# Electron-Microscope Study of Cloud and Fog Nuclei

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*Abstract*

Droplets of clouds on a mountain and of fog in an urban area were captured and the form, nature and size of their nuclei were studied by means of an electron-microscope and by a chamber of constant humidity. These nuclei have similar form and nature to the hygroscopic particles in haze and to the artificially produced combustion particles. No sea-salt nuclei were found in our observations, therefore sea-spray appears to be an insignificant source of condensation nuclei. It was found that both the cloud and the fog nuclei originated in combustion products which were the mixture of hygroscopic and non-hygroscopic substances, and that the greater part of the nuclei did not contain pure sulfuric acid.

## I. Introduction

The nature and origin of cloud and fog nuclei have been studied by many investigators and SIMPSON (1941) concluded that "sea-salt, sulfuric acid and nitrous acid drops were important nuclei in the atmosphere". However, these nuclei are so small that their size and nature cannot be studied by an optical microscope. Recently KUROIWA (1951) observed the sea fog nuclei by an electron microscope and found that many of them were solid particles insoluble in water and that only a small fraction of them were sea-salt particles. No sea-salt nuclei were found on Mt. Niseko (1,300 m) by the same author (KUROIWA and TADANO 1951). However, he did not refer to the origin of the nuclei.

From May to July, 1951 we observed the cloud nuclei on Mt. Zaō and the fog nuclei in Sendai (fig. 1). Observations and experiments were made mainly by an electron microscope and by a chamber of constant humidity. To study the nature and origin of nuclei we compared them with haze particles, sea-salt particles and artificially produced combustion particles.

## II. Observation of cloud and fog nuclei

### *1. Capture of cloud and fog droplets*

Cloud and droplets were captured on a thin collodion film coated on the sample holder (Träger) of the electron microscope. As the droplets thus captured on the Träger evaporated quite rapidly the following technique was employed:

A small metal case was placed on the stage of an optical microscope as shown in fig. 2 (a) and the Träger was put into it. Drawing the air by a syringe which was connected to the case, the droplets in the air were captured upon the collodion film and their locations on the film were easily detected by the microscope.

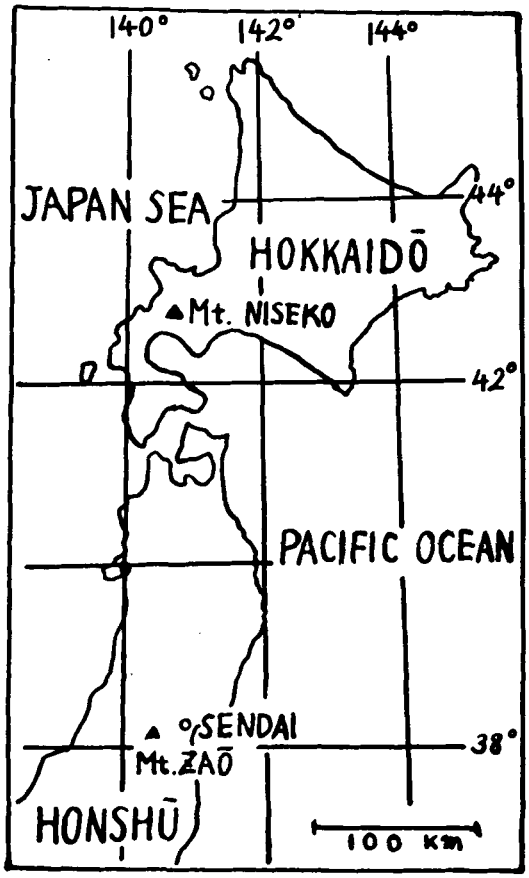



Fig. 1.

The total number of droplets captured were 60 on Mt. Zaō and 15 in Sendai, respectively. The time and station of observation and the weather conditions are shown in Table 1.

