Silver Concentration in Rainwater from Seeded and Nonseeded Florida Cumuli: 1973–1975 Results

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ABSTRACT

The main objective of the Florida Area Cumulus Experiments was to seed groups of supercooled tropical cumulus clouds to convert enough water to ice so that sufficient heat could be released to enhance cloud growth and augment rainfall over a specific south Florida target area. During three consecutive summers (1973–75), subprograms to monitor the rainwater silver concentrations were conducted. Results showed that rainwater silver concentrations can be detected in significantly higher concentrations in seeded vs nonseeded samples with a carefully positioned network and with precise care in the preparation, collection, handling, storage and analysis of the samples. In addition, the silver concentrations in the seeded rainwater samples are low enough to present no ecological or environmental problems whatsoever under the present seeding rates in south Florida.

Stratification of the rainwater silver concentrations, using two different trajectory techniques, showed silver concentrations collected under continental influenced regimes to be almost identical to those collected under seeding conditions. This is a crucial result since, if rainwater silver concentrations are being used to determine whether the seeding material is being targeted in the optimum areas for maximum seeding efficiency, then the results may be biased under these continental influenced regimes.

1. Introduction

The dynamic seeding technique used in NOAA's Florida Area Cumulus Experiment (FACE) relies upon the massive infusion of silver iodide (AgI) nucleant into supercooled cumulus towers in order to induce, through the conversion of water to ice, additional buoyancy and, with attendant cloud growth and merger, greater convective organization and a more efficient processing of the available moisture. Pyrotechnics emitting either 50, 70 or 100 g of AgI effluent have been used during the FACE program (Sax *et al.*, 1977), the general procedure being direct injection into the active, moist updraft regions of suitable towers at about the −10°C isotherm level. While in the updraft the pyrotechnic release rate generally has been one flare per 150–200 m, resulting in the injection of the order of 1 kg of AgI into a typical tower of updraft diameter about 2 km. Generally between 150 and 300 pyrotechnics were released on a given seeded GO day, thereby resulting in the order of 10 kg of AgI being released into the atmosphere during the approximately 6 h of afternoon experimentation. The project was randomized by day on a double blind basis, with the scientists responsible for the flare release not knowing if a seed decision had been drawn. All selected clouds on GO days were treated consistently in terms of aircraft penetration procedures, although on roughly half the days no actual release of AgI occurred. A complete description of the rationale, theory and procedures of FACE has been provided by Woodley and Sax (1976).

Prior to the beginning of the FACE 1973 field season, it was decided that a subprogram would be undertaken to monitor the changes in the rainwater silver concentrations resulting from dynamic seeding activities. A number of silver scavenging programs have been conducted in several countries through the collection of rain, hail and snow samples. These programs showed a great deal of variability in manner of sample collection and analysis, along with interpretation of the data. Table 1 lists the known experimental silver tracer studies conducted through 1976; this table giving each study's principal investigator(s), location, types of precipitation samples collected, silver analytical technique, silver detection limit and the mean or range of silver concentrations found in the seeded or no-seed samples. A more detailed discussion of each of these studies with emphasis on each program's successes and failures has been overviewed by Wisniewski (1977, 1979).

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TABLE 1. Synopsis of silver tracing studies.

References	Study location	Type of samples collected	Ag analytical technique	Ag detection limit (ng ℓ^{-1})	Mean or range of Ag concentrations (ng ℓ^{-1})
Warburton (1963)	New South Wales, Australia	Surface rain	Ion-exchange	5	Seeded $\sim$10 Nonseeded <5
Warburton and Maher (1965)	New South Wales, Australia	Surface rain	Ion-exchange	3	Seeded $\sim$10 Nonseeded <3
Schaefer and Fuquay (1965)	Yellowstone Park, Wyoming Steamboat Springs, Colorado	Surface snow	Neutron activation	5	Seeded $\sim$2000 Nonseeded $\sim$30
Bollay (1965)	Western United States	Surface rain and snow	Neutron activation	5	Seeded $\sim$20 to 200 Nonseeded $\sim$5 to 20
Warburton and Young (1968a, b)	Five western United States sites	Surface hail, rain and snow	Neutron activation	3	Seeded $\sim$6000 to 20 Nonseeded $\sim$10
Parungo and Robertson (1969)	Park Range, Colorado	Surface snow	Atomic absorption	10	Seeded $\sim$1000 Nonseeded $\sim$20
Ostlund and Stearns (1970)	South Florida	Aircraft rain	Neutron activation and atomic absorption	1000	Seeded <1000
Warburton and Owens (1972)	Lake Erie	Surface snow	Neutron activation	2	Seeded $\sim$361 Nonseeded $\sim$19
Warburton (1973)	Central Alberta, Canada	Surface hail and rain	Neutron activation	2	Seeded $\sim$31 Nonseeded $\sim$5
Gatz (1973)	South Illinois	Surface rain	Atomic absorption	2	Nonseeded $\sim$73
Summers (1973)	Central Alberta, Canada	Surface hail and rain	Atomic absorption	100	Seeded $\sim$3800 to <100 Nonseeded <100
Wisniewski (1974 a, b)*	South Florida	Aircraft and surface rain	Atomic absorption	2	Seeded aircraft $\sim$160 Seeded surface $\sim$69 Nonseeded aircraft $\sim$518 Nonseeded surface $\sim$51
Zacharuk and Goyer (1975)	Central Alberta, Canada	Surface hail and rain	Atomic absorption	3	Seeded $\sim$16 Nonseeded $\sim$3
Zacharuk and Goyer (1976)	Central Alberta, Canada	Surface hail and rain	Atomic absorption	3	Seeded $\sim$14 Nonseeded $\sim$3
Zacharuk and Erb (1977)	Central Alberta, Canada	Surface hail and rain	Atomic absorption	3	Seeded $\sim$15 Nonseeded $\sim$3
Wisniewski (1977)*	South Florida	Surface rain	Atomic absorption	2	Nonseeded $\sim$51
Wisniewski (1977)*	South Florida	Aircraft and surface rain	Atomic absorption	2	Aircraft seeded $\sim$71 Surface seeded $\sim$81 Aircraft nonseeded $\sim$48 Surface nonseeded $\sim$49

* These studies are discussed in this paper.

