and new experimental techniques were developed.

3.1.1 The 960 Liter Isothermal Cloud Chamber

The Cloud Simulation and Aerosol Laboratory ICC has been described in the literature by Grant and Steele (1966), Garvey (1975), and, more recently, by DeMott et. al. (1983) and DeMott (1982). The ICC simulates a slowly settling supercooled stratus cloud. It has an effective volume of 960 liters, cloud temperature can be controlled from 0 to -25 ^{o}C, and cloud liquid water content from 0.3 to 3.0 g m^{-3}. The cloud is generated by atomization of distilled water in a commercial ultrasonic humidifier and is mixed with cold filtered air through a 10 cm diameter stand tube extending up into the chamber (figure 1). The cloud rises through the stand tube and spills out into the chamber. An array of 22 thermocouples monitors the temperature throughout the chamber and cloud density is calibrated against dewpoint from a Cambridge cooled-mirror type dewpoint hygrometer.

Measurements of the size distribution of cloud droplets by a Particle Measuring Systems Forward Scattering Spectrometer Probe (FSSP) at 0.5 and 1.5 g m^{-3} liquid water contents for several temperatures are shown in figure 2. Modal radii are consistently in the 3.0 to 3.5 micron size range and the spectra do not show much variation with temperature. Cloud droplet concentrations are shown in figure 3 as a function of cloud temperature. They are generally on the order of 4300 cm^{-3} at a liquid water content of 1.5 g m^{-3}.

3.1.2 Measurements of Ice Crystal Yield and Rate of Production

Ice crystal yields are measured by visually counting the number of crystals that fall onto standard microscope slides after injecting or exposing the cloud to a seeding agent. The slides are removed from a vertically stacked tray at regular intervals through a port in the chamber wall uncovering a fresh slide each time. They are then rapidly transferred to the microscope in a -20 ^{o}C cold box next to the chamber for ice crystal counting.

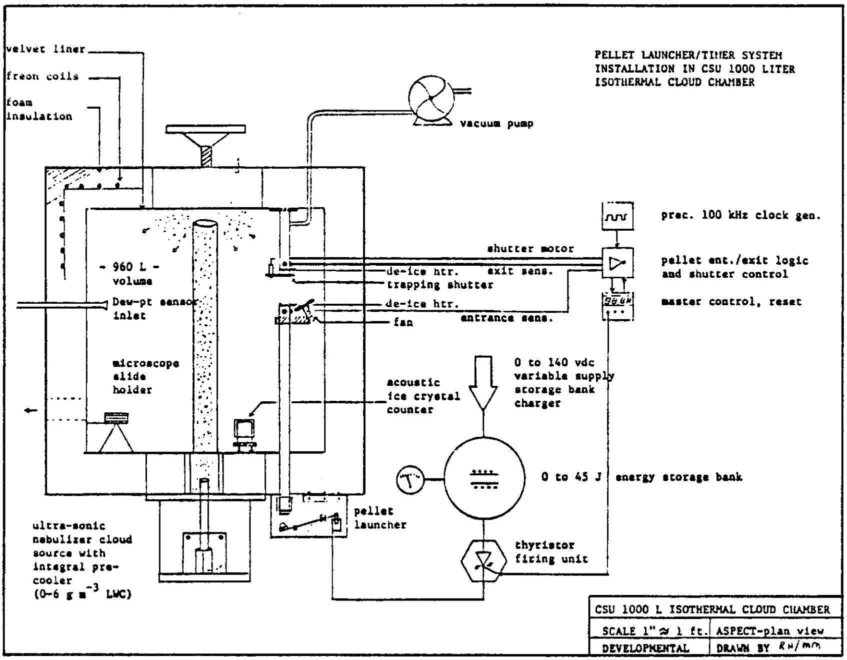

Figure 1. Schematic of 960 liter Isothermal Cloud Chamber and apparatus used in the dry ice pellet experiments (not to scale).

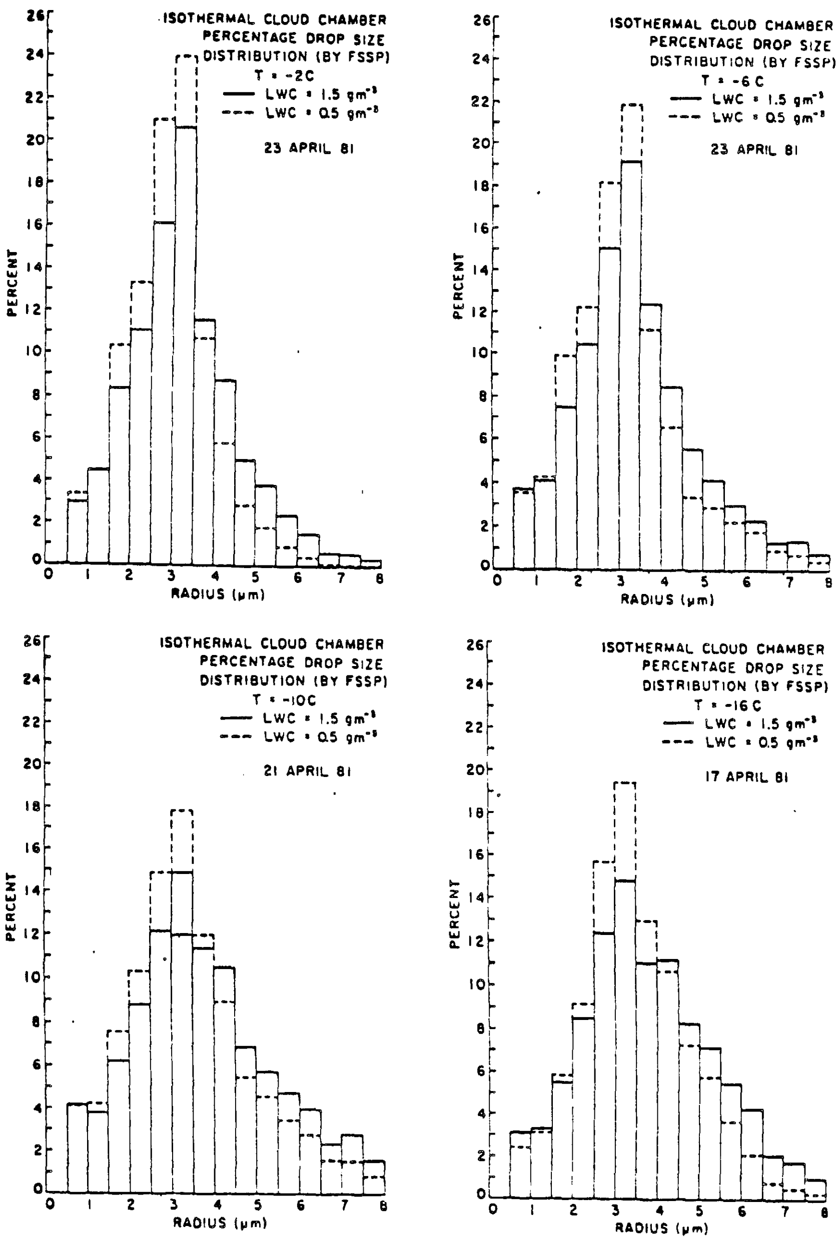

Figure 2. Forward Scattering Spectrometer Probe (FSSP) measurements of cloud chamber droplet size distributions for selected temperatures (from DeMott, 1982).

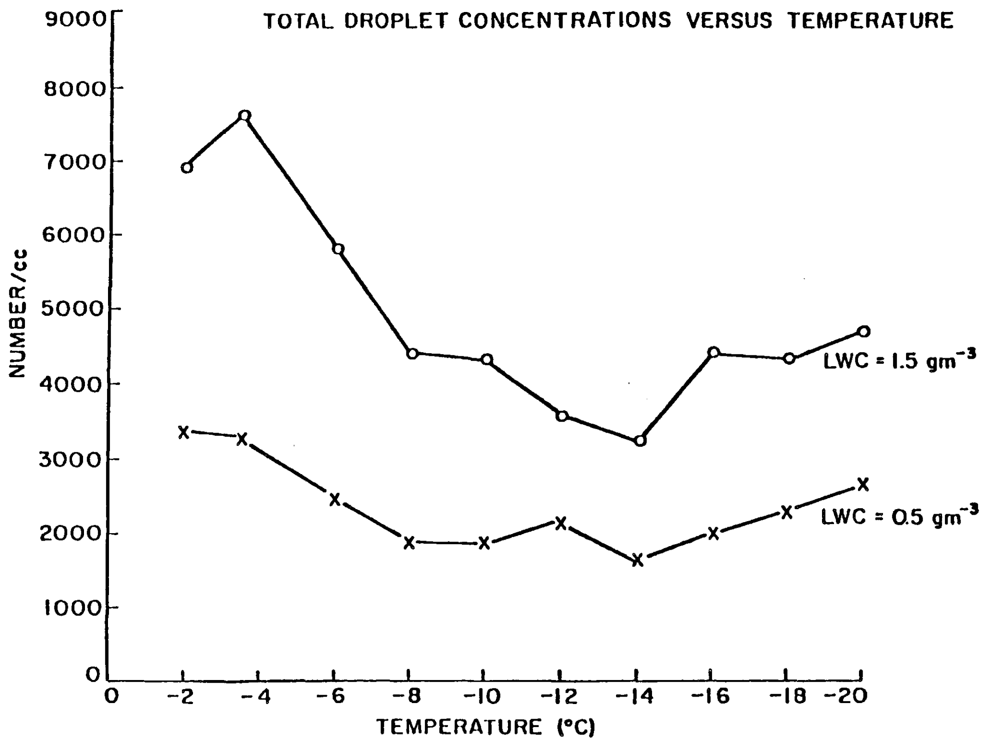

Figure 3. Isothermal Cloud Chamber cloud droplet concentrations as a function of chamber temperature and liquid water content (from DeMott, 1982).

At best, visual counting can provide ice crystal production rate measurements with a temporal resolution of 1.0 minute. This is insufficient for the dry ice experiments which had time scales of about 2.5 minutes. Therefore, a capillary-type acoustic ice crystal counter was used to provide production rate data at a 0.1 minute resolution. The acoustic counter was built at the Cloud Simulation and Aerosol Laboratory based on the concepts of Langer (1965). It senses ice crystals larger than about 20 microns at a rate of up to 200 Hz. The highest rates encountered in the dry ice experiments were about 175 Hz.

3.1.3 Dry Ice Pellet Manufacture

Catapult injection of dry ice pellets into the ICC necessitated the use of spherical pellets for ballistic reliability and accuracy. Molds for forming spherical pellets of 5, 7 and 11 mm diameters were made by pressing greased steel ball-bearings into cylindrical sections of solid aluminum bars using a ten-ton hydraulic press. These molds were then tapped and could be bolted into the jaws of a large shop vise. The resulting apparatus allowed homogeneous pellets to be quickly and easily pressed from small irregular chunks of dry ice prior to the start of each experiment.

3.1.4 Pellet Launcher and Catcher

Realistic "seeding" in the ICC was accomplished by shooting a spherical dry ice pellet into the chamber at nearly terminal velocity and then catching and isolating the pellet. The main requirements for a pellet "launcher" were that it should provide a smooth acceleration to minimize the risk of shattering the pellet, permit reproducible pellet speeds over a wide range up to and including terminal velocity, and be accurate enough to maintain consistent pellet trajectories. An electro-mechanical catapult system was built to meet these specifications (see figure 1). Spherical dry ice pellets are placed on a dimple in a small disk of rubber cemented to one end of a 36 cm Nickel-Silver lever arm. A pivot point was located at about 30 cm up the lever arm and the other end was attached to the moving element of a 12 volt DC Guardian-type open frame solenoid. This solenoid was overdriven by a controllable voltage (0 to 140 volts) from a rechargeable 4500 microfarad capacitor storage bank allowing for consistent accelerations at any given voltage. The catapult mechanism was mounted on a 6.4 mm aluminum plate and bolted underneath the chamber. A loose aluminum clip was used to hold the lever arm down prior to firing the solenoid.

Pellets were launched from the catapult into a plexiglas "entry" tube extending up into the chamber. They then traverse a portion of the cloud and enter a "catcher" consisting of a plexiglas tube hanging from the chamber roof with a brass plate motor-driven shutter installed at the open end. The aperture between the entry and trapping tubes was initially set for a 33.0 cm path length through the cloud, but, due to the occurrence of heavy overseeding, was reduced to 10.3 cm. A rubber cork was used to seal the bottom opening of the entrance tube before and after pellet launch.

Originally, dual small infrared photosensors were installed at the mouths of the entrance and trapping tubes to sense pellet transit. They would trigger a precision clock to measure the transit time of the pellet and energize the trapping shutter. Unfortunately, the photosensor system proved unreliable for sensing the smaller pellets probably due to an insufficient field of view. A major reconfiguration of this system was not possible and it was decided to override the photosensors and drive the trapping shutter with a variable delay timing system.

Lacking an operational photodetector system, speeds for the smaller pellets were determined from stroboscopic photos of pellet transits across the aperture between the entry and trapping tubes over several trials. Speeds for the large 11 mm pellets were, however, successfully obtained from the photosensor system. The results from both methods are summarized in table 3 and figure 4.

Included in table 3 is the Reynolds Number computed for each pellet diameter and speed.

Table 3

PELLET SPEED RESULTS

Pellet Diameter (mm)	Number of Successful Trials	Avg Solenoid Voltage (V DC)	Avg Pellet Speed ($m\ s^{-1}$)	Cloud Exposure Time (ms)	Reynolds Number
5	8	121	12.3	8.3	3420
5	2	105	11.8	8.6	3280
5	3	90	11.1	9.2	3080
5	7	80	9.8	10.4	2720
5	4	66	7.7	13.2	2140
5	5	56	6.5	15.7	1800
5	2	50	5.7	17.9	1580
5	5	45	4.7	21.7	1310
7	4	122	12.4	8.2	4820
7	3	111	12.0	8.5	4670
7	6	91	10.7	9.5	4160
7	3	81	9.8	10.4	3810
7	3	76	9.0	11.3	3500
11	35	120	11.9	8.6	7270

The terminal velocity for a dry ice sphere can be formulated in the following manner. The drag force exerted on a sphere moving through a viscous fluid is:

$$F_D = \frac{\pi}{8} d^2\ u^2\ \rho_a\ C_D \qquad (3.1)$$

where d = diameter (cm)

u = velocity ($cm\ s^{-1}$)

ρ_a = fluid density ($g\ cm^{-3}$)

C_D = drag coefficient

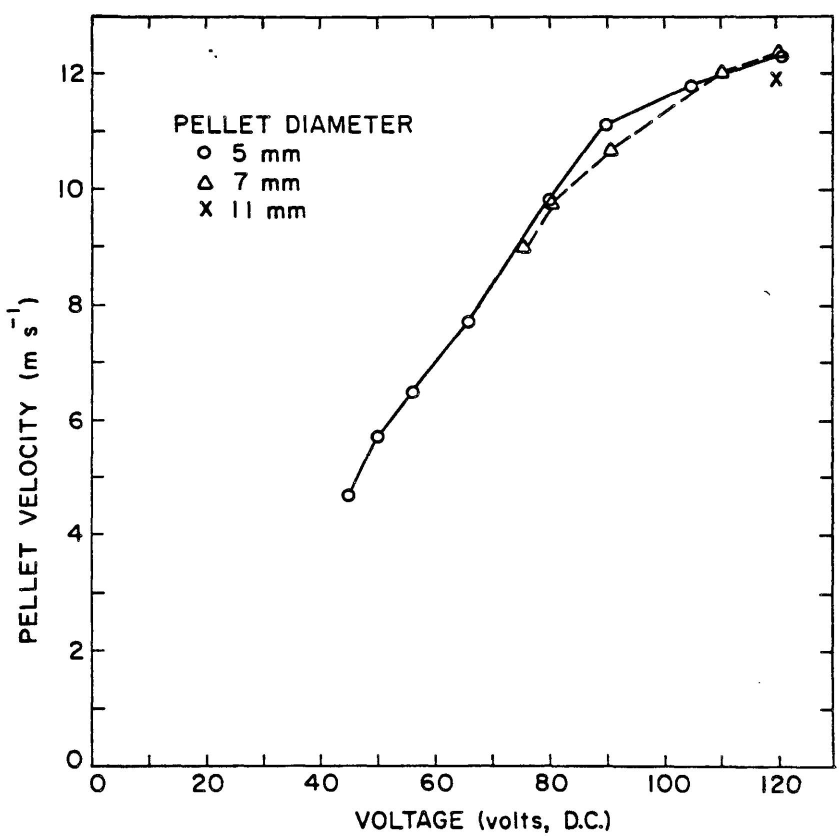

Figure 4. Pellet speed calibration curves as a function of solenoid voltage for three different pellet sizes.

The gravitational force on the sphere is:

$$F_g = \frac{\pi}{6} d^3 g (\rho - \rho_a) \tag{3.2}$$

where g = acceleration of gravity (981 cm s^{-2})

ρ = density of dry ice (1.52 g cm^{-3})

At terminal velocity $F_D = F_g$. Therefore, by equating the two equations above, noting that $\rho_a << \rho$ and solving for u, we get:

$$u_T = \left[\frac{4}{3} \frac{d\ g\ \rho}{\rho_a\ C_D} \right]^{0.5} \tag{3.3}$$

At sufficiently high Reynolds Numbers C_D for spheres becomes constant at about 0.45 (Rogers, 1979). Using the above formula, the terminal velocities for our pellets are computed and compared against the velocities obtained with the launcher in table 4.

Before construction of the launcher, the solenoid was tested and found to provide enough force to accelerate an 11 mm pellet to terminal velocity. Unfortunately, in operation there was apparently a significant loss of energy through flexing of the lever arm so that, at best, the 11 mm pellets could reach only 54% of terminal velocity. The smaller 5 mm pellets, however, do much better to within 83%. Replacement with an improved system was not done as it would have required a major reconfiguration of the launcher and added further delays

Table 4

COMPARISON OF PELLET SPEEDS WITH TERMINAL VELOCITIES

Pellet Diameter (mm)	Avg Solenoid Voltage (Volts, D.C.)	Avg Pellet Speed ($m\ s^{-1}$)	Terminal Velocity ($m\ s^{-1}$)	Percent of Terminal Velocity
5	121	12.3	14.8	83
5	105	11.8	14.8	80
5	90	11.1	14.8	75
5	80	9.8	14.8	66
5	66	7.7	14.8	52
5	56	6.5	14.8	44
5	50	5.7	14.8	39
5	45	4.7	14.8	32
7	122	12.4	17.5	71
7	111	12.0	17.5	69
7	91	10.7	17.5	61
7	81	9.8	17.5	56
7	76	9.0	17.5	51
11	120	11.9	21.9	54

to the experiments. The 5 mm pellets are close enough to terminal velocity for experimental purposes.

