

14. *Chemical Composition of Prehistoric Lavas
at Makaopuhi Crater, Kilauea Volcano,
and Periodic Change in Alkali Content
of Hawaiian Tholeiitic Lavas.*

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Abstract

The K_2O/Na_2O ratio and, to a lesser degree, Na_2O+K_2O of prehistoric tholeiitic lavas exposed on the wall of Makaopuhi crater, Kilauea volcano, show periodic change with time. Similar variation is found in the lower member of Waianae volcano, Oahu Island. Marked increase in K_2O/Na_2O ratio of the historic lavas of Kilauea as compared with that of early historic and many prehistoric lavas of the same volcano may correspond to a part of such periodic variations in K_2O/Na_2O ratio as represented by the above two examples. As one possible mechanism for such increase in K_2O/Na_2O ratio, fractional crystallization of K_2O -poor plagioclase is proposed.

Locality and Sampling

Many prehistoric lava flows of Kilauea volcano are exposed on the wall of Makaopuhi crater; one of the largest pit craters on the east rift zone of Kilauea. The lava flows are superposed without any detectable unconformity and lie parallel with the slope of the volcano. Makaopuhi crater consists of two partially overlapping collapse pits of different ages. The eastern older pit is larger and filled with a prehistoric consolidated lava lake (MOORE and EVANS, 1967). The western pit was formed after the lava lake solidified and the western part of the lava lake was lost by the collapse. A near-vertical section of lava flows about 300 m high was exposed on the wall of the western pit at the time of sampling by us in the summer of 1963. Samples were collected from the succession of pahoehoe lava flows both on the north

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Table 1. Alkali content, thickness, and petrography of prehistoric lava flows exposed on north and south walls of Makaopuhi crater, Kilauea volcano.

Double lines between sample numbers indicate one or more unsampled lava flows are present between sampled flows. The new lava lake formed during the 1965 eruption has buried the lava flows below sample number 2 of this table. Symbols used in table: Ol=olivine, Cpx=clinopyroxene, Plg=plagioclase, Opq=opaque minerals, —=absent, +=present, +=present in abundance, (+)=present in very small amount, m=olivine rimmed with fine opaque minerals, w=slight oxidation, brownish staining on mafic minerals, ww=very slight oxidation.

Number	Vertical distance in feet above (+) or below (—) top surface of prehistoric lava lake	Thickness of flow in feet	Na ₂ O	K ₂ O	K ₂ O/Na ₂ O	Phenocrysts		Average size of groundmass pyroxene (×0.01 mm)	State of oxidation	Groundmass				
						Ol	Cpx			Plg	Plg	Cpx	Ol	Opq
20	+295	3?	1.89 ³⁾	0.345 ³⁾	0.18	+	—	1	—	+	—	+	+	?
19	+251	16	1.99	0.39	0.20	+m	—	2	—	+	—	+	+	+
18	+208	42	2.00	0.36	0.18	+	+	3	—	+	—	—?	—	—
17	+190	8	2.04	0.47	0.23	+	—	4	w	+	—	—?	—	—
16	+175	3	2.07	0.46	0.22	+	—	8	ww	+	—	—	—	—
15	+160	3	2.00	0.45	0.23	+m	—	4	w	+	—	+	+	—
14	+60	?	2.36 ³⁾	0.485 ³⁾	0.21	—	+	3	—	+	—	+	+	—
13	+42	19	2.60	0.68	0.26	—	+	3	w	+	—	+	+	—
12	+26	18	2.40	0.57	0.24	—	—	3	—	+	—	—	—	+
11	—11	37	2.45	0.72	0.29	—	(+)	4	w	+	+m	+	?	—
10	—19	6	2.38	0.77	0.32	—	+	2	w	+	+	+	?	?

(to be continued)

Table 1. (continued)