*2. Nature of nuclei.*

The size of the nuclei which remained on the collodion film after the cloud and fog droplets evaporated was measured by the optical microscope. The humidity in the room was 60 to 70 % and the magnification of the microscope was 900. Some of the nuclei were too small to be detected by the optical microscope.

Then the nuclei were put into a small chamber of constant humidity shown in fig. 2 (b). The chamber was a tapered glass vessel whose diameter was about 4 cm, and on its bottom a sheet of filter paper soaked with distilled water was placed. In the chamber almost all nuclei increased their diameter by absorbing the water vapour and the increment continued for 3 or 4 minutes. The final diameter was about 2 to 3 times as large as the initial one. On the small nuclei, which could not be detected at the room humidity, water droplets were also formed. Two nuclei, however, retained their original size in the chamber.

The temperature of the filter paper was equal to room temperature and the experiment was made in an underground dark room to avoid temperature fluctuations. Therefore the humidity in the chamber will be considered as just 100 %. As the nuclei which grew in the chamber are able to form cloud and fog droplets in the atmosphere with a humidity

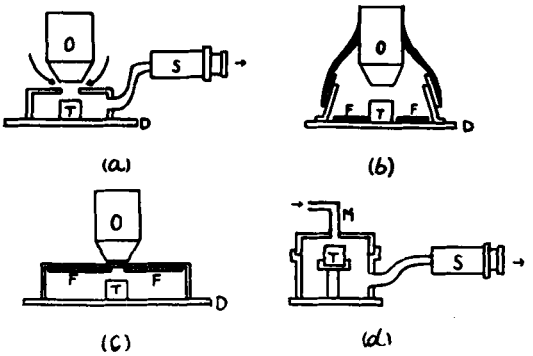



Fig. 2.

**Table 1. Observation of cloud and fog.**

| Date | Time h | m | Station of Observation | $T_{dry}$ (°C) | $T_{wet}$ (°C) | R. H. (%) | Visibility (m) | Wind Direction | Cloud Form |
|---|---|---|---|---|---|---|---|---|---|
| 11 May | 03 | 30 | Tōhoku University..... | 8.8 | 8.8 | 100 | — | — | Fog |
| 5 June | 07 | 00 | » » ..... | 17.4 | 17.2 | 98 | 250 | — | » |
| 15 » | 04 | 00 | » » ..... | 17.5 | 17.3 | 98 | 250 | — | » |
| 17 » | 05 | 00 | » » ..... | 15.4 | 15.4 | 100 | 250 | — | » |
| 19 July | 05 | 50 | Mt. Zaō (Hütte 1,400 m) . | 13.0 | 13.0 | 100 | 200 | WSW | Cu |
| 23 » | 08 | 00 | Mt. Zaō (Mt. Katta 1,750 m | — | — | — | — | » | » |

of 100 % or below we shall refer to them as "hygroscopic nuclei". The two nuclei referred to above are incapable of acting as condensation nuclei except in the supersaturated atmosphere, shall be referred to as "non-hygroscopic nuclei". The above results show that cloud and fog nuclei are formed in the atmosphere with a humidity of 100 % or below. There was one cloud sample in which no nuclei were found either by the optical or by electron microscope.

After the above experiment was finished, the form and size of the nuclei were observed by the electron microscope. Miscellaneous forms of the nuclei were observed and typical forms of them are shown in the photographs on page 236 and 237. Those from (1) to (13) are hygroscopic nuclei and (14) is a non-hygroscopic one.

(1) . . . Nearly circular thin filmy substance, in about half of this type of nuclei 1 to 10 small dark spots are also seen. These nuclei range in diameter from 0.3 to 1 $\mu$.
(3) and (6) . . . A large well defined dark particle and thin filmy substance around it. The former is of polygonal, circular or ragged form and its diameter is 0.3 to 1.2 $\mu$.
(8) . . . Special form as seen in the photograph.
(9) . . . Spotted thin substances, in the centre of which a dark particle is seen.
(10) and (12) . . . Resemble (3) etc. . . As will be described later, however, they do not readily lose their hygroscopic nature by electron-microphotographing.
(14) . . . Non-hygroscopic nucleus. Presumably it is a clump of carbon particles of oil as shown in (15). It should be noted that most nuclei belong to the former two types.

