in an in-line closed filtering system constructed entirely of polyethylene and polypropylene. The collected particulate material appeared to be largely biogenic. The "particulate" sample and filter were dissolved in a mixture of perchloric and nitric acid for subsequent atomic absorption (AA) analysis. The filtrate from the particulate sample was extracted with chloroform. The chloroform, along with any interfacial material, was filtered through a Millipore HA filter (24 mm, 0.45 μm) to remove the interfacial material. The chloroform extract was concentrated in a rotary evaporator, and the final few milliliters were transferred to the 24-mm filter and evaporated. The "organic" phase on the filter was then processed in the same manner as the "particulate" sample for AA analysis. After filtration and chloroform extraction, the remaining aqueous portion of the sample was acidified and buffered to a pH of 3.0. The solution was then extracted with a 5 percent (by weight) solution of diethyldithiocarbamic acid into methyl isobutyl ketone. This "inorganic" phase was analyzed directly by AA. All AA analyses were performed with an atomic absorption spectrophotometer (Perkin-Elmer model 303) with standards prepared in a seawater matrix.

We have collected several surface microlayer samples in Narragansett Bay and analyzed them for trace metals, lipids, and chlorinated hydrocarbons. The results of the analysis of two of these surface slicks and subsurface waters are presented in Table 1. Sample 1 was collected in a heavy frothy surface slick and showed enrichment factors in the surface microlayer relative to those in the subsurface water ranging from 3 to 50 for fatty acids, PCB's, and "particulate" and "organic" trace metals. Sample 2, from a less pronounced slick, had lower enrichment factors varying from slightly over 1 to 15. With the exception of iron in sample 1, no enrichment was found for trace metals in the "inorganic" phase for these or other surface microlayer samples collected in Narragansett Bay. In general, the fatty acids were normal saturated and monounsaturated acids and ranged from 12 to 20 carbons in chain length. Only three hydrocarbons were detected in these samples, and they were tentatively identified as having carbon numbers of $C_{21.0}$, $C_{22.5}$, and $C_{24.0}$ relative to n-alkanes on an Apiezon L column. The only chlorinated hydrocarbons found were PCB's which were measured as Aroclor 1254.

An enrichment factor given in Table 1 for any of these substances in the top 100 to 150 μm of the water surface suggests a much greater enrichment in the film material itself. If the film layer is monomolecular, as has been suggested (*1*), it should have a thickness of about 2×10^{-3} μm. If the film thickness is estimated conservatively as five molecular layers (10^{-2} μm) and all the chemical enrichment is in this layer, the actual concentration in the film would be 1.5×10^4 times the concentration in the top 150 μm. In sample 1, where the enrichment of PCB's in the top 100 to 150 μm is 28, the PCB concentration would be $\sim$ 60 parts per million, which would represent an enrichment of 4×10^4 in the film layer itself. The effects of the high concentrations of chlorinated hydrocarbons, lipids, and heavy metals on the diversity and species composition of the bacteria, phytoplankton, and zooplankton living in the surface microlayer are unknown.

Pollutants present in the surface microlayer of the coastal zone may easily be introduced into the atmosphere for subsequent transport to open ocean waters. The surface of the ocean is a major source of atmospheric particulate matter. The primary production mechanism for these particles is the breaking of bubbles, which ejects particles both from a central jet and from the ruptured bubble film cap (*6*). There is evidence that these particles are chemically more representative of the surface microlayer than the bulk water underneath (*7*), and thus this study suggests that the particles may be considerably enriched in pollutants if they are generated in highly polluted nearshore areas.

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Cloud Seeding Experiments: Lack of Bias in Florida Series

Abstract. *There has been concern about the possibility of selection bias in cloud seeding experiments. Covariates and experimental design have been used to obtain an estimate of this bias. The results indicate that there was no selection bias in the Caribbean and Florida series of cloud seeding experiments.*

Stigler (*1*) has made the general point that selection bias may be introduced when suitable experimental subjects arrive sequentially; he referred in particular to the cloud seeding experiments in Florida. We present evidence here that such a bias is not detectable in the experiments over the Caribbean Sea (*2*) in 1965 and over Florida (*3*) in 1968 and 1970. These references should be consulted for a description of the experimental details. In addition, we wish to make a suggestion about what to do in some experimental situations where selection bias has not been (or cannot be) eliminated by some appropriate design.