Regarding the specific FACE operations, it can be calculated using measured values of the mean AgI particle diameter ($\sim$0.03 μm) emitted from FACE pyrotechnics and assuming a uniform distribution of material throughout a 10^{15} cm^3 volume of cloud updraft, that the order of 1000 particles cm^{-3} should be available for scavenging through diffusion processes. If only 10% of these particles are scavenged in 10 min (Wisniewski, 1977), then the concentration of AgI particulate matter collected by the cloud water is of the order 100 cm^{-3}. This converts to a silver (Ag) mass content of the order 10^{-15} g cm^{-3} which, when consumed by a cloud water content of the order 10^{-6} g cm^{-3} (1 g m^{-3}), results in a silver concentration in water of the order 1000 ng ℓ^{-1}. Although further dilution due to coalescence with "uncontaminated" water in other portions of the cloud is probable, it is not unreasonable to expect that the use of atomic absorption (AA) technology, which prior to the start of FACE 1973 had advanced to the point of allowing a silver detection sensitivity of 1 ng ℓ^{-1}, would enable us readily to detect a seeding signal several orders of magnitude above background. It should thus be possible to measure reliably the concentration of silver present in the rainwater collected both from individual seeded clouds (mobile aircraft and surface samples) and from fixed surface sites located within the 13 000 km^2 FACE target area (Fig. 1).

Following the conclusion of FACE 1973, it was decided to collect and analyze rainwater samples from fixed surface locations for silver content during the summer of 1974, a year in which no cloud seeding

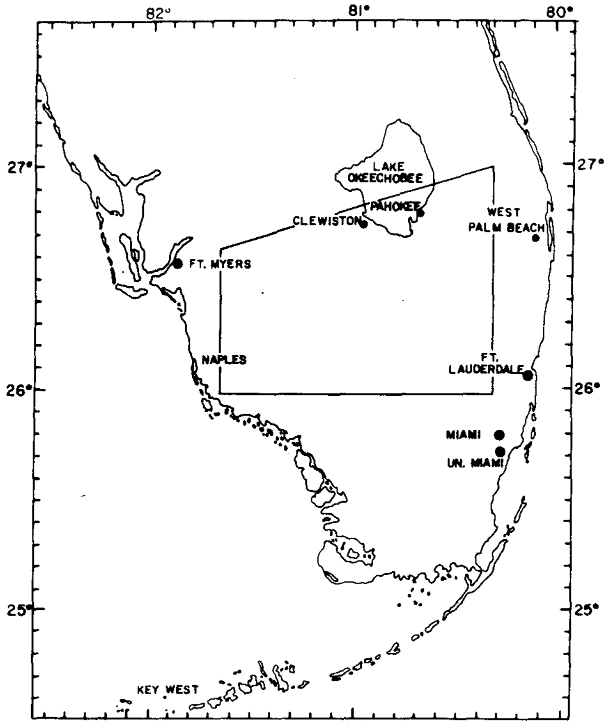

FIG. 1. The 13 000 km² quadrilateral experimental area in which all samples were collected.

activity took place anywhere in south Florida, for the purpose of acquiring background data. The rainwater collection program continued during the FACE 1975 field season. Data relating to silver concentration in rainwater in the south Florida area, therefore, are available from a nonseeded background (control) season sandwiched between two seasons with randomized seeding. In this paper, we examine the 3 year (1973–75) data set with the purpose of determining whether or not a significant detectable increase in silver in rainwater can be correlated with dynamic seeding activities.

2. Procedure and operations

During the FACE field programs of 1973 and 1975, rainwater was collected in three different ways. First, an aircraft (the NRL S-2D and NOAA-DC6 during FACE 1973 and the NCAR 304-Delta during FACE 1975) operated just below the cloud-base level (600 m) in an attempt to collect rainwater from individually treated clouds with a scoop mounted on the top front position of the aircraft fuselage (Fig. 2). The aircraft, vectored into position underneath the cloud system being seeded[1] by the upper level (6 km) NOAA aircraft, carried out repeated penetrations on reciprocal headings through the rainshaft. With the scoop presenting a cross-sectional area of approximately 100 cm² and titled upward 15°, it was possible to collect between 250–1000 ml of rainwater on each traverse through moderate or heavy precipitation. In addition to collecting rainwater, the scoop also collected cloud water and aerosols; therefore, comparison of the aircraft results to those obtained at the surface (described below) should be made with the above considerations in mind.

The second method of collecting rainwater samples was through utilizing air-to-ground communications which permitted the mobile collection of rainwater at the surface. On these occasions, a van was maneuvered underneath the cloud being seeded, and a polyethylene beaker was mounted on a platform tower (~2 m above the ground) and exposed for a 5–15 min period. Third, rainwater samples were collected at a number of fixed sites spread throughout the experimental area, utilizing the same type collector as for the mobile samples, except that these samples were exposed for a 24 h period. A log of each aircraft and surface sample collected during FACE 1973 and 1975 (time, location and volume) can be found in two reports by Wisniewski (1974, 1976b).

During the nonseeding summer of 1974, rainwater samples were collected (in the same manner described above) from a fixed number of surface sites located within the experimental area (Fig. 3). These samples were collected in cooperation with personnel of the South Florida Water Management District.

Due to the suspected contamination of a number of FACE 1973 aircraft rainwater samples (detailed in Section 3), a set procedure was developed and stringently adhered to in the collection and analysis of all samples collected during the 1974 and 1975 field programs. The first consideration was the collection material. It was found through extensive laboratory testing that both polyethylene and polypropylene materials presented no significant silver

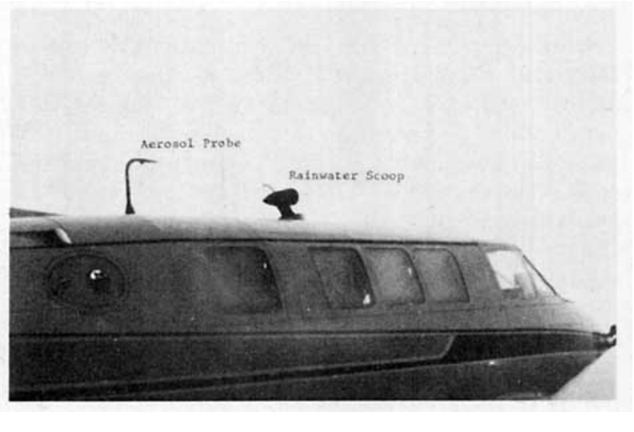

FIG. 2. Rainwater collection scoop on top of the aircraft fuselage. The intake for aerosols and ice nuclei was located directly behind the rainwater collection scoop.

