Electron Microscope Study of Cloud and Fog Nuclei

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Electron Microscope Study of Cloud and Fog Nuclei

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Abstract

Specimen changes due to electron bombardment in the electron microscope were investigated on numerous artificially produced nuclei. It was noticed that nuclei of different substances show the specimen changes characteristic to the respective substances. With use of this results, the nature of fog nuclei captured in Sendai and of cloud nuclei on Mt.Zaō was investigated. Several statistical results concerning the nature and size of fog and cloud nuclei were also described.

1. Introduction.

Recent development of the electron microscope technique seems to open a new field in the investigation of cloud physics. The investigations along this line have been carried out by KUROIWA [6, 7, 8] and OGIWARA and ŌKITA [10] on cloud and fog nuclei, by KUMAI [5] on snow crystal nuclei, and by JACOBI, JUNGE, and LIPPERT [3] and JUNGE [4] on atmospheric aerosols.

In the investigation of sea fog nuclei, KUROIWA [6] found that a considerable number of the observed sea fog nuclei were insoluble in water and that, contrary to the expectation, sea salt nuclei were rather scarce in number than combustion nuclei and soil particles. Also in the investigation of cloud nuclei at Mt. Niseko [8], he found, out of 41 particles observed, 6 sea salt nuclei, 14 soil particles, and 21 combustion nuclei. It was noticed that 4 cloud droplets had no nucleus. OGIWARA and ŌKITA [10] had also investigated the cloud nuclei on Mt.Zaō (1500m) in north eastern part of Japan and the fog nuclei near Sendai. According to them no sea-salt nuclei were found and the majority of cloud and fog nuclei observed were composed of hygroscopic and non-hygroscopic substances which originated in combustion. They further noticed that the greater part of the hygroscopic substances contained in the nuclei were not pure sulphuric acid. The recent investigation by JUNGE [4] of atmospheric aerosols with use of the electron diffraction pattern of the nuclei and by direct chemical analysis is one of the most excellent ones in this field. According to the result, most of the nuclei with a radius less than 1μ seem to consist of ammonium sulphate, while nuclei originating from sea salt in most cases are larger than 1μ.

These hitherto obtained results are not necessarily in fair agreement with each other, so that more works will be necessary to clarify the nature and size of atmospheric nuclei. In this context it will be most convincing to utilize the electron diffraction pattern of nuclei in detecting the nature of nuclei, as actually done by JUNGE. However, even with use of the electron diffraction pattern, it is practically difficult to take a clear picutre of the pattern when the specimen is smaller than, say, 1μ in diameter. Besides, at present, we

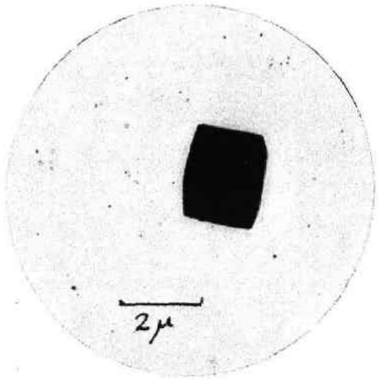


Fig. 1a Sodium chloride crystal under weak bombardment.

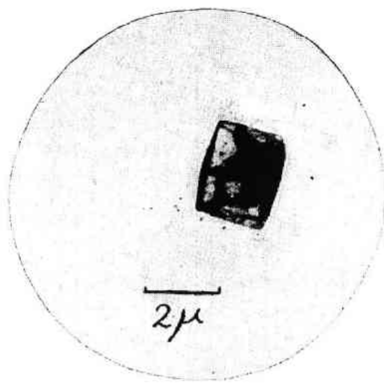


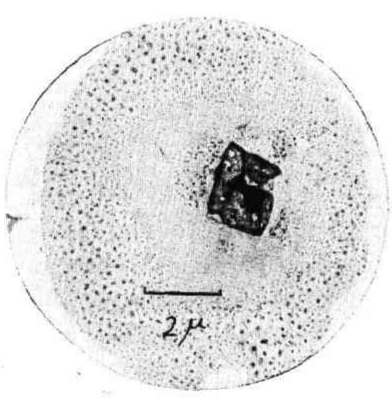
Fig. 1b The same crystal under intense bombardment.

are not in such a fabourable condition as to utilize the electron microscopic diffraction apparatus. So in the present investigation we tried to detect the nature of cloud and fog nuclei from their electron microscope pictures alone, but with higher approximation than the earlier works.

2. Changes of the artificially produced nuclei due to electron bombardment.

It is reported that a number of substances, chiefly ionic crystals, suffers changes when subject to intense electron bombardment in the electron microscope. For instance, according to BURTON, SENNETT and ELLIS [1], the crystals of soduim chloride, sodium chlorate, potassium bromide and potassium iodide, originally opaque, become transparent under intense electron bombardment. At first sight the effects seem due to the sublimation or melting of the crystals by the heating effect of the electron bombardment [11]. There are, however, some evidences that the temperature of the crystals under bombardment is too low for sublimation at pressures of the order of 10^{-4} to 10^{-5} mm of mercury. So the results are not interpretable by the hypothesis of sublimation alone and BURTON, SENNET and ELLIS [1] have considered that ionic migartion may play a part in the changes. While these changes are not readily interpretable, the phenomenon itself deserves attention for our present purpose of the detection of the nature of the unknown nuclei. If the changes of, for instance, sodium chloride crystal with increase of the intensity of electron beams are previously well known, we shall be surer in identifying a given fog nucleus with sodium chloride crystal, by bombarding the nucleus with weak, intense and more intense electron beams and by comparing the changes with the known changes of the sodium chloride crystal's.

Fig. 1c The same crystal under more intense bombardment



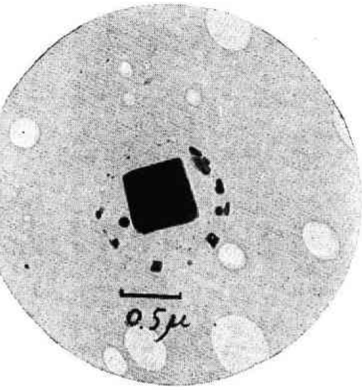


Fig. 2a Sodium chloride crystal under weak bombardment.

