# A Change in the Size-Distribution of Dust falling in the Polar regions over the Past fourteen centuries

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**Abstract**

The size-distribution of particulate fallout in the 0.6 to 3 microns diameter size range has been measured in samples of ice and snow from Greenland and Antarctica. The ages of the samples range from about 2 to almost 1400 years. The shape of the average size distribution curve exhibits a pronounced change with time. In older samples, the log-log cumulative frequency distribution is usually convex upward, but there is a progressive tendency toward straight or convex downward distributions in younger samples. Most of the change seems to be due to the particle population smaller than 1.5 microns diameter.

The change is believed to be due to the recently increasing abundance of a well-sorted particle population with a modal diameter of 0.6 micron or less. Microscope investigations of dust in recent samples give conflicting results with some indication that the new population may be associated with an increased abundance of black spherules, clusters of black angular particles, or angular particles with metallic luster.

It is also possible that the smaller size fraction slowly dissolves in ice and is consequently less abundant in older samples.

The evidence does not rule out the third possibility that the greater relative abundance of the sub-micron particles is related to increased man-caused air pollution.

## Background

The size-distribution of dust particles between 0.6 and 3 microns diameter has been measured in hundreds of samples of polar ice from different sites on, and different depths within, the Greenland and Antarctic ice sheets. These measurements have been made with the Coulter Counter, an electrical sensing zone size-analyzer, on melted specimens of polar ice and snow. The results upon which this report is based have been given by Bader, Hamilton and Brown (1965), Hamilton (1966), Hamilton and O'Kelley (submitted for publication), and Hamilton and Langway (submitted for publication). The first article contains a review of the techniques of particle counting. Other size-distribution analyses have been made by Taylor and Gliozzi (1964) and Gliozzi (1966).

During these analyses, various size-distributions of particles have been detected. Early work, done on old ice from deep beneath Byrd and Little America Stations, Antarctica, yielded distribution curves which were mainly like those illustrated in Figure 1a. Most older samples gave log-log cumulative frequency vs. size-distribution plots which increase in slope toward the larger sizes. Sometimes this increase is gradual; sometimes it takes place abruptly, forming an elbow joining two essentially straight lines.

Hamilton and Bull (1966) presented arguments indicating that the elbow may result from an essentially monosize population of particles with diameter of about 1.5 microns, believed to be spherules derived from ablation of meteoric dust.

### *Change in the size-distribution*

As more samples of recent polar snow and ice have been analyzed a new shape of distribution curve has become more common. The slope of the log-log cumulative frequency vs. size-distribution *decreases* toward larger sizes in many of these new distributions (Figure 1b).

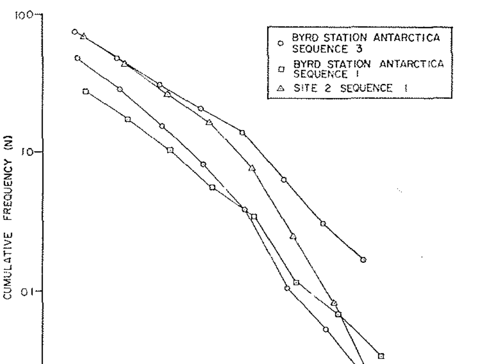


Figure 1a

Representative cumulative frequency curves for which slope increased toward larger sizes. The Site 2 counts have been multiplied by 0.1 to fit them on the graph. N is the number of particles larger than stated size V per microliter of sample.

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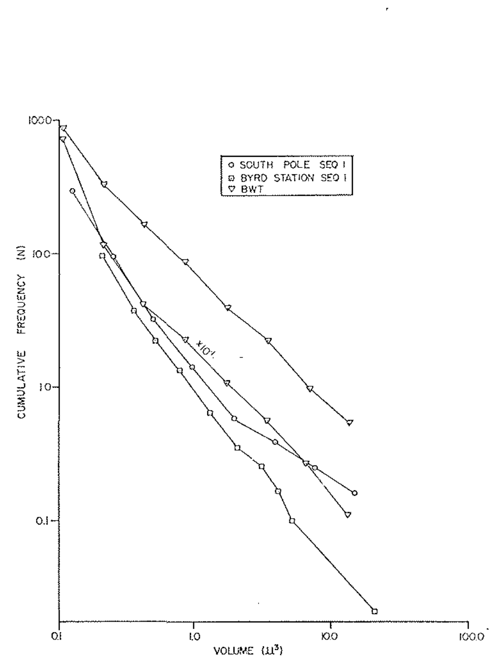



Figure 1b

Representative cumulative frequency curves for which slope decreases toward larger sizes. The indicated Byrd-Whitmore Traverse (BWT) curve has been multiplied by 0.1 to fit it on the graph. N is the number of particles larger than stated size V per micromiter of sample.

