The Disposition of Silver Released from Soviet Oblako Rockets in Precipitation during the Hail Suppression Experiment Grossversuch IV. Part I: Measurements of Background and a Preliminary Seeding Test

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ABSTRACT

In association with Grossversuch IV, a program designed to test the Soviet hail suppression method by seeding clouds with AgI from Oblako rockets, a complementary program was conducted by l'Observatoire du Puy-de-Dôme and the Desert Research Institute to study the diffusion of the seeding material (AgI) in the clouds, based on the analysis of silver in precipitation. This program covered the summers of 1977 and 1978, and this paper describes the results of measurements of natural background silver concentrations in unseeded precipitation. It also describes a new automatic precipitation collector, five of which were first tested in the field in 1977. A more extensive network of 15 collectors was deployed during two months of the 1978 summer.

Based on the analysis of 118 unseeded precipitation samples collected in 1977, the natural background concentration of silver was estimated as 0.9×10^{-11} g mL^{-1} ($\sigma = 0.6 \times 10^{-11}$ g mL^{-1}). Although the standard deviations overlap, the 1978 season results appear to indicate a lower background of 0.5×10^{-11} g mL^{-1} ($\sigma = 0.3 \times 10^{-11}$ g mL^{-1}), based on the analysis of 414 rain samples. The average value for the two seasons was 0.6×10^{-11} g mL^{-1} with a standard deviation of 0.5×10^{-11} g mL^{-1}. These background concentrations were found to be independent of both the length of sampling period and the precipitation intensity, averaged over the sampling periods of the collectors.

The background is sufficiently low to permit the detection of the presence of silver iodide emitted from the Soviet rockets in the precipitation. The preliminary resuls from one case study are presented to support this conclusion.

1. Introduction

Grossversuch IV is a randomized experiment designed to test the Soviet OBLAKO rocket technique for seeding hail-forming convective clouds. These rockets are designed to deliver the silver iodide particulates with some degree of precision to specific locations in the clouds.

The plan for the five-year experiment, 1977 being the first year, is to evaluate the effects of seeding by comparing the kinetic energies of the hail for seeded and non-seeded days (Federer *et al.*, 1978), using statistical methods. A set of criteria have been established by Federer *et al.* which are identical to those used by the Soviets. These criteria involve radar reflectivity, echo geometry and temperatures of the cloud systems.

A complementary program of study of the disposition of the silver iodide throughout the precipitating system, based on measurements of the silver content of the precipitation falling from the seeded clouds, was conducted in 1977 and 1978 by the Observatoire du Puy-de-Dôme, and the Desert Research Institute.

The spatial-temporal variations of the silver content of precipitation have been investigated in a number of earlier studies (Lacaux, 1972; Warburton, 1973), and interpreted as a signature of the interaction of the AgI particles with the cloud particulates. The analysis of the precipitation for the presence or otherwise of the seeding material is to determine, as far as feasible by such methods, what happens to the seeding agent and so form some estimate of the seeding effectiveness. In particular, the measurements made by Linkletter and Warburton (1977) in the U.S. National Hail Research Experiment (NHRE) demonstrated that the convective cloud cells are not uniformly affected by extensive aircraft seeding, and that the silver iodide particles follow somewhat priveleged trajectories in the clouds. Those studies were made under conditions where the NHRE program had also established seeding criteria related to radar reflectivity and storm motion. In the

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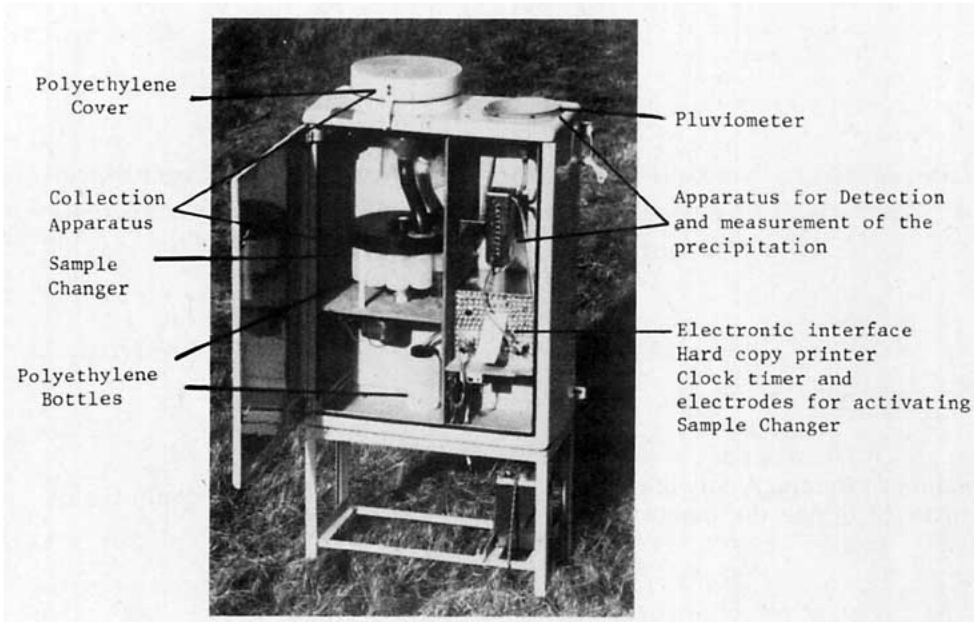

