Measurements of Ice Nucleus and Associated Chloride Particle Concentrations at Mauna Loa Observatory

DWIGHT B. KLINE[1]

Environmental Research Laboratories, NOAA, Boulder, Colo.

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ABSTRACT

A series of ice nucleus measurements at Mauna Loa Observatory obtained concurrently with Millipore filter samples which were subjected to chemical analyses for chloride particles are briefly described. These aerosol concentrations displayed a pronounced relationship with flow regimes at the Observatory. The results emphasize the need for care in collecting data at this unique site to insure against contamination from low-level sources. Both aerosols increased up to nearly three orders of magnitude during sustained diurnal upslope flow conditions. The depletion of chloride particle counts with distance from the shoreline of the Island of Hawaii was in general agreement with similar data obtained in the Puerto Rican environment.

1. Introduction

The increasingly recognized need for careful and sustained monitoring of certain atmospheric constituents at sites remote from the effects of inhabited areas requires that "benchmark" observational sites be thoroughly evaluated to insure that the data are representative of conditions uncontaminated by possible local sources of pollutants. Since Mauna Loa Observatory (MLO), Hawaii, has been assigned a prominent role in a planned global network of atmospheric monitoring stations, it seems appropriate to publish the details of some measurements obtained at this site which appear to be of value in designing and conducting future observational programs at this unique location. The data reported here were obtained during a series of aerosol measurements from 1 January to 4 February 1961. Those portions of the data pertaining to ice nucleus observations have been reported elsewhere in summary form (Kline, 1963). When interpreted in the context of similar measurements obtained at other sites in the United States and, especially, when compared with a parallel series of results at Hilo, Hawaii, these data gave evidence consistent with the hypothesis that the origins of abnormal ice nucleus concentrations at MLO are dominated by, or at least associated with, low-level influences. This interpretation was questioned by Bigg (1964) and Droessler and Heffernan (1965), partly on the basis of further measurements, but primarily because of concern about the influence of low ambient humidity at MLO on the reliability of the refrigerated expansion cloud chamber technique used.

[1] Present affiliation: National Severe Storms Laboratory, NOAA, Norman, Okla.

Knowing that a subsequent series of measurement by a method independent of that used in 1961 was being planned at MLO, the results of which were subsequently published by Nagamoto *et al.* (1967), I deferred offering further details of my data for publication since there appeared to be no way of proving that the instrumental technique used was beyond dispute. In a physically rigorous sense this situation still exists. For example, the exchange of comments by Bigg (1968) and Langer (1968) seems to point to a continuing uncertainty about the interpretation of ice nucleus measurements at MLO. The results presented here, while not decisively resolving the confusion, are offered as a further contribution toward ultimate clarification of the problem of interpreting measurements of atmospheric constituents at that site. A more detailed examination of the chloride particle data reported here is presented in a companion paper by Semonin (1972).

2. Experimental approach

The original purpose in undertaking this series of ice nucleus measurements at the isolated site of MLO was to bring the question of terrestrial vs extraterrestrial influences under scrutiny. The experiment was designed to take advantage of the regularity with which thermally induced changes in local flow regimes occur at MLO, as described by Price and Pales (1963), for performing replicated measurements. That is, efforts were made to take full advantage of the opportunity for detecting any differences in the ice nucleus data between upslope and downslope flow regimes, if such existed, by selective timing of the observations

