Sea salt particles transported to the land

By SHIZUO TSUNOGAI, *Department of Chemistry, Faculty of Fisheries, Hokkaido University, Hakodate, Japan*

(Manuscript received January 28, 1974)

ABSTRACT

Large quantities of sea salt particles are suddenly transported to the land by the discrete events of a violent storm. In Japan these events are the typhoon in summer on the Pacific coast region and the north-west monsoon in winter on the Japan Sea coastal region. The share of "sedimentation", one of the mechanisms of the transport of sea salt particles to the land, is only about 20 % of "precipitation" over various parts of Japan. The contribution of "impaction" with surface obstacles is negligibly small, as observed in the example of the Ogōchi Basin. This conclusion was also confirmed by analysing the concentration of chloride in rivers undisturbed by other sources. Moreover, the analysis reveals the following facts. The half-decrease distance of the concentration is about 20 km from the coast in both coastal regions of Japan. The total deposition of sea salt was calculated to be 2.7×10^{12} g/yr in the whole of Japan (area 3.7×10^5 km^2) and the mean concentration of chloride in rain to be 2.4 ppm over Japan as a whole, but the concentration is about 3 times larger over the Japan Sea coastal region than over the Pacific coast region, owing to the violent north-west monsoon in winter.

Introduction

Some attempts to estimate the amount of sea salt transported from sea to land have been made. Toba (1965*a*, *b*) analysed the variation in the vertical profile of sea salt particles over land and concluded that the scavenging of surface obstacles may be an efficient process, although it was not directly comparable to the rainout process.

Eriksson (1960) estimated the proportion of wet removal (precipitation) of chloride in the atmosphere and dry removal (sedimentation and impaction), using an observational network over Scandinavia. His results show that impaction is dominant and precipitation accounts for less than 30% of total fallout.

Sugawara (1964) found that dry fallout accounts for two-thirds of total deposition, by calculating the total deposition from the balance between river discharge (mean concentration of chloride in Japanese rivers, 5.2 ppm) and the other sources on land such as hot and mineral springs and artificial sources, and comparing it with wet precipitation (mean concentration of chloride in rain over Japan, 1.1 ppm).

Gorhem (1961), however, suggested that dry fallout is of little importance, finding that the amounts of chloride in precipiatation tallied well with river discharges of chloride in Australia.

It is difficult to estimate the absolute amounts of total deposition from Toba's calculation, since his method needs some assumptions which largely affect the absolute value and he has used number concentration in his equation instead of mass concentration. Moreover, the balance calculation, using concentrations in river and rain, is apt to cause large errors in the estimates of the mean concentration and of the amounts supplied from sources other than meteoric precipitation.

First of all, therefore, I will discuss the importance of discrete events for the transport of sea salt particles. Secondly, I will show the minor importance of dry fallout, using results of observations for one year at Ogōchi Basin where the supply of salts from sources other than air-borne is little and the amounts of water discharged are well known because the dam at the opening of the basin has been constructed to supply Tokyo with water. Fi-

Table 1. *Fallout of chloride in precipitation including sedimentation in the various parts of Japan*

Location	Distance from the coast (km)	Level above the sea (m)	Year[a]	Annual fall ($g/m^2/yr$)	Partial fall: Event or period	Partial fall: g/m^2
The Pacific coast region						
Kawane, Shizuoka	25	190	1963	3.49		
			1964	2.80	Typhoon No. 20	0.87
					In Sept.	1.06
Koenji, Tokyo	12	40	1964	2.14	Typhoon No. 20	0.22
				1.78[b]	In Sept.	0.38
			1965	—	Typhoon No. 24	0.79
Saku, Nagano	150	650	1964	—	Typhoon No. 20	0.010
Taba I, Yamanashi	70	620	1964	0.84_5	In Sept.	0.11_1
				0.71[b]		
Taba II, Yamanashi	70	1 000	1964	0.70	In Sept.	0.07_2
The Japan Sea side region						
Shishiku, Ishikawa	15	600	1966	—	Feb. 3–Feb. 6	0.98
						0.74[c]
Assabu, Hokkaido	20	95	1970		May–Aug.	1.41
				10.52 (Mean)		(Mean)
			1971		Nov.–Feb.	6.45
						(Mean)

[a] From April to March.
[b] Excluding sedimentation.
[c] Deposition from 21.00 on Feb. 3 till 04.00 on Feb. 4.

nally, I will re-examine the river discharges using the data from Japanese small rivers which are not greatly affected by any land sources such as hot springs, industries, or other human activities.