Number	Vertical distance in feet above (+) or below (-) top surface of prehistoric lava lake	Thickness of flow in feet	Na ₂ O	K ₂ O	K ₂ O/Na ₂ O	Phenocrysts			Average size of groundmass pyroxene (×0.01 mm)	State of oxidation	Groundmass					
						Ol	Cpx	Plg			Plg	Cpx	Ol	Opq	Glass	Silica mineral
9	-26	6	2.35	0.83	0.35	(+)	-	-	2	w	+	+	+	+	+	-
8	-43	18	2.56 ³⁾	0.90 ³⁾	0.35	-	-	-	3	ww	-	+	+	+	-	-
7	-52	11	2.07	0.64	0.31	+	-	-	2	w	+	+	-	+	?	?
6	-90	36	2.31	0.59	0.26	-	-	-	3	-	+	+	-	+	+	+
5	-123	55	1.97	0.44	0.22	+	-	-	4	w	+	+	-	+	-	?
4	-145		2.27	0.49	0.22	-	-	-	2	w	+	+	-	+	?	?
3	-204	58	2.37	0.55	0.23	-	+ ¹⁾	+	3	ww	+	+	-	+	+	+
2	-226	100	2.30	0.47	0.20	-	-	-	10	ww	+	+	-	+	+	+
1	-249		2.40	0.54	0.23	-	-	-	7	ww	+	+	-	+	+	+
21	-296		2.475 ³⁾	0.56 ³⁾	0.23	-	+ ¹⁾	(+)	4	-	+	+	-	+	+	+
22	-318	14	2.27	0.41	0.18	-	-	-	4	w	+	+	-	+	-	?
23	-332	13	2.33	0.48	0.21	-	-	-	2	w	+	+	-	+	-	?
24	-341	10	2.27	0.53	0.23	+	-	-	3	-	+	+	-	+	+	+
25	-349	11	2.00	0.44	0.22	+	+	-	1	ww	+	+	-	+	+	+
26	-373	>20	2.19	0.47	0.22	+	+	+	0.5	-	+	+	-	+	+	+

Note: 1) Samples Nos. 3 and 21 contain hypersthene phenocryst rimmed with clinopyroxene. 2) Sample No. 19 contains fine prisms of deep yellowish brown crystals in groundmass whose optical properties agree with those of pseudobrookite. 3) Average of two separate determinations.

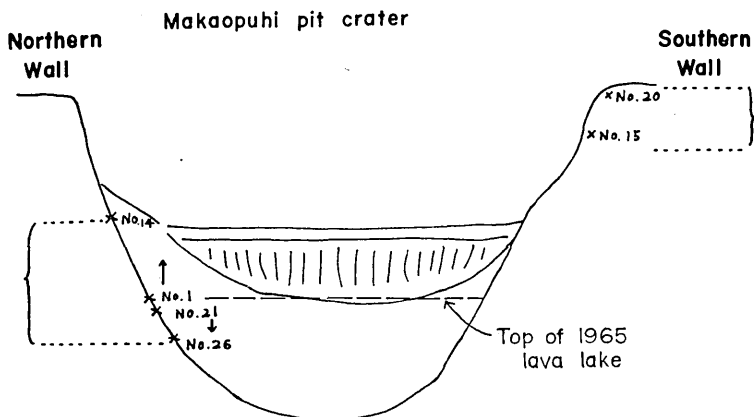


Fig. 1. Schematic cross section of the western part of Makaopuhi pit crater. Numbers indicate the position of specimens. The 1965 lava lake filled the lower part of the pit and buried the site of specimens from No. 21 to No. 26, and Nos. 1 and 2.

and south walls of the pit (Figs. 1 and 7). From the north wall every accessible lava flow was sampled (Nos. 1-14, and 21-26). From the south wall, where many thin flows are superposed, six flows were sampled (Nos. 15-20). Out of the six, two flows (Nos. 18 and 19) are in direct contact, and the other five flows are separated from each other by many thin flows which were not sampled. These lavas are probably younger than 17,000 years because they are believed to lie on top of the Pahala Ash, a thick widespread ash from Kilauea that has been determined to be between 10,000 and 17,000 years old by radio-carbon dating (RUBIN and BERTHOLD, 1961). The Pahala Ash is not exposed in the pit crater but is present a few miles south where it crops out in the high fault scarps of the Hilina Pali fault system. The lavas of the crater wall are older than those of the prehistoric lava lake which partially fills the pit crater.

In March, 1965, an eruption occurred within Makaopuhi crater and formed a lava lake 120 m deep in the west pit. The sites of the lower crater wall samples (up to No. 2 in Table 1) were covered by the new lava (Fig. 1).

Chemical Composition of the Lava Samples

A total of 30 specimens were collected from 27 individual flow units. All of these were chemically analyzed for Na_2O and K_2O by H. HARAMURA, Geological Institute, University of Tokyo, and one of us (S.A.). The chemical data and the microscopic petrography of the samples are shown in Table 1. Specimens 8, 9, and 10 were analyzed for SiO_2 , and Nos. 14, 20, and 21 for 13 components (Table 2). In Fig. 2, $\text{Na}_2\text{O} +$

K_2O , K_2O/Na_2O , and the thickness of each lava flow are shown in stratigraphic order.

From Fig. 2, it is apparent that the K_2O/Na_2O ratio increases from about 0.2 at the lowest sampled flows to about 0.35 near the center flows, and then decreases to about 0.2 at the top. Total alkalis show

Table 2. Chemical composition and norms of lavas from Makaopuhi crater wall. Sample numbers correspond with those in Table 1. Analyst: H. HARAMURA, Geological Institute, University of Tokyo.

