# Sources of origin and meteorological importance of hygroscopic and ice-forming nuclei[1]

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ABSTRACT

The hygroscopic nuclei whose source region during the monsoon is the sea penetrate far into the interior and play important role in development of precipitation well inland. The fraction of such nuclei present in the total aerosol constitutes a more dependable criterion for distinguishing maritime airmasses from those of others.

Development of rain frequently by all-water and ice-crystal mechanisms have been suggested by the large concentrations noticed of the hygroscopic and ice-forming aerosols respectively.

While it is seen that there is a major identifiable singular source, which is the sea, for hygroscopic aerosols at Delhi, it does not appear to be so in the case of ice-forming nuclei. The latter are of varied origin and might be maritime, continental, stratospheric etc.

## Introduction

A knowledge of aerosol state of the atmosphere helps furnish information on the possible origin of airmasses which influence the area and the mechanisms which contribute to rain development in clouds over the region. The size spectrum of aerosols which is of vital concern for investigation in this connection extends from the conventional large (radius 0.1 to 1 micron) to the giant size (radius more than 1 micron). Of special importance for development of rain by all-water mechanism in the tropics are the particles in the latter size range which are also hygroscopic by nature.

Observations made in ground air layers at Delhi on hygroscopic, non-hygroscopic and ice-forming nuclei are presented and discussed from the above points of view. Also, possible sources of origin of ice-forming aerosols at Delhi have been discussed.

## Measurements

The equipment used for the study of hygroscopic and nonhygroscopic aerosols is the Cascade Impactor (May 1945). Slide 2 in the Impactor which collects most of the aerosols present in the giant size range has been examined at 95 per cent humidity under high power microscope. The droplets formed around hygroscopic particles collected and solid insoluble particles which have not turned into liquid have been counted separately. Particles collected on slide 3 (this slide collects only a small fraction of total aerosol present in the large size range) have also been counted on some occasions. Ice-forming nuclei have been estimated during 1960–61 with 1 litre mixing-type cold box (PRABHAKAR & RAMANA MURTY, 1962) and subsequently by millipore technique (BIGG *et al.*, 1963). The concentrations of atmospheric chlorides and sulphates have been measured using millipore, the methods adopted for detecting these particles having been those due to GEORGII & METNIEKS (1958) and LODGE (1954) respectively.

The site of measurements is the second floor of the National Physical Laboratory at New Delhi (Lat. 28°35′ N, Long 77°12′ E, A.S.L. 714′). The place being far away from the main

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industrial areas of the city is expected to be reasonably free from pollutants in the nature of industrial smoke particles. The contaminating effects from foliage and vegetation may be considered to be relatively small on account of the open surroundings at the measuring site.

## Results and discussion

### (i) *Source of hygroscopic nuclei*

Some of the important features in respect of giant size hygroscopic aerosols at Delhi have been reported earlier (RAMANA MURTY *et al.*, 1962). The day-to-day concentration of these particles has been found to vary from almost nil to as much as about 100 per litre. The general level of the nuclei activity is highest during the monsoon season (Fig. 1). As the feed into the region of air during monsoon season is from the sea the observed influx of hygroscopic nuclei into the area during the monsoon is explained by the hypothesis that the sea is the main source of hygroscopic nuclei in the air. As the distance of nearest approach to the sea from Delhi is about 1000 km, the observations substantiate the finding by earlier workers (CROZIER *et al.*, 1952, and TWOMEY, 1955) that the aerosols produced over the oceans travel upwards and in the horizontal considerable distances inland.

### (ii) *Identification of airmasses*

The majority of hygroscopic aerosols during the monsoon are indicated to be of sea origin. These are produced over the oceans presumably by processes such as wave breaking and bubble bursting (BLANCHARD & WOODCOCK, 1957). As sodium chloride is considered to be the main constituent of these particles, an estimate of chloride content in the air should serve as a convenient indicator of influx of relatively fresh maritime airmasses. However, an examination of the data collected has indicated no such trend, specially during the transition period, summer to monsoon.

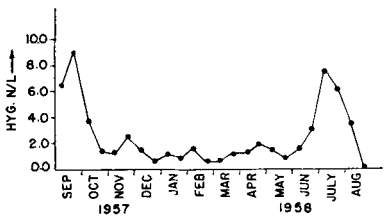


FIG. 1. Average half monthly concentration of giant hygroscopic nuclei (1957–58).