The accuracy of the launcher was marginal with less than about 50% of the shots on target. The remainder went off track and smashed against the trapping tube. This resulted in a major slowdown of the experimentation as the time scale for the chamber to return to equilibrium was on the order of 25 minutes after a "miss" compared to 5 minutes for a successful run. A more reliable launcher should be built for any future experimentation.

During the initial experiments it was determined that ice embryos were probably "leaking" out of the shutter due to an imperfect seal and vibration of the shutter as the trapped pellet sublimes on the shutter surface. A small vacuum pump was hooked up to a CO_2 venting hose already connected to the trapping tube and operated with a 6 liter min^{-1} airflow. This supplied enough negative pressure in the trapping tube when the shutter was closed to effectively seal the trapping tube. The pump was activated just prior to launch and kept in operation long enough for the pellet to completely sublime.

3.1.5 Mixing and Dispersion of Ice Embryos

The concentration of ice embryos in the dry ice pellet plume is very high, on the order of 3×10^7 cm^{-3} ($T = -10^{\circ}C$, $LWC = 1.5$ g m^{-3}). In the absence of turbulent mixing the plume will only gradually diffuse radially outwards and ice embryo losses due to aggregation will occur. To minimize this effect a "stirrer" was made by cementing a low-mass model airplane propellor to the shaft of a small DC motor which was then strapped next to the outlet of the entrance tube. The stirrer is activated a split second after pellet launch to mix the ice embryos uniformly across the chamber.

Mixing tests were run to determine the best length of time to operate the stirrer so that the cloud is well mixed after injection of a 5 mm pellet without undue wall losses of ice crystals. The results shown in figure 5 indicate that the stirrer must be operated for a minimum

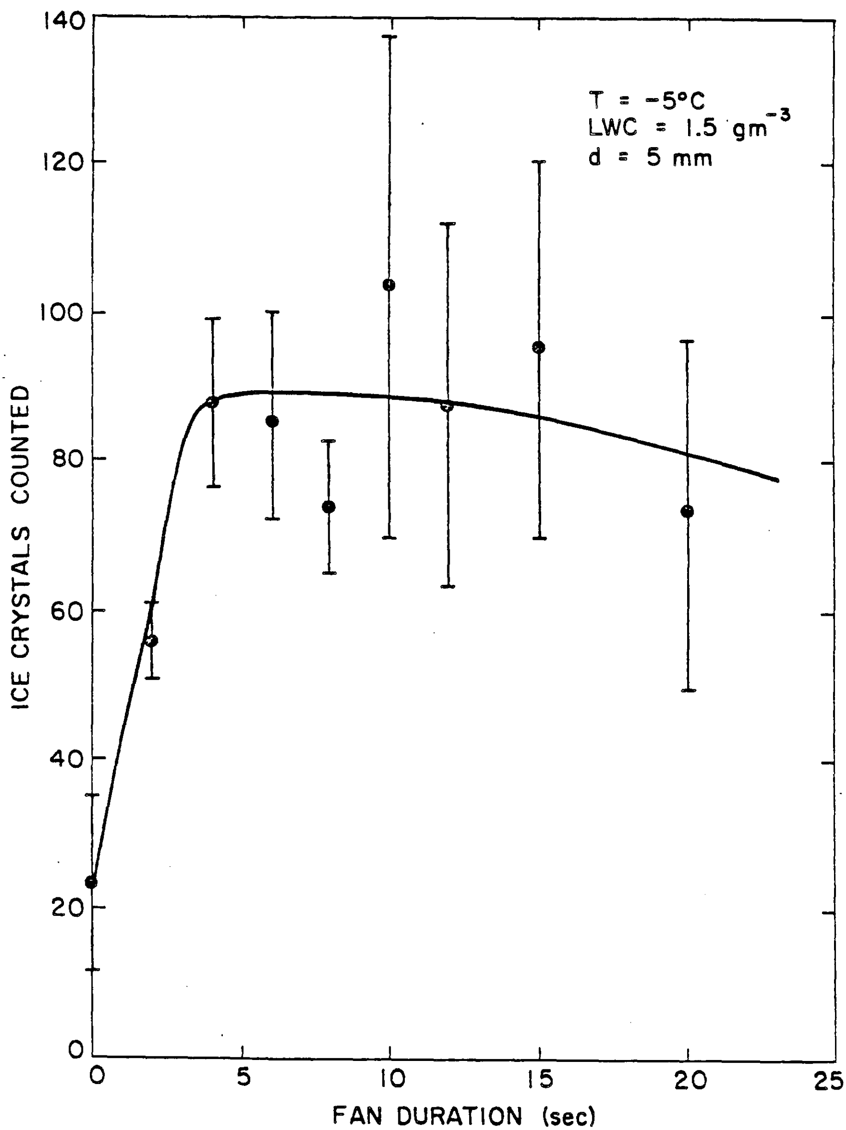

Figure 5. Ice embryo mixing test.

of 4 seconds before the ice crystal count reaches a maximum and levels off. An apparent downward trend after 12 seconds may indicate occurrence of wall losses. Throughout all the experiments the stirrer was operated for 4 to 5 seconds immediately after launch.

The potential loss of ice embryos due to aggregation in the plume was demonstrated in another series of tests where the stirrer activation was delayed over increasing time intervals after pellet launch (figure 6). The decrease in ice crystal yield is very pronounced within a second or less. Ideally, it would be optimal to start the stirrer just prior to pellet launch to ensure immediate dispersion of the ice embryos, but the smaller pellets are blown off trajectory by the stirrer air flow.

3.2 Experimental Procedure

Once the cloud temperature and liquid water content come to equilibrium at the desired values, a rheostat on the control box is reset (if necessary) to charge the capacitor bank to the desired voltage for firing the solenoid (usually 121 volts). The trapping tube shutter is opened, electronics reset, and the trapping tube vacuum pump started. A pellet is then pressed from a small chunk of dry ice and rapidly placed onto the launcher. A microscope slide is removed from the stack in the chamber to uncover a fresh slide and the acoustic ice crystal counter strip chart recorder is turned on and zeroed. The

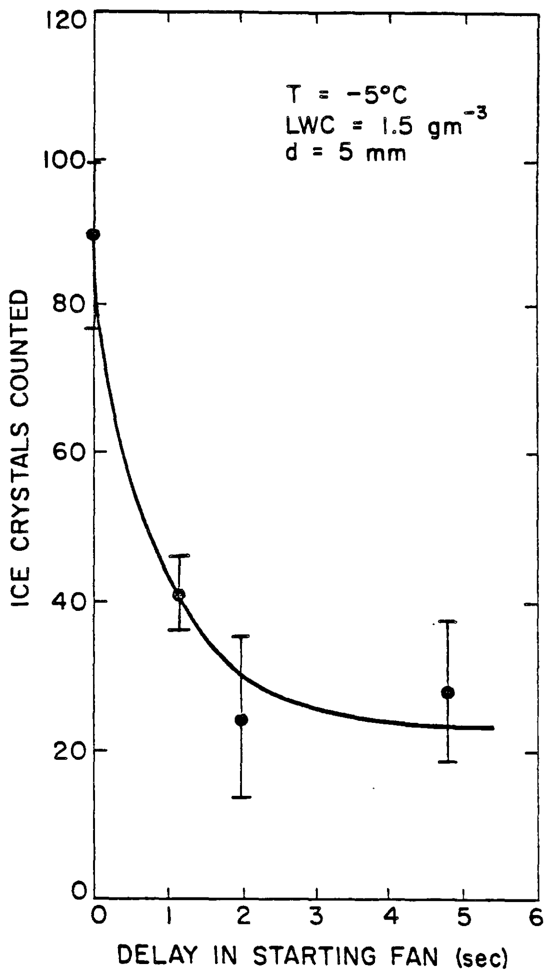

Figure 6. Test for aggregation of ice embryos in the dry ice pellet plume.

solenoid is then fired launching the pellet into the chamber while the stirrer and a stopwatch are simultaneously activated.

Microscope slides are removed from the chamber at one minute intervals and ice crystals counted using the microscope in the cold box. Typically, ice crystals are counted in five separate viewing areas on the slide and the average is taken to give the mean number of ice crystals collected per viewing area. Slides are continually pulled until the ice crystal flux becomes negligible.

3.3 Spherical Dry Ice Pellet Sublimation Rate

Mee and Eadie (1963) give the following empirical expression for the time required for the complete sublimation of a spherical dry ice pellet falling at terminal velocity as a function of initial pellet mass:

$$t = 153\ m^{0.42}\ \frac{T_s - 25}{T_s - T_a} \qquad (3.4)$$

where t = time (s)

m = initial pellet mass (g)

T_s = pellet surface temperature (^oC)

T_a = ambient air temperature (^oC)

A pellet surface temperature of $-78.5\ ^oC$ was assumed and average pellet density was measured to be $1.4\ g\ cm^{-3}$. The equation was based on vertical wind tunnel studies of pellet sublimation in clear air at terminal velocity. Holroyd *et. al.* (1978) solved the above equation for mass and differentiated with respect to time to obtain an expression for sublimation rate:

$$\frac{dm}{dt} = -1.556\times10^{-2}\, m^{0.58}\, \frac{T_s - T_a}{T_s - 25} \tag{3.5}$$

Fukuta *et. al.* (1971) also proposed a sublimation rate for spherical pellets falling at terminal velocity that is derived from theory:

$$\frac{dm}{dt} = -\,(8.16\times10^{-8}\, d\,/\,P)\,(0.097\, T_a + 9.4\, P + 137.3)\,(0.097\, T_a + 9.4\, P - 25.4)\,(1 + 6.66\times10^{2}\, d^{3/4}\, P^{1/4}\, T_a^{-5/12}\,(T_a - 125.9)^{-1/6}) \tag{3.6}$$

where T_a = ambient air temperature (K)

P = ambient air pressure (atmospheres)

d = spherical pellet diameter (cm)

A pellet density of 1.56 g cm^{-3} was assumed. The pellet surface temperature was taken to be -100 ^{o}C as a consequence of ventilation.

The question of dry ice pellet surface temperature has recently been experimentally addressed. An icing wind tunnel study by Kochtubajda and Lozowski (1985) produced results showing that the ventilated equilibrium temperature of cylindrical dry ice pellets at terminal velocity does indeed drop to nearly -100 °C. Similar experiments by G.D. Emmitt of Simpson Weather Associates in Charlottesville, Virginia (personal communication, 1985), however, have detected no drop in surface temperature at all.

We are then faced with the dilemma as to which sublimation rate formula to use. Figure 7 shows that the difference between the two formulae for 5 mm diameter pellets is a factor of two or more over the experimental range. Unfortunately, an independent sublimation rate study was beyond the scope, time and resources available for this research. Lacking any reasonable justification for selecting one formula over the other, both have been used in this study together.

3.4 Effectiveness Computation

Effectiveness, E, in terms of ice crystals produced per gram of dry ice sublimed, and ice crystal yield, Y, are computed by:

$$E = \frac{Y}{m_s} \tag{3.7}$$

where Y = total ice crystal yield in the chamber

m_s = mass of dry ice sublimed during pellet transit across the aperture

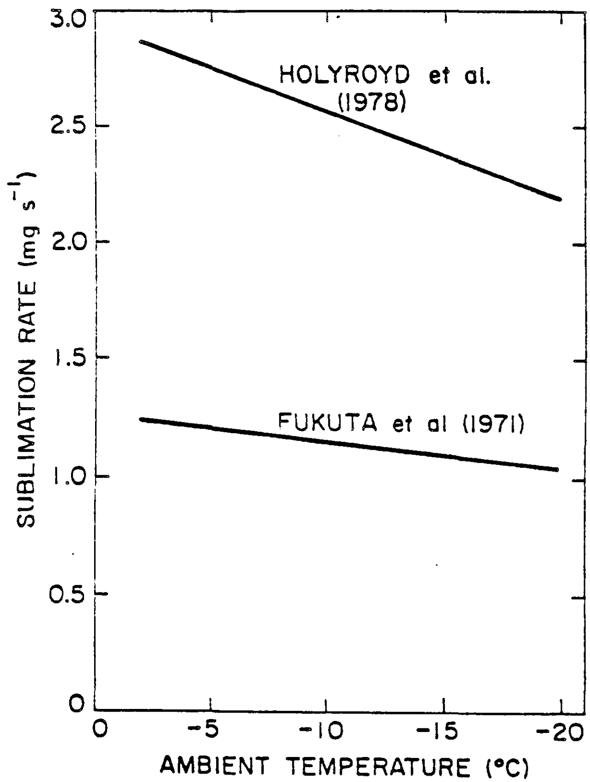

Figure 7. Sublimation rates for 5 mm diameter spherical dry ice pellets using two different sublimation rate equations.

$$Y = N_{IC} \frac{A_c}{A_v} \qquad (3.8)$$

where N_{IC} = mean number of ice crystals counted per slide viewing area

A_c = chamber cross-sectional area (8.35×10^3 cm^2)

A_v = microscope viewing area (2.01×10^{-2} cm^2)

The mass of dry ice sublimed is calculated by multiplying the sublimation rate of the dry ice pellet by the time it takes for pellet transit across the 10.2 cm aperture. The need to correct effectiveness values due to chamber airflow losses of ice nuclei has been discussed in DeMott _et_. _al_. (1983). The rate of ice crystal production from dry ice is so rapid, however, that there is little time for airflow losses to occur and the correction can therefore be neglected.

3.5 Composite Kinetics Plots

The measured rates of ice crystal production are best illustrated and analyzed in the form of kinetics plots (DeMott _et_. _al_., 1983). In these plots the total number of ice crystals sensed by the acoustic counter is taken to be the total number of viable ice embryos present after pellet transit through the cloud at time t = 0. Then the depletion of these embryos as they grow and fall out is plotted from the ice crystal production data (from the acoustic counter strip charts) on a natural log scale as a function of time. The rates of ice crystal production (or, alternatively, viable ice embryo depletion) are proportional to the slope of the plot. An example of the kinetics plots for a "family" of several dry ice seeding runs at the same cloud temperature and liquid water content is shown in figure 8. The time resolution is 0.1 minutes.

In order to obtain a representative kinetics plot for the same set of cloud (temperature, liquid water content) and seeding (pellet size, velocity) conditions, each "family" of plots is averaged together to form a composite plot (figure 9). Initially this was done graphically but did not provide enough accuracy. Therefore, the ice crystal count for each run every 0.1 minutes was arithmetically averaged. The average total count at t = 0 is taken as 100% on the natural log scale (ln 100% = 4.605) and the percentage depletion of viable embryos is plotted against time.

The slopes of the kinetics plots are also subject to correction for airflow losses of ice embryos. However, as already mentioned, the dry ice reaction is so rapid that airflow losses are negligible. For instance, the slope of kinetics plots for 5 mm pellets ($v = 12.3$ m s^{-1}, LWC = 1.5 g m^{-3}) at a cloud temperature of -20 °C would decrease by 4.3% and at -6 °C by only about 1.5%.

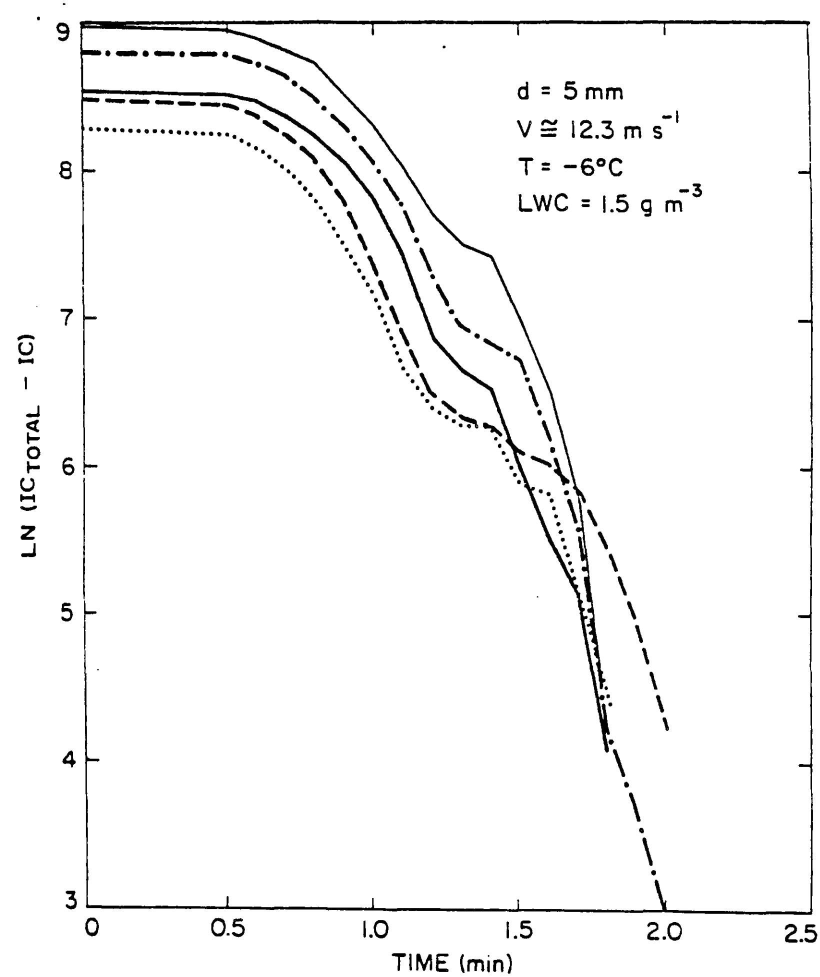

Figure 8. Example of the kinetics plots for a "family" of several dry ice tests at the same cloud and seeding conditions.

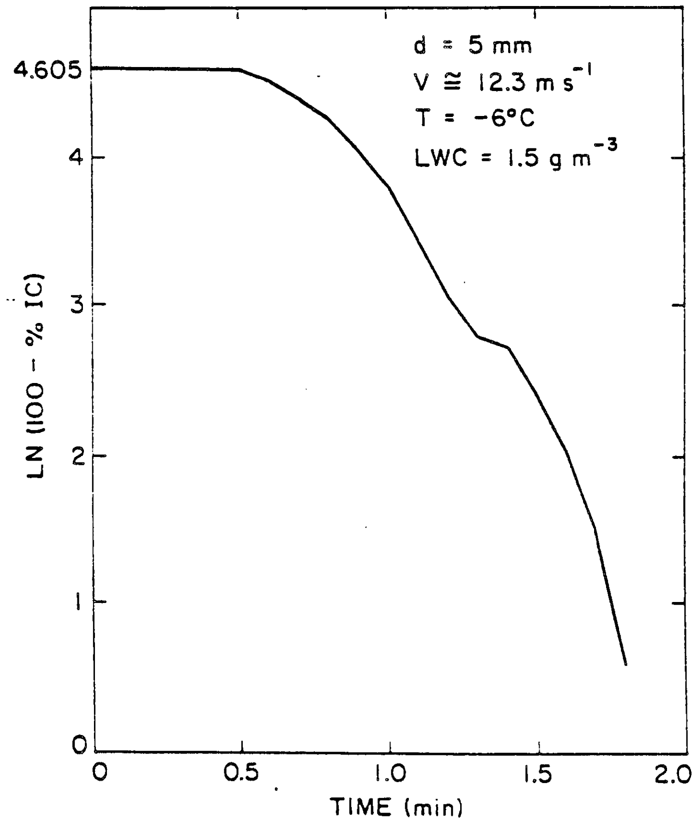

Figure 9. Composite kinetics plot formed from the plots in figure 8.