Chemical analyses						
Column	1	2	3	4	5	6
Sample No.	20	21	14	8	9	10
SiO ₂	49.85	51.96	50.90	49.22	48.78	49.14
Al ₂ O ₃	10.87	11.66	12.63			
TiO ₂	2.72	4.08	3.54			
Fe ₂ O ₃	4.81	2.83	2.95			
FeO	7.44	10.25	9.37			
MnO	0.21	0.22	0.21			
MgO	11.76	5.71	6.40			
CaO	9.64	9.79	10.81			
Na ₂ O	1.91	2.50	2.41	2.56	2.35	2.38
K ₂ O	0.33	0.55	0.48	0.90	0.83	0.77
P ₂ O ₅	0.24	0.38	0.27			
H ₂ O ⁻	0.05	0.08	0.05			
H ₂ O ⁺	0.28	0.28	0.35			
Total	100.11	100.29	100.37			
Norms						
Column	1	2	3			
Sample No.	20	21	14			
Q	3.09	8.11	5.29			
Or	1.95	3.23	2.84			
Ab	16.15	21.11	20.40			
An	20.07	18.96	22.24			
Wo	10.91	11.40	12.36			
En	29.28	14.21	15.93			
Fs	5.61	10.16	9.31			
Il	5.16	7.76	6.73			
Mt	6.97	4.09	4.29			
Ap	0.57	0.91	0.64			
Total	99.76	99.94	100.03			

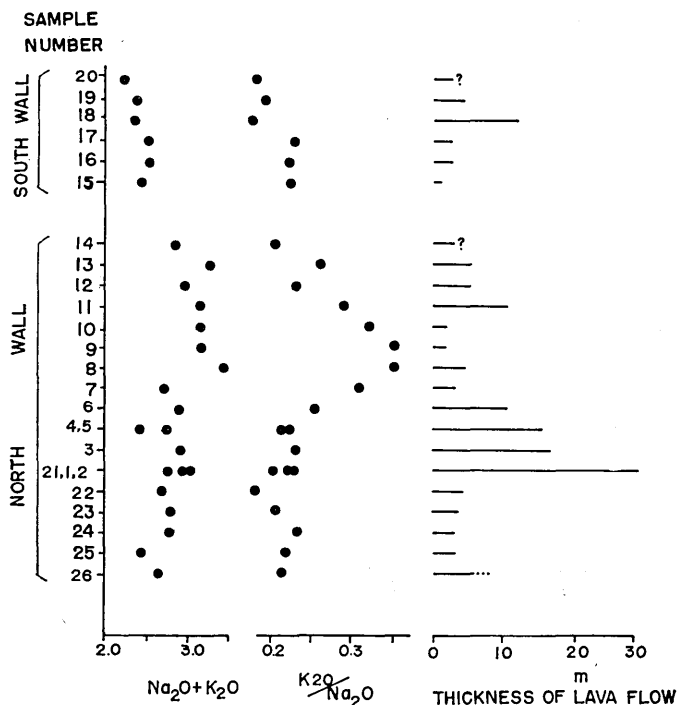


Fig. 2. Na₂O+K₂O, K₂O/Na₂O, and thickness of lava flows in stratigraphic succession exposed in Makaopuhi crater wall, Kilauea volcano.

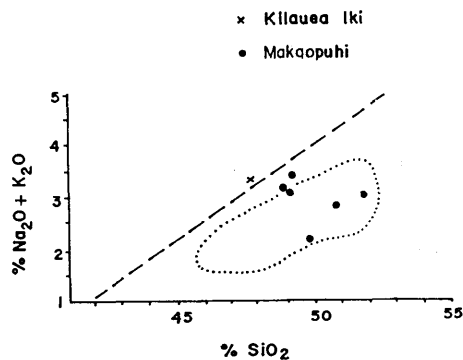


Fig. 3. Plot of Na₂O+K₂O against SiO₂. Broken line is the boundary between tholeiite and alkali olivine basalt fields as given by MACDONALD and KATSURA (1964, Fig. 1). × is the liquid that rose into the drill hole through the crust on the 1959 Kilauea Iki lava lake (MACDONALD and KATSURA, 1961). Filled circles are specimens from Makaopuhi crater wall (Table 1). Bulk of the Hawaiian tholeiites fall within the area indicated by dotted curve.

a similar though less pronounced trend. The SiO_2 content of the samples Nos. 8, 9, and 10 (Table 2) and the microscopic petrography of the high $\text{K}_2\text{O}/\text{Na}_2\text{O}$ samples (Table 1) indicate that the high $\text{K}_2\text{O}/\text{Na}_2\text{O}$ values are not due to an advanced stage of crystallization differentiation. In Fig. 3, total alkalis are plotted against SiO_2 . Though the three high $\text{K}_2\text{O}/\text{Na}_2\text{O}$ samples fall close to the broken line which is the boundary between the fields of tholeiites and alkali olivine basalts (MACDONALD and KATSURA, 1964), all six samples from the Makaopuhi crater wall fall within the tholeiitic field. The presence of the ground-mass olivine in the high $\text{K}_2\text{O}/\text{Na}_2\text{O}$ lavas (Nos. 8, 9, 10, and 11) may be related to the higher content of alkalis in these rocks.