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[1] Seeding action was either real or simulated, depending on randomized decision for clouds on that day.

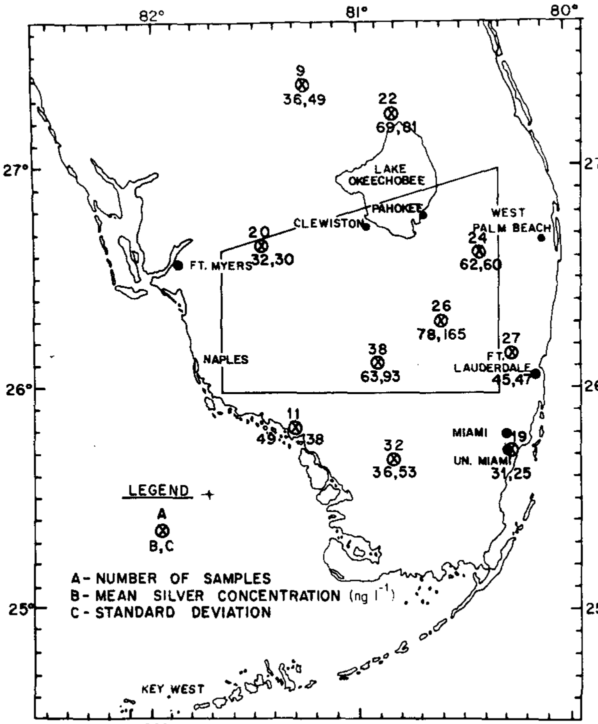

FIG. 3. Silver concentrations for fixed surface samples collected throughout south Florida during summer 1974, a year completely free of cloud seeding.

adsorption problems. For this reason, any material that had even a remote chance of contact with the rainwater sample was made of either polyethylene and/or polypropylene material. This included the aircraft rainwater scoop, the tubing running into the aircraft, all collection bottles and beakers and all storage vials. The aircraft rainwater samples were collected in bottles, while surface samples were collected in beakers. In both cases, a small portion of the sample was poured into a 5 ml vial and kept for silver analysis. All bottles, beakers and vials were washed with high purity nitric acid in the laboratory before being utilized in the field. The second consideration was the acidification of all samples on collection, as laboratory testing showed this to be an excellent method of keeping the silver in solution, this being a secondary precaution against adsorption. Therefore a 5 μl drop of high purity nitric acid was deposited into each 5 ml vial while in the laboratory before samples were actually collected. The third consideration in preventing the adsorption of silver was to freeze all samples immediately in dry ice after collection. In summation, if this three-pronged approach is utilized in collecting rainwater samples for silver analysis, the adsorption problems are almost nonexistent. This is a most important factor in dealing with the extremely low-level silver concentrations that results in this experiment.

The rainwater samples collected were all analyzed for silver concentrations using AA spectroscopy techniques. After being allowed to melt, the rainwater samples were analyzed[2] with a Perkin-Elmer 403 flameless atomic absorption spectrometer equipped with a HGA-2100 heated graphite atomizer and a deuterium arc background corrector. The instrumentation and analysis techniques have been fully detailed by Segar and Cantillo (1976). However, the procedure used for silver analysis is described here.

A sample of 100 μl volume was found to be optimum for silver analysis. Using a single-injection stopped-gas-flow analysis (Kahn and Slavin, 1971), and with careful optimization of instrument parameters, the detection limit varied from 5–20 ng ℓ^{-1} (80% confidence), depending on the condition of the instrument, the age of the atomized tube, etc. The deuterium arc background corrector was necessary to compensate for low-level absorption.

The analysis procedure was simple and straightforward. Samples were introduced through an irrigation port and were dried for 38 s at 160°C to evaporate the water. The remaining salts were charred to 560°C for 22 s to remove the volatile "background" salts. Atomization was accomplished via 15 s rapid heating to 2400°C. The cycle was completed by a cooling-down period. Water was allowed to flow around the tube, followed by flushing with argon gas. This cooling period took 15–20 s. The entire cycle duration of this analytical procedure was approximately 1.5 min and the results were plotted on an instantaneous peak readout. Each sample was analyzed either three times or until some consistency appeared in the results.

3. Results and discussion

A summary of the sample stratifications and analytical results are given in Tables 2–7. Each of the tables contains a description of the sample set of interest, the number of samples collected within each set, the mean silver concentration, the standard deviation, standard error of the mean, and the median, maximum and minimum silver concentrations within each set. All silver concentrations are expressed in ng ℓ^{-1}.

a. *FACE field program—1973*

Table 2 gives the results of all samples collected during FACE 1973. The table is subdivided into seeded versus nonseeded comparisons for three groupings: aircraft samples collected aboard the NRL S-2D and the NOAA DC-6, mobile samples collected at the

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[2] Rainwater analysis was carried out under contract with Professor Douglas Segar (FACE 1973 data) and Professor Carroll Smith (FACE 1975 and summer 1974 data) of the Rosenstiel School of Marine and Atmospheric Sciences, University of Miami.

TABLE 2. Silver concentrations (ng ℓ^{-1}) samples collected during FACE 1973.

Group	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
* Seeded aircraft	68	160	386	47	46	3000	14
* Nonseeded aircraft	55	518	1357	183	130	7200	BD**
Seeded mobile surface	24	43	65	13	36	320	BD
Nonseeded mobile surface	32	45	64	11	22	320	BD
Seeded fixed surface	10	130	30	94	13	970	BD
Nonseeded fixed surface	13	66	148	41	23	540	BD

* Data excluding one seed and three non-seed samples showing anomalously high concentrations of silver.
** BD signifies a level below detection limit of the atomic absorption spectiophotometer.

surface, and samples collected from fixed surface sites.

