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DISTRIBUTION OF GOLD IN QUATERNARY VOLCANIC ROCKS OF THE KURIL ISLAND ARC

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This paper presents the results of the first study of the Au distribution in the Quaternary volcanics of the southern and central Kuril-arc volcanic zones: 205 samples were analyzed from 29 volcanoes of varying differentiation, including 19 submarine structures. Although there may be a positive correlation between the Au distribution and the silica and Fe-Mg contents in the rocks, it is controlled to a greater extent by the enrichment of the parental magmas with volatiles, e.g. S and Cl. The different patterns found in the Au distribution across the arc were attributed to the differences of the parental magmas in alkalinity and also to different scales of tectonic activity which was responsible for variability in Au removal from the magma together with volatiles. These Au distribution patterns may serve as important guides in assessing the region for potential gold mineralization. The frontal (fore-arc) volcanic zone, where tectonic activity has lasted a longer time and was more intense than in the other zones, and where the lavas are less alkalic, can be considered to be a more favorable area for gold deposition and ore formation.

INTRODUCTION

The geochemistry of gold from the Quaternary volcanics of the Kuril island arc is practically unknown except for some scanty and unrepresentative evidence from the products of a few land volcanoes that occur on the largest of the Kuril islands [1], [7]. No publications are available on the Au distribution in submarine volcanic rocks, abundant in the back-arc area. This hampered the understanding of the Au distribution mechanism in the course of the magmatic process and precluded the assessment of potential gold mineralization in the region.

A regional survey was recently carried out in an extensive area to investigate the geologic structure of the submarine portion of the arc and study the mineralogy and geochemistry of Quaternary

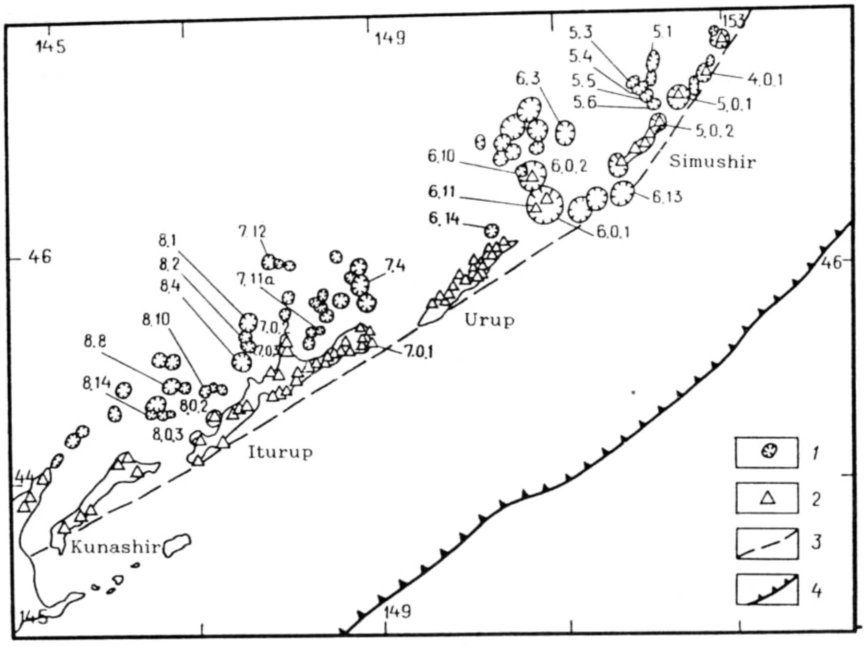

Figure 1 Map showing location of volcanoes in the southern and central Kuril island arc. 1 – submarine volcanoes; 2 – land volcanoes; 3 – volcanic front; 4 – Kuril-Kamchatka trench axis. Indices of submarine volcanoes are given after the catalog [5].