After the electron microphotographs of the specimens were taken, the hygroscopic nuclei were again put into the chamber of constant humidity. Then it was found unexpectedly that the moisture in the chamber did not condense on the greater part of the nuclei, that is to say, these nuclei had then lost their hygroscopic nature. It is supposed that by electron bombardment the hygroscopic substances contained in the nuclei were evaporated during photographing. Examples of these nuclei are shown in photos (1), (3), (6), (8) and (9). On the other hand three nuclei did not readily lose their hygroscopic nature, two of which are shown in (10) and (12). It should be noted that whether the nuclei lost their hygroscopic nature or not did not primarily depend upon their size, since, even some small nuclei did not necessarily lose their hygroscopic nature, while some nuclei with large diameters lost it by photographing. These results may be explained by assuming that generally several kinds of hygroscopic substances are contained in the nuclei and the rate of evaporation of these substances by electron bombardment differs from one kind to another.

To see whether the residues of the nuclei which lost their hygroscopic nature were soluble in water the following experiment was made. The residues were placed in a sealed glass vessel lined with moistened filter-paper as shown in fig. 2 (c). When the temperature of the filter paper was higher than the room temperature the moisture in the vessel condensed on the residues and water droplets were seen by the optical microscope. After the residues were wetted with water like this and then dried, their photographs were again taken by the electronmicroscope.

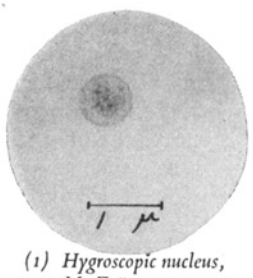
Comparing the image of the nuclei before and after wetting, it was found that most nuclei retained their original form as shown in (1) and (2), (3) and (4), (6) and (7). In three nuclei which retained their hygroscopic nature, however, the filmy substance changed form considerably, as shown in (10) and (11), (12) and (13).

From the experimental study described above it may be concluded that cloud and fog nuclei are composed of filmy substances and non-hygroscopic solid particles. A part of the substances with thin filmy appearance are also non-hygroscopic and insoluble in water, while others are hygroscopic and soluble in water.

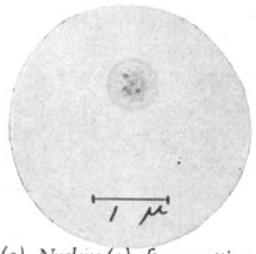
### III. Observation of haze particles

#### 1. *Capture of particles*

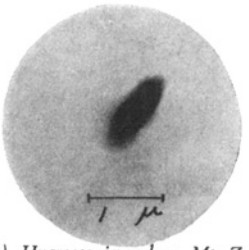
For comparison with cloud and fog nuclei, haze particles were captured in Sendai. Near the ground there were large particles exceeding several microns in diameter. However, most particles captured on the roof, about 10 m above the ground, were comparable in



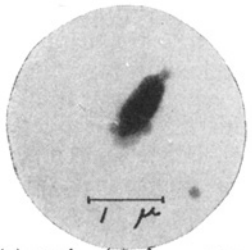

(1) Hygroscopic nucleus, Mt. Zaö.




(2) Nucleus (1) after rewetting.

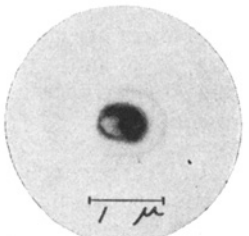



(3) Hygroscopic nucleus, Mt. Zaö.

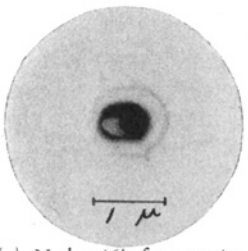



(4) Nucleus (3) after rewetting.