These results have previously been published (Wisniewski *et al.*, 1976) and will be discussed here only for the sake of completeness and to point out areas of discrepancies. In looking at Table 2, several points become immediately obvious:

1) The aircraft data, even excluding the four samples (three of which are no-seed) with anomalously high concentrations of silver, indicate mean silver concentrations of several hundred nanograms per liter. Surprisingly, the samples obtained on nonseeded days contained a factor of 3 more silver than those obtained on seeded days with the differences significant at the 99.5% confidence level using the Mann-Whitney-Wilcoxon nonparametric *U*-test.

2) The population of rainwater samples collected at the surface by the mobile van showed essentially no difference in silver concentration when stratified on the basis of seeded and nonseeded days, with the means in both instances approaching 50 ng ℓ^{-1}.

3) A relatively small number of fixed surface samples indicated a factor of 2 more silver in the mean on seeded days than on nonseeded days, the nonseeded values being slightly greater than 50 ng ℓ^{-1}. However, the differences are not statistically significant at the 95% confidence level.

Wisniewski *et al.* (1976) have discussed the possibility of contamination in preparation and analysis of the aircraft samples, while also pointing out the difficulties involved in vectoring the NOAA DC-6 aircraft into position underneath seeded clouds. These problems were magnified by the fact that in 1973 the NOAA DC-6 was not dedicated to rainwater collection, and an attempt was made to carry out several other programs simultaneously. In retrospect, we have become even more suspicious of the meaningfulness of the FACE 1973 aircraft rainwater data, particularly in view of the consistent results obtained during FACE 1975 discussed below.

b. Background—1974

Table 3 gives the results of all samples collected during the summer of 1974, a period in which no seeding activities were conducted. The samples were collected at ten fixed sites. The location, number of samples collected at each site, as well as the mean and standard deviation of the silver content are detailed in Fig. 3.

From Fig. 3, it can be seen that the means ranged from a low of 31 ng ℓ^{-1} on the southeast coast (near Miami) to a high of 78 ng ℓ^{-1} in the southeast corner of the FACE target area. Table 3 shows the overall mean for 228 samples was approximately 50 ng ℓ^{-1}. The maximum value of silver was 846 ng ℓ^{-1}, with only about 10% of the total number of samples containing silver in excess of 100 ng ℓ^{-1}. It would appear from the variations in the means that the highest silver concentrations occur within the interior agricultural portions of south Florida, with lower concentrations occurring on the coastal sites or within the Everglades National Park region (southernmost sites). It should be emphasized that the data collected from the fixed sites were obtained in beakers which were left exposed for 24 h periods and thus represent an integration of several individual showers which may have occurred from time to time during the day.

TABLE 3. Silver concentrations (ng ℓ^{-1}) for samples collected during the nonseeding summer of 1974 (16 June–30 September).

Group	Number of sampling days	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Fixed surface	69	228	51	80	5	25	846	BD

Table 4. Silver concentrations (ng ℓ^{-1}) for samples collected during FACE 1975 (23 June–18 July, aircraft; 28 June–19 August, mobile; 11 July–9 September, fixed).

Group	Number of sampling days	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Seeded aircraft	4	33	71	60	10	51	281	11
Nonseeded aircraft	18	265	48	43	3	39	296	BD
Seeded mobile surface	5	18	108	202	48	24	786	BD
Nonseeded mobile surface	11	27	47	59	11	18	264	BD
Seeded fixed surface	8	16	51	49	12	37	115	BD
Nonseeded fixed surface	42	85	50	47	5	32	169	BD

Dry deposition contamination at the fixed sites was also possible, although the rather low concentrations of silver indicate that, at least with respect to silver, this is not a major problem.

c. *FACE field program—1975*

Table 4 gives the results of all samples collected during FACE 1975. The table is subdivided into seeded versus nonseeded comparisons for three groupings: aircraft samples collected aboard the NCAR Queenair, mobile surface samples and fixed surface samples.

It can be seen that the silver concentrations in rainwater collected by the aircraft are consistent with those occurring in rainwater collected at the surface. The rather large difference in the number of nonseeded rainwater samples relative to seeded was due partly to the chance control-weighted randomization during the 3-week period of data collection and partly to the change in procedure (relative to FACE 1973), which allowed rainwater from nonseeded clouds to be collected on seeded days (prior to the beginning of the day's seeding activities). This additional flexibility was allowed because in FACE 1975 the NCAR aircraft was dedicated to the rainwater and aerosol collection program and thus could operate in a manner independent from the NOAA seeding project aircraft. It can be seen that the mean silver concentration of 71 ng ℓ^{-1} in rainwater collected by aircraft from the seeded clouds exceeded that of 48 ng ℓ^{-1} collected in nonseeded clouds. These differences are statistically significant at the 95% confidence level using a modified version of the standard t test called the Welch test, is one of a multitude of approximate solutions that can be used to test for equality of means when variances are unequal (Welch, 1938).

Results from the mobile surface rainwater collection program during FACE 1975 showed a mean silver concentration of 108 ng ℓ^{-1} reaching the ground from seeded clouds to be slightly more than twice that from nonseeded clouds (Table 4). These differences, however, were not statistically significant at the 95% confidence level.

The rainwater collected on a daily basis at the fixed surface sites failed to show any increase in silver due to the seeding. The mean concentration of silver obtained on both seeded and nonseeded days from the fixed surface sampling network was about 50 ng ℓ^{-1}, a value consistent with that obtained in the background year of 1974 and with the mobile surface nonseeded sample sets.

d. *Interyear consistency*

The sample set of nonseeded data obtained during the FACE 1975 field program presents a consistent picture when compared to the background data obtained during 1974. This is true regardless of the mode of sampling (aircraft, mobile surface or fixed surface). Rainwater silver concentrations obtained during the FACE 1973 field program are consistent with that from the two previous years for the surface data, but are sharply inconsistent for the aircraft data. The extraordinarily high values of silver found in the aircraft-collected rainfall on nonseeded days in FACE 1973 were not found in FACE 1975, a program which was carried out using dedicated resources with a much greater degree of care taken in the preparation, collection, handling and storage of the samples. Because of the very likely possibility of contamination in at least some of the 1973 aircraft samples, those data will not be discussed further.