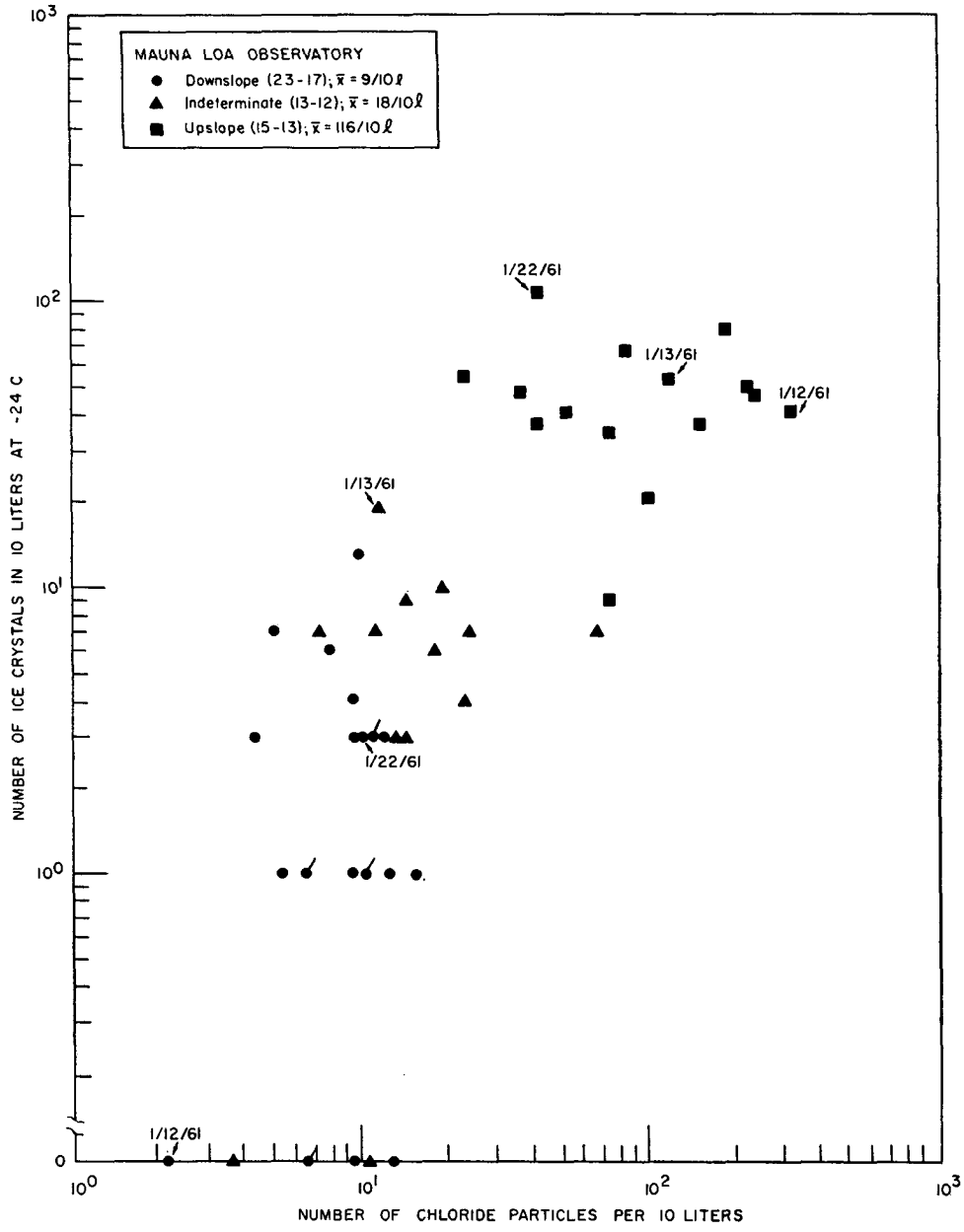

FIG. 1. Scatter diagram of the association between ice crystal counts at Mauna Loa Observatory at an expansion temperature of −24C and the number of chloride particles. The legend in the box insert (upper left) shows the number of filter samples (or observations) on the number of days during each flow regime and the arithmetic means of the chloride concentrations normalized to a 10-liter sampling volume. Tick marks designate overlapping data points, and the indicated dates are those measurements on days corresponding to the "Bowen January singularity" periods.