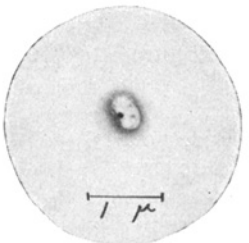

(5) Nucleus (4) after reaction with HF.



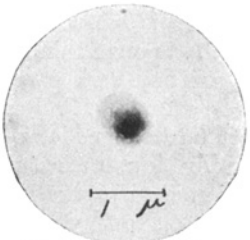

(6) Hygroscopic nucleus, Sendai.



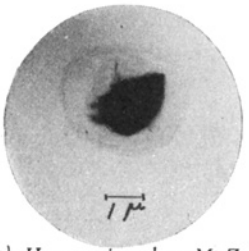

(7) Nucleus (6) after rewetting.



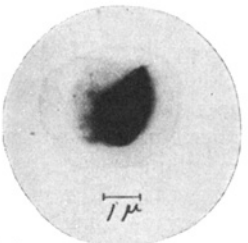

(8) Hygroscopic nucleus, Mt. Zaö.



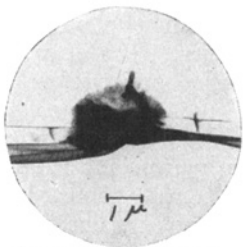

(9) Hygroscopic nucleus, Sendai



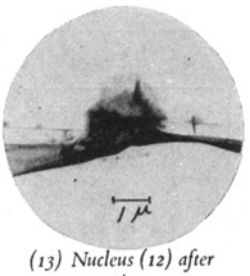

(10) Hygroscopic nucleus, Mt. Zaö.



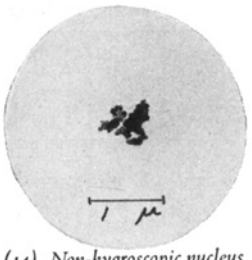

(11) Nucleus (10) after rewetting



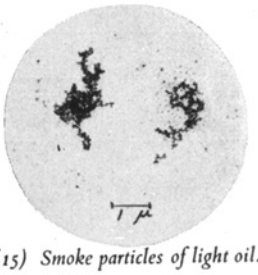

(12) Hygroscopic nucleus, Mt. Zaö.



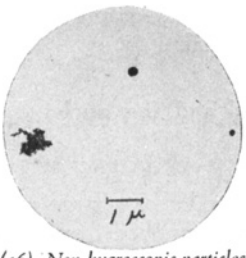

(13) Nucleus (12) after rewetting



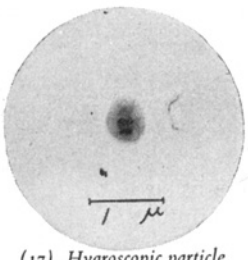

(14) Non-hygroscopic nucleus, Sendai.



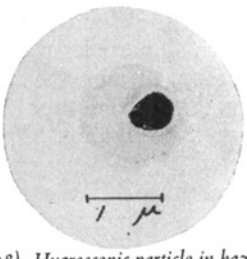

(15) Smoke particles of light oil.



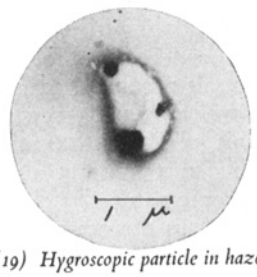

(16) Non-hygroscopic particles in haze.



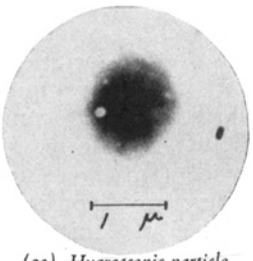

(17) Hygroscopic particle in haze.