volcanics in the region. It was a teamwork of three institutes: the Institute of Volcanology, Far East Division, Russian Academy of Sciences, the Institute of Geochemistry, Siberian Division, Russian Academy of Sciences, and the Institute of the Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry (IGEM), Russian Academy of Sciences. The results were published first in a few papers [2], [3], [4] and later in a book [5]. Although the publications present information on the distribution of over 40 trace elements and of all major elements, the Au distribution was not reported because of a delay in getting analytical data. This paper is an attempt to make up for the gap. It presents data on the Au distribution in the rocks of 29 variously differentiated volcanic structures, including 19 submarine volcanoes, from the volcanic zones of the southern and central Kuril arc (Figure 1). Overall, 205 samples were examined.

ANALYTICAL METHODS

The Au concentrations in the rocks were determined at the Institute of Geochemistry, Siberian Division, Russian Academy of Sciences, by the atomic absorption analysis. 1 to 5 g specimens were chemically enriched by extraction using 0.5 ml of methylbenzene. Measurements were made using an atomizer with a cell (Perkin-Elmer-503). The precision was 0.0002 g/ton (0.2 mg/ton); analysts, S. E. Chernigova and A. A. Khlebnikova.

The analytical data for all volcanoes are presented in a graphical form in Figure 2. For the convenience of comparison the results are summarized in Tables 1 and 2 where the Au variation ranges and average concentrations are given for all types of rocks, from basalts to rhyolites. The data are tabulated for individual volcanoes and for the groups of them localized in the fore-arc, central and back-arc zones to demonstrate the Au behavior with distance from the volcanic front landward and with the increasing overall alkalinity, potassium content and other parameters of the rocks [2], [3].

To minimize the statistical estimate of the Au distribution, the Au concentration was determined in 3 to 5 specimens of rocks containing varying amounts of silica for each volcano, except for 7 or 8 volcanoes where one or two specimens were used for lack of samples (six of them were submarine volcanoes). Although this impairs the quality of the data, the general assessment of the Au distribution remains valid. As many samples showed anomalous (too high or too low) Au concentrations because of the secondary alteration of rocks, specimens were collected for analysis from samples of best preservation. The localization and geologic structure of the Kuril volcanoes have been described in a book by Gorshkov [10] and also in our book [5].

RESULTS

As follows from Tables 1 and 2 and Figure 2, the Au concentrations vary from < 0.5 to 16 mg/ton. The higher concentrations, as well as the maximum values of variance for the same rock types from one volcano, were found in the basalts, the rocks that are compositionally closer to the primary magmas. The average Au concentrations in the basalts of 15 volcanic structures (out of 22 where basalt was found) are higher than 2-3 mg/ton irrespective of the zone the volcano is located. Yet, five volcanoes yielded the average values of 1 to 1.5 mg/ton and two less than 1 mg/ton. It is not unlikely that the low Au concentrations in at least half of the cases resulted from an insufficient number of specimens (one per a volcano). As can be seen in Figure 2, and has been confirmed by our petrographical examination, the nonuniform Au distribution in the basalts is governed by two relationships. On the one hand, the Au percentage is higher in the rocks of the lowest silica content. On the other hand, the basalts of a similar (high or low) silica content are almost always (in 18 out of 21 cases) more enriched in Au, provided they contain abundant early phenocrysts of pyroxene and, especially, olivine and contain few or no plagioclase phenocrysts. The plagioclase-bearing basalts that occur largely in the frontal volcanic zone (e. g., Lvinaya Past caldera and

