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ATMOSPHERIC ICE NUCLEATION — A REVIEW

by

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RÉSUMÉ

On résume brièvement l'état actuel des connaissances sur les sources et sur les mécanismes d'activation des noyaux atmosphériques. On établit des comparaisons avec des déductions tirées d'observations sur la formation de la glace dans les nuages. La discussion laisse de côté de nombreux autres points tels que ceux concernant la composition, les dimensions, le transport et la préactivation ou la désactivation des noyaux.

ABSTRACT

The current status of understanding of the sources and activation mechanisms of atmospheric nuclei are briefly reviewed. Comparisons are made with deductions drawn from observations of ice formation in clouds. The composition, size distribution, transport, pre- and de-activation of nuclei and many other topics are not discussed.

INTRODUCTION.

A discussion of the topic of atmospheric(*) ice nuclei should include quantitative descriptions of the sources, distribution and temporal variations of nuclei and, also, of the ice formation that may be caused by these nuclei in the atmosphere. Such a broad goal is not now within reach however, as it has to be recognized that the expression of ice nucleating activity in the atmosphere can follow various paths (nucleation modes, mechanisms) and that the probability for each path depends on the characteristics of the nuclei and on the conditions to which they become exposed in the atmosphere. A major consequence of this fact is that measurements of ice nuclei, and the use of those measurements for predicting primary (**) atmospheric ice formation, become extremely difficult. In fact, accomplishments in this field to date must be accepted as being mostly

(*) The emphasis is on nuclei of the troposphere even though the more general term of atmospheric ice nuclei is used. For stratospheric nuclei some of the discussion applies although different aspects would have to be considered regarding sources, sinks and activation mechanisms.

(**) The distinction is from secondary mechanisms, or ice multiplication processes of ice formation.

exploratory steps rather than definite findings. Needless to say, the topic remains a subject of active research even though the pace of activity is now perhaps slower (more deliberate ?) than a decade ago. The purpose of this review is to briefly summarize recent investigations, to examine what generalizations may be warranted, and to suggest directions for future efforts. Emphasis is on the major characteristics and the measurement of atmospheric ice nucleus populations. Topics like the composition, sizes, and pre-activation of nuclei, nucleation time-lags and other details are not taken up, as there are no major new developments to report and because the interpretation of the evidence on these topics will remain somewhat vague until the major questions discussed in this paper are clarified. The activities of nucleating substances, the influences of chemical interactions, the properties of nucleating sites and related topics are not discussed either ; these aspects of the subject are covered in other recent reviews [5, 78, 88]. Predominantly, the findings of the last 5-10 years are considered here. For a more complete historical perspective, for greater depth on many of the topics, and for alternate points of view, the reader should consult earlier reviews [5, 78, 89, 90, 91, 92, 93, 94, 95] which this work merely intends to supplement, not replace.

MECHANISMS AND MEASUREMENTS.

Faced with the complication of having various possible mechanisms of activity in the atmosphere there are two major options for performing measurements of ice nuclei : the simulation approach [1] and the mechanistic approach [2]. In the former, one tries to recreate particular atmospheric conditions in an instrument and observe the numbers of ice crystals that form. In the latter, separate measurements are performed for each mode of nucleation, and atmospheric ice formation is calculated for the appropriate conditions as a superposition of the individual modes.

The simulation approach was at the heart of the earliest ice nucleus measurements [1] and was implicitly assumed in the use of many other subsequent cloud chamber devices. Many years of development of the various types of smaller cloud chamber instruments led to the elimination of some basic shortcomings like extreme supersaturations and short cloud lifetimes and, later, to the realization that the precise details of the cloud formation process and of the characteristics of the cloud influence the outcome of the measurements significantly [3]. Thus, it was concluded [4, 5, 6, and others] that for most chambers the simulation of cloud processes was invalidated by instrumental limitations or artifacts and that it was not possible to simulate at will different types of natural cloud situations. This conclusion holds for chambers based on either the expansion or mixing principles, both of which have been used in a large variety of forms. The instruments which held the greatest promise of reliable simulation, namely the slow-expansion chambers of 300 liter to several m^3 volumes, have not proven very productive for studies of atmospheric ice nuclei, probably because of their lack of portability and difficulty of operation. Recent results from cloud chamber devices focus on the interpretation of the observations in terms of nucleation mechanisms ([7] for example) so that the simulation approach seems essentially abandoned at present.

