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Mineralization of late Neogene Tertiary to Quaternary period related to the formation of sulphur, iron-sulphide and limonite ores in Hokkaido, Japan

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Abstract

Hokkaido is abundant in sulphur, iron-sulphide and limonite deposits believed to have been formed during Neogene Tertiary to Quaternary ages. These mineral deposits were studied geologically and the two mineralization epochs, Plio-Pleistocene and Holocene, were distinguished. The mutual relation between this Plio-Pleistocene mineralization and the Miocene mineralization related to the formation of Cu-Pb-Zn-Mn ores was then investigated from the geochemical point of view.

The results obtained are outlined as follows:

1. Sulphur, iron-sulphide and limonite deposits formed during Plio-Pleistocene age are genetically in an intimate association and frequently form composite deposits. Some of the sedimentary type limonite deposits lacking in sulphur or iron-sulphide ore, and some of the sulphur ore deposits lacking in limonite ore were interpreted as varieties of the composite type deposit, judging from the characteristics of wall rock alteration.
2. Wad deposits occasionally associated with sulphur ore deposits of Holocene age occur exclusively within the Miocene manganese metallogenic province, suggesting that the wad ore is a product of regenerating mineralization.
3. Mineralization of Miocene time in west Hokkaido initially deposited Cu-Pb-Zn-Mn ores at the Toyoha, Oe, and Inakuraishi mines along the axis of so-called Shakotan-Toya dome, and later,Bi bearing Au-Ag-Ba ores were formed at the Teine, Chitose, Koryu, Minami-Shiraoi and Otaru-Matsukura mines along the outer zone of the above-mentioned dome. Afterwards, deposition of sulphur took place at the Horobetsu, Abuta and many other mines near the center of the dome. Considerable amount of Bi is detected as minor element of the sulphur ores, implying that the sulphur ore may represent the final product of the ore solution that has precipitated the base metals during Miocene mineralization.

Introduction

Volcanism of late Neogene Tertiary to Quaternary period in Japan, especially in northeastern Japan including Hokkaido island, yielded some mineral resources such as sulphur, iron-sulphide, limonite and wad deposits as post-volcanic products. Among them, the sulphur deposit of Plio-Pleistocene age is economically most important owing to its purity and amount. Remarkably concentrated bedded sulphur ore deposits associated with massive iron-sulphide ores of this age have attracted the attention of foreign geologists, because this kind of sulphur ore deposit is found almost

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exclusively in the Japanese Island arc.

In general, limonite deposits, occasionally containing jarosite, are found around the above-mentioned sulphur ore deposits. Thus the present author attempts to discuss the genesis of these sulphur ore deposits together with the iron-sulphide, limonite and wad ores.

It is also the aim of this paper to discuss the mutual relation between the sulphur mineralization and Miocene mineralization of some metallic ores, because these two kinds of mineralization are occasionally overlapped in the same area and the behaviour of minor element suggests that there exists a close relationship between these two mineralizations.

Geologic significance of the composite type of sulphur, iron-sulphide, limonite ore deposits of late Neogene Tertiary to Quaternary period and the results obtained by geochemical study of the ores are given in the paper.

1. Previous work

There have been many reports on the origin of sulphur, iron-sulphide and limonite ores of late Neogene Tertiary to Quaternary period of Japan. Of these, the following reports may be most important.

During, the 1930's, KATO (1934, 1940) studied the genesis of sulphur and iron-sulphide ore deposits of the Matsuo, Horobetsu, Yonago, and Ogushi mines, concluding that some of the ore deposits were sedimentary type. The hanging wall of this kind of sulphur ore deposits in the Matsuo and Horobetsu mines was interpreted as the lava flow of post-mineralization age. He proposed that this kind of sedimentary type sulphur ore deposit should be called "Matsuo-Horobetsu type". WATANABE et al. (1937) and WATANABE (1940) reported the mode of occurrence of native sulphur flow erupted from Mt. Shiretoko-Iwozan in 1936.

Unfortunately, the World War II disturbed the successive geological investigation of the sulphur ore deposit in this country. However, many valuable reports on the mineralization of sulphur have been published since 1950. First, sulphur ore deposits of the Matsuo and Horobetsu mines have been restudied and the view opposed to Kato was proposed by HAYASE (1951, 1956). His impregnation theory played a role of basement for the present idea on the origin of sulphur ore deposit.

YAMAGUCHI (1952) reported the sulphur ore deposits of Hokkaido from the view point of economic mineral resources.

MUKAIYAMA (1954a, 1954b) studied the wall rock alteration related to the sulphur mineralization in the Nishiazuma and Zawo mines, and later he (1959) summarized the sulphur ore deposits of this country from the genetic view point.

In 1962, TAKAHASHI interpreted the origin of sulphur ore deposit of the Matsuo mine as impregnation-replacement type. SUZUKI (1962) pointed out that more than 70% of sulphur ore deposits of this country occur in relation with the Vd-type pyroxene andesite belonging to Nasu volcanic belt. ABE (1962) made detailed description on the modes of occurrence of sulphur, iron-sulphide and limonite ore deposits of the Abuta mine.

Origin of the sulphur ore deposit of the Ishizu mine, Gumma Prefecture has been discussed by MADO (1965) and HORIKOSHI (1965). MADO presented the genesis based on impregnation-replacement theory, while HORIKOSHI had an opposing view of exhalative sedimentary theory. As noted above, there have been conflicts between syngenetists and epigenetists on the origin and the genesis of sulphur ore deposit of Plio-Pleistocene time.

MUKAIYAMA (1970) summarized the genetic relation between volcanism and related sulphur mineralization on the basis of the properties of host rock, especially petrographic characters of altered rock, and made a classification of sulphur ore deposit.

During the same period, some authors attempted to discuss the genesis of limonite and jarosite deposits in the relation with sulphur mineralization (SAITO (1949), KATAYAMA (1953), SHIIKAWA (1960), SAITO et al. (1967) and SAITO (1967)).

Some informations on the environmental condition of formation and the mode of crystallization of the ferruginous precipitates which absorb various substances from spring water have been provided by ITO et al. (1961) and ICHIKUNI (1965). Later, SHIIKAWA (1970) investigated the origin of limonite deposits of this country using modern techniques and a developed idea.

IGARASHI et al. (1963) studied the sulphur, iron-sulphide and limonite ore deposits of the Iwao mine and concluded that these three kinds of ore deposit are genetically related to one another and that underground water and occasionally surface water play an important role to form these ores. Since then, they extended the study in Niseko mountains where a geochemical study has been carried out for water, sulphur ore and some wall rocks and the genetic relation between Neogene Tertiary mineralization and Plio-Pleistocene mineralization has been investigated. Recently, IGARASHI (1974) described a peculiar occurrence of pisolitic iron-sulphide ore from the Abuta mine, proposing a more detailed model to explain the genesis of sulphur and iron-sulphide ore deposits.

2. Volcanism and related mineralization of late Neogene Tertiary to Quaternary period

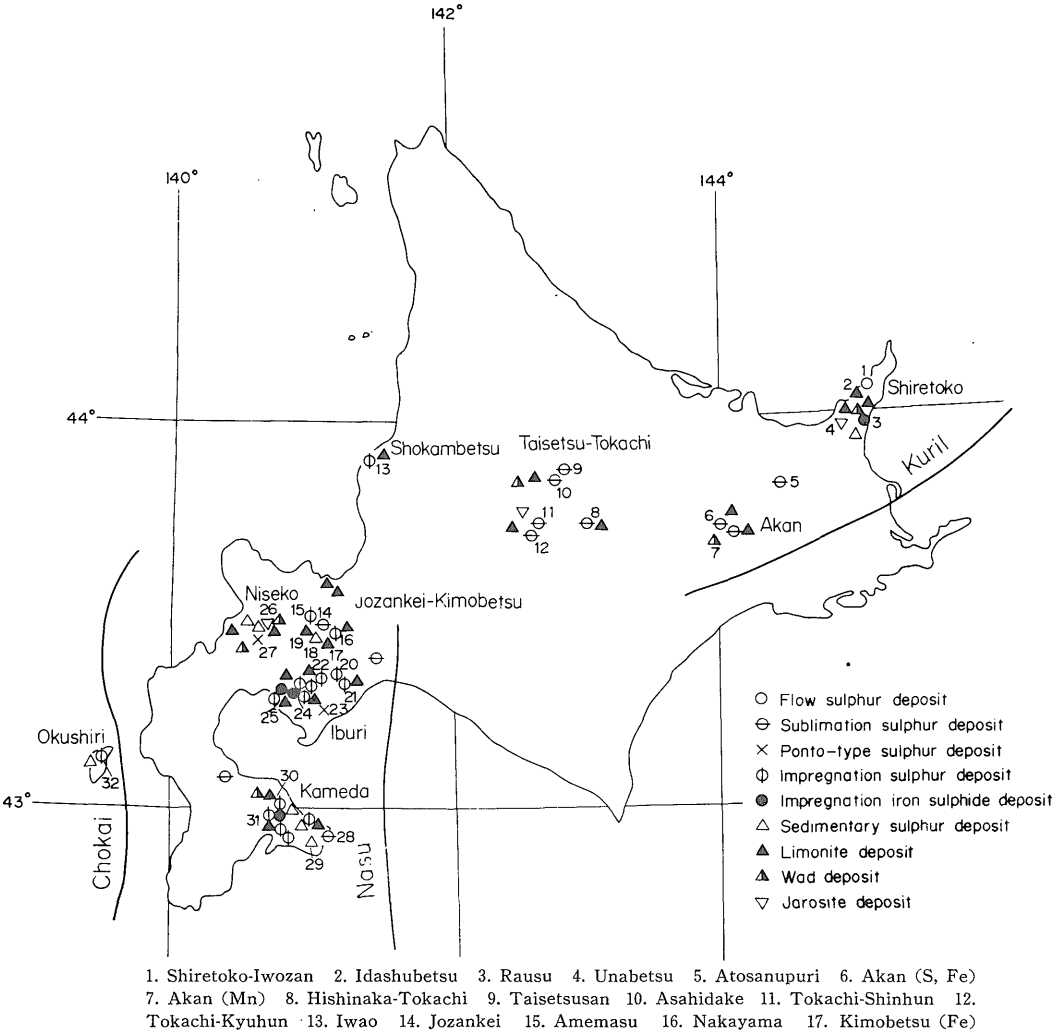
So-called green tuff region, the inner zone of Japanese island arc is characterized by the violent volcanism from Miocene time up to the present. MINATO et al. (1956) mentioned that the Quaternary volcanoes of the region represent a final geologic episode of the so-called green tuff orogenesis.

The Quaternary volcanoes in Hokkaido are divided into two major zones, i.e. the one comprises those along the Kuril arc and the other those along the northeast Honshu arc.

Petrographically, these Quaternary volcanoes are composed of a basalt - andesite - dacite - rhyolite suit, of which andesite and dacite of the calc-alkali rock series are predominant. It has been noticed that these rocks remarkably vary in nature from the Pacific side (calcic) to the marginal sea side (more alkalic) (KATSUI, 1959).

Although andesite and dacite of the calc-alkali rock series are the most abundant rocks in the Quaternary volcanic regions of Hokkaido, unnegligible amounts of basaltic rocks are intimately associated with them in many volcanoes. On the basis of occurrence of characteristic basaltic types, the above each major volcanic zone could be divided into three zones of tholeiite, high-alumina basalt, and alkali basalt from the Pacific side to the marginal sea side (YAGI and KATSUI, 1965; KUNO, 1968). Zonal classification of late Neogene Tertiary period in Hokkaido has not yet been investigated from the above-mentioned petrographic point of view.

Volcanism of Quaternary period is somewhat different from that of Tertiary period; e.g. Quaternary volcanism is seen in a limited area characterized by the presence of fractures related to the final tectonic movement of the green tuff orogenesis, and the



1. Shiretoko-Iwozan 2. Idashubetsu 3. Rausu 4. Unabetsu 5. Atosanupuri 6. Akan (S, Fe) 7. Akan (Mn) 8. Hishinaka-Tokachi 9. Taisetsusan 10. Asahidake 11. Tokachi-Shinhun 12. Tokachi-Kyuhun 13. Iwao 14. Jozankei 15. Amemasu 16. Nakayama 17. Kimobetsu (Fe) 18. Kimobetsu (S) 19. Kutchan 20. Shiraoi 21. Nittetsu-Shiraoi 22. Tokushunbetsu 23. Noboribetsu 24. Horobetsu 25. Abuta 26. Niseko-Iwaonupuri 27. Niseko-Konbu 28. Esan 29. Kobui 30. Amemasugawa 31. Shojingawa 32. Okushiri

Fig. 1 Map of Hokkaido showing the Quaternary petrographic province and the distribution of sulphur, iron-sulphide, limonite, wad and jarosite deposits of late Neogene Tertiary to Quaternary period.

majority of middle to late Pleistocene volcanoes associated with calderas have been active until present time. The representative active volcanoes in Hokkaido include Shiretoko-Iwo, Atosanupuri, Meakan, Taisetsu, Tokachi, Tarumae, Usu, Esan, Komagadake and Oshima-Oshima. On the other hand, the volcanoes of Yotei, Eniwa and Mashu are dormant though they are believed to have been formed in the same period.