In this article, I tentatively assume that the chemical composition of sea salt particles is the same as that of sea water and that the sources of the atmospheric chloride other than sea salt are negligibly small. That is to say, chloride is considered as the representative of sea salt particles. This is based on the findings of Gorham (1955) and Tsunogai et al. (1972), that the composition of oceanic rain is similar to that of sea water. On the other hand, Komabayashi (1964) observed an enrichment of major cations relative to sodium content in sea water, with an enrichment factor of 1.4 to 2.1 for particles of radius 0.4 μ, which may originate from the film of breaking bubbles, whereas no fractionation was found between major ions for particles of radius larger than 0.8 μ.

Junge (1972) critically reviewed the reports on this subject and stated "fractionation is possible but it may occur only under certain conditions, of which the presence of organic film is perhaps only one". Gorham (1958) demonstrated the atmospheric pollution by hydrochloric acid. Tsunogai et al. (1972) found an occasional contribution of chlorine other than sea salt in air and rain samples, especially over industrial areas, whereas the degree of the contribution in rain over rural area is very small, compared with that of sea salt particles. Thus, the assumption is practically valid and the amount of sea salt is obtained by multiplying 1.8 by that of chloride.

I. Events affecting the supply of sea salt particles

Amounts of wet deposition including sedimentation were estimated by determining the chloride concentrations in the samples taken at various stations in Japan. Cylindrical samplers made of glass or plastic were placed 1–2 meters above level ground. The precipitation samples, including dry sedimentation, were collected every month or at certain periods, and analysed

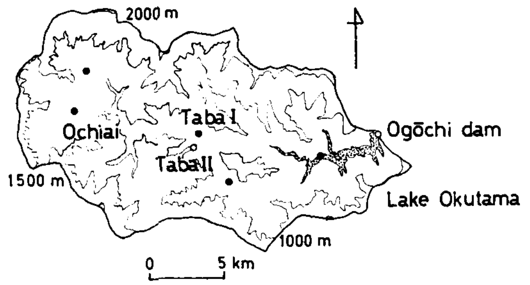

Fig. 1. Ogōchi Basin. Solid circles show locations where the amounts of rainfall were determined.

for dissolved major cations and anions. The method and analytical data were reported in detail by Tsunogai (1966) and Tsunogai et al. (1972). The summarized results are shown here in Table 1.

In the Pacific coast region of Japan, the annual deposition rate decreases with increasing distance from the coast. A remarkable feature is found when the annual deposition rate is compared with the partial deposition rate. At Kawane (Shizuoka Prefecture) in 1964, for example, about one-third of the annual deposition was transported by one typhoon alone, Typhoon No. 20, 1964. The phenomenon seems to be rather local, as the amount deposited by the same typhoon at Koenji, Tokyo, was not so large and was negligibly small at Saku, Nagano Prefecture, inland area of Japan. The event of a concentrated deposition, however, may be not so rare, as in the following year the amount of sea salt transported to Koenji by Typhoon No. 24, 1965, is nearly the same as that by Typhoon No. 20, 1964, at Kawane.

In contrast, large quantities of sea salt were carried down to the coastal region of the Japan Sea in winter, when north-west monsoon prevailed. At Assabu, Hokkaido, the amount of deposited chloride was several times larger than that at the Pacific coast region, 60% of which fell during 4 months in winter (Table 1). A remarkable event, which may occur not so rarely, was the strong north-west wind lasting only 7 hours, but which conveyed 0.74 g/m^2 of chloride together with 15 g/m^2 of soil dust (loess from the continent) to Shishiku, Ishikawa Prefecture and consequently the snow was colored (Table 1).