FIG. 1a. Precipitation sampler used in Grossversuch IV.

case of Grossversuch IV, the rocket seeding method may tend to simplify the study of such AgI dispersion and its involvement in the precipitation processes. In effect each rocket releases a well-estimated number of particles along a simple predetermined trajectory during a time which is short compared with the lifetime of the cloud. This was not the case for the aircraft-type seeding used in NHRE, in which the clouds were probably treated in some complex manner almost throughout their lifetimes.

In order to carry out such investigations it is important to study the natural background of silver in

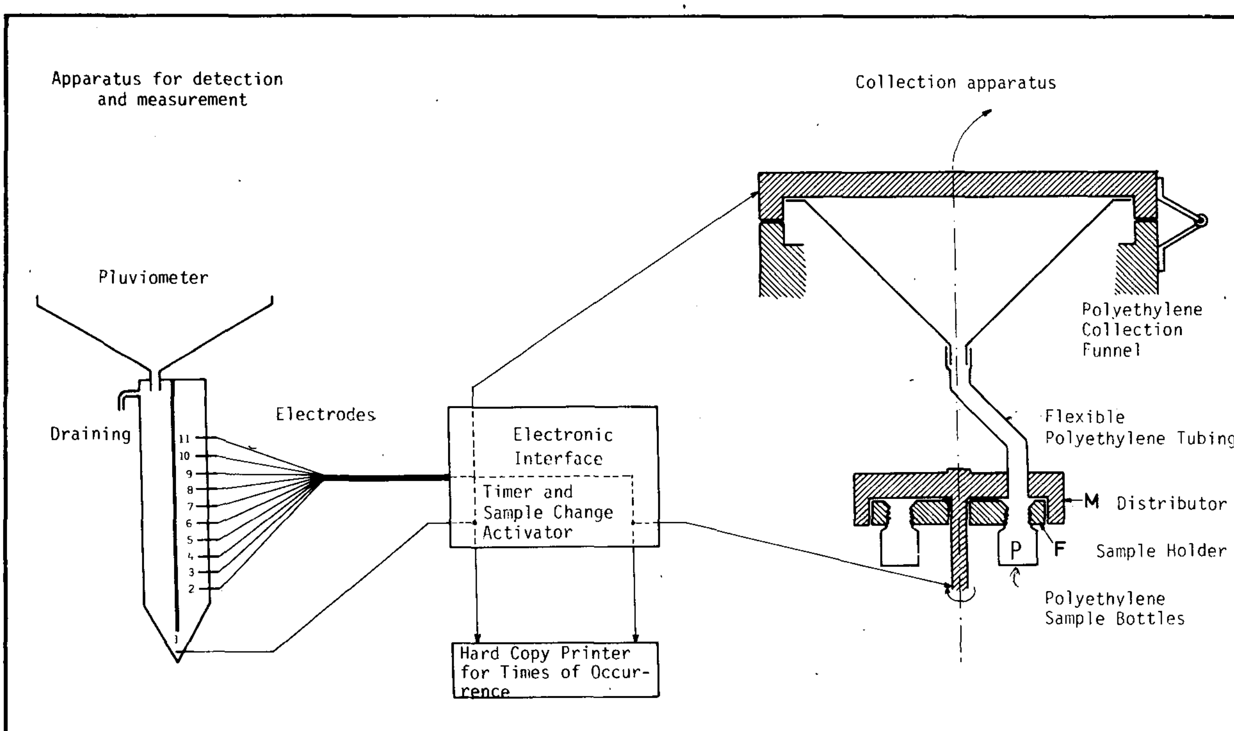

FIG. 1b. Block schematic of precipitation collection and measurement apparatus.

TABLE 1. Technical characteristics of automatic collector.

Surface area of collector	490 cm^2
Volume of each sample	49–196 cm^3 (adjustable)
Corresponding depth of rainfall	1 to 4 mm
Number of samples	10 (+1)
Total depth of rainfall sampled	10 to 40 mm (adjustable)
Register of time of opening of sampler	to nearest second
Collection of hailstones up to 30 mm diameter	
Battery operated for periods of more than one month	
Dimensions	70 × 40 × 120 cm
Weight	40 kg

precipitation in the project area and to test new automatic precipitation collectors. A network of five collectors was installed during the month of August 1977 in the target area of Grossversuch IV, and a more extensive network of 15 collectors was deployed for the 1978 season. The purpose of this note is to describe the precipitation collectors, and the measurements of background silver in precipitation on non-seeded days. Measurements of the silver content of samples collected at one site for one seeded day during the observational period are also presented. The results of all seeded day samples collected during the 1978 season will be described in Part II of this series.