IV EXPERIMENTAL APPROACH - NUMERICAL MODEL

A simple computer model was developed in an attempt to provide an independent estimate of dry ice effectiveness based on aircraft observations of dry ice seeding curtains for comparison with the laboratory results. The model tracks the fall and sublimation of a cylindrical dry ice pellet dropped into a cumulus congestus turret and the vertical advection of its products over two second time steps. It estimates the nucleation temperature of the products in the portion of the dry ice plume sampled by the aircraft and computes the sample volume. Given the ice crystal concentrations in the portion of the plume sampled by the aircraft, the model can compute observed effectiveness values as a function of nucleation temperature. Conversely, by using the laboratory derived effectiveness spectrum, the model can predict the ice crystal concentration in the portion of the seeding curtain sampled by the aircraft.

4.1 Dry Ice Seeding Curtain Data

Aircraft measurements have been made with an instrumented Learjet 24 by Cansas International Corporation (Pty) Limited since 1978 as part of a weather modification research program in the eastern Transvaal of the Republic of South Africa sponsored by the South African Water Research Commission (Morrison et. al., 1985 and 1986; Garstang et. al., 1981). The data used in the numerical model are from 17 cumulus congestus cloud turrets which were penetrated and seeded with dry ice at temperatures of between -9 and -18 ^{o}C. After seeding they were then repenetrated at near-orthogonal angles to the initial penetration a few minutes later to sample the seeding curtain. The data are summarized in table 5 which shows the following parameters for the treatment pass (cloud not previously penetrated) and the subsequent repenetration: time of penetration, aircraft true air speed (TAS), ambient air pressure and temperature, time in cloud, average turbulent dissipation coefficient through the cloud, average updraft speed, and the average and maximum cloud particle concentrations. The turbulent dissipation coefficient is computed from a fourier transformation of the normalized TAS time series. The cloud particle concentrations are computed using data from a Particle Measuring Systems 2D-C optical array probe (35 micron resolution) processed using the particle-weighting technique with standard artifact rejections. Characteristics of the dry ice pellet dispensing system used on the Learjet are summarized in table 6. The numbers in parentheses denote standard deviations.

Table 5

LEARJET DATA USED IN NUMERICAL SEEDING SIMULATION

Cloud/ Pass	Date	Time (SAST)	TAS ($m\ s^{-1}$)	Pressure (mb)	Temp (°C)	Time in-Cloud (sec)	ε ($cm^2\ s^3$)	Avg.updraft ($m\ s^{-1}$)	Avg.2D Conc(ℓ^{-1})	Max 2D Con (ℓ^{-1})
1/1	31 Dec. 1980	16:50:46	187	391	-18	28	426	5.1	104	270
/2		16:55:32	187	387	-18	27	312	3.3	264	540
2/1	7 Jan 1981	15:04:40	146	423	-13	25	888	7.2	104	350
/2		15:07:08	140	426	-12	27	353	5.3	382	1360
3/1	25 Feb. 1981	14:26:46	158	397	-17	12	455	5.4	1	1
/2		14:29:00	144	402	-16	18	196	3.4	874	3740
4/1	27 Feb 1981	14:22:02	152	414	-15	17	461	6.0	19	60
/2		14:26:00	155	403	-16	25	468	6.5	610	1670
5/1	27 Feb 1981	14:37:26	166	426	-14	23	486	5.4	45	110
/2		14:40:58	155	427	-14	24	269	3.9	335	960
6/1	27 Feb 1981	14:54:54	164	401	-16	17	423	4.0	4	10
/2		14:57:30	172	419	-16	45	423	4.4	141	2710
7/1	28 Feb 1981	16:08:12	136	462	-9	20	254	3.5	16	50
		16:11:04	140	467	-8	10	163	0.8	52	140
8/1	28 Feb 1981	16:17:42	140	468	-9	7	69	1.0	45	70
/2		16:20:24	129	512	-4	14	178	0.5	91	250
9/1	28 Feb 1981	16:24:48	129	447	-10	16	296	4.5	20	90
/2		16:27:38	142	448	-10	20	157	3.3	75	400
10/1	28 Feb 1981	16:31:42	156	422	-13	17	429	6.3	60	250
/2		16:34:22	156	419	-13	21	401	4.8	289	1060
11/1	3 Mar 1981	14:42:02	163	381	-17	6	181	4.8	12	20
/2		14:45:48	154	382	-17	23	1917	7.9	148	700
12/1	11 Mar 1981	14:43:52	139	420	-17	17	1174	9.6	0	1
/2		14:46:18	140	431	-16	16	525	5.2	1159	3830
13/1	11 Mar 1981	15:37:06	147	412	-17	10	389	8.3	9	20
/2		15:38:44	154	408	-19	34	960	6.0	248	3830
14/1	7 May 1981	15:32:46	160	532	-11	10	259	6.1	0	0
/2		15:36:06	150	531	-11	18	444	9.5	132	510
15/1	12 May 1981	16:55:02	157	495	-15	36	515	4.6	5	20
/2		16:58:24	144	496	-15	26	213	2.2	332	1380

Table 5 (cont/)

LEARJET DATA USED IN NUMERICAL SEEDING SIMULATION

Cloud/ Pass No	Date	Time (SAST)	TAS ($m\ s^{-1}$)	Pressure (mb)	Temp (°C)	Time in-Cloud (sec)	ε ($cm^2\ s^{-3}$)	Avg. updraft ($m\ s^{-1}$)	Avg. 2D Conc (ℓ^{-1})	Max 2D con (ℓ^{-1})
16/1	28 Nov 1984	15:53:27	129	467	-8	10	2016	5.1	0	0
/2		15:56:05	139	462	-9	10	838	1.1	14	42
17/1	12 Mar 1985	15:24:48	152	453	-9	26	1166	6.9	25	104
/2		15:29:24	165	358	-22	15	835	6.4	427	882

Table 6

CHARACTERISTICS OF CIC LEARJET DRY ICE PELLET DISPENSING SYSTEM *

Average pellet diameter	0.90 (0.01) cm
Average pellet length	0.80 (0.40) cm
Pellet density	1.52 $g\ cm^{-3}$
Pellet number density	1300 kg^{-1}
Dispensing number density	0.19 $kg\ s^{-1}$
Pellet line density	1 - 2 m^{-1}

* From Emmitt et. al. (1984)

4.2 Terminal Velocity and Sublimation Rate of Cylindrical Dry Ice Pellets

A model of cylindrical dry ice pellet sublimation was developed in Emmitt et. al. (1984) utilizing observations from field experiments of pellets falling in clear air at warm temperatures. This model, with certain refinements, forms a part of the seeding model used in the present study.

In the field experiments 22 drops of cylindrical dry ice pellets were made over a period of 4 days from an aircraft over a 150000 m^2 sports field. The time for the pellets to reach the ground was measured to compute average fall speeds. The 3 to 5 cm high grass cushioned pellet impact so that pellet sizes and diameters could be measured after the fall. The results of these experiments are shown in table 7.

Table 7

RESULTS OF CLEAR AIR DRY ICE PELLET FALL EXPERIMENTS *

Drop Height (km AGL)	Air Temp (^{o}C)	Pressure (mb)	Avg Pellet Radius (cm)	Avg Fall Speed ($m\ s^{-1}$)
2.0	11	720	0.15	12.6
1.5	14	770	0.23	15.3
1.0	17	820	0.30	17.2
0.5	20	870	0.36	17.8
0	25	920	0.45	-

* After Emmitt et. al. (1984)

Lozowski and Kochtubajda (1980) give the terminal fall velocity for a cylindrical dry ice pellet as:

$$u_T = \left[\frac{\pi\ d\ \rho_c\ g}{2\ C_D\ \rho_a} \right]^{0.5} \tag{4.1}$$

where d = pellet diameter (cm)

ρ_c = pellet density ($g\ cm^{-3}$)

g = acceleration of gravity ($981\ cm\ s^{-2}$)

C_D = drag coefficient

ρ_a = air density ($g\ cm^{-3}$)

The coefficient of drag, C_D can be computed directly as a function of Reynolds Number (Re). Rather than using an empirical relationship for finite cylinders, the functional relationship between Re and C_D for the dry ice pellets has been approximated from the data in table 7 using linearly interpolated values between levels. Solving for C_D in equation (4.1) we get:

$$C_D = \frac{\pi \; d \; \rho_c \; g}{2 \; u_T^2 \; \rho_a} \qquad (4.2)$$

A fictitious dewpoint typical of local summer morning soundings is used to compute more realistic air densities than assuming a dry atmosphere. The results are given in table 8.

Table 8

Re vs C_D ESTIMATES FOR CYLINDRICAL DRY ICE PELLETS

Pressure (mb)	Air Temp (^oC)	Air Dewpoint (^oC)	Pellet radius (cm)	Fall speed (m s^{-1})	Re	C_D
820	18.0	12.0	0.30	12.6	4080	0.91
840	19.5	15.5	0.34	15.3	5690	0.69
870	21.0	13.0	0.38	17.2	7250	0.58
895	2.5	12.0	0.41	17.8	8250	0.57

The relationship between Reynolds Number and C_D can then be approximated by the polynomial regression:

$$C_D = 1.997 - 3.587\text{x}10^{-4} \text{ Re} + 2.254\text{x}10^{-8} \text{ Re}^2 \qquad (4.3)$$

The theoretical sublimation rate formulas for cylindrical dry ice pellets are given by Emmitt *et. al.* (1984) as:

$$\frac{dm}{dt} = \frac{\pi \; k}{4} (2 \; d \; l + a \; d^2) \frac{dd}{dt} \qquad (4.4)$$

$$\frac{dd}{dt} = \frac{4\ l\ k_a\ Nu}{\rho_c\ l_s} (T_s - T_a) \frac{1}{(2\ d\ l + a\ d^2)} \qquad (4.5)$$

where l = pellet length (cm)

k_a = thermal conductivity of air ($cal\ cm^{-1}\ {}^oC^{-1}$)

Nu = Nusselt Number

l_s = specific latent heat of sublimation for dry ice

T_p = pellet surface temperature (^oC)

T_a = ambient air temperature (^oC)

a = dimensionless constant ($\sim$ 0.2)

Using equations 4.1, 4.3, 4.4, and 4.5 the fall of individual dry ice pellets from the drop altitudes in table 7 is simulated in a simple model using one-second time steps. The results are shown in figures 10 and 11. The average fall velocity estimate error is -0.3 m s^{-1} and the average computed radius error is 0.01 cm. We can conclude that this model of dry ice pellet sublimation and fall velocity in clear air is in good agreement with observations.

Laboratory experiments by Kochtubajda and Lozowski (1985) have shown that although cloud and saturated conditions can enhance the dry ice sublimation rate at warm temperatures, the effect at subzero temperatures is

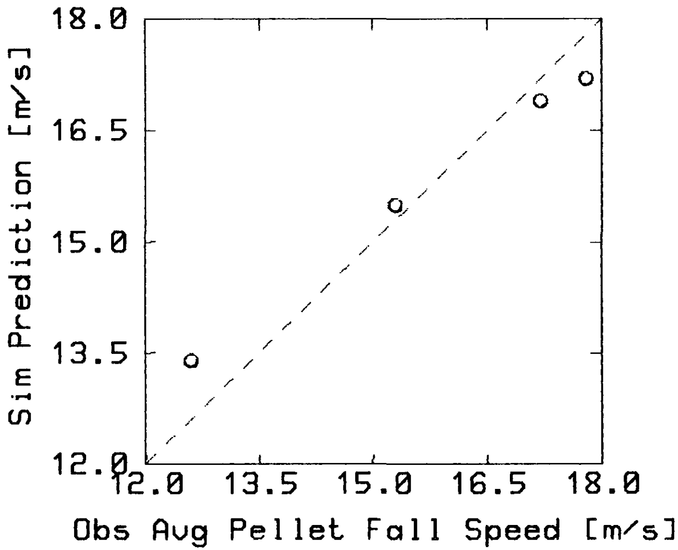

Figure 10. Observed dry ice pellet fall speeds from clear air drop experiments versus simulation prediction.

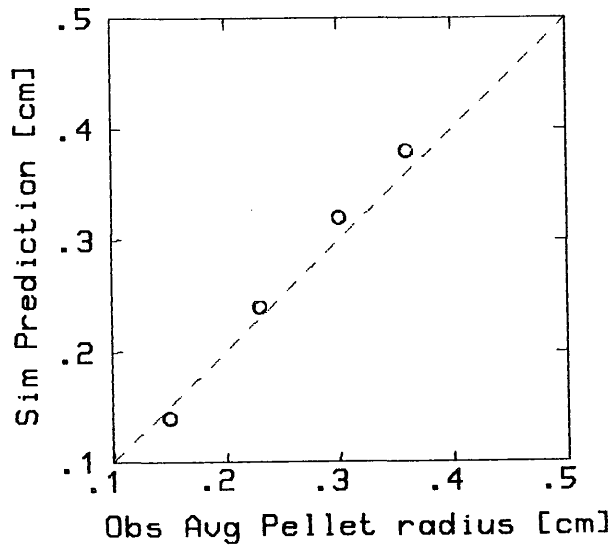

Figure 11. Observed dry ice pellet radius from clear air drop experiments versus simulation prediction.

insignificant. Therefore at seeding altitudes the effects of cloudy air on sublimation rate can be neglected.

4.3 Model Structure

At the beginning of each time step the dry ice pellet sublimation rate is computed for ambient conditions and a new pellet size and mass is calculated. Then a pellet fall speed is computed as the difference between terminal fall speed and updraft velocity. The updraft is assumed to vary linearly in time between the seeding run and the subsequent repenetration. The pellet falls over the time step and a new pellet altitude, Z, and air temperature (assuming a saturated adiabatic in-cloud lapse rate), T_a, are computed.

It is assumed in the model that the products in the seeding plume are advected vertically with the updraft. Then after each time step, the altitude that the portion of the plume nucleated at temperature T_a would reach in the time remaining before aircraft repenetration is calculated. If this altitude is above repenetration altitude a new time step is begun. However, if it is not, we assume that the aircraft has intercepted the portion of the plume nucleated at T_a and the program exits the pellet fall loop.

The volume of the portion of the plume intercepted by the aircraft is taken to be the products of the fall distance of the pellet over the last time step, the aircraft path length through the turret, and the width of the plume. The width of the plume, D, can be predicted using turbulent diffusion theory (Batchelor, 1950) as:

$$D = (C \varepsilon t^3)^{1/2} + D_o \qquad (4.6)$$

where C = dimensionless constant

ε = turbulent dissipation coefficient interpolated in time ($cm^2 s^{-3}$)

t = time (s)

D_o = initial plume width due to turbulence in the aircraft wake (100 m)

A value of 2 for C was used in Garstang et. al. (1981) for cumulus congestus turrets in the eastern Transvaal region. The mass of dry ice sublimed in this volume is the product of the mass of the simulated pellet sublimed over the last time step and the total number of pellets dropped.

Given the dry ice mass sublimed in the plume volume intercepted by the aircraft as calculated from the model, and given the concentration of ice crystals from the Learjet 2D-C probe on repenetration (after subtracting out the value on the seeding run as "natural background") the effectiveness of dry ice is estimated using:

$$E_{model} = CONC_{2D} \frac{Vol}{m} \qquad (4.7)$$

where $CONC_{2D}$ = total 2D-C probe ice crystal concentration ($IC\ cm^{-3}$)

Vol = sampled plume volume (cm^3)

m = mass of dry ice sublimed in the sample volume (g)

This assumes the ice crystal counting efficiency of the 2D-C probe is 100%. Since this is not the case, the model effectiveness values will be underestimated.

Conversely, the ice crystal concentration in the sampled plume volume can be predicted using the model by taking the product of the mass of dry ice sublimed and the laboratory derived value of effectiveness for the corresponding nucleation temperature.

A correction to the model predicted ice crystal concentrations for each cloud should be applied if the plume age is less than the time required to produce 90% of the counted ice crystals by the time the plume is sampled by the aircraft. The correction is based on the nucleation temperature using the data presented in section 5.3 of this study. However, in all 17 cases, the plume was always old enough for complete ice crystal production to occur and so no corrections were necessary.

V. RESULTS

5.1 The Overseeding Problem

The most significant problem in previous laboratory studies of dry ice by other investigators has been the probable loss of ice embryos from overseeding because of the inability of the cloud chambers used to remain at water saturation. Indeed, this was recognized as a serious problem in the initial phases of the experiments presented here and a great deal of time and effort were devoted to its elimination resulting in two changes in the experimental design: The pellet path length through the cloud, originally set at 32.9 cm, was reduced to its smallest possible setting of 10.2 cm, and the smallest pellet size of 5 mm in diameter was used instead of the 11 mm size chosen initially. Despite these changes, overseeding still occurred when the pellets went off trajectory and smashed against the pellet "catcher", or when pellets partially fell apart during transit through the cloud. These instances, however, were easily identified by their long ice crystal production times relative to other runs under the same conditions and were rejected from the data set. Was overseeding, then, still a problem?

Horn *et. al.* (1982) note that at all operating temperatures in the ICC an ice crystal production rate of about 100000 per liter (9.6×10^7 ice crystals) in one minute can completely deplete the cloud liquid water content in spite of continual cloud replenishment. In the event of such overseeding, sublimation loss of ice embryos below a critical radius can occur. The average ice crystal yields obtained in these experiments for 5 mm diameter pellets at a cloud liquid water content of 1.5 g m^{-3} are listed in table 9. As will be discussed in section 5.3, the nucleation rate of dry ice is so fast that the entire population of ice embryos probably appear if not instantaneously, certainly well within one minute, after seeding. The ice crystal yields in table 9, then, do approach values where overseeding could occur, particularly at the colder temperatures.