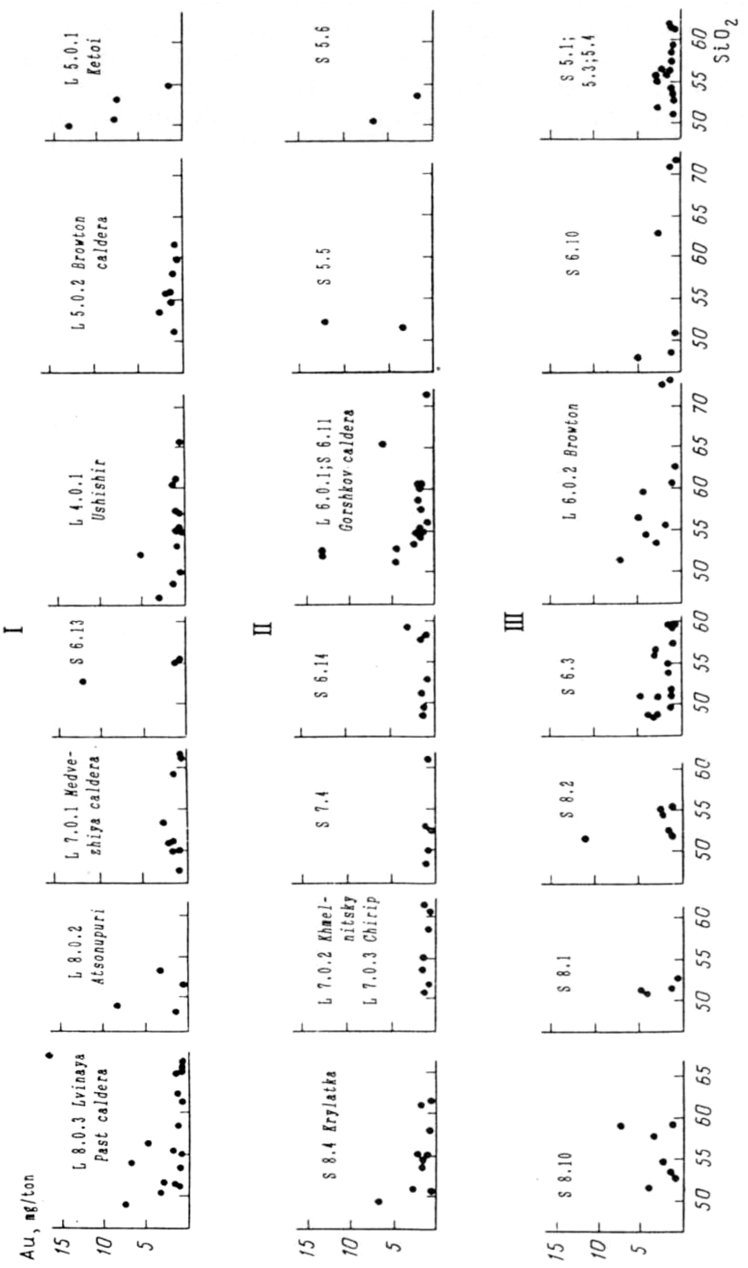

Figure 2 Plots showing Au and SiO_2 distribution in Quaternary volcanics of some Kuril-arc volcanoes, Indices of volcanoes are as in Figure 1. Letters S and L in the indices denote submarine and land volcanoes, respectively: I is fore-arc, II central, and III back-arc volcanic zone.

Atsonupuri Volcano) showed the lowest, < 0.5 mg/t Au quantities. At the same time, high average Au percentages are most frequent in the least alkaline tholeiitic basalts of the frontal zone (see Table 1). Apparently, this last feature accounts for the highest average Au concentrations (4.58 mg/ton) in the basalts of the frontal zone, somewhat lower values (3.58 mg/ton) in the central, and the lowest (3.15 mg/ton) in the back-arc zone. The average Au values are close to the clarke of Au concentration (4 mg/ton) in the basic rocks (after A. P. Vinogradov). They are notably higher than the average Au values in the Cenozoic basalts of the Kuril-Kamchatka province (1.73 mg/ton) and of Paramushir Island (1.85 mg/ton, average of eight analyses) and are close to the average values for the basalts of Simushir Island (4.25 mg/ton, average of seven analyses) reported by Anoshin [7]. Besides, they resemble the average Au concentrations in the magnesian basalts (4.3 mg/ton) produced by the Northern fissure eruption of Tolbachik, Kamchatka, [8] and are markedly lower than those of the subalkalic aluminous basalts produced by the Southern fissure eruption of the same Tolbachik event (7 mg/ton). They are also lower than the average Au values for the continental tholeiitic basalts of the Siberian Craton [7] and of the Cascade Range, USA, [16].