2. The automatic precipitation collector

Because of the suddenness and the uncertain localized character of the precipitation produced by convective clouds, it is difficult, in the field, to collect a set of precipitation samples which is representative of the lifetime of the cloud and of its movement, even when mobile units guided by radar are used for the sample collection. It seemed preferable to us to use entirely automatic collectors (Fig. 1a), set out in a network at fixed positions. The collectors used were conceived and constructed for the 1977 Swiss program (Lacaux, 1979). The collector operates on the following principles:

1) Collects precipitation with a high degree of cleanliness, avoiding, in particular, aerosol deposition before the onset of the rainfall.

2) Functions automatically as a precipitation detection system and measures the precipitation intensity which controls the collection system.

3) Collects samples sequentially during the precipitation period, as a function of the precipitation intensity. The samples have constant volumes col-

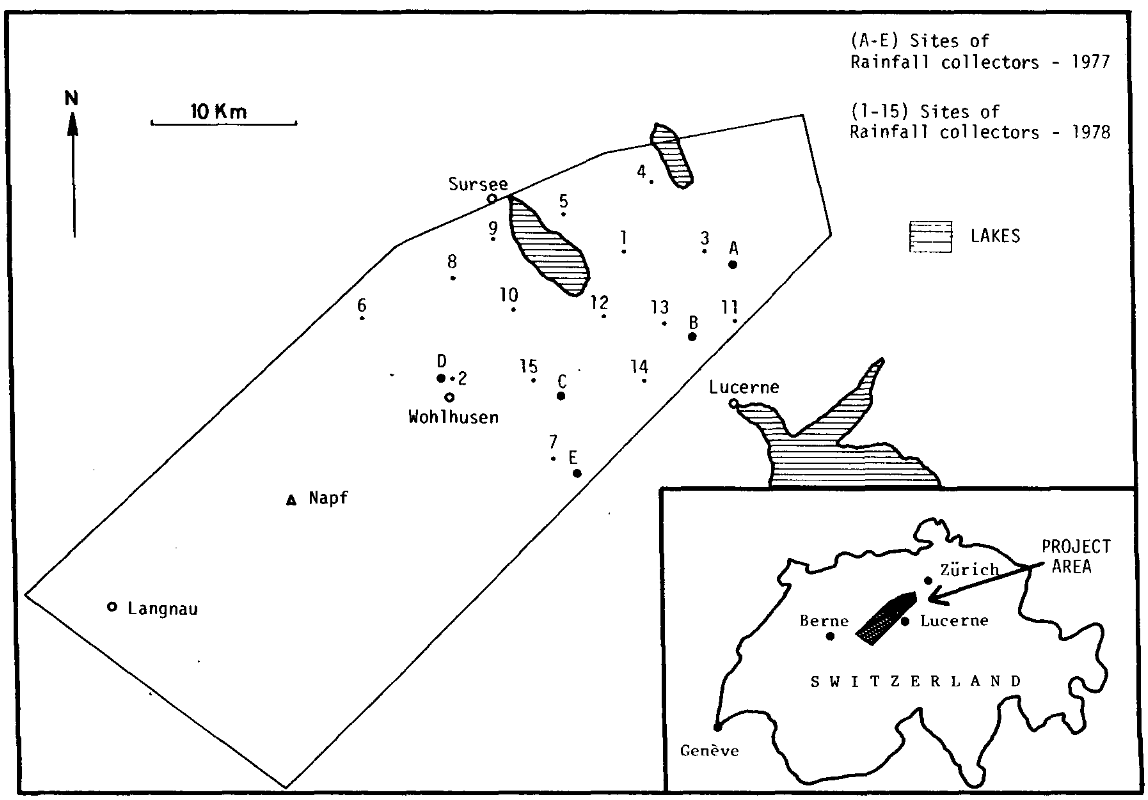

FIG. 2. Grossversuch IV project area—Switzerland—showing locations of sampling sites.

TABLE 2. Ranges of silver concentration and precipitation intensities measured.

