The Chlorine Content of Hawaiian Lavas

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Studies of the chlorine content of Hawaiian llavas have been published by Shepherd,1) Iwasaki et al.,2) and Yoshida et al.3) Nevertheless, the rock samples used in their studies were small in number and collected from only a few localities. Recently, 141 new chemical analyses of Hawaiian lavas systematically collected have been reported by Macdonald and Katsura.4)

In the present paper, the distribution of chlorine in the Hawaiian lavas will be investigated in order to answer the following principal questions: Is there any change in the chlorine content of rocks which were extruded at different times in volcanic history? Is it clear that chlorine tends to be enriched in rock of an alkalic type? Does the frequency distribution of chlorine show a lognormality? Is there any regional change in the chlorine content of the rocks?

Experimental

Rock Specimens.-The rock specimens used for the present study cover all of the rock types found in the Hawaiian Islands, e.g., tholeiitic basalt, tholeiitic olivine basalt, alkalic basalt, alkalic olivine basalt, basanite, basanitoid, ankaramite, hawaiite, mugearite, and trachyte, as defined by Macdonald and Katsura.4) Most of them were collected by Macdonald during the 1960~1961 period, during which time one of the authors of this paper joined him under a National Science Foundation .grant.5,6)

Method of Analysis. - The total chlorine was determined by the method of Iwasaki et al.7) with some improvements. About 200 mg. of a fine powdered rock sample was fused with anhydrous sodium carbonate in a platinum crucible. After cooling, the chlorine was completely extracted with hot redistilled water and was determined photometrically by the thiocyanate method.8)

The water-soluble chlorine in rocks was determined as follows: About 1.00 g. of a fine powdered (100 to 150 mesh) rock sample was put into a centrifugal vessel containing 15 ml. of redistilled water. The vessel was then vigorously shaken about 1 min. After the mixture had stood overnight at room temperature, the solution in the vessel was separated from the rock sample by centrifuge. Almost all of the soluble chlorine was extracted by this procedure. About 0.1 ml. of concentrated perchloric acid was then added to the solution. The total soluble chlorine thus extracted was determined by the method shown above.

The total chlorine in approximately 160 Hawaiian volcanic rocks was duplicatedly determined two or more times, but the water-soluble chlorine was determined only one or two times. It may safely be said that the precision for the present determination of chlorine is within $\pm 5\%$ over the whole range of chlorine contents obtained here.

In addition to the reproducibility of the results of the analyses, a cross-check analysis of one sample was made by Dorothy Powers, with the aid of Dr. H. A. Powers and under the supervision of L. C. Peck, in the laboratory of the Geological Survey in Denver, Colorado, U.S.A. The value of 0.018% chlorine in a rock specimen, G-2907 in Table I-b obtained by the present authors is in good agreement with that obtained by Mrs. Powers, (0.02% chlorine).

According to Shepherd,1) the chlorine content in the 1926 Mauna Loan volcanic rocks was 0.01% chlorine; as may be seen in Table I-b, the corresponding value for the same rock obtained by the present authors was 0.008% chlorine (C-133). Taking account of the difference in rock specimens, it may be said that these values are in good accordance.

Results

Tables I-a, I-b give both the insoluble and total chlorine contents in the Hawaiian lavas. The insoluble chlorine content was calculated by subtracting the water-soluble chlorine from the total chlorine.

Discussion

Water-soluble Chlorine in Rocks.—It has been found by Kuroda and Sandell9) and by Iwasaki

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⁵⁾ C. A. Macdonald and T. Katsura, Pacific Science, 15, .358 (1961).

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7) I. Iwasaki, T. Katsura and N. Sakato, J. Chem. Soc.

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⁸⁾ A. Tomonari, ibid., 83, 693 (1962).

⁹⁾ P. K. Kuroda and E. B. Sandell, Bull. Geol. Soc. Am., 64, 879 (1953).