In the green tuff region, especially in the above-mentioned volcanic rock area, the ore deposits displaying a trinity association of sulphur, iron-sulphide and limonite ores are known. Jarosite and wad are occasionally accompanied by these ore deposits. It is believed that these ores are the post-volcanic products, and have been formed through various processes such as impregnation, sublimation, eruption, replacement, precipitation and sedimentation. It is notable that the preponderant sulphur ore deposits generally occur in the volcanic regions such as Akan-Shiretoko and Daisetsu-Tokachi (the Kuril belt) as well as the Pacific side of west Hokkaido (the Nasu belt). These regions are of the tholeiitic and the high-alumina basaltic provinces respectively. On the con-

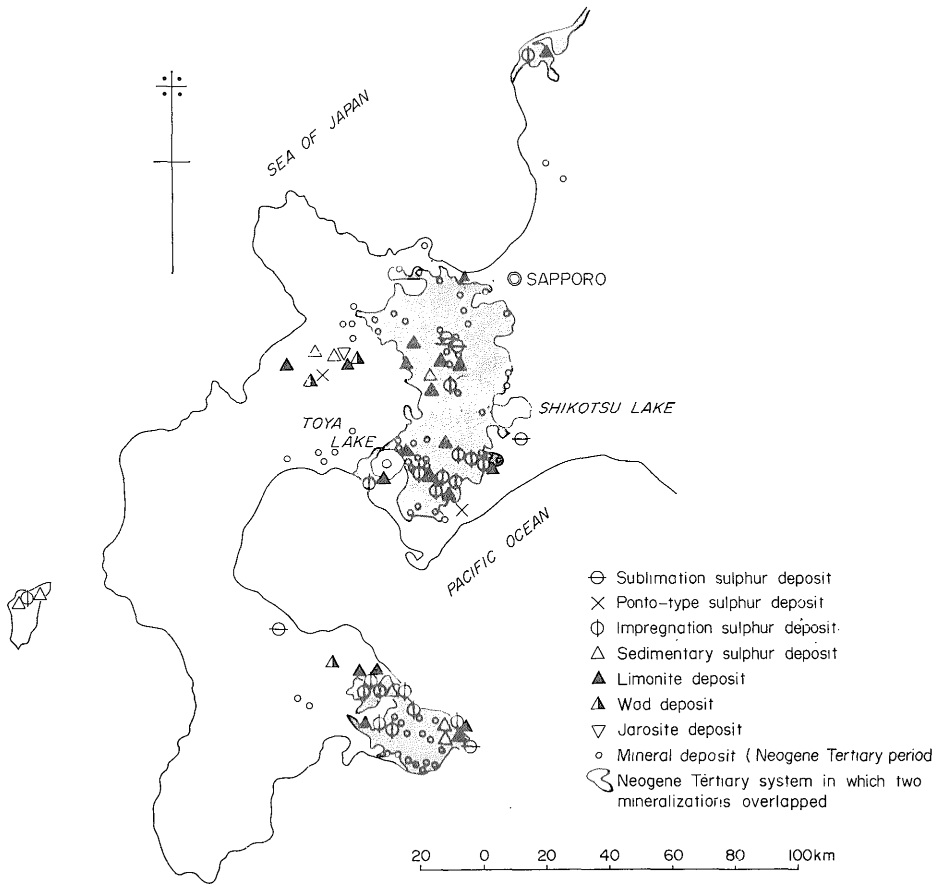


Fig. 2 Map of west Hokkaido showing the distribution of some overlapped ore deposits of Neogene Tertiary and of late Neogene Tertiary to Quaternary periods. The grey-colored field represents the extent of the area where overlapped two mineralizations are observed.

trary, in the alkali basalt province of the Japan Sea side (the Chokai belt), sulphur ore deposits are generally depleted and small scale if any (Fig. 1).

Formerly, the mineralization epoch of the above-mentioned ore deposits was briefly interpreted as late Neogene Tertiary to Quaternary period or more briefly as Quaternary period. The present author however, has proposed that the mineralization epochs of this kind of ore deposits should be distinguished as 1) Plio-Pleistocene and 2) Holocene or recent on the basis of the following evidences. That is, the sulphur ore deposits of impregnation or replacement type associated with iron-sulphide ore are predominant in Iburi and Kameda districts, west Hokkaido (Fig. 2), where the host rocks of these ore deposits are occasionally sedimentary rocks belonging to Miocene to Pliocene time. These ore deposits are usually controlled by geologic structure which is believed to have been formed during Miocene age, whereas no alteration is observed at the top of the Pleistocene formation covering the ore deposits. Based on these evidences the ore formation of these sulphur and iron-sulphide ore deposits has been interpreted as Plio-Pleistocene time. On the other hand, there are also some sulphur, iron-sulphide or limonite ores which are believed to have been formed in the recent. The sulphur erupted from Mt. Shiretoko-Iwozan in 1936 is a typical example of this type. We can also observe the pisolitic iron-sulphide ore which is growing in the hot water lake of Noboribetsu spa, and further we can find limonite ores accumulating in the swampy ground at the piedmont district of some volcanoes. These Holocene or recent mineralizations have been distinguished from the above-mentioned older one by the present author (IGARASHI, 1967).

2.1 Mineralization of Plio-Pleistocene age

Sulphur, iron-sulphide and limonite ore deposits formed during Plio-Pleistocene age are exclusively found in the Miocene green tuff region, especially in the area where the Miocene mineralization is predominant, e.g., Kutchan-Toya, Kameda peninsula area along the Nasu volcanic belt and Shiretoko peninsula area along the Kuril volcanic belt.

Massive replacement type sulphur ore deposits associated with limonite deposits are observable in sedimentary rocks of late Miocene to Pliocene time and contemporaneous andesitic lavas. Impregnation type sulphur ore is generally accompanied by massive iron-sulphide ore but these two ores are often separated from each other. However, it is considered that both ores were continuously formed under the same geologic condition.

Pseudo-stratification is frequently observed as a mode of occurrence of the sulphur, iron-sulphide and limonite ores. In general, impregnation type limonite ore occupies the top and sulphur ore the bottom of the "trinity" ore deposit. Compact iron-sulphide ore is commonly observed, being intercalated with these two ores.

It is notable that hydrothermally altered rocks are developing extensively around the ore body. This wall rock alteration is characterized by the presence of opal, cristobalite and alunite. This is one of the special characters on the mineralization of Plio-Pleistocene time because no such a widespread wall rock alteration can be seen in the

Miocene mineralization.

Sedimentary type limonite deposits are usually found in the nearby stream or swampy ground of the above-mentioned ore deposits. Mode of occurrence of these limonite ores and their topographical position indicate that the sedimentary type limonite deposit is a secondary product derived from the massive replacement type or impregnation type iron-sulphide ore deposits situated at higher levels.

As to the formation process of the sulphur, iron-sulphide and limonite deposits of massive replacement type or impregnation type, the following model would be acceptable.

1. Strong acidic and osmotic sulphuric gas migrated into underground water during its ascending and reacted with wall rocks to form the impregnation replacement type sulphur and iron-sulphide ore deposits, accompanying perhaps the wall rock alteration.
2. Subsequently, Fe ion contained in the altered wall rock migrated into acidic underground water and iron-containing sulphuric acid water was brought up to the surface. During this process, most of the replacement type limonite deposits were formed in subsurface levels.

On the other hand, the sedimentary type limonite ore is believed to have been formed on the surface by hydrous ferric oxides separated from iron-containing sulphuric acid water. In order to explain the geologic environments related to the formation of sedimentary type sulphur ore, the hot water lake such as Oyunuma of Noboribetsu spa may be presumed.

2.2 Mineralization of Holocene age

Near the top of active volcanoes belonging to the Kuril and the Nasu volcanic belts, sulphur ore deposits of sublimation-impregnation type, eruption or flow type and sedimentary type of Holocene age are observed. On the other hand, sedimentary type limonite, jarosite and wad deposits of Holocene are found in the piedmont district of active volcanoes. Among them, the sulphur ore erupted in 1936 on the slope of Mt. Shiretoko-Iwozan has been famous since the erupted sulphur ore amounted to 200,000 metric tons. In 1974, a newly erupted sulphur of small scale was observed by the present author at the same locality.

The mineralization of Holocene sulphur, iron-sulphide, and limonite ores is thought to have begun with sublimation or impregnation type of sulphur and iron-sulphide ores accompanying extensive wall rock alteration around the crater of active volcano. The iron in the altered rocks was then leached and carried by underground water as far away to the piedmont area of the volcano. The origin of limonite deposit at the foot of the active volcano can be interpreted as above. Jarosite deposit frequently associated with limonite ore and wad deposit found in the neighbouring area are both presumed to have been formed as the final product of this process.

It is concluded that the underground water plays an important role in the formation of the limonite and wad ores of this age.

3. Metallogenic province of late Neogene Tertiary to Quaternary period

In Hokkaido, sulphur ore deposits including iron-sulphide ores of late Neogene Tertiary to Quaternary period and limonite deposits of the same period are known in more than fifty localities, though some of them are overlapped (Fig. 1). Jarosite deposits in four localities and wad deposits in five localities are also known. Annual production of these deposits is given in Figure 3.

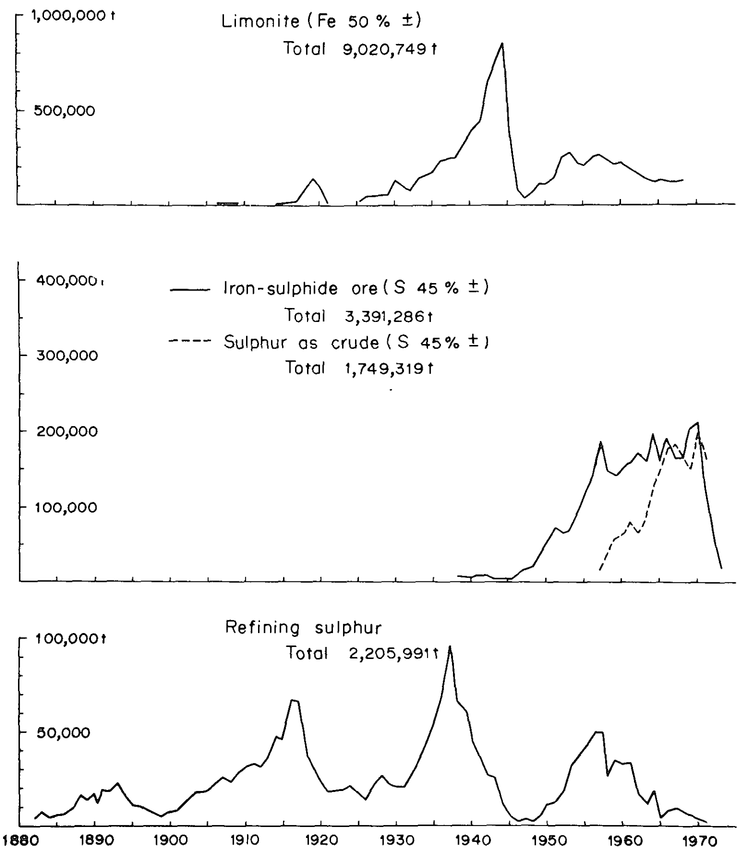


Fig. 3 Shifts of annual production of limonite, iron-sulphide and sulphur ores from Hokkaido excepting iron-sulphide ores from cupriferous pyrite and vein type deposits.

These ore deposits are distributed in some restricted areas, indicating the presence of metallogenic provinces as shown in Fig. 1. As these ore deposits were formed in relation to the volcanism of Plio-Pleistocene to Holocene ages, the metallogenic provinces are not always coincident to the Quaternary petrographic provinces.

3. 1 Kuril islands metallogenic province

1) Shiretoko peninsula district

Geology of this district is composed of Miocene-Pliocene sedimentary rocks and

Plio-Pleistocene pyroclastic materials. These formations run with NE-SW trend along the direction of the peninsula and show an anticlinal structure.

Along the ridge, active or dormant volcanoes such as Mts. Shiretoko-Iwozan, Shiretokodake, Rausudake and Unabetsudake are known. It is one of the topographic characteristics that most of the craters of these volcanoes are facing to the sea of Okhotsk. There exist sulphur ore deposits of different two ages. The sulphur ore deposit of Mt. Shiretoko-Iwozan is flow-type and that of Unabetsudake is sublimation type, both formed recently, while the deposit of Musadake situated at the southern edge of this district is sedimentary type of deposit formed in Pleistocene time.

Sedimentary type limonite deposits are also found at the piedmont areas of Plio-Pleistocene volcanoes where swampy ground is developed between Miocene-Pliocene basal sediments and overlying Plio-Pleistocene volcanic materials. The representative limonite deposits exploited to date include the Utoro, Idashubetsu, Shiretoko, Nippo and Horodomari mines.

The eroded surface of the limonite deposit of Horodomari mine is found under Pleistocene terrace deposit. The basement rock of the limonite ore is thought to be Miocene. From these evidences, this kind of limonite ore is believed to have been formed during Pleistocene time.

Iron-sulphide ore deposit of the Rausu mine situated at the southeastern corner of this peninsula is a sole occurrence in the district.

2) Akan district

Akan district running with NEE-SWW trend is situated at west of the above-stated Shiretoko district, being distributed in echelon. In this district, active volcanoes of caldera type such as Mts. Atosanupuri and Meakandake are known. Growing sulphur, limonite and wad ores are found along the caldera walls.