Measurements of the different modes of activity for atmospheric ice nuclei have been, and continue to be, under development. The filter method [8] with exposure of the sample to a controlled vapor field became widely used over the last 20 years. This method was viewed by different researchers either as a way of observing several nucleation modes or as responding primarily to deposition or to condensation-freezing nuclei. In fact, the large number of particles compacted onto the filter interact with the vapor field in complex ways leading to rather unpredictable results when working with aerosols whose composition is not known *a priori.* This situation has been quite well documented, mostly through experiments with artificial aerosols [9, 10, 11, 12, 13, 14]. One general finding is that the concentrations of atmospheric ice nuclei detected on the filters in vapor chambers exhibit an exponential dependence on supersaturation [15, 16] with no discontinuity at water saturation. The reason for this latter fact is not clear ; it may result

from the lack of specificity of the technique for deposition, or could be a true characteristic of the nuclei.

Over the past five years, a number of new instruments have been constructed for specific mechanisms of nucleation. Schaller and Fukuta [17] used an ingenuous wedge-shaped chamber to simultaneously create a range of different saturation conditions. When used with artificial aerosols the observations yielded threshold values for deposition and for condensation-freezing activity. While numerically different, the shapes of the activity boundaries for different substances were found to be similar, so that the results have some noteworthy generally, possibly valid also for atmospheric aerosols. The instrument does not seem suitable for direct observations of natural nuclei.

Condensation-freezing activity was investigated with continuous-flow chambers in which ice coated plates at different temperatures provide the controlled supersaturations [18, 19, 20]. The same conditions can be covered in these instruments as in the static diffusion chambers used for filter processing, but with the advantage that the aerosol remains in its natural dispersed state. Deposition nuclei also become activated in these devices and very small (rapidly diffusing) contact nuclei can also make a contribution to the ice crystal count, but each of these components can be separated out with reasonable definition via specific tests. It is noteworthy that the supersaturation spectra from these observations also cross water saturation without marked discontinuities just as with filter samples, lessening the chance that this is an artifact. The potential demonstrated by these continuous flow devices certainly warrants further development (in the detection of the ice crystals, portability, etc.) and continued use.

For the detection of immersion-freezing nuclei, Schnell [21] developed a method in which aerosols are collected by a membrane filter and the freezing of nucleus-free water drops on the filter is observed as the filter is gradually cooled on a cold stage. Perhaps to a lesser degree than in the vapor activation of filter samples, but some interference from the highly compacted aerosol can still be expected (chemical interactions, masking, clustering) and this aspect needs further evaluations. Also, some deposition nuclei near the drops might become activated and the resultant crystals may eventually touch and freeze the drops. In spite of these misgivings the method seems to offer a useful technique for observing immersion-freezing nuclei and is capable of doing so even at temperatures close to 0 °C. Comparison with an earlier, similar method [22] would also seem to be worthwhile.

The detection of contact nuclei was attempted by three newer techniques [23, 24, 25]. It is likely that the settling cloud chamber [26] also responded primarily to contact nuclei. The difficulty with Deshler's [25] and with Ohtake's [26] measurements is that the deduced nucleus concentrations depend on what sizes are assumed for the nuclei ; the instrument of Vali [23] loses detection efficiency for particles near 0.01 μm. In Cooper's experiments [24], cloud droplets were allowed to settle onto a filter bearing the sample aerosol. This technique avoids size dependence but the effect of particle penetration into the filter and the reasons for the reported aging of the filter samples need elucidation. It is not readily evident which of the three methods, or perhaps yet another, could be most easily perfected in the future and used as the basis for constructing an instrument for field use.