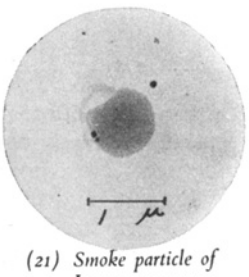

(18) Hygroscopic particle in haze



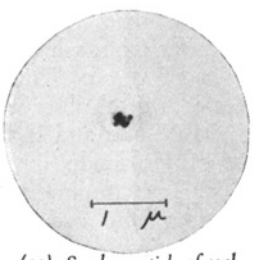

(19) Hygroscopic particle in haze.



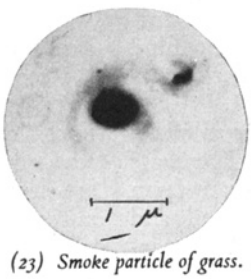

(20) Hygroscopic particle in haze



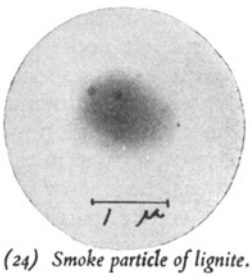

(21) Smoke particle of Japanese cypress.




(22) Smoke particle of coal




(23) Smoke particle of grass.




(24) Smoke particle of lignite.

**Table 2. Observation of haze (1951).**

| Date | Time | Number of Particles | | $T_{dry}$ | $T_{wet}$ | R. H. | Visibility | Wind | Cloud | |
|---|---|---|---|---|---|---|---|---|---|---|
| | h m | Hygroscopic | Non-hygroscopic | (°C) | (°C) | (%) | (m) | | Amount | Form |
| 19 June | 05 30 | 14 | 13 | — | — | — | — | — | — | — |
| 19 » | 06 00 | 11 | 40 | 18.0 | 14.3 | 67 | — | Calm | 9 | as, ci, cc |
| 20 » | 06 30 | 3 | 14 | 19.8 | 16.2 | 69 | — | » | 8 | ac, as, sc, ci |
| 23 » | 05 40 | 9 | 16 | 17.5 | 14.4 | 72 | 2,500 | » | 1 | cs, ci, cu |
| 24 » | 05 40 | 14 | 18 | 15.5 | 14.0 | 85 | 3,500 | — | 0 | — |
| 25 » | 06 00 | 35 | 35 | 17.4 | 15.3 | 80 | 2,000 | NW | 5 | cs, ci |
| 4 Sep. | 06 40 | 41 | 154 | 19.3 | 16.9 | 79 | 4,000 | WNW | 8 | ac, as, cu |
| 9 » | 07 00 | 10 | 37 | 20.9 | 18.7 | 81 | 7,000 | — | 10 | sc |
| 23 Oct. | 06 30 | 1 | 47 | 6.3 | 5.2 | 84 | 1,200 | NNW | 9 | cs, ac, sc |

size with the cloud and fog nuclei. To capture these small particles we used a collector shown in fig. 2 (d), in which the particles passed through a metal tube *M* at high speed and struck the collodion film, to which they then adhered.

The time and the weather conditions are shown in Table 2 together with the number of haze particles captured by the collector. It will be seen from the table that the haze is composed of hygroscopic and non-hygroscopic particles in various proportions. Experimental procedure to decide whether the particles were hygroscopic was quite the same as that for cloud and fog nuclei.

### 2. *Nature of haze particles*

Electron-microphotographs of hygroscopic haze particles are shown in (17) to (20). As will be seen by comparing (17) with (1), (18) with (6), and (19) with (8) they are similar in form to the hygroscopic nuclei of clouds and fog. Furthermore these particles lost their hygroscopic nature by photographing. Thus hygroscopic particles in haze are similar both in form and in nature to cloud and fog nuclei.

Two hygroscopic particles evaporated completely as a result of the electron bombardment and left nothing on the collodion film. These particles probably consisted entirely of hygroscopic substances.

One example of non-hygroscopic particles in haze is shown in photo (16). There are three particles one of which is similar in form to the smoke particles of fuel (?) oil and others have circular form. Since these particles remained unchanged under exposure to hydrogen fluoride vapour it is presumed that they consisted of carbon.