Thirteen out 23 volcanoes showed a progressive Au decrease in the Kuril volcanic rocks as the silica content grows from the basalts to the andesites and further to the dacites and the rhyolites. Seven volcanoes showed that the average Au concentrations were at the same, usually low level (0.5 to 1.5 mg/ton) throughout the whole series, while in six volcanoes the average values might grow slightly from the basalts to the basaltic andesites, then drop in the andesites, and continue to the decrease further on.

The Au concentrations in the basaltic andesites are generally lower than in the basalts and vary as a function of the petrography and geochemistry of the rocks in the same manner as do the basalts. The average concentrations do not show any appreciable differences across the arc, being about 1.22-2 mg/ton in all three volcanic zones.

The average Au concentrations in the andesites of all volcanic zones are lower than in the basaltic andesites (0.74 to 1.58 mg/ton) but in contrast to the basalts they increase progressively from the fore-arc to the back-arc andesites (see Table 1). Variability of the Au values increases in the same direction. For instance, the Au variation range in the andesites of the submarine volcano S 8.10 is as great as 1.2 to 7.6 mg/ton, which is larger, like the average value (3.42 mg/ton), than in the basalts of the same volcano: 0.9 to 4 mg/ton and 2.45 mg/ton, respectively. Abnormally high Au concentrations (up to 42.6 mg/ton) were observed in the andesites of some homeogenic inclusions in the rocks of the back-arc zone (S 5.1 volcano), even though no relationship was found in the Au distribution between the total amount and the ratio of early phenocrysts in the andesites.

All of our average Au concentrations in the andesites are lower than the values reported by Anoshin [7] for the rocks of a similar

Table 1 Au distribution in Quaternary volcanics of the basalt-andesite series from the Kuril- arc volcanic zones, mg/ton.

Volcanic zone	Index and name of volcano	Basalts			Basaltic andesites			Andesites		
		Au distribution								
		n	Range	Average	n	Range	Average	n	Range	Average
Fore-arc	L8.0.3 Lvinaya Past caldera	5	0,72—7,00	2,88	5	0,48—6,60	2,80	3	0,40—0,90	0.70
	L8.0.2 Atsonupuri	3	0,32—8,00	3,21	1	—	2,80	—	—	-
	L7.0.1 Medvezhiya caldera	5	0,58—1,80	1,26	1	—	2,40	3	0,29—1,20	0.67
	S6.1.3	1	—	11,70	2	0,40—1,08	0,74	—	—	-
	L5.0.2 Browton caldera	1	—	1,04	4	1,48—2,50	1,78	3	0,32—1,06	0.69
	L5.0.1 Ketoi	3	7,80—13,00	9,53	1	—	1,52	—	—	-
	L4.0.1 Ushishir	4	0,63—5,00	2,47	4	0,53—1,00	0,75	5	0,60—1,20	0.89
	Average for the zone	22	—	4,58	18	—	1,83	14	—	0.74
Central	S8.4 Krylatka	3	0,26—6,46	3,13	4	0,90—2,08	1,56	3	0,34—1,70	0.88
	L7.0.2 Chirip	1	—	1,00	1	—	1,07	2	0,60—0,60	0.60
	L7.0.3 Khmelnitsky	1	—	0,53	2	1,08—1,40	1,24	1	—	1.00
	S7.11	—	—	—	2	0,96—1,20	1,08	3	0,58—2,82	1.40
	S7.4	4	0,34—0,58	0,46	—	—	—	2	0,60—0,61	0.60
	S6.14	4	0,78—1,20	1,03	1		0,52	3	0,53—3,00	1.74
	L6.0.1 Gorshkov caldera	4	4,00—13,00	8,50	7	0,50—2,13	1,40	5	1,00—1,50	1.15
	S5.5	2	3,00—12,00	7,50	—	—	—	—	—	-
	S5.6	1	—	6,50	1	—	1,70	—	—	-
	Average for the zone	20	—	3,58	18	—	1,22	19	—	1.05
Back-arc	S8.1	4	0,54—4,86	2,46	—	—	—	—	—	-
	S8.2	3	0,99—11,25	4,51	3	0,72—2,20	1,64	—	—	-
	S8.10	2	0,90—4,00	2,45	3	1,25—2,30	1,78	4	1,20—7,60	3.42
	S8.8	—	—	—	2	0,56—0,90	0,73	—	—	-
	S7.12	—	—	—	—	—	—	2	0,53—1,30	0.92
	L6.0.2 Browton	1	—	6,90	5	1,68—4,50	2,89	3	0,45—4,13	1.76
	S6.10	3	0,26—4,80	1,97	—	—	—	1	—	2.40
	S6.3	8	0,72—4,38	2,38	4	1,32—3,00	2,13	4	0,50—1,28	0.84
	S5.1	3	0,68—2,68	1,39	6	0,80—2,70	1,51	3	0,84—1,60	1.19
	S5.3	—	—	—	1	—	2,20	1	—	1.25
	S5.4	—	—	—	1	—	3,10	2	0,80—1,00	0.90
	Average for the zone	24	—	3,15	25	—	2,00	20	—	1.58