Sublimation type sulphur ore deposit of Mt. Atosanupuri, sublimation-sedimentary type sulphur ore deposit at the crater of Mt. Meakandake are both notable. Sedimentary type limonite and wad deposits are also found at the piedmont areas of these volcanoes.

3) Taisetsu-Tokachi district

Mountainous region in the central part of Hokkaido, which runs NE to SW and comprises Mts. Taisetsuzan and Tokachidake is called "Taisetsu-Tokachi district" in this paper. This region is geologically situated at the southwestern margin of the "Kitami green tuff region". Thus the propylite mass overlying the Hidaka Super Group is seen as the basement of Plio-Pleistocene volcanics.

Several deposits of growing sulphur, limonite, jarosite and wad related to the recent volcanism are present in this district. Sublimation-sedimentary type sulphur ore deposit is being formed inside of the crater of Mt. Taisetsuzan and sublimation type sulphur ore deposits are seen near the tops of Mts. Asahidake, Tokachidake and Maruyama.

Limonite and wad deposits have been formed at the piedmont of Mt. Tokachidake, where the limonite layer is intercalated with jarosite bands, a few centimeter in total thickness. This mineralization zone runs in NE-SW direction and is associated with many hot springs such as Yukomambetsu and Sounkyo.

3.2 Nasu metallogenic province

The province situated at the eastern half of west Hokkaido is characterized by the violent volcanism of Miocene time which formed a number of hydrothermal deposits of Au, Ag, Cu, Pb, Zn, Mn and barite. However, in the province, especially in Iburi and Kameda districts, the mineralization of late Neogene Tertiary to Quaternary period formed sulphur, iron-sulphide and limonite deposits, which are partially overlapping with the preceding metallic ore deposits. Four metallogenic districts may be recognized in the province as follows:

1) Jozankei-Kimobetsu district

In this district, a number of profitable sulphur, iron-sulphide and limonite ore deposits of Plio-Pleistocene age are known. In general, these three kinds of ore deposits occur in intimate association with one another. However, there also exist some exceptions, thus the ore deposits in Kutchan, Pepenai and Kimobetsu are composed only of limonite ores. The sedimentary type limonite deposit of the Kutchan mine had been the biggest of this type in this country. The exploited refined ore from the mine amounted to 3 million metric tons.

It is worthy to note that the widespread hydrothermally altered rock consisting of quartz, pyrite and clayey minerals occurs beneath the limonite deposits, even of sedimentary type. This fact indicates that the mineralization forming the limonite ore is partially related to replacement processes by hydrothermal solution.

Sulphur ore deposits of impregnation-replacement type of Plio-Pleistocene time are found near the Nakayama-pass between Sapporo city and Toya-lake. They are small in scale but the hydrothermal alteration is very distinct around the ore deposits.

Sublimation type sulphur ore deposit of Holocene age is observed in the vicinity of Toyoha mine. It occurs in Miocene propylite as stockwork veins along faults. The same kind of sulphur mineralization is also found at Ogawa, Yunosawa and Sokeishuomanai, but all of them are economically not very important. In the case of Ogawa, lead zinc ore veins are observable together with sulphur ore.

It is notable that the ores such as sulphur, iron-sulphide and limonite related to the younger Holocene mineralization are limited within the area of Jozankei and the ores related to the Plio-Pleistocene mineralization are found only outside of the Jozankei area. It is also notable that Pleistocene volcanics forming flat plateau develop along the boundary between the above-mentioned two areas.

2) Iburi district

Profitable sulphur, iron-sulphide and limonite ore deposits of post-Pliocene period are concentrated in this district, thus this district has been considered as one of the most important metallogenic areas for sulphur and iron resources in Hokkaido. Sulphur, iron-sulphide and limonite ore deposits in large scale occur in the area extended southwards from the line connecting Toya-lake and Shikotsu-lake. In the mineralized zone running with NEE-SWW trend, the volcanoes of Plio-Pleistocene age such as Mt. Orofure and Mt. Tokushumbetsu are known. The important ore deposits are localized at the piedmont areas of these volcanoes.

Impregnation-replacement type sulphur ore deposits of the Horobetsu and Benkei mines are located at the foot of Mt. Orofure and sulphur and limonite ore deposits of the Shiraoi, Nittetsu-Shiraoi and Tokushumbetsu mines are situated at the foot of Mt. Tokushumbetsu.

In the Horobetsu mine, large scale sulphur ore deposit is associated with iron-sulphide ore. On the other hand, in the Benkei mine, sulphur ore deposit is small in scale but is associated with limonite ore.

Limonite deposit recently formed is found in the Konai mine situated at the western foot of Mt. Orofure. The ore bodies are distributed in the swampy ground which was formed by land sliding. Iron-containing acid water is still depositing limonite ores around some welling orifices. Estimated crude ore of this mine amounts to around 180,000 metric tons.

Around Mt. Tokushumbetsu, impregnation-replacement type sulphur ore deposits associated with large scale limonite deposits are known. They occur in andesitic rocks of Miocene and Plio-Pleistocene ages. The ore deposits are overlaid with Pleistocene unaltered andesitic lava. Thus it is considered that the deposits were formed during Pliocene age. Limonite deposits of the Tokushumbetsu and Nittetsu-Shiraoi mines are of sedimentary type and have remarkable extents.

The Abuta mine situated at the western side of Toya-lake had been exploited for impregnation-replacement type sulphur, iron-sulphide and limonite ores. The host rocks of the ore deposits are Miocene to Pliocene sedimentary and volcanic rocks. A typical trinity association of sulphur, iron-sulphide and limonite ore deposits is observed in this mine. The mode of occurrence of these ores was described by ABE (1962), and the detailed description of the peculiar pisolitic iron-sulphide ore and its genesis have been given by the present author (IGARASHI, 1974).

In Noboribetsu spa, the sublimation type sulphur and pisolitic sulphur are still growing in a hot water lake, Oyunuma.

3) Niseko district

Around Niseko volcanoes, sedimentary and sublimation type sulphur ore deposits of Pleistocene age, and sedimentary type limonite, jarosite and wad ore deposits of Holocene age are known. The distribution of these ore deposits is restricted to two areas: Mts. Iwaonupuri-Chisenupuri area (the east area) and Mt. Raiden area (the west area). The former is economically more promising than the latter, and most of the profitable ore deposits to date have been found in the former area. In the west area, only small scale sedimentary type limonite and jarosite deposits are known.

4) Kameda peninsula district

Geology and ore deposits of this district are similar to those of Iburi district.

The basement of the district is composed of pyroclastic rocks of Miocene to Pleistocene ages, running parallel to the elongation of the peninsula, or, NW-SE. Volcanoes Yokotsudake and Maruyama of probably Plio-Pleistocene age have sulphur and limonite deposits at their piedmont areas. On the other hand, active volcanoes such as Mts. Komagadake and Esan occuring in the north-western part and the southeastern margin of the peninsula respectively, yield some sulphur and wad ores, e.g., sublimation type sulphur ore deposits in Kaminigorigawa and Esan, and wad deposit in Komagadake.

At the foot of Mt. Yokotsudake, impregnation-replacement type sulphur ore deposits of Plio-Pleistocene age are known. They occur in sedimentary rocks of Miocene to Pliocene time and the contemporaneous volcanic rocks. These ore deposits had been exploited at the Hakodate, Shojingawa and Amemasu mines.

Sedimentary type sulphur ore deposits of Pleistocene age are found in Kumadomari and at the foot of Mt. Maruyama. The ores were exploited by the Kobui, Okaji and Kinaoshi mines. Small scale limonite deposits of sedimentary type found in Akanuma, Kikyono, Amemasugawa, Furube and Todohokke areas are considered to have been formed during Plio-Pleistocene time.

5) Shokambetsu district

Geology of this district consists mainly of andesite lavas of Miocene to Pliocene age, in which impregnation type sulphur and iron-sulphide ore deposits and sedimentary type limonite deposits are known. They are not so profitable due to their small scale, while the wall rock alteration characterized by silicification is widespread. The alteration zone extends with a trend of N70°W, being parallel to the anticlinal axis of the basement volcanic breccia.

3. 3 Chokai metallogenic province

The area contiguous to the west of the so-called west Hokkaido belongs to the alkali-basalt petrographic province. In the area, especially in the northern part of the Okushiri island, several sulphur ore deposits are found.

1) Okushiri island district

The Horonai mine previously exploited for sulphur ore is located in the northern part of this island. Although the detail of the deposit is not very clear today, the sulphur ore seems to be of sedimentary type because the remnant of the ore is concordantly found in the Plio-Pleistocene Katsuma Formation.

Along the Oiwaoi-river, a small scale impregnation-replacement type sulphur ore deposit of Plio-Pleistocene time has been known. But it is not workable due to the small quantity of reserve.

4. Behaviour of the trace elements related to the mineralization of late Neogene Tertiary to Quaternary period

The impregnation-replacement type sulphur and iron-sulphide ore deposits of Plio-Pleistocene age are generally distributed in the area where the mineralization of Miocene age is predominant, implying that there exists an intimate relation between the two mineralizations. The similar situation may also be expected with the Holocene mineralization in the district (IGARASHI and YOKOTA, 1970; IGARASHI, 1971).

In order to throw some light on this problem, the author has examined the trace elements of sulphur ores. The ores from sedimentary type sulphur and limonite deposits were omitted in this study because of their possible complication caused by the involvement of water or by the long process of sedimentation.

The trace element analysis was carried out with the emission spectrograph method using Shimazu QF-60 quartz spectrograph with a universal source unit. The specimens were ground to 250 mesh and were mixed with sodium chloride in the ratio of 1:1. About 0.1g of the mixed sample was loaded into a carbon cathode. A pointed carbon anode was used to give an electrode gap of 3mm and the exposure time was 60 seconds. Intensities of the spectral lines were estimated by Shimazu projecting photometer PD-20. Quantitative analyses of some trace elements were carried out by the atomic absorption spectrograph method for comparison.

The sulphur ore is composed mainly of sulphur and iron-sulphide but is always associated with quartz, opal, tridymite, cristobalite, alunite and some clay minerals. Considering the possible interference due to these accompanying minerals, the analysis was carried out only on pure sulphur ores. Three types of ore were chosen for the examination: (1) the sublimation type sulphur ore (younger), (2) the replacement type sulphur ore (older) and (3) the impregnation type sulphur ore (so-called Takanome). Purification of the type (1) and (3) ores were done by hand picking under a binocular microscope. The ore of the type (2) was analysed as a whole. The numbers of analysed samples are 96 for the type (1) ore, 295 for the type (2) ore and 117 for the type (3) ore.

The trace elements detected in the type (1) sulphur ore include Ag, As, Ba, Bi, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sb, Sn, V, Zn and Zr. Among them, Ag, As, Bi, Co, Cu, Mn,

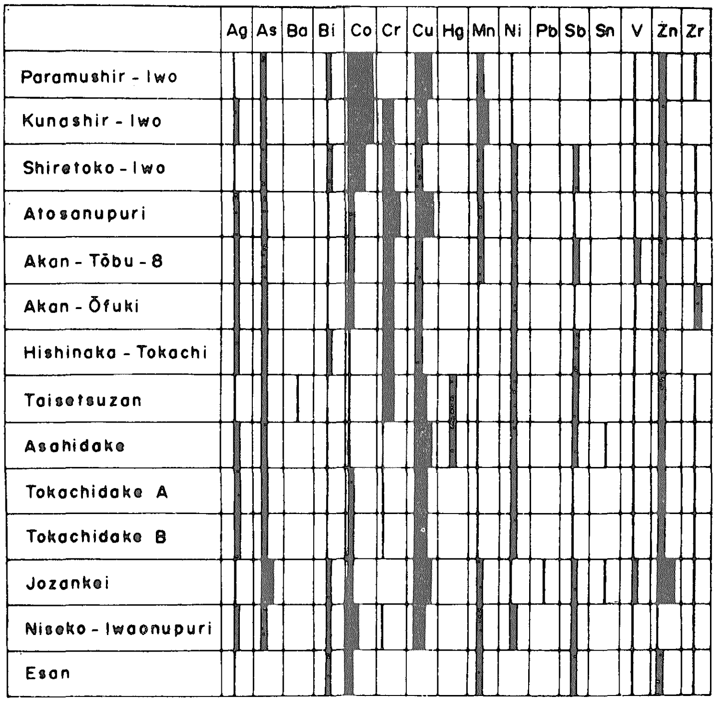


Fig. 4 Trace elements in the sublimation type sulphur ores from Hokkaido.

Ni, V and Zn are found throughout the region. Some areal characteristics may be recognized in the result (Fig. 4).

