

Estimation of Amounts of Weathered Products through Chemical Composition of Waters in the Kamenose Landslide Area

By Ryuma YOSHIOKA

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Abstract

136 water samples have been collected from the Kamenose landslide area, Osaka Prefecture, to estimate the amounts of weathered products through the chemical composition of the waters.

We have examined the relationships among the chemical species contents of the waters and the interaction of silicate minerals with the waters, and the following have been obtained: The amounts of montmorillonite and kaolinite produced by CO₂-bearing waters reacting with parent rocks are estimated as 49 tons/year (60 g/m²/year) and 40 tons/year (50 g/m²/year), respectively; the total amount of weathering rocks is estimated as 150 tons/year (190 g/m²/year); the waters are in equilibrium with kaolinite and montmorillonite; coexistence of both kaolinite and montmorillonite is maintained only under the condition of bicarbonate content over about 60 mg/l.

1. Introduction

Most natural waters in the hydrologic cycle contact and react with silicate minerals, altering them, and forming new minerals. Natural waters are a primary agent of rock weathering and offers us various informations concerning underground environment, such as whether rock-forming silicates are weathered or not, whether bedrock consists of silicate rocks or limestone etc..

Probably formation and existence of clay minerals at landslide areas bring about lowering of cohesion through their expansion and/or fragility of constitution material by ion exchange. Therefore, it is very important to estimate the production rate of clay minerals at a landslide area. We have investigated the distribution of dissolved constituents in natural waters of various scar and landslide areas in order to estimate the amounts of weathered products through the chemical composition of the waters^{1)~5)}.

Many geological and geophysical studies on the Kamenose landslide area have been reported, but very few geochemical studies have been done^{2),4),6)}.

2. Outline of the Kamenose landslide area

The Kamenose landslide area is situated at the intersecting point of the Kongo-Ikoma mountain range and the Yamato River, which gathers the water of the Nara basin, flows through the Kawachi plain and empties into Osaka Bay. This area lies

to the northern slope of the Yamato River, is sunny and well drained. Therefore, this area is suited to the cultivation of various fruits. Especially, grape is widely cultivated.

Serious landslides occurred in the Toge district from 1931 to 1932 and covered a total area of about 32 ha⁷⁾. The horizontal slide movement of this land was 53 meters (max. 52.3 cm/day), its dipping 16 meters (max. 18.6 cm/day), the upheaval of river bed 36 meters (max. 38.5 cm/day). In 1951, a total area of about 3 ha slid along the downstream part of the Shimizudani district, and on February 1967 some cracks broke out at the upper Shimizudani district and as the days went by, a strong slide movement was activated, developing into a large scale landslide, covering a total area of about 50 ha in both Shimizudani and Toge districts (max. movement 25.5 cm/day).

A geological map of the Kamenose landslide area is shown in Fig. 1. Bed rock consists of Ryōke granites, and Nijō and Osaka groups cover on the granites. Granite forming the bed rock is exposed on the western side of landslide area and the boundary between granite and Nijō group is approximately consistent with a crack of western edge in this landslide area. Kasugayama volcanic rock covers directly granite of base rock, and mainly distributes on the left bank of the Yamato River and does not seem to have an immediate relation to the phenomena of these landslides⁷⁾.

Donzurubo layer consists chiefly of unconsolidated tuff or tuff-breccia with green or purplish red colours. When this layer is weathered, it alters to a clayey material with light gray or yellow colours and becomes very impermeable to water. Once this layer contains water, it becomes weak rock, and so it seems to have a close relation to the occurrence of landslide. 55 borings were carried out to grasp accurately the

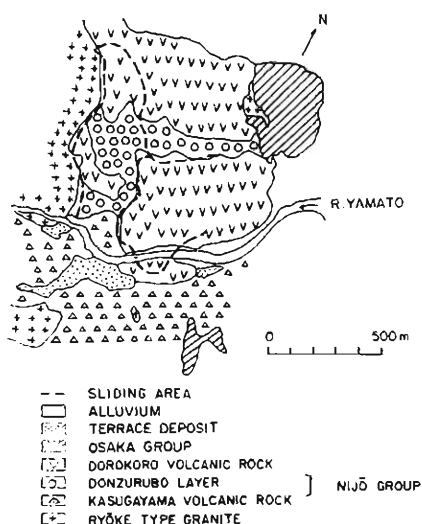


Fig. 1 A geological map of the Kamenose landslide area, Osaka Prefecture (after KINKI REGIONAL CONSTRUCTION BUREAU¹⁴⁾).

situations and shapes of slip surfaces. It was found that the slip surfaces exist an average of 30—40 meters under the surface but at times exceeds depths of 80 meters. They are found in the tuff itself which is sandwiched thinly between andesite layers, or on the upper layers of tuff or tuff breccia contacting with the lower part of overlying tuff. These seem to be closely connected with the fact that though the tuff of Donzurubo layer is very impermeable to water, once it contains water, it expands, weakens and develops easily into slide, while Donzurubo volcanic rock layer is very permeable to water.

Besides geological conditions, the effect of ground water is much accounted as one of various causes of the occurrence of landslide.

In the Kamenose landslide area, it is widely recognized that tuff or tuff breccia alter to clay minerals, being one of the causes having influence on the occurrence of landslide. Ground water plays an important role in the formation of clay minerals. The directional map of ground water and landslide flows shows that the ground water flows correspond considerably to the landslide flows (Fig. 2)¹⁾.

