

# SIMPLE TECHNIQUE FOR THE MEASUREMENT OF GIANT SEA-SALT PARTICLES BY USE OF A HAND-OPERATED IMPACTOR AND A CHLORIDE REAGENT FILM

By

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## Abstract

The design and use of a hand-operated impactor, by which giant sea-salt particles in six different volumes of air may be sampled on a chloride reagent film of 1 cm<sup>2</sup>, are described. Some practical notes on the preparation and use of Farlow's chloride reagent gelatin film, which has been used with the impactor to determine dry masses of the giant sea-salt particles, are also presented.

## 1. Introduction

Measurement of aerosols by the use of a jet impactor has a long history since May [1945]. We devised a simple but handy impactor, with which we can sample giant sea-salt particles in six different volumes of air, on a chloride reagent film of 1 cm<sup>2</sup>, and have used it for these several years. We participated in the International Meeting of Comparison of Condensation and Ice-Forming Nuclei Generators and Counters, held in Lannemezan, France, from September 8 to September 22, 1967, and took the impactor with us, together with a continuous sampler; the impactor in its improved form. Since that time the hand-operated impactor has come to be used in institutions in several countries. Under these circumstances, we have felt encouraged to publish a short note on the design and use of the simple impactor, as well as on a slight improvement in the preparation and use of Farlow's chloride reagent gelatin film, which we have been using as the sampling surface.

## 2. Hand-operated impactor for giant sea-salt particles

### (a) *Design and use of the impactor*

The design of the impactor is shown in Fig. 1. The part marked A is the

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air intake with an orifice of 1 mm diameter. On the surface of stage C, a reagent film D of 1 cm×1 cm is set by use of spring E. The distance between the film and the orifice is 1.5 mm. The setting of the film is performed by taking off the air intake. By use of a 100-cc hypodermic syringe G, an air jet is formed at the orifice, and giant particles impact on a circular area of 1 mm diameter on the reagent film. By rotating stage C by use of handle F, six different volumes of air may be sampled successively as shown in the left hand part of Fig. 1. Needle marks, which are made by pushing a needle through the orifice, alternately with the sampling as shown in the figure, are very useful when identifying the areas of halos in a microscope examination. According to the number concentration of the particles at the time of sampling, one or two out of the 6 samples of different air volumes are selected and read off. Sometimes the 8 l sample contains an appropriate number of halos, while the 2 l sample, for example, contains too few. But in other case, the 1 l is appropriate, while the 4 l sample has so many halos that many of them coalesce. According to the locality (distance inland etc.) the volumes of air sampled may be shifted to smaller or larger volumes. An O-ring B, silicone rubber and 2 washers at handle F, keep the inside airtight. The air once drawn into the syringe is slowly discharged through the same orifice.

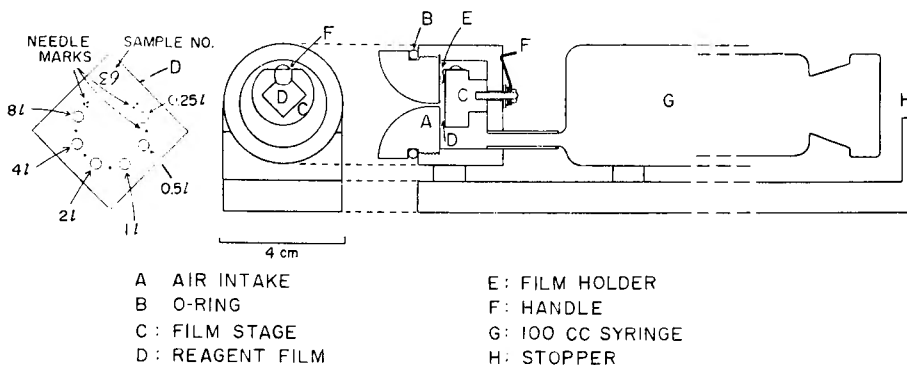


Fig. 1. Design of the hand-operated impactor for giant sea-salt particles.

Using a theoretical and experimental study by Ranz and Wong [1952], it is calculated that sea-salt particles larger than  $1\mu$  diameter are collected perfectly by the jet impactor of a round orifice of 1 mm diameter when the jet speed is higher than 40 m/sec. For the present impactor, a jet speed of 42 m/sec or larger is attained if the syringe is drawn at a speed of 3 sec per 100 cc or faster, and in this case efficiency of impaction should be considered 100% for giant sea-salt particles. If an air jet with a very high speed such as 100 m/sec is used, the circular area of the halos becomes much smaller than 1 mm, and the halos

tend to be grouped together. This is not a desirable condition. It is also recommended that the impactor be used with the air intake against the wind, since otherwise the particles may escape because of their inertia from the wind.