Table 9

ICE CRYSTAL YIELDS AND IWC PRODUCTION RATE ESTIMATES

Temp (°C)	mass growth rate at 50 s (ng s^{-1})	avg ice crystal yield *	IWC production rate (g m^{-3} s^{-1})	IWC after 50 s (g m^{-3})
-2	no data	5.2×10^6	------	----
-3	0.12	2.3×10^7	2.9×10^{-3}	0.14
-4	0.28	2.8×10^7	8.2×10^{-3}	0.41
-5	0.42	3.4×10^7	1.5×10^{-2}	0.74
-6	0.55	5.0×10^7	2.9×10^{-2}	1.40
-8	0.48	5.0×10^7	2.5×10^{-2}	1.30
-9	0.45	5.4×10^7	2.5×10^{-2}	1.30
-10	0.45	6.0×10^7	2.8×10^{-2}	1.40
-12	0.47	7.4×10^7	3.6×10^{-2}	1.80
-15	1.01	6.4×10^7	6.7×10^{-2}	3.40
-16	0.65	9.1×10^7	6.2×10^{-2}	3.10
-20	0.48	1.6×10^8	8.0×10^{-2}	4.00

* pellet diameter = 5 mm, LWC = 1.5 g m^{-3}, v = 12.3 m s^{-1}

To add further perspective, it is useful to make a rough estimate of the order of magnitude of the ice water content nearly a minute after seeding for each given yield. Using the empirical ice crystal mass growth rates from Ryan et. al. (1976) at 50 seconds after seeding, we can approximate the ice water content production rate using:

$$\frac{d(IWC)}{dt} = \frac{dm}{dt} \times \frac{Y}{Vol} \qquad (5.1)$$

where IWC = ice water content ($g\ m^{-3}$)

dm/dt = ice crystal mass growth rate ($g\ s^{-1}$)

Y = total ice crystal yield

Vol = total volume of the ICC ($0.960\ m^3$)

Multiplying the above by 50 seconds, we obtain the results also listed in table 9. It can be seen that the ice water content after nearly one minute approaches the initial chamber liquid water content at -6 °C and is well in excess at temperatures of -15 °C and colder.

These results suggest that some degree of overseeding which can cause the sublimation loss of some ice embryos could have occurred at temperatures as warm as -6 °C and most likely did occur at temperatures of -15 °C and colder. As a consequence, the ice crystal yield data at these temperatures for a 1.5 $g\ m^{-3}$ liquid water content may be underestimated.

5.2 Dry Ice Effectiveness

A complete effectiveness spectrum was obtained for 5 mm diameter dry ice pellets at 83% terminal velocity with a cloud liquid water content of 1.5 g m^{-3}. The results are shown in figure 12 and are tabulated in table 10. The effectiveness values have been computed using both the Holroyd _et_. _al_. (1978) and Fukuta _et_. _al_. (1971) sublimation rate formulas. As noted in the previous section, some overseeding may have occurred at temperatures as warm as -6 ^{o}C. The results from -6 to -20 ^{o}C must therefore be regarded as conservative estimates. Contrary to the Fukuta _et_. _al_. effectiveness spectrum after correction for a computational error noted in Horn _et_. _al_. (1982), the results presented here show a moderate temperature dependence ranging over two orders of magnitude.

The dip in the laboratory effectiveness spectrum at -15 ^{o}C could be due to overseeding. The depositional growth rate of ice crystals is a maximum at this temperature. If overseeding is indeed the cause, we would expect to see an increase in ice crystal yield at a higher cloud liquid water content which would better maintain the vapor supply. When the liquid water content was stepped up to 2.5 g m^{-3} there was a small increase in ice crystal yield of 6%. This is far short of the approximately 38% increase needed to eliminate the dip in the effectiveness curve.

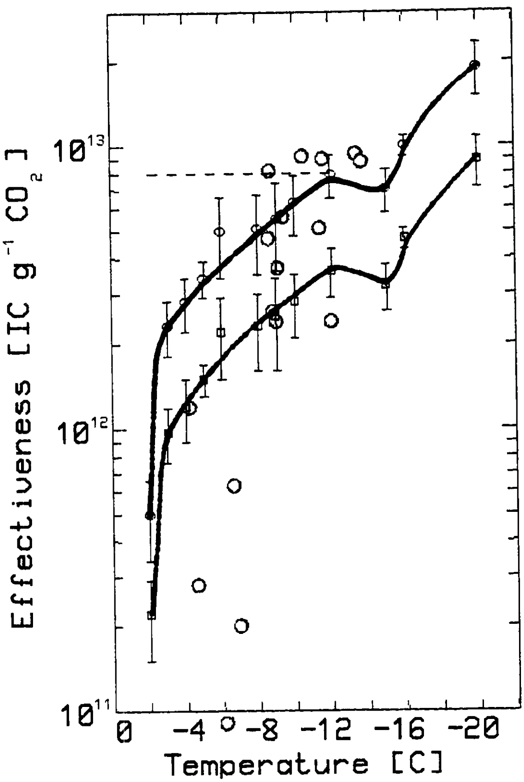

Figure 12. Laboratory derived effectiveness spectrum for 5 mm diameter dry ice pellets at a cloud liquid water content of 1.5 g m^{-3} using the Fukuta et. al. (1971) sublimation rate formula (top solid line) and the Holroyd et. al. (1978) formula (bottom solid line). Numerical model effectiveness estimates using the maximum 2D-C ice crystal concentrations observed in 17 clouds are shown as solid circles. The Fukuta et. al. effectiveness spectrum after correction by Horn et. al. (1982) is shown as the dashed line for comparison.

Table 10

Laboratory Effectiveness Results

(d = 5 mm, v = 12.3 m s^{-1}, LWC = 1.5 g m^{-3})

Temp (°C)	Num of Successful Trials	Avg Ice Crystal Yield	Method No. 1 * Sublimation Rate (mg s^{-1})	Method No. 1 * Effect-iveness (IC g^{-1} CO_2)	Method No. 2 ** Sublimation Rate (mg s^{-1})	Method No. 2 ** Effect-iveness (IC g^{-1} CO_2)
-2	4	5.2×10^{6}	2.8755	2.2×10^{11}	1.2444	5.0×10^{11}
-3	4	2.3×10^{7}	2.8379	9.7×10^{11}	1.2329	2.3×10^{12}
-4	3	2.8×10^{7}	2.8003	1.2×10^{12}	1.2215	2.8×10^{12}
-5	9	3.4×10^{7}	2.7627	1.5×10^{12}	1.2100	3.4×10^{12}
-6	5	5.0×10^{7}	2.7251	2.2×10^{12}	1.1984	5.0×10^{12}
-8	5	5.0×10^{7}	2.6500	2.3×10^{12}	1.1752	5.1×10^{12}
-9	5	5.4×10^{7}	2.6124	2.5×10^{12}	1.1635	5.5×10^{12}
-10	8	6.0×10^{7}	2.5748	2.8×10^{12}	1.1518	6.3×10^{12}
-12	5	7.4×10^{7}	2.4996	3.6×10^{12}	1.1283	7.9×10^{12}
-15	6	6.4×10^{7}	2.3868	3.2×10^{12}	1.0928	7.0×10^{12}
-16	4	9.1×10^{7}	2.3493	4.7×10^{12}	1.0809	1.0×10^{13}
-20	4	1.6×10^{8}	2.1989	8.9×10^{12}	1.0328	1.9×10^{13}

* Using Holroyd et. al. (1978) sublimation rate equation

** Using Fukuta et. al. (1971) sublimation rate equation

The effectiveness estimates from the model runs using the maximum 2D-C ice crystal concentrations from 17 seeded cumulus congestus turrets are also shown in figure 12 and are tabulated in table 11. They are generally within an order of magnitude of the laboratory derived effectiveness spectrum and also imply a temperature dependence. This is remarkably good agreement considering the simplicity of the model, the large number of assumptions involved, and the limitations in sampling the dry ice plumes with an aircraft.

Table 11

MODEL EFFECTIVENESS ESTIMATES

Cloud Num	Nucleation Temp (^{o}C)	Effectiveness using 2D-C Avg Conc	Effectiveness using 2D-C Max Conc
1	-12.0	1.4×10^{12}	2.4×10^{12}
2	- 8.8	7.2×10^{11}	2.6×10^{12}
3	-13.4	2.2×10^{12}	9.4×10^{12}
4	- 8.7	3.0×10^{12}	8.2×10^{12}
5	- 9.1	1.3×10^{12}	3.7×10^{12}
6	-10.5	4.6×10^{11}	9.2×10^{12}
7	- 6.9	7.9×10^{10}	2.0×10^{11}
8	- 4.6	7.1×10^{10}	2.8×10^{11}
9	- 6.6	1.1×10^{11}	6.3×10^{11}
10	- 9.0	6.9×10^{11}	2.4×10^{12}
11	- 9.4	1.1×10^{12}	5.6×10^{12}
12	-11.6	2.7×10^{12}	9.0×10^{12}
13	-13.7	5.5×10^{11}	8.8×10^{12}
14	- 4.2	3.1×10^{11}	1.2×10^{12}
15	-11.4	1.2×10^{12}	5.1×10^{12}
16	- 6.1	3.0×10^{10}	9.1×10^{10}
17	- 8.6	2.4×10^{12}	4.7×10^{12}

The model has also been used to compute the expected ice crystal concentration in each sampled plume volume using the laboratory effectiveness value corresponding to the estimated nucleation temperature. These results are plotted in figures 13 and 14 against observed average and maximum 2D-C ice crystal concentrations (after subtracting out the concentrations measured on the seeding penetrations as "background"). In 11 of the cases the predicted ice crystal concentration lies between the observed average and maximum values, in 5 cases it exceeds the observed maximum and in the remaining case it is below the observed average.

5.3 Rates of Ice Crystal Production

The overall rate of ice crystal production from dry ice seeding is very fast. The times required to produce 90% of the counted ice crystals by seeding with 5 mm diameter pellets at a cloud liquid water content of 1.5 g m^{-3} are listed in table 12. They range from 2.40 minutes at -2 ^{o}C to 1.24 minutes at -16 ^{o}C. These rates are much faster than those observed for most aerosol type nucleants under the same cloud conditions at water saturation (chapter 6).

The composite kinetics plots for 5 mm dry ice pellets at a cloud liquid water content of 1.5 g m^{-3} from -2 to -20 ^{o}C are shown in figure 15. Recalling that the slopes

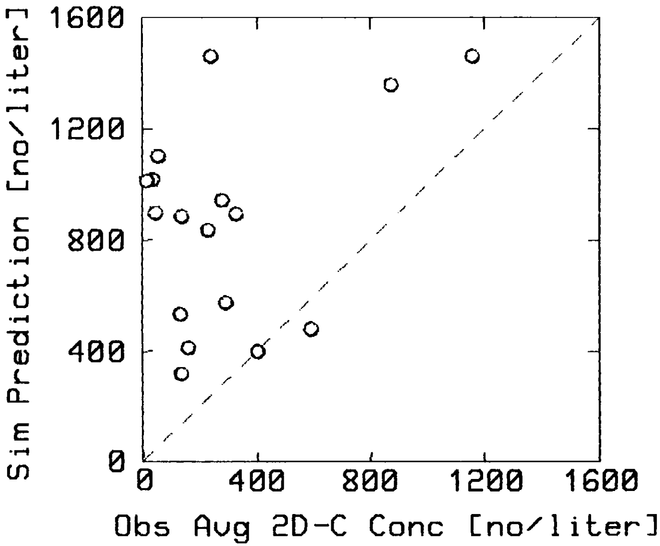

Figure 13. Observed average 2D-C ice crystal concentrations in the seeding curtains of 17 cumulus congestus clouds versus the model predicted values applying the laboratory effectiveness spectrum using the Holroyd et. al. (1978) sublimation rate formula.

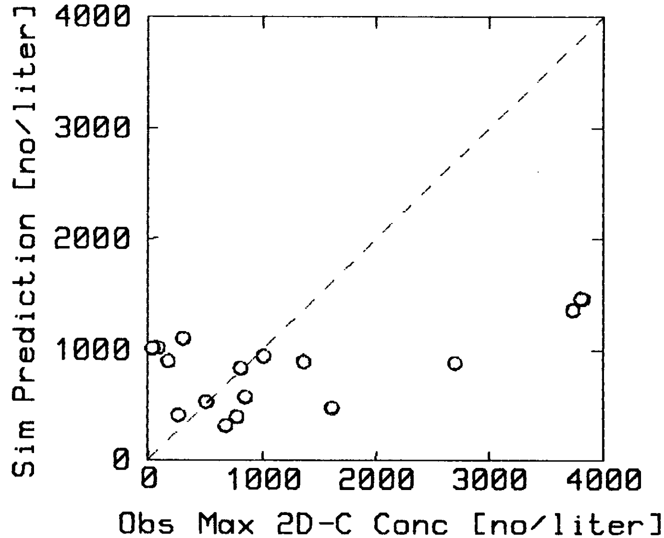

Figure 14. Same as figure 13 except for observed maximum 2D-C ice crystal concentrations.

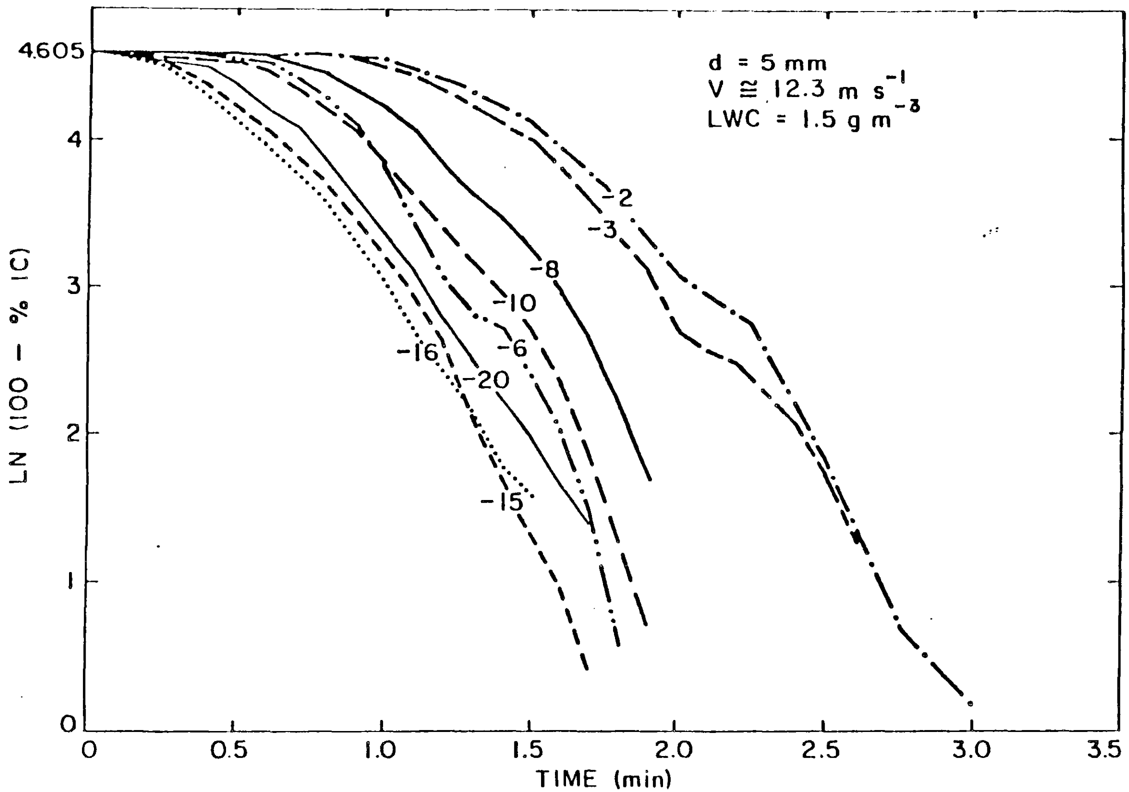

Figure 15. Composite kinetics plots for 5 mm dry ice pellets over a range of temperatures. The plots for -5, -9, and -12 °C have not been included in order to reduce confusion in the figure.

Table 12

ICE CRYSTAL PRODUCTION RATE DATA

FOR 5 mm PELLETS *

Temp (oC)	Time Required to Produce 90% of the Counted Ice Crystals (min)	Percentage of Ice Crystals Produced in 0.8 minutes (%)
- 2	2.40	2.5
- 3	2.29	2.2
- 5	1.43	29.0
- 6	1.53	28.2
- 8	1.78	15.8
- 9	1.53	32.9
-10	1.61	32.3
-12	1.47	44.8
-15	1.26	59.8
-16	1.24	63.9
-20	1.38	53.0

* $v = 12.3$ m s^{-1}, LWC = 1.5 g m^{-3}

are proportional to the ice crystal production rates, the plots can be characterized as having a slow, but short, linear phase followed by a fast curved phase. The flat first phase represents the time delay between the injection of the dry ice pellet and first detection of ice crystals greater than 20 microns in diameter by the acoustic counter. This time delay is temperature dependent. The shapes of the curves in the second phase are similar irrespective of temperature. It is the first phase, then, that determines the overall rate of ice crystal production.

Also listed in table 12 are the percentages of the total number of ice crystals counted in 0.8 minutes which is plotted against the ice crystal mass diffusional growth rates after 50 seconds (0.83 minutes) from Ryan _et_. _al_. (1976) in figure 16. Clearly, the initial rate of ice crystal production is reasonably well correlated (r = 0.817) with the rate of ice crystal growth. This suggests that the rate of production is not determined by nucleation, but by factors such as diffusional ice crystal growth. If the effect of ice crystal fall speeds during fallout were included, this correlation might be even stronger.

There are three primary processes that can occur simultaneously in the ICC under saturated conditions after seeding: Nucleation, ice crystal growth, and ice crystal fallout. It is the slowest process that determines the rate of ice crystal production at any instant. Since it is the initial phase that determines the overall rate of ice crystal production, and since the rate of ice crystal production in this initial phase is not determined by nucleation, then the process of nucleation occurs too quickly to govern the rate of reaction.