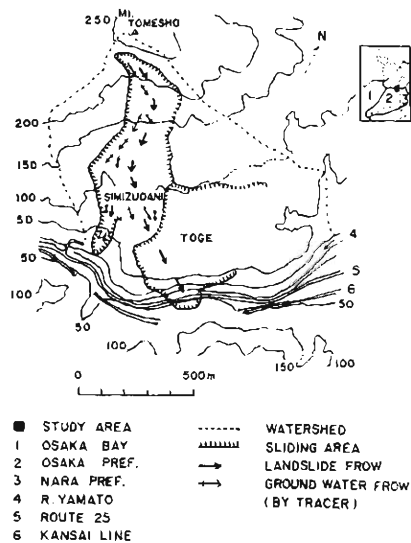


Fig. 2 A directional map of ground water and landslide flows (after KINKI REGIONAL CONSTRUCTION BUREAU,¹⁾).

3. Experimental

Samples: 54 ground water samples draining from bore holes in the tunnel were collected from the Kamenose landslide area on June 1971, 4 stream and 6 shallow well water samples from 1967 to 1969, and 7 rain water samples from June to August 1968 (Fig. 3). Moreover, 65 ground water samples were collected monthly at six fixed stations from July 1972 to July 1973 to examine the relationship between the concentration of dissolved chemical species and ground water discharge.

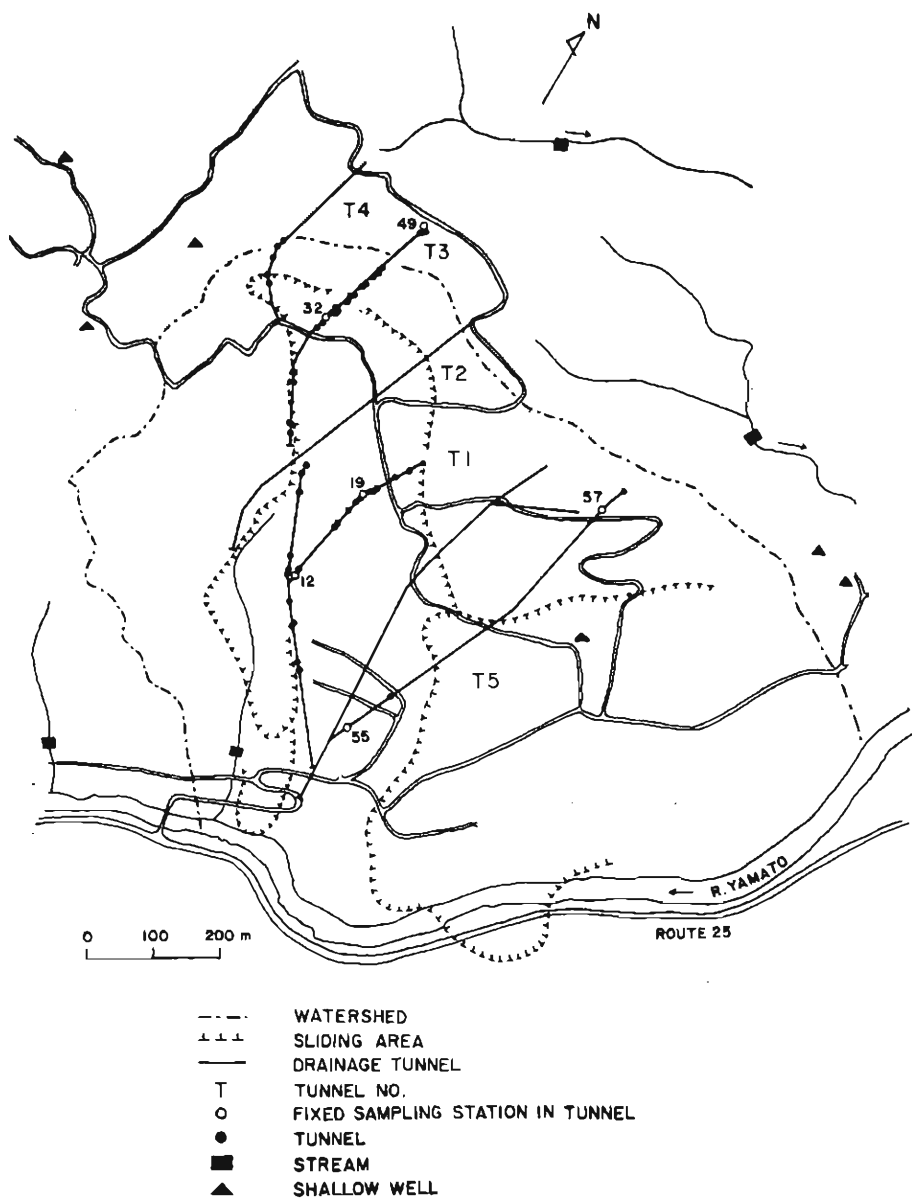


Fig. 3 A map showing sample localities.

Analytical method: Water temperature and pH values were measured in the field and chemical constituents, such as Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , HCO_3^- and soluble SiO_2 were determined in the laboratory.

pH: Colorimetric method.

Na^+ : Flame photometric method.