(b) *A note on the absolute measurement of the particle number concentration*

There is a possibility that particles once collected on the sampling surface may be blown off again by the air jet. As a matter of fact, by examination of the particle number for various volumes of air, sampled on a film, it is found that the counted particle number is not twofold for a twofold sampled volume of air.

Black circles in Fig. 2 are a combination of several data on the relation between the sampled volume of air on one spot and the counted particle number. If particles once collected on the sampling film are not blown off, the counted number  $N$  is to be expressed by

$$\frac{dN}{dV} = r, \quad N = rV \quad (1)$$

where  $V$  is the sampled volume of air and  $r$  the number concentration of the particles, and the equation (1) is the straight line shown by  $a$  in Fig. 2. So a "retention factor"  $\epsilon$  shown in the lower part of the figure should be considered: the value becomes 0.3 to 0.25 for a very large sampled volume of air in the present example.

If we assume that the rate of being blown off is proportional to the number of particles which are on the film surface at the time considered, with a factor of proportionality  $k$ , the relation would be expressed as

$$\frac{dN}{dV} = r - kN, \quad N = \frac{r}{k}(1 - e^{-kV}) \quad (2)$$

and  $N$  would approach a constant value  $r/k$  for large  $V$ . Actually, it is not the case, but the  $N$  continues to increase with  $V$ .

So we are to consider that the particles consist of two categories: one composed of particles not blown off (group I) and the other having the rate of being

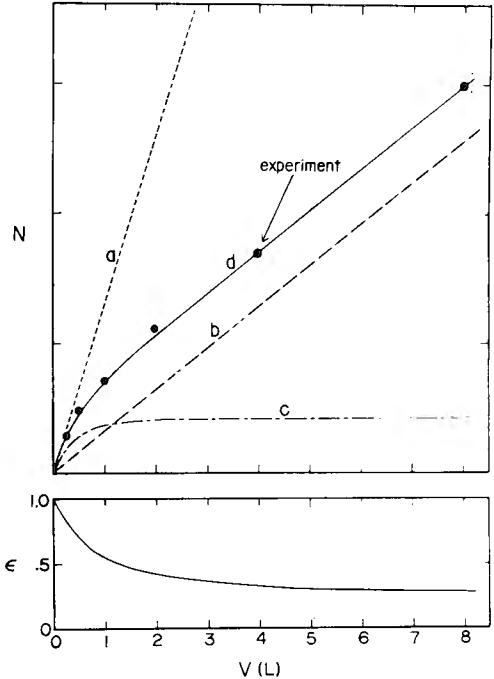


Fig. 2. Relation between the sampled volume of air on one spot and the counted particle number. See context.

blown off  $k$  (group II). The particles of group I might be wet particles. Then we have

$$\frac{dN_1}{dV} = \epsilon_{\infty} r, \quad N_1 = \epsilon_{\infty} r V \quad (3)$$

$$\frac{dN_2}{dV} = (1 - \epsilon_{\infty}) r - k N_2, \quad N_2 = \frac{(1 - \epsilon_{\infty})}{k} r (1 - e^{-kV}) \quad (4)$$

$$N = N_1 + N_2 = \epsilon_{\infty} r V + \frac{(1 - \epsilon_{\infty})}{k} r (1 - e^{-kV}). \quad (5)$$

If we determine the coefficients from the black circles of Fig. 2, we have

$$\epsilon_{\infty} = 0.25 \quad \text{and} \quad k = 2.3, \quad (6)$$

and the equation (5) is shown by a curve  $d$ ,  $N_1$  and  $N_2$  being shown by curves  $b$  and  $c$ , respectively. These coefficients, however, must vary according to different conditions of humidity, the size of the particles, etc. Consequently the above values are just a preliminary estimate. But it seems necessary that this kind of retention factor should be taken into account.

At the same time, it is shown that the above mentioned decrease in the particle number is not caused by the effect of the overlapping of the reaction halos. Denoting the number of particles collected on the sampling film by  $N_*$  and the average ratio of the area of a reaction halo to the total area of the film by  $S$ , the rate of increase of the number of halos  $N$  is expressed by

$$\frac{dN}{dV} = r(1 - SN_*). \quad (7)$$

On the other hand, the rate of increase of the ratio of the total area of halos to the total area of the film  $SN_*$ , is expressed by

$$\frac{d(SN_*)}{dV} = Sr. \quad (8)$$

From which it follows that

$$N = \left(1 - \frac{1}{2} SN_*\right) N_*, \quad (9)$$

and relative error of the apparent (counted) number  $N$  compared to the true (collected) number  $N_*$  of the particles is expressed by  $SN_*/2$ . Consequently, if we have 100 halos of  $20 \mu$  diameter in  $1 \text{ mm}^2$  of the film area, the error in the number is smaller than 2%.