Meaningful distinctions among nucleation modes have been taken for granted in the foregoing discussion of instrument responses. This is perhaps warranted for operational definitions, i.e. for considering sequences of events leading to nucleation in the atmosphere. There is an element of the simulation approach in this — the simulation of individual processes is the intent of the definitions. The theoretical distinctions are much less clear, as discussed by Knight [5], for example. The observed time lag for freezing after contact with the nucleating particles [23, 27] also calls into question, to some extent, the meaning of « contact » nucleation. Clearly, the ultimate test of whether any definition is meaningful or not and whether an instrumental observation is useful or not is the degree to which the results can be transposed into descriptions of atmospheric

processes. One should not exclude the possibility that the currently used definitions will have to be modified and augmented if analyses of the processes at work in the measuring devices, or observations of atmospheric processes of nucleation, reveal fundamentally new information.

SOURCES AND CONCENTRATIONS.

In view of the difficulties in obtaining meaningful measurements, and especially because of the major influence exerted by aerosol and gas composition, by humidity, and by other factors on ice nucleus measurements, it is necessary to be very cautious in accepting evidence regarding the sources and atmospheric concentrations of ice nuclei. This attitude is well reflected in the evident struggle of most authors in evaluating their observations and in drawing conclusions. Hence, this overview also is presented with a critical attitude, but not without some optimism that the main patterns focused on here are strong enough to be confirmed rather than overturned by future findings. Ice nuclei will be referred to here only in a generic sense, since in the majority of measurements the mode of nucleation could not be well defined. Furthermore, it must be remembered that essentially all available data are for temperatures of — 15 to — 20 °C. There is a dearth of information concerning nuclei active at the warmer temperatures and this fact alone seriously limits the usefulness of the available data for the prediction of cloud glaciation.

Evidence continues to mount that terrestrial sources provide the major input to the atmosphere. Studies at coastal sites showed that air masses which travelled over land contained more ice nuclei than those having trajectories over the ocean [28, 29, 30]. Long-term averages of ice nucleus concentrations in a network of stations over North America showed increases with increasing distance inland [31]. Even at the South Pole the nuclei of ice crystals were found to be clay particles [32], just as has been the case with similar examinations elsewhere [33, 34, 74]. The concentrations of nuclei over land and close to shorelines decrease with altitude [45, 58, 59, 60, 61, 77] consistent with the source of nuclei being at the surface.

As a counterpart to the results quoted above one could surmise that the oceans are not important sources of atmospheric ice nuclei. A direct examination of this point [35] revealed a variable situation, with nuclei active at warm temperatures sometimes encountered. The rather complex argument [36, 76] according to which nuclei are generated in continental air as that air passes over the ocean needs re-examination to assure that it was not based on instrumental artifacts. Measurement difficulties in maritime air were clearly identified by Wisniewski and Langer [28]. Perhaps the most compelling evidence that oceans can be sources of nuclei, even if of lesser importance than terrestrial sources, is the interpretation of Bigg's data [37] in terms of biogenic sources [38], and the isolation of nucleating substances from ocean waters [39, 40].

The third major possibility, that of extra-terrestrial sources of ice nuclei, continues to receive no support [64, 75].

Returning to the discussion of terrestrial sources, evidence keeps emerging in favor of both mineral and of biogenic origins of the nuclei. Observations of high ice nucleus concentrations in desert dust transported from long distances [96, 97, 98] identified the potential importance of desert soils as sources of ice nuclei, with the implication that minerals are the nucleating substances in the dust. Laboratory tests with minerals gave somewhat weak support to this possibility (cf. summary in [91]). As mentioned already, ice crystal centers are almost always identified as clay particles [32, 33, 74] even in atmospheric nuclei activated on filters [34] and also in crystals produced in a cloud chamber from aerosolized soils [74]. In the latter reference it is stated that < 10 % of the nuclei were organic material, which is undoubtedly an underestimate since in that work, as in many others, the techniques used for identifying the central particles in ice crystals cannot effectively detect organic substances. On the other hand, the original findings demonstrating the existence of biogenic sources [38, 41] are receiving some further

support [42, 43, 44]. An extensive series of measurements in New York State [45] indicated a correlation of ice nucleus concentrations with soil type in a direction that may well be due to the influence of organic/biogenic sources. Overall, evidence concerning the atmospheric role of biogenic nuclei is not yet comprehensive and direct enough to be convincing ; nonetheless, the motivation for some further work is clearly substantial.

