Comments on "Freezing Nuclei Derived from Soil Particles"

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21 August 1973

Rosinski, Nagamoto, Kerrigan and Langer (1973), to be referred to as RNK&L, claim to have found evidence showing that the freezing nuclei in precipitation samples originate primarily from the breakup of large soil particles upon entering into water drops. Such particles transferred into water drops in the laboratory were found to be capable of releasing small satellite particles and to thereby create large numbers of freezing nuclei. By considering various mechanisms for the transfer of aerosol particles to precipitation elements, RNK&L concluded that satellite production in natural cloud droplets will be the most prolific source of small particles in rain, overshadowing other contributions. Thus, they purport, measurements of freezing nucleus contents in precipitation cannot provide useful information on the role of the freezing nuclei in the precipitation processes.

The purpose of this note is to point out additional facts and considerations which tend to contradict the deductions of RNK&L. Since many of the points to be discussed have not been presented previously in the open literature, somewhat more detail is included here than what would be necessary otherwise.

Some comments are in order regarding the laboratory experiments. For simulating atmospheric aerosols, the use of soil particles separated by sieving has some disadvantages, especially when the physical state of the soil grains is the main aspect to be examined, since soil particles ablated under natural conditions may be selected rather differently than in the sieving process. Another problem is associated with the determinations of the numbers of the satellite particles. Optical and electron microscopy were used to make these determinations. For the smaller particles, it seems likely that the suspensions were evaporated onto electron microscope grids. In that case, precipitation of dissolved matter could have produced additional particles; the amount of dissolved material reported in the paper is sufficient to produce 10^{11} cm^{-3} particles of 0.05 μm diameter, a number well in excess of the number reported by the authors.

An additional factor is worth noting in connection with the nucleating abilities of soil particles. Schnell and Vali (1972) reported that organic matter in soils seems to be the greatest contributor to the nucleus contents of soils. The organic nuclei are small in size (<0.05 μm). These nuclei exhibit a yet poorly understood "saturation" phenomenon (Schnell, 1972), i.e., the concentration of nuclei in a suspension appears to reach an upper limit with increasing amounts of material in the suspension. The data of RNK&L show a similar effect in the lack of proportionality between particle and nucleus concentrations. Thus, the presence of organic nuclei in soils and some of the resulting effects can be used to give alternative interpretations for some of the results given by RNK&L.

Important to the discussion is to examine the nucleus contents that were observed to result from large

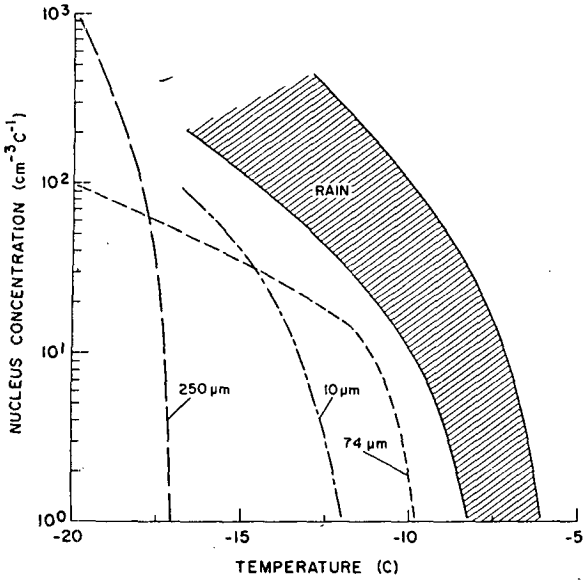

FIG. 1. Comparison of the nucleus concentrations in three different suspensions of soil particles with the nucleus contents of rain samples.

soil particles upon immersion in water and to compare these observations with the nucleus content of natural precipitation. Using data from Rosinski (1967), three of the experiments of RNK&L can be selected as being roughly representative of natural rain and hail with respect to the concentrations of large particles. These are for 10 cm^{-3} of 250-μm particles, 100 cm^{-3} of 74-μm particles, and 1000 cm^{-3} of 10-μm particles. Results for these tests (from Fig. 2 of the original paper) are replotted in Fig. 1; the range of concentrations is also given for rain from the same geographical location from which the soil samples used by RNK&L originated. The relative positions of the curves reveal that the artificial preparations had considerably fewer active freezing nuclei than what is normally found in rain samples. In terms of temperatures, the soil suspensions had freezing temperatures which are from 4 to 10C colder than for the rain. Only in very rare instances are rain samples found with nucleus contents as low as those of the soil preparations; these are usually rain samples from long periods of steady rain for which no particle concentrations are known but which can be expected to contain relatively few large particles. It is thus seen that the activity produced by the hydrosolization of large soil particles is far below that found in rain samples; therefore, the suggestion that satellite particles are responsible for the nucleation activity observed in rain samples appears untenable.