## IV. Origin of cloud and fog nuclei

### 1. *Comparison with sea-salt particles*

Is has been considered that a large number of sea-salt particles are supplied from the sea-spray and that they are the most important condensation nuclei in the atmosphere. To decide whether the cloud and fog nuclei captured by us were sea-salt particles, small sea-salt particles with a diameter of about 0.5 $\mu$ were produced by spouting a water solution of sea-salt. Following the same experimental procedure as described before it was found that these salt particles had images of comparatively well defined crystalline form. Furthermore it was found that they were not only soluble in water but also did not lose their hygroscopic nature by electron-microphotographing. Thus the sea-salt particles have different form and nature from the greater part of cloud and fog nuclei.

As already described, however, a few nuclei of cloud and fog did not lose their hygroscopic nature by electron bombardment. But hygroscopic substances contained in these nuclei were composed of those with thin filmy appearance, which were far from salt crystals.

Recently KUROIWA and TADANO (KUROIWA 1951, KUROIWA and TADANO 1951) observed the nuclei of clouds on Mt. Niseko and those of sea fog on the coast of Hokkaido by using an electron microscope. They found no sea-salt nuclei on Mt. Niseko, and even in the sea fog only a small fraction of the nuclei were sea-salt particles. These results lead one

to the conclusion that sea-spray is not an important source of condensation nuclei of cloud and fog, at least not in these cases.

It is well known, however, that a considerable amount of sodium chloride is contained in cloud and fog water (KÖHLER 1922, HOUGHTON and RADFORD 1938, MIYAKE 1948), and the sea-salt nucleus theory has been based on this fact. Therefore it is necessary to explain the origin of sodium chloride in cloud and fog water. From some preliminary experiments sea-salt particles were found to have equal hygroscopic activity to combustion particles of the same size. Observations of sea-salt particles on the coast of Honshū (OGIWARA and ŌKITA 1951) showed that the number of particles larger than cloud and fog nuclei was about 5 per c.c. and was far less than that of cloud and fog droplets in the atmosphere. Therefore it seems reasonable to conclude that the greater part of cloud and fog droplets are formed on combustion nuclei, and that small sea-salt particles cannot act as condensation nuclei in the atmosphere. When cloud or fog water is collected by wire screen or other kinds of collectors, small sea-salt particles floating in the atmosphere should be captured together with the cloud or fog droplets. Large particles, which would become condensation nuclei, but are not detected in the cloud and fog droplets owing to the smallness of their number, would be added to the sodium chloride content in the cloud and fog water. Quantitative explanation for the sodium chloride content requires further investigation.

### 2. *Comparison with combustion products*

As it was found that cloud and fog nuclei are not sea-salt particles but are similar in form and nature to hygroscopic haze particles, the most probable source of the nuclei is considered to be combustion. Smoke particles produced by combustion of many kinds of fuel, such as coal, lignite, leaf of Japanese cypress, pine tree and grass were observed following the method described in chapter II. Some of these smoke particles are shown in photos (21) to (24). It was found that they too are a mixture of hygroscopic and non-hygroscopic substances and that they may lose their hygroscopic nature by electron-microphotographing. Cloud and fog nuclei are therefore similar in form and nature to combustion products. From the results described it may be concluded that most nuclei of cloud and fog originate in combustion.

It remains a question, whether the non-hygroscopic particles contained in cloud or fog nuclei are those captured by the cloud or fog droplets which have formed on the hygroscopic nuclei. As cloud and fog nuclei have similar form, nature and size to combustion products, the greater part of these non-hygroscopic particles are considered to have been adhering to the hygroscopic substances from the beginning.

### 3. *Chemical composition of combustion nuclei*

Generally combustion products are composed of many kinds of substances, therefore it is not easy to study their chemical composition in detail.