When plotted in a $\text{CaO}-\text{SiO}_2$ diagram such as given by TILLEY and SCOON (1961, Fig. 3), out of three specimens analyzed for both CaO and SiO_2 (Table 2), No. 21 falls in the field of hypersthene bearing lavas while Nos. 14 and 20 fall in the field of lavas without hypersthene. It is interesting to note that in specimen No. 21 rare hypersthene phenocrysts rimmed with clinopyroxene are present while in Nos. 14 and 20 no hypersthene was noted (Table 1).

Discussion

The $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratio of 0.35 obtained from some of the Makaopuhi lavas is exceptionally high for the Hawaiian tholeiites. In Fig. 4, the

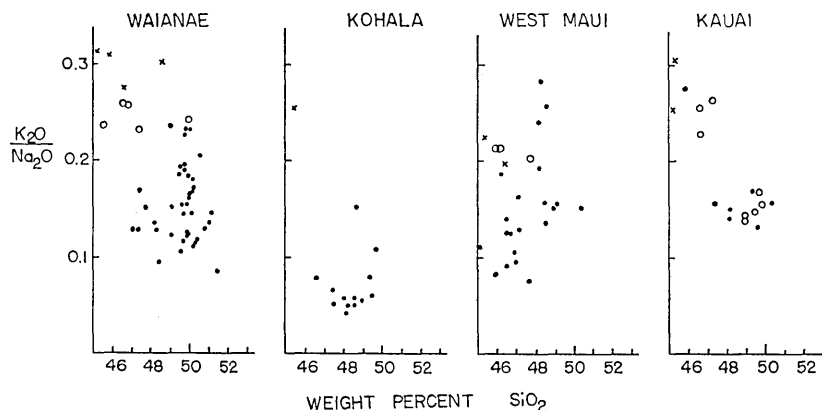


Fig. 4. $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratio and SiO_2 content of lavas from some dormant Hawaiian volcanoes. Dots, early tholeiitic lavas; circles, later tholeiitic lavas; and crosses, alkalic lavas. Analyses from MACDONALD and KATSURA (1964) and MACDONALD et al (1960). Waianae, Oahu: dots, lower member; circles and crosses, upper member. Kohala, Hawaii: dots, lower member; crosses, upper member. West Maui: dots, main part Wailuku volcanic series; circles, uppermost part Wailuku volcanic series; crosses, Wailuku volcanic series. Kauai: dots, lower Waimea Canyon volcanic series (Napali formation); circles, upper Waimea Canyon volcanic series (Olokele and Makaweli formations); crosses, Koloa volcanic series.

K_2O/Na_2O values are plotted against SiO_2 for four major tholeiitic primitive shields of Hawaiian Islands (data from MACDONALD and KATSURA, 1964). The K_2O/Na_2O ratio of the bulk of tholeiites of these four volcanoes fall in the range 0.05 to 0.25 for SiO_2 content of about 50 weight percent. Many lavas show a K_2O/Na_2O ratio less than 0.2.

In Fig. 5, K_2O/Na_2O ratios of the tholeiites of Kilauea volcano are plotted against SiO_2 . In view of the poor quality of alkali determination in some of the older chemical analyses, only those published after 1949 are plotted in the figure. The submarine lavas of the east rift zone of Kilauea volcano were collected from the surface of the rift