TABLE I-a. CHLORINE CONTENT OF THE VOLCANIC ROCKS IN THE ISLANDS OF HAWAII

Speci- men	Volcano	Type of rock	Total Cl	Insoluble Cl, %	Speci- men	Volcano	Type of rock	Total Cl	Insoluble Cl, %
C-1	Waianae	Tb	0.010	0.009	C-57	Kohala	Tb	0.008	0.006
C-2		Tb	0.008	0.007	C-58		Tb	0.007	0.006
C-3		Tb	0.015	0.014	C-59		Tb	0.007	0.006
C-4		Tb	0.009	0.007	C-60		Tb	0.006	0.005
C-5		Tb	0.019	0.007	C-61		Tb	0.010	0.004
C-6		Tb	0.010	0.006	C-62		Tb	0.006	0.005
C -7		Tb	0.010	0.006	C-63		Tb	0.012	0.003
C -8		Tb	0.007	0.006	C-64		Tb	0.007	0.006
C-9		Tb	0.011	0.010	C-65		Fpb	0.006	0.004
C-10		Tb	0.010	0.009	C-66		Tb	0.007	0.007
C-11		Tb	0.008	0.006	C-70		Aob	0.008	0.007
C-12		Тb	0.019	0.006	C-69		Aob	0.007	0.005
C-13		Tb	0.009	0.007	C-68		Ha	0.037	0.034
C-14		Tb	0.008	0.007	C-134		Mu	0.008	0.006
C-15		ТЬ	0.008	0.006	C-77	Mauna Kea	Aob	0.050	0.010
C-16		Tb	0.012	0.010	C-78		Aob	0.062	0.007
C-17		Tb	0.012	0.008	C-76		Ak	0.010	0.008
C-18		Tb	0.017	0.008	C-75		Tob	0.007	0.006
C-19		Tb	0.008	0.007	C-74		Tb	0.007	0.005
C-20		Tb	0.012	0.007	C-71		Ak	0.011	0.009
C-21		Tb	0.008	0.006	C-67		Ak	0.010	0.009
C-22		Tb	0.026	0.006	C-72		Ha	0.024	0.021
C-23		Tb	0.011	0.008	C-73		Ha	0.008	0.006
C-24		Tb	0.019	0.011	C-79		Ha	0.009	0.007
C-25		Tb	0.029	0.007	1902		Ha	0.013	0.012
C-26		Tb	0.012	0.008	TK8200		Aob	0.009	0.007
C-27		Tb	0.012	0.007	C-80	Kauai	Tb	0.006	0.005
C-28		Tb	0.021	0.007	C-81		Tb	0.007	0.006
C-29		Tb	0.020	0.008	C-82		Tb	0.006	0.005
C-33		Tb	0.158	0.020	C-83		Tb	0.007	0.006
C-34		Tb	0.041	0.006	C-84		Oc	0.007	0.004
C-35		Tb	0.015	0.011	C-85		Tb	0.012	0.010
C-36		Tb	0.015	0.008	C-86		Tb	0.008	0.007
C-37		Tb	0.015	0.008	C-87		Tob_{r}	0.007	0.005
C-38		Tb	0.009	0.007	C-88		Tb	0.008	0.007
C-39		Tb	0.016	0.010	C-89		Tb	0.006	0.005
C-40		Tb	0.016	0.007	C-90		$\mathbf{B}\mathbf{s}$	0.008	0.006
C-41		Tb	0.051	0.013	C-91		Aob,	0.013	0.011
C-46		Tb	0.012	0.010	C-93	West Maui	Tb	0.008	0.007
C-47		Tb	0.009	0.006	C-94		Tb	0.010	0.007
C-48		ТЬ	0.011	0.010	C-95		Tb	0.007	0.006
C-49		Tob	0.023	0.013	C-96		Tb	0.007	0.006
C-50		Tob	0.013	0.010	C-97		Tb	0.020	0.006
C-51		Tb	0.010	0.009	C-98		Tb	0.007	0.006
C-52		Ha	0.008	0.006	C-99		Tb .	0.008	0.007
C-30		Aob	0.007	0.006	C-100		Tb	0.218	0.012
C-31		Fpb	0.006	0.005	C-101		Tb	0.007	0.005
C-31'		G	0.008	n.d.	C-102		Tb	0.006	0.005
C-42	X . 1	На	0.024	0.023	C-103		Tb	0.008	0.007
C-44	Molokai	Aob	0.014	0.010	C-104		Tb	0.011	0.006
C-45	YZ =1: -1	Ha	0.023	0.020	C-105		Tob	0.009	0.007
C-53	Kohala	Tb	0.098	0.006	C-106		Tb	0.006	0.005
C-54		Tb	0.065	0.010	C-107		Tb	0.012	0.009
C-55		Tob	0.006	0.005	C-108		Tob	0.006	0.004
C-56		Tb	0.007	0.006	C-109		Tb	0.017	0.014

TABLE I-a. (Continued)