### 3. Notes on preparation and reading of Farlow's chloride reagent film

#### (a) Preparation of the film

A chloride reagent film prepared by Farlow's method (Farlow [1954, 1957]) was conveniently used with the impactor. Our preparation procedure is as fol-

lows. Commercial Fuji Gravure Film Normal (cabinet size) is fixed to get a colorless transparent film with gelatin layers on both sides. The film is washed with distilled water and dried. It is dipped into a solution of 0.5% sodium dichromate and 2% polyvinyl alcohol in distilled water for 2 min, drained for 15 sec, dipped into a solution of 5% silver nitrate (70 cc) for 2 min, drained for 15 sec, coarse sediment on the surfaces is rubbed off with a soft sponge in distilled water for 30 sec and it is dried in a dark box: we then get a gelatin film containing reddish brown sediment of silver dichromate. When sea-salt particles are collected on the surface, white halos of silver chloride are developed, the diameters of which correspond to the salt masses.

The temperature at the time of preparation seems to affect the nature of the film obtained. Room temperature of 15°C or somewhat cooler seems preferable. The film obtained is slightly photosensitive and is preferably stored in a dark desiccator.

In the course of preparation, patterns of a different depth of color are apt to develop. We consider the error caused by the lack of uniformity in the quality of the film obtained. Assuming that the depth of color is proportionate to the amount of sediment of silver dichromate contained in a unit area of the film, we here consider a case where the depth of the color is  $a$  times deeper than the standard depth. In this case the area of the halo must be  $1/a$  of the standard case. If we measure this halo as that of the standard case, we obtain a value of the volume smaller by a factor of  $1/a$ . If we express the dry mass of the particle in  $\log m$  ( $m$  in  $10^{-12}$  gm unit), the error will be  $-\log a$ . In Table 1 the error for a few values of  $a$  is shown.

Table 1. Errors appearing in measured salt masses when the depth of color of the prepared reagent film is  $a$  times deeper than the standard.

$a$	1.0	1.1	1.2	1.5	2.0
Error in $\log m$ ( $m$ in $10^{-12}$ gm unit)	0	0.041	0.079	0.18	0.30

(b) *Processing of the reagent film after sampling particles*

After sampling the sea-salt particles on the reagent film, water vapor should be applied in order to fully develop white halos of silver chloride. During this procedure, one should take care not to expose the film to fog particles. For this purpose, it is recommended that the film be put into a glass desiccator containing a low percentage salt solution, which keeps the inside of the desiccator a few % below 100% relative humidity, to prevent fog formation due to

change in the room temperature, for one day or so.

When the halos have fully developed, they look like homogeneous white disks. Then it is essential to cover the film with a transparent coating by dipping the dried film into a solution of collodion of about 2%, in order to prevent

further contamination by salt particles. If we store the film in a dark place, e.g. in an opaque polyethylene bag, the halos will remain unaltered for several years.

(c) *Calibration and reading of the reagent film*

For the calibration of the relation between the dry masses of sea-salt particles and the sizes of the halos, a method which seems most reliable is the isopiestic method (cf. Toba, [1966]). Droplets of NaCl solution are sprayed on a hydrophobic surface, and placed in an isopiestic method control chamber, and a microphotograph is taken. Then the reagent film is brought into contact with the hydrophobic

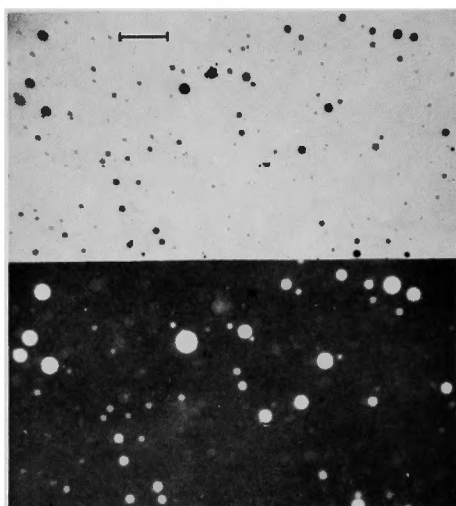


Fig. 3. An example showing the calibration procedure of the reagent film. Upper: particles in the control chamber of the isopiestic method. Lower: halos developed on the reagent film by the same particles. One-to-one correspondence is seen. The line entered indicates  $100\mu$ .