These facts suggest that a considerable quantity of sea salt particles is transported from sea to land by the discrete events of a violent storm rather than by continuous events. According to Tsunogai et al. (1972), chloride in oceanic rain or maritime air increases by a factor of 10 with each 4 to 7 m/sec increase of wind velocity. As discussed by Toba (1965*b*), the more windy it is, the more sea salt particles are produced and the farther they are transported. The events of storm are more frequent in the Japan Sea region in winter than in the Pacific coast region in summer. This is the reason why snow in Japan generally contains more sea salt than does rain in summer. It is another conclusion that a mean calculated arithmetically from concentrations in normal rain may be an underestimated value.

II. An estimate of sea salt particles removed by impaction

As stated previously, a proportion of the sea salt particles is removed from the atmosphere by impaction with surface obstacles such as vegetation and buildings. It is not easy to determine quantitatively the contribution of impaction. Therefore, Ogōchi Basin (drainage area, 263 km^2) was chosen as an experimental field. It is located 70 km west of central Tokyo. As shown in Fig. 1, three observation stations were established in the basin. Observation was started on April 1, 1964 and ended on May 31, 1965. Sampling was carried out once a month at Taba I, where samples of precipitation including or excluding dry sedimentation and river water were collected, and at Taba II where samples of precipitation including sedimentation were collected. These samples were analysed for some major cations and anions. A detailed description of the method was given by Tsunogai (1966).

The annual budget of chemical constituents in the Ogōchi Basin is calculated as shown in Table 2. The amount of chloride discharged was determined by multiplying the amount of water discharged by its chloride concentration, and adding the amount of chloride in the increased storage water at the start and end of the observations. The chloride deposition was calculated by integrating the products of the area of each 100 m contour interval and the assumed deposition at its level from the straight line in Fig. 2. This assumption is strengthened

Table 2. *A budget of chemical constituents in the Ogôchi Basin*[a]

	Rainfall (mm/yr)	Cl	SO_4	Na	Ca	Mg	K	F
From the dam	753	189	1 453	544	1 618	157	139	17.6
Discharged amount	675	167	1 397	490	1 421	141	125	16.0
Change in storage amount	78	22	156	54	197	16	14	1.6
To the dam	753	189	1 453	544	1 618	157	139	17.6
Fallout		169	699	73	124	28	43	10.0
Precipitation	1 082	145	436	59	53	20	25	6.9
Sedimentation	−329 (Evaporn)	24	263	14	71	8	18	3.1
Impaction		~0	—	—	—	—	—	—
Other		20	754	471	1 494	129	96	7.6
Cf. common salt used	—	35	—	23	—	—	—	—

[a] Unit in tons/yr except for rainfall.

by the following fact. The line connecting two points of deposition at Taba I and Taba II fits well with the estimated deposition in the Taba river basin (mean height, 1 360 m), which is obtained from the mean concentration of chloride in Taba river (0.64 ppm), the mean rainfall (1 229 mm/yr at Ochiai) and the mean evaporation (329 mm/yr in the whole basin). The proportion of sedimentation to total fall in the whole basin is assumed to be the same as that at Taba I (16% for chloride).

The difference between the discharged amount at the dam and the deposition in the basin (Table 2) is due to impaction and other sources in the basin such as human activities and the dissolution of soil. For the case of chloride, the difference is only 20 tons/yr in the whole basin, which is nearly equal to the amount of common salt used by people in the basin. According to the Japan Monopoly Corporation, Japanese use 30 g salt/person·day or a mean consumption of 11 kg salt/person·yr for food, and the population in this basin was 5 300 at that time. Impaction, therefore, does not contribute significantly to the total deposition. I think this conclusion does not necessarily contradict that of Toba (1965*a*) who has predicted the contribution of impaction, since the sea salt particles which fell in the basin were transported by air over mountains higher than 1 000 m. The extent of impaction for other chemical constituents is obscure, owing to the extensive interaction with earth's surface after rainfall.