K^+ : Flame photometric method.

Ca^{2+} : Titration method with EDTA standard solution and Dotite NN (2-hydroxy-1-(2-hydroxy-4-sulfo-1-naphthylazo)-3-naphthonic acid) indicator or colorimetric method with Dotite GHA(glyoxal bis-(2-hydroxyanil)⁸⁾.

Mg^{2+} : Titration method with EDTA standard solution and Dotite NN and EBT indicators or colorimetric method with Dotite XB-1(1-azo-2-hydroxy-3-(2, 4-dimethylcarboxanilido)-naphthalene-1'-(2-hydroxybenzene-5-sulfonic acid)⁹⁾.

Cl^- : Colorimetric method with mercury thiocyanate¹⁰⁾.

SO_4^{2-} : Colorimetric method with barium chromate and diphenylcarbazide¹¹⁾.

HCO_3^- : Titration method with hydrochloric acid standard solution and bromocresol purple indicator.

Soluble SiO_2 : Colorimetric method with ammonium molybdate.

4. Results and Discussion

(1) **Trilinear diagram**: The trilinear diagram representing relative percentage of milligram equivalents of major ionic constituents is shown in Fig. 4 to examine the water quality of ground waters in the Kamenose landslide area. It is clear from this figure that waters in tunnels at high elevation give the high proportions of sodium, chloride and sulfate ions but the proportions of calcium, magnesium and bicarbonate ions increase with decreasing elevation. Consequently, the type of water quality in the tunnel at high elevation shows $\text{Ca}^{2+}-\text{SO}_4^{2-}$ and Na^+-Cl^- types but at lower elevation the type changes to $\text{Ca}^{2+}-\text{HCO}_3^-$ and $\text{Mg}^{2+}-\text{HCO}_3^-$ types. Sodium ion of shallow well and stream waters in the granite area is higher than that of

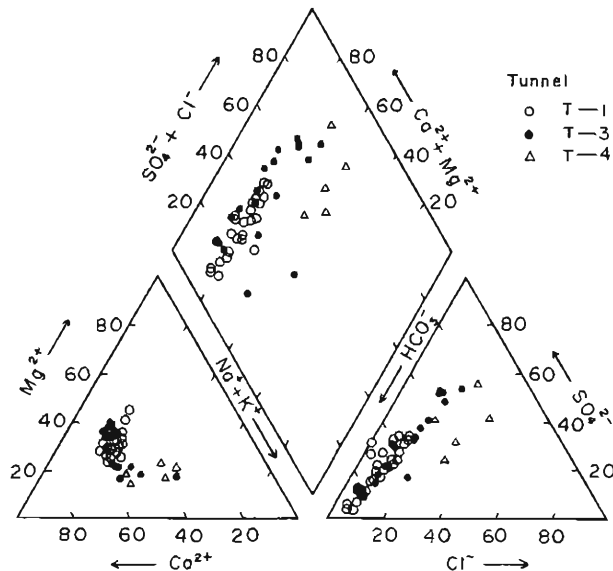


Fig. 4 Trilinear diagram used to represent relative percentage of milligram equivalents of major dissolved ionic constituents in ground waters.

ground waters from andesite, but magnesium ion in the granite area is lower than that in the andesite area.

(2) **Mineral-water interactions:** The minerals which constitute the rocks in this landslide area consist of plagioclase (An_{30-40} or An_{60-70}), orthoclase, pyroxene, hypersthene, quartz with minor amounts of amphibole and calcite^{(2), (3)}. The bulk chemical compositions of the unweathered, weathered andesites and slip clay are given in Table 1. The clay minerals consist mainly of montmorillonite and kaolinite. Minor amounts of halloysite and saponite were observed in several samples⁽⁴⁾.

In general, the chemical composition of land water is partly affected by precipitation, dry fallout, thermal-mineral springs and human activities, but the most dominant influence is exerted by parent rocks with which water comes in contact. Silicate rocks react with CO_2 -bearing water to form clay minerals, releasing cations, silica and bicarbonate ions to solution. An aluminum in most silicates is not at all mobile, so that reactions with natural waters must produce new mineral products higher in aluminum than the primary silicates⁽⁵⁾. The new minerals formed in silicate rocks include kaolinite, montmorillonite, chlorite, illite and some others. The chemical reactions continue until the minerals in the system reach a state of equilibrium with water. Therefore, a knowledge of the processes of chemical reactions between silicate rocks and water will contribute to some understanding of the rock weathering and soil formation processes found in nature.

In order to understand weathering reactions, it is convenient to use stability

Table 1 Bulk chemical composition of unweathered, weathered andesites and clay.

	Unweathered andesite	Weathered andesite	Clay
	Wt%	Wt%	Wt%
SiO ₂	60.22	43.49	48.15
TiO ₂	0.50	0.72	1.15
Al ₂ O ₃	15.34	22.90	23.88
Fe ₂ O ₃	2.73	1.62	1.23
FeO	2.89	0.88	1.19
MnO	0.13	0.10	0.09
MgO	5.34	1.64	1.77
CaO	5.60	1.51	1.53
Na ₂ O	2.77	0.88	0.98
K ₂ O	1.72	0.10	0.86
H ₂ O ⁺	1.42	7.85	7.43
H ₂ O ⁻	1.30	18.23	12.27
P ₂ O ₅	0.12	—	—
CO ₂	nd	—	—
Total	100.08	99.92	100.53
	YAMAZAKI and ONUKI ⁽³⁾	ITIHARA ⁽⁵⁾	

diagrams for the common silicates on which the phases are drawn in terms of dissolved constituents of the coexisting solution. The stability diagrams should be constructed by means of the thermodynamic data for various minerals^{(16)~(21)}. But this method can be applied only in the case when we know the chemical compositions of the minerals of primary rock and the compositions of secondary minerals produced during weathering⁽²¹⁾. Normally, although it is difficult to solve this problem in nature, this method may be effective in the case of a relatively simple field.