The nonseeded surface data from the field seasons of 1973 and 1975, both from fixed and mobile sources, show a mean concentration of silver in rainwater in nonseeded samples of approximately 50 ng ℓ^{-1}, a value quite consistent with the control data set from 1974. The surface seeded sample set appears to reverse (relative to the nonseeded sample set) between FACE 1973 and FACE 1975 in that, in the former field season, no difference was detected in the mobile samples with the fixed samples showing an increase on seeded days, while in the latter field season no difference was evident from the fixed samples with the mobile samples showing a substantial increase from seeded cases. Because the rainwater samples at the fixed surface sites can collect several showers

TABLE 5. Silver concentrations (ng ℓ^{-1}) of FACE 1975 persistence versus background samples.

Group	Number of sampling days	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Persistence aircraft	3	90	46	50	5	33	296	BD
Background aircraft	15	175	50	40	3	39	247	BD
Persistence mobile surface	4	11	44	52	14	BD	213	BD
Background mobile surface	6	13	52	68	19	28	264	BD

during the course of a day, thus perhaps diluting a seeded sample with "clean" rain from nonseeded clouds, it is reasonable to expect that any effect of seeding on the silver concentration in rainwater would be more difficult to detect. In theory, with a well-placed mobile (van or aircraft) collector, higher silver rainwater concentrations should be detected since collection is being performed directly under treated cloud or convective systems. In this regard, the FACE 1975 data show an expected tendency, with the aircraft and mobile surface collected seeded samples containing more silver in the rainwater (up to a factor of 2 in the mean) than the nonseeded samples.

e. *The ecological impact*

In all three field seasons, the mean concentration of silver observed in rainwater from seeded clouds (or on seeded days) did not exceed 130 ng ℓ^{-1}, with a maximum concentration (ignoring the FACE 1973 aircraft data) of 970 ng ℓ^{-1} (see Tables 2–4). It can also be shown from these tables that the median silver concentrations from both the seed and no-seed samples, regardless of collection procedure, are generally in the 25–50 ng ℓ^{-1} range. While these concentrations are slightly lower then those found in other silver tracing studies (Table 1), overall agreement is reasonable.

These results are most encouraging from an ecological and/or environmental consideration since, regardless of how the samples are collected, the concentrations are well below the values that would cause concern to humans, animals, plants and lower forms of organisms (Klein, 1978) at the present time, under the now existing silver iodide output in south Florida.

f. *The persistence problem*

An important consideration is whether the silver iodide released may persist in the atmosphere to continue to act as seeding material at a later time. In an earlier FACE 1973 study (Wisniewski *et al.*, 1976), it was suggested that this may be a problem. Samples were collected at one fixed upwind site and stratified by seeding and nonseeding periods. Results showed silver concentrations to be higher during the seeding period than the nonseeding period, the differences being significant at the 99.5% confidence level.

During FACE 1975, the persistence problem was studied in more detail. Data in the "persistence" stratification were compiled by averaging the rainwater silver concentrations obtained on days following those which were massively seeded ($\geqslant$3 kg of AgI released). Table 5 compares the persistence versus background silver concentrations for the aircraft and mobile surface sample sets. Background samples are defined as those collected within the experimental area which were 2 days or more removed from a seeded day. In both cases, the mean concentrations are virtually identical, a result which strongly suggests that carry-over of the nucleating material from day to day does not appear to be a detectable source of contamination. This result is not surprising in view of the calculations discussed earlier, showing an "idealized" silver signature of the order 1000 ng ℓ^{-1} for seeded rainfall. Even without bulk transport processes being considered, it could be expected that plume dispersion alone will dilute the concentration of AgI particulate matter remaining in the atmosphere by several orders of magnitude over the course of a 24 h period. Such a reduction, of course, would bring the concentration of silver in rainwater back down to background levels which, from the data collected during 1975, appears to have been the case.

g. *Air mass trajectory analyses*

Wisniewski *et al.* (1976) suggested that higher rainwater silver concentrations may occur naturally in south Florida when air masses pass through polluted or industrial continental areas and eventually rainout these contaminants in the south Florida area. The conclusions were reached by stratifying analyses of synoptic maps on dates on which rainwater silver concentrations were collected. In light of these implications from the FACE 1973 data, it was decided to take more detailed look at this problem during FACE 1975.

Two different air mass trajectory analyses were performed for FACE 1975 experimental days on which the NCAR aircraft operated. The first set of analyses utilized the average wind block-layer trajectory model described by Heffter and Taylor (1975). These trajectory computations used winds at any desired layer above averaged terrain. The wind input was obtained from observed winds at reporting pibal and radiosonde

TABLE 6. Silver concentrations (mg ℓ^{-1}) of nonseeded aircraft samples for the lower level average wind and isentropic trajectories.

Group	Number of sampling days	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
			Average wind trajectories					
Continental influenced	2	22	68	36	8	65	179	14
Maritime	11	195	48	42	3	37	256	BD
			Isentropic trajectories					
Continental influenced	2	22	68	36	8	65	179	14
Maritime	6	108	46	48	5	36	256	BD

stations and from analyzed wind fields at selected grid points. This technique may be somewhat limited by its failure to incorporate vertical motions and vertical shears into the analysis.

The second set of analyses utilized an isentropic trajectory scheme as described by Danielsen (1961, 1974) and further developed by Deaven (1977, personal communication). These computations employed a vertical coordinate based on those of potential temperature (θ). Since the air ascends and descends through the pressure surfaces, the computed trajectories can, on occasion, differ substantially from those computed by the method of average wind analysis. This technique has the limitation of failing to incorporate diabatic temperature changes. Extreme surface heating, therefore, can sometimes result in the low-level trajectories intersecting the surface abruptly within the first day of transport which, of course, eliminates that case from further analysis.

The rainwater silver concentrations from all nonseeded days were stratified using both trajectory method. The stratification was accomplished by classifying data as either maritime or continental influenced, depending on both the origin of the air mass and the path it followed during its 3-day trek which terminated in south Florida. In a previous paper, Wisniewski (1976a) stratified the FACE 1975 data using a less stringent approach; however, after careful consideration it was felt that the maritime versus continental classification scheme was much more meaningful.