Date 1977	Number of collectors used	Range of [Ag] (10^{-11} g mL^{-1})	Mean [Ag] ± σ (10^{-11} g mL^{-1})	Range of precipitation intensity (mm h^{-1})
9 August	5	<0.3 to 2.8	0.8(0.7)	0.8 to 15.8
12 August	2	0.7 to 3.2	1.3(0.7)	3.4 to 81.3
13 August	4	<0.3 to 1.9	0.8(0.5)	0.5 to 36.4
16 August	2	<0.3 to 3.3	1.3(0.9)	1.3 to 85.4
18 August	4	<0.3 to 1.1	0.6(0.3)	0.4 to 43.9
21 August	4	<0.3 to 0.8	0.5(0.3)	0.1 to 0.8
22 August	3	0.4 to 1.6	1.0(0.4)	1.2 to 8.6
23 August	4	<0.3 to 2.1	0.9(0.6)	0.3 to 8.6

Date 1978	Number of collectors used	Range of [Ag] (10^{-11} g mL^{-1})	Mean [Ag] ± σ (10^{-11} g mL^{-1})	Range of precipitation intensity (mm h^{-1})
6 June	15	0.2 to 1.0	0.4(0.2)	0.3 to 59.4
7–8 June	6	0.1 to 1.5	0.4(0.2)	0.2 to 72.8
9–10 June	14	0.2 to 1.4	0.5(0.3)	0.1 to 88.7
15 June	15	0.2 to 1.3	0.5(0.4)	0.3 to 4.7
19 June	15	0.2 to 1.4	0.5(0.3)	0.1 to 94.2
21 June	1	0.4 to 0.5	0.5(0.1)	0.3
18–19 July	2	0.2 to 1.6	0.6(0.4)	0.1 to 41.1
26–27 July	15	0.1 to 1.2	0.4(0.2)	0.2 to 35.4

lected over varying time intervals which are recorded.

4) Collects hailstones up to 30 mm in diameter, but does not separate them from the rainwater.

a. The apparatus for detection and measurement

This system controls the opening of the cover which is over the collection funnel. Opening occurs at the onset of the precipitation (after the first 0.1 mm has fallen). The system also measures the depth of precipitation, and in so doing allows the control of the distribution of the precipitation in the collection apparatus. The apparatus for detection and measurement, shown in Fig. 1b, is composed of a pluviometer of 290 cm^2 reception surface area which has an opening to allow the water to flow from the bottom. When this water reaches electrode 1, an electrical impulse is transmitted to the electronic interface causing the opening of the cover of the collection funnel and recording the time of occurrence with an accuracy of 1 s. At a later time, when the water reaches the next electrode, the automatic sample changer is activated and the time of occurrence is recorded.

b. The collection apparatus

This system (Fig. 1b) consists of a polyethylene funnel of 490 cm^2 surface area, and a distributor (M) which partitions the precipitation into 11 polyethylene bottles (P). When not in use the funnel is covered by the pivoted nylon lid. The precipitation flows from the funnel along a bent polyethylene tube which is kept very short to minimize contamination of one sample by the preceding one. This tube is fixed to a plate M which moves in rotation from one sample bottle to the next. These 250 mL polyethylene bottles are housed in the fixed sample holder (F), and are located at the exits of each of the first 10 orifices of the plate. The eleventh position is occupied by a 1 L bottle which receives the final portion of the precipitation.

The principal technical characteristics of the collector are summarized in Table 1.

This apparatus was built at a cost of approximately $2000 per unit. It is not commercially available.

3. Collection procedures—1977 and 1978

During the 1977 and 1978 summer field seasons, automatic collectors were installed in the target area of the Grossversuch IV experiment (Fig. 2). Mobile trucks were used to pick up the collected samples, to immediately freeze these samples with dry ice (CO_2) and to prepare the sampler for a new set of collections by rinsing the collectors with deionized distilled water. About 2 h were required to carry out these operations for each group of 5 collectors following the end of the precipitation period.

4. Silver content of precipitation

The technique used for the silver analysis by the Center of Atmospheric Research in Lannemezan was flameless atomic absorption. The minimum detectable concentration depends on the volume of the treated dose of water; with 1000 μL the detectable concentration is 0.3×10^{-12} g mL^{-1} using a Perkin Elmer 305 coupled with an HGA 70 graphite furnace. For reducing the time of analysis, we introduce only 250 μL which corresponds to a minimum detectable concentration of nearly 10^{-12} g mL^{-1}. The analytical conditions are summarized below:

- The hollow cathode lamp, operated under 15 mA current, emits the silver wavelength 328.1 μm.
- The opening of the slit for the beam is 1 mm.
- The volume of the treated dose is 5 × 50 μL which are delivered to the furnace with an Eppendorf micropipette with interchangeable tips.
- For each aliquot (50 μL), the sample is dried for 1 min at a temperature of 230°C, followed by 1 min at 490°C. At the end of the drying cycle of the fifth aliquot, atomization is produced in a period of 5 s at 2600°C.
- The flux of argon gas is stopped in the system 2 s before atomization.
- A deuterium lamp is used to compensate for non-specific absorption.