in conjunction with a completely independent series of aerosol measurements. For the latter purpose, Type HA grid Millipore filters were exposed simultaneously with the operation of the expansion chamber equipment for analysis of chloride particle content. Wind direction and humidity were continuously monitored during each sampling period to permit stratification of the data by presumed flow regimes. Although a northerly wind component is usually construed as *a priori* evidence of upslope flow at the Observatory, the only cases defined as such for purposes of this observational experiment were those where continuous records from an infrared hygrometer in operation at MLO during the data-collection period showed that the moisture content of the air moving over MLO had stabilized at values representative of sub-inversion layers. Samples obtained during periods with fluctuating humidity values and erratic wind conditions were classified as

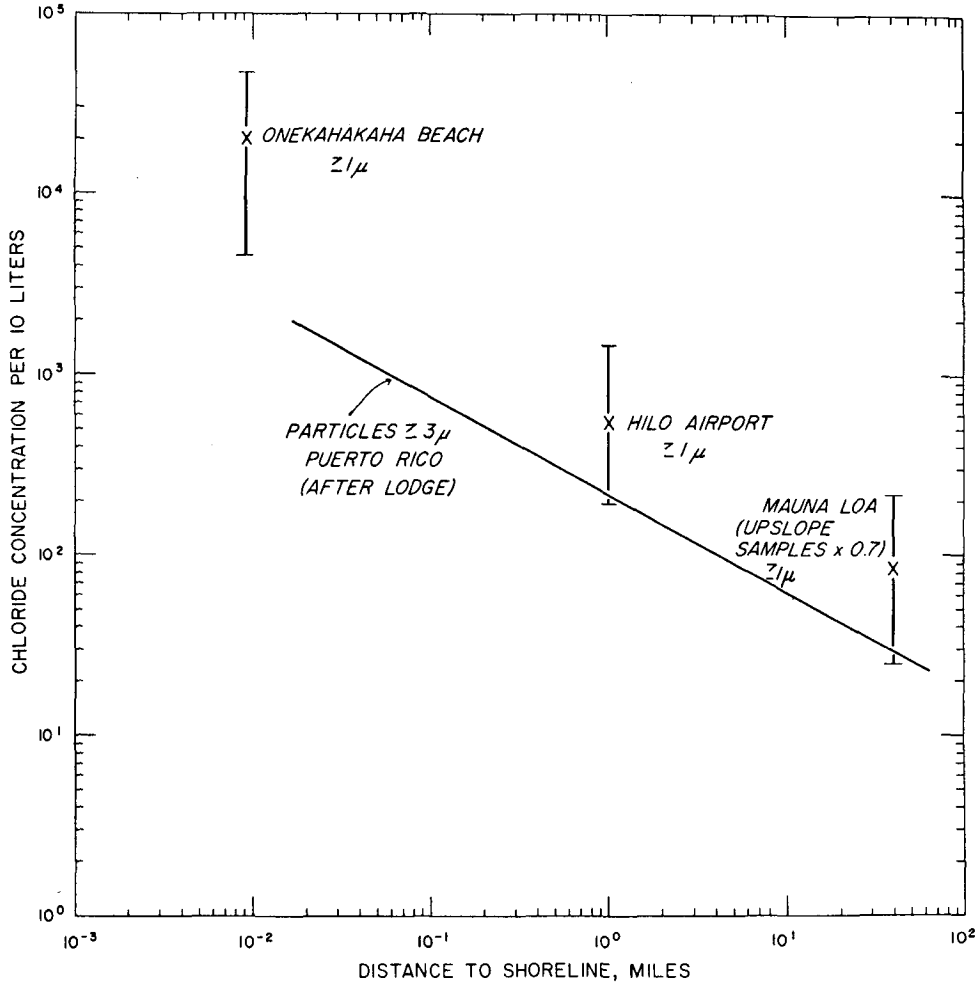

FIG. 2. Depletion of average chloride particle concentration $\geqslant 1\,\mu$ diameter as a function of distance inland to Mauna Loa Observatory from the Hawaii shoreline compared with Lodge's Puerto Rican measurements. Fiducial marks indicate the range of particle counts per 10-liter sampling volume on filters exposed at each Hawaiian location.

"indeterminate" for purposes of this investigation; samples obtained during periods with both low moisture values and non-northerly wind directions were considered "downslope" cases.