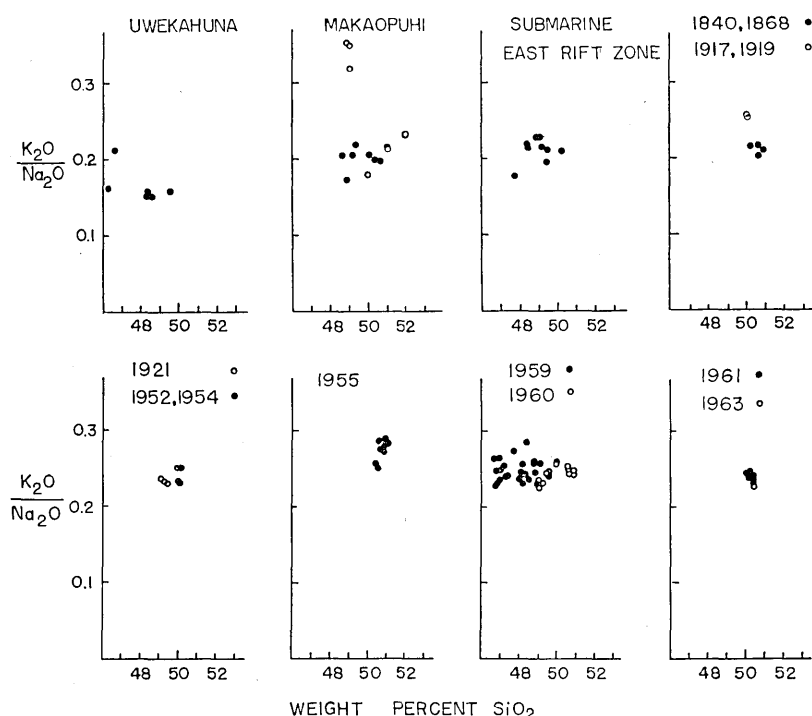


Fig. 5. K_2O/Na_2O ratio and SiO_2 content of prehistoric and historic lavas and intrusive masses Kilauea volcano. Sources of analyses and symbols are: Uwekahuna laccolith, MURATA and RICHTER (1961); Makaopuhi prehistoric lava lake (filled circles), (MOORE and EVANS, 1967); prehistoric lavas exposed on the wall of Makaopuhi crater (open circles), this report (Tables 1 and 2), three points with K_2O/Na_2O ratios higher than 0.3 are Nos. 8, 9, and 10 in Table 1; lavas dredged from the submarine part of the east rift zone of Kilauea volcano (MOORE, 1965). Historic lavas: 1840 (MACDONALD, 1949, and MOORE and AULT, 1965); 1868 (TILLEY and SCOON, 1961); 1917 and 1919 (MACDONALD and EATON, 1955); 1921 (TILLEY, 1960); 1952 (MACDONALD, 1955b); 1954 (MACDONALD and EATON, 1957); 1955 (MACDONALD, 1955a); 1959 (MURATA and RICHTER, 1966; and MACDONALD and KATSURA, 1961); 1960 (MURATA and RICHTER, 1966); 1961 (RICHTER et al, 1964); 1963 (PECK et al, 1965).

zone by dredging from water depths of 490 to 5,000 m (MOORE, 1965). Since the subaerial part of the same rift zone is mantled by historic flows over roughly one-half of its length, probably about one-half of these submarine samples are historic. Hence this suite of samples must be considered as a mixture of young surface flows intermediate between late prehistoric and historic.

From Fig. 5, it is apparent that K_2O/Na_2O ratios are higher in lavas erupted after 1917 than those erupted before the 20th century and in prehistoric time. The K_2O/Na_2O ratios of most of the early and prehistoric lavas range from 0.15 to 0.24 while those of recent lavas range from 0.23 to 0.29. The SiO_2 content of most lavas cluster around 50 percent while picritic varieties show lower SiO_2 contents. Thus the K_2O/Na_2O ratio appears to have increased temporarily in recent Kilauean lavas as compared with prehistoric lavas of the same volcano.

In Fig. 6, the variation of K_2O/Na_2O and K_2O+Na_2O is shown for successive lavas of the lower member of Waianae volcano, Oahu Island (data from MACDONALD and KATSURA, 1964). Like the lava succession on the crater wall of Makaopuhi, Kilauea, the K_2O/Na_2O ratio changes periodically with time. The range of the K_2O/Na_2O variation is more than 0.15. The variations shown in Figs. 2 and 6 indicate that in both the Kilauea and lower Waianae magmas, there were epochs of marked enrichment of K_2O and to a lesser degree of Na_2O .

No definite estimate can be made as to the time intervals between the emplacement of successive lava flows exposed on the Makaopuhi crater walls and on Waianae volcano. However it is possible that the recent and probably temporary increase in K_2O/Na_2O ratio in historic lavas of Kilauea vol-

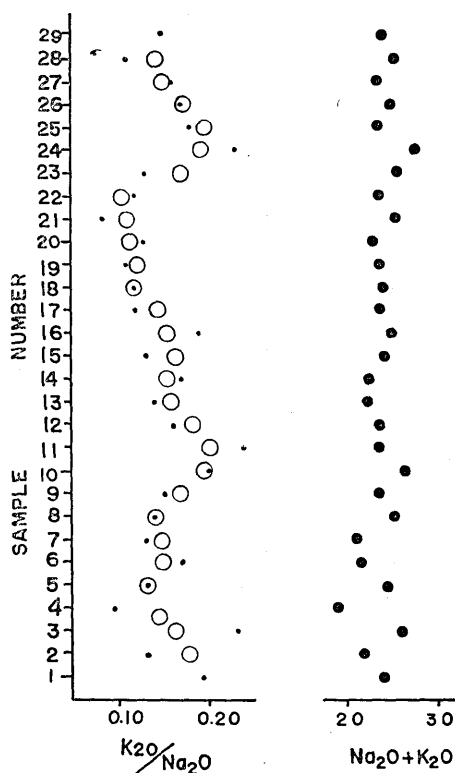


Fig. 6. Variation of Na_2O+K_2O and K_2O/Na_2O in the lower member of Waianae volcano, Oahu (data from MACDONALD and KATSURA, 1964). Open circles are overlapping means of three consecutive points.