Speci- men	Volcano	Type of rock	Total Cl %	Insoluble Cl, %	Speci- men	Volcano	Type of rock	Total Cl	Insoluble Cl, %
C-110	West Maui	Tb	0.030	0.025	C-128	West Maui	Tr	0.021	0.001
C-111		Tb	0.013	0.012	C-115		$\mathbf{B}\mathbf{s}$	0.067	0.066
C-112		Tob	0.025	0.012	C-130		Bs	0.042	0.017
C-113		Tob	0.041	0.010	C-123	Haleakala	Oc	0.006	0.006
C-114		Aob	0.017	0.013	C-122		Tob	0.006	0.006
C-117		Tob	0.072	0.019	C-124		Ab	0.009	0.008
C-118		Aob	0.007	0.005	C-125		Aob	0.008	0.007
C-119		Tob	0.017	0.013	C-126		Aob	0.007	0.007
C-120		Tob	0.009	0.006	C-121		Tob	0.008	0.007
C-92		Mu	0.038	0.019	C-127		Ha	0.033	0.031
C-116		Tr	0.009	0.003	C-129		Bt	0.032	0.027

Abbreviation used in Table I has the following meaning: Tb, tholeiite basalt. Tob, tholeiite olivine basalt. Ab, alkalic basalt. Aob, alkalic olivine basalt. Fpb, feldspar phyric basalt. Ha, hawaiite. G, groundmass. Mu, mugearite. Ak, ankaramite. Oc, oceanite. Bs, basanite. Bt, basanitoid. Tr, trachyte.

TABLE I-b. CHLORINE CONTENT OF THE DATED HAWAIIAN VOLCANIC ROCKS

Specimen	Volcano	Date (year)	Type of rock	Total Cl, %	Insoluble Cl, %
2005	Hualalai	1801	Aob	0.015	0.012
2204	Mauna Loa	1843	Tb	0.008	0.006
2202		1855	Tb	0.007	0.006
2003		1859	Tb	0.008	0.007
2015		1868	Tb	0.006	0.005
2201		1881	Tb	0.008	0.007
2014		1887	Tb	0.007	0.005
2203		1899	Tb	0.006	0.005
2013		1907	Tb	0.007	0.006
2012		1907	Tb	0.007	0.006
2010		1919	Tb	0.009	0.009
C-133		1926	Tb	0.008	0.007
2205		1935	Tb	0.008	0.006
2206-a		1935	Tb	0.008	0.007
2206-ь		1935	Tb	0.007	0.005
2007		1950	Tb	0.007	0.007
1803	Kilauea	1750	Tb	0.010	0.009
2103		1921	Tb	0.013	0.011
2104		1954	Tb	0.014	0.012
C-131		1955	Tb	0.017	0.017
C-132		1955	Tb	0.017	0.016
1802		1955	Tb	0.017	0.016
G-2907		1955	Tb	0.018	n.d.
2101-a		1959	Tb	0.011	0.009
2101-b		1959	Tb	0.012	n.d.
Tachylite		1959		0.018	n.d.
Melt		1959		0.016	0.014
1805		1960	Tb	0.016	0.014
1806		1960	Tb	0.018	0.014
1808		1960	Tb	0.015	0.014
Pele's hair		1959		0.016	n.d.

et al.²⁾ that volcanic rocks generally contain a very small amount of water-soluble chlorine compared with amount of total chlorine when the specimens are fresh. Table II shows the amounts of both the total and the water-soluble chlorines of the historic fresh lavas from Mauna Loa and Kilauea. The amount of water-soluble chlorine varies from 0.000 to 0.004% chlorine, as may be seen in Table II,

TABLE II. VARIATION OF CHLORINE CONTENT IN ROCK SAMPLES AT DIFFERENT PERIODS OF AN ERUPTION

	Total	Insoluble
	Cl %	Cl %
The 1955 Kilauea	Eruption (Puna)	
C-131	0.017	0.016
C-132	0.017	0.016
G-2907	0.018	n.d.
1802	0.017	0.016
The 1960 Kilauea I	Eruption (Kapoh	0)
1805	0.016	0.014
1806	0.018	0.014
1808	0.015	0.014
The 1935 Mauna L	oa Eruption	
2205	0.008	0.006
2206	0.008	0.007
The 1959 Kilauea I	ki Eruption	
2101	0.011	0.009
2101-a	0.012	n.d.