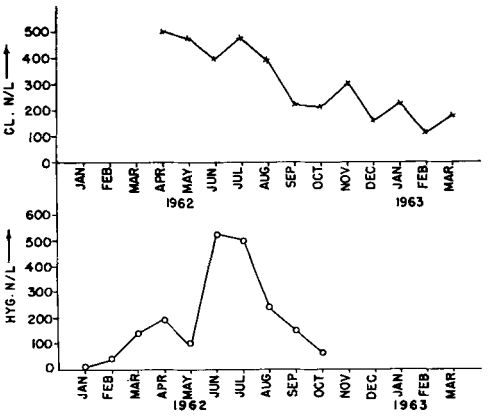


FIG. 2. Monthly mean concentration of hygroscopic (large size) and chloride aerosols (1962–63).

Measurements made on chloride particle concentrations (monthly means) with millipore filter have been shown plotted in Fig. 2, along with measurements on hygroscopic nuclei (slide 3 in the impactor). The chloride particle count has not shown any rise during the monsoon (July–September), but instead shown a decrease as compared to the value noticed during the relatively dry period, summer, which is April–June (average concentration during the monsoon and dry periods are 326/l and 478/l respectively). The values during winter, i.e. December–February, when airmasses over the region are typically continental in nature are found to be considerably low (average concentration: 184/l). The feature suggests that chloride particle concentration in the air can be used as a useful guide for identifying winter airmasses (continental). It cannot, however, distinguish maritime air of the south west monsoon from the hot relatively dry air of summer period, perhaps because the strong convectional currents due to insolation at that time (summer) make the place chloride-rich with particles of land and industrial origin. The findings suggest that the chloride particle concentration cannot be considered as a dependable indicator of the influx of maritime air into the region in areas well inland, such as Delhi. Investigations by METNIEKS (1958) in Dublin, RAU (1955) in

TABLE 1. *Mean concentration of sulphate and chloride aerosols. (Number per c.c.)*

| Period | $SO_4$ | Cl |
|---|---|---|
| Summer | 0.26 | 0.39 |
| Monsoon | 0.23 | 0.42 |
| Winter | 0.38 | 0.19 |

South West Germany and PODZIMEK (1959) and PODZIMEK & CERNOH (1961) in Czechoslovakia also point in this direction.

As sulphate aerosol is considered predominantly more continental in origin, simultaneous measurements made on sulphate and chloride aerosols have been examined with a view to finding out if a knowledge of $Cl/SO_4$ in the air might enable a criterion to be evolved for judging the airmass character during the three principal seasons at Delhi.

The ratio of Cl to $SO_4$ counts is about 2 during monsoon, 1.5 during summer and 0.5 during winter. The prevailing dry continental airmasses during winter which have their source region over Central Asia may be identified by their smaller $Cl/SO_4$ concentrations. However, the criterion does not help differentiate the strong maritime airflow during the monsoon from the dry air of land origin during summer.

The fraction of total aerosol constituting the hygroscopic component in different seasons has been examined, and it is found to provide a more definite clue to distinguish summer air from that of monsoon.

The very large fraction of the hygroscopic component in the air during monsoon characterizes the monsoon airflow from that of summer. The very low value obtained during winter denotes advection of continental airmasses into the region.

TABLE 2. *Mean concentrations (number per litre) of hygroscopic and total aerosols in the giant size range (slide 2 of Cascade Impactor); measurements: 1963–64.*

| Period | Total aerosol | Hygroscopic aerosol | Fraction constituting hygroscopic component X $10^3$ |
|---|---|---|---|
| Summer | 72.0 | 2.2 | 31 |
| Monsoon | 34.5 | 9.0 | 261 |
| Winter | 76.6 | 1.2 | 16 |

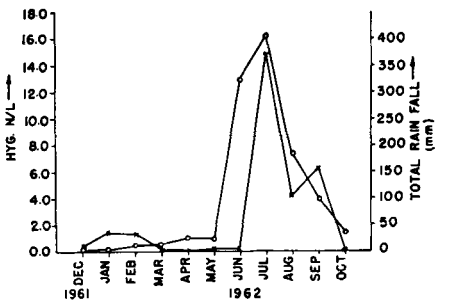


FIG. 3. Monthly mean concentration of giant hygroscopic aerosols (○) and total monthly rainfall (×) (measurements Dec. 1961 to Oct. 1962).