Anthropogenic production of ice nuclei has been demonstrated for certain steel and copper smelters ([46] for example) but coal-fired power plants were found [47, 48] not to produce nuclei (or detection to be interfered with). Urban areas, with their mixture of gaseous and aerosol products, were found to either decrease ice nuclei concentrations [50, 51], to not be detectably strong sources [30, 45, 52] or to provide very high nucleus concentrations [66, 86]. The reason for such disparate findings is not known ; the many possible explanations would have to be tested by careful experimentation. Clearly, this is not an insignificant question. Concerning one possible urban source, the recent indications are that automobile exhaust does not form ice nuclei [30, 53], counter to early evidence on profuse ice nucleation by some reaction product of exhaust gas with iodine [54]. Another anthropogenic product, the smoke from sugar-cane fires was reported to be an important source [55], suggesting that forest or prairie fires in general might produce ice nuclei.

Two examinations of active volcanoes [56, 49] failed to yield indications of the presence of ice nuclei in the plumes. It is not clear how to reconcile these observations with earlier reports [99, 100] and with laboratory experiments which revealed moderate activity for volcanic ash [14]. It bears repetition though, that when dealing with the sampling of concentrated aerosol and gas plumes, the problems of obtaining valid measurements of ice nuclei become especially amplified.

Most of the deductions discussed above regarding ice nuclei sources were drawn from *comparisons* of measurements made under different conditions with a given instrument, either cloud chambers (simulation) or filters. For such comparisons repeatability of instrumental conditions is the prime requisite, and that reduces, though certainly not alleviates the impacts of the uncertainties introduced by the measurement techniques. When considering the *absolute values* of observed ice nuclei concentrations those uncertainties become debilitating. The large ranges of variability of meteorological parameters which interact with the generation transport and removal of ice nuclei does not help matters either. Furthermore, most of the available data were collected at surface sites where maximum variabilities can be expected. In light of all these problems it is probably not warranted to accept at face value the geographical variations in ice nucleus concentrations, implied by the apparent large differences in the data reported, for example, for Kenya [62], Colorado [61], Florida [28, 36], Italy [63], the Western U.S. [59, 65, 31, 67], the Ivory Coast [29], Germany [52], The United Kingdom [20], the USSR [13], New York State [45] or for Antarctica [64]. The range of measured concentrations is large, nearly two orders of magnitude (see Fig. 1), but many data sets have average values for — 20 °C between 0.1 and 1 l^{-1} with factors of 3 to 8 lower concentrations at — 16 °C. It is noteworthy that long-term variability is very similar in almost all of the data sets : factors of 15 to 100 to encompass 90 % of the observed values [57, 67]. This large variability is evident demonstration of the influences of sources, of sinks (clouds and precipitation) and of variable transport trajectories (horizontal and vertical). On shorter time scales, hours to days, the ice nuclei concentrations can remain more constant [61, 68] although very rapid and large (factor 10) changes can also occur, for example at the passage of thunderstorm outflow gusts [68, 69]. Otherwise, correlations with meteorological variables are usually quite poor which is fairly surprising and is in seeming contradiction with the notion that nuclei originate from specific sources. Maybe the generation of ice nuclei is more complex than that simple notion ; perhaps the ice nuclei form from the reactions of several components, or complications possibly arise from the development or decay of nucleating sites with time under the influence of environmental conditions.