5.4 Nucleation Mechanism

The widespread misconception that dry ice functions primarily by the freezing of supercooled water droplets has been noted in chapter 2. This study confirms the findings of

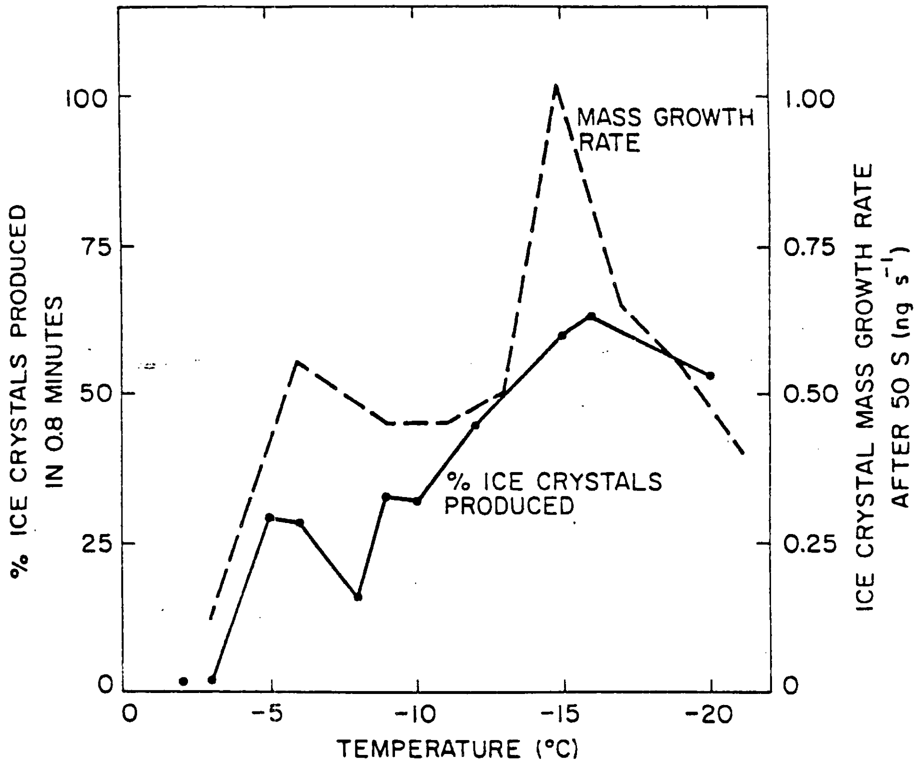

Figure 16. Percentage of the total number of ice crystals counted that were observed in 0.8 minutes for 5 mm dry ice pellets at a cloud liquid water content of 1.5 g m^{-3} (solid line). The pellets velocity was 12.3 m s^{-1}. The mass growth rate for ice crystals after Ryan et. al. (1976) is plotted as the dashed line.

Horn *et. al.* (1982) and Leonard and Detwiler (1983) that the number of water droplets that could be frozen by dry ice is several orders of magnitude less than the observed ice crystal yields. Table 13 shows the number of cloud droplets that could be frozen if we assume a 100% freezing efficiency within an effective radius equal to four times the pellet diameter of 5 mm. This freezing could occur either by contact nucleation of the drops colliding with the pellet, or by homogeneous freezing of the droplets in the portions of the pellet plume colder than -40 °C. It is clear that the number of possible frozen cloud droplets can at best only account for a small percentage of the total ice crystal yield. Although droplet freezing certainly can occur, it is not the dominant nucleation mechanism.

Table 13

PERCENTAGE OF ICE CRYSTAL YIELD POSSIBLE FROM CLOUD DROPLET FREEZING *

LWC ($g\ m^{-3}$)	Temp (°C)	Observed Yield	Cloud Droplet Conc(cm^{-3})	Number of Possible Frozen Droplets**	Percentage of Yield
1.5	- 5	3.4×10^7	6550	8.4×10^5	2.5
1.5	-10	6.0×10^7	4300	5.5×10^5	0.9
1.5	-15	6.4×10^7	3750	4.8×10^5	0.8

* d = 5 mm, v = 12.3 m s^{-1}
** assuming r_{eff} = 20 mm

Excluding a droplet dependent nucleation mechanism as discussed above, the mechanism must be vapor dependent. One possibility is heterogenous nucleation by vapor deposition onto natural ice nuclei in the highly supersaturated region next to the dry ice pellet. However, the presence of natural ice nuclei in the ICC is minimized by filtering the cold air entering the chamber. Even allowing for some "leakage" of nuclei into the chamber, it is most unlikely that they could explain the very high ice crystal concentrations (approximately 3×10^7 cm^{-3} at -10 ^oC) in the dry ice pellet plume. The remaining possibility is homogeneous nucleation --- either homogeneous condensation-freezing or homogeneous vapor to ice phase. Evidence from other investigators (Anderson et. al., 1980) indicates that the former possibility is the most likely.

One of the primary objectives of this research was to attempt to use chemical kinetics methodologies in the hope of gaining insight into the mechanism of nucleation by dry ice seeding. However, this approach can only be used if nucleation is the rate-determining step in the production of ice crystals. This has not been the case for these experiments. Consequently, the kinetics data produced from these experiments through the variation of water vapor (as a function of temperature) and the variation of cloud droplets (as a function of liquid water content) show no consistent responses that are interpretable in the context of ice nucleation.

5.5 Volume Effect

The expression of dry ice "effectiveness" in terms of ice crystals produced per gram of CO_2 sublimed is generally used because the traditional way of expressing glaciogenic aerosol effectiveness is in terms of ice crystals produced per gram of substance consumed. This is misleading if dry ice functions primarily through the cooling of cloudy air rather than by any process dependent upon its chemical composition. In such a case, dry ice effectiveness would be more usefully expressed in terms of ice crystals produced per unit volume of air cooled per unit time that the air remains cooled below a certain temperature. It is then important to assess if there is a volume effect and a velocity (time) effect upon the ice crystal yield from dry ice seeding. Velocity effect experiments are presented in the following section.

The difficulty in obtaining data with the larger 7 mm pellets has been noted in section 5.1. Overseeding at temperatures colder than -7 ^{o}C was quite pronounced and the data were rejected. A comparison of the yields between the 5 and 7 mm pellets is then available over only a small range of warm temperatures (table 14). Since the speeds of the 5 and 7 mm pellets are very close, only the volumes of air cooled should be significantly different. It is thus surprising to note that the percentage change in yields are not constant with temperature and, in fact, decrease with decreasing temperature. This would suggest that there is no clear volume effect.

Table 14

COMPARISON OF ICE CRYSTAL YIELDS FOR 5 AND 7 mm PELLETS *

Temp (^{o}C)	5 mm	7 mm	% change
-2	5.2×10^{6}	1.3×10^{7}	+ 150
-3	2.3×10^{7}	4.2×10^{7}	+ 83
-4	2.8×10^{7}	4.1×10^{7}	+ 46
-5	3.4×10^{7}	4.4×10^{7}	+ 29
-6	5.0×10^{7}	5.1×10^{7}	+ 2
-7	no data	6.5×10^{7}	---

* LWC = 1.5 g m^{-3}, v (5 mm) = 12.3 m s^{-1}, v (7 mm) = 12.4 m s^{-1}

The composite kinetics plots for the 5 and 7 mm pellets (figure 17), however, imply that at -5 and -6 ^{o}C there was still some overseeding occurring for the 7 mm pellets. This is evidenced by the slower ice crystal production rates relative to the 5 mm pellets despite identical cloud and seeding conditions. Kinetics data for 5 mm pellets at -4 ^{o}C are insufficient to form a reasonable composite plot for comparison to the 7 mm data. Therefore, only the -2 and -3 ^{o}C comparisons can be considered valid.

It is interesting to note that the percentage increase in volume swept out from the 5 to 7 mm pellets is 96% which is comparable to the percentage change in yield

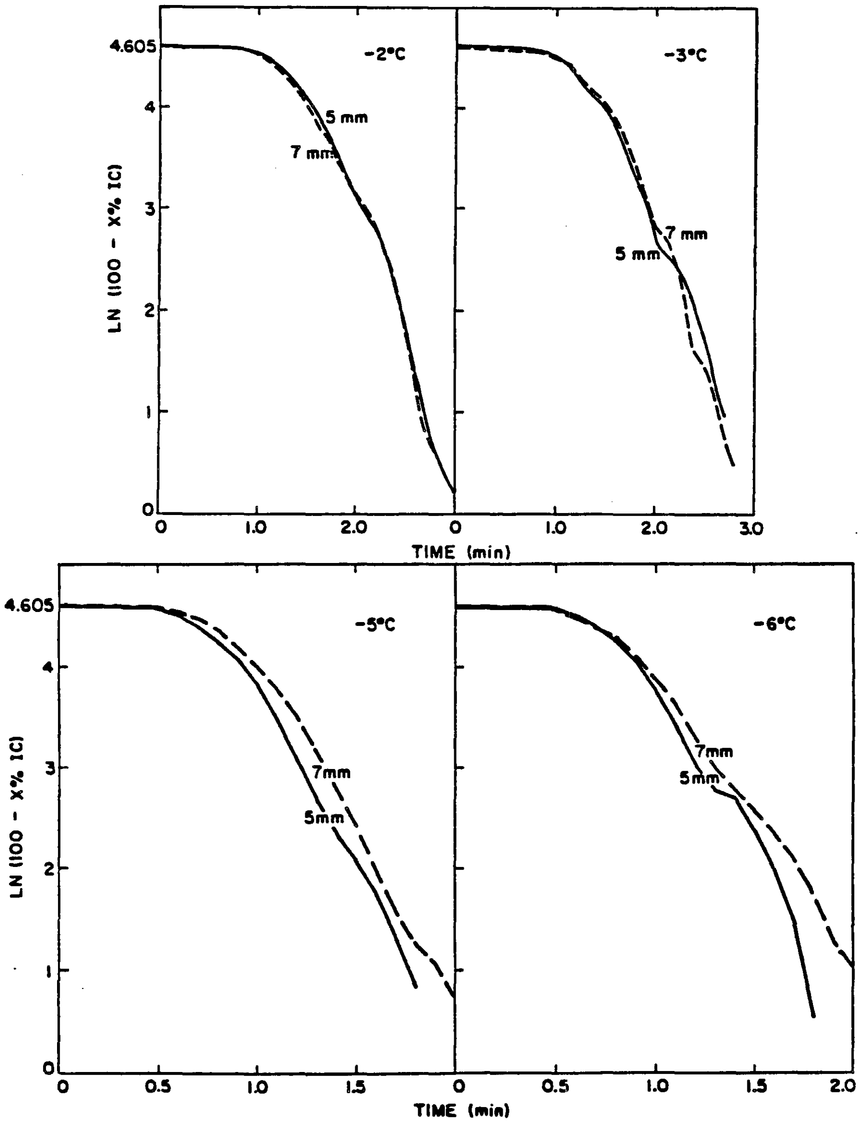

Figure 17. Composite kinetics plots for 5 and 7 mm dry ice pellets for several warm temperatures at a cloud liquid water content of 1.5 g m^{-3}.

at -2 and -3 °C. This lends support for a volume effect, but, without better data at colder temperatures, is not conclusive.

5.6 Velocity Effect

To test for a velocity effect, data were taken for 5 mm pellets at a speed of 6.5 m s^{-1} (44% of terminal velocity) in addition to the complete data set at 12.3 m s^{-1} (83% terminal velocity). Slower pellet speeds had a very poor "catch" success rate and could not be used. The results from these experiments are shown in table 15. Whereas there was no significant change in ice crystal yield at -5 °C, there was an increase in yield observed from the slower to higher speeds at the colder temperatures on the order of 40%. This result is contrary to those of Eadie and Mee (1963) who found no significant differences in ice crystal yields at temperatures colder

Table 15

COMPARISON OF ICE CRYSTAL YIELDS FOR 5 mm PELLETS

AT 6.5 AND 12.3 m s^{-1} *

	v = 6.5 m s^{-1}		v = 12.3 m s^{-1}		
Temp (°C)	n	Avg Yield	n	Avg Yield	% change
- 5	5	3.3×10^7	9	3.4×10^7	+ 3
-10	5	4.3×10^7	8	6.0×10^7	+ 40
-15	5	4.5×10^7	6	6.4×10^7	+ 42

* d = 5 mm, LWC = 1.5 g m^{-3}

than about -8 ^{o}C for pellets moving at 0.8 m s^{-1} versus 16 m s^{-1}, while at warmer temperatures the faster pellets showed a steep dropoff in yield and the slower pellets did not.

The reason for these results is not clear. Much more work needs to be done on this problem.

5.7 Liquid Water Content Effect

As discussed in section 5.4, the nucleation mechanism of dry ice is primarily droplet independent. We would therefore expect to observe no significant change in yield with changing cloud droplet concentrations as long as the vapor concentration is unchanged. This can be tested by varying the chamber cloud liquid water content while holding cloud temperature constant.

Unfortunately, no experiments could be run at a 0.5 g m^{-3} liquid water content due to the occurrence of persistent overseeding. These tests then had to be conducted at 2.5 g m^{-3} where it is somewhat more difficult to minimize temperature gradients in the chamber. The results are given in table 16, which shows an increase in ice crystal yield at the higher liquid water contents.

Could this result have been due to droplet dependent nucleation, i.e. droplet freezing? Although droplet concentrations at 0.5 and 1.5 g m^{-3} liquid water contents are known, there is no data available at 2.5 g m^{-3}. In order to make some quantitative discussion possible, cloud

Table 16

COMPARISON OF ICE CRYSTAL YIELDS AT 1.5 AND 2.5 g m^{-3}
CLOUD LIQUID WATER CONTENTS *

	LWC = 1.5		LWC = 2.5		
Temp (°C)	n	Avg Yield	n	Avg Yield	% change in yield
- 5	9	3.4×10^7	5	4.4×10^7	+ 29
-10	8	6.0×10^7	5	6.8×10^7	+ 13
-15	6	6.4×10^7	5	6.8×10^7	+ 6

* d = 5 mm, v = 12.3 m s^{-1}

droplet concentrations at 2.5 g m^{-3} have been extrapolated as shown in figure 18 from the data in figure 3. These values are listed in table 17. We can estimate the number of frozen droplets as in section 5.4 by making the very liberal assumption of a 100% freezing efficiency in a radius equal to four times the pellet diameter around the center of the pellet. As is shown in table 18, the increase in yield that we might expect due to the freezing of more water droplets is 5% or less of the actual increase in yield.

It seems more likely that the results implied from table 15 are artifacts due to overseeding. The composite kinetics plots comparing the ice crystal production rates, at the two liquid water contents are shown in figure 19. In all of these plots the rates of ice crystal production are slightly slower at 1.5 g m^{-3} compared to 2.5 g m^{-3} which could imply a relative degree of overseeding at 1.5

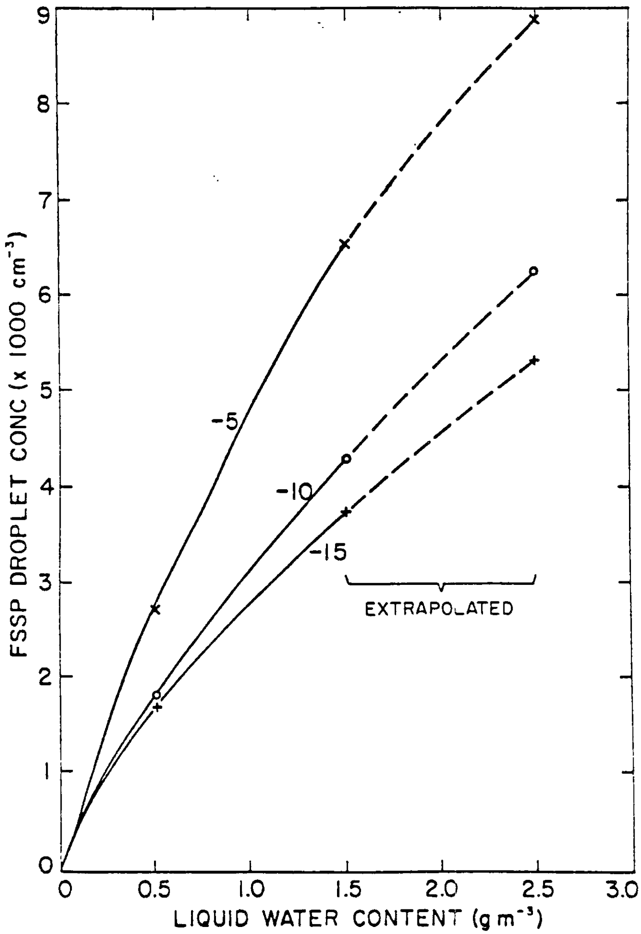

Figure 18. Extrapolation of cloud droplet concentrations for -5, -10, and -15 ^{o}C at a cloud liquid water content of 2.5 g m^{-3}. The data points at 0.5 and 1.5 g m^{-3} were taken from figure 3. The curves shown are hand-fitted.

Table 17

CHANGE IN CLOUD DROPLET CONCENTRATION WITH LIQUID WATER CONTENT

Temp (°C)	Cloud Droplet Conc (cm^{-3}) LWC = 1.5	LWC = 2.5*	% change
- 5	6550	8850	+ 35
-10	4300	6250	+ 45
-15	3750	5300	+ 41

* extrapolated values

Table 18

PERCENTAGE CHANGE IN LIQUID WATER CONTENT EFFECT YIELD DUE TO CLOUD DROPLET FREEZING

Temp (°C)	Num Frozen Droplets ** LWC = 1.5	LWC = 2.5	Difference	Yield 2.5 -Yield 1.5	% Yield Change Due To Droplet Freezing
- 5	8.4×10^5	1.1×10^6	2.6×10^5	1.0×10^7	+ 2.6
-10	5.5×10^5	8.0×10^5	2.5×10^5	0.8×10^7	+ 3.1
-15	4.8×10^5	6.8×10^5	2.0×10^5	0.4×10^7	+ 5.0

* d = 5 mm, v = 12.3 m s^{-1}
** assuming an effective freezing radius of 20 mm

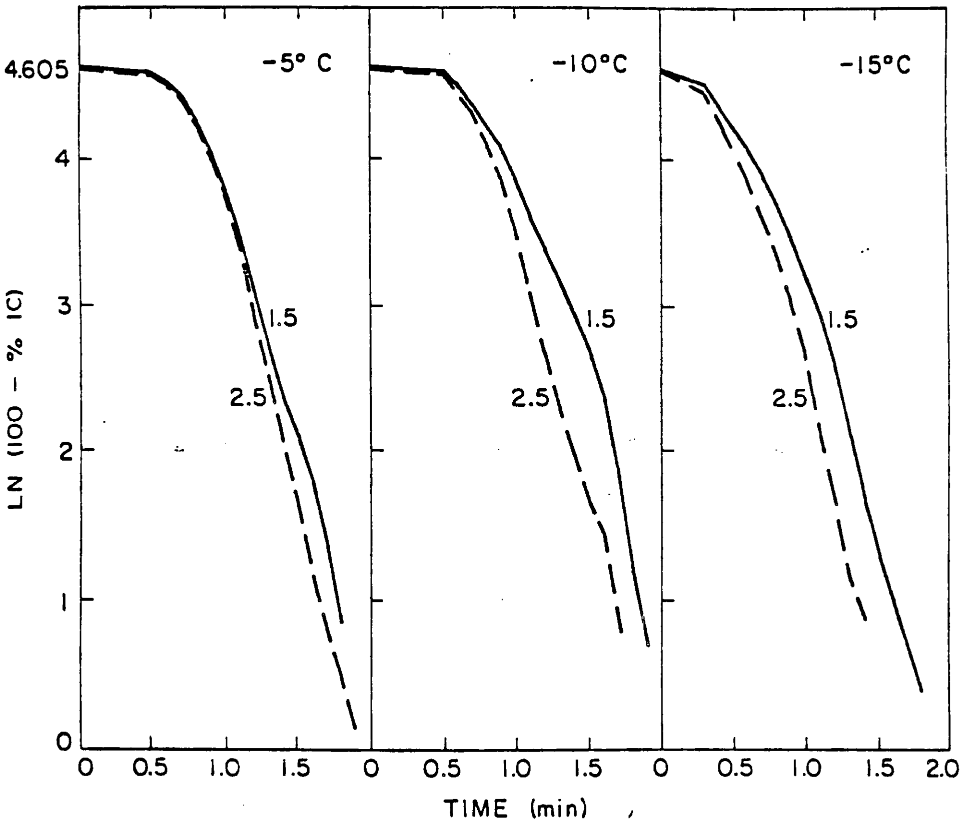

Figure 19. Composite kinetics plots for 5 mm dry ice pellets at 1.5 and 2.5 g m^{-3} cloud liquid water contents. Pellet velocities are approximately 12.3 m s^{-1}.

g m^{-3}. If this is the case, the differences in the percentage changes in yield in table 15 would be more consistently explained since the liquid water deficit is much more severe at -15 ^{o}C than at -5 ^{o}C.