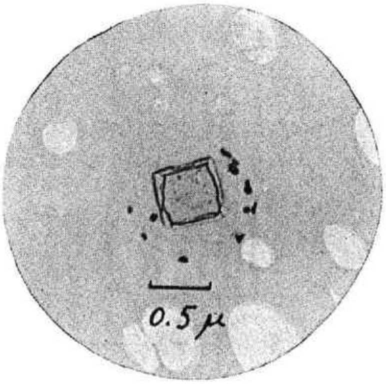
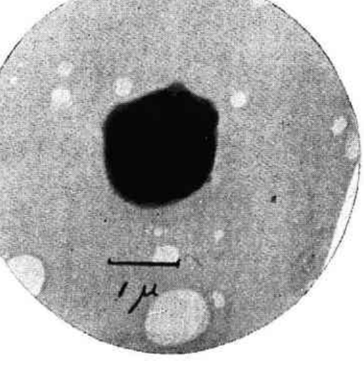


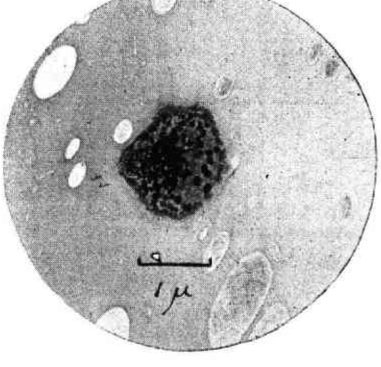
Fig. 2b The same crystal under intense bombardment.

With use of the electron microscope manufactured at Hitachi Co. LTD., specimen changes due to electron bombardment were studied for the artificially produced nuclei or crystals. Fig. 1a is the electron microscope picture of the weakly bombarded sodium chloride crystal which is artificially produced from a drop of sodium chloride solution by slow evaporation. When it is subject to intense electron bombardment the inner parts of the crystal become transparent as shown in Fig. 1b. The forms of the transparent portions are manifold ; some of them, like cumulus clouds, others, like window frosts, and sometimes small square-like transparent portions appear in the mother crystal. When the crystal is subject to more intense electron bombardment, there appears a large number of minute particular deposites around the mother crystal, which, on the whole, seem like 'halo'. (Fig. 1c). These halo particles will probably be composed of metallic sodium, and the mechanism of halo formation was investigated by HIBI [2] on the case of silver bromide. In the stage of intense electron bombardment, sometimes the crystal of less than about 1μ in size becomes wholly transparent as shown in Figs. 2a and 2b. On the case of a still smaller crystal, it becomes transparent even under weak bombardment.

Fig. 3a Magnesium chloride crystal under weak bombardment.

Fig. 3b The same crystal under intense bombardment.





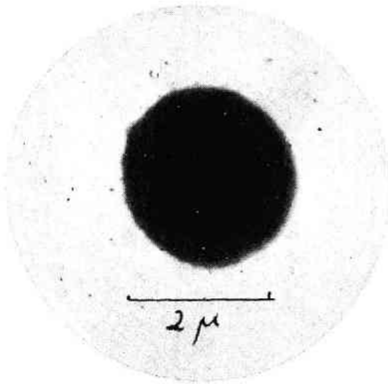


Fig. 4a Magnesium sulphate particle under weak bombardment.

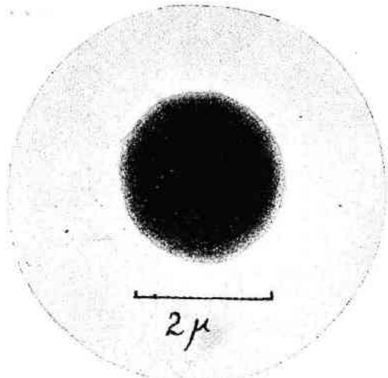


Fig. 4b The same particle under intense bombardment.

Specimen changes due to electron bombardment of some other compor ents of sea salt were also studied. The changes of the magnesium chloride crystal are shown in Figs. 3a and 3b. When subject to intense bombardment, the crystal becomes partly transparent with black spotts in it. The changes of magnesium suplhate particle are shown in Figs. 4a, 4b and 4c. The particle of magnesium sulphate crystalizes very rarely, and when crystalized, it contains crystal water, its chemical composition being mainly $MgSO_4.7H_2O$ and its crystal habit being rhombic. The particle shown in Fig. 4a was obtained from a drop of dilute solution of magnesium sulphate by slow evaporation. As the particle is nearly spherical, so it will probably be non-crystalline magnesium suplhate particle. When it is subject to intense electron bombardment, it becomes spottedly transparent near the edge of the particle, and under more intense electron bombardment, the transparent small spotts spreads into the inner part of the globe. The electron microscope picture of calcium sulphate particle is as shown in Fig. 5. The particle does not suffer changes with increase of the intensity of electron beams. The pictures of potassium sulphate particle are shown in Figs. 6a, 6b and 6c, from which we can see that the particle suffers changes similar to

Fig. 4c The same particle under more intense bombardment.

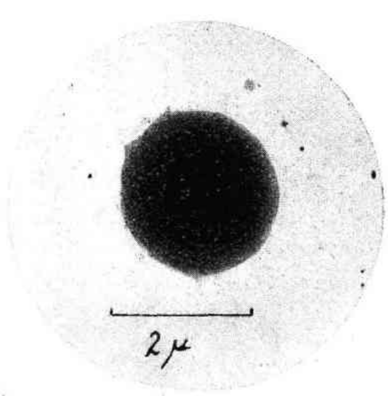
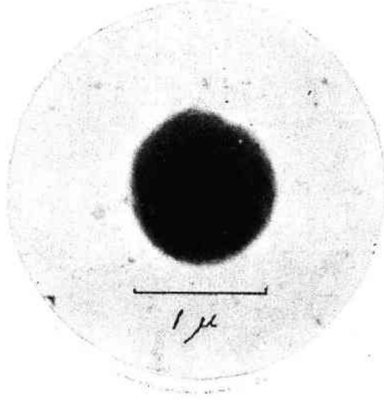


Fig. 5 Calcium sulphate particle. It does not suffer changes under intense bombardment.



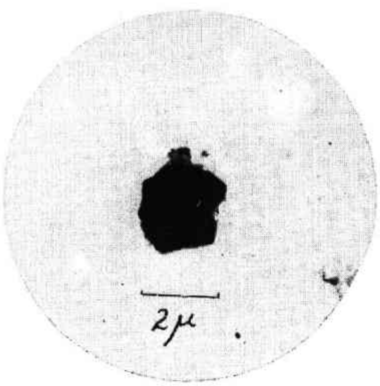


Fig. 6a Potassium sulphate crystal under weak bombardment.

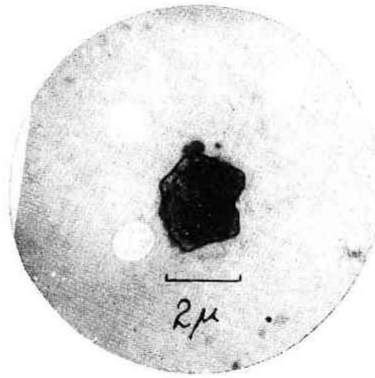


Fig. 6b The same crystal under intense bombardment.