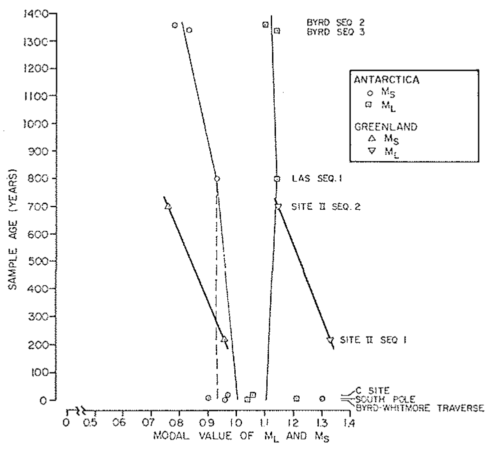



Figure 2

Sample age vs. proportion (%) of samples with $m_l$ less than $m_s$.

The change of the distribution curve shape with time is illustrated in Figure 2. The symbol m denotes slope, given by

$$-m = \frac{d(\log N)}{d(\log V)}$$

where N is the number of particles per microliter of sample which are larger than volume V. The subscript s refers to the small-size portion of the cumulative curve, between 0.6 and about 1.5 microns diameter, and the subscript l refers to the large-size portion, between 1.5 and 3 microns diameter. The ration $m_l/m_s$ is useful in comparing samples in order to observe changes in the convexity of the elbow.

In Figure 2, the increase with time in the abundance (percent) of samples with $m_l/m_s$ (distribution curves which are convex downward) is evident for the Antarctic sequences. The limited results from Greenland are consistant with the Antarctic results, but not independently convincing. Curves have been hand-fitted through the points.

*Increasing abundance of sub-micron particles*

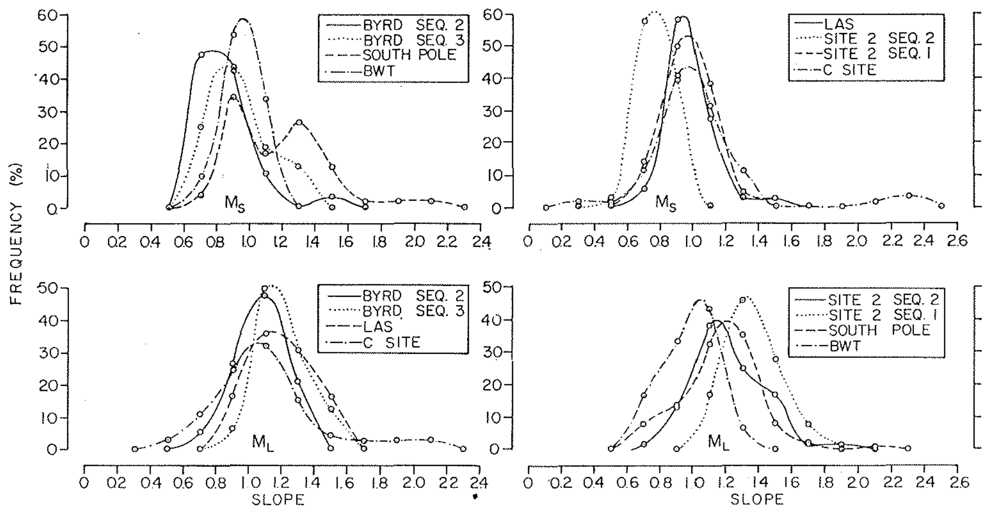
To determine whether the change in distribution was due mainly to changes in the contribution of smaller sizes (less than 1.5 micron) or of larger sizes (greater than 1.5 micron), the variation in time of $m_l$ and $m_s$ was examined.

Averages of $m_l$ and $m_s$ in Figure 3 are given in the form of modal values taken from the frequency distributions of $m_l$ and $m_s$ given in Figure 4. Two values of $m_s$ are given for the South Pole samples because the frequency distribution of $m_s$ was bimodal. In Figure 3 the dashed portion of the $m_s$ growth curve goes through a point calculated without including the South Pole $m_s$ mode at 1.3. The solid curve goes through a point calculated by including that value. The reasons for regional variation are not yet understood.

The number of samples in each sequence is given in Table I together with the age of the samples. Sequences are listed in the order in which they were analyzed.