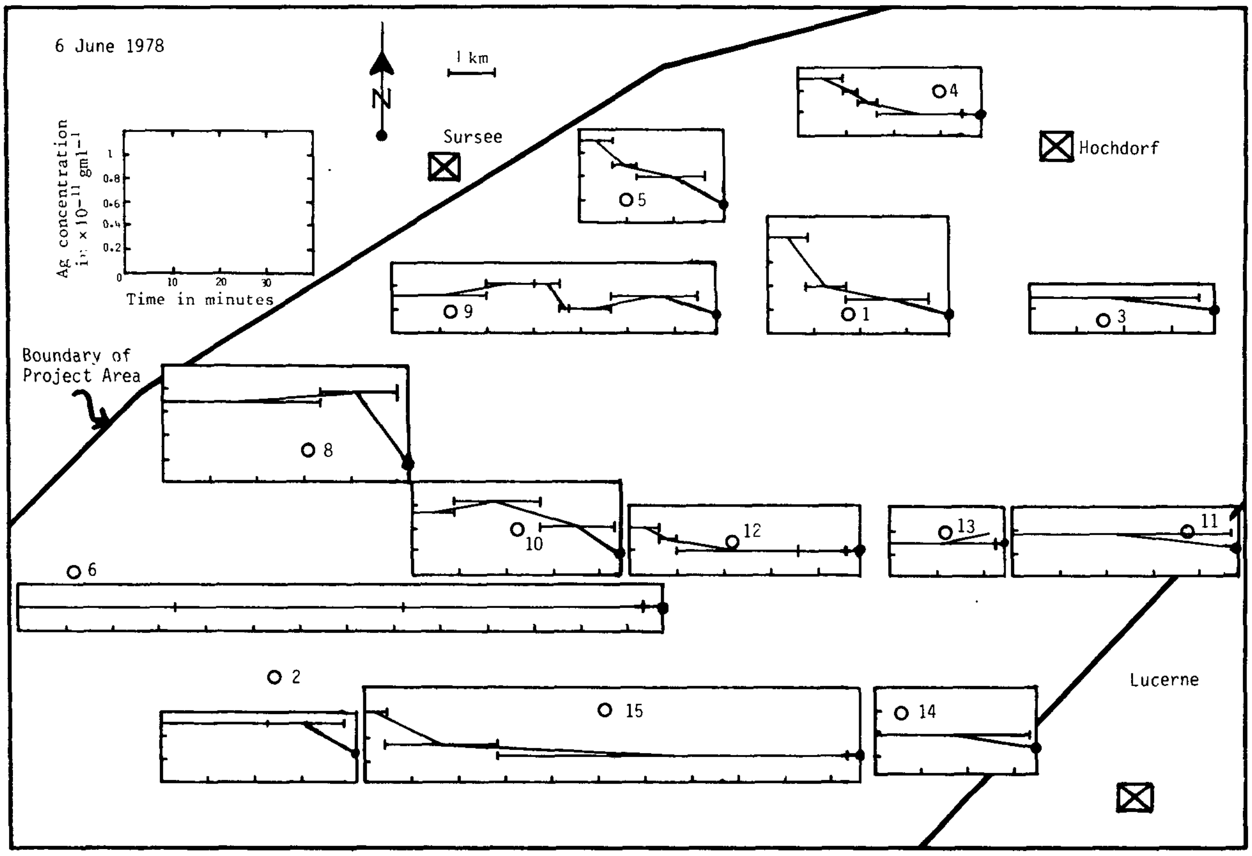

FIG. 3. Concentrations of silver in sampled precipitation as function of sampling time at all sites.

A large number of samples collected on non-seeded days in the summer of 1978 were shipped frozen from France to the United States for analysis in the class 100 cleanroom laboratories of DRI in Reno, Nevada. The flameless atomic absorption technique used was similar to that employed by the Center of Atmospheric Research in Lannemezan. The minimum detectable concentration for silver is 0.3 $\times 10^{-12}$ g mL^{-1} using a Varian Tektron Spectrophotometer, Model AA5 in conjunction with an Instrumentation Laboratory Flameless Atomizer, Model 455. This atomizer uses 50 μL capacity carbon microboats. For the background determinations, each microboat was loaded 20 times, representing 1 mL of the sample. Five boats were loaded in this manner for each sample analyzed. The standard silver concentrations used for the determination of the standard calibration line, both prior to and immediately after the unknown sample analyses, used silver nitrate of concentrations 16, 32, 64 and 128 $\times 10^{-12}$ g mL^{-1}. Each standard used 50 μL for each microboat and five analyses were performed for each standard concentration. The rain samples being analyzed were first melted from the ice phase to the liquid into a cleaned polyvial. Analyses of blanks were performed at regular intervals throughout the analyses. Such blanks, covering the status of the micropipettes, the exchangeable tips, the deionized water and the polyvials, always showed less than the minimum detectable amount of silver. All analyses were performed by atomizing the samples in an argon gas atmosphere.