Table I gives the following six facts:

- 1) Aluminum and titanium are enriched in weathered andesite and produced clay minerals.
- 2) Alkali, alkaline earth metals, ferrous and ferric irons are readily leached from primary rock.
- 3) The potassium released from primary rock is fixed in clay mineral products.
- 4) Silicate is released from primary rock and is rich in clay minerals.
- 5) Manganous content does not change during weathering processes.
- 6) The water contents of weathered andesite and clay mineral are very high as

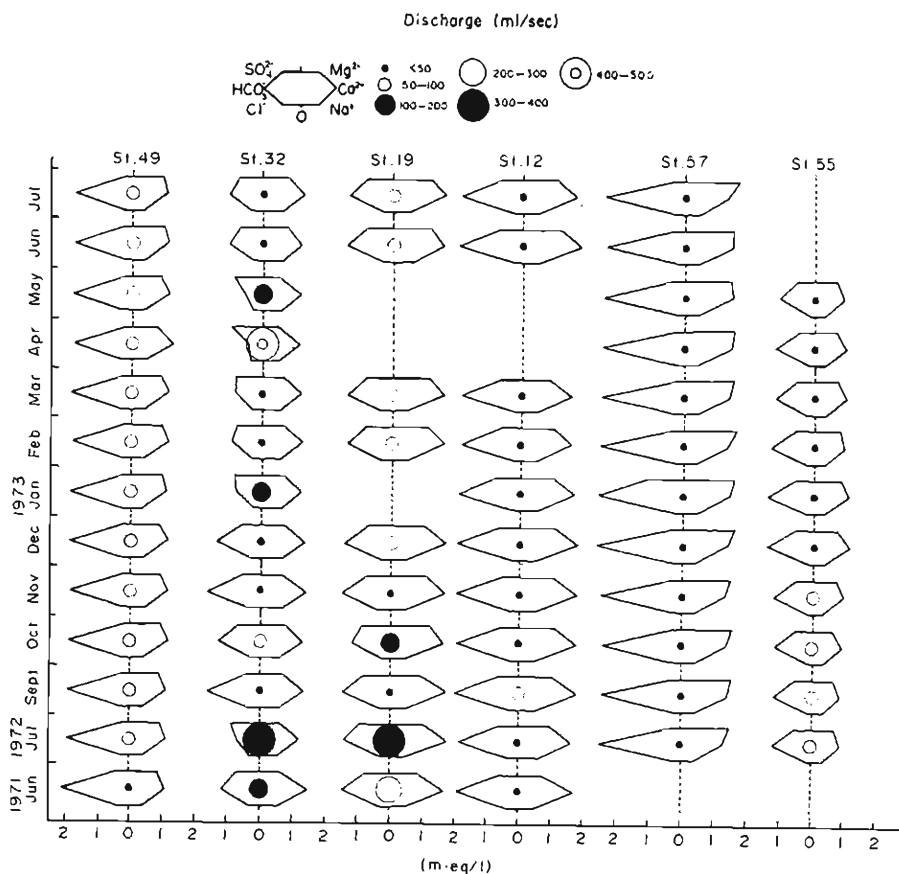


Fig. 5 Variation with time in the concentrations of major dissolved ionic constituents and discharge.

compared with fresh rock.

It seems that the high content of silica in clay mineral may be due to mixing very fine quartz or colloidal silica with clay mineral, the high content of ferric iron and the low content of ferrous iron are affected by a reducing environment of ground water, the high content of titanium is caused by deposits of titanium oxide, and the high content of potassium is attributable to adsorption on clay minerals⁶⁾.

In Fig. 5 is hexadiagrams showing the relation between chemical composition and discharge of ground waters in three tunnels (T. 1, T. 2 and T. 3). Except St. 32, chemical compositions of ground waters have almost been remained constant during the observation periods. At St. 32, the concentration of sulfate ions shows a trend to increase with the increasing discharge, but that of bicarbonate ions a trend to decrease with increasing discharge.

Generally, as the residence time of ground water is large, dissolved oxygen is consumed by decomposition of organic matters under ground and gradually decreases if it is not supplied, and sulfate ions disappear from water by reduction to hydrogen sulfide which combines with ferrous iron²²⁾. Judging from these, this behaviour of sulfate ions at St. 32 should be interpretable as follows. With a large amount of discharge, the increase of discharge depends on ground water velocity, therefore its residence time should be small. In that case the content of dissolved oxygen should be large and that of the sulfide ions may be oxidized. With increasing CO₂ content of water in contact with rock and sediment, the bicarbonate ions content increases. And with increasing discharge, the CO₂ content of water decreases and the bicarbonate ions content decreases. This may explain the observation of relation between bicarbonate ions content and discharge of water.