**Table I**

| Sequence Sample | Number of sample. | Approximate sample age (years) |
|---|---|---|
| Byrd I | 68 | 465 |
| Byrd II | 19 | 1360 |
| Little America | 38 | 800 |
| Byrd III | 16 | 1340 |
| Byrd IV | 78 | 450 |
| Site 2, Seq. I | 89 | 220 |
| C Site | 114 | 18 |
| South Pole | 98 | 7 |
| Site 2, Seq. II | 62 | 700 |
| Byrd- Whitmore | 30 | 2 |



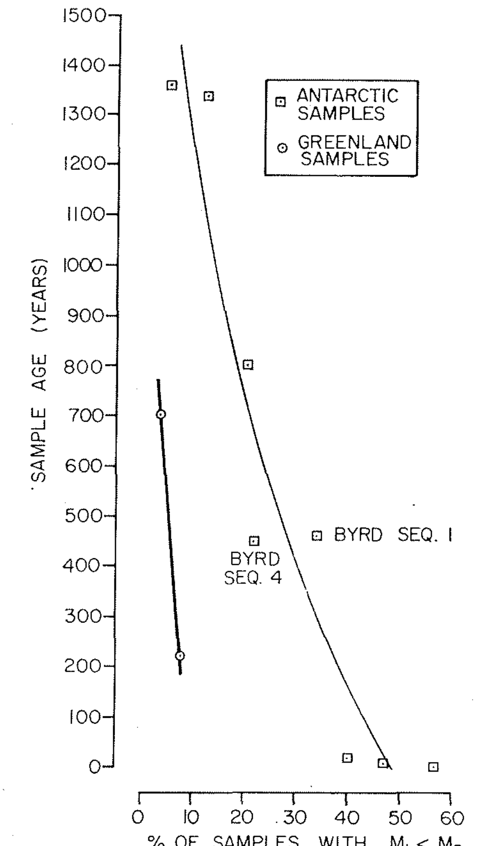




Figure 4
Frequency distributions of $m_s$ and $m_l$. In each drawing the order of age of sample sequences, from oldest to youngest, is given by the sequence: solid, dotted, dashed and dot-dashed lines. BWT and LAS refer to samples from the Byrd-Whitmore Traverse and Little America Station.

Figure 3
Sample age vs. modal value of $m_l$ and $m_s$ taken from the curves in Figure 3. Lines pass through points representing arithmetic mean value for samples of comparable age. Dashed line goes through arithmetic mean value calculated without including South Pole $m_s$ mode at 1.3.

The values $m_s$ and $m_l$ have been calculated by least squares regression in the case of the 700-year-old Greenland ice and the C Site, South Pole and Byrd-Whitmore Traverse samples. Values for the other sequences were calculated by taking the arithmetic means of the slopes between data points on the portions of the curves under consideration. This shortout introduces a maximum error of about 2 % when data points are most widely scattered. However, the arithmetic mean slope is usually greater than the slope of the regression line; less than 1 % greater in the case of distributions with $m_s$ less than $m_l$ and about 1.5 % in the case of distributions with $m_s$ frequently larger than $m_l$. Therefore the error, small as it es, is in the wrong direction to account for the smaller values of $m_s$ in older samples from Antarctica. The error could account for a slight amount of the decrease in $m_s$ in the older Greenland samples, but it is by no means as significant as possible random sampling errors.

Figure 3 shows that most of the change in Antarctica can be attributed to an increase in slope of the small-size end of the distribution. This is the effect which would result from the addition of a new population of particles with a modal size somewhat smaller than 0.6 micron diameter, just out of the range of our Coulter Counter.

The Greenland samples show an increase in the slope of both the large- and small-size ends of the distribution.

*Comparison of distributions with microscope counts*

Because microscope examination of older particulate material is lacking, it is not yet possible to directly attribute the change in the distribution to any particular types of particles. Also, it might be impossible to detect the new population because it is composed of particles smaller than the resolution limit of the light microscope.

In spite of these difficulties, microscope counts of particles from the 1965 annual layer from six sites along the Byrd-Whitmore Traverse (1965—66) were divided into counts of samples which had $m_l$ greater than $m_s$ and those which had $m_l$ less than $m_s$, as determined by Coulter Counter. These results will be discussed in greater detail elsewhere, but the result which may apply here relates to the types of particles which were relatively more common in the samples having $m_s$ greater than $m_l$. Table II lists the particle types counted.

Of 30 samples, 5 from each site along the traverse, 17 had $m_s$ greater than $m_l$. These 17 samples contained an average of 1.2% black spherules (Class II), 50 % more than in the other 13 samples. There were also 11.8 % angular particles with metallic luster (Class VC) and 2.9 % clusters of black, angular particles (Class VA); respectively 50 % and 45 % more than in the other samples.