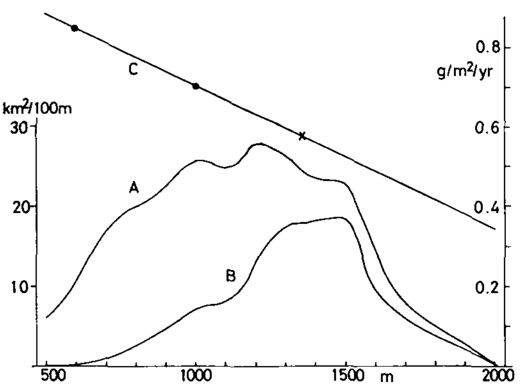

Fig. 2. The height distributions of drainage area of Ogōchi Basin and the assumed deposition rate of chloride. *A* and *B*, the height distributions of the whole basin and of the Taba river basin (upstream from Taba I) in km²/100 m height, respectively. Solid circles are deposition rates observed at Taba I and Taba II and a cross marks that estimated from chloride in Taba river water.

III. Chloride in small rivers reflecting the relation between sea salt particles and distance from the coast.

As shown in Table 2, air-borne sea salt in river water is the predominant source for the supply of chloride to the water, provided there are not many hot and mineral springs, surface areas covered by freshly deposited volcanic ash, factories, or inhabitants in the basin. I therefore chose 34 small rivers in Japan from 225 rivers where the chloride concentration has

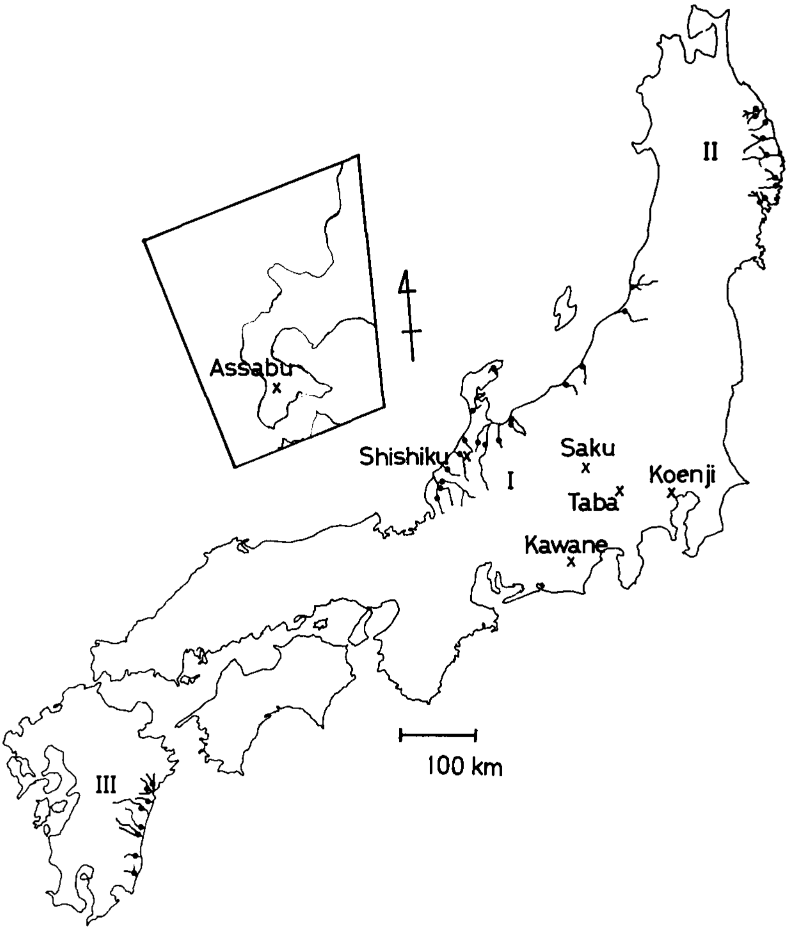

Fig. 3. Locations of rivers used in the analysis. Solid circles show the sampling stations.

been determined by Kobayashi (1960) (Fig. 3). He took monthly samples during the period 1943 to 1957. The mean concentrations of chloride are shown in Table 3. These rivers satisfy the above conditions and their direction of flow are almost parallel with the direction of the monsoon. The rivers chosen from the Japan Sea coastal region are situated in the central part of Japan and those from the Pacific side are situated in the northern and southern parts of Japan. The results of the following analyses obtained from these rivers must be regarded as an approximate mean for the whole of Japan.