(iii) *Frequent development of rain by warm process and by Bergeron mechanism at Delhi*

Rain from clouds in tropical regions, as at Delhi, may not entirely be of 'warm' type. Bergeron mechanism also may play significant role in the release of precipitation. Simultaneous measurements on hygroscopic and ice-forming nuclei should help indicate how important the all-water and ice-crystal mechanisms could be for development of natural rain in clouds during different seasons.

Observations on giant size hygroscopic aerosols made during the period December, 1961 to October, 1962, have shown that the aerosol content is closely associated with total rainfall received (Fig. 3). The hygroscopic component recorded its maximum value during the monsoon (average concentration for the three months July to September: 9.2/l) which is the peak rainfall period of the year. During winter the concentration decreased conspicuously (average concentration during December, 1961, to February, 1962: 0.3/l). If the content of giant hygroscopic nuclei in the air is some indicator of prospects of rain formation by warm mechanism, observations suggest that the frequency of such rain could be very high during the monsoon, as compared to what it could be during winter. It would be of interest to note in this connection that the estimates based on radar observations (RAMANA MURTY *et al.*, 1960) have also suggested that quite a large proportion (about 30 to 40 per cent) of

precipitating clouds in Delhi area during monsoon could be of warm type, in contrast with rare occurrence of such rain cells during winter.

(iv) *Rain by Bergeron mechanism*

If rain initiation by Bergeron mechanism in super-cooled clouds is facilitated by ice-forming nuclei, the counts made in air of such nuclei could, perhaps, be treated as a useful index of prospects of rain development by that mechanism. Observations with 1-litre mixing type cold box have shown that the activity of ice-forming nuclei (number per litre at −20°C) at Delhi during the monsoon season is remarkably high (PRABHAKAR & RAMANA MURTY, 1962). Also, it is seen that the trend of variations of the total monthly rainfall is closely associated with the monthly mean concentration of the ice-forming aerosol (number per litre at −20°C) as shown in Fig. 4. The features suggest that Bergeron mechanism may play a significant role in the release of precipitation from clouds in the Delhi area. Observations by radar during the season which show frequent development of Bright Band (RAMANA MURTY *et al.*, 1960), characteristic of rain by Bergeron mechanism, lend support to this finding.

It is suggested, therefore, that both the known processes of rain development, namely, warm as well as cold, could become significant for rain formation in clouds in tropical areas as at Delhi. However, it may not be possible to estimate the relative contributions by the two mechanisms to season's total rainfall from measurements on aerosols alone.

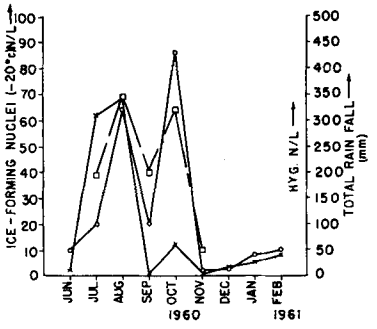


FIG. 4. Monthly mean concentration of ice-forming nuclei (○), large hygroscopic nuclei (□) and total monthly rainfall (×) (measurements 1960–61).

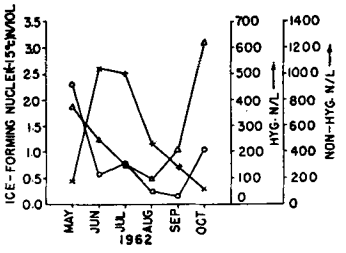


FIG. 5. Monthly mean concentration of ice-forming (○), large hygroscopic (×) and large non-hygroscopic aerosols (△), (measurements 1962).

(v) *Suggested sources of origin of ice-forming nuclei*

While it is agreed generally that the majority of hygroscopic aerosols owe their origin to the sea, we are not at all clear about the probable source of origin of ice-forming aerosols. Divergent views have been expressed in this regard by different investigators (KLINE, 1960; BATTAN & RILEY, 1960; ISONO *et al.*, 1959; GEORGII & METNIEKS, 1958). An attempt is made in the following to find out which of the possible sources of origin suggested would apply in the case of ice-forming aerosols at Delhi.