**Table II**

| Class | Description |
|---|---|
| I | Opaque spherules with metallic luster |
| II | Opaque spherules with non-metallic lustor (black) |
| III | Transparent and translucent, colorless or amber spherules and discs |
| IVA | Transparent, colorless, angular particles with vitreous luster |
| IVB1 | Translucent, colorless or amber angular particles with dull luster |
| IVB2 | Translucent, colored angular particles (usually appears to be hematite or limonite) |
| IVC | Transparent and translucent, colorless, gelatinous blebs with inclusions and attachments |
| VA | Clusters of opaque, angular particles (black and brown) |
| VB | Single opaque, angular particles (black and brown) |
| VC | Opaque, angular particles with metallic luster |
| VI | Clusters of opaque spherules in a matrix which is usually opaque, but may be transparent or translucent, colorless |

Values of $m_l$ and $m_s$ from the South Pole sample sequence were compared with microscope spherule counts made by Taylor (submitted for publication). In this case samples with $m_l$ less than $m_s$ contained 5 % fewer Class II spherules than the other samples.

These results, if applicable to the problem of identifying the recently increasing sub-micron particle population, are inconclusive. The Byrd-Whitmore Traverse results suggest that the increasing population may be comprised of, or associated with, black spherules, clusters of black angular particles, or angular metallic particles. The South Pole results do not support the idea of increased relative abundance of black spherules. Langway (1966) reported a 3- to 4-fold increase in the rate of black spherule accretion in Greenland over the past 700 years. While this adds some support to the tentative conclusion based on the Byrd-Whitmore Traverse results, it is still too early to say what kinds of particles comprise the recently increasing sub-micron population.

*Solubility of sub-micron fraction*

The possibility definitely exists that the smaller size-fraction may be soluble in ice. If this were true, older samples would contain a smaller relative abundance of sub-micron particles, and samples of equal age might contain more or less sub-micron particles depending on the thermal history of the ice.

This argument is favored by the fact that older samples from both ice sheets contain relatively fewer sub-micron particles, and also by the fact that the two sequences from Greenland, warmer in general than Antarctica, contain relatively fewer small particles than Antarctic samples of comparable age (Figure 2). However, the relative abundance of small particles appears to be changing less rapidly in Greenland than in Antarctica as indicated by the slopes of the lines in Figure 2. This is because the value of $m_l$ has increased almost as much as the value of $m_s$ in Greenland between about 1270 and 1750 A.D. (Figure 3). It seems unlikely that the distribution of large sizes could be changed as much as that of the small sizes by dissolution, and a change of the size-distribution of particles falling out of the atmosphere in this 480-year period is thought to be a more likely possibility.

*Increasing man-caused pollution*

There is also the possibility that the increased contribution of sub-micron particles might be due to increased man-caused pollution such as smoke from combustion processes. This possibility was tested by examining in detail the data from South Pole Station. The sample sequence there spans the interval 1956 to 1965. No evidence of increased incidence of large values of $m_s$ was noted for samples from snow which fell after the station was occupied in November, 1956. This does not rule out the possibility, however, because prevailing winds usually carry diesel smoke from the station generators away from the site where the samples were taken. Another argument against pollution is that the sub-micron population is more common in the Antarctic samples (Figure 2) than in those from Greenland, whereas one would expect Greenland to receive relatively more industrial contamination than Antarctica.

However, major industrialization took place more recently than 1750, the time from which the youngest Greenland samples date. Therefore, the possibility of smoke contamination cannot be ruled out.

*Sources of data*

The values of slopes have been calculated from the following data: Byrd Station, Antarctica, Sequence 2 (Bader, Hamilton and Brown, 1965); Byrd Station, Sequence 3, Little America Station, Antarctica, Sequence 1, and Site 2, Greenland, Sequence 1 (Hamilton, 1966); C Site, Antarctic Peninsula and South Pole Station, Sequence 1 (Hamilton and O'Kelley, submitted for publication); Site 2, Sequence 2 (Hamilton and Langway, submitted for publication); and Byrd-Whitmore Traverse (Hamilton, in preparation). Data for Byrd Sequences 1 and 4, described respectively in Bader, Hamilton and Brown (1965) and Hamilton (1966), were temporarily unavailable in digital form. Therefore, size distribution plots were used to calculate the points shown in Figure 2.