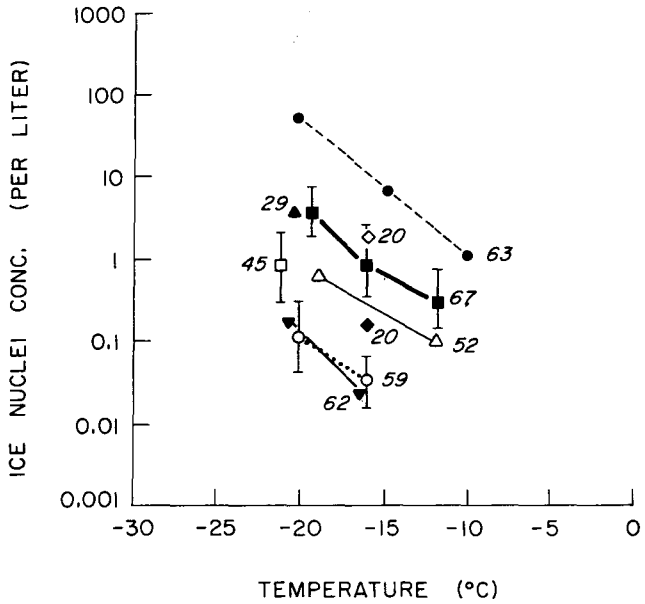

FIG. 1. — Examples of observed ice nuclei concentrations. Numbers for each data set indicate the data source in the reference list. From reference [20] two points are shown, each for — 16 °C and water saturation but from two different instruments. From reference [67] the data for contact-freezing nuclei are shown. In all cases the original references should be consulted for details. The vertical bars are geometric standard deviations for the frequency distributions of the observed concentrations.

To reiterate, it is clear that progress in the identification of the origins of atmospheric ice nuclei and in the quantification of their concentrations is critically dependent on the development of measurement techniques which are free from influences of other atmospheric variables or have those influences well characterized. In the latter case, independent measurements of the order parameters must accompany the ice nuclei measurements (more extensively, but not unlike the way simultaneous CCN measurements were used to obtain better corrections for the filter method of ice nucleus measurements). In the meantime, only very crude and tentative statements can be made concerning the atmospheric ice nucleus populations.

EVIDENCE FROM CLOUDS.

Since the overriding interest in studying tropospheric ice nuclei (setting aside for a moment interest in the role of ice nuclei in frost injury to plants) is to predict ice formation in clouds, it is worthwhile to briefly examine what can be concluded from observations of ice particles in clouds. This reverse path is not an easy one either ; it is made complicated by the processes of secondary ice particle production and mixing, plus the difficulties of following the trajectories of ice particles back to their point of origin where they are too small to be directly observable. In spite of these difficulties, and because of the considerable spurt of in-cloud studies from aircraft over the past decade, some results are beginning to emerge. Of interest here are data which were collected in clouds where it was possible to identify primary ice particles by their morphology and where cases with conditions suitable for ice multiplication could be excluded (similarly to the classification given in [80]). The results of a summary recently prepared by Cooper [70] are shown in Figure 2. The concentrations of ice particles observed at different geographical locations and in a variety of cloud types are shown as a function of temperature. The concentrations were directly measured for particles which have grown to 100 to 300 μm in

size, i.e. 2 to 5 minutes after nucleation ; the temperatures refer to those at the point of origin of the ice particles as best as they could be determined. The fact that concentrations in Figure 2 rise monotonically with decreasing temperatures is corroborative evidence that indeed primary ice particles only are included in the data sets. For reasons already mentioned, the accuracy of this type of data is not great, perhaps within one order of magnitude for individual points. The typical spread of observations for any temperature is usually greater than that, as shown in the figure for one of the data sets, no doubt due to the combination of meteorological variations with fluctuations in ice nucleus concentrations.