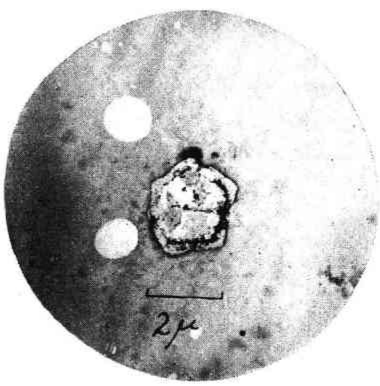


Fig. 6c The same crystal under more intense bombardment.

Fig. 7a Sea salt paritcle under weak bombardment.

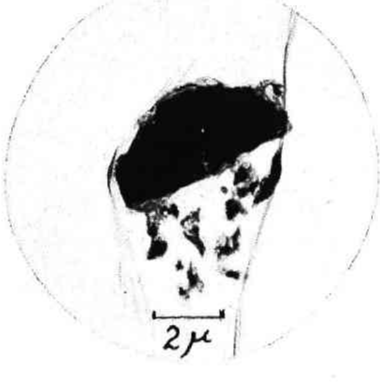


Fig. 7b The same particle under intense bombardment.

Fig. 7c The same particle under more intense bombardment.

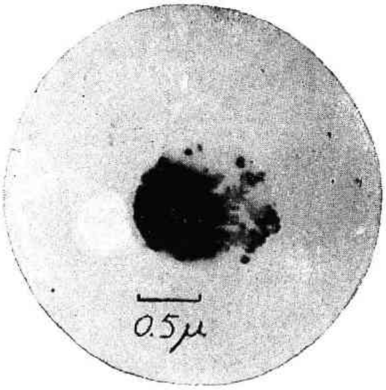


Fig. 8a Sea salt particle under weak bombardment.

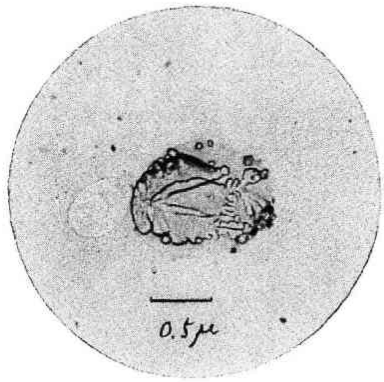


Fig. 8b The same particle under intense bombardment.

the sodium chloride crystal.

Next we examined the changes of sea salt nuclei, which were also artificially produced from droplets of sea water by slow evaporation. Figs. 7a, 7b and 7c show the changes of large sea salt nuclei. Generally large sea salt nuclei take irregular shapes. So that, if the electron microphotograph of Fig. 7a, which was taken under weak electron bombardment, alone were shown, we would be bewildered in identifying the particle to be a sea salt nucleus. However, by observing the changes shown in Figs. 7b and 7c, which exhibit the characteristic feature of sodium choloride crystal, we may easily be convinced that the particle is the sea salt nucleus. Another example of the changes of sea salt nucleus which was obtained from a small drop of sea water is shown in Figs. 8a and 8b. The particle is irregular and blurred at the boundary even under weak bombardment as shown in Fig. 8a. When subject to intense bombardment the particle becomes transparent and there appear ferny veins as shown in Fig. 8b.

We further examined the specimen changes due to electron bombardment of combustion products and industrial products. There are many sorts of nuclei which may be classified in this category. However, according to JUNGE [4] most important ones among them are ammonium sulphate and ammonium sulphite particles. So we shall show several electron microscope pictures of them in Figs. 9a, 9b, 9c, 10a, 10b, 11, 12, 13, 14 and 15. The particles are very sensitive to electron bombardment, that is to say, they suffer changes even under weak bombardment. Accordingly, the specimens shown in Figs. 11~15 do not suffer changes any more with increase of the intensity of electron beams. Unfortunate for us, the general aspect of the specimen changes of the particles are very similar to those of some of sea salt nuclei. However, by the precise comparison of pictures of both ammonium sulphate and sea salt particles, we can find several different points of both particles. First, ammonium sulphate particles are more blurred at the boundary than sea salt particles even under weak bombardment. Second, under intense bombardment ammonium sulphate particles do not show the exudation from the mother particles or halo, which is frequently observed on the collodion film around the sodium chloride or sea salt particles. When both particles are small these differing features disappear and the distinction of both particles becomes very difficult. Ammonium sulphite

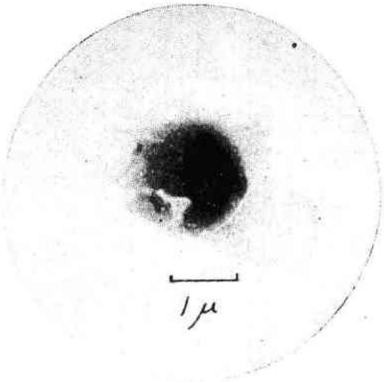


Fig. 9a Ammonium sulphate particle under weak bombardment.

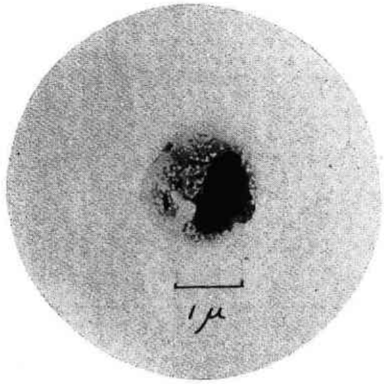


Fig. 9b The same particle under intense bombardment.

particles too show similar features to ammonium sulphate particles.* Another important combustion product is carbon particle, whose electron microscope picture is very characteristic as shown in Figs. 16 and 17, and which does not suffer appreciable changes under intense bombardment of short duration.

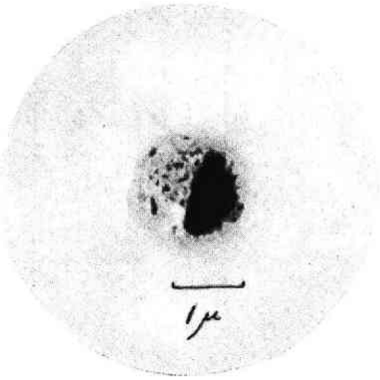


Fig. 9c The same particle under more intense bombardment.

Finally some remarks may be added as to the electron microscope picture of soil particles. Generally soil particles have the crystalline structure and are non-hygroscopic and insoluble in water and do not suffer changes under electron bombardment. The electron microscope pictures of them are already shown by KUROIWA [6, 7], and KUMAI [5].

3. The identification of the natural nuclei as the known ones.

The above described specimen changes due to electron bombardment in the electron microscope were utilized by us in identifying the natural nuclei as the known ones. We also

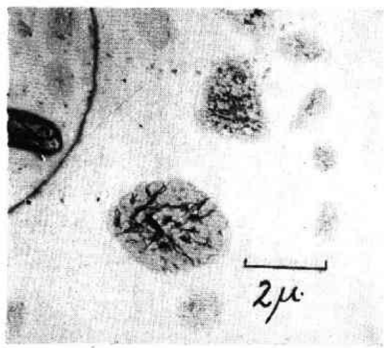


Fig. 38 Fuming sulphuric acid.