while the total chlorine content has a slightly wider range, from 0.006 to 0.018% chlorine. The amount of water-soluble chlorine is presumably independent of the total chlorine content, and it seems to be almost constant. On the other hand, a rock sample such as C-33 has a high amount of water-soluble chlorine (0.16% chlorine). The specimen was collected about 2 feet above sea level at the end of a spur; as has been pointed out by Macdonald the specimen has without doubt been washed with sea-water, but it appeared very fresh. Considering this, a rock specimen with a high amount of water-soluble chlorine has possibly been contaminated by sea-water, etc. Indeed, there was only a very small amount of water-soluble chlorine even in a fresh a rock specimen, and we can hardly distinguish the chlorine which was primarily present in a rock sample just after the extrusion from the chlorine due to contamination; however, we may presume that the contaminated chlorine could be completely extracted with water. In the following discussion we will employ the amount of insoluble chlorine, though it is tedious work to determine the soluble chlorine as well as the total chlorine.

The Manner of Occurence of Chlorine in Rocks.—Based on the studies by Shepherd.¹⁾ Behne,10) and Kuroda and Sandell,9) Correns11) summarized the manner of occurence of chlorine in igneous rocks, and pointed out that in all probability the chlorine content of igneous rocks does depend either upon their mineralogical composition or upon their modes of crystallization. Iwasaki et al.2) determined the chlorine content of about 190 volcanic rocks from Japan, Korea, Manchuria (China), and Hawaii, and of pebbles dredged near Jimmu Seamount,12) and made it clear that there seems to be no distinct relationship between the chlorine content and the major chemical constituents, especially with respect to the phosphorous and water contents on which Kuroda and Sandell⁹⁾ focussed their attention. Iwasaki et al.,23 therefore, concluded that the chlorine distribution may largely be attributed to the quenching condition at high temperature, at which a magma is equilibrated with the gaseous phase.

The chlorine content of apatite in igneous rocks other than volcanic rocks was determined by Kind,13) Behne,10) and Vasilieva,14) and the average chlorine content of apatite in igneous rocks was estimated by Behne¹⁰⁾ as 0.30% chlorine. Assuming that the Hawaiian volcanic rocks contain apatite in which 0.3% chlorine is contained as an essential constituent on the average, the calculations will lead to the result that the contribution of chlorine due to the appatite present in rocks is limited only to the magnitude of 0.002% chlorine provided the average phosphorus content in Hawaiian tholeiitic basalts is 0.26% P₂O₅.⁴⁾ In fact, no close relationship existed between the chlorine content and the phosphorus content. For instance, specimen C-73 in Table I-a is low in chlorine in spite of a high phosphorus content. It is, therefore, clear that apatite in Hawaiian volcanic rocks is of little importance in the distribution of chlorine.

Mica is rarely found in Hawaiian volcanic rocks; as may be seen in Table I-a, the rock specimens, C-73, and C-79, which contain abundant dark mica are low in chlorine (0.006 and 0.007% respectively). Therefore, mica is also of little importance as a chlorine-carrier. The chlorine content of olivine separated from the 1959 lava of Kilauea Iki was very low (0.003% chlorine, as determined by T. Ozawa)

¹⁰⁾ W. Behne, Geochim. et Cosmochim. Acta, 3, 186 (1953).
11) C. W. Correns, "Physics and Chemistry of the Earth," Vol. I, Ed. by L. H. Ahrens, K. Rankama and S. K. Runcon, McGraw-Hill, New York (1956), p. 180.
12) H. Kuno, R. L. Fisher and N. Nasu, Deep-Sea Research, 3, 126 (1956).

¹³⁾ A. Kind, Chemie der Erde, 12, 50 (1938).
14) Z. V. Vasilieva, Geokhimiya, 704 (1957.)

in comparison with that of the original rock (0.012% chlorine).

In contrast to the facts mentioned above, the chlorine content of a ground mass in specimen C-31 (0.008% chlorine), which is a feldspar-phyric basalt and 50% occupied by feldspar phenocryst, is higher than that of the original rocks (0.005% chlorine). It is clear that almost all the chlorine in the bulk is provided by the ground mass, which represents the liquid part of the rock when it was extruded at high temperature. According to Yoshida, 15) the chlorine compounds are evolved at 600°C, and the amount of evolved chlorine compounds increases gradually with an elevation of temperature. It may be considered that the rock samples studied by Yoshida contain an appreciable amount of groundmass, that the glassy parts begin crystallizing at 600°C at a significant speed, and that the chlorine dissolved in the glassy parts is forced to escape to the atmosphere. In addition, it has been found by Iwasaki¹⁶) that the enrichment of chlorine in a rock sample collected from the middle part of one thick basaltic lava flow resulted from the larger amount of glassy parts solidified at the latest stage of the cooling process.