Table 3. SiO₂ and alkali contents of segregation veins and filter-pressed "oozes" of historic or prehistoric lava lakes, laccoliths, and lava flows of Kilauea volcano.

Specimen	Host rock body	SiO ₂	Na ₂ O	K ₂ O	Na ₂ O + K ₂ O	$\frac{K_2O}{Na_2O}$	Reference
Segregation vein	Uwekahuna laccolith	52.04	2.79	0.83	3.62	0.30	MURATA and RICHTER (1961)
"	"	51.68	2.46	0.70	3.16	0.28	ARAMAKI (1968)
"	Prehistoric lava flow on Kilauea caldera wall	52.36	2.99	0.80	3.79	0.27	KUNO et al. (1957)
"	Prehistoric lava lake in Makao-puhi crater	52.72	2.85	0.68	3.53	0.24	MOORE and EVANS (1967)
Filter-pressed ooze in drill hole	Crust on 1959 lava lake Kilauea Iki	48.43	2.10	0.60	2.70	0.29	MACDONALD and KATSURA (1961)
		47.73	2.43	0.98	3.41	0.40	
"	Crust on 1963 lava lake Alae pit crater	50.62	2.65	0.88	3.53	0.33	PECK et al. (1966)
		50.25	2.62	0.80	3.42	0.31	
		53.30	3.26	1.57	4.83	0.46	
		50.72	2.77	0.96	3.73	0.35	
		51.73	2.83	1.16	3.99	0.41	
		51.34	2.80	1.07	3.87	0.38	
		51.68	3.10	1.17	4.27	0.38	

cano is similar to such peaks in the K₂O/Na₂O ratio as are found in lavas of Makaopuhi crater wall and Waianae volcano.

The anomalously high K₂O/Na₂O ratios are also found in segregation veins or filter-pressed "oozes" of historic or prehistoric lava lakes, laccoliths, and lava flows of Kilauea volcano. They are tabulated in Table 3. Most of the authors cited in Table 3 consider advanced stages of fractional crystallization as the main cause of enrichment in K₂O and Na₂O, but MACDONALD and KATSURA (1961) and others remark that volatile transfer and thermodiffusion may have operated to some degree during differentiation.

ARAMAKI (1965) showed that the plagioclase phenocrysts (labradorite) of the 1955 lava flow on the east rift zone of Kilauea volcano are low in K₂O. They contain 0.22 weight percent K₂O (1.2 molecular percent Or) and 4.09 percent Na₂O. All of the analyses of the lavas of Kilauea volcano presently known to us have K₂O contents higher than 0.22 percent and Na₂O lower than 4.09 percent. Therefore subtraction of plagioclase with such a composition should always increase the K₂O/Na₂O ratio of the residual liquid. For example, the subtraction of 5 weight