The Kuril island metallogenic province is rather rich in Co, Cr, Mn, Ni and the ores from Paramushir island and Kunashir island are most abundant in Co and Cu among the whole specimens. Cr shows its maximum in Atosanupuri of Akan district and Zr in Meakan. Hg is found only in the specimens from the central volcanoes of Taisetsuzan and Asahidake of Taisetsu district throughout the region. In the Nasu metallogenic province, much As, Co, Cu, Pb and Zn are found in the sublimation type sulphur ore from Yunosawa and Pb is exclusively found in Jozankei area. Niseko district is rather rich in Co. The ores from Esan of Kameda district show little, if any, content of As, Cu and

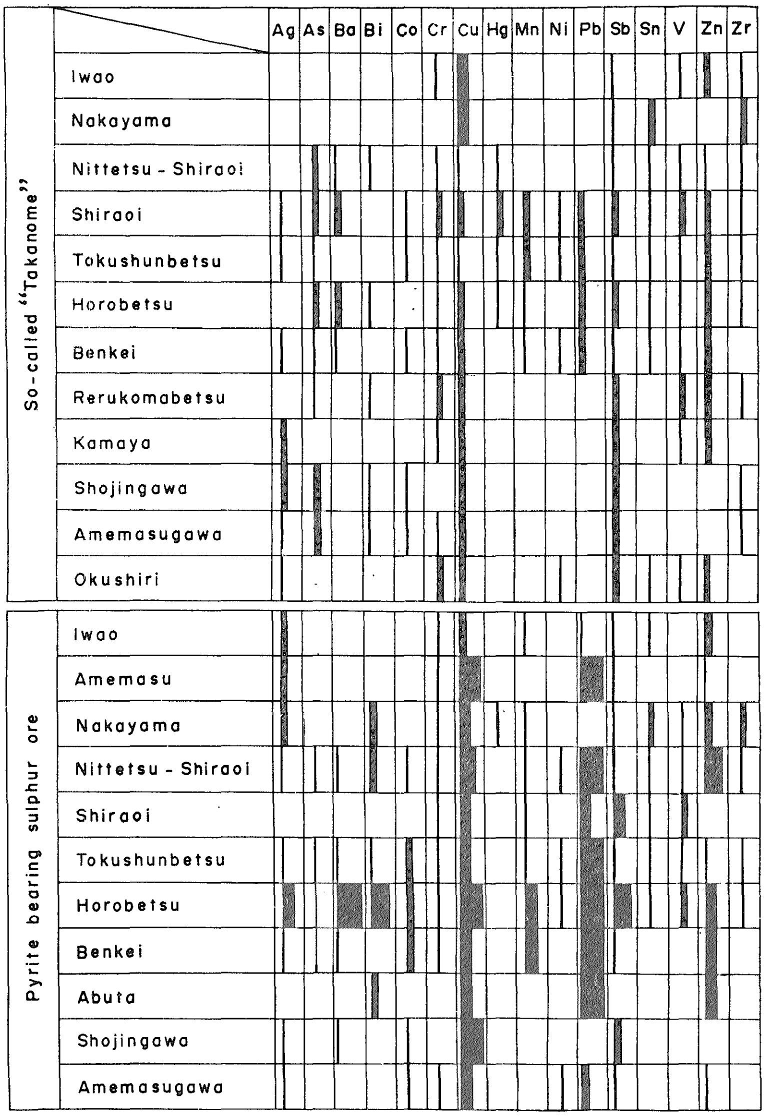


Fig. 5 Trace elements in pyrite bearing sulphur ores (type 2) and pure sulphur (so-called Takanome) from Hokkaido.

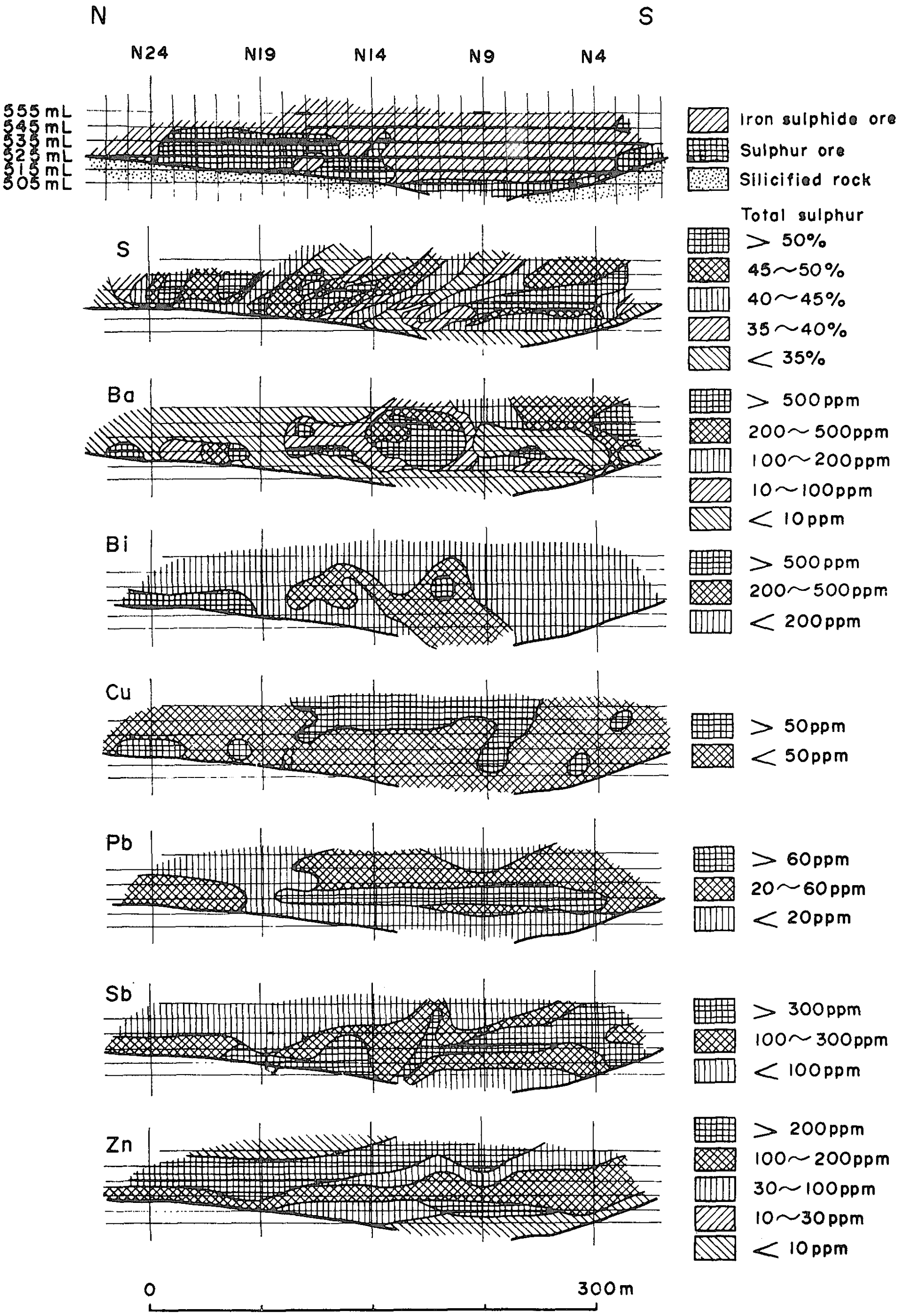


Fig. 6 The uppermost profile shows the vertical distribution of sulphur, iron-sulphide ores and altered wall rock. The second is assay section of sulphur, and the others represent modes of distribution of some detected trace elements in sulphur ores from the gallery E-25, Daisan ore body, Horobetsu mine.

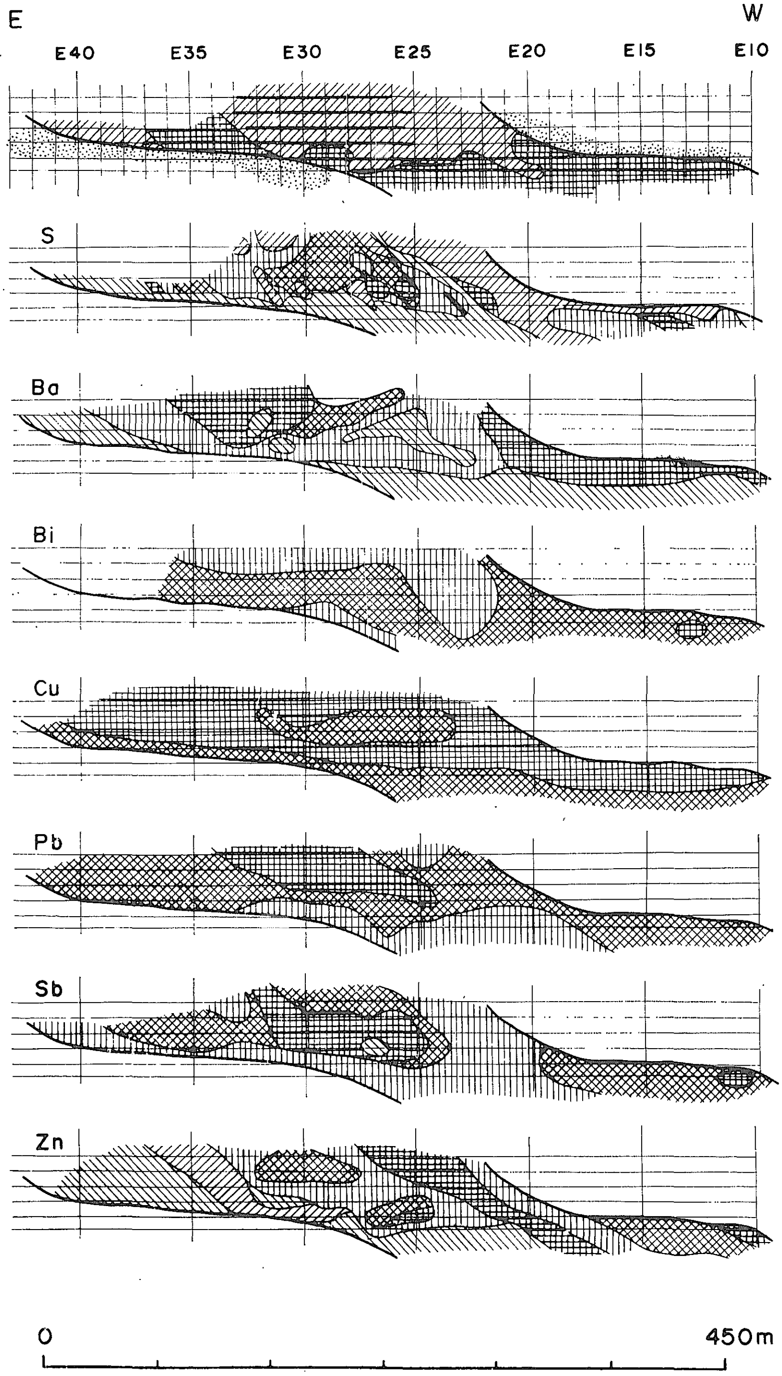


Fig. 7 The uppermost profile shows the vertical distribution of sulphur, iron-sulphide ores and silicified rock. The second is assay section of sulphur and the others represent modes of distribution of some detected trace elements in sulphur ores from the gallery N-9, Daisan ore body, Horobetsu mine. Legend is same as Fig. 6.

Ni, although these elements are common in other district.

The kinds of trace element in the type (3) sulphur ore are almost the same with those observed in the type (1) ore. Cu and Sb are found throughout the region. As, Cr, Sn, V, Zn and Zr are also found regionally. In the vicinity of Iburi, the Nasu metallogenic province, the ores of this type from the deposits, such as Nittetsu-Shiraoi, Tokushumbetsu, Benkei, Shiraoi and Horobetsu have fairly high contents of As, Ba, Bi, Cr, Cu, Pb and Zn. The Shiraoi deposit is characterized by the abundance of Hg (Fig. 5).

The trend in the type (2) ore is rather similar to those of pure sulphur ores of the type (1) and (3). Sulphur ores from the Shiraoi, Nittetsu-Shiraoi, Tokushumbetsu, Benkei and Horobetsu mines have relatively high contents of Cu, Pb and Zn. Especially the ore from the Horobetsu mine has considerable amounts of Ag, Ba, Bi and Sb (Fig. 5).

The variation in the trace element distribution in a single ore body has been observed in the Horobetsu and Nittetsu-Shiraoi ore deposits. Ores from the central to upper part of the ore body of the Horobetsu mine has higher contents of Bi, Cu, Cr, Pb and Sb compared with those from the marginal part. In the Nittetsu-Shiraoi mine, Cu increases to the midst of the ore body, while Ba, Bi, Cr, Pb and Sb are found to increase toward the lower part. Figs. 6 and 7 are examples of the trace elements variation in one ore body (the Daisan-ore body, Horobetsu mine).

It was found that the trace element distribution in the sulphur ores shows some regularities with regard to their mineralization age and metallogenic province. It is noteworthy that the ores of Plio-Pleistocene replacement type deposits are characterized by the presence of metal elements which are found in the preceding Miocene mineralization.

5. Type classification of ore deposits and their characteristics

5.1 Type classification of ore deposits with reference to their genesis

The ore deposits formed during late Neogene Tertiary to Quaternary period in Hokkaido include native sulphur, iron-sulphide, limonite, jarosite and wad deposits. These ore deposits are genetically classified as follows:

1) impregnation-replacement type
2) sublimation type
3) sublimation-sedimentary type
4) flow type
5) sedimentary type

Table 1 shows the classified types of the ore deposits.

It is characteristic that in most cases two or three kinds of these ore deposits occur together, suggesting a close genetical relationship between them. There often occurs, in Plio-Pleistocene time, the composite deposit consisting of impregnation-replacement type sulphur ores and sedimentary type limonite ores accompanying occasionally iron-sulphide and/or limonite ores of impregnation-replacement type.