To avoid the possibility of personal bias in evaluations of these data, arrangements were made with the director and staff of the Atmospheric Sciences Section, Illinois State Water Survey (ISWS), to analyze the filters for chloride particle concentrations by the Lodge-Tufts chemical "spot-test" technique. A stratification of the filters by flow regime was furnished ISWS before any results were communicated to me. ISWS in turn did not inform the person performing the tedious chemical and microscopic analytical work of the filter stratification until the analyses had been completed. The results reported here are, without exception, in accordance with the classification of flow regimes specified before the filter analyses. A supplementary and exploratory series of 18 filter samples was obtained during the period 2 January to 10 February 1961 at General Lyman Field and Onekahakaha Beach near Hilo, Hawaii, to provide background data representative of sea level and littoral zone conditions.

3. Results and conclusions

A scatter diagram of the ice nucleus counts at -24C plotted against the concentrations of chloride particles observed on 51 filters exposed simultaneously with the periods of ice nucleus measurements at MLO are shown in Fig. 1. All data have been normalized to a 10-liter sampling volume (the volume of the refrigerated expansion cloud chamber used). Also indicated for their possible intrinsic value are the January singularity dates as indicated in earlier published results of ice nucleus measurements, for example, the data summarized by Bowen (1956) and Kline and Brier (1958), which motivated this investigation. For reasons evident

from Fig. 1, any inferences about an extraterrestrial source of nucleating agents at MLO must take into account these indications of a striking association between both the chloride particle and ice nucleus concentrations and the trajectory of the air reaching the Observatory. Although all known precautions were taken to insure the reliable operation of the refrigerated expansion chamber equipment, these results should not be construed as implying that the ice nucleus data per se are totally beyond question. Nevertheless, to the extent that the chloride particle content can be regarded as a natural atmospheric tracer in the Hawaii environment, the association reflected in these data is interpreted as demonstrating that any aerosol data or other measurements of atmospheric constituents at MLO should be obtained during carefully selected periods to minimize the confounding of results by contamination from sources at lower elevations. Data obtained during the early phases of the onset of the diurnal upslope flow regime at MLO are likely to lead to uncertainties in any interpretation of results. In this series of measurements, high ice nucleus and chloride particle counts were consistently observed only where the upslope flow regime was sustained for several hours, and the ambient humidity reached steady or quasi-steady values representative of the sub-inversion layer. Similar trends occurred with respect to ice nucleus measurements at −20C. However, values at −24C were selected for routine monitoring at MLO in view of the rarity of ice nuclei activated at warmer expansion cloud chamber temperatures, especially during downslope flow regimes.

Although the filter samples obtained in the Hilo area after the observational experiment at MLO had been essentially completed were intended only for exploratory investigations of the background aerosol content at sea level, it is of interest to compare the trends in chloride depletion with respect to distance from the shoreline with the results reported by Lodge (1955) based on limited samples in the vicinity of Puerto Rico, as shown in Fig. 2. The results appear to be compatible, considering minor differences in analytical technique, the small number of samples in both series, and the fact that the Hawaiian shoreline samples were obtained nearer to the littoral zone and during abnormally heavy surf conditions. For purposes of the graphical presentation in Fig. 2, the MLO chloride data have been reduced by a factor of 0.7 to adjust for the compression of sampling volume if reduced to sea level conditions. Semonin's companion paper provides further insight into the depletion phenomena. This presentation assumes, of course, that the movement of the air up the slope of Mauna Loa is a direct trajectory from the Hilo area rather than by a circuitous route.

Although the quantitative values of ice nucleus concentrations are known to be significantly influenced by the characteristics of the particular measuring technique used, there appears to be no basis in the published literature for challenging the validity of the gross differences found in chloride concentrations in this experiment. The fact that high ice nucleus counts were associated with abnormal chloride particle concentrations at MLO should not be construed as a causal relationship. Data not reported here indicated no obvious connection between simultaneous ice nucleus and chloride particle counts at sea level in the Hilo area, or in the littoral zone of the island.

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