silica content from the Glavnaya volcanic zone of the Kuril arc (2 mg/ton) and from individual islands: Paramushir (1.88 mg/ton), Simushir (1.7 mg/ton), and Kunashir (2.2 mg/ton). Yet, they are comparable with the Au values in single specimens of andesites from the island Chirpoi (1.52 mg/ton) and Browton (0.83 mg/ton) but are considerably lower than in the andesites of Tyatya Volcano on Kunashir [1].

Our data on the Au distribution in the rocks of the dacite-rhyolite series are limited: we had only 15 samples collected from six volcanoes (see Table 2). As seen from Table 1, the Au concentrations in the silicic lavas are commonly low, < 1 mg/ton, especially in the rocks of genetically related lava complexes. The values are notably below the clarke of Au concentration in silicic igneous rocks, which is 4.5 mg/ton after A. P. Vinogradov, and are lower than the average Au concentrations in the silicic volcanics of Kunashir and of the Glavnaya volcanic zone (1.96 mg/ton) reported by Anoshin [7].

It is important to note that dacitic pumice from calderas of some volcanoes (e.g., Lvinaya Past and Gorshkov), as well as some extrusive dacites from back-arc submarine volcanoes (S 8.14), showed abnormally high Au concentrations (5.5 to 16 mg/ton).

DISCUSSION

There is plentiful literature on the geochemistry of gold, but data on its concentrations in igneous rocks of varying compositions are often contradictory. As follows from more recent publications [12], most of the investigators report higher average concentrations from basic rocks. At the same time, according to other authors, e.g. Anoshin [7], there is not much difference between silicic and basic rocks in the Au content. Our data indicate that the Au distribution in the evolution series of the Quaternary Kuril volcanics suits both patterns, though the latter is manifested less frequently. An important point here is that whereas the decrease of average values from basalts to more silicic rocks may be caused by a decrease in the concentration of iron as a more active element-modifier [12], our observations of higher and lower Au concentrations (noted by other investigators as well), unrelated to the iron or silica content, seem to be caused by other processes.

As demonstrated by observational and experimental data [12], [13], [14], the Au distribution in the magmatic process is controlled largely by the association of gold with volatiles, primarily sulfur and chlorine. The behavior of volatiles in a magmatic system is closely related to the *PT* conditions of magma generation and evolution, the alkalinity and oxygen fugacity of crystallizing magma, and other factors. For instance, in ultrabasic and basic melts, Au migration occurs largely on account of sulfur and in the reducing environment where gold may exist as Au^{1+}. In silicic melts, however, where the part of sulfur is not as significant as in ultrabasic and basic melts,

Table 2 Au distribution in Quaternary volcanics of the dacite-rhyolite series from the Kuril-arc volcanic zones.