In Holocene age, the composite deposits of sublimation, sublimation-sedimentary or flow type sulphur ores and sedimentary type limonite ores are common, the former being

Table 1 Types of sulphur, iron-sulphide, limonite, jarosite, wad ore deposits of late Neogene Tertiary to Quaternary period and related mines.

ore deposits of Plio-Pleistocene	
ore deposits mainly formed by impregnation-replacement	iron-sulphide ore deposit (Rausu)
	sulphur ore deposit (Shiraoi, Hakodate)
	sedimentary type limonite deposit (Nittetsu-Shiraoi)
	sulphur, iron-sulphide ore deposit (Horobetsu, Takinosawa)
	sedimentary type limonite deposit (Shojingawa, Amemasu)
	sulphur, iron-sulphide, limonite deposit
	sedimentary type limonite deposit (Iwao, Abuta)
	sulphur, limonite deposit (Benkei)
	sedimentary type limonite deposit (Tokushumbetsu)
	limonite deposit (Asari)
ore deposits mainly formed by sedimentation	sulphur ore deposit (Kimobetsu, Niseko-Chisenupuri)
	**impregnation type sulphur ore deposit (Musa, Niseko-Iwaonupuri, Kobui, Okushiri)
	*limonite deposit (Idashubetsu, Kutchan, Konai, Nakatoya)
	*jarosite deposit (Unabetsu, Horodomari)
ore deposits of Holocene	
ore deposits mainly formed by sublimation-impregnation	sulphur ore deposit (Unabetsu, Atosanupuri)
	sedimentary limonite deposit (Esan)
	sedimentary type wad deposit (Asahidake)
	sedimentary type limonite, jarosite, wad deposit (Tokachidake)
	***composite deposit with sedimentary type sulphur ore deposit (Niseko-Kombu, Taisetsu, Noboribetsu)
	sedimentary type limonite, wad deposit (Meakandake)
	**sedimentary type limonite, jarosite, wad deposit (Niseko-Iwaonupuri)
	flow type sulphur ore deposit (Shiretoko-Iwozan)
sedimentary type wad deposit (Nishikioka, Komagadake)	

After Igarashi, T. (1974)

* These ore deposits are associated with widespread altered rocks characterized by silicification and argillization.

** Sedimentary type and sublimation-impregnation type sulphur ore deposits of Niseko-iwaonupuri mine are believed to have been formed in late Pleistocene time, and sublimation type sulphur, sedimentary type limonite, jarosite, wad deposits of the same mine are in Holocene time.

*** Among them, sulphur ore deposits of Niseko-Kombu and Noboribetsu belong to so-called "Ponto type".

located in or around the craters and the latter at the piedmont areas of volcanoes. The sedimentary type limonite deposit is occasionally accompanied by jarosite and wad ores. Jarosite is considered to have been formed at the earliest stage and wad at the last stage of the precipitation process of limonite from acid sulphuric water. Jarosite and wad, however, sometimes form independent ore bodies from limonite.

It is notable that the ore deposit of Plio-Pleistocene time is usually lacking in wad, and the Holocene mineralization has little iron-sulphide ore except the small scale impregnation around sublimation sulphur ore or the precipitate in hot water lake.

The ore forming processes of these deposits may have been as follows: ascending sulphuric gas had initially formed native sulphur, iron-sulphide and limonite ores through the processes of impregnation, sublimation and replacement, and then limonite, jarosite and occasionally wad ores were formed by precipitation from acid sulphuric water containing these materials. Though there exist some chronological and spatial differences between them, the wall rock alteration connecting these two groups of ore deposit suggests that the two processes have been closely related with each other. However, there also exist some examples in which all the deposits seem to have been formed only by one continuous process.

Impregnation-replacement type sulphur ore deposits in the Horobetsu mine occur without limonite ore and, on the other hand, in the Kutchan and the Nakatoya mines, limonite deposits free of sulphur and iron-sulphide ores occur. The genesis of these ore deposits may be explained as follows. The sulphuric gas of volcanic origin formed not only sulphur ore but also the widespread altered zone showing zonal structure. The iron contained in the host rock reacted easily with the successively ascending sulphur to form iron-sulphide ores displaying a pseudostratified structure overlying the sulphur ore body. When the amount of iron in the alteration aureole had been large, massive pyrite ores were formed instead of disseminated pyrite. If the sulphur ore deposit was formed in deep niveau, or if the deposit was overlaid with lava flows immediately after the ore formation, the ores should have escaped from oxidation (Horobetsu mine). On the contrary, if the iron-sulphide ores were erroded out and carried to the other place by surface water, then a sedimentary type limonite deposit would have formed (Tokushumbetsu mine). If the sulphuric gas moves up near the surface, the materials contained in the gas diffuse away, and the altered rock with scanty sulphur and iron seem to be formed in great quantity. Leaching of sulphur and iron in these rocks leads to the for-

Table 2 Sequence of volcanism and mineralization of Neogene Tertiary to Quaternary period related to the formation of sulphur, limonite, jarosite and wad deposits in the Niseko district, west Hokkaido.

		volcanism	mineralization
Holocene	later		sedimentary type sulphur or Pontoh type sedimentary type wad sedimentary type limonite, jarosite sublimation-impregnation type sulphur sedimentary type sulphur
	earlier	Koiwaonupuri dome Iwaonupuri dome Chisenupuri mud flow Chisenupuri dome	sublimation-impregnation type sulphur
Pleistocene	later	Iwaonupuri ejecta Chisenupuri ejecta Nitonupuri ejecta Shakunagiyama ejecta Nisekoannupuri ejecta Weiss-Horn ejecta	sedimentary type sulphur sublimation-impregnation type sulphur
	earlier	breccia	

(After IGARASHI, T. 1970)

mation of a peculiar type of altered zone characterized by the presence of opal. Ore deposits of the Kutchan and the Nakatoya mines are characterized by such widespread opalized rocks accompanying scanty pyrite and native sulphur. It appears that this opalized rock once had the iron-sulphide ore in it. Therefore, though the limonite deposit lacks in sulphur or iron-sulphide ore, it seems likely that the deposit had been formed in close relation with the mineralization which formed sulphur or iron-sulphide ore.

The relation between the two mineralizations of Plio-Pleistocene time and of Holocene time has been clarified in Niseko volcano area by the present author (IGARASHI and YOKOTA, 1970). Here the volcanism began in Pliocene time and the main volcanic bodies were formed in Pleistocene. Finally the lava domes in craters were formed in the volcanoes such as Chisenupuri, Koiwaonupuri and Iwaonupuri. Some sublimation-impregnation type sulphur ore deposits and, in certain places, sedimentary type sulphur ore deposits were formed during this period by the post-volcanic activity. They were then followed by the formation of sedimentary type limonite, jarosite and wad deposits, and this mineralization is still observable at present in some places. Thus it is concluded that some of the Plio-Pleistocene mineralizations have been continuing up to present.

The relation between volcanism and related mineralization in Niseko volcano area is summarized in Table 2.

5. 2 Notes on some typical ore deposits

1) Flow type sulphur ore deposit of the Shiretoko-Iwo mine

The mine is situated at Kamuiwakka, Shari town. An eruptive crater exists on the northern slope of Mt. Shiretoko-Iwozan at about 620m above sea level. Sulphuric gas and water vapour always fill the crater and sublimated sulphur is seen to be depositing. The crater erupted molten sulphur in 1876 and 1889, up to the amounts of several tens of thousand tons. After a period of quiescence, then, in February, 1936, a big eruption came to form two hundred thousand tons of flowed sulphur deposit in eight months. The eruptions were repeated intermittently in a few tens of times bringing out several hundred to fifteen hundred tons of sulphur in each eruption. Consequently, the river Kamuiwakka located on the western slope of the crater changed into a big river of native sulphur. Such a large scale intermittent eruption of sulphur has not been known in the world.

The mechanism of sulphur eruption is considered as follows by WATANABE (1937, 1940): Prolonged sulphur impregnation in the vicinity of crater was followed by melting of sulphur by an activated volcanism. When the molten sulphur accumulated in the cavity under the crater, the sulphur went to erupt by the action of water vapour and underground water, by the mechanism analogous to that of a geyser. The process might be called the natural dressing of sulphur by water vapour.

LYMAN (1877) reported that he already recognized the presence of molten sulphur in the crater of this volcano when he visited there in 1874, before the first eruption in

1876.

The erupted sulphur in 1876 had been mined for three years from 1878. Tens of thousand tons of flowed sulphur from the 1889 eruption was mined out within the succeeding four years. The big river of sulphur formed in 1936 had been worked until 1943 and produced 116,523 tons of crude ore, that is, 96,208 tons as refined sulphur (S content: 82.5%).

2) Sublimation-sedimentary type sulphur ore deposit of the Akan-Iwo mine

The mine is situated at Moashoro, Ashoro town, Ashoro district. The sulphur ore deposit of this mine is in an explosion crater of Mt. Nakamachineshiri, located at 1 km northeast from the top of Mt. Meakandake. The mode of occurrence indicates that the mineralization is presumably related to the formation of central cone in the crater.

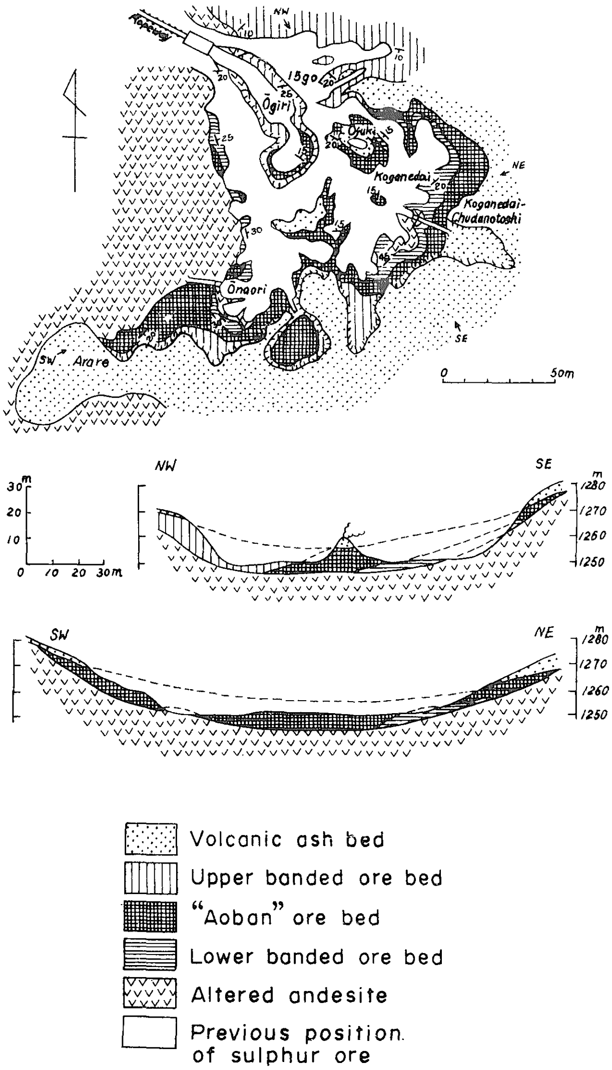


Fig. 8 Geological map of the Ofuki sulphur deposit, Akan-Iwo mine (after J. SUZUKI, 1957).

The explosion crater having dimensions of 150 m (EW) by 200 m (NS) led to the formation of sedimentary type sulphur ore deposit related to a hot water lake. In the central part of the lake, sublimation of sulphur occurs along a channel way. This particular area forming sublimation type sulphur ore is called "Ofuki" (Fig. 8).

Sedimentary type sulphur ore deposit occurred in the basin of crater is more than 20m thick at its center and the thickness decreases gradually towards the margin of the basin. Before exploitation, the growing sublimation type sulphur ore deposit was only observed in the explosion crater. Thus nobody has assumed the presence of sedimentary type sulphur ore deposit beneath it.

The sedimentary type sulphur ore deposit is composed of (a) lower banded ore bed, (b) blue coloured ore bed (Aoban ore bed) and (c) upper banded ore bed in ascending order as illustrated in Fig. 8. These are overlaid with the sublimation type sulphur ore.

Lower banded ore bed is compact and grey to grey-white in colour. This consists of tuff and sulphur intercalated with clays derived from tuff. The clay layers are found rather in the marginal part of the deposit where the bed shows the dip of 10–20° towards the center. Grade of this sulphur ore is estimated as 25 to 30%.

Blue coloured ore bed is composed of sulphur, tuff, volcanic breccia, pumice and scoria erupted from Mts. Nakamachineshiri and Ponmachineshiri. The grade of this sulphur ore is highest among all the ores of this mine, being estimated as 50 to 60%. Main portion of this ore is regarded to have been formed by impregnation or replacement. The pores, 0.5cm in diameter, are occasionally filled with native sulphur. Microscopic study has clarified that the ground mass of ore is mostly composed of sulphur and rock fragments characterized by the presence of opal.

Upper banded ore bed consists of coarser-grained materials compared with the lower banded ore, and is intercalated with iron-sulphide bands. The grade of sulphur is estimated around 30%. The sublimation type sulphur ore occupying the top of the composite deposit is composed of aggregates of acicular form sulphur and pisolitic sulphur having 1–2cm in diameter.