*Summary*

The modal average of the slope ($m_s$) of the log-log cumulative frequency vs. size-distribution between 0.6 and 1.5 microns diameter in samples from the Antarctic ice sheet has increased from about 0.82 to about 1 in the past 14 centuries. In Greenland the value increased from 0.76 to 0.96 between about 1270 and 1750 A.D.

There was a corresponding increase in the slope ($m_l$) of the distribution of particles between 1.5 and 3 microns diameter in the Greenland samples only. The distribution of these larger particles has remained essentially constant in Antarctica.

The increase in $m_s$ is believed due to the increased relative abundance of a sub-micron population of particles. These particles may be either natural fallout or man-caused contamination. The evidence is less strongly in favor of the latter possibility.

It is also possible that the sub-micron population has been depleted in older samples by dissolution in the ice, but this seems less likely in view of the change in $m_l$ accompanying the change in $m_s$ in Greenland ice.

The proportion of samples with distributions having $m_l$ less than $m_s$ has increased from about 8 % to almost 50 % in Antarctica in the past 14 centuries. Between 1270 and 1750 A.D. the proportion increased from 3.5 % to 7.5 % in Greenland.

*Acknowledgements*

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## Prof. Dr. Walter Wundt †

Nach einem reich erfüllten Leben, das einen gnadenvollen, leichten Abschluß ohne vorherige Krankheit fand, verschied unerwartet am 24. August 1967 in Genf, noch erfüllt von den Eindrücken einer Reise in die Gletscherwelt der Schweizer Alpen, unser langjähriges Mitglied, Prof. Dr. Walter Wundt, Freiburg i. Br. Er nahm als Vortragender an allen Polartagungen unserer Gesellschaft teil und war Mitarbeiter an unserer Zeitschrift „Polarforschung".

Walter Wundt wurde am 6. Mai 1883 in Schorndorf/Württ. als Sohn des Oberbaurates Georg Wundt geboren. Nach dem Besuch des humanistischen Gymnasiums widmete er sich dem Studium der Mathematik, Physik und Naturwissenschaften in Stuttgart, Göttingen, Berlin und Tübingen.

Im Jahre 1904 promovierte er in Berlin zum Dr. phil. und war dort Assistent am Preußischen Meteorologischen Institut. Nach seiner Prüfung für das höhere Lehramt war er an mehreren Schulen in Württemberg tätig, zuletzt als Direktor der Oberrealschule in Schwenningen. Im Jahre 1934 wurde er wegen Krankheit in den Ruhestand versetzt und siedelte nach Freiburg i. Br. über. Dort erhielt er auf Grund seiner wissenschaftlichen Arbeiten bis 1958 Lehraufträge an der Universität. Im Jahre 1942 wurde er zum Mitglied der Akademie der Naturforscher in Halle und 1947 zum Honorarprofessor an der Universität Freiburg ernannt. Im Jahre 1961 wurde ihm das Bundesverdienstkreuz 1. Klasse verliehen.

Seine wissenschaftliche Tätigkeit erstreckte sich zunächst auf das Gebiet der Meteorologie. Im Jahre 1910 entwickelte er eine allgemeine Methode zur kartographischen Darstellung von Abflußzuständen und stellte die Bedeutung der Trockenwetter-Abflußkurve heraus. Im Jahre 1938 gibt er ein allgemeines Bild des Wasserkreislaufes und im Jahre 1950 wendet er sich der Untersuchung des Grundwassers zu und gibt Karten für die Tiefe des Grundwasser-Spiegels heraus.

Sein im Jahre 1953 erschienenes Werk „Gewässerkunde" bringt eine zusammenfassende Darstellung der festländischen Hydrographie. In anderen Arbeiten beschäftigt sich Wundt mit der Chronologie der Erdgeschichte, speziell mit der Eiszeit und Nacheiszeit. Seine letzte Veröffentlichung zur Eiszeit (1960/61) behandelt das Zusammenwirken der beiden Erdhalbkugeln bei der Ausbildung der allgemeinen Zirkulation. Ein weiteres Arbeitsgebiet waren rhythmische Erscheinungen in der Erdgeschichte, für die physikalische Begründungen gegeben werden, und erstmalig wurde die Kühlboden-Hypothese eingeführt, nach der die Ozeanböden eine dauernde Tendenz zur Vertiefung zeigen, die ihrerseits wieder an den Rändern Erdbeben, Eruptionen und Hebungen erzeugen und zusammen mit der Abtragung des Festlandes einen Materialkreislauf zur Folge haben.

Prof. Dr. Walter Wundt wird durch seine Arbeiten und sein Wirken stets Vorbild und unvergessen bleiben. Ruthe