The rainwater silver concentrations were also stratified on the basis of trajectory level. The lower level average wind trajectory used observed or computed winds in a layer which extended from the surface to 1500 m, while the isentropic trajectory traced air motion along the 300 K isentrope, which usually fell within the surface to 1500 m layer. The middle level average wind trajectory used winds within the layer from 1500–5700 m, while the isentropic trajectory traced the air motions at both the 310 and 320 K isentropes, both of which fell within the 1500–5700 m layer.

A summary of the rainwater silver concentrations as stratified utilizing the two trajectory methods is presented in Tables 6 and 7. The total number of samples is different between the average wind and isentropic analyses and are less than the total on all nonseeded dates because, in both cases, days with a "stagnating" trajectory in the Florida area were excluded from analysis and, in the isentropic case, some trajectories were lost early due to surface heating. The data are stratified by whether the air mass was either 1) continental influenced, i.e., the air mass followed a continental path along some portion of its trek into south Florida; 2) maritime, i.e., the air mass originated in a maritime area and flowed only over water into south Florida. An example of the scheme used to stratify the trajectories is illustrated in Figs. 4 and 5. Fig. 4 corresponds to the lower and middle-level trajectories, respectively, from 5 July 1975, while Fig. 5 corresponds to those from 16 July 1975. Fig. 4

TABLE 7. Silver concentrations (ng ℓ^{-1}) of nonseeded aircraft samples for the middle-level average wind and isentropic trajectories.

Group	Number of sampling days	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
			Average wind trajectories					
Continental influenced	4	44	62	43	6	48	204	11
Maritime	9	173	47	41	3	35	256	BD
			Isentropic trajectories					
Continental influenced	3	39	62	42	7	52	204	11
Maritime	7	129	49	45	4	37	256	BD

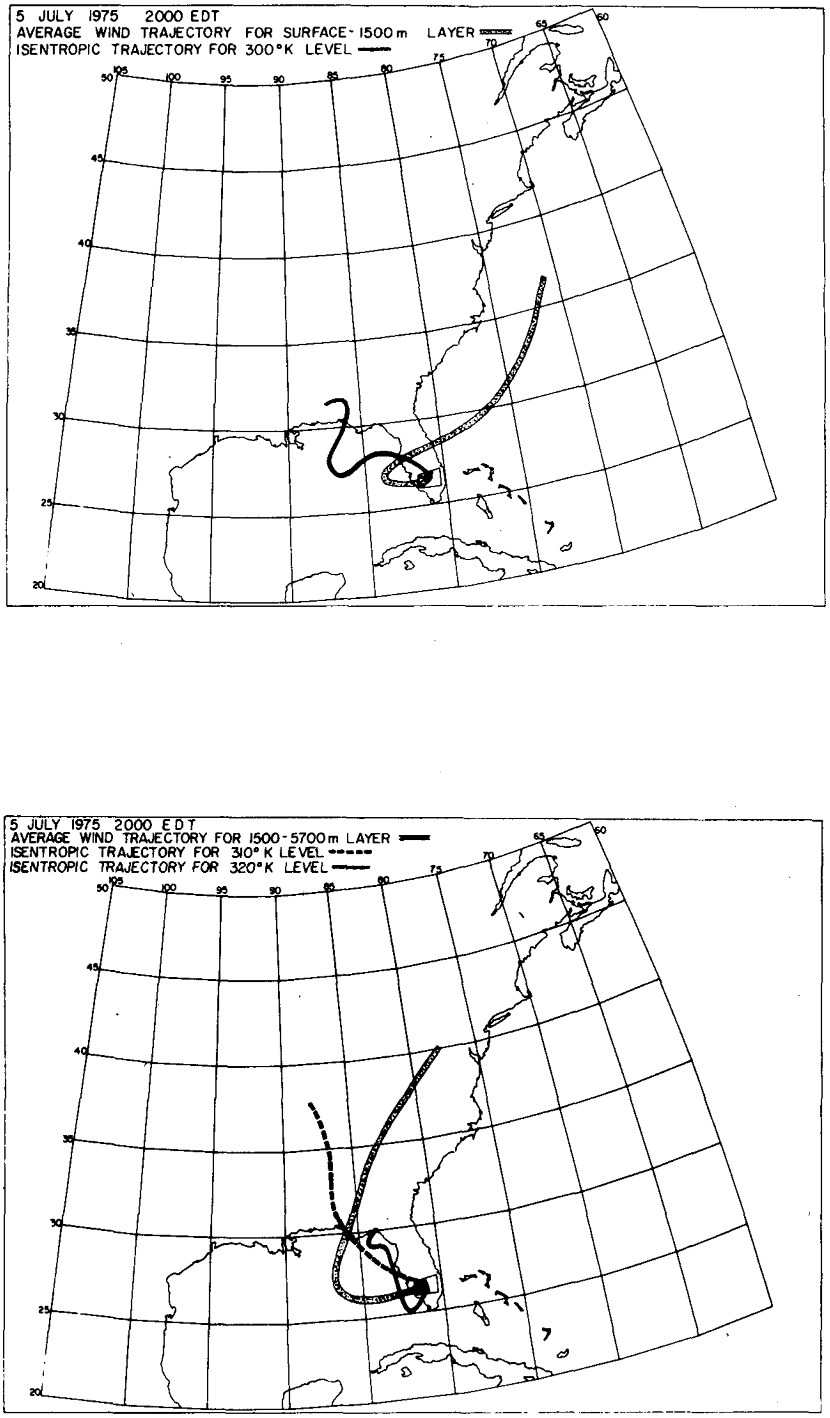

FIG. 4. Lower and middle level trajectories utilizing both the average wind and isentropic methods of analysis for 5 July 1975, a date in which all trajectories can be classified as continental influenced.

shows the average wind and isentropic trajectories to be different; however, each is classified as being a continental influenced trajectory. Fig. 5 depicts maritime trajectories.

In all four cases shown in Tables 6 and 7, the mean and median rainwater silver concentrations are substantially greater when the air mass has a continental trajectory, the differences being significant at the 95%

confidence level. These differences may be due to various industrial activities pumping silver particulate matter into the atmosphere which are then transported over long distances as suggested by Ranticelli and Perkins (1970).