A part of the chloride in river water is derived from salt used for food, which can be corrected by the following equation

$$\Delta Cl = P/F \times 6.1$$

where ΔCl is the chloride concentration in ppm which is supplied from sea salt used by human beings at a rate of 30 g NaCl/person·day, P is the population density in person/km^2 and F is the flow rate of river water in mm/yr. On the basis of the mean precipitation estimated by Ishihara (1972), F is assumed to be 2 200 mm/yr for the Japan Sea coast region and the southern part of the Pacific coast region and 900 mm/yr for the northern part of the Pacific coast region. The corrected concentrations of chloride are also given in Table 3.

The river drainage area is cut into two exact halves by drawing a line parallel to the coast line, or perpendicular to the direction of the

Table 3. *Concentrations of chloride observed and corrected for the common salt consumed and the distance from the coast for 35 Japanese rivers which are used in the analysis*

Name of river[a]	Cl observed (ppm)	Cl corrected (ppm)	Mean distance from the coast (km)
I. *The Japan Sea coast region*			
1. Miomote	7.4	7.3	20
2. Kaji	7.5	7.4	24
3. Sabaishi	8.0	7.7	16
4. Kokura	7.7	7.4	19
5. Katakai	2.1	2.0	69
6. Hayatsuki	2.4	2.3	69
7. Jinzū	2.4	2.2	60
8. Shō	3.2	3.1	54
9. Oyabe	5.9	5.7	29
10. Wakayama	17.3	17.1	5
11. Hakui	15.2	14.6	12
12. Sai	6.7	6.4	21
13. Tedori	6.7	6.6	30
14. Daishoji	7.3	7.1	18
15. Kuzuryū	3.6	3.4	51
16. Asuwa	5.4	5.3	35
17. Hino	6.2	6.1	20
II. *The Pacific coast region—northern*			
1. Kuji	4.2	4.0	19
2. Osanai	3.6	3.3	18
3. Akka	2.3	2.3	24
4. Omoto	2.4	2.3	33
5. Hei	1.7	1.6	36
6. Ohzuchi	3.1	2.6	21
7. Unosumai	2.3	2.0	23
8. Sakari	3.6	3.4	18
9. Kesen	3.4	3.1	25
III. *The Pacific coast region—southern*			
1. Kita	3.1	3.0	23
2. Hōri	2.9	2.8	19
3. Isuzu	3.4	3.3	17
4. Mimi	1.7	1.6	39
5. Omaru	2.5	2.3	26
6. Hitotsuse	1.8	1.7	35
7. Kiyotake	4.2	3.8	13
8. Sakatani	3.8	3.7	15

[a] The rivers are numbered in order from north to south, as shown in Fig. 3.

monsoon. The distance from the coast to this line is defined in Table 3 as the mean distance from the coast.

As shown in Fig. 4, the plot of logarithms of the corrected chloride in river water versus mean distance from the coast yields a straight line for rivers in nearly the same meteorological and topographical conditions. This fact can be explained as follows. If sea salt is removed from maritime air in proportion to its concentration in the atmosphere, the concentration of chloride in the air transported to the land is given by

$$dC/dZ = -\lambda C$$

$$\therefore C = C_0 \exp(-\lambda Z)$$

where C is the concentration of chloride in the air, C_0 is that at the coast, Z is the distance from the coast and λ is the removal rate of chloride from the atmosphere. Though this assumption may not be the case for every rainfall, it may well be valid for a mean value obtained statistically from a long period observation. Therefore, the concentration of chloride in precipitation or in river water is considered to be proportional to its concentration in the air, which decrease with the rate λ, from the coast, as written by

$$Y = fC$$

$$Y = Y_0 \exp(-\lambda Z)$$

where Y is the concentration of chloride in precipitation or in river water, $Y_0(=fC_0)$ is that at the coast and f is the proportionality constant converting the dimension at the same time. For the case of river water, Z is approxi-