The real question that remains is: What is the origin of the freezing nuclei that are being found in precipitation samples? Undoubtedly these nuclei derive from aerosols transferred to the precipitation, but a complete, or even partially realistic description of the transfer mechanism would require knowledge of the size distribution of the *active* aerosols in their original form. RNK&L attempted to deduce the origins of freezing nuclei in rain by comparing three possible sources, namely 1) small particles attaching themselves to condensation nuclei, 2) direct diffusional attachment of small particles to cloud droplets, and 3) the breakup of large soil particles in water droplets. They concluded that 3) dominates over 1) and 2) by a factor of 100 or more. Quantitative as well as qualitative arguments can be raised with the description given by RNK&L. First, the input aerosol size distribution assumed by RNK&L is distorted due to the fact that the concentrations of large particles (>10 μm) were taken from data pertaining to dust-storm conditions whereas for small particles (<1 μm) clear-air background measurements were used. The resulting composite distribution corresponds to a slope of -1, contrary to the generally accepted -3 or -4. This distortion favored the argument presented by RNK&L. Second, not all mechanisms of aerosol scavenging were considered. This is important because the role of soil particle breakup in affecting nucleation is critically dependent on the stage of precipitation development at which the large soil particles get into water.

Some elaboration of the second point given above is worthwhile. The prime distinction that needs be made is whether the nucleating particles entered the precipitation elements during development of the cloud or during fallout. If the aerosol transfer was directly to cloud droplets, nuclei that may be released from large soil particles can initiate ice the same way as if they had gotten into the precipitation elements as separate entities. The detection of these nuclei in the rain would then provide relevant information. If the aerosol trans-

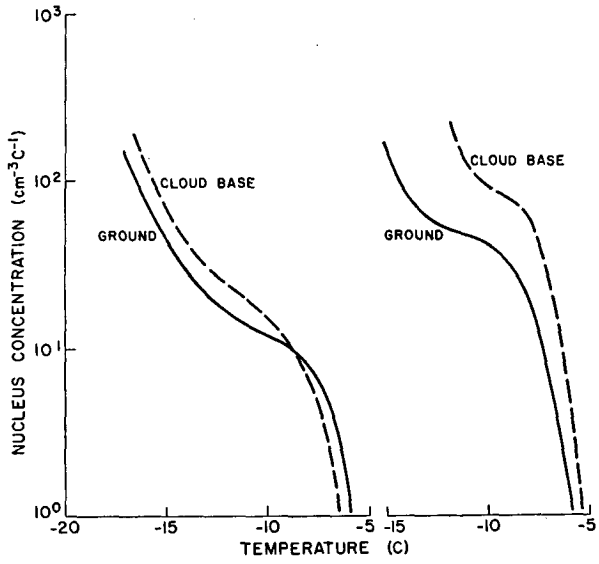

FIG. 2. Two pairs of nucleus spectra for near-simultaneous samples at cloud base and at ground level from measurements on 11 July 1972.

fer takes place during fallout, satellite particles would indeed endanger the usefulness of nucleation measurements in rain. A comparison of the nucleus contents of precipitation at cloud base and at ground level can answer this question. Fig. 2 shows nucleus spectra for two typical pairs of such samples. The freezing nucleus content of rain at cloud base is generally somewhat higher than at the ground. Thus, losses rather than increases of nucleus content are evident. Loss of nucleating activity in water has been demonstrated before (Vali, 1968). The possibility still remains that nuclei are collected during fallout and that this contribution is offset by a loss of those nuclei previously contained in the rain. The very systematic relation between cloud base and ground samples, and especially the recognizable similarity in the shapes of the corresponding spectra, make such a fortuitious cancellation of two effects very unlikely. Therefore, it may be concluded that the majority of freezing nuclei found in rain have entered the precipitation elements during the formative stages of the precipitation. RNK&L correctly assumed this to be the case.

Further insight into the ways that freezing nuclei enter precipitation elements may be obtained from a comparison of the nucleus contents of simultaneously collected rain and hail samples. Hailstones acquire most of their final mass while growing as ice. Raindrops were either never frozen or were small enough ice particles to melt before reaching the ground; in either case, the raindrops were obviously in the liquid state much longer than hailstones. Observations show nearly identical nucleus contents in rain and hail collected simultaneously. A thorough explanation of this observation is not yet possible, but the rain-hail comparison points to the fact that the nuclei were present in the cloud droplets which compose the larger precipitation elements (raindrops or hailstones) and that no additional nuclei were released into the raindrops by satellite production. This being the case, it may be argued that the nuclei found in precipitation were indeed effective in the cloud also. An important point of this argument rests on the fact that the numbers of freezing nuclei in rain and hail are much smaller (on a unit volume basis) than the numbers of cloud droplets which formed the precipitation and is just comparable to the number of raindrops. The only other way to reconcile our observations with the hypothesis of satellite nuclei would be to assume that satellites are produced during the brief period (~5 min) during which the hailstones are melted in preparation for analysis. If, however, the production of satellite nuclei reaches its final equilibrium in such a short time, then is is reasonable to think that the release would have taken place while the soil particles were in cloud drops, in which case, again, the nuclei would have had an opportunity to cause the cloud drops to freeze.

To recapitulate, evidence obtained from examinations of the freezing nucleus contents of natural precipitation samples seem to contradict the suggestions of RNK&L. The question of origin of the nuclei in precipitation is far from resolved. The mechanism discussed by RNK&L [and previously mentioned by Mason (1971, p. 164)] should be considered, but the data so far presented are insufficient to find the actual contribution of the process. To usefully comment on the origins of freezing nuclei in precipitation, size distribution data for the satellite nuclei (not total particles) will have to be compared with more realistic estimates of the magnitudes of aerosol-to-hydrometeor transfer processes and with the size distributions of nuclei in natural rain.

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