In planning the Caribbean cumulus experiments, careful attention was given to the design. This included consultations with experts in experimental design, one of whom, W. J. Youden (now deceased), prepared the randomization scheme. These experiments represent the first time that a numerical model of cumulus dynamics was used to make predictions of cloud growth and to provide a measure of "seedability" of selected clouds. The power of the experiments was greatly increased by incorporating the model prediction into their design as a covariate, since the covariate accounted for more than 90 percent of the variance. Because the covariate made use of information available up to the time of cloud selection, it could be examined for evidence of bias in selecting the clouds for seeding or for evidence that randomization failed to balance out uncontrolled variation and to provide equivalent clouds for the two test groups (seeded and unseeded clouds). No significant difference in seedability was found, that is, the samples of seeded and unseeded clouds apparently came from the same population.

In 1968 and 1970 the site of the dynamic seeding experiments was transferred to southern Florida, where it was possible to evaluate amounts of precipitation as well as the other cloud parameters. For the statistical analysis of precipitation the important covariate was the amount of rainfall in the 10-minute period before "seeding." As before, no significant difference was found between the two groups, although the seeded clouds were very slightly wetter in this 10-minute period than the unseeded clouds. However, this slight difference (whatever its source might be) is automatically allowed for in the classical covariance analysis procedure, which makes the test on the adjusted means. Again, this illustrates the importance of making use of a significant covariate if one is available, not only to increase the sensitivity of the experiment but to indicate in the treatment groups evidence of selection bias or failure of the randomization scheme to give "equal" groups. Braham *et al.* (*4*) suggest that some of the ambiguity in the results of the Whitetop experiment (*5*) may be due to an uncontrolled background effect that was present and not balanced out by the rigorous randomization scheme.

In 1968 the randomization series was generated so that the ratio of seeded to unseeded clouds would be approximately 2/1 (it turned out to be 14/5). This was to allow for more seeded clouds so that further study could be made of the physical model. Operationally, up to five experimentally acceptable clouds were "seeded" each day. The decisions on each day were known after the aircraft landed, not after each cloud. Furthermore, in all of the experiments carried out in Florida, the selection of the clouds was often turned over to the pilots or other members of the flight crew since they had neither a vested interest in the outcome of the experiment nor a theoretical knowledge of cloud behavior.

In 1970 the original design for the single-cloud experiment involved randomizing in pairs. The first cloud of the experiment was to be selected and treated according to the randomized instruction. The second cloud was then to be given the opposite treatment. For cases in which the second cloud was chosen after experimentation with the first cloud, there might have been a strong possibility of selection bias if the experimenters were able to guess the correct instruction for the first cloud from visual observation of the resulting behavior of that cloud. We had planned to test for this bias by comparing the seeding effect of the first clouds of the randomization pairs with that of second clouds. (The seeding effect is the mean amount of precipitation from seeded clouds less the mean amount from unseeded clouds.) However, randomization by pairs did not work out because the decision for seeding or not seeding in the single-cloud experiment most often had to be contingent on the decision in the area experiment. That is, the area decision governed that for single clouds on that day. Those within the area were given the treatment required by the randomized decision for the area, while those outside the area were given the opposite treatment, so that area instructions and single-cloud instructions were intermixed in a manner complicating any rational guessing strategy of the experimenters.

It can be seen from the design and problems of execution of the 1968 and 1970 single-cloud experiments that the optimum strategy for guessing the seeding instruction before choosing the experimental cloud would be difficult to devise. However, it might be suggested that it would be easiest to make this guess for the first cloud on each day of the experiment and more difficult for subsequent clouds as the number of these increases. A comparison of the seeding effect for first clouds with the seeding effect for second clouds, third clouds, and so on does not support this contention. In fact, the seeding effect computed for first clouds was actually slightly less than that for the remaining clouds.

To demonstrate the limitations of an experimenter's ability to bias the experiment in favor of seeded clouds, the two principal experimenters guessed what the seeding instruction was after visually observing the outcome on each day of the 1971 experiment (similar guesses were made in the 1970 experiment but these were not routinely recorded). Only 3 out of 11 guesses were correct, which shows how difficult it is to draw inferences from visual observations of cloud behavior, a fact recognized by those familiar with the large variability of cumlus clouds.

In the area experiment one might suspect a possibility for bias in that it is randomized by days and the scientists learn the decision at the end of each day. However, since all the eligible clouds in the area are "seeded" on each experimentally acceptable day, there is no choice and hence no possibility for selection bias. Since the termination of the experimental periods is forced by the end of the aircraft assignment and never chosen by the scientists, it might be possible in future years not to reveal the decisions until the end of the whole period. In the past this has been unavoidable because of public concern about drought and other potential environmental disturbances in the seeding area. There have been only 12 acceptable days in the area experiment to date, and, although the results are encouraging, these are too few cases for definite conclusions.

We have been very much concerned with the possibilities of selection bias in cloud seeding experiments. We agree with Stigler (*1*) in his concern "to alert future experimenters to the dangers of bias associated with this type of experiment." However, we do not believe that selection bias was present in the Florida series of cloud seeding experiments.

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