Volcanic zone	*Index and name of volcano*	*Rock*	*n*	*Au*	
				Range	*Ave-rage*
Fore - arc	L8. 0.3 Lvinaya Past caldera	Dacite, lava, flows	4	0.40 - -1.20	0.70
		Dacite, pumice	1	–	16.0
	L4. 0.1 Ushishir	Dacite extrusion	1	–	0.50
Central	L6. 0.1 Gorshkov caldera	Dacite, pumice	1	–	5.50
		Rhyolite, pumice	1	–	0.60
Back-arc	S8. 14	Dacite, extrusion I	2	0.48 - -0.74	0.61
		Dacite, extrusion II	1	–	12.60
	L6. 0.2 Browton	Rhyolite, Lava flows	2	1.00 - -2.00	1.50
	S6. 10	Rhyodacite, pumice	2	0.40- -1.11	0.73

and the part of chlorine is quite essential, a rather high oxygen potential is required for the formation of $AuCl_4^-$, for example, (to oxidize Au to Au^{3+}). Moreover, Au is rather uniformly dispersed in rock-forming minerals during magma crystallization, and the ratio of its distribution between the solid phase and the melt is about one, even though it depends to a degree on the structural constitutions of the minerals [7], [9], [12].

Regrettably, we do not have data on the distribution of sulfur and chlorine in the Kuril volcanic rocks, that have been analyzed for Au, and hence are unable to examine more or less quantitatively a relationship between them. Yet, the scanty information we do have on the S and Cl concentrations in the Quaternary volcanics of some structures from the frontal volcanic zone of the Kuril islands (Table 3) can be correlated, at least qualitatively, with the intricate behavior of volatiles in the magmatic systems of the respective volcanoes and with the distribution of gold in them. For instance, the concentrations of S and Cl may vary notably (by a factor of 1.5 to 2.5) even in the rocks of identical silica content, thus testifying to the possibility that magmatic melts can undergo differentiation in terms of these elements and may lose them under certain tectonic conditions. These data might as well be indicative of an important role of sulfur during all phases of magma evolution (rather close average S concentrations in rocks of different silica contents) and of a somewhat greater role of

Table 3 S and Cl distribution in the Quaternary products of some volcanoes from the oceanward zone of the Kuril island arc, g/ton.

Rock	*Sulfur*			*Chlorine*		
	n	*Range*	*Average*	*n*	*Range*	*Average*
Basalt	5	90 – 230	160	5	310–420	350
Basaltic andesite	2	110 – 120	120	3	340–670	460
Andesite	4	140 – 240	180	5	310–700	430
Dacite	6	90 – 210	150	6	250–590	400
Rhyodacite	–	–	–	1	–	590
Rhyolite	1	–	120	1	–	470

Note. The S and Cl concentrations were determined in the rocks of the volcanoes: Ebeko (Paramushir I.), Hemo and Tao-Rusyr (Onekotan I.), Ushishir (Yankich I.), Zavaritsky (Simushir I.), Tyatya, Mendeleev, and Golovnin (Kunashir I.). The concentrations of these elements in individual samples are presented in [5].

chlorine in magmas during the late stages of their evolution (increase of the Cl average with an increasing silica content in the rocks).