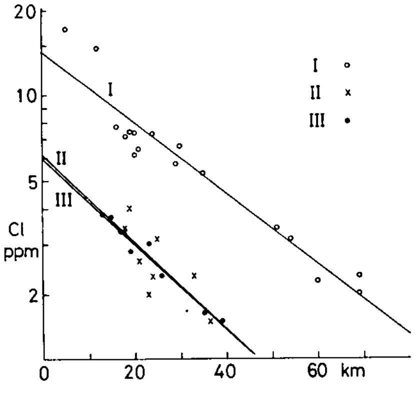

Fig. 4. The concentration of chloride in river water versus the mean distance from the coast. I, II and III in the figure are the indexes of rivers in the Japan Sea, the northern part of the Pacific and the southern part of the Pacific coast regions, respectively.

Table 4. *Calculated concentration at the coast* (Y_0) *and decrease rate constant* (λ) *of chloride in Japanese rivers*

	No. of rivers	Y_0 (ppm)	λ (km^{-1})	Half-decrease distance (km)
The Japan Sea coast region				
I	17	14.1	0.0288	24
The Pacific coast region				
II	9	6.0	0.0345	20
III	8	6.1	0.0352	20
Mean		6.1	0.0349	20

mately equal to the mean distance from the coast, estimated from the drainage area upstream from the sampling point.

The values Y_0 and λ are determined from the intercept and gradient, respectively, of the straight line in Fig. 4, and which are reproduced in Table 4. These values of the Pacific side of northern Japan agree closely with those of southern Japan. Thus, mean values of these two regions are regarded as representative of the Pacific side of Japan. When the regional differences of the Pacific side and the Japan Sea side are compared, Y_0 is remarkably larger on the Japan Sea side, but λ is nearly equal. This seems to indicate that the Japan Sea in winter is more violent than Pacific in summer and that sea salt particles are removed from the atmosphere by nearly the same process on both sides of Japan. Of course, the half-decrease distance ($0.693/\lambda$) may be influenced by many factors, such as topography. The distance, therefore, may be larger than 20 km on the plains.

Next, the deposition of chloride is calculated in the following way.

Total deposition in Japan (g/yr)

$$= a\int_0^b cY\,dZ = acY_0\int_0^b e^{-\lambda Z}dZ$$

Mean deposition ($g/m^2/yr$)

$$= c\int_0^b Y/b\,dZ$$

Mean concentration in precipitation (ppm)

$$= c\int_0^b Y/bd\,dZ$$

where a is the length of the coastline in meters, b the distance in meters from the coast to the central ridge which cuts Japan into two almost exact halves, and c the mean flow rate of rivers (including underground water) in $m^3/m^2/yr$, which is obtained by subtracting evaporation from rainfall. For simplicity in calculating, the shape of Japan (its area, 3.7×10^5 km^2) is approximated by two narrow rectangles of equal area, divided by the central ridge, and the small regional differences in mean rainfall and mean evaporation between the Japan Sea side and the Pacific side are neglected on the basis of Ishihara's (1972) estimation. The assumed input data and the calculated results are given in Table 5.

Although sources other than air-borne salt contribute to the above estimates somewhat, the estimated concentrations of chloride in precipitation coincide well with those including sedimentation observed in various parts of Japan (cf. Table 1). This fact also indicates that impaction is not predominant in Japan. The deposition of sea salt particles is about 3 times

Table 5. *Assumed data for the calculation of sea salt particles transported to Japan and the results of the calculation*

	The Japan Sea coast region	The Pacific coast region	Whole Japan
Input data			
Rainfall (mm)	1 750	1 750	1 750
Evaporation (mm)	450	450	450
Length of the coastal line (km)	1 850	1 850	—
Integrated distance from the coast (km)	100	100	—
Results of the calculation for chloride			
Total fallout (10^{12}g/yr)	1.1	0.4	1.5
Mean fallout ($g/m^2/yr$)	6.0	2.2	4.1
Mean concentration in precipitation (ppm)	3.45	1.25	2.35

greater on the Japan Sea side than on the Pacific side. The total deposition of air-borne sea salt in Japan is one order of magnitude larger than common salt used for food. In the developed area of Japan, however, salt used for food and industry is equal in amount to, or more than, the air-borne sea salt. Thus, large errors would be introduced if the contribution of impaction were estimated by analysing data on rivers in developed areas, where the sampling is easier than in the undeveloped areas.