VI SUMMARY AND CONCLUSIONS

6.1 Summary of experimental results

The results of the laboratory and numerical modeling experiments in the context of the objectives set forward for this research may be summarized as follows:

A) Effectiveness

1. A complete dry ice effectiveness spectrum for 5 mm diameter spherical pellets at a 1.5 g m^{-3} liquid water content has been established using the isothermal cloud chamber. Effectiveness is moderately temperature dependent ranging from 2.2 x 10^{11} ice crystals produced per gram CO_2 sublimed at -2 °C to 8.9 x 10^{12} at -20 °C using the Holroyd _et._ _al._ (1978) sublimation rate formula. Alternative use of the Fukuta _et._ _al._ (1971) sublimation rate formula gives a spectrum from 5.0 x 10^{11} to 1.9 x 10^{13} over the same temperature range. There is a likelihood that a certain degree of overseeding occurred during these experiments. If this is the case the effectiveness values colder than about -6 °C may be underestimated and at -15 °C and colder are almost certainly underestimated.

2. Effectiveness values computed using a numerical model based on aircraft measurements of dry ice seeding curtains in 17 cumulus congestus turrets are generally within an order of magnitude of the laboratory derived values. The model results also imply a temperature dependence. This lends a certain degree of confidence to the laboratory effectiveness results despite the overseeding problem and demonstrates that some transferability of the laboratory results to the field is warranted.
3. Model calculated ice crystal concentrations using the laboratory effectiveness spectrum are generally within an order of magnitude of the observed ice crystal concentrations in the seeding plumes from the 17 cumulus turrets. This suggests that the high concentrations of ice crystals often observed in these plumes can be explained solely by dry ice nucleation.
4. Studies of the effects of dry ice pellet speed and size on effectiveness have been inconclusive apparently due to problems with overseeding.

B)Rates of ice crystal production

1. The rates of ice crystal production for 5 mm dry ice pellets at 1.5 g m^{-3} cloud liquid water content are very fast, ranging from 90% of the

counted ice crystals being produced in 2.4 minutes at -2 °C, to 1.3 minutes at -15 °C.

2. The rates of ice crystal production are not completely determined by dry ice nucleation, but involve other factors such as ice crystal growth and fallout.

C) Nucleation mechanism

1. The ice crystal yields in the ICC cannot be accounted for by a predominant droplet freezing nucleation mechanism, nor by heterogeneous nucleation by vapor deposition onto natural ice nuclei. The predominant mechanism must be vapor dependent, most likely homogeneous condensation-freezing.

2. Because dry ice nucleation apparently does not determine the rates of ice crystal production observed in the ICC experiments, chemical kinetics methodology could not be applied in this instance to study the mechanism of nucleation.

6.2 A Comparison of Dry Ice With Some Other Glaciogenic Seeding Agents

To help put dry ice into perspective as a glaciogenic seeding agent for weather modification, it is useful to compare it to other nucleants. The effectiveness and ice crystal production rates data of four other nucleants studied at the CSU Cloud Simulation and Aerosol Laboratory

(Sim Lab) using the ICC are presented here. These nucleants are:

1. Composite Silver Iodide - Silver Chloride - Sodium Chloride (AgI-AgCl-NaCl). This is an experimental "state-of-the-art" solution aerosol developed at the Cloud Simulation and Aerosol Laboratory. It functions by homogeneous condensation-freezing (Finnegan, et. al., 1984).
2. Mixed Silver Iodide - Silver Chloride (AgI-AgCl). This aerosol was used in pyrotechnic form during the Florida Area Cumulus Experiment. It functions by contact nucleation at temperatures of -16 ^{o}C and warmer (DeMott et. al., 1983; DeMott, 1982).
3. Silver Iodide - Sodium Iodide (2AgI-NaI). Used in the Israeli and Climax program, this solution aerosol functions primarily by condensation freezing (Blumenstein et. al., 1983).
4. Swartklip TB-1 with Hexachlorobenzene (HCB) pyrotechnic aerosol. These pyrotechnics were used during the 1979/80 and 1980/81 summer seasons of the Nelspruit, South Africa hail-suppression program. Effectiveness data is from calibration tests run at the Sim Lab for Cansas International Corporation (Pty) Ltd. in 1979. The rough kinetics data presented in this study are extracted from those tests. The mode of

nucleation has been deduced to be contact freezing.

The first three nucleants listed have been studied and characterized successfully using chemical kinetics methodology and are documented in the literature.

The comparative effectiveness spectra are shown in figure 20. The dry ice curve is from the results obtained for 5 mm diameter pellets using the Holroyd et. al. (1978) sublimation rate formula. Except for the Swartklip TB-1 data, dry ice effectiveness is a few orders of magnitude less than the other nucleants at temperatures colder than about -5 ^{o}C. While the aerosol nucleants show little significant effectiveness at temperatures warmer than about -4.5 ^{o}C, dry ice has demonstrable effectiveness up to at least -2 ^{o}C. The dry ice effectiveness curve shows a moderate temperature dependence over one order of magnitude from -3 to -20 ^{o}C.

Examination of the kinetics plots in figure 21 and the data in table 19 shows that dry ice has a much faster rate of ice crystal production than the aerosol nucleants at water saturation with the exception of AgI-AgCl-NaCl at -16 ^{o}C. It must be noted that the two contact nucleants (AgI-AgCl and the Swartklip TB-1 pyrotechnics) have nucleation rates that are dependent on cloud droplet concentration. In clouds with much lower droplet concentrations than that of the ICC their nucleation rates would be even slower.

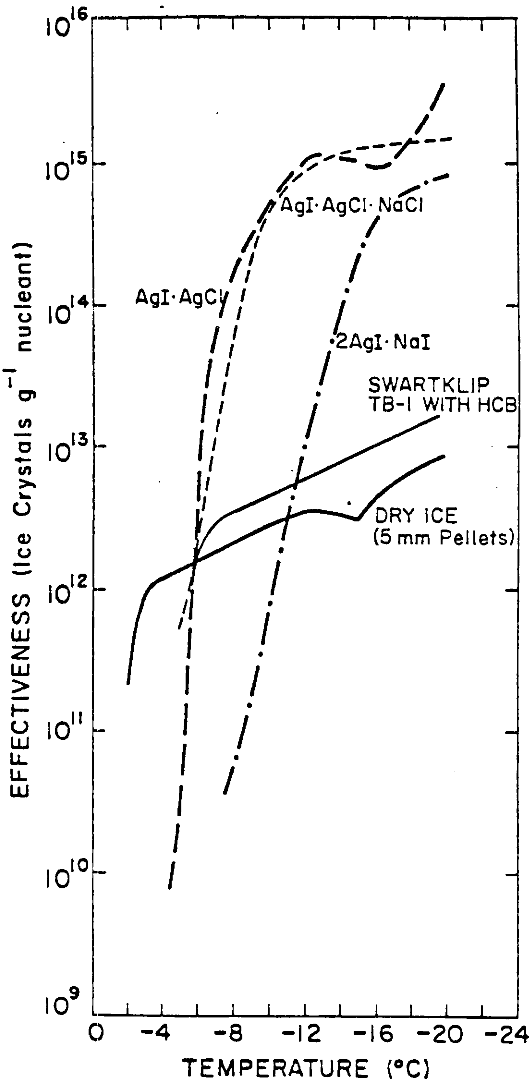

Figure 20. Effectiveness spectrum for 5 mm dry ice pellets using the Holroyd et. al. (1978) sublimation rate formula compared against the effectivities of several glaciogenic aerosols. Cloud liquid water content is 1.5 g m^{-3}.

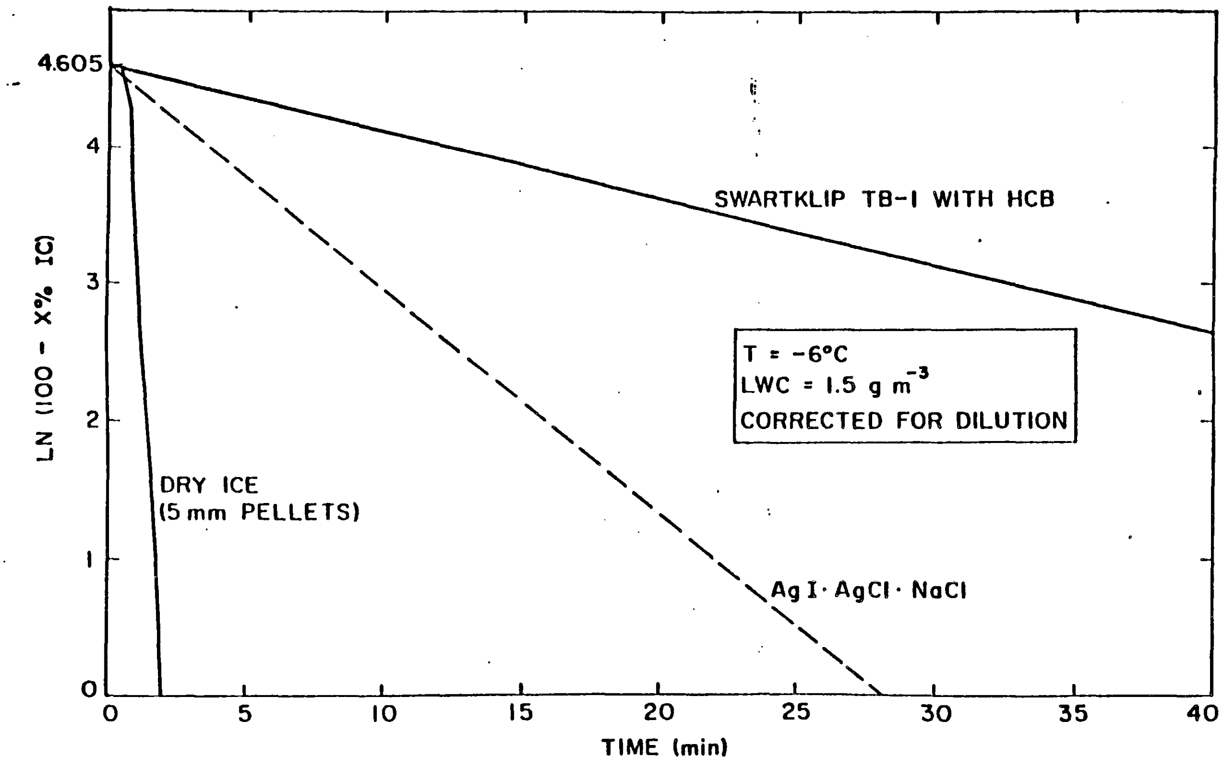

Figure 21. Kinetics plots comparing the rates of ice crystal production for dry ice to that of several glaciogenic aerosols at water saturation.

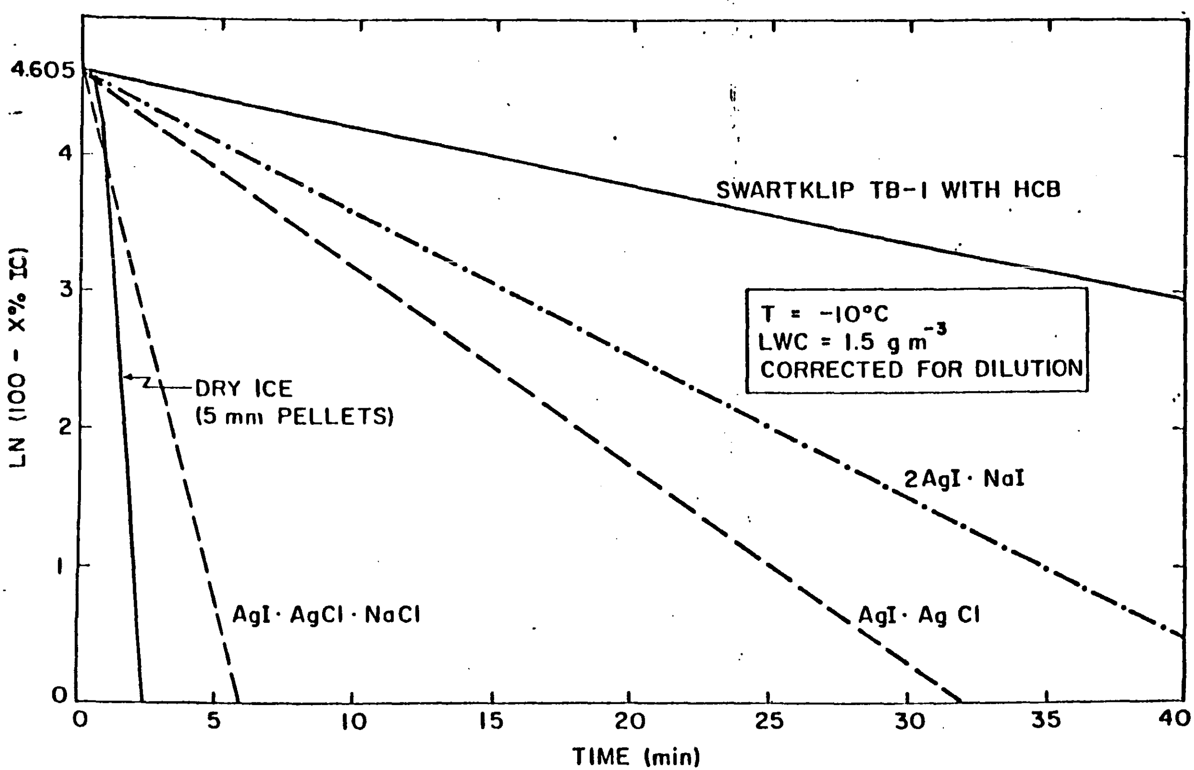

Figure 21 continued.

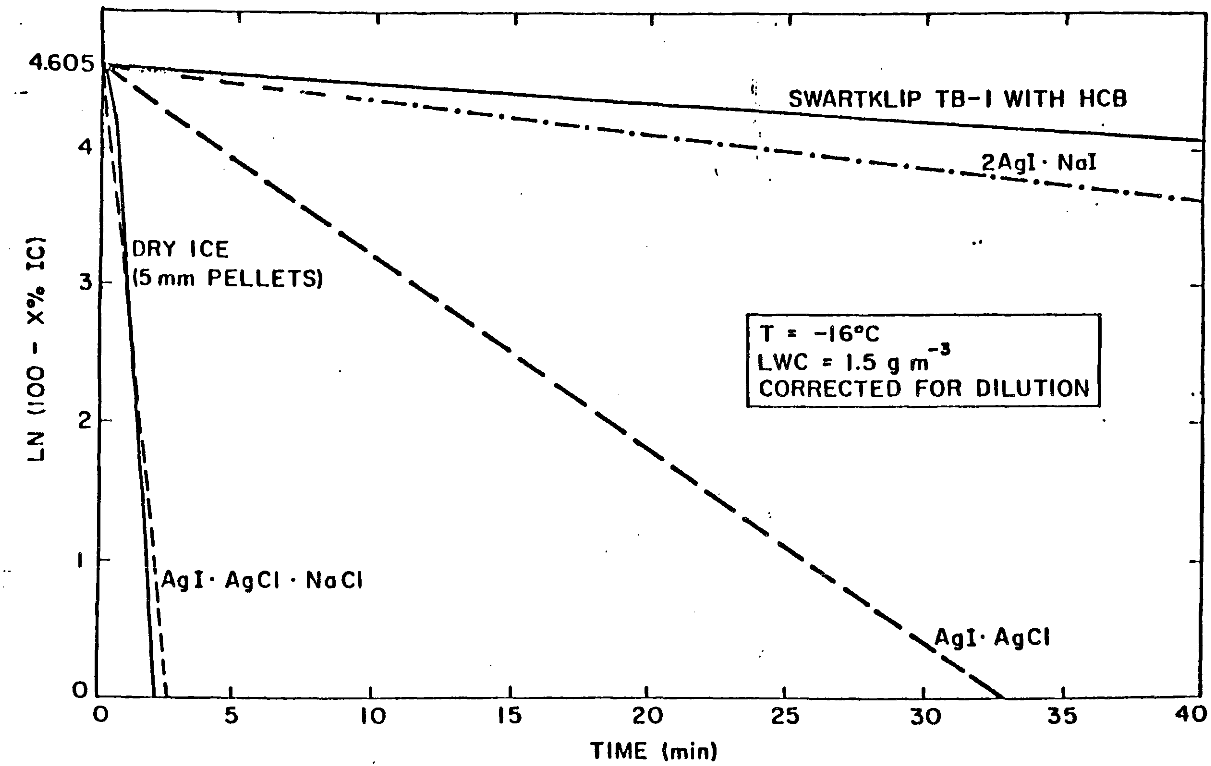

Figure 21 continued.

Table 19

TIME TO PRODUCE 90% OF TOTAL ICE CRYSTALS FOR DRY ICE AND SEVERAL GLACIOGENIC AEROSOLS AT WATER SATURATION

	Time (minutes) *			
Nucleant	T=-6°C	T=-10°C	T=-16°C	T=-20°C
Dry Ice (5 mm pellets)	1.5	1.6	1.2	1.4
AgI-AgCl-NaCl	17.8	3.9	1.8	0.9
AgI-AgCl	no data	15.8	16.5	13.0
2AgI-NaI	no data	22.0	89.0	no data
Swartklip TB-1 with HCB	47.0	56.0	329.0	no data

* all times corrected for chamber dilution effect

6.3 Implications for Weather Modification

Dry ice has several characteristics that make it an attractive on-top glaciogenic seeding agent in situations requiring very rapid pulses of artificial ice nucleation such as convective cloud modification through dynamic seeding:

* Dry ice has a warmer effectiveness threshold (up to at least -2 °C) than most glaciogenic aerosols allowing for the initiation of the ice phase lower in convective clouds which in turn permits more time for particle growth in the updraft.