*Maritime source-measurements by mixing chamber.*—As already pointed out, the general level of activity of ice-forming nuclei at Delhi is markedly high during the monsoon season (July–September) when the feed of air into the region is from the sea. The mean temperature at which 10 ice crystals have been observed in one litre of air is noticed to be −19°C during this period as compared to −22°C and −24°C during summer (April–June) and winter (December–February) respectively. Also, the ice-nuclei concentrations have been found to be closely associated with hygroscopic nuclei content (Fig. 4). These features suggest marine origin of the nuclei.

*Continental source-measurements by millipore filter.*—The day-to-day concentration of ice-forming nuclei at −15°C during the period May to October, 1962 has been estimated by millipore technique, and the monthly mean concentrations have been shown plotted in Fig. 5. Also, the concentrations of the hygroscopic and non-hygroscopic aerosols in the large size range as obtained from measurements on slide 3 in the impactor have been shown in the figure. Observations indicate that the trend of varia-

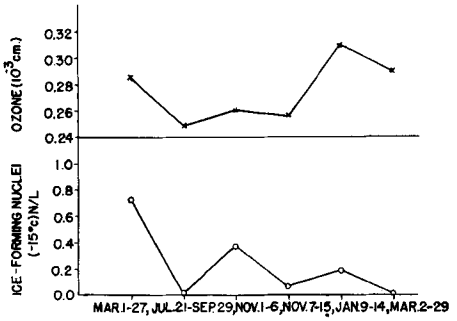


FIG. 6. Ice nuclei concentration and atmospheric ozone content (1962–63).

tion of ice-nuclei content follows more closely that of non-hygroscopic aerosols. The finding points out land origin of the ice-forming nuclei.

*Stratospheric source—atmospheric ozone and ice-forming nuclei.*—If atmospheric ozone, whose seat of origin is in the stratosphere, descends into the troposphere in "rivers" (KROENING & NEY, 1962), and if ice-nuclei in the same air would likewise descend, a certain relation between ice-nuclei content and total ozone amount is to be expected, if such nuclei are mainly of stratospheric origin, as envisaged by BIGG & MILES (1963). Estimates of vertical distribution of ozone by Umkehr data indicate relatively high ozone amounts in lower levels on occasions of high total ozone content, presumably due to air exchange mechanisms across the tropopause or through the tropopause gaps.

Measurements made on concentration of ice-forming aerosols at −15°C with millipore filter during 1962–63 have been examined vis-a-vis the values of total ozone content for Delhi as available from the published data by the India Meteorological Department. The periods chosen for the study are when alternately prominent peaks and troughs have been noticed in the value of the aerosol content. Also, the occasions were such that the maximum value recorded for the aerosol content during the trough period was smaller than the minimum value recorded during the peak period. The ozone amounts for the corresponding periods have been considered. Table 3 and Fig. 6 summarize the result. Peak and trough values both in ice-nuclei content and in ozone amount are seen to occur simultaneously. The feature lends some qualitative support to the finding by BIGG & MILES (1963) that ice-forming aerosols are largely of stratospheric origin.

It is felt that there is perhaps no one single source of origin for these aerosols with ice-forming property. Some of these may be of marine origin, some of land origin, and some of stratospheric origin too. In an earlier investigation (RAMANA MURTY & ROY, 1961), the possibility of ice-nuclei at Delhi being of meteoric origin (extra-terrestrial) was brought out. All these possibilities seem to be relevant. The important question, however, is whether nuclei from each one of these sources are equally effective, or there are significant differences in their activation temperatures. Our knowledge in this regard is still very scanty and will remain so until the difficulties involved in obtaining these aerosols for study in their pure and fresh

TABLE 3. *Ice-nuclei and atmospheric ozone.*

| Period | Ice-nuclei concentration at Delhi Number/litre | | Ozone content at Delhi ($10^{-3}$ cm) | |
|---|---|---|---|---|
| | Range of variation | Average value | Range of variation | Average value |
| 1962 | | | | |
| March 1–27 | 0.10 –2.1 | 0.73 | 0.271–0.309 | 0.286 |
| July 21 to Sept. 29 | 0.0 –0.07 | 0.02 | 0.231–0.287 | 0.249 |
| Nov. 1–6 | 0.26 –0.60 | 0.38 | 0.253–0.279 | 0.261 |
| Nov. 7–15 | 0.04 –0.09 | 0.07 | 0.239–0.271 | 0.256 |
| 1963 | | | | |
| Jan. 9–14 | 0.15 –0.26 | 0.20 | 0.297–0.320 | 0.310 |
| March 2–29 | 0.005–0.025 | 0.012 | 0.264–0.319 | 0.290 |

form are overcome. These particles undergo complex transformations under the influence of various atmospheric phenomena when once they move away from their source regions, and any subsequent observations made on them have to be interpreted with due caution. For example, SOULAGE (1961) is inclined to attribute the marked activity of ice-forming aerosols, observed by him on the south west coast of France, to the possible activation of continental nuclei during their passage over ocean, but not to sea spray.