Acknowledgements

I would like to thank Prof. M. Nishimura for his helpful suggestions. I am also indebted to Mr S. Nakaya who analysed the concentration of chloride in some rain samples and to Miss M. Nambu who estimated the mean distance of the rivers from the coast.

REFERENCES

Eriksson, E. 1960. The yearly circulation of chloride and sulfur in nature, meteorological, geochemical and pedological implications. Part II. *Tellus 12*, 63–109.

Gorham, E. 1955. On the acidity and salinity of rain. *Geochim. Cosmochim. Acta 7*, 231–239.

Gorham, E. 1958. Atmospheric pollution by hydrochloric acid. *Quart. J. Roy. Meteorol. Soc. 84*, 274–276.

Gorham, E. 1961. Factors influencing supply of major ions to inland waters with special references to the atmosphere. *Bull. Geol. Soc. Am. 72*, 795–840.

Ishihara, K. 1972. An estimate of mean rainfall amount in Japan. *Environmental Information Sciences (Kankyo Jōhō Kagaku) 1*, 15–20.

Junge, C. E. 1972. Our knowledge of the physicochemistry of aerosols in the undisturbed marine environment. *J. Geophys. Res. 77*, 5183–5200.

Kobayashi, J. 1960. Average composition and its features of Japanese rivers. *Studies on Agriculture (Nōgaku Kenkyū) 48*, 63–106.

Komabayashi, M. 1964. Primary fractionation of submicron spray drops from sea salt solution. *J. Meteorol. Soc. Japan 42*, 309–316.

Sugawara, K. 1964. Migration of elements through phases of atmosphere and hydrosphere. *J. Water and Waste (Yōsui to Haisui) 6*, 317–321.

Toba, Y. 1965*a*. On the giant sea salt particles in the atmosphere. I. General features of the distribution. *Tellus 17*, 131–145.

Toba, Y. 1965*b*. On the giant sea salt particles in the atmosphere. II. Theory of the vertical distribution in the 10-m layer over the ocean. *Tellus 17*, 365–382.

Tsunogai, S. 1966. Chemical study of precipitation: Concentration, fall rate and sources of chemical constituents in precipitation and air-borne dust. Thesis for doctorate submitted to Tokyo Kyoiku University, p. 226.

Tsunogai, S., Saito, O., Yamada, K. & Nakaya, S. 1972. Chemical composition of oceanic aerosol. *J. Geophys. Res. 77*, 5283–5292.

ЧАСТИЦЫ МОРСКОЙ СОЛИ, ПЕРЕНОСИМЫЕ НА ЗЕМЛЮ

Большие количества частиц морской соли иногда внезапно приносятся на землю сильными штормами. В Японии примерами таких событий служат тайфуны летом в областях, обращенных к Тихому океану и северо-западные муссоны зимой в областях побережья Японского моря. Доля «седиментации», являющейся одним из механизмов переноса частиц морской соли на землю, составляет лишь около 20% «осадков» в различных частях Японии. Вклад «соударений» частиц с препятствиями на поверхности также пренебрежимо мал, как это наблюдается на примере бассейна Огочи. Этот вывод также подтверждается анализом концентрации хлоридов в реках, не подвергшихся влиянию других источников. Более того, анализ выявляет следующие факты. Расстояние, на котором концентрация частиц уменьшается наполовину, составляет порядка 20 *км* от берега для обеих береговых областей Японии. Общее выпадение морской соли вычислено равным $2{,}7 \cdot 10^{12}$ грамм в год для всей Японии (площадь $3{,}7 \cdot 10^{5}$ *км*^{2}) и средняя концентрация хлоридов в дождях найдена равной $2{,}4 \cdot 10^{-6}$ для всей страны, однако эта концентрация примерно в 3 раза больше на побережьи Японского морчя, чем на побережьи Тихого океана, что имеет место благодаря силе северо-западного муссона зимой.