* After the present paper was written, we got in hand the electron microscope picture of fuming sulphuric acid as shown in Fig. 38 by courtesy of Professor Hibi. The particles shown in the figure are also very similar to some pictures of sea salt nuclei and ammonium sulphate nuclei. So that it becomes increasingly difficult to distinguish the nature of the small natural nuclei by electron microscopic method. The result of the present investigation, however, may not be altered owing to this fact, because most of the small nuclei were reserved by us as unknown nuclei.

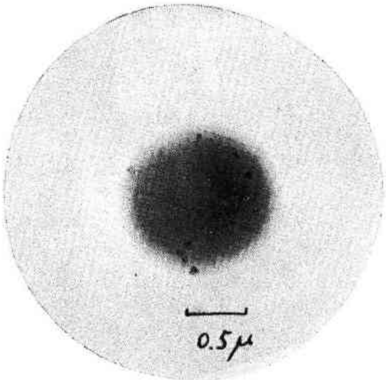


Fig. 10a Ammonium sulphate particle under weak bombardment.

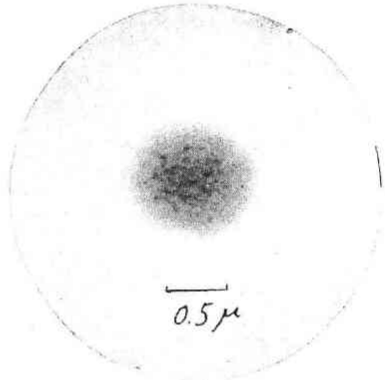


Fig. 10b The same particle under intense bombardment.

carried out the hygroscopicity test of the nuclei by the method described in OGIWARA and ŌKITA's paper [10], and no further description on it will be given here.

Some examples of the electron microphotograph of cloud and fog (or haze) nuclei captured by us are shown in Figs. 18~34. Fig. 18a shows a fog nucleus, captured in Sendai, under weak electron bombardment. When subject to intense bombardment, it suffers changes (Fig. 18b) characteristic to sea salt nuclei, and the nucleus is hygroscopic and water soluble. Accordingly we presumed that it is a sea salt nucleus. Fig. 19a shows antoher fog particle, captured in Sendai, under weak bombardment. With increase of the intensity of electron beams it suffers changes as shown in Figs. 19b and 19c. The pictures show 'halo' characteristic to sea salt, and the transparent portions in the particle show straight faces which suggest the cubic structure of the transparent portions. In

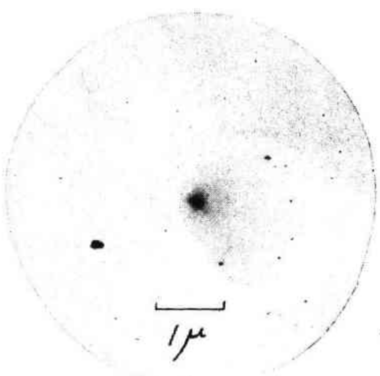


Fig. 11 Ammonium sulphate particle. Pictures of Figs. 11~17 were all taken under weak bombardment. These paritcles did not suffer appreciable changes under intense bombardment.

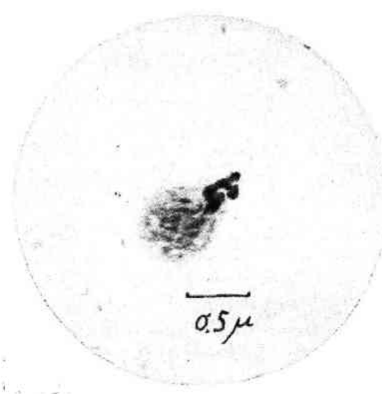


Fig. 12 Ammonium sulphate particle.

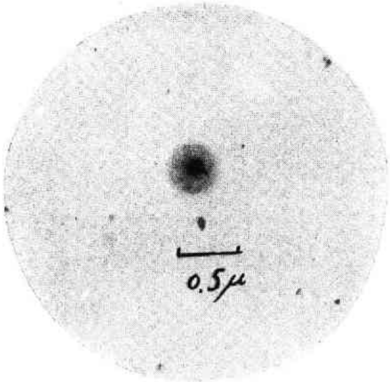


Fig. 13 Ammonium sulphate particle.

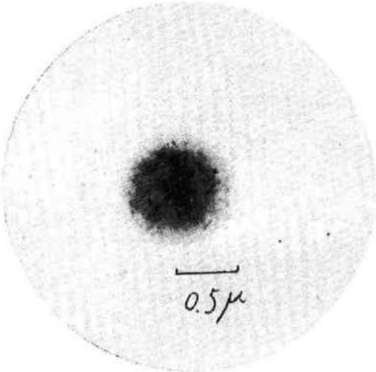


Fig. 14 Ammonium sulphite particle.

addition the particle is hygroscopic and water soluble, hence we presumed that the particle is sea salt. Fig. 20a shows the picture of the largest fog nucleus observed by us in Sendai. In this case the hygroscopicity test was first carried out and the test of the specimen changes was carried out on the deformed sample as shown in Figs. 20b and 20c which show the characteristic features of sea salt nuclei definitely. Fig. 21a shows a fog particle captured in Sendai under weak bombardment. When subject to intense bombardment, the particle suffers changes as shown in Fig. 21b. By the transparent, ferny-like figure and the weak exudation around the particle it seems likely that the particle is sea salt. But, in this case there is a tolerable possibility that it is a particle of ammonium sulphate or sulphite. Several numbers of similar nuclei were captured in fog in Sendai as shown in Figs. 22 and 23. In these cases the transparent, ferny-like figures appeared very clearly even under weak bombardment, probably owing to the smallness and thinness of the nuclei. We are inclined to conceive them to be sea salt nuclei, but still some question remains that they may be ammonium sulphate or sulphite. For the sake of clarifying the point we made the electron diffraction pattern inspection, but the

Fig. 15 Ammonium sulphite particle.

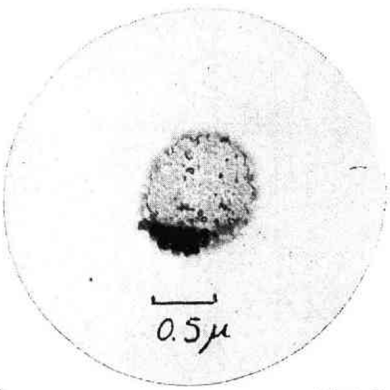
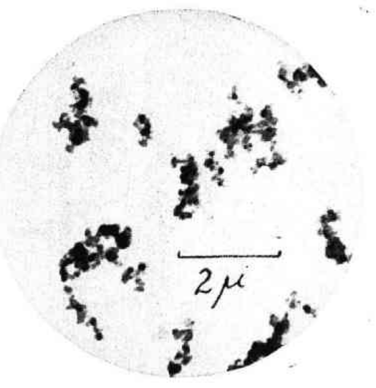


Fig. 16 Carbon particles.