It may be observed in this connection that measurements made relative to one temperature may not be true indicator of the entire aerosol spectrum. Aerosols at Delhi during monsoon may be classified under three categories, (1) non-hygroscopic (2) hygroscopic and (3) mixed (combination of both). The latter two categories have been grouped under hygroscopic aerosol when examined under microscope. If a certain proportion of non-hygroscopic nuclei be ice-forming in nature and be activated at a relatively warm temperature, say $-15°C$, the content of such ice-forming nuclei would be expected to decrease during monsoon season, when total concentration of non-hygroscopic aerosols in air is found to be so much smaller (Fig. 5). Again, if those amongst the nuclei which are treated as hygroscopic but really come under the category of 'mixed nuclei' be activated at lower temperatures, the concentration of such aerosols showing ice-forming property would increase with increase in concentration of the hygroscopic nuclei, as shown by measurements with mixing type cold box at $-20°C$. Further, if some amongst the soluble particles in maritime air serve as ice-forming nuclei by a process suggested by POWER & POWER (1962), it is considered that these would be activated at relatively cold temperatures. The content of such ice-forming aerosols would increase with progressive incursion into the region of moist airmass of marine origin.

Ice-forming aerosols indicated to be of stratospheric origin may have actually been produced in the stratosphere or may have originated as terrestrial dusts or may be of extra-terrestrial origin, as pointed out by BIGG & MILES (1963). However, MOSSOP (1963) is inclined to consider that these particles are of extra-terrestrial origin as they might be meteor ablation products or broken cometary aggregates of fine dust.

## Conclusion

The features brought out by the present study have been based on measurements made at ground level only. The important point to be considered is how representative are such measurements of the actual conditions aloft. Aerial observations at Delhi recently commenced and in progress have, on many occasions, indicated close association in the trend of variation of aerosol concentrations as measured in surface air layers and those observed at higher levels. The feature would need to be examined further and in greater detail by analysis of data collected during different periods of the year before we could come to a reasonably firm conclusion.

In course of study on ice-forming nuclei, measurements made both by mixing type cold box (used in the earlier stages) and by millipore filter (used later) have been considered. Both the methods of measurements have their own merits and demerits and the values obtained by them are not comparable (BIGG *et al.*, 1963). One of the factors considered responsible for the observed underestimation of ice-nuclei content by the millipore technique is the deactivation effect due to hygroscopic nuclei present in the air (MOSSOP, 1965). It is not known whether the general level of activity of the hygroscopic aerosols as at Delhi during the monsoon is high enough to be considered important for this purpose. The conclusions which have been based on measurements by millipore filter, therefore, are to be considered only as tentative until this point has been further examined.

## Acknowledgement

The millipore filters exposed at Delhi are being processed in the Radio Physics Laboratory, C.S.I.R.O., Sydney. The authors wish to record their deep sense of indebtedness to Dr. S. C. Mossop and to his colleagues in the laboratory for their generous help in this regard.

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## ИСТОЧНИКИ ОБРАЗОВАНИЯ И МЕТЕРЕОЛОГИЧЕСКОЕ ЗНАЧЕНИЕ ГИДРОСКОПИЧЕСКИХ И ФОРМИРУЮЩИХ ЛЕД ЯДЕР


Гидроскопические ядра, образующиеся над морем во время муссонов, проникают далеко вглубь континента и играют важную роль в выпадении осадков над материком. Доля таких ядер от полного количества присутствующих в воздухе аэрозолей является надежным критерием, позволяющим различать одну воздушную массу от другой в зависимости от времени нахождения их над морем.

Развитие дождя в виде капель или кристаликов льда часто сопровождалось наблюдением большой концентрации соответственно гигроскопических или формирующих лед ядер.

В то время как очевидно, что единственным источником гигроскопических аэрозолей в Delhi является море, формирующие лед ядра имеют различное происхождение: морское, континентальное, стратосферное и т. д.