The origin of the sulphur ore deposit of this mine is presumed to be as follows. First, the explosion crater was filled by hot water and banded sulphur ore precipitated in the bottom of the lake. Afterwards, the sedimentary type banded ore was overlaid with the materials from new eruptions. At the channel way of sulphuric gas, central part of the crater, the accumulated material was thickned according to the sinking of the basement. Sulphuric gas replaced the erupted materials forming the blue coloured ore bed. Hot water lake was newly formed at the surface of the blue coloured ore bed and the upper banded sulphur ore bed was formed. This newly formed ore bed played a role of cap rock for the successively ascending sulphuric gas. Consequently, enrichment of sulphur was enhanced in the blue-coloured ore bed. In addition, sublimation type sulphur ores were formed as the result of reaction between hydrogen sulphide, sulphurous acid gas and oxygen or water.

The shape of the channel way for ascending sulphuric gas, or the so-called Ofuki, has been clarified to be chimney-like, having about 15m in height and 2m in diameter, as the exploitation of ore has proceeded.

According to some authors, this sulphur ore deposit has been known since 1802. After B. S. LYMAN made a first geologic investigation, the exploration was carried out since 1876. Modern era of sulphur production from this mine began in 1951. Since then 777,362 metric tons of crude sulphur ore (25.9%) had been mined till 1963. The amount is the biggest among the sedimentary type sulphur ore deposits of Neogene Tertiary to Quaternary period in Hokkaido.

3) Sedimentary type sulphur ore deposit of the Kobui mine

The mine is situated at Hinohama, Shirikishinai town, Kameda district. An outcrop of impregnation type sulphur ore deposit is known in a tributary, Ishibashi-zawa, of the Kobui river, 8km north from Hinohama.

Small scale sulphur ore deposit of sedimentary type is also found in another tributary, Kyuyama-zawa in the neighbouring area. The sulphur ore deposit of the Kobui mine is situated at upper stream of the Kyuyama-zawa, 1km from the outcrop mentioned above. This is the biggest sulphur ore deposit in the Kameda peninsula district. Though the ores of this mine have been mostly mined out, remnants of the ore in the open-pit suggest that the extent of ore bed was as long as 400m along the strike (N-S), and 100m to the dip (15–25E).

OHINATA (1911) previously reported the mode of occurrence of this ore deposit as follows. The thickness of the ore bed was 0.6m to 7m and the ore bodies were intercalated with tuffaceous layers so that lamella and banded structures were distinct. Occasionally native sulphur occurred in veins or as scattered patches.

Sulphur ore deposit of the Kyuyama-zawa mine occupied a hollow, 50m in diameter, where opal zone and kaolin zone were extensive around the ore bodies.

The sulphur ores of these mines were mostly exploited during Meiji era. After the World War II, a small amount of remnant sulphur ore was mined. The production of each period has been reported as follows:

1904–1917: 169,935t (as refined sulphur)
1951–1952: 1,295t (as crude ore).

Grade of crude ore was between 25 and 60% S.

4) Impregnation-replacement type sulphur, iron-sulphide ore deposit of the Horobetsu mine

This mine is situated at Wokei, Sobetsu town, Usu district. The mining area is located in the midstream of the Benkei river being 530m above sea level.

Geology of the mine is composed of green tuff, plagioclase-bearing rhyolite tuff and andesitic lava belonging to the Osaru formation of Miocene time. This formation is overlaid with a pyroxene andesitic lava intercalated with tuff breccias of Pliocene age. These formations are covered with Pleistocene andesitic lava. Alteration related to the sulphur and iron-sulphide mineralization of this area is observed in the older two formations. The Pleistocene formation, however, has not suffered any alteration.

The sulphur and iron-sulphide ore deposit of this mine is impregnation-replacement type (HAYASE, 1957), being formed in Miocene dacitic lava and Pliocene pyroxene andesitic lava. The deposit extends in N60°E direction. Its overall dimension is estimated as 1,300m long, 200–400m wide and 20–60m thick (Fig. 9). Lower part of the ore body is mainly composed of sulphur ore[1] (Fig. 10), while the upper part consists of iron-sulphide ore (Fig. 6 and 7). There exist three ore bodies in the deposit. They are called Daiichi ore body (sulphur), Daini ore body (sulphur, iron-sulphide ore) and Daisan ore body (sulphur, iron-sulphide ore) from west (Fig. 9).

Altered rocks around the ore body can be discriminated into the following four zones on the basis of their characteristic alteration minerals. They are outwardly (a) powdery silicified zone, (b) opal-alunite zone, (c) kaolin zone and (d) montmorillonite zone.

From the appearance, the ores can be classified into three kinds, i.e., grey coloured ore, yellow coloured ore and banded ore. The sulphur ores are generally accompanied by such minerals as pyrite, marcasite, orpiment, realgar, opal, cristobalite and tridymite. The banded ore, most predominant, is further discriminated into rhythmic ring ore, fine-grained ore and coarse-grained ore.

The iron-sulphide ore is generally powdery and loose, and the colour is greyish black. When quartz is associated, the ore is compact showing a shiny yellow colour. Such a

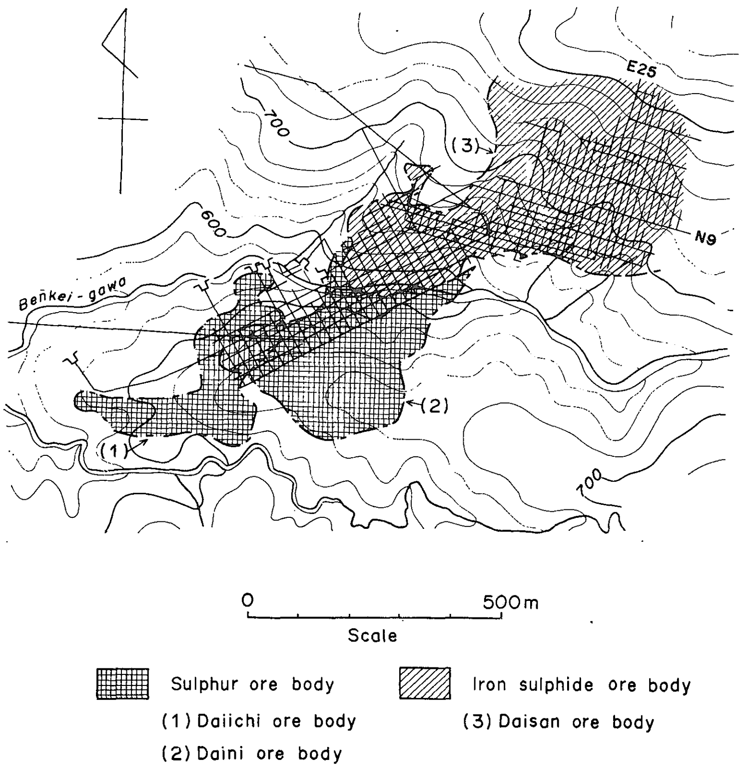


Fig. 9 Map showing the distribution of Daiichi, Daini and Daisan ore bodies in the Horobetsu mine.

1) In this mine, the ore contained more than 20% of free sulphur is called sulphur ore and the ore of lower contents is called iron-sulphide ore.

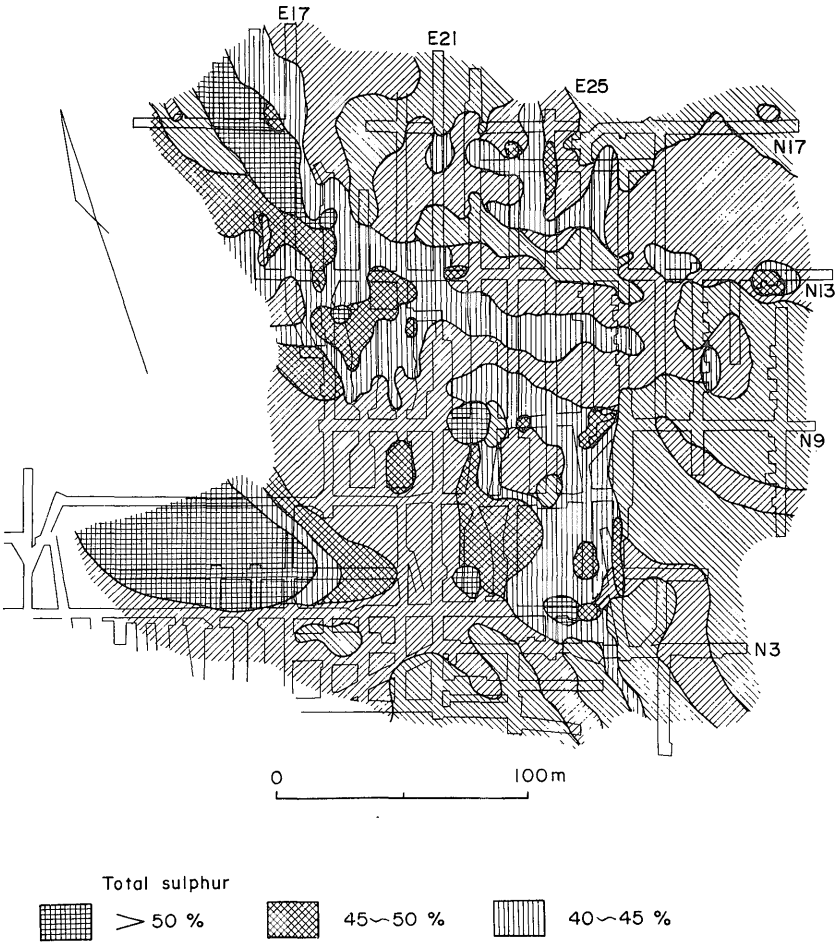


Fig. 10 Assay map of the sulphur ore deposit of the Daisan ore body at 515m level in the Horobetsu mine.

compact ore is exclusively found in the Daisan ore body. In this ore, barite and horobetsuite showing acicular crystals are occasionally observed. The horobetsuite described by HAYASE (1952, 1955) is a solid solution mineral between Sb_2S_4 and Bi_2S_3, being regarded to be a new variety of stibnite. It is notable that trace elements such as Sb and Bi are remarkably concentrated in the sulphur ore where horobetsuite exists. Sulphur content of the compact ore from the Daisan ore body is 40–47%.

The western part of this sulphur ore deposit was discovered in 1902, when mining was started. During three years from 1913 to 1916, the annual production of sulphur from this mine was in the first rank of this country. The exploitation for iron-sulphide ore began in 1938 and continued for more than thirty years. But in recent years, many sulphur mines of this country have been compelled to close due to the entry of sulphur from petroleum refinery. The Horobetsu mine was also closed in 1973 by the same reason though it has still the ore reserve enough for mining.

The total production of sulphur from this mine is given below:

Sulphur:

1913–1930	173,749t (as refined sulphur)
1931–1969	1,769,695t (as crude ore, S 37.69%)

Iron-sulphide ore:

1938–1972	1,602,572t (as crude ore, S 39.42%).

5) Composite type sulphur, iron-sulphide and limonite ore deposit of the Abuta mine

This mine is situated at Mitoyo, Abuta town, Abuta district, located to the south-

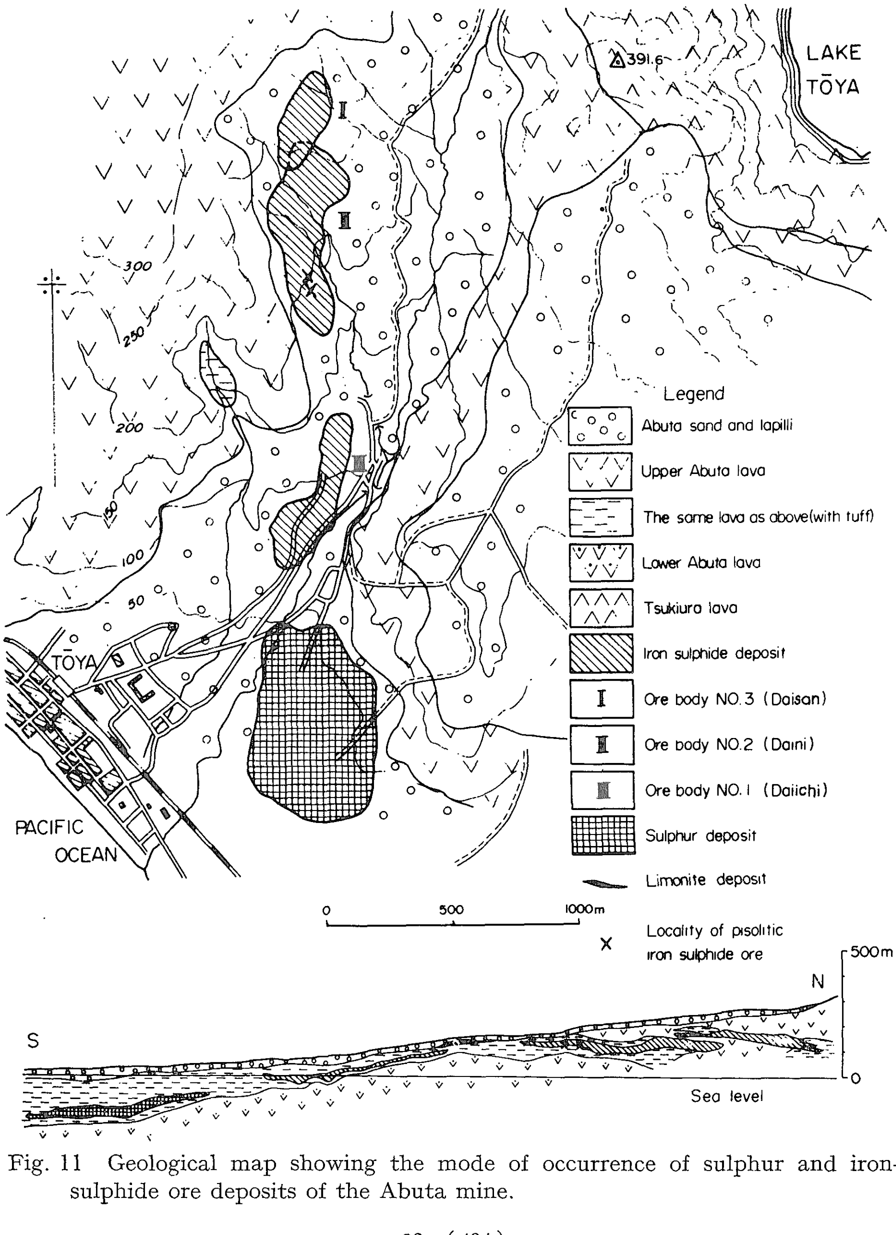


Fig. 11 Geological map showing the mode of occurrence of sulphur and iron-sulphide ore deposits of the Abuta mine.

west of Toya-lake. The exploited hill, 100–300m above sea level, is facing to the Pacific Ocean.