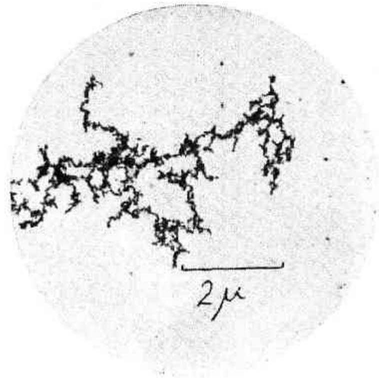


Fig. 17 Carbon particles.

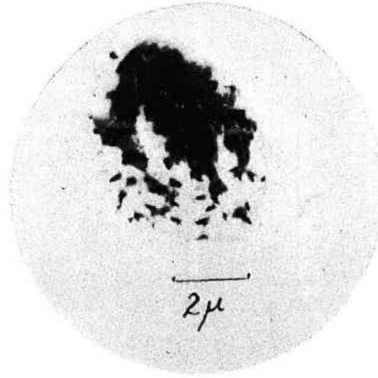
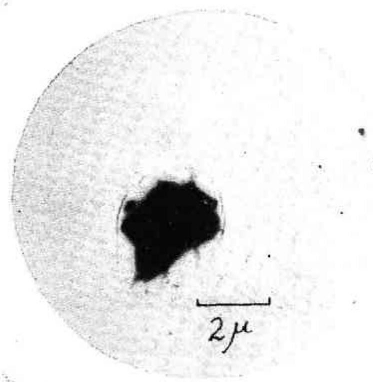


Fig. 18a A fog nucleus in Sendai, under weak bombardment.

result was unsatisfactory owing to the smallness of the specimens and to the unfabourable apparatus, so that at present we reserved them in the category of indistinguishable nuclei. In our observation sea salt nuclei were also found in cloud droplets on Mt. Zaō. Figs. 24a and 24b show the specimen changes of one of the nuclei, which show the decided feature that the particle is sea salt. Figs. 25a and 25b show another example of sea salt nuclei in which black spotts are seen scattered around the mother particle. These spotts do not suffer changes under electron bombardment and they are quite similar to the artificially produced calcium sulphate particle shown in Fig. 5, with the exception of the size. So it may be conceived that they are particles of calcium sulphate separated from the mother sea salt nucleus. Similar separation of salt components likely other than sodium chloride are frequently observed in the case of large sea salt nuclei. Some years ago MIYAKE [9] noticed in his observation of saline matter in the atmosphere that, when the percentage compositions are compared, the relative amount of sulphate and calcium is much larger in the salt contained in the precipitation water than in the oceanic salt. He attributed the

Fig. 18b The same nucleus under intense bombardment. By the changes it is presumed to be a sea salt nucleus.

Fig. 19a A fog nucleus in Sendai, under weak bombardment.



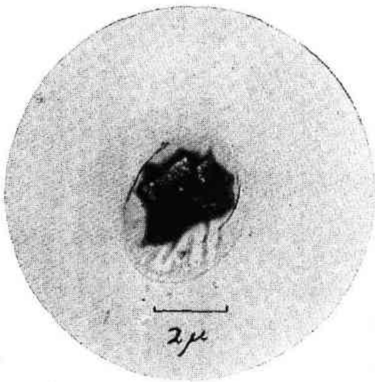


Fig. 19b The same nucleus under intense bombardment.

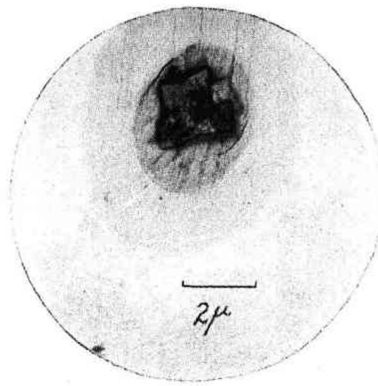


Fig. 19c The same nucleus under more intense bombardment. By the changes it is presumed to be a sea salt nucleus.

cause to the separation of some salts having small solubility such as calcium sulphate from the mother droplet during evaporation and to the possible early rain-out of the mother droplet. It seems that above mentioned electron microscope observation on large sea salt nuclei ascertains MIYAKE's explanation of his observed results. Because the electron microscope picture under weak bombardment may be conceived to reveal the evaporated state itself of the droplet.

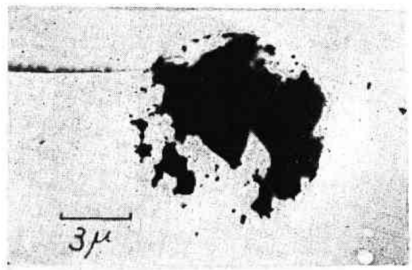


Fig. 20a A fog nucleus in Sendai under weak bombardment.

Another interesting example of cloud nuclei is shown in Figs. 26a and 26b. Inspite of its smallness of size it appeared as a black particle under weak bombardment and changed to transparent under intense bombardment. By its sharp bounded figure we conceived it to be a sea salt particle and not ammonium sulphate. Similar small sea salt nuclei were found several times in

Fig. 20b Modified figure of the same nucleus by the hygroscopicity test, under weak bombardment.

Fig. 20c The same sample under intense bombardment. By the changes it is presumed to be a sea salt nucleus.

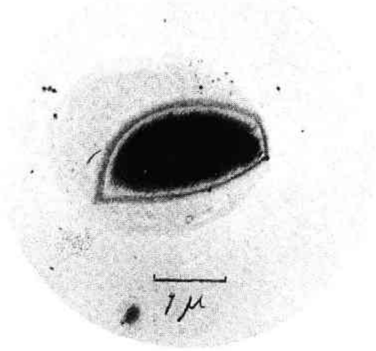


Fig. 21a A fog nucleus in Sendai under weak bombardment.

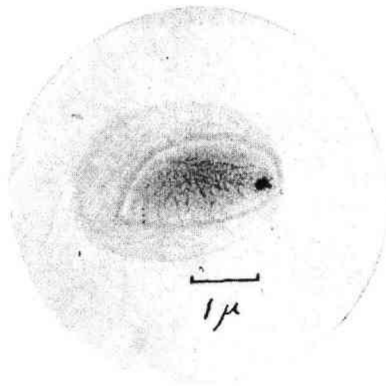


Fig. 21b The same nucleus under intense bombardment. By the changes it seems very likely to be a sea salt nucleus, but with consideration of the possibility of its being ammonium sulphate, it was reserved as an indistinguishable one.

cloud droplets on Mt. Zaō.