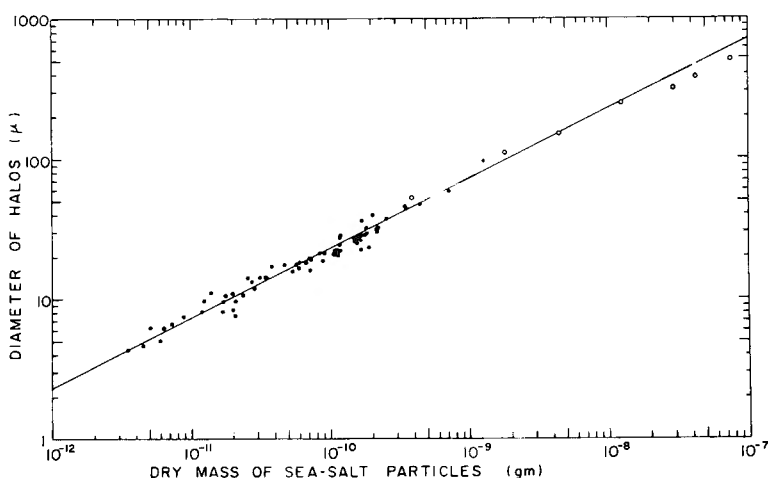


Fig. 4. Calibration of the chloride reagent film. Black circles are the calibration by use of the isopiestic method, and open circles those reported in Toba and Tanaka [1963].

surface to transfer particles onto the film. Thus there is a one-to-one correspondence between the mass of chloride determined by the isopiestic method and the diameter of the halo. Fig. 3 shows an example of the one-to-one correspondence. As the hydrophobic surface, the same transparent film as that used to prepare the reagent film, coated with silicone oil may be successfully used.

Fig. 4 shows the result of the calibration for our film. The straight line entered is

$$\log m = 2 \log D - 0.714,$$

where  $m$  is the dry mass of sea-salt particles in  $10^{-12}$  gm unit, and  $D$  diameter of the halos in  $\mu$ . By use of our film, we may determine the mass of sea-salt particles larger than  $10^{-11}$  gm safely, and may count the particles larger than about  $10^{-11.5}$  gm.

The reading of the halos is easily performed by use of a transparent scale shown in Fig. 5, on  $\times 100$  enlarged prints of microphotographs. A moving microscope with  $1\mu$  vernier is also used for counting halos directly from the

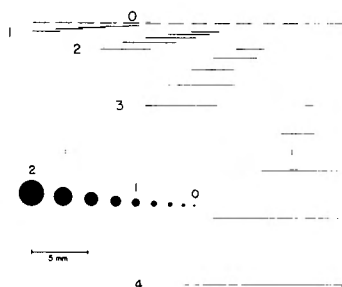


Fig. 5. Scale for reading the halos from  $\times 100$  enlarged prints of the microphotograph. The numbers represent the value in  $\log m$ .



Fig. 6. An example of the halo area on the reagent film provided by the impactor. The line entered indicates  $100\mu$ .

Table 2. Mass distribution of sea-salt particles read off from Fig. 6.

Mass range of dry particles (log $m$ ; $m$ in $10^{-12}$ gm unit)	0.5		1		2				3				4	
Number in 4 l sample	(74)	(59)	31	18	11	8	6	2	0	1	0	0	0	0

gelatin film.

In Fig. 6 is shown an example of the halos obtained by use of the impactor on the reagent film. It is a 4-liter sampling of natural air in Kyoto on the afternoon of June 20, 1963. The mass distribution obtained is shown in Table 2.

It should be noted here that, if we use, with the impactor, the transparent film coated with silicone instead of the reagent film, we may measure much smaller particles including other hygroscopic particles than sea-salt ones, by use of the isopiestic method.

### References

- Farlow, N. H., 1954; An improved halide ion-sensitive sampling surface for water aerosols, *Rev. Sci. Instr.*, **25**, 1109-1111.
- Farlow, N. H., 1957; Quantitative determination of chloride ion in  $10^{-6}$ -to  $10^{-12}$ -gram particles, *Anal. Chem.*, **29**, 883-885.
- May K. R., 1945; The cascade impactor. An instrument for sampling coarse aerosols, *Jour. Sci. Instr.*, **22**, 187-195.
- Ranz, W. E., and J. B. Wong, 1952; Impaction of dust and smoke particles, *Ind. Engr. Chem.*, **44**, 1371-1381.
- Toba, Y. and M. Tanaka, 1963; Study on dry fallout and its distribution of giant sea-salt particles in Japan, *Jour. Met. Soc. Japan*, Ser. II, **41**, 135-144.
- Toba, Y., 1966; Critical examination of the isopiestic method for the measurement of sea-salt nuclei masses, *Special Contr., Geophys. Inst., Kyoto Univ.*, **6**, 59-67.