The composite type deposit consisting of impregnation-replacement type sulphur, iron-sulphide and limonite ore deposits associated with sedimentary type limonite deposit of late Pliocene to Pleistocene time occurs in so-called Abuta lava believed to be of late Neogene Tertiary period. This lava is composed of pyroxene andesite and andesitic tuff breccia.

As shown in Figure 11, the ore bodies being called by the names of Daiichi, Daini and Daisan, run with N-S trend. The Daiichi ore body consisting of iron-sulphide ore is overlaid with sedimentary type limonite deposit which occurs in Pleistocene loam formation. The iron-sulphide ore of the Daiichi ore body shows gradational change to the sulphur ore at its lower level according to an increase of free sulphur.

The extent of each ore body is given below:

	length	width	thickness
Daiichi ore body	700m	100–200m	40m
Daini ore body	600–800m	150m	20m
Daisan ore body	450m	150m	20m
Sulphur ore body	800m	500m	35m

The exploitation of this mine started in 1892 when sedimentary type limonite deposit was discovered. During 45 years from 1903 to 1947, the limonite ore of 530,000t (50% Fe) was intermittently mined.

In 1951, sulphur and iron-sulphide ore deposit lying beneath the limonite ore bed was newly discovered as the result of prospecting by drilling. Thus the presence of a "trinity" composite deposit consisting of limonite, iron-sulphide and sulphur ore deposits was confirmed in this mine. Soon after, the exploitation of iron-sulphide ore began and 1,690,000t of iron-sulphide ore (40% S) was mined during 16 years since 1955. In 1971, the mine was closed though the reserve of sulphur ore at subsurface levels was still much expected.

Altered rocks related to the sulphur and iron-sulphide mineralization show zonal arrangements which have the following outward sequences (Fig. 12).

(a) iron-sulphide ore body → silicified zone → alunite zone →kaolin zone → montmorillonite zone → saponite zone → chlorite zone → unaltered rock

(b) sulphur ore body → silicified zone → quartz, alunite zone → alunite, kaolin zone → kaoline zone → kaolin, montmorillonite zone

The iron-sulphide ore body is composed of several different types of ore. They are outwardly (1) compact ore, (2) rounded ore, (3) brecciated ore, and (4) powdery ore. In addition, a peculiar ore called "pisolitic iron-sulphide ore" is found in the lower part of the powdery ore (IGARASHI, 1974). Besides, small scale vein-type ores believed to belong to the last stage of mineralization are observed.

The iron-sulphide ore of this mine is mainly composed of pyrite. Marcasite is an

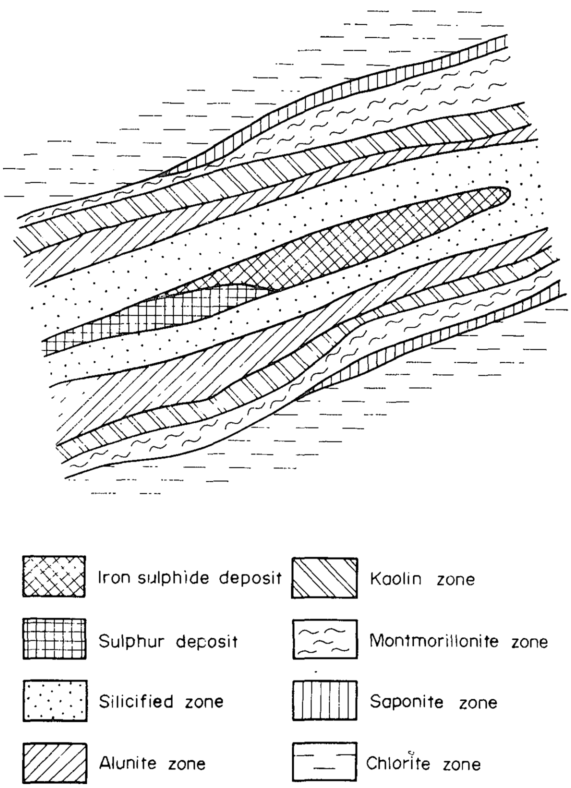


Fig. 12 Schematic profile of the altered zones in Abuta mine (after ABE, 1962).

accessory mineral. Microscopically the pyrite shows good crystallinity taking cube or octahedral form of the size of 10–15μ, occasionally up to 200μ. When gangue minerals such as calcedonic quartz, opal, tridymite, cristobalite, alunite, kaolinite or halloysite are abundant, xenomorphic pyrite crystals are rather common, showing granular aggregates. In this case, small amounts of cinnabar, and bismuthinite are occasionally found. In the montmorillonite zone lying beneath the ore bed, gypsum is associated. Sulphur content of the compact iron-sulphide ore is around 45%, whereas the others show lower grades.

Sulphur ores are generally compact and are yellow, white-yellow or greysh yellow in color. Pyrite, marcasite, opal, quartz, alunite and kaolinite are almost always and barite is occasionally observed as the accessory minerals. The grade of sulphur is 40% in average.

Limonite ore can be classified into two types: one is impregnation-replacement type and the other is sedimentary type. Supergene residual sulphur ore is rarely found near the surface where the upper most part of the iron-sulphide ore body occurs.

Impregnation-replacement type limonite ore deposit of this mine occurs overlying the above-mentioned iron-sulphide ore body. Intercalation of silicified rock or argillized rock is observable between the limonite and iron-sulphide ore bodies. The limonite ore is mainly composed of goethite and hematite. Hydrous hematite is frequently associated with the limonite ore and jarosite is less commonly observed. The grade of this ore is 50% Fe at the maximum.

Sedimentary type limonite ore is characterized by the presence of brown colored loose crystals of goethite and hydrous hematite replacing some kinds of leaves or trunks of tree or moss.

6) Sedimentary type limonite deposit of the Kutchan mine

This mine is situated at Wakikata, Kyogoku town, Abuta district. Geology of the mining area consists of various kinds of volcanic rocks of Neogene Tertiary and of Plio-Pleistocene periods. This system is overlaid with Makkaribetsu Formation consisting of volcanic tuff, scoria and clayey material of Pleistocene age (Fig. 13).

The limonite deposit of this mine is sedimentary type, and is conformably lying in

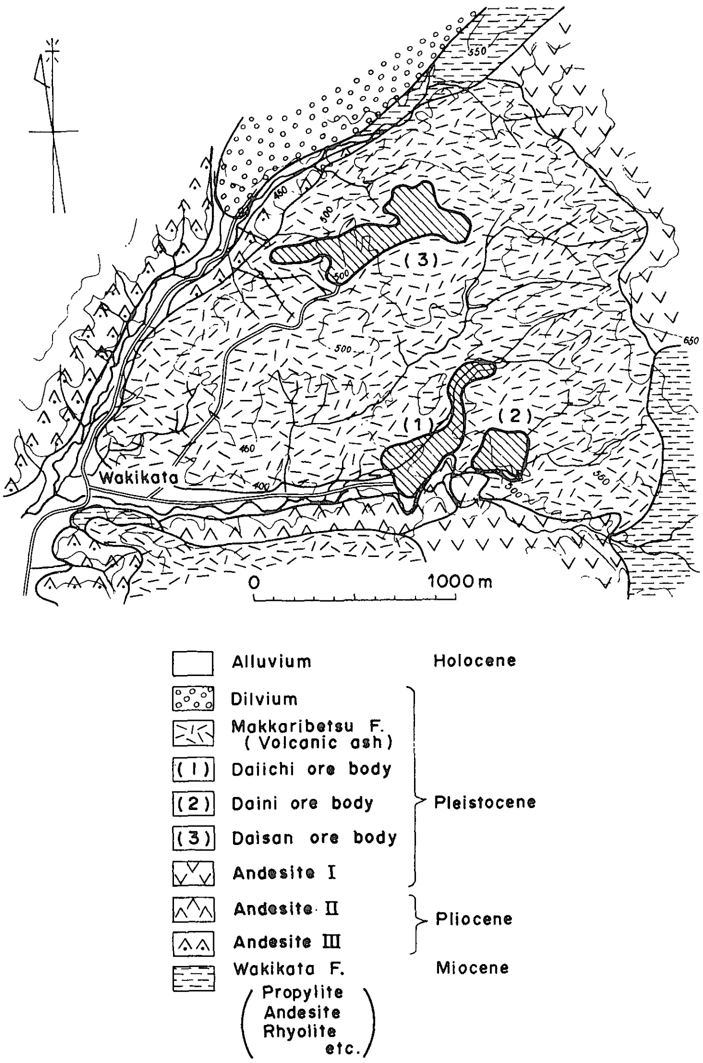


Fig. 13 Geological map of the Kutchan mine representing the stratigraphic position of limonite ore deposits.

the formation. The ore reserve has been the biggest of this type in this country. The exploited limonite ore during fifty years from 1898 to 1969 when the mine was closed amounted to more than five million metric tons. Following is the reported production of ore from this mine.

1906–1930: 464,396t (as refined ore)

1931–1969: 5,087,446t (as crude ore, 50.47% Fe)

The ore deposit is composed of independent three ore bodies. The Daiichi and the Daini ore bodies are found at the slope facing Wakatasatsupu river at the height of 430–500m above sea level, while the Daisan ore body is found at another slope facing Shirai river at 460–580m above sea level, 1km north to the former two ore bodies.

Mode of occurrence of the ore deposit and intercalating clayey layers in the ore bed suggests that the precipitation of limonite took place repeatedly several times.

The extents of these three ore bodies are estimated as follows:

	length	width	thickness
Daiichi ore body	700m	200m	10m
Daini ore body	300m	200m	10m
Daisan ore body	1,200m	500m	6m

The ore is massive and brown to chocolate in color. Replaced plants such as bamboo leaves or moss are commonly observed in the ore. Wall rock alteration is remarkable, which is characterized by silicification and argillization associated with scattered grains of pyrite. This alteration is rather predominant in the foot wall, especially in the eastern side of the mining area.

6. Mutual relation between Cu-Pb-Zn-Mn mineralization of Miocene time and sulphur, iron-sulphide, limonite mineralization of late Neogene Tertiary to Quaternary period

The present author (IGARASHI, 1967; IGARASHI and YOKOTA, 1970; IGARASHI et al., 1974) has proposed that there exists some close relationship between the mineralizations of Miocene time and of late Neogene Tertiary to Quaternary period. In this chapter, it is attempted to interprete the mutual relation between the above-mentioned two mineralizations on the basis of the geochemical data obtained in the area extending from Shakotan-peninsula to Toya-lake. In this area the two kinds of mineralization are observed to overlap with each other (Figs. 2 and 14).