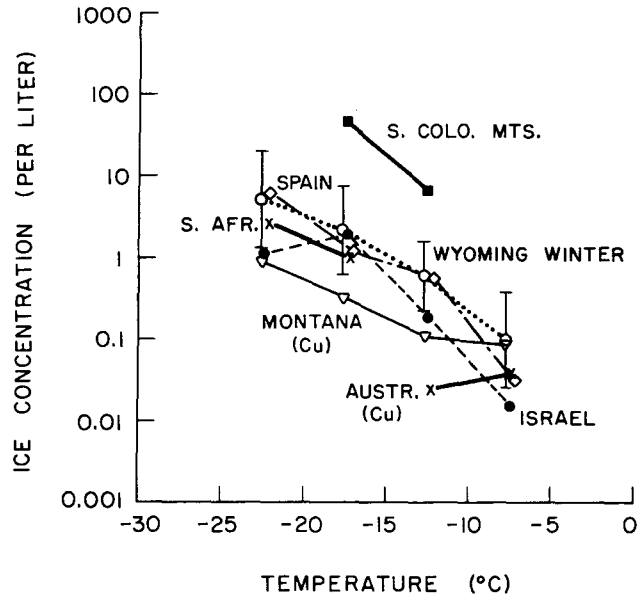

FIG. 2. — Summary of observed ice particle concentrations for clouds in which secondary processes of ice formation are believed to have made no significant contribution. Temperatures refer to the origins of the ice particles ; the data are averaged over 5 °C intervals. The error bars for the Wyoming data (one standard deviation) show the spread in concentrations typically encountered in such data. Sources are : Israel [81] ; Australia [82] ; Wyoming [83] ; S. Colorado [84] ; Montana and S. Africa [85] ; Spain [87]. (Figure compiled by Cooper [70]).

The most remarkable fact shown in Figure 2 is the similarity of the various curves (with one exception), in spite of the diversity of locations and cloud types involved. This could almost be interpreted as indicative of a worldwide uniformity of nucleus concentrations and of cloud activation mechanisms, but that argument is weakened considerably by the relatively large spreads of observations about the means in all of the data sets. On the whole, the situation with respect to primary ice particle concentrations is not altogether different from that of ice nucleus concentrations : large variabilities but worldwide similarities of averages. The absolute values at the center of the cluster of lines lie about an order of magnitude higher than the corresponding modes of the ice nucleus concentrations quoted earlier. The slopes (temperature dependence) for ice particles and for ice nuclei are quite similar, although the range of temperatures for most of the ice nucleus observations is quite small. Both ice crystal and ice nucleus concentrations should decrease sharply as 0 °C is approached, but there are no data available to show this, except perhaps the freezing nucleus content of precipitation [71, 72].

Cloud studies can also afford some insight into the mechanisms of nucleation. In mountain cap clouds, where the air particle trajectories can be reasonably followed by research aircraft, it was found that some ice crystals formed in the ice-supersaturated

zones (deposition nucleation) before a water cloud formed and that there was a rapid increase in ice concentrations after passing the condensation level [73]. This points to condensation-freezing activity, or to contact-nucleation by rapidly diffusing particles in these clouds. Some model predictions, with assumed ice generation processes, have produced a measure of agreement with these observations, so that credence can be given to the reality of the assumptions. It is certainly to be hoped that cloud observations and model descriptions will continue to help approach a better understanding of atmospheric ice nucleation.

SUMMARY.

When taking a bird's-eye view of the data on atmospheric ice nuclei and making a comparison with the data on ice particles from primary mechanisms in clouds, one can note some convergence of the two data sets. Certainly, the situation is not as grave as was noted in the early 70's when it wasn't yet clear that a distinction has to be drawn between primary and secondary mechanisms of ice formation in clouds. Yet, as has been summarized in this paper, there are many major problems to overcome before truly meaningful comparisons can be made. Even apart from details, the comparisons now possible for any data set look at statistical measures of ice nucleus populations and of ice crystal populations from observations taken over long periods of time and in varying degrees of special correspondence with one another. Eventually, the technology will have to be developed that will enable a near-instantaneous measurement of ice nucleus concentration in a parcel of air (most likely from aircraft) and an observation of ice crystal formation in the same parcel of air immediately thereafter. Only when such one-to-one comparisons will be available ([79] is such an attempt) and will be understood,and when we learn more about the sources, transport and transformation of atmospheric ice nuclei will ice nucleus measurements become major inputs to cloud physics studies. Indeed, there is a very clear need to understand the influence of ice nuclei on precipitation and hail formation, cirrus clouds, ice fogs, precipitation efficiency, on the margin available for increasing precipitation efficiency by cloud seeding, and other key questions.

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