Figs. 6—8 show the stability diagrams for potassium, sodium and calcium silicates

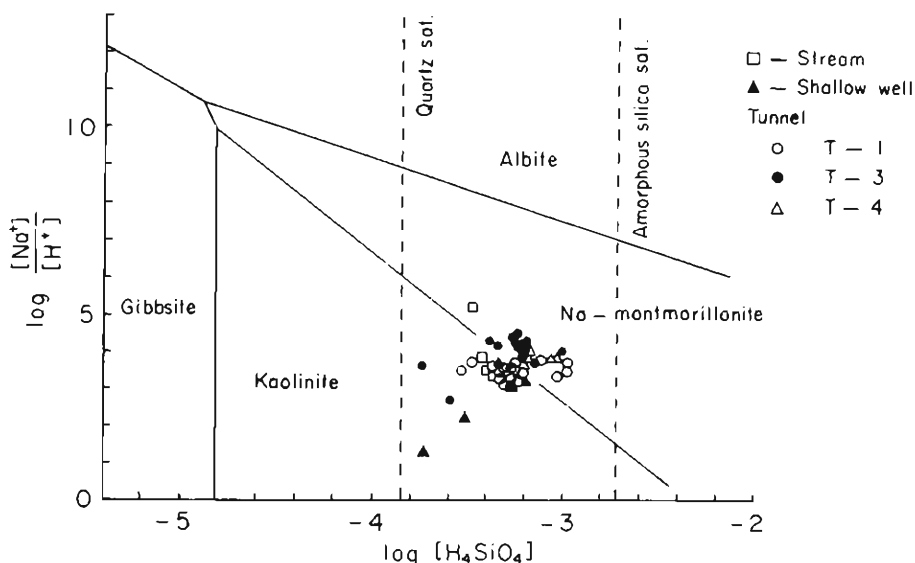


Fig. 6 Stability relations of albite, Na-montmorillonite, kaolinite and gibbsite as functions of $[Na^+]$, $[H^+]$ and $[H_4SiO_4]$ (at 25°C and 1 atm.).

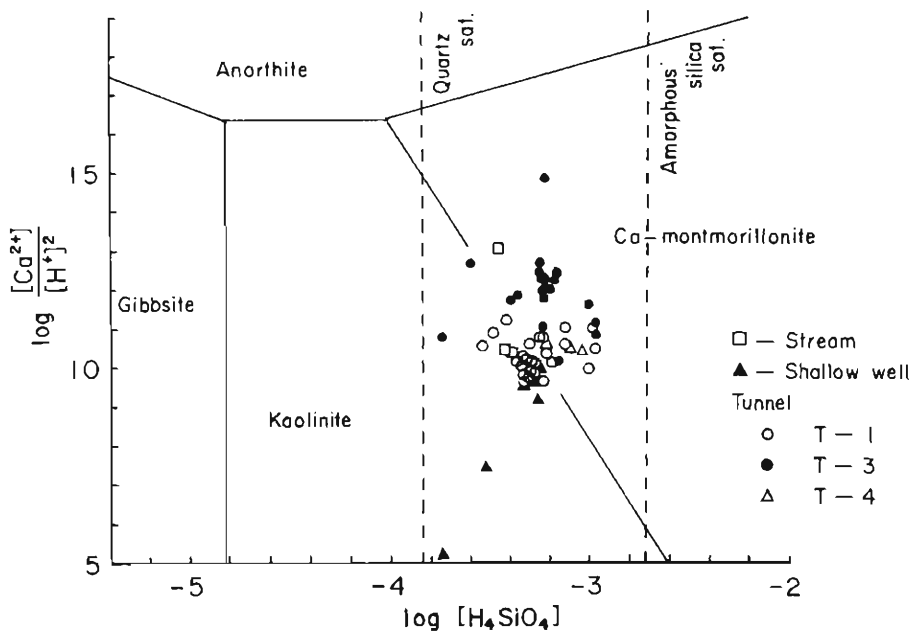


Fig. 7 Stability relations of anorthite, Ca-montmorillonite, kaolinite and gibbsite as functions of $[Ca^{2+}]$, $[H^+]$ and $[H_4SiO_4]$ (at 25°C and 1 atm.).

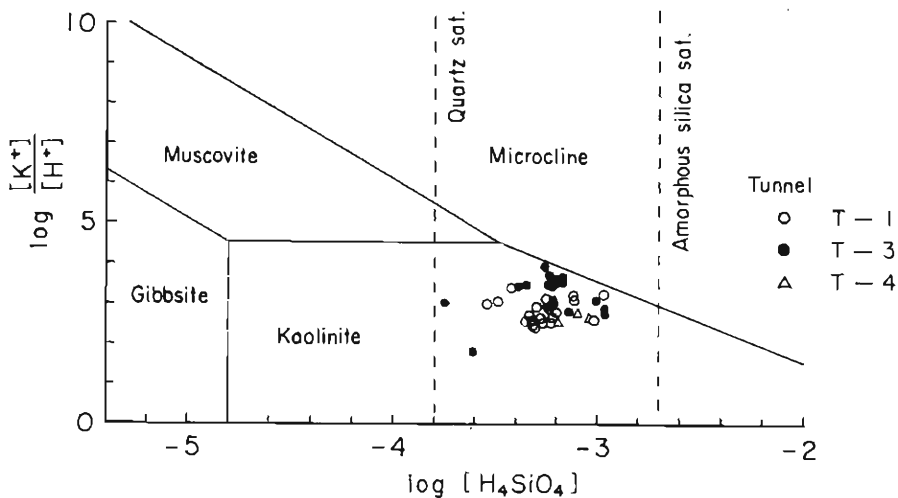
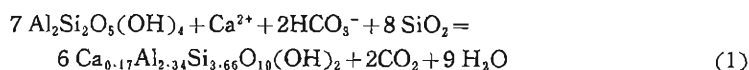