The Variation in the Chlorine Content of the Rock Samples of One Eruption.-Table II shows some examples of the variation in chlorine content of rock samples representing different periods of the time during each dated eruption of Kilauea and Mauna Loa. These samples all belong to aphyric-type basalt, and the amount of phenocrysts is less than 5% (tholeiitic basalt). It may, therefore, be assumed that the chlorine content of a lava flow extruded at a definite period of year will be almost constant when we take rocks with the same texture. In addition, Iwasaki and Ozawa¹⁷⁾ made it clear that the chlorine contents of approximately 120 rock samples of the 1950~1951 lava flow of O-sima Volcano, Japan, were quite homogeneous and ranged from 0.029 to 0.036% chlorine, irrespective of their appearance.

The Variation in the Chlorine Content of Rocks in Stratigraphical Sequences.—From the preceding discussion, we could expect that the chlorine content of a rock will depend mainly on the chlorine dissolved in an original silicate melt which came in contact with a gaseous phase containing a small amount of chlorine compounds. It would be valuable, in studying the descent of magmas, to see whether

or not there is any variation in the chlorine content of rocks of each volcano in stratigraphical sequence. In Table I-a, the rock samples are presented in order of stratigraphical sequences; for instance, C-1 represents the lower parts in the stratigraphy of the Waianae Range, and C-52, the uppermost part.

As may be seen in Table I-a, there is a small variation in the insoluble chlorine content of rocks in most of volcanoes with the exception of the West Maui Volcano. Roughly speaking, these variations range from 0.005 to 0.01% chlorine, and there seems no distinct variation scheme with respect to the stratigraphical sequences. In contrast, it may be seen in Table I-b that the rock specimens of Kilauea are higher in chlorine content than those of Mauna Loa. This may be explained, in one way, by the two different magma batches proposed by Powers¹⁸⁾ which are different in chemical composition. Another explanation rests on the difference in volatileescaping tendency during the movement of a melt from a magma chamber to the surface of the earth; the path in Mauna Loa is longer than in Kilauea.

In the case of West Maui, there seems to be an increase in the chlorine content of rocks toward the end of the period of volcanic eruption.

The Relationship between the Rock Types and the Chlorine Content.—A study by Macdonald19) has elucidated many detail of the relationships of the tholeitic rocks of the main shield volcano, which make up about 99 per cent of the volume as an over-all average in Hawaii; only a small percentage of Hawaiian rocks consist of the alkalic rocks which covered the late-stage cap of the volcanic mountains. The numbers of analyzed rock specimens in this study are not proportional to the volumes of each rock type, but the frequency diagrams of insoluble chlorine content in both tholeiitic and non-tholeiitic rocks are given in Figs. 1-a, 1-b. As may be seen in Figs. 1-a, 1b- the chlorine content of both tholeitic and alkalic basalts range from 0.005 to 0.014%, with a few exceptions in tholeitic basalt. On the other hand, the chlorine contents of hawaiite are scattered over a wide range from 0.006 to 0.034%. The same distribution will be seen in basanite and group, ranging from 0.006 to 0.066%, though there are only four samples. The highest chlorine content was found in trachyte obsidian of Hualalai (0.12%). The other trachytes contain very small amounts of chlorine

¹⁵⁾ M. Yoshida, This Bulletin, 36, 773 (1963).

¹⁶⁾ B. Iwasaki, Read at the Meeting of the Volcanological Society of Japan, October, 1963.

¹⁷⁾ I. Iwasaki and T. Ozawa, Read at the Meeting of the Volcanological Society of Japan, May, 1959.

¹⁸⁾ H. A. Powers, Geochim. et. Cosmochim. Acta, 7, 77 (1955).

¹⁹⁾ G. A. Macdonald, J. Geophys. Res., 68, 5100 (1963).

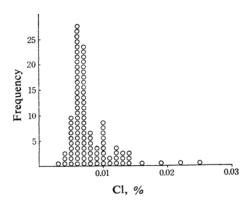


Fig. 1-a. Frequency diagram of insoluble chlorine content in tholeitic basalts.