The most crucial result that appears in the trajectory stratifications is that the rainwater silver concentrations which occur on nonseeded days during continental trajectories are nearly identical to those which occur during seeding activities. This is im-

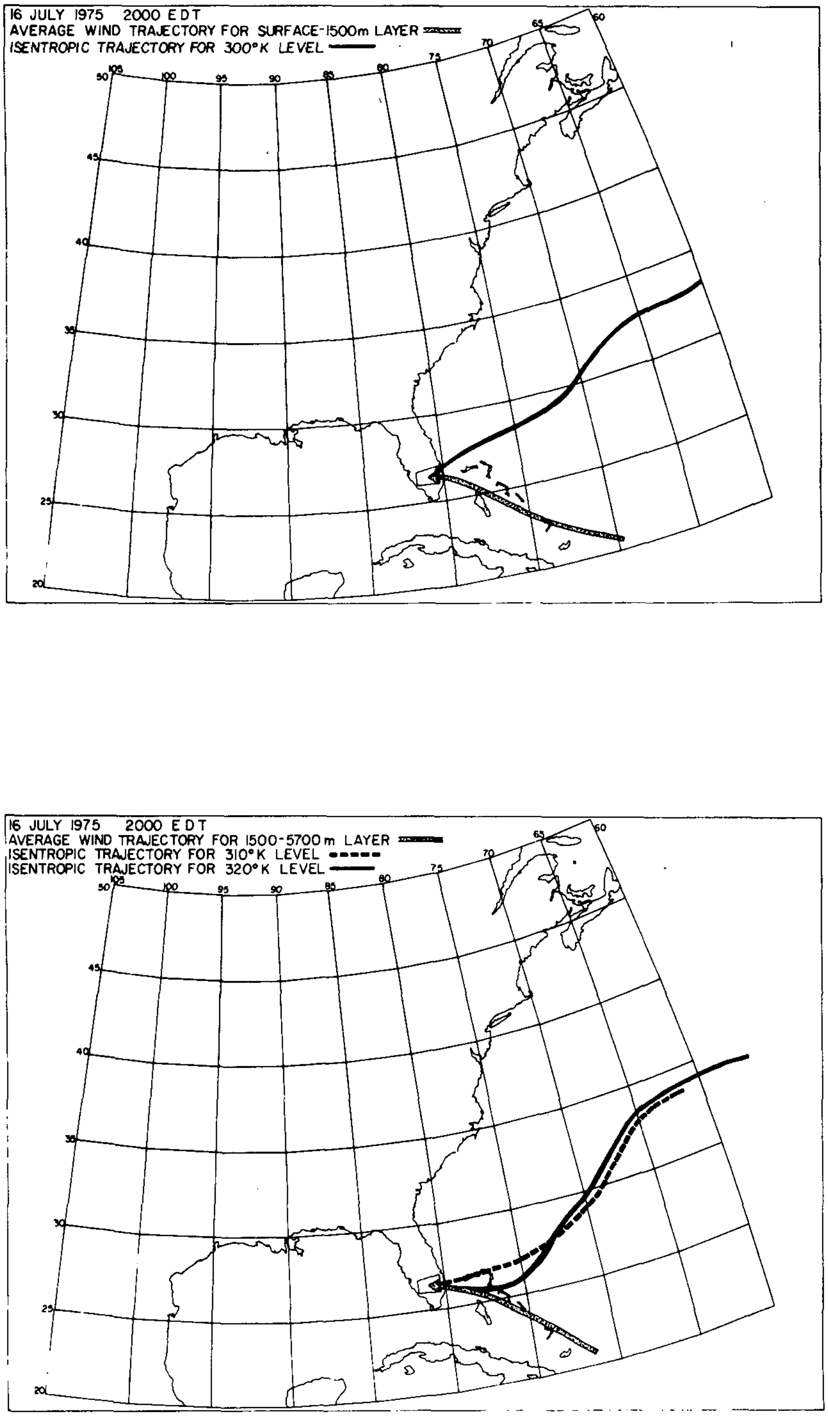

FIG. 5. As in Fig. 4 except for 16 July 1975, a date in which all trajectories can be classified as maritime.

portant since, if the technique of measuring rainwater silver concentrations is being used to infer the presence of the silver iodide seeding agent, erroneous conclusions may result unless the origin of the air mass in which the rainfall is sampled is taken into account. For the 1975 rainfall data, all seeded days had maritime air mass trajectories. The magnitude of seeding, therefore, appeared to be no more than to bring the silver levels up to those experienced on nonseeded days with continental air mass trajectories.

4. Summary and conclusions

Rainfall data obtained during portions of three consecutive summers (1973–75) in south Florida have shown that the mean concentrations of silver resulting from the FACE cloud seeding activities are not elevated by more than about a factor of 2 above background levels and remain well below the tolerance limits that would cause ecological and/or environmental concern. Excluding the aircraft-collected data from 1973 which are suspected of having contaminated samples, the maximum concentration of silver observed on any seeded day was 970 ng ℓ^{-1}. The maximum concentration of silver observed on any nonseeded day was found to be 846 ng ℓ^{-1}, which occurred in 1974, a year in which there was no seeding program.

The persistence of silver in the atmosphere from seeding activities from one day to the next is not a detectable feature in south Florida. A careful analysis of the 1975 samples showed silver concentrations to be almost identical between background and persistence data sets.

It was shown that days with air masses of continental origin (at low and mid-tropospheric levels) rainout a higher concentration of silver than those with air masses of maritime origin. The concentration of silver in "continental rain" on nonseeded days approaches that of silver in "maritime rain" on seeded days. The effect of seeding, therefore, does not appear to produce changes in rainwater silver concentrations which are outside the spectrum of natural variability.

It can be concluded from this series of measurement studies that the dynamic seeding technique as used in FACE (which involves very heavy dosages of AgI ($\sim$1 kg per cloud)) does increase the mean silver concentration in rainwater on a localized shower basis by about a factor of 2, but this is within the natural variability brought about by differences in trajectories of the air mass in which the shower is occurring. The measured factor of 2 increase falls far short of a calculated (using admittedly very simplified assumptions) seeding signature of several orders of magnitude above background. This may well be due to substantial dilution of the seeded water with "clean" water prior to collection of the rain, spatial and temporal sampling problems in collecting water originating from the main seeded portion of cloud, transport of some seeding material upward and outward to new portions of cloud making less particles per unit volume available for collection, and/or gross errors in the amount of material theoretically available for collection due to burn peculiarities of the flare, causing slag products which might not be sampled during particle size distribution analyses in the wind tunnel.