* Although the effectiveness of dry ice appears to be moderately temperature dependent, it may be of sufficiently narrow range to make it relatively easy to achieve a desired artificial ice crystal concentration over large vertical temperature gradients.
* Since the mechanism of dry ice nucleation is droplet independent, the ice crystal yield from dry ice seeding in clouds with the same temperature and seeding conditions should be similar regardless of the cloud droplet concentrations and size distributions as long as the vapor supply is sufficient to prevent overseeding.
* The very rapid nucleation rate and subsequent rate of ice crystal production will produce a large microphysical response in the target cloud over a very short period of time. This can be crucial in the generation of latent heat release for a dynamical seeding effect.
* It can be dispersed in a continuous line along a flight track allowing for more ready dispersion as a seeding "curtain" compared to periodic injection of droppable pyrotechnics.
* The fall of dry ice pellets and the dispersion of the seeding plume in cumulus clouds can be realistically modeled and predicted.

* There can be no residual artificial ice nuclei from dry ice seeding to contaminate clouds other than the target.
* Logistically it is very suitable for airborne application. Sufficient amounts can be carried in light aircraft for the treatment of a large number of clouds. It is stable and safe. Unlike pyrotechnics or solution aerosols, it does not require explosives, combustion, nor the use of any hazardous chemicals.
* There are no real or perceived environmental hazards in using dry ice for cloud seeding.
* Dry ice is more economical than most commercially available glaciogenic aerosol seeding agents.
* Dry ice pellets can be easily manufactured on-site with commercially available equipment requiring no special skills or knowledge to operate.

Most important for weather modification experiments testing specific hypotheses of cloud microphysical responses to glaciogenic seeding is that the functioning of dry ice in terms of effectiveness and rates of ice crystal production is fairly well defined and understood. It can be accurately targetted and its dispersion reasonably estimated. As the simple model developed in the course of this research has shown, dry ice seeding in convective clouds can be numerically simulated with a good degree of skill.

6.4 Suggestions for Future Work

Unless a radically different experimental methodology is devised, the Isothermal Cloud Chamber may be unsuitable for conducting quantitative studies of dry ice nucleation due to the overseeding problems encountered in the course of this research. Obtaining a complete data set at a cloud liquid water content of 2.5 g m^{-3} would probably mitigate this problem to some extent, particularly in the -6 to -12 ^{o}C temperature range. However, it is much more difficult to minimize the temperature gradients in the chamber at this higher liquid water content and, more importantly, limits the usefulness of the results for comparative purposes since most other seeding agent characterizations have been conducted at 0.5 and 1.5 g m^{-3} liquid water contents only. Nevertheless, data could probably be obtained at the 2.5 g m^{-3} liquid water content which could conclusively answer the important questions regarding volume and fall speed effects.

