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Indium and gallium enrichments in zinc orebodies in the Taebaeksan region, Korea

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Indium (In) and gallium (Ga) have been regarded as the critical elements due to a large recent demand on high-tech applications but their relatively low crustal abundance. The presence of variable In and Ga contained in sphalerite has recently been reported from numerous magmatic-hydrothermal zinc orebodies in the Taebaeksan metallogenic region in Korea. Here we report chemical compositions of lithologies including granitoid intrusions and sedimentary sequences in the region and compare with associated sphalerites to investigate geological distributions and enrichment processes of In and Ga. While In in the lithologies are not generally detected by LA-ICP-MS, high concentrations up to 630 ppm in sphalerite suggest a significant In enrichment by magmatic-hydrothermal fluids. Major zinc mines show high In concentrations of an average 100 ppm suggesting a potential of economic In orebodies in the region. Despite relatively moderate Ga contents up to 24 ppm in the hosting lithologies, Ga enrichment in sphalerite up to 66 ppm is rather limited compared to the In enrichment. The types and mineral assemblages of magmatic-hydrothermal sphalerite-bearing ores indicate that In is concentrated in relatively higher temperature, whereas Ga tends to be enriched in lower temperature. The enrichment processes of In and Ga in sphalerite might be strongly related to their complexations in the magmatic-hydrothermal fluids. High salinity in the fluid is favorable to form an In-chloride complex for mobilization and enrichment of In into sphalerite. Conversely, Ga-hydroxide complex might not be stable at relatively high temperature which could limit an economic enrichment of Ga in sphalerite in the region.

Introduction

Antimony, barite, beryllium, cobalt, fluorine, gallium, germanium,

graphite, hafnium, indium, lithium, manganese, niobium, platinum-group elements, rare-earth elements, rhenium, selenium, tantalum, tellurium, tin, titanium, vanadium, and zirconium have been and assigned as the “critical element or mineral” due to their low crustal abundances and potential instabilities in the global supply chain (Schulz et al., 2017). In particular, indium (In) and gallium (Ga) are the key elements for the high-tech industrial applications such as LED flat panels, smartphones, electrical materials, photovoltaic, and batteries. Indium is mainly consumed for the indium tin oxide (ITO) as a transparent electrode, which is essential for a display industry. Chemical properties such as low melting points of In and Ga allow us to apply it as a liquid metal (eutectic Ga-In alloy; EGaln) which is a promising material for a flexible electrode.

Indium is produced as a byproduct of Zn smelting mainly in China, Japan, and Korea, while the global In resources are mostly distributed in China (Lokanc et al., 2015; USGS, 2019). Gallium is mainly recovered as a byproduct of a processing of bauxite Al ore and lesser amounts are produced from a Zn smelting (Foley et al., 2017), and China accounts for most of the global Ga production (USGS, 2019). The average abundance of In and Ga in the upper crust are reported as 0.05 ppm (Taylor and McLennan, 1995) and 17.5 ppm (Rudnick and Gao, 2003), respectively. Due to the low crustal abundance, In and Ga generally substitute for elements in minerals including sphalerite (e.g., $2\text{Zn}^{2+} \leftrightarrow \text{Cu}^+ + \text{In}^{3+}$; Johan, 1988) rather than presenting in the stoichiometry in minerals. Sphalerite (ZnS) is the primary Zn ore containing a variety of minor and trace elements such as Ag, Cd, Co, Cu, Fe