It has been considered that sulfur dioxide produced by combustion of coal etc. becomes sulfuric acid droplets by oxidation, and that these hygroscopic droplets can act as effective condensation nuclei of cloud and fog. To decide whether the hygroscopic substance in the nuclei is sulfuric acid the following experiments were made.

When a droplet of sulfuric acid with a diameter of about 0.5 $\mu$ was put into the chamber of constant humidity it increased its diameter by 2 to 3 times. Thus the increment of the droplet was of quite the same order as that of cloud and fog nuclei. Therefore the quantity of hygroscopic substances in cloud and fog nuclei cannot be considered to be far smaller than that of sulfuric acid. The droplet was then exposed to the electron beam of the microscope. Even after the electron bombardment, it was found that the droplet did not change its size nor its hygroscopic nature. From the above results it is obvious that the greater part of cloud and fog nuclei do not contain sulfuric acid. Chemical engineers have found that many kinds of sulfuric compound are contained in the smoke of coal and that some of them are hygroscopic. Therefore it may be considered that the sulfuric ions dissolved in cloud and fog water would originate in these sulfuric compounds but not in pure sulfuric acid droplets.

Next, to study the chemical composition of the nonhygroscopic solid particles in the nuclei, they were exposed to the vapour of hydrogen fluoride. Some particles were attacked by the vapour as shown in photo (5), while others were unchanged. By the same technique it was found that the solid particles in the smoke of light oil, leaf of Japanese cypress and grass were proof against hydrogen fluoride, while some of those from smoke of coal and lignite were attacked by hydrogen fluoride. It is presumed that the former is a pure carbon particle and the latter is a silicate or carbonate, and that the solid particles in the nuclei are composed of these substances.

## V. Conclusion

From the investigations set forth above it is concluded that the greater part of cloud and fog nuclei are composed of hygroscopic and non-hygroscopic substances which originate in combustion and that sea-spray is not a significant source of condensation nuclei. It was also shown that the greater part of the hygroscopic substances contained in the nuclei are not pure sulfuric acid. It is important to bear in mind that, contrary to the opinion which has been vigorously advocated so far, the greater part of cloud and fog nuclei are composed of neither sea-salt nor sulfuric acid.

A question will arise: If the combustion products are the most important nuclei, what was the most effective nucleus before man used fire? Even at that time combustion nuclei would be supplied by the eruption of volcanos or by forest fires. Or it may be more probable to consider that cloud and fog droplets would be formed on other kinds of less effective nuclei such as small sea-salt nuclei at a slightly higher atmospheric humidity than at present. As it has been assumed that by the appearance of life the chemical composition of the air has changed its proportions, it is perhaps no too bold to assume that by the appearance o man the most effective nucleus had changed from sea-salt to combustion products.

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## REFERENCES

HOUGHTON, H. G., and RADFORD, W. H., 1938: On the Measurement of Drop Size and Liquid Water Content in Fogs and Clouds. *Papers in Physical Oceanography and Meteorology*, **6**, No. 4.

KÖHLER, H., 1922: Zur Kondensation des Wasserdampfes in der Atmosphäre. 1 and 2. *Geofysiske Publikationer*, **2**, 1 and 2.

KUROIWA, D., 1951: Electron-Microscope Study of Fog Nuclei. *Journ. Meteor.*, **8**, 157—160.

KUROIWA, D., and TADANO, B., 1951: Investigations on the Condensation Nuclei of Fog by an Electron-Microscope. *Low Temperature Science*, **7**, 51—66.

MIYAKE, Y., 1948: The Chemical Nature of the Saline Matter in the Atmosphere. *Geophysical Magazine*, **16**, 64—65.

OGIWARA, S., and ŌKITA, T., 1951: The Size Distribution of Sea-Salt Nuclei and Its Contribution to Visibility. *Science Report Tōhoku Univ.*, Series **5**, 3, 78—86.

SIMPSON, G. C., 1941: On the Formation of Cloud and Rain. *Quart. Journ. Roy. Met. Soc.*, **67**, 99—133.