Next we shall show the pictures of nuclei other than the sea salt, although the study of their nature is at present not satisfactory. Pictures of nuclei presumed to be soil particles are shown in Figs. 27, 28 and 29. The former two particles were obtained from fog droplets captured in Sendai and the latter one was found in cloud droplets on Mt. Zaō. The causes that we presume them to be soil particles are that they have crystalline structure and that they do not suffer changes with increase of the intensity of electron beams and that they are non-hygroscopic and water-insoluble. Fig. 30 shows a cloud nucleus captured on Mt. Zaō. The particle is presumed to be a sulphur particle, because it is quite similar to the artificially produced sulphur particle, which is shown in Fig. 31, and the existence

Fig. 22 A fog nucleus in Sendai under weak bombardment. presumed to be an indistinguishable one.

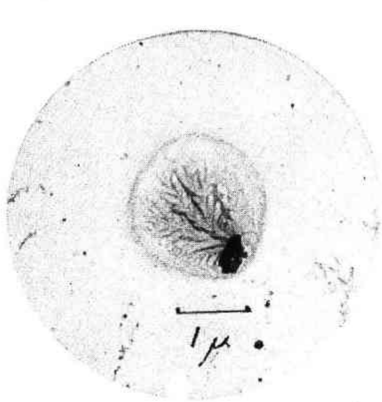


Fig. 23 A fog nucleus in Sendai under weak bombarmdent, presumed to be an distinguishable one.

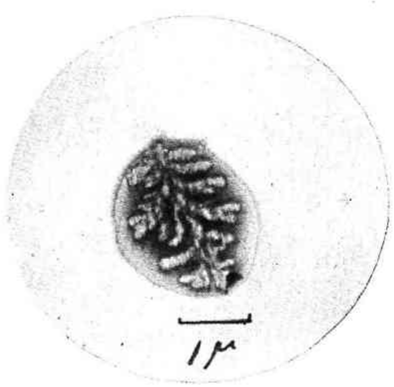


Fig. 24a A cloud nucleus on Mt. Zaō under weak bombardment.

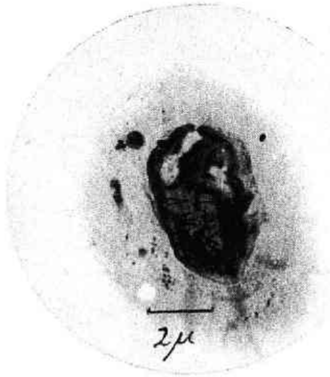


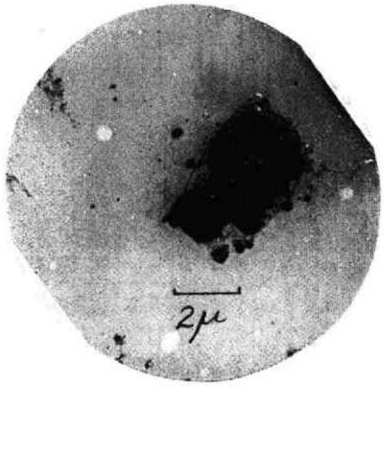
Fig. 24b The same nucleus under intense bombardment. By the changes it is presumed to be a sea salt nucleus.

of sulphur nuclei in cloud droplets on Mt. Zaō will be due to the fact that sulphur gas is emitted here and there on Mt. Zaō.

Pictures of paritcles which may be conceived to be combustion products or industrial products will be shown next. In this context JUNGE's results were very useful to us. Fig. 32 shows a fog nucleus captured in Sendai and Fig. 33 shows a cloud nucleus captured on Mt. Zaō both under weak bombardment. These pictures are very similar to the picture shown by JUNGE (Fig. 6 of his paper [4]) as the typical example of the ammonium sulphate nuclei. Figs. 34a and 34b show a fog nucleus captured in Sendai, 35a under weak bombardment and 34b under intense bombardment. As there seem not so appreciable changes between the two pictures, and as the particle is hygroscopic, so it may possibly be a combustion products, other than ammonium sulphate or sulphite.

Fig. 25a A cloud nucleus on Mt. Zaō under weak bombardment.

Fig. 25b The same nucleus under intense bombardment. By the changes it is presumed to be a sea salt nucleus.



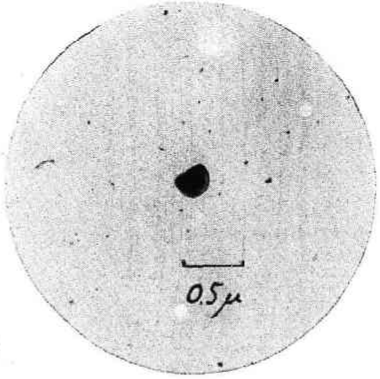


Fig. 26a A cloud nculeus on Mt. Zaō under weak bombardment.

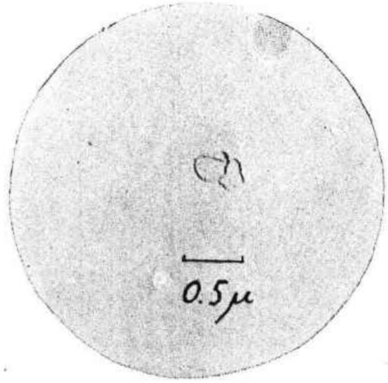


Fig. 26b The same nucleus under intense bombardment. By the changes it seems to be an example of small sea salt nuclei.

4. Statistical discussion of the results

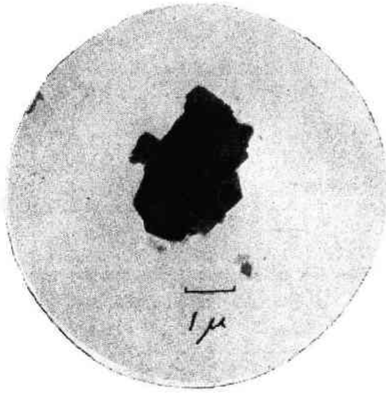
The results of observation for cloud and fog (or haze) are listed in Table 1.

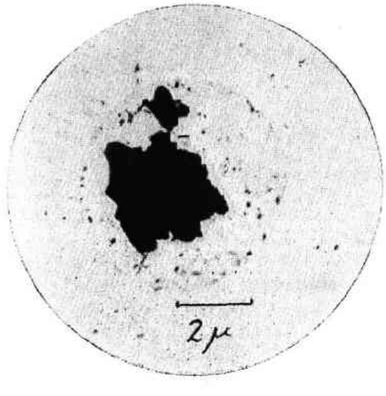
Table 1. Classification of observed nuclei.

	fog (or haze) nuclei in Sendai		cloud nuclei on Mt. Zaō	
	number	percent	number	percent
sea salt	14	23	11	16
combustion product	18	30	25	36
soil matter	12	20	16	23
unknown	16	27	17	25
Total	60	100	69	100

Fig. 27 A fog nucleus in Sendai under intense bombardment, presumed to be a soil particle.