O Tholeiitic basalt

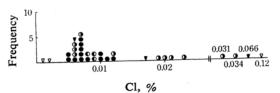


Fig. 1-b. Frequency diagram of insoluble chlorine content in non-tholeitic basalts.

- Alkalic basalt
- Hawaiite and mugearite
- Ankaramite
- △ Trachyte
- ▲ Basanite and basanitoid

(0.001 and 0.003%); thus the range of chlorine content here is larger than in the basanite group.

In general, it is difficult to say that alkalirich rocks have more chlorine than tholeitic rocks. We can, however, safely say that the rocks containing much chlorine are without exception of alkalirich types.

The Frequency Distribution of the Chlorine Contents in Hawaiian Lavas.—Figure 2 gives three cumulative frequency distributions of insoluble chlorine contents of rocks from Waianae, from West Maui, and from all Hawaian volconoes. They show approximately lognormalities, rather than normalities. Strictly speaking, a bi-log-normality is appropriate for Waianae and for all Hawaiian volcanoes. This is in striking contrast to the finding of Iwasaki16) that the frequency diagram of the chlorine content of the 1950~1951 lava flow of O-sima, Izu, shows a normality. Provided that the frequency diagram of the insoluble chlorine contents in Hawaiian lavas is lognormal, and that the water-soluble chlorine content of a rock is 0.001% chlorine, we obtain the mean chlorine content of Hawaiian lavas as, 0.008% chlorine.

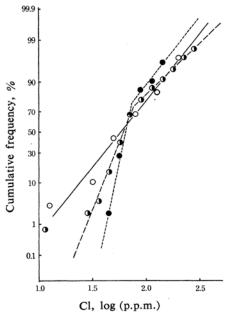


Fig. 2. Cumulative frequency distribution of insoluble chlorine content of rocks.

- O West Maui Volcano
- Waianae Volcano
- All Hawaiian lavas

It is valuable to know whether or not the chlorine content of rocks of one region differs from that of others. With respect to this regional variation, Kuroda and Sandell⁹⁾ mentioned that Japanese lavas are lower in chlorine content than those of the U.S.A. On the other hand, Iwasaki et al.2) mentioned that Japanese volcanic rocks do not significantly differ in chlorine content from igneous rocks elsewhere in the world. Here we will compare the chlorine contents of Hawaiian and Japanese basalts. The total chlorine content, that is, 0.001% plus insoluble chlorine, is used in Hawaiian basalts. For the present purpose, we will adopt the available data of the volcanic rocks in Japan studied by Iwasaki et al.23 Figure 3 shows two cumulative frequency distributions of the chlorine contents of Hawaiian and Japanese basalts. They show approximately lognormal distributions. Japanese basalts are clearly higher in chlorine than are Hawaiian As may be seen in the slope of basalts. straight lines in Fig. 3, the former is greater in the dispersion of chlorine content than is the latter. The mean chlorine contents of two basalts are 0.015% in Japan and 0.008% in Hawaii. The former is, therefore, about twice as high as the latter. Judging from the results obtained, it can be confidently stated that a regional difference in chlorine content exists between Hawaiian and Japanese basalts.

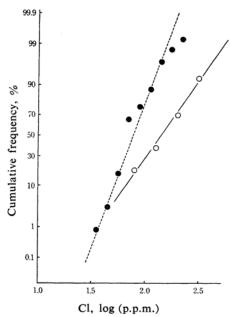


Fig. 3. Cumulative frequency distribution of chlorine content in Hawaiian and Japanese basalts.

- O Japanese basalt
- Hawaiian basalt

Conclusion

The foregoing evidence: (1) makes it clear that a narrow variation in the insoluble chlo-

rine contents of rocks is common in the history of most Hawaiian volcanoes, especially in the tholeiitic rock series; (2) confirms that the alkali-rich rocks containing much insoluble chlorine are erupted at nearly the end of the volcanic history, although the alkalirich rocks are not always higher in chlorine content than the tholeitic rocks; (3) shows, however, that the high chlorine contents are selectively found in the alkali-rich rocks without exception; (4) indicates that the frequency diagram of the insoluble chlorine in Hawaiian lavas is bi-lognormally distributed, while the mean value of chlorine content is 0.008%; (5) shows that a regional difference in chlorine content may be seen in basalts between the islands of Hawaii and Japan, and (6) suggests that the chlorine in rocks is concentrated in the glassy parts, irrespective of their mineral assemblages.

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