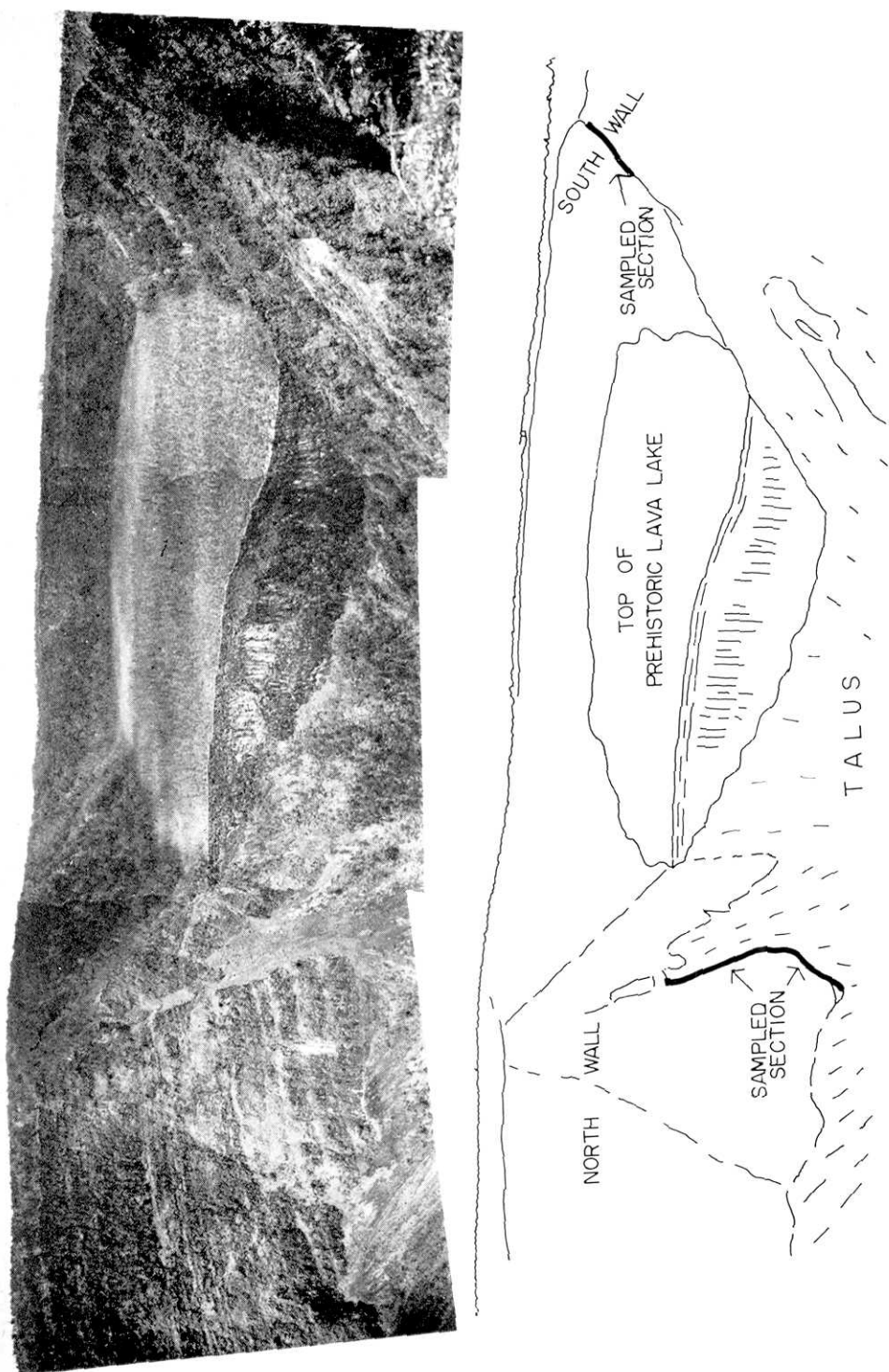


Fig. 7. Panorama photograph and sketch of Makaopuhi pit crater viewed from the lookout point on the southwestern rim. Photo taken in September, 1963.

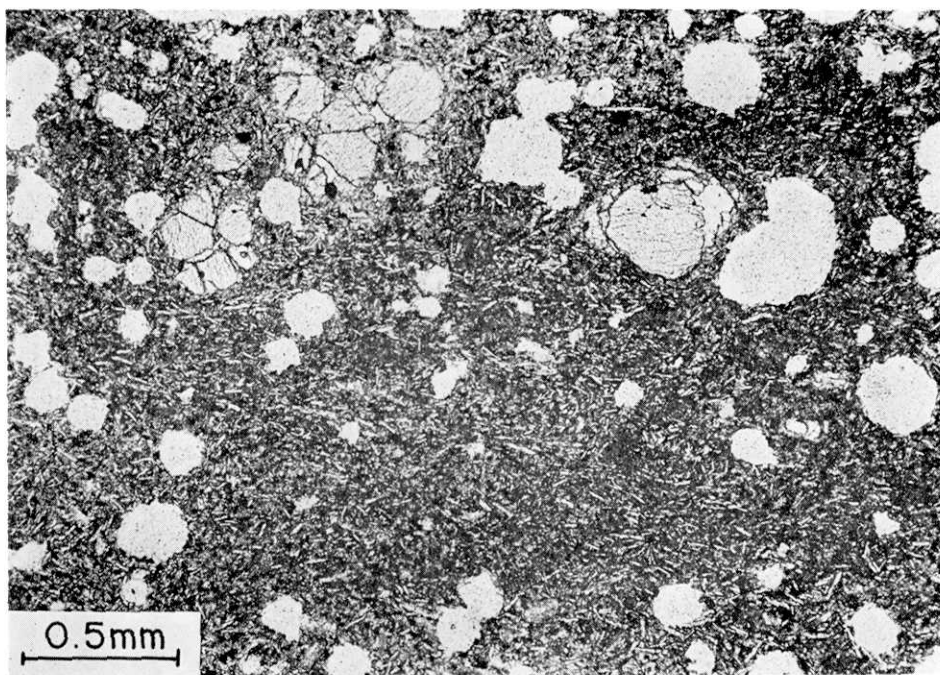


Fig. 8. Photomicrograph of lava No. 20 (Table 1), Makaopuhi crater wall. Plain light. Note olivine phenocrysts.