Fig. 28 A fog nucleus in Sendai under intense bombardment, presumed to be a soil particle.





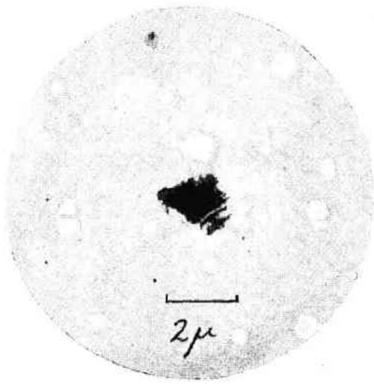


Fig. 29 A cloud nucleus on Mt. Zaō under intense bombardment, presumed to be a soil particle.

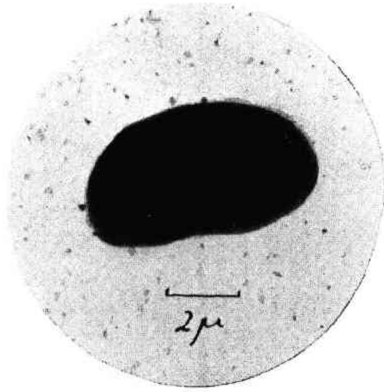


Fig. 30 A cloud nucleus on Mt. Zaō, presumed to be a sulphur particle.

It will be seen that considerable numbers of sea salt nuclei are found in fog of Sendai and in cloud of Mt. Zaō. These results of us are not in agreement with the results of OGIWARA and ŌKITA [10], who found no sea selt nuclei in the same places and our results are in the same tendency to KUROIWA's results. It will also be seen in Table 1 that the percentage of sea salt nuclei is larger in fog of Sendai than in cloud of Mt. Zaō. For the explanation of this fact it must be remarked that Sendai-city is situated only about 12 kms far from the Pacific Ocean and the formation of fog in Sendai is generally accompanied with the breeze from the sea, while Mt. Zaō stands in the middle part of the Tōhoku district about 50 kms far from the Pacific Ocean and 80 kms far from the Japan Sea. Now the decrease of sea salt nuclei over the land area was recently pointed out by WOODCOCK [12]. He attributed the cause of the decrease to rain-out of the large sea salt nuclei in the windward high rain areas. In the cases of our observation of cloud nuclei on Mt. Zaō, sometimes

Fig. 31 Sulphur particle artificially produced.

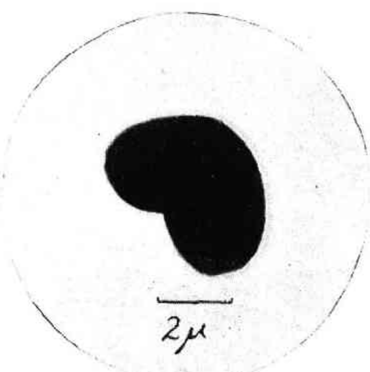
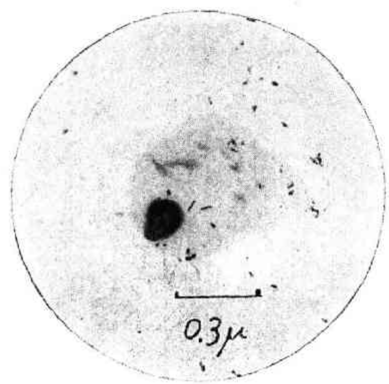


Fig. 32 A cloud nucleus on Mt. Zaō under weak bombardment, presumed to be a combustion product.



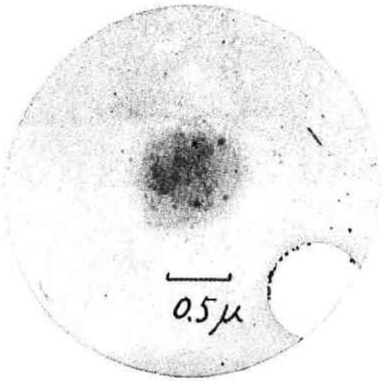


Fig. 33 A cloud nucleus on Mt. Zaō under weak bombardment, presumed to be a combustion product.

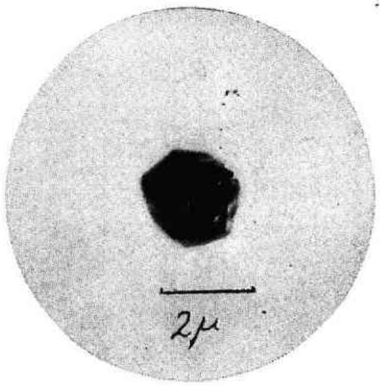


Fig. 34a A fog nucleus in Sendai under weak bombardment.

clouds were really accompanied by rain areas in the windward, but at other times clouds were found only on Mt. Zaō and the weather was fine in the mountainless region. So that, although the main cause of the decrease will be the rain-out, even the mere fall down of the large fog (or haze) droplets to ground will also play some part on the decrease of the sea salt nuclei over the land area.

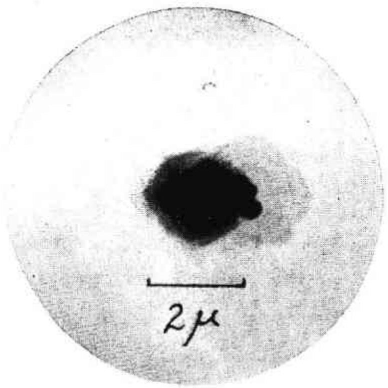


Fig. 34b The same nuclues under intense bombardment. It is presumed to be a combustion product.

Next we shall show the size-frequency curves for cloud and fog nuclei respectively (Fig. 35). Here the size of the irregularly shaped nucleus was determined by its mean diameter and in the case of the ferny-like figure such as shown in Figs. 21~23, not only the black portion but also the ferny-like portion were counted in the size. As this ferny-like

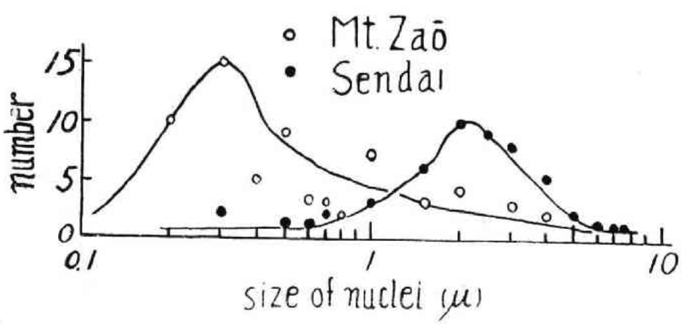


Fig. 35 The size-frequency curves for cloud and fog nuclei.

portion will be very thin, the size, here assumed, represents only one dimensional scale of the nucleus, and do not represent its volume. It is noticed in Fig. 35 that the cloud nuclei on Mt. Zaō are generally smaller than the fog nuclei in Sendai. Most frequently observed sizes for cloud and fog nuclei are respectively 0.3μ, and 2μ. It has also shown by WOODCOCK [12] that the size of sea salt particles decreases with increase of altitude. Fig. 35 indicates that this tendency holds not only of sea salt nuclei but of all sorts of nuclei.