Details of the sample collections and of the silver analyses for the non-seeded days in 1977 and 1978 are given in Table 2.

5. Discussion

The analysis of the samples collected sequentially during the precipitation periods, showed that, in a general way, the natural background of silver decreased regularly throughout the precipitation and that the first few samples had concentrations higher than the ones which followed. For example, the silver concentrations in relation to time of day after precipitation commencement, have been plotted in Fig. 3 for 6–7 June 1978, where the unseeded convective clouds moved from East to West between

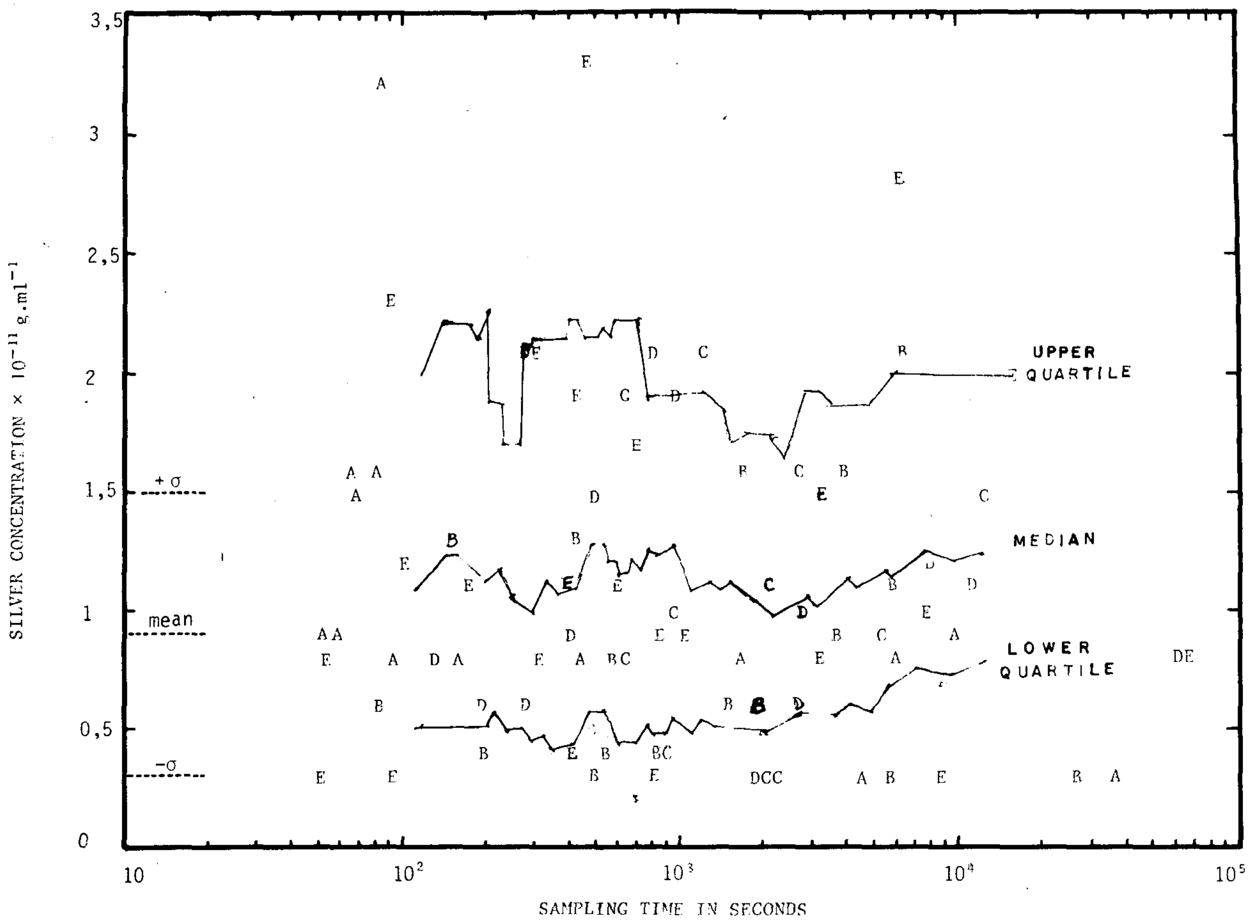

FIG. 4. Silver concentration as function of sampling time duration (1977 season), showing medians, upper and lower quantities.