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APPENDIX A

FORTRAN 77 LISTING OF SEEDING SIMULATION

```
C       PROGRAM SEEDING_SIMULATION
C
C***********************************************************************
C                                                                      *
C       Brian J. Morrison                                              *
C       July 1985 (Last updated 2Ø March 1986)                         *
C                                                                      *
C       Cansas International Corporation (Pty) Ltd.                    *
C       P.O. Box 1135                                                  *
C       Nelspruit 12ØØ                                                 *
C       Republic of South Africa                                       *
C                                                                      *
C       Purpose:  This program simulates the fall and sublimation of   *
C                 a cylindrical dry ice pellet in the updraft of a     *
C                 supercooled cumulus congestus cloud.  Measurements   *
C                 of 17 cumulus turrets over the eastern Transvaal of  *
C                 South Africa from an instrumented Learjet 24 are     *
C                 used to provide cloud property data to initialize    *
C                 and run the model.                                   *
C                                                                      *
C       Specifications:  Fortran 77; Data General MV4ØØØ Computer;     *
C                 AOS/VS Operating System                              *
C                                                                      *
C       Subroutines or functions required :                            *
C                                                                      *
C                 DYNAMIC_VISCOSITY, INTERPOLATE, MIXING_RATIO,        *
C                 MOIST_AIR_DENSITY, REYNOLDS_NUMBER, SALR2            *
C                 INTERPOLATE, VAPOR_PRESSURE, EFFECTIVENESS           *
C                                                                      *
C       References :                                                   *
C                                                                      *
C               Emmitt, G.D., D.S. Roos, and B.J. Morrison, 1984:  Dry *
C                  ice pellet experiments.  Report to the South        *
C                  African Water Research Commission, Pretoria, 6Ø pp.*
C                                                                      *
C               Haltiner, G.J., and F.L. Martin, 1957:  Dynamical and  *
C                  physical meteorology.  McGraw-Hill, New York.  47Ø  *
C                  pp.                                                 *
C                                                                      *
C               Lozowski, E.P., and B. Kochtubajda, 198Ø:  Theory and  *
C                  measurements of dry ice sublimation in clear air    *
C                  and simulated cloud.  Third WMO Sci. Conf. on Wea.  *
C                  Modif., Clermont-Ferrand, pp. 4Ø9-416.              *
C                                                                      *
C               Pruppacher, H.R., and J.D. Klett, 1978 :  Microphysics *
C                  of clouds and precipitation.  D. Reidel Publishing  *
C                  Co., Boston.  714 pp.  See page 418.                *
C                                                                      *
C               Rogers, R.R., 1979 :  A short course in cloud physics. *
C                  Pergamon Press, New York.  235 pp.  See page 21.    *
C                                                                      *
C***********************************************************************
C
        INTEGER SYSTEM_DATE(3), SYSTEM_TIME(3), IDATE(3), DELTA_T
        INTEGER TIME1(3), DELTA_T1, PRESS1, TEMP1, TAS1, EPSILON1,
     A          AVG_2DC_CONC1, MAX_2DC_CONC1
        INTEGER TIME2(3), DELTA_T2, PRESS2, TEMP2, TAS2, EPSILON2,
     A          AVG_2DC_CONC2, MAX_2DC_CONC2

        REAL MIXING_RATIO, NUSSELT_NUMBER, LS, MASS, MASS_PER_PELLET

        CHARACTER*3  MONTH(12)
        CHARACTER*55 SCRATCH
```

```
      DATA MONTH/ "Jan", "Feb", "Mar", "Apr", "May", "Jun",
     A            "Jul", "Aug", "Sep", "Oct", "Nov", "Dec"/
      REV = 1.Ø4

C
C----- Define Constants ---------------------------------------------------
C
      CP                 = Ø.24Ø           ! Sp. Heat dry air [cal/(g K)]
      PI                 = 4.Ø*ATAN(1.Ø)   ! Pi
      G                  = 981             ! Accel of gravity [cm/s**2]
      DRY_ICE_DENSITY    = 1.52            ! Pellet density [g/cm**3]
      T_DRY_ICE          = -1ØØ.Ø          ! Pellet surface temp [C]
      D_INITIAL          = Ø.9             ! Initial pellet diameter [cm]
      X_INITIAL          = Ø.8             ! Initial pellet length [cm]
      A                  = .2Ø             ! Constant
      NUM_BINS           = 1               ! Number of dispensing bins used
      DISPENSE_RATE      = Ø.19            ! Pellet dispensing rate per bin
                                           ! [kg/s]
      NUM_DENSITY_PELLETS = 13ØØ           ! Number density - pellets [/kg]
      WIDTHØ             = 1Ø5.Ø           ! Initial pellet plume width [m]

C
C----- Open listing file for output of results ------------------------
C
      OPEN(3, FILE = ":UDD:BRIAN:DRY_ICE:SEEDING_SIMULATION_RESULTS",
     A     STATUS = "FRESH", IOSTAT = IER)
      CALL ERRCODE (IER)

C
C----- Open data file containing Learjet data -------------------------
C
      OPEN(2, FILE = ":UDD:BRIAN:DRY_ICE:SEEDING_SIMULATION_DATA",
     A     MAXRECL = 56, PAD = "YES", STATUS = "OLD",
     B     IOINTENT = "INPUT", IOSTAT = IER)
      CALL ERRCODE (IER)
      READ (2, "(A55)") SCRATCH                  ! Skip header records
      READ (2, "(A55)") SCRATCH                  !  "     "      "
      READ (2, "(A55)") SCRATCH                  !  "     "      "

C
C----- Read in aircraft data for a seed pass and a repenetration pass --
C
1ØØ   READ (2,11Ø) IDATE
11Ø   FORMAT(I2,1X,I2,1X,I2)
      IF (IDATE(1) .EQ. 99) GO TO 9999         ! Test for end-of-file
      READ (2,12Ø) TIME1, DELTA_T1, PRESS1, TEMP1, TAS1, EPSILON1,
     A             UPDRAFT_SPEED1, AVG_2DC_CONC1, MAX_2DC_CONC1
      READ (2,12Ø) TIME2, DELTA_T2, PRESS2, TEMP2, TAS2, EPSILON2,
     A             UPDRAFT_SPEED2, AVG_2DC_CONC2, MAX_2DC_CONC2
12Ø   FORMAT (I2,1X,I2,1X,I2,1X,I2,1X,I3,1X,I3,1X,I3,1X,I4,1X,F4.1,
     A        9X,I6,1X,I6)

C
C----- Estimate aircraft altitude from static pressure using equations
C      for NACA standard atmosphere from Haltiner and Martin (1957).
C      This is only an approximation, but we are only interested in
C      relative changes
C
      Z1 = 443Ø8 * (1 - (PRESS1 / 1Ø13.25)**.19Ø23)   ! [meters]
      Z2 = 443Ø8 * (1 - (PRESS2 / 1Ø13.25)**.19Ø23)   ! [meters]
      DELTA_Z = Z2 - Z1                               ! [meters]

C
C----- Write banner to output listing file ----------------------------
```

```
C
          CALL DATE (SYSTEM_DATE)
          IDAY3   = SYSTEM_DATE(3)
          IMONTH3 = SYSTEM_DATE(2)
          IYEAR3  = SYSTEM_DATE(1)
          IF (IYEAR3.GT.1900) IYEAR3 = IYEAR3 - 1900
          CALL TIME (SYSTEM_TIME)
          TIME3 = SYSTEM_TIME(1) * 100 + SYSTEM_TIME(2) + SYSTEM_TIME(3) /
     a          100.0
          WRITE (3,"('<FF>')")
          WRITE (3,20) REV, IDAY3, MONTH(IMONTH3), IYEAR3, TIME3
20        FORMAT("SEEDING SIMULATION",6X,"Rev",F7.2,6X,
     A           "Run on ",I2,"-",A3,"-",I2," at ",F7.2," SAST",34X,
     B           "Cansas International Corp."//)

C
C----- Write aircraft data to output listing file ---------------------
C
          WRITE (3,150) IDATE(1), MONTH(IDATE(2)), IDATE(3)
150       FORMAT(/1X,I2,"-",A3,"-",I2,11X,"Time",4X,"DT",3X,"P",4X,
     A           "PALT",4X,"T",2X,"TAS",5X,"E",3X,"UP",3X,"LWJ",2X,
     B           2X,"LWF",3X,"2D",2X,"2DMAX"/)
          WRITE (3,160) 1, TIME1, DELTA_T1, PRESS1, Z1, TEMP1, TAS1,
     A                 EPSILON1, UPDRAFT_SPEED1, AVG_2DC_CONC1,
     B                 MAX_2DC_CONC1
          WRITE (3,160) 2, TIME2, DELTA_T2, PRESS2, Z2, TEMP2, TAS2,
     A                 EPSILON2, UPDRAFT_SPEED2, AVG_2DC_CONC2,
     B                 MAX_2DC_CONC2
160       FORMAT(1H ,9X,"PASS ",I1,I5,":",I2,":",I2,I4,I5,2X,F5.0,2X,I3,
     A           I5,2X,I4,F6.1,12X,I5,1X,I6)

C
C----- Initializations ------------------------------------------------
C
          DELTA_T = 2                        ! Time step [s]
          FALLV   = 0.0                      ! Fall velocity [cm/s]
          VTERM   = 10000.0                  ! Terminal velocity [cm/s]
          ELT     = 0.0                      ! Elapsed time [s]
          R       = D_INITIAL / 2.0          ! Pellet radius [cm]
          D       = R * 2.0                  ! Pellet diameter [cm]
          X       = X_INITIAL                ! Pellet length [cm]
          T       = TEMP1                    ! Temperature [C]
          DEW     = T                        ! Assume saturation in-cloud
          Z       = Z1                       ! Altitude [m]
          P       = PRESS1                   ! Pressure [mb]
          CALL DYNAMIC_VISCOSITY (T, DYN_VIS)  ! [g/(cm s)]

          TIME1_SECS = TIME1(1) * 3600.0 + TIME1(2) * 60.0 + TIME1(3) +
     A                 DELTA_T1 / 2.0
          TIME2_SECS = TIME2(1) * 3600.0 + TIME2(2) * 60.0 + TIME2(3) +
     A                 DELTA_T2 / 2.0
          TIME_LIMIT_SECS = TIME2_SECS - TIME1_SECS
          ELT = -1.0 * DELTA_T

C
C----- Listing File Headings ------------------------------------------
C
          WRITE (3,170) TIME_LIMIT_SECS
170       FORMAT (/10X,"Time between passes = ",F5.1," s"//)
          WRITE (3,180)
180       FORMAT (10X,"ELT",5X,"D",3X,"PELL VEL",2X,"UPDRAFT",3X,
     A             "FALLV",3X,"DELTAZ",7X,"Z",7X,"TC",5X,"DENSAIR",2X,
     B             "PLUME Z"/10X,"===",5X,"=",3X,"========",2X,"=======",
     C             3X,"=====",3X,"======",7X,"=",7X,"==",5X,"=======",
```

```
     D               2X,"======="//)

C*=====================================================================
C                                                                     *
C                        BEGIN PELLET FALL LOOP                      *
C                                                                     *
C----------------------------------------------------------------------

200       ELT = ELT + DELTA_T               ! ELT = elapsed time [sec]

          IF (ELT .GE. TIME_LIMIT_SECS) THEN
             WRITE (3,210)
210          FORMAT (12X," *** Pellet plume never reaches repenetration ",
     A                "altitude ***")
             GO TO 100
          END IF

          IF (ELT .EQ. 0) GO TO 250    ! Compute initial secondary params?

C
C----- Estimate updraft velocity by linear interpolation in time -------
C
          CALL INTERPOLATE (0, TIME_LIMIT_SECS, UPDRAFT_SPEED1,
     A                      UPDRAFT_SPEED2, ELT, UPDRAFT_SPEED)
          UPDRAFT_SPEED = UPDRAFT_SPEED * 100.0        ! [m/s] to [cm/s]

C
C----- Compute saturated adiabatic lapse rate -------------------------
C
          DT_DZ = SALR2 (T, P)                               ! [C/km]
          DT_DZ = DT_DZ / 1000.0                             ! [C/m]

C
C----- Cylindrical dry ice pellet still air fall speed ----------------
C
          TERM1 = ((DRY_ICE_DENSITY - AIR_DENSITY) / DRY_ICE_DENSITY) * G
          TERM2 = (1.0 / (2.0 * MASS)) * AIR_DENSITY * FALLV * FALLV *
     A             AREA * CD
          FALLV = (TERM1 + TERM2) * DELTA_T                  ! [cm/s]

C
C----- Cylindrical dry ice pellet terminal velocity (Lozowski and
C      Kochtubajda, 1980)
C
          VTERM = ((PI * 2 * R * DRY_ICE_DENSITY * G) / (2 * CD *
     A            AIR_DENSITY)) ** 0.5
          IF (FALLV .GT. VTERM) FALLV = VTERM

C
C----- Compute fall speed of pellet in updraft and vertical displacement
C
          W       = FALLV - UPDRAFT_SPEED                    ! [cm/s]
          DELTA_Z = (W / 100.0) * DELTA_T                    ! [m/s]
          Z       = Z - DELTA_Z

C
C----- List out iteration results.  Do another iteration if pellet is
C      above repenetration altitude
C
          IF (Z .GT. Z2) THEN
             WRITE (3,220) ELT, D, FALLV/100.0, UPDRAFT_SPEED/100.0,
     A                  W/100.0, DELTA_Z, Z, T, AIR_DENSITY
220          FORMAT (7X,F6.1,3X,F4.2,3X,F6.2,3X,F6.2,3X,F6.2,3X,F6.2,3X,
     A            F7.1.3X,F5.1,3X,F8.6)
```

```
            GO TO 24Ø
         END IF
C
C----- Pellet is below repenetration altitude.  Assume ice crystal plume
C       is advected upwards with the updraft interpolated linearly in
C       time.  If plume does not overshoot repenetration altitude at
C       repenetration time, assume it is close enough to repenetration
C       altitude to exit pellet fall loop
C
         ZZ    = Z
         I     = ELT + Ø.5
         LIMIT = TIME_LIMIT_SECS + Ø.5

         DO NØ = I, LIMIT, DELTA_T
            XNØ       = NØ
            CALL INTERPOLATE (Ø, TIME_LIMIT_SECS, UPDRAFT_SPEED1,
     A                        UPDRAFT_SPEED2, XNØ, WW)
            DELTA_ZZ = WW * DELTA_T
            ZZ       = ZZ + DELTA_ZZ
         END DO

         WRITE (3,225) ELT, D, FALLV/1ØØ.Ø, UPDRAFT_SPEED/1ØØ.Ø,
     A                 W/1ØØ.Ø, DELTA_Z, Z, T, AIR_DENSITY,ZZ
225      FORMAT (7X,F6.1,3X,F4.2,3X,F6.2,3X,F6.2,3X,F6.2,3X,F6.2,3X,
     A           F7.1,3X,F5.1,3X,F8.6,3X,F7.1)

         IF (ZZ .LE. Z2) GO TO 1ØØØ                ! Exit pellet fall loop?

C
C----- Pellet plume has overshot repenetration altitude at repenetration
C       time.  So, let's carry on with pellet fall loop by computing
C       parameters necessary for pellet fall computations
C
C----- Compute temperature, dewpoint, and pressure altitude ------------
C
24Ø      T   = T + (DT_DZ * DELTA_Z)                                  ! [C]
         DEW = T                                                      ! [C]
C
         P   = 1Ø13.25 * (1.Ø - (Z / 443Ø8.Ø))**(1.Ø / Ø.19Ø23)  ! [mb]

C
C----- Compute dynamic viscosity of air --------------------------------
C
25Ø      CALL DYNAMIC_VISCOSITY (T, DYN_VIS)          ! [g / (cm s)]

C
C----- Compute thermal conductivity of air (Pruppacher and Klett, 1978)
C
         THERM_CON = (5.69 + Ø.Ø17 * T) * 1E-5        ! [cal / (cm s C)]

C
C----- Compute specific heat of moist air (Rogers, 1979) ---------------
C
         XMIX = MIXING_RATIO (P, DEW)                  ! [g/kg]
         XMIX = XMIX / 1ØØØ.Ø                          ! [g/g]
         CPM = CP * (1.Ø + Ø.8 * XMIX)                 ! [cal / (g K)]

C
C----- Compute Prandtl Number for air (Emmitt, et. al., 1984) ----------
C
         PRANDTL_NUMBER = CPM * DYN_VIS / THERM_CON

C
```

```
C----- Compute Reynolds Number ------------------------------------------
C
        CALL MOIST_AIR_DENSITY (P, T, DEW, AIR_DENSITY)      ! [g/m**3]
        AIR_DENSITY = AIR_DENSITY * 1E-6                     ! [g/cm**3]
        RE = REYNOLDS_NUMBER (FALLV, R, AIR_DENSITY, DYN_VIS)

C
C----- Compute drag coefficient using empirical polynomial from clear
C      air fall experiments
C
        CD = 1.99719 - (3.58661E-4 * RE) + (2.25381E-8 * RE * RE)

        IF (ELT .EQ. Ø) GO TO 29Ø

C
C----- Compute Nusselt Number (Lozowski and Kochtubajda, 198Ø) ---------
C
        NUSSELT_NUMBER = Ø.37 + Ø.37 * (PRANDTL_NUMBER**(1.Ø / 3.Ø)) *
     A                   (RE**Ø.5)

C
C----- Compute latent heat of sublimation for dry ice [cal/g] ----------
C
        LS = 45.3 + (52.8681 - Ø.961288 * T - 7.Ø8333E-3 * T * T)

C
C----- Compute sublimation rate (Emmitt, et. al., 1984) and then new
C      pellet diameter and length
C
        D = 2.Ø * R
        DD_DT = ((-4.Ø * X * THERM_CON * NUSSELT_NUMBER) /
     A          (DRY_ICE_DENSITY * LS)) * ((T_DRY_ICE - T) /
     B          (2.Ø * D * X + A * D * D))                   ! [cm/s]
        D = D - (DD_DT * DELTA_T)                            ! [cm]
        R = D / 2.Ø                                          ! [cm]
        DL_DT = A * DD_DT                                    ! [cm]
        X = X - (DL_DT * DELTA_T)                            ! [cm]

C
C----- Compute new pellet mass -----------------------------------------
C
29Ø     MASS = PI * R * R * X * DRY_ICE_DENSITY

C
C----- Compute equivalent area of half cylinder ------------------------
C
        AREA = (2.Ø * R) * X                                 ! [cm**2]
        GO TO 2ØØ

C-----------------------------------------------------------------------
C                                                                      *
C                       END PELLET FALL LOOP                           *
C                                                                      *
C=======================================================================

C=======================================================================
C                                                                      *
C            SUCCESSFULL INTERCEPTION OF ICE CRYSTAL PLUME             *
C                                                                      *
C-----------------------------------------------------------------------

C
C----- Determine mass of dry ice sublimed (Emmitt, et. al., 1984) over
C      last time step
```

```
C
1000     DM_DT = (PI * DRY_ICE_DENSITY / 4) * (2 * D * X + A * D * D) *
     A           DD_DT
         MASS_PER_PELLET = DM_DT * DELTA_T

C
C----- Compute total number of pellets dropped in seeding run ----------
C
         PELLETS = NUM_DENSITY_PELLETS * DISPENSE_RATE * NUM_BINS *
     A             DELTA_T1

C
C----- Compute total dry ice mass sublimed [g] in last time step -------
C
         TOTAL_MASS = MASS_PER_PELLET * PELLETS

C
C----- Compute depth of plume volume [m] over last time step -----------
C
         DEPTH = (FALLV/100.0) * DELTA_T

C
C----- Compute plume width [m] using turbulent dispersion formula ------
C
         XEPSILON1 = EPSILON1
         XEPSILON2 = EPSILON2
         CALL INTERPOLATE (0, TIME_LIMIT_SECS, XEPSILON1, XEPSILON2,
     A                     ELT, EPSILON)
         EPSILON = (EPSILON + XEPSILON2) / 2.0
         XTIME_SECS = (TIME_LIMIT_SECS - ELT) + 1
         WIDTH = WIDTH0 + 2.0 * (SQRT(EPSILON * XTIME_SECS**3.0) / 100.0)

C
C----- Compute plume length [m] using aircraft TAS ---------------------
C
         PLUME_LENGTH = TAS1 * DELTA_T1

C
C----- Compute plume volume over last time step ------------------------
C
         PLUME_VOLUME = DEPTH * WIDTH * PLUME_LENGTH              ! [m**3]
         PLUME_VOLUME = PLUME_VOLUME * 1000.0                     ! [l]

C
C----- Compute dry ice effectiveness from 2D-C concentrations ----------
C
         EFF_2D_AVG = (AVG_2DC_CONC2 - AVG_2DC_CONC1) * PLUME_VOLUME /
     A                TOTAL_MASS
         EFF_2D_MAX = (MAX_2DC_CONC2 - MAX_2DC_CONC1) * PLUME_VOLUME /
     A                TOTAL_MASS

C
C----- For simulation nucleation temp, get lab effectiveness value -----
C
         CALL EFFECTIVENESS (T, EFF_LAB)

C
C----- Compute simulation predicted ice crystal conc using lab effect
C      value
C
         XNUMIC = TOTAL_MASS * EFF_LAB
         CONC = XNUMIC / PLUME_VOLUME                        ! [no/liter]

C
```

```
C----- Write results to listing file -----------------------------------
C
      WRITE (3,3ØØØ) D, X, T
3ØØØ  FORMAT (///" Final Values ..."//" Diam = ",F4.2," cm",16X,
     A        "Length = ",F4.2," cm",14X,"T = ",F5.1," C")
      WRITE (3,3Ø1Ø) DEPTH, WIDTH, PLUME_LENGTH
3Ø1Ø  FORMAT (" Pl depth = ",F5.1," m",12X,"Pl width = ",F6.1," m",
     A        11X,"Pl length = ",F6.1," m")
      WRITE (3,3Ø2Ø) PLUME_VOLUME, PELLETS, TOTAL_MASS
3Ø2Ø  FORMAT (" Pl volume = ",E9.3," l",7X,"N pellets = ",F6.Ø,
     A        12X,"Tot mass = ",F6.2," g")
      WRITE (3,3Ø3Ø) NUM_BINS, DM_DT*1ØØØ.Ø
3Ø3Ø  FORMAT (" Bins = ",I1,22X,"Sub rate = ",F6.3," mg/s")
      WRITE (3,3Ø4Ø) EFF_2D_AVG, EFF_2D_MAX
3Ø4Ø  FORMAT (" EFF_2D_AVG = ",E7.2,1ØX,"EFF_2D_MAX = ",E7.2)
      WRITE (3,3Ø5Ø) EFF_LAB, XNUMIC, CONC
3Ø5Ø  FORMAT (" EFF_LAB = ",E7.2,13X,"Num IC = ",E8.3,13X,"Conc = ",
     A        F6.1///)
      GO TO 1ØØ

9999  STOP
      END
```

```
      SUBROUTINE DYNAMIC_VISCOSITY (TEMPC, DYN_VIS)
C
C***********************************************************************
C
C     B.J. Morrison
C     July 1985 (Last updated 4 July 1985)
C
C     Purpose :  Computes the dynamic viscosity of air.
C
C     Specs   :  Fortran 77 --- MV4ØØØ Computer --- AOS/VS Oper System
C
C     Variables passed to this subroutine :
C
C             TEMPC      Ambient temperature [C]
C
C     Variables returned from this subroutine :
C
C             DYN_VIS    Dynamic viscosity [Poise = (g / s cm)]
C
C     References :
C
C             Pruppacher, H.R., and J.D. Klett, 1978 :  Microphysics
C                 of clouds and precipitation.  D. Reidel Publishing
C                 Co., Boston.  714 pp.  See page 323.
C
C     Notes        :
C
C             The formulae used in this subroutine were based on data
C             from the Smithsonian Meteorological Tables.  Accuracy
C             is +- Ø.ØØ2E-4 poise.  Note that pressure does not
C             affect the dynamic viscosity of air and that water
C             vapor has a negligible effect.
C
C***********************************************************************
C
      IF (TEMPC .GE. Ø) THEN
         DYN_VIS = (1.718 + Ø.ØØ49 * TEMPC) * 1E-4     ! [g/(s cm)]
      ELSE
         DYN_VIS = (1.718 + Ø.ØØ49 * TEMPC - 1.2E-5 *
     A             TEMPC * TEMPC) * 1E-4               ! [g/(s cm)]
      END IF
      RETURN
      END
```

```
      SUBROUTINE INTERPOLATE(XHI,XLO,YHI,YLO,X,Y)
C
C**********************************************************************
C
C     B.J. MORRISON
C     SEPT 1984 (LAST REVISED 27 SEPT. 1984)
C
C           THIS IS A SIMPLE LINEAR INTERPOLATION ROUTINE.  GIVEN
C           XHI, XLO, X, YHI, AND YLO, IT INTERPOLATES FOR Y.
C
C**********************************************************************
C
      Y=YHI-(YHI-YLO)*((XHI-X)/(XHI-XLO))
      RETURN
      END
```

```
      REAL FUNCTION MIXING_RATIO (PRESS,TEMPC)
C
C**********************************************************************
C
C     B. J. Morrisor
C     February 1985
C
C     Cansas International Corp. (PTY) Ltd.
C     P.O. Box 1135
C     Nelspruit 12ØØ
C     REPUBLIC OF SOUTH AFRICA
C
C     Purpose  :  Computes mixing ratio or saturation mixing ratio
C
C     Specs    :  FORTRAN 77 --- MV4ØØØ COMPUTER --- AOS/VS OP SYS
C
C     Subroutines or functions called :  NONE
C
C     Arguments:
C
C           PRESS --------> Pressure (mb)
C           TEMPC --------> Dew point (C) to get Mixing Ratio or
C                           Temperature (C) to get Saturation Mixing
C                           Ratio.
C
C     Returned value:  Mixing Ratio or Saturation Mixing Ratio (g/kg)
C          depending on TEMPC above.  NOTE:  MIXING_RATIO must be
C          declared as a REAL variable in the calling program.
C
C     NOTE:  Vapor pressure equation taken from subroutine
C            VAPOR_PRESSURE.
C
C**********************************************************************
C
      VAPOR_PRESSURE=6.112*EXP((17.67*TEMPC)/(TEMPC+243.5))
      MIXING_RATIO=(622.Ø*VAPOR_PRESSURE)/(PRESS-VAPOR_PRESSURE)
      RETURN
      END
```

```
      SUBROUTINE MOIST_AIR_DENSITY (PRESS_MB, TEMPC, DEWC, AIR_DENSITY)
C
C************************************************************************
C
C       B.J. Morrison
C       July 1985 (Last revised 4 July 1985)
C
C       Purpose :  Computes density of moist air.
C
C       Specs   :  Fortran 77 --- MV4000 Computer --- AOS/VS Oper System
C
C       Subroutines or subprograms required :
C
C               VAPOR_PRESSURE (from CIC MET Library)
C
C       Variables passed to this subroutine :
C
C               PRESS_MB        Ambient air pressure [mb]
C               TEMPC           Dry-bulb temperature of moist air [C]
C               DEWC            Dewpoint of moist air [C]
C
C       Variables returned from this subroutine :
C
C               AIR_DENSITY     Moist air density [g/m**3]
C
C       NOTE  :  For saturated air set DEWC = TEMPC.
C
C       References :
C
C               Wallace, J.M., and P.V. Hobbs, 1977 :  Atmospheric
C                   science.  Academic Press, Inc., New York.  467 pp.
C                   See page 52.
C
C************************************************************************
C
C               CONSTANTS
C
        RD = 287.0                  !Indiv gas con dry air [J/(kg K)]
C
C               COMPUTATIONS
C

        CALL VAPOR_PRESSURE (DEWC, E_MB)         !Comp vap press [mb]
        E_PA = E_MB * 100                        !Convert mb to Pa

        PRESS_PASCALS = PRESS_MB * 100           !Convert mb to Pa
        TEMPK = TEMPC + 273.15                   !Convert C to K

        TERM1 = PRESS_PASCALS / (RD * TEMPK)
        TERM2 = 1.0 - (E_PA / PRESS_PASCALS) * (1.0 - 0.622)
        AIR_DENSITY = TERM1 * TERM2              !kg/m**3
        AIR_DENSITY = AIR_DENSITY * 1000         !kg/m**3 to g/m**3

        RETURN
        END
```

```
      FUNCTION REYNOLDS_NUMBER (SPEED, RADIUS, AIR_DENSITY,
     A                             DYNAMIC_VISCOSITY)
C
C***********************************************************************
C
C       B.J. Morrison
C       July 1985 (Last updated 4 July 1985)
C
C       Purpose  :  Computes the Reynolds Number for a falling object
C                   in air.  This is the non-dimensional ratio of
C                   the inertial force to the viscous force in fluid
C                   motion.  It is of importance in the theory of
C                   hydrodynamic stability and the origin of turbulence.
C                   It is usefull in establishing flow similarity among
C                   differing size scales.
C
C       Variables passed to this function :
C
C               SPEED                Fall speed of object [cm/s]
C               RADIUS               Characteristic radius of object [cm]
C               AIR_DENSITY          Density of ambient air [g/cm**3]
C               DYNAMIC_VISCOSITY    Dynamic viscosity of ambient air
C                                    [g/(cm s)]
C
C***********************************************************************
C
      REYNOLDS_NUMBER = 2.Ø * SPEED * RADIUS * AIR_DENSITY /
     A                  DYNAMIC_VISCOSITY
      RETURN
      END
```

```
      SUBROUTINE VAPOR_PRESSURE (TEMPC,E)
C
C***********************************************************************
C
C       B.J. MORRISON AND G. MORGAN
C       NOVEMBER 1984 (LAST REVISED 5 NOV 1984)
C
C       PURPOSE  :  THIS SUBROUTINE COMPUTES VAPOR PRESSURE GIVEN DEW-
C                   POINT TEMPERATURE IN DEGREES C OR SATURATION VAPOR
C                   PRESSURE GIVEN TEMPERATURE IN DEGREES C.
C
C       SPECS    :  FORTRAN 77 --- MV4ØØØ COMPUTER --- AOS/VS OP SYSTEM
C
C       SUBROUTINES CALLED :  NONE
C
C       INPUT VARIABLES:
C
C          TEMPC --->  TEMP (C) OR DEWPOINT (C)
C
C       RETURNED VARIABLES:
C
C          E ------->  SAT VAPR PRESSURE (MB) OR VAPOR PRESSURE (MB)
C
C
C       REFERENCES:
C
C               Bolton, D., 198Ø:  The computation of equivalent
C                  potential temperature.  MON. WEA. REV., 1Ø8,
C                  1Ø46-1Ø53.
C
C***********************************************************************
C
       E=6.112*EXP((17.67*TEMPC)/(TEMPC+243.5))
       RETURN
       END
```

```
      FUNCTION SALR2 (TEMPC,PRESS_MB)
C
C***********************************************************************
C
C       G. Morgan and B.J. Morrison
C       March 1985 (Last revised 21 March 1985)
C
C       Cansas International Corp. (Pty) Ltd.
C       P.O. Box 1135
C       Nelspruit 1200
C       REPUBLIC OF SOUTH AFRICA
C
C       Purpose :  Computes an approximate saturated adiabatic lapse
C                  rate for a given temperature and pressure.
C
C       Specs   :  Fortran 77 --- MV4000 Computer --- AOS/VS Oper System
C
C       Subroutines or special functions required:  Real function
C               MIXING_RATIO
C
C       Variables passed to this subroutine from calling program:
C
C               TEMPC           Temperature in degrees C
C               PRESS_MB        Pressure in millibars
C
C       Variables returned:
C
C               SALR2           Saturated adiabatic lapse rate in
C                               degrees C per km.
C
C***********************************************************************
C
      REAL LV,MIXING_RATIO
C
C             CONSTANTS
C
      CP=1004.0         !Specific heat of dry air at con P [J/(K kg)]
      RD=287.0          !Gas constant dry air [J/(K kg)]
      RV=461.0          !Gas constant for water vapor [J/(K kg)]
C
C             CONVERSIONS
C
      TEMPK=TEMPC+273.15
C
C             COMPUTE LATENT HEAT OF VAPORIZATION AS FUNCTION OF TEMP
C
      GAMMA=0.167+(TEMPK*3.67E-4)
      LV=597.3*((273.15/TEMPK)**GAMMA)            ![cal/g]
      LV=LV*4.18684*1000.0                        ![J/kg]
C
C             COMPUTE SATURATED MIXING RATIO
C
      WS=MIXING_RATIO(PRESS_MB,TEMPC)             !Sat mixing ratio [g/kg]
      WS=WS/1000.0                                !Sat mixing ratio [g/g]

C
C             COMPUTE PSUEDOADIABATIC LAPSE RATE
C
      TERM1=1.0+((LV*WS)/(RD*TEMPK))
      TERM2=1.0+((LV*LV*0.622*WS)/(RD*TEMPK*TEMPK*CP))
      SALR2=9.8*TERM1/TERM2

      RETURN
      END
```

```
      SUBROUTINE EFFECTIVENESS (TC, E)
C
C*********************************************************************
C
C       B.J. Morrison
C       July 1985
C
C       Purpose :  Computes dry ice effectiveness value for a given
C                  temperature based on laboratory results.
C
C       Specs   :  Fortran 77 --- MV4ØØØ Computer --- AOS/VS Oper System
C
C       Subroutines or subprograms required :  INTERPOLATE
C
C       Variables passed to this subroutine :
C
C               TC      Cloud Temperature [C]
C
C       Variables returned from this subroutine :
C
C               E       Dry ice effectiveness [Ice crystals / g dry ice]
C
C*********************************************************************
C
      DIMENSION T(12), EFF(12)
      DATA T/-2.Ø,-3.Ø,-4.Ø,-5.Ø,-6.Ø,-8.Ø,-9.Ø,-1Ø.Ø,-12.Ø,-15.Ø,
     A       -16.Ø,-2Ø.Ø/
      DATA EFF/Ø.22,Ø.97,1.2,1.5,2.2,2.3,2.5,2.8,3.6,3.2,4.7,8.9/

      N = 12
      DO NØ = 1, N-1
         IF (TC .LE. T(NØ) .AND. TC .GT. T(NØ+1)) THEN
            CALL INTERPOLATE (T(NØ), T(NØ+1), EFF(NØ), EFF(NØ+1),
     A                        TC, E)
            E = E * 1.ØE12
            RETURN
         END IF
      END DO

      WRITE (3,1ØØ)
1ØØ   FORMAT (//" **** Final temp outside lab effectiveness range! "/)
      E = Ø
      RETURN
      END
```