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REFERENCES

Bollay, E., 1965: Neutron activation analysis of silver content in precipitation samples. Final Rep. Contract 14-06-D-5573 (submitted to the Bureau of Reclamation).

Danielsen, E. F., 1961: Trajectories: Isobaric, isentropic and actual. *J. Meteor.*, **18**, 479–486.

——, 1974: Review of trajectory methods. *Advances in Geophysics*, Vol. 18B, Academic Press, 73–94.

Gatz, D. F., 1975: Background silver concentrations in Illinois precipitation and river water. *J. Appl. Meteor.*, **14**, 217–221.

Heffter, J. L., and D. Taylor, 1975: A regional-continental scale transport, diffusion and deposition model. NOAA Tech. Memo. ERL-ARL 50, 28 pp.

Kahn, H. L., and S. Slavin, 1971: Static method of analysis with graphite furnace (HGA-70). *Atom. Absorp. Newslett.*, **10**, 125–126.

Klein, D. A., 1978: *The Environmental Impacts of Artificial Ice Nucleating Agents.* Dowden, Hutchinson and Rose, Stroudsburg, Pa., 257 pp.

Ostlund, H. G., and R. D. Stearns, 1970: Florida Cumulus Experiment 1970—silver content of precipitation. Final Report, Naval Research Laboratory, 9 pp.

Parungo, F. P., and C. E. Robertson, 1969: Silver analysis of seeded snow by atomic absorption spectrophotometry. *J. Appl. Meteor.*, **8**, 315–321.

Rancitelli, L. A., and R. W. Perkins, 1970: Trace elements of concentrations in the troposphere and lower stratosphere. *Geophys. Res.*, **75**, 3055–3064.

Sax, R. I., D. M. Garvey, F. P. Parungo and T. W. Slusher, 1977: Characteristics of the AgI nucleant used in NOAA's

Florida Area Cumulus Experiment. *Preprints Sixth Conf. Planned and Inadvertent Weather Modification*, Champaign-Urbana, Amer. Meteor. Soc., 198–201.

Schaefer, U. J., and J. J. Fuquay, 1965: The detection of silver iodide in snow by neutron activation analysis. *J. Rech. Atmos.*, **11**, 49–52.

Segar, D. A., and A. Y. Cantillo, 1976: Direct determination of trace metals in seawater by flameless atom absorption spectrometry. *Anal. Methods Oceanogr.*, **147**, 56–81.

Summers, P. W., 1972: The silver fallout patterns in precipitation from seeded convective storms. *Preprints Third Conf. Weather Modification*, Rapid City, Amer. Meteor. Soc., 279–286.

Warburton, J. A., 1963: The detection of silver in rainwater from cloud seeding experiments in Australia. *J. Appl. Meteor.*, **2**, 569–573.

——, 1965: The detection of silver in rainwater: Further developments of technique. *J. Appl. Meteor.*, **4**, 564–568.

——, 1973: The distribution of silver in precipitation from two seeded Alberta hailstorms. *J. Appl. Meteor.*, **12**, 677–682.

——, and C. T. Maher, 1965: The detection of silver in rainwater: Analysis of precipitation collected from cloud-seeding experiments. *J. Appl. Meteor.*, **4**, 560–564.

——, and L. G. Young, 1968a: Evidence of persistence on cross contamination in cloud seeding experiments. Reprint Ser. No. 54, Desert Research Institute, University of Nevada, 14 pp.

——, and ——, 1968b: Neutron and activation procedures for silver iodide in seeded lake effect storms. *J. Rech. Atmos.*, **6**, 679–692.

Welch, B. L., 1938: The significance of the difference between two means when the population variances are unequal. *Biometrika*, **29**, 350–359.

Wisniewski, J., 1974a: Tracing of silver iodide released during the 1973 Florida Area Cumulus Experiment (FACE). M.S. thesis, Dept. of Atmospheric Sciences, University of Miami, Coral Gables, 83 pp.

——, 1974b: Tracing of silver iodide and other chemical constituents during the 1973 Florida Area Cumulus Experiment (FACE). NOAA Tech. Memo. ERL WMPO-12, 141–186.

——, 1976a: Variability of rainwater silver concentrations in south Florida. *Preprints Int. Conf. Weather Modification*, Boulder, WMO, 563–568.

——, 1976b: Tracing of silver, ice nuclei and other chemical constituents. NOAA Tech. Memo. ERL WMPO-28, 149–180.

——, 1977: Silver concentrations in rainwater from seeded and nonseeded south Florida cumuli. Ph.D. dissertation, University of Virginia, 98 pp.

——, 1979: A review of silver tracing programs. *Preprints Seventh Conf. Inadvertent and Planned Weather Modification*, Banff, Alberta, Amer. Meteor. Soc.

——, W. R. Cotton and R. I. Sax, 1974: Tracing silver iodide in the south Florida area. *Preprints Fourth Conf. Weather Modification*, Ft. Lauderdale, Amer. Meteor. Soc.

——, —— and ——, 1976: Silver content of precipitation from seeded and nonseeded Florida cumuli. *J. Appl. Meteor.*, **15**, 1004–1001.

Woodley, W. L., and R. I. Sax, 1976: The Florida Area Cumulus Experiment: Rationale, design, procedures, results, and future course. NOAA Tech. Rep. ERL 354-WMPO 6, 204 pp.

Zacharuk, E. V., and G. G. Goyer, 1975: Silver iodide disposition in 1974 samples, current research on Alberta hailstorms. Prog. Rep. 1974–1975, Atmos. Sci. Div., Alberta Research Council, Edmonton, Alberta, 70–94.

——, and ——, 1976: Silver iodide disposition in 1975 storms, current research on hailstorms and their modification. Rep. 76-1, Atmos. Sci. Div., Alberta Research Council, Edmonton, Alberta, 103–119.

——, and R. Erb, 1977: Silver deposition. Rep. 77-3, Atmos. Sci. Div., Alberta Research Council, Edmonton, Alberta, 63–72.