2230 and 0100 h (local time). Except for collector 13, the silver background decreased, regularly for collectors 1-2-3-4-5-11-12-14-15 and with some fluctuations for collectors 8-9-10, down to 0.2×10^{-11} g mL^{-1} for the last sample collected at each site, as shown by the heavy black dots. The time periods over which these last samples were collected at each site are not known because the samples do not record the end of the precipitation period but only the time of commencement and the times of changes of sample bottle. The measurements of silver concentration presented in Fig. 3 have standard deviations of 0.5 $\times 10^{-12}$ g mL^{-1}. All values presented represent the mean of three to six separate determinations for each sample.

The values of the silver concentrations for all samples collected at all sites in 1977 have been plotted in Fig. 4, as a function of the length of period of collection. Using statistical techniques described by Katz (1977), Fig. 4 shows that the median and upper and lower quartiles contain no obvious trends, and it is concluded that the background silver is independent of the length of collection period. This suggests in the first place, that the dry aerosol deposition prior to the precipitation does not intervene with what is observed after the collector is opened and, in the second place, that the precipitation intensity, which is directly related to the duration of the collection period (the depth of water collected being practically constant and of the order of 1.5 mm per sample) does not influence the concentration of background silver. The site of the collector does not affect the concentrations for values between 0.3 and 2×10^{-11} g mL^{-1}; however, collector 5 in 1977 did collect 50% of the samples having concentrations equal to or greater than 2×10^{-11} g mL^{-1}. In the same way, the values of the natural silver concentrations for the samples collected in the fifteen sites in 1978 have been plotted in Fig. 5, in relation to collection period. The same statistical techniques as used in Fig. 4 were applied to the 1978 data and results shown in Fig. 5. The upper quartile plot contains a "trend" which produces the suggestion of a minimum in silver background for collection times around