Fig. 8 Stability relations of microcline, muscovite, kaolinite and gibbsite as functions of $[K^+]$, $[H^+]$ and $[H_4SiO_4]$ (at 25°C and 1 atm.).

which are calculated by Trady²¹⁾, and these diagrams include the field of stability of anorthite, albite, microcline, muscovite, gibbsite, kaolinite and montmorillonites.

It is seen from these diagrams that stream waters and shallow well waters are placed in the field of kaolinite in the stability diagrams except one sample, but tunnel waters are on the phase-boundary between kaolinite and montmorillonites. These relations imply that the waters may be in equilibrium with these clay minerals.

From these diagrams, by X-ray diffraction and differential thermal analysis, it becomes evident that both kaolinite and montmorillonite exist in this area. If water is in equilibrium with both kaolinite and calcium montmorillonite, the reaction can be written according to Garrel²³⁾:



And the following relation is obtained:

$$\log [\text{Ca}^{2+}] + 8 \log [\text{SiO}_2] + 2 \text{pH} = \text{constant} \quad (2)$$

here, []: molalities

Consequently, the equilibrium of kaolinite and montmorillonite in water can be tested by a constancy of the left-hand terms in Eq. (2)²³⁾. Fig. 9 showing the relation between bicarbonate content and the value of constant indicates that the value becomes constant over the region of bicarbonate content in about 60 mg/l. And it suggests that the state approaches to equilibrium between kaolinite and calcium montmorillonite. Waters low in bicarbonate content are active in kaolinizing the rocks, and waters in more than bicarbonate content 60 mg/l are montmorillonizing the rocks. It is necessary for bicarbonate content of about 60 mg/l to maintain equilibrium between kaolinite and montmorillonite. Montmorillonite is generally formed

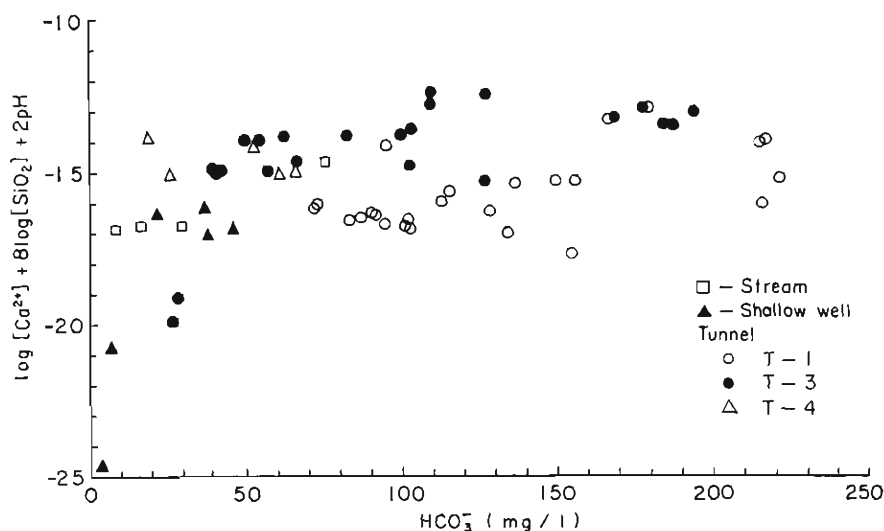


Fig. 9 Plots of $(\log [\text{Ca}^{2+}] + 8 \log [\text{SiO}_2] + 2 \text{pH})$ versus $\text{HCO}_3^- (\text{mg/l})$.

in landslide areas. The above result suggests that the formation of the swelling montmorillonite clays seems to be affected by the concentration of bicarbonate ions in water. We regard the treatment, involving a relatively simple chemical equilibrium between mineral and water, as one of useful methods for understanding the phenomena of landslides.

(3) **Estimation of amounts of produced clay minerals:** Generally, the most sodium and calcium ions in the ground waters of igneous rock areas are supplied through chemical weathering of plagioclase except for those waters with large concentrations of chloride or sulfate ions²³⁾.