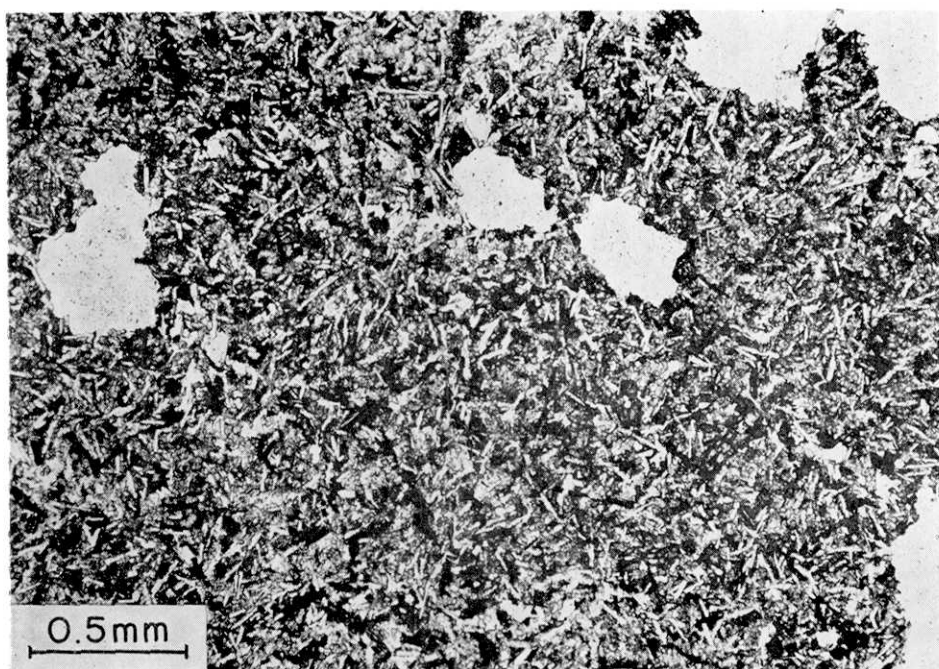


Fig. 9. Photomicrograph of lava No. 9 (Table 1), Makaopuhi crater wall. Plain light. White areas are gas pores.

percent of this plagioclase from a liquid whose composition is represented by an analysis of the 1955 Kii flow of Kilauea (MACDONALD, 1955a, Table 13, column 3) will increase the K_2O/Na_2O ratio from 0.29 to 0.31. The composition of the Kilauean tholeiitic magmas is generally very close to the boundary between fields of olivine, clinopyroxene, and plagioclase on the liquidus "surface" of the magma. The presence of these three minerals as phenocrysts in the lavas of Makaopuhi crater wall (Table 1) also indicates that the prehistoric Kilauean liquids fluctuated between these fields. Hence, a slight change in physical-chemical conditions in the shallow magma chamber may change the order of appearance of these minerals from the liquid.

SHIBATA and KATSURA (written communication) have shown experimentally that in a tholeiite melt from Kilauea volcano, the order of appearance of phenocrysts is olivine→clinopyroxene→plagioclase at atmospheric total pressure and at relatively high partial pressure of oxygen, while at lower partial pressure of oxygen, the order is olivine→plagioclase→clinopyroxene. Thus it is interesting to note that in the relatively shallow magma chamber within the shield of Kilauea volcano local or temporary change in partial pressure of oxygen alone could result in a change in kinds of precipitating crystals and hence the amount of alkalis and the K_2O/Na_2O ratio.

The sampling of the lava specimens was made when one of us (S.A.) visited the Hawaii Volcano Observatory, U.S. Geological Survey, in 1963 under the program of the U.S.-Japan scientific cooperation. The Japanese party was led by Professor T. MINAKAMI.

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14. キラウエア火山マカオプヒ火口の先史時代の溶岩の

化学成分とハワイのソレライト質溶岩の

アルカリ量の周期的変化

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キラウエア火山マカオプヒ火口壁に露出する、一連の先史時代のソレライト質溶岩を採集し、30個の標本について Na_2O と K_2O 、そのうち3個については更に SiO_2 、他の3個については13成分の定量を行った。その結果、 $\text{K}_2\text{O}/\text{Na}_2\text{O}$ 比が時間と共に顕著な周期的変化を示すことが明らかになった。 $\text{Na}_2\text{O}+\text{K}_2\text{O}$ もほぼ同様の変化を示す。同じような変化はオアフ島ワイアナイ火山においても見られる。キラウエア火山の最近の溶岩の $\text{K}_2\text{O}/\text{Na}_2\text{O}$ 比は、同火山の20世紀以前の溶岩のそれに比べ著しく高いが、これは、上に述べた周期的変化の2例と同じ現象かも知れない。これを説明する1つの可能性として、 K_2O に乏しい斜長石がマグマから晶出分離するモデルを提唱した。