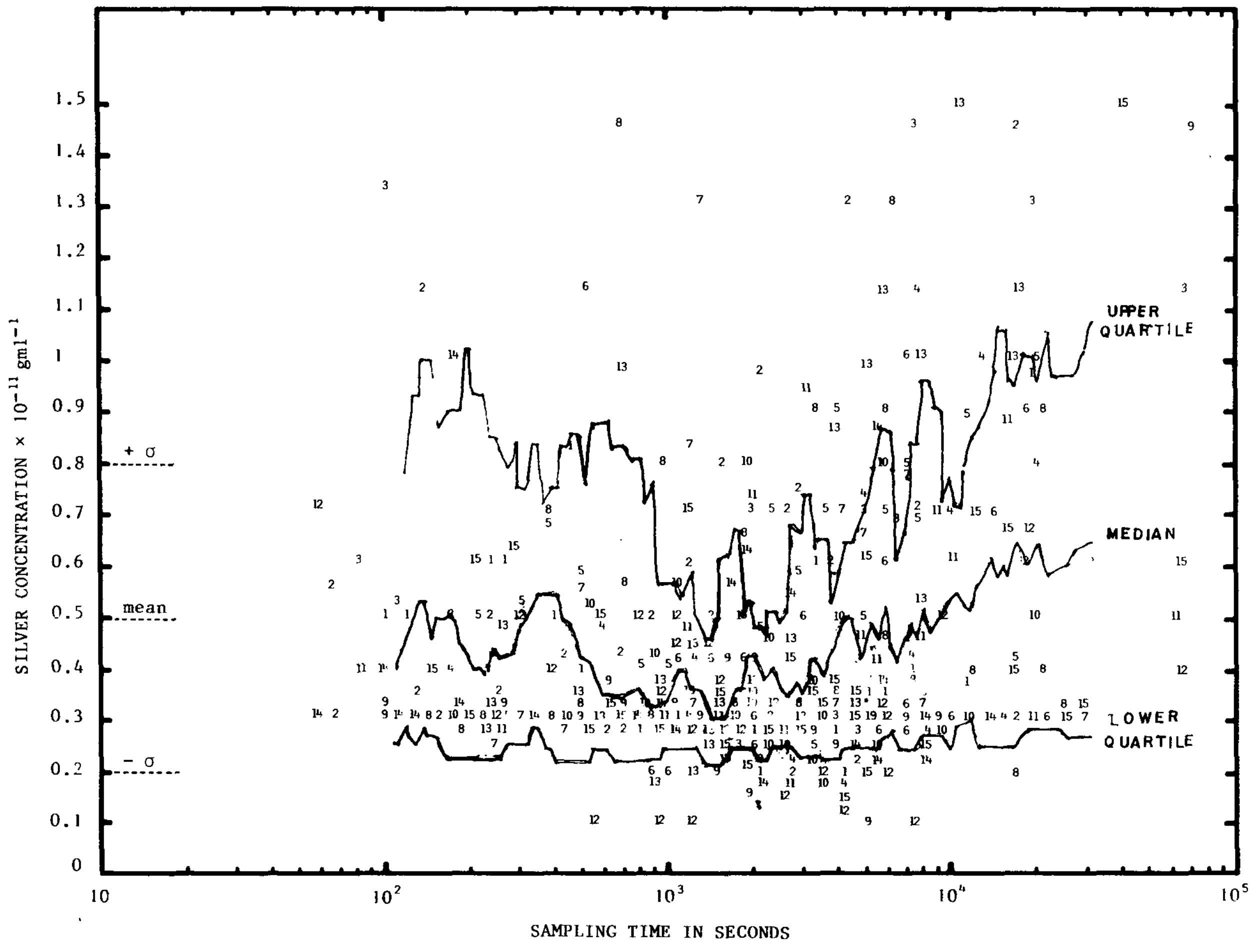

FIG. 5. Silver concentration as function of sampling time duration (1978 season), showing medians, upper and lower quantities.

10^3 s. However, this "trend" spans less than one standard deviation, and since we are not able to attribute any physical significance to this, it is concluded that the site of the collection and the length of the collection period do not influence the silver concentration and that all values are less than 1.5 $\times$ 10^{-11} g mL^{-1}.

In the same way, the variation of the mean concentrations on a site by site basis is also very small; from 0.7 $\times$ 10^{-11} to 1.1 $\times$ 10^{-11} g mL^{-1} in 1977, and 0.3 to 0.6 $\times$ 10^{-11} g mL^{-1} in 1978. The mean of the total 118 precipitation samples analyzed for 1977 is 0.9 $\times$ 10^{-11} g mL^{-1}, with a standard deviation of 0.6 $\times$ 10^{-11} g mL^{-1}. The mean for the 414 samples analyzed for 1978 is 0.5 $\times$ 10^{-11} g mL^{-1}, with a standard deviation of 0.3 $\times$ 10^{-11} g mL^{-1}. These natural silver background values are reproduced in Table 3 with those measured in other regions. Based on the ability of other studies to detect the silver from AgI particles released for seeding above these levels of background, the same possibility appears to exist in this project area.

Without making hypotheses about the trajectories of the AgI particulates in clouds seeded with the Soviet OBLAKO rockets, a comparison was made of the silver concentrations of precipitation samples which fell from clouds on a seeded day (7 August 1977), with the mean value of the natural background silver determined for 1977 (see Table 4). The quantity of silver iodide released was 672 g, coming from 8 OBLAKO fusee rockets each containing 84 g of AgI. The "seeding silver" was observed above the background on this day in the first three samples of the set collected at Site A. The restricted time span over which the "seeding silver" was observed at this site is not unlike the observations made by Linkletter and Warburton (1977) in the NHRE experiment in Colorado in which it was demonstrated that the silver appears in narrow time and spatial bands.

6. Conclusions

The principal functions of the 15 automatic collectors have been quite satisfactory with a minimum

TABLE 3. Background of natural silver (10^{-11} g mL^{-1}) in precipitation from different regions.

Region	Concentration of Ag	Number of samples	Reference
Sierra Nevada (USA)	0.4	169	Warburton and Young (1972)
Florida (USA)	5.2	45	Wisniewski *et al.* (1976)
Illinois (USA)	7.3	82	Gatz (1975)
Colorado (USA)	12.5	19	Parungo and Robertson (1969)
Alberta (Canada)	0.3	494	Zacharuk (1976)
Languedoc (France)	1.4	39	Lacaux and Ribet (1977)
Switzerland (1977)	0.9	118	This study
Switzerland (1978)	0.5	414	This study
Switzerland (1977 + 1978)	0.6	532	This study

TABLE 4. Silver concentrations in precipitation at Site A from clouds seeded on 7 August 1977.

Sample	Observed [Ag] (10^{-11} g mL^{-1})	Ratio of observed [Ag] to mean 1977 background [Ag]
1	46.0	51.1
2	5.6	6.2
3	4.5	5.0
4	2.7	3.0
5	2.8	3.1
6	2.4	2.7
7	1.3	1.4
8	1.2	1.3
9	1.1	1.2
10	<0.3	<0.4

of electronic and mechanism maintenance in the field. In particular, it has been possible to follow and sample high intensity precipitation (80 mm h^{-1} on 12 August, 1977).

The average background concentration for the two seasons of 0.6×10^{-11} g mL^{-1} ($\sigma = 0.5 \times 10^{-11}$ g mL^{-1}), is sufficiently low that the detection of the presence of AgI released from the Soviet Oblako rockets in the precipitation on seeded days should be possible.

Using the new automatic collectors, it has been shown that the observed background concentrations of silver are independent both of the length of sampling period and of the precipitation intensity averaged over the sampling period of the collectors.

In 1978, the network of 15 automatic collectors installed in the experimental zone for two months, permitted the collection of 500 precipitation samples on seeded days with a reliability approaching 100%. The analysis of those seeded samples is in progress and the results of this work will be reported in Part II of this paper.

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