A plot of the mole ratio of sodium to calcium ions versus bicarbonate ions to silica for ground waters from the Kamenose landslide area is shown in Fig. 10 with four theoretical curves (A, B, C and D). They were calculated from various ratios of silica to aluminum oxide in the plagioclase. The three curves (A, B and D) were calculated by Garrels²³⁾. In the figure, the typical equations used in constructing the plagioclase-gibbsite, -kaolinite, -Na-montmorillonite and -Ca-montmorillonite curves are

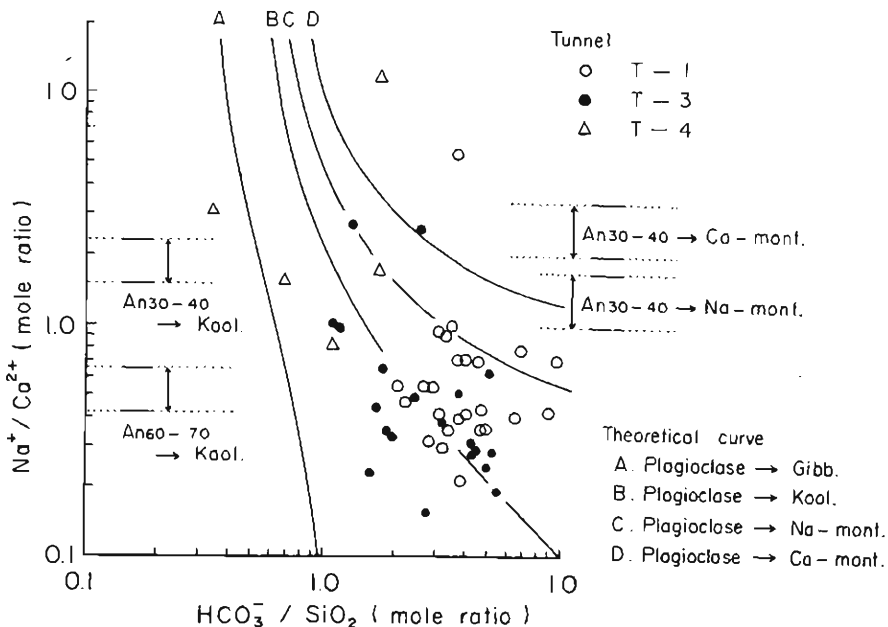
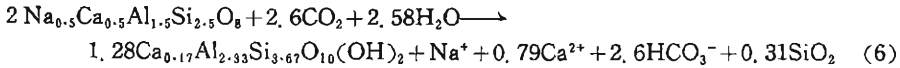
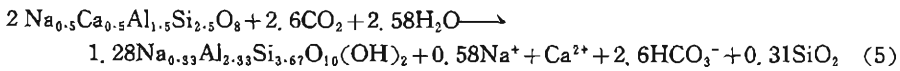
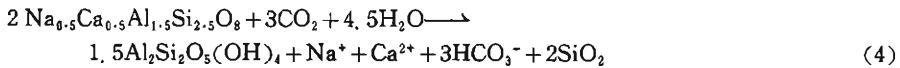
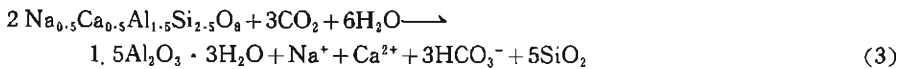


Fig. 10 $\text{Na}^+/\text{Ca}^{2+}$ mole ratio plotted versus $\text{HCO}_3^-/\text{SiO}_2$ mole ratio.

An_{30-40} and An_{60-70} in this figure represent the plagioclases which exist abundantly in this landslide area⁽²⁾. The points plotted in the figure were after the correction of contribution from rain and human activities, that is, with Garrels and MacKenzie's⁽²⁴⁾ thought that chloride ions are derived from sodium chloride, and sulfate ions from calcium sulfate. Most of the chemical compositions at T-1 tunnel distribute between the theoretical curve derived from kaolinization of plagioclase and that from Na-montmorillonization of plagioclase. The waters at T-3 tunnel fall mainly on the theoretical curve for the alteration of plagioclase to kaolinite. The most points plotted are much in range of sodium to calcium ions ratio derived from alterations of plagioclase of An_{60-70} to kaolinite and montmorillonites. Also, there are considerable points between 0.2 and 0.4 of sodium to calcium ions ratio. This suggests that the plagioclase of An_{70-80} exists considerably, when ion exchange between soil, and sodium and calcium ions in waters, dissolution and/or precipitation of calcium carbonate, quartz and others are neglected. If only two kinds of the plagioclase mentioned above exist, it can be considered that the chemical reactions of ion exchange, dissolution and precipitation occur significantly. In any case, it seems true that CO_2 -bearing water reacts with plagioclase and produces kaolinite and montmorillonite in this area.

In order to estimate the amounts of clay minerals produced by weathering processes through the chemical composition of ground water, it is necessary to determine the altering minerals, the clay minerals and dissolved constituents. As mentioned in the previous section, the dissolved constituents in this area are supposed to be supplied by weathering of the five minerals, plagioclase, biotite, hypersthene, pyroxene and quartz, and most of weathered products are kaolinite and montmorillonite. And, the amounts of clay minerals through weathering processes can be estimated by reconstituting the original minerals by back-reaction of ground water constituents and weathering products^{(24), (25)}. The calculated results of the Kamenore landslide area are shown in Table 2:

- a) The contribution from rain water is subtracted from the chemical constituents of the ground water. The remaining chloride and sulfate ions in the ground water are removed as sodium chloride and calcium sulfate, respectively. The total electrical charge of cations is adjusted by bicarbonate ions.
- b) The amounts of weathered parent minerals and produced clay minerals were calculated by use of the chemical reactions shown in Table 2.
- c) From the remaining calcium, magnesium and silica, the amounts of $MgSiO_3$ and $CaCO_3$ were calculated.