The size-frequency relation was also examined respectively for sea salt nuclei, combustion products and soil particles in each case of cloud on Mt. Zaō and fog in Sendai

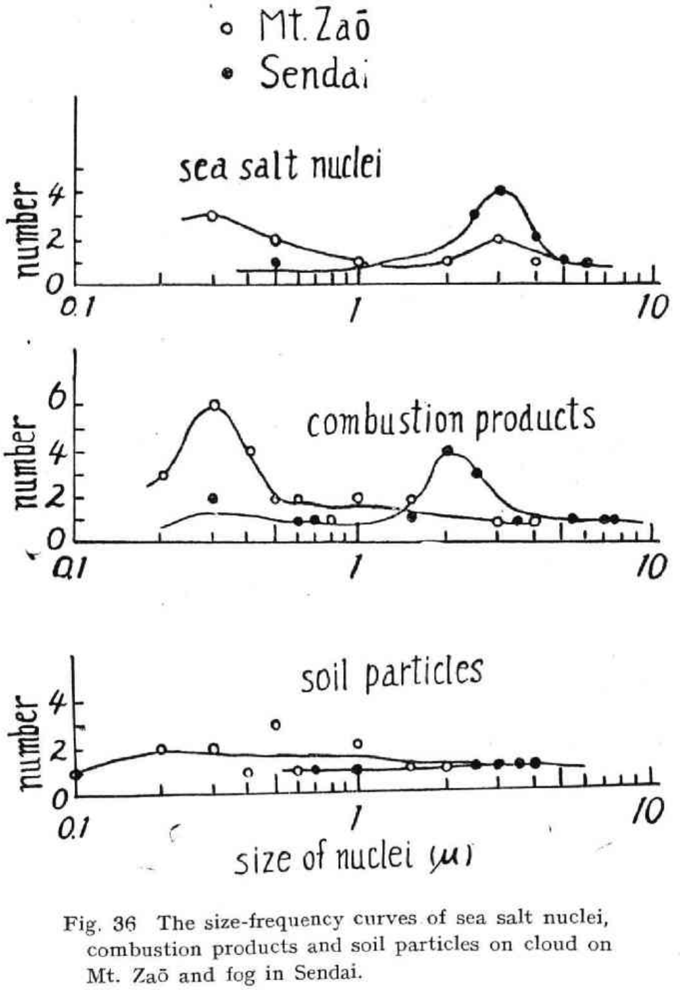


Fig. 36 The size-frequency curves of sea salt nuclei, combustion products and soil particles on cloud on Mt. Zaō and fog in Sendai.

(Fig. 36). In the case of fog in Sendai, sea salt curve shows maximum at 3μ and combustion product curve at 2μ, while soil particle curve is nearly flat. In the case of cloud on Mt. Zaō, in addition to soil particle curve, sea salt curve also becomes nearly flat, and the maximum of combustion product removes to 0.3μ. This result will mean that not only of sea salt nuclei but also of combustion nuclei which are mainly produced in populated region the larger ones rain-out earlier than the smaller ones in the course of drifting aloft the sky.

Next, if we construct the size-frequency curves for the sum of cloud nuclei on Mt. Zaō and fog nuclei in Sendai, we have curves shown in Fig. 37. In this case soil particle curve

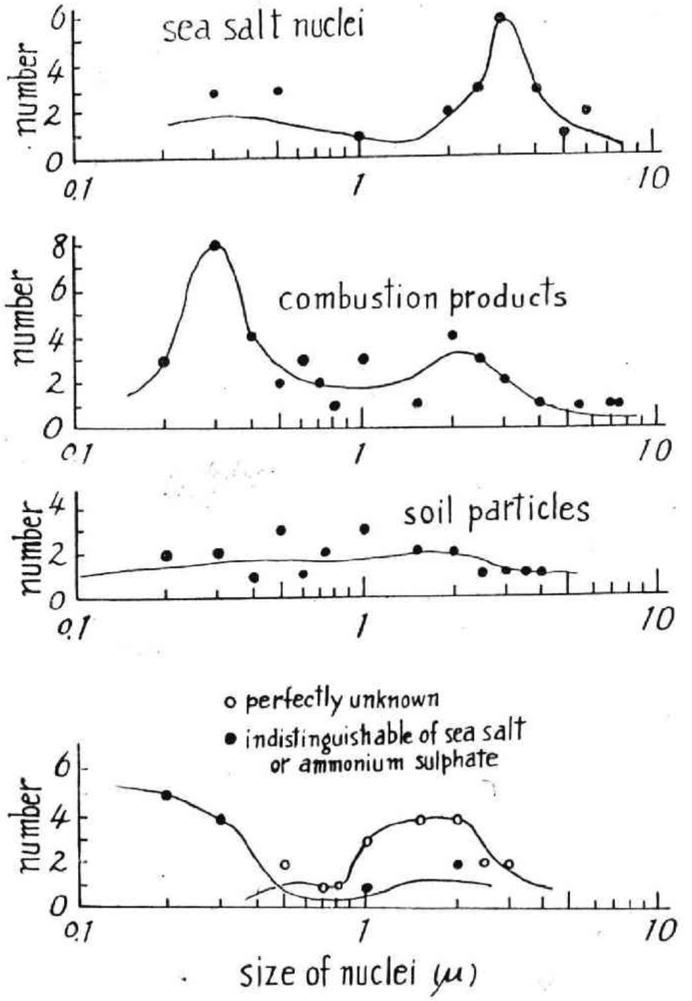


Fig. 37 Size-frequency curves irrespective of cloud and fog.

is of course flat, while the sea salt curve has maximum at 3μ and the combustion product curve, at 0.3μ. This result is in good agreement with JUNGE's result that, "most of the nuclei with a radius less than 1μ seem to consist of ammonium sulphate, while nuclei originating from sea salt in most cases are larger than 1μ." In Fig. 37 are also shown the size-frequency curve for perfectly unknown nuclei and the curve for nuclei indistinguishable whether they are sea salt or ammonium suplhate. As will be seen the former, perfectly unknown nuclei, are mainly small nuclei, and the latter curve has maximum at 2μ.

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