It has been well known that the area of 3,000km² from Shakotan-peninsula to Toya-lake is an up-heaved mass consisting mainly of pyroclastic green rocks in which the doming structures represented by Shakotan dome and/or Jozankei dome etc. are observed. Quartz porphyry and propylite are characteristically found along the axis of the domes. It seems that the structure is connected with the zonal distribution of some metallic mineral deposits. For instance, Pb, Zn, Mn, Cu ore deposits of the Toyoha, Inakuraishi and Oe mines appear near the center of the dome and Bi, Sb bearing Au, Ag ore deposits

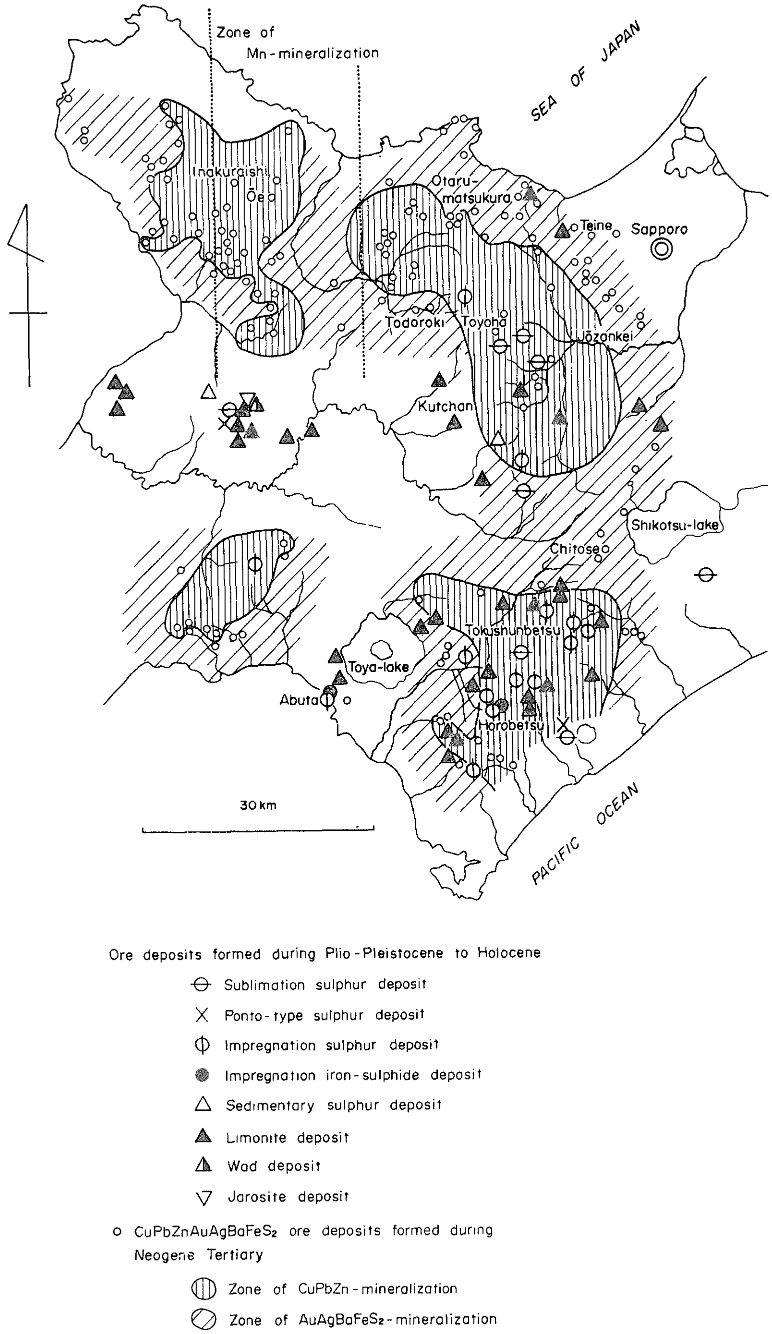


Fig. 14 Map showing the overlapped distribution of ore deposits related to the mineralizations of Neogene Tertiary and of Plio-Pleistocene to Holocene periods.

of the Chitose, Todoroki, Teine and Koryu mines are known in the peripheral area. Furthermore, these ore deposits are surrounded by barite deposits of the Otaru-Matsukura and Minami-Shiraoi mines (Fig. 14). Wall rock alteration in the Pb-Zn-Mn-

Cu zone is characterized by the presence of quartz-sericite-chlorite or chlorite-albite assemblage. On the other hand, in the Au-Ag-Bi-Sb-barite zone, the wall rock alteration represented by quartz-adularia assemblage is rather predominant.

Relatively high abundance of As, Ba, Bi, Cu, Pb, Zn was detected in the sulphur ore or the native sulphur from the Horobetsu, Abuta, Benkei, Tokushumbetsu, Shiraoi and Nittetsu-Shiraoi mines. Sb is common in the sulphur ore from the Horobetsu mine. "Horobetsuite" is observed in this mine and bismuthinite is known in the Abuta and Shiraoi mines. In the vicinity of Toyoha mine, some sublimation type sulphur ore deposits have been known, in which As, Co, Pb and Zn are detected. Especially, the presence of Pb is notable because this element is exclusively found in the sulphur ore from this area. This fact indicates that Pb from the sulphur ore should have been related to the Miocene Pb-Zn mineralization that had formed the Toyoha deposit.

Consequently, the following conclusions have been obtained:

1. Geologic structure of the eastern area of west Hokkaido formed during Miocene time might have played a role of channel way not only for the ascending ore solution of the Miocene to Pliocene Au-Ag-Pb-Zn-Cu-Mn-Ba mineralization but also for the sulphuric gas or solution that formed the sulphur, iron-sulphide and limonite ores of Plio-Pleistocene time. Thus the metallogenic provinces of Miocene and of Plio-Pleistocene ages were overlapped to each other.
2. Various kinds of metallic elements related to the Miocene mineralization were migrated into the sulphur ores of Plio-Pleistocene time. Thus, Pb, Zn, As, Co were detected in sulphur ores from the vicinity of the Toyoha mine. This fact, as far as concerning Pb and Zn, may affirm the existence of a regenerative process.
3. On the other hand, specific element such as Bi was detected in the sulphur ores from the Horobetsu, Tokushumbetsu, Nittetsu-Shiraoi and Shiraoi mines. Bi containing ore minerals have been restrictedly known in the Au-Ag zone including the Teine, Koryu mines but have never been found in the Pb-Zn-Cu-Mn zone including the Toyoha, Oe, Inakuraishi mines. It is, therefore, rather difficult to explain Bi in the Horobetsu, Tokushumbetsu, Shiraoi and Nittetsu-Shiraoi ores as a product of regenerating process.

These observations suggest that the ore forming processes of Miocene to Plio-Pleistocene times in this part of Hokkaido may be summarized as follows. The first effective movement of ore solution had formed Pb-Zn-(Cu)-(Mn) ores in the inner zone of the dome in Shakotan-peninsula to Toya-lake area. Then the mineralization moved to the outer zone including the Teine, Koryu and Chitose mines precipitating Au, Ag, Bi, Sb, Ba ores, afterwards returned to the inner zone again to deposit sulphur, iron-sulphide, limonite ores resulting the overlapped mineralizations of two different ages in the same area. During this process, Pb, Zn and some other elements in the older mineralization were migrated into the newly formed sulphur ores, whereas the origin of Bi in the sulphur ores is considered to be in the residue of the solution that had precipitated Au, Ag and a part of Bi during late-Miocene to early Pliocene time.

The distribution of wad deposits of Holocene age is limited to the area west of the

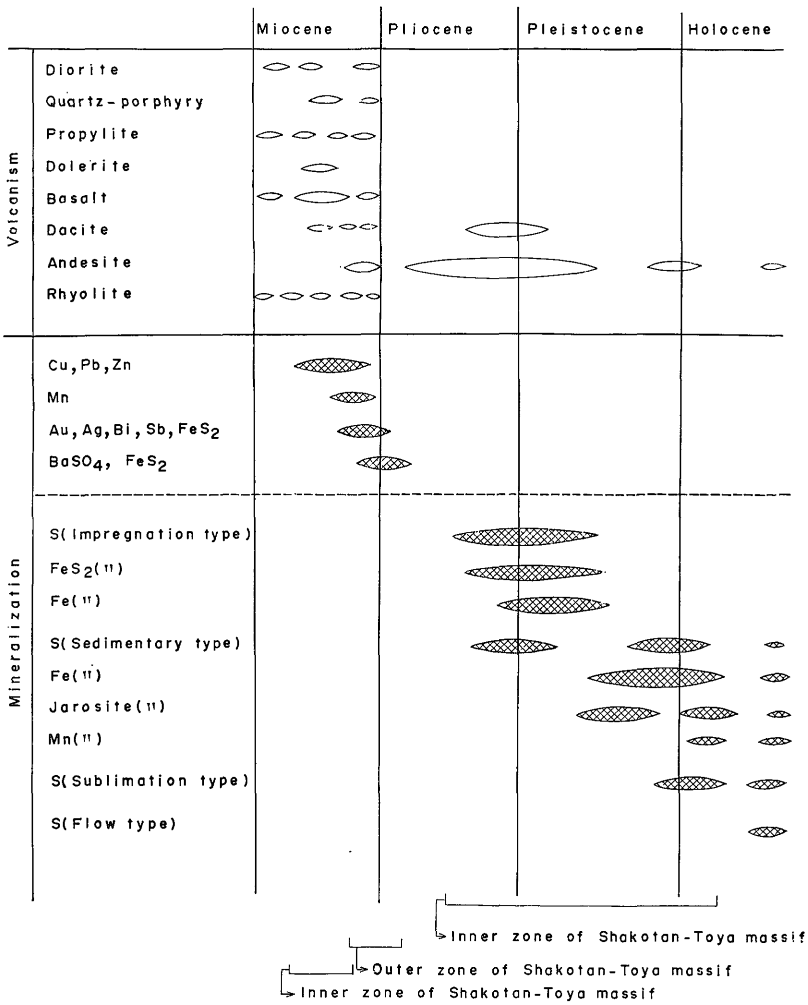


Fig. 15 Schematic diagram to explain the activity of volcanic rocks related to the formation of sulphur, iron-sulphide and limonite ores of Plio-Pleistocene time and of base metal ores of Miocene time.

line connecting Mt. Yotei and Mt. Komagadake where Miocene Mn ore deposit predominates. This indicates that the wad may also be a product of regenerating process and the mineralization forming wad deposit is independent from those that yielded sulphur, iron-sulphide and limonite ores.

Conclusion

The present geological and geochemical studies on the sulphur, iron-sulphide and limonite ore deposits in Hokkaido has yielded the following results.

1. The mineralization age can be divided into two: Plio-Pleistocene time and Holocene time.
2. The type of sulphur ore deposit is classified into four: sublimation type, impregnation-replacement type, flow type and sedimentary type.

3. Though wad deposit is occasionally accompanied by sulphur, iron-sulphide and limonite ore deposits, the orogin of wad deposit is interpreted as a kind of regenerated deposit, and it must be distinguished from the others.
4. Detailed description of the composite type sulphur, iron-sulphide and limonite ore deposit and the comparative study of the limonite deposit free from sulphur and iron-sulphide ores and the sulphur ore deposit lacking limonite ore indicate that the origin of the last two kinds of ore deposit should be explained as varieties of the composite type deposit.
5. The mutual geochemical relation between Miocene base metal mineralization and Plio-Pleistocene sulphur, iron-sulphide and limonite mineralization points out the existence of a regenerating process as well as an evolution of a single ore solution.

The relation between volcanism and sulphur mineralization, an important subject of the ore genesis, has not been touched in this paper, though a series of volcanism of Miocene to Holocene time in west Hokkaido is summarized in Fig. 15 to emphasize the time relation between the mineralization and the volcanism.

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北海道の新第三紀後期－第四紀の硫黄，硫化鉄鉱，褐鉄鉱鉱化作用

五十嵐 昭明

要　旨

北海道には新第三紀後期－第四紀に形成された多数の硫黄，硫化鉄鉱，褐鉄鉱鉱床が分布する．本論文では，これらの鉱化作用を地質学的，地球化学的観点から考察した結果をのべるとともに新第三紀鉱化作用との関連についても言及した．

本論では，まず硫黄，硫化鉄鉱，褐鉄鉱鉱床の生成時期を鮮新世－更新世と完新世との2時期に分けた．完新世の硫黄，褐鉄鉱鉱床には，ときにマンガン土鉱床が伴われるが，マンガン土鉱床は，中新世マンガン鉱化帯と重複して出現し，その起源は一種の再生鉱床と考えられる．

鮮新世－更新世の硫黄，硫化鉄鉱，褐鉄鉱鉱床は，一連の鉱化作用によって形成された複合型鉱床であり，本論では，その産状を詳細に記載した．従来，独立した鉱床とみなされていた硫黄，硫化鉄鉱鉱床を伴わない堆積型褐鉄鉱鉱床や，褐鉄鉱鉱床を伴わない硫黄鉱床などの産状および地質環境を検討し，このような単独に存在する鉱床も複合型鉱床の一変種としてみるのが適当であることを明らかにした．

賤金属鉱石を生成した中新世の鉱化作用と硫黄，硫化鉄鉱，褐鉄鉱鉱石をもたらした鮮新世－更新世の鉱化作用との相互関係を地球化学的に検討し，とくに硫黄鉱石に伴われる微量元素の挙動から，微量元素の起源には再生過程によるものと，鉱液それ自身の進化過程によるもののあることを指摘した．

中新世における鉱液の最初の沈殿は，ドーム構造をなす積丹—洞爺地塊の中心部で実現され，そこに大江，稲倉石，豊羽鉱山などの Pb-Zn-Cu-Mn 鉱床を形成し，ついで鉱液はドームの外帯に移動し，手稲，光竜，千才，小樽松倉，南白老鉱山などで Au, Ag, Bi, Sb, Ba を沈殿した．その後，鉱液沈殿の場は鮮新世の火山活動の激化に伴い再び内側に移動し，硫黄，硫化鉄鉱，褐鉄鉱鉱床を形成し，結果的に異なる2時期の鉱化作用を重複せしめた．この過程で Cu, Pb, Zn ほか 2, 3 の元素は，硫黄鉱石の中に再生的な物質として移動した．

一方，硫黄鉱石中に卓越する Bi の起源は，中新世後期－鮮新世初期に鉱液が Au, Ag, Bi の一部を沈殿したあとの稀薄化溶液中の残存物であろうと考察した．このことから鮮新世－更新世の鉱化作用に関係した微量元素には，再生過程によってもたらされたものと，鉱液それ自身の進化過程によってもたらされたものとがあると結論した．

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