Then, the rate of chemical weathering in this area is estimated. The annual rainfall average about 1,400 mm/year, and the area of this watershed is $8.0 \times 10^5 m^2$. By assuming that the direct runoff is about 10% of rainfall and the evapotranspiration loss is about 30%⁽²⁶⁾, about $7.1 \times 10^5 m^3$ is estimated as the ground water recharge. The total amount of weathered materials is 150 tons/year ($190 g/m^2/year$), and the amounts of kaolinite and montmorillonite produced by CO_2 -bearing waters reacting with silicate rock are estimated as 49 tons/year ($60 g/m^2/year$) and 40 tons/year ($50 g/m^2/year$), respectively.

Table 2 Reconstitution of original minerals by back-reaction of ground water constituents and weathering products.

	Concentrations in moles/liter × 10 ⁴								
	Na ⁺	Ca ²⁺	K ⁺	Mg ²⁺	SiO ₂	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	
Ground water	5.22	7.70	0.95	4.38	5.93	17.93	2.76	3.26	
Rain water	0.25	0.09	0.08	0.08	0.00	0.00	0.19	0.54	
Ground water - rain water	4.97	7.61	0.87	4.30	5.93	17.93	2.57	2.72	Amount of material produced by the back reaction of ground water with weathered rock (mole /liter × 10 ⁴)
Remove Na ⁺ =Cl ⁻ and Ca ²⁺ =SO ₄ ²⁻	2.40	4.89	0.87	4.30	5.93	17.93	0.00	0.00	
Adjust HCO ₃ ⁻ =total Cations	2.40	4.89	0.87	4.30	5.93	21.64	0.00	0.00	
Kaolinite → plagioclase (1)	1.20	3.69	0.87	4.30	3.53	18.04	0.00	0.00	
Montmorillonite → plagioclase (2)	0.00	2.74	0.87	4.30	3.16	14.92	0.00	0.00	2.4 plagioclase → 1.54 montmorillonite
Kaolinite → biotite (3)	0.00	2.74	0.00	1.69	1.42	8.83	0.00	0.00	0.87 biotite → 0.87 kaolinite
Form CaCO ₃ (4)	0.00	0.00	0.00	1.69	1.42	3.35	0.00	0.00	2.74 calcite
Form MgSiO ₃ (5)	0.00	0.00	0.00	0.00	-0.27	-0.05	0.00	0.00	1.69 enstatite
Residual	0.00	0.00	0.00	0.00	-0.27	-0.05	0.00	0.00	

Reactions (coefficients × 10⁴)

- (1) $1.8\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + 1.2\text{Na}^+ + 1.2\text{Ca}^{2+} + 3.6\text{HCO}_3^- + 2.4\text{SiO}_2 \rightarrow 2.4\text{Na}_{0.5}\text{Ca}_{0.5}\text{Al}_{1.5}\text{Si}_{2.5}\text{O}_8 + 3.6\text{CO}_2 + 5.4\text{H}_2\text{O}$
- (2) $1.54\text{Ca}_{0.17}\text{Al}_{2.33}\text{Si}_{3.61}\text{O}_{10}(\text{OH})_2 + 1.2\text{Na}^+ + 0.95\text{Ca}^{2+} + 3.12\text{HCO}_3^- + 0.37\text{SiO}_2 \rightarrow 2.4\text{Na}_{0.5}\text{Ca}_{0.5}\text{Al}_{1.5}\text{Si}_{2.5}\text{O}_8 + 3.12\text{CO}_2 + 3.1\text{H}_2\text{O}$
- (3) $0.87\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4 + 0.87\text{K}^+ + 2.61\text{Mg}^{2+} + 6.09\text{HCO}_3^- + 1.74\text{SiO}_2 \rightarrow 0.87\text{KMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2 + 6.09\text{CO}_2 + 3.05\text{H}_2\text{O}$
- (4) $2.74\text{Ca}^{2+} + 5.48\text{HCO}_3^- \rightarrow 2.74\text{CaCO}_3 + 2.74\text{CO}_2 + 2.74\text{H}_2\text{O}$
- (5) $1.69\text{MgSiO}_3 + 3.38\text{CO}_2 + 1.69\text{H}_2\text{O} \rightarrow 1.69\text{Mg}^{2+} + 3.38\text{HCO}_3^- + 1.69\text{SiO}_2$

5. Conclusions

From the dissolved constituents of natural waters the following results have been obtained:

- 1) The quality of ground water changes from $\text{Ca}^{2+}-\text{SO}_4^{2-}$ and Na^+-Cl^- types to $\text{Ca}^{2+}-\text{HCO}_3^-$ and $\text{Mg}^{2+}-\text{HCO}_3^-$ types as the elevation decreases.
- 2) The chemical compositions of ground waters have remained almost constant during our observation periods, except for one station.
- 3) Stream and shallow well waters are in equilibrium with kaolinite, and ground waters with kaolinite and montmorillonites.
- 4) Equilibrium between kaolinite and montmorillonite can be maintained only under the condition of bicarbonate content over about 60 mg/l.
- 5) The total amount of chemical weathered products is about 150 tons/year (190 g/m²/year) and the amounts of kaolinite and montmorillonite produced through weathering are estimated as 49 tons/year (60 g/m²/year) and 40 tons/year (50 g/m²/year), respectively.

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