Almukhamedov and Medvedev [6] demonstrated that in chambers of evolving basaltic melts, that exist largely in closed systems and quiet tectonic environments, sulfur may not be uniformly distributed but may be restricted to one or even two enriched strata. One of them, a major stratum, is usually localized in the lower part of the chamber, where magma is most enriched with iron (and Mg) and hence possesses a greater capacity for sulfur [17]. Here, sulfur occurs predominantly as sulfides and precipitates gravitationally together with early Fe-Mg minerals. In the upper part of the chamber, sulfur may accumulate too, as it migrates together with the other volatiles, primarily H_2O, Cl and F, during emanation differentiation (when it is deposited also as sulfides during magma crystallization). Obviously, these processes are responsible for the fact that maximum Au contents are observed most often either in the Kuril volcanics of lowest silica content, containing maximum amounts of phenocrysts of Fe-Mg minerals (obviously products of the lower portions of magma chambers), or in some leucocratic basalts, basaltic andesites, andesites, or even dacites, that may be considered as differentiates of basaltic magmas enriched with sulfur and other volatiles. Supportive of the latter supposition is the fact that abnormally high Au concentrations were found not in lavas but in pumice and extrusions enriched in volatiles (see Table 2). Moreover, high Au concentrations are observed more frequently in the basalts of the frontal arc zone, which were most likely derived from large and long-lived magma chambers, where the process of Au precipitation to the lower levels together with sulfur might be most active.

It is important to note that in active volcanic areas magma does not always evolve in a closed system, because magma chamers may be unlocked by tectonic movements, apart from eruptions, resulting in repeated escapes of volatiles from the melt. As shown by Frolova *et al.* [15], the "drying" of sodic tholeiitic basalt magmas in the Kurils that crystallize in crustal chambers often leads to the anorthosite trend of their differentiation, the process during which large amounts of plagioclase phenocrysts are formed in more silicic basalt differentiates with minor amounts or absence of Fe-Mg mineral phenocrysts.

Apparently, the smallest Au concentrations found in the plagioclase-rich basalts and basaltic andesites of the Kurils can be attributed to the active removal of gold together with volatiles (primarily with sulfur) during large-scale tectonic adjustments even during the early stages of tholeiitic magma evolution. These low-Au, plagioclase-bearing rocks have only been found in the frontal volcanic zone where tectonic activity was most intensive. This conclusion is supported by the occurrence of long-lived volcanic centers, intricately differentiated calderas, and earthquake epicenters in that zone, which are more numerous there than elsewhere. So, it is most likely that intensive tectonic activity promoted the removal of gold during all stages of magma evolution in that zone.

The higher average Au concentrations in the back-arc andesites might be related to a less intensive tectonic activity in that zone and hence to the enrichment of magmas in the chambers with volatiles and gold. The Au accumulation in the back-arc andesites might be facilitated by a higher alkalinity of andesitic magmas: the activity of iron (principal gold modifying agent) grows with increasing alkalinity. For this reason, as indicated in [9], gold is more actively fixed in early Fe minerals and hence is less prone to be removed from magma by volatiles.

It has been recognized by many investigators, e.g. [11], that aqueous fluids of long-lived volcanic centers are among the major Au transporting agents in the crust. Such fluids may localize gold deposits, provided Au precipitates in sufficient amounts at some geochemical boundaries. So, the ability of gold to accumulate from emanations and separate during magma evolution, these processes being promoted by a low alkalinity of the melt and a long-lived considerable tectonic activity, can and must be regarded as a weighty indication of gold mineralization. In that case, as follows from the data reported here, the frontal volcanic zone of the Kuril island arc must be recognized as an area that is more favorable than the other zones for the localization of gold deposits (this has been confirmed by some discoveries).

To sum up, our research was the first attempt to systematize and compare the gold content of Quaternary volcanics from 29 variously differentiated volcanoes (from basalts to rhyolites) in the three major volcanic zones of the southern and central Kuril island arc The Au distribution was found to be nonuniform and less dependent on the silica content than on the iron content and volatile enrichment. The differences that have been established in this research between the Au distributions in the volcanics of different volcanic zones of the arc can be correlated with differences in the alkalinity of the parental magmas and also with differences in the tectonic activity of the volcanic zones, responsible for the removal of gold from the magma together with the volatiles. These differences may be instrumental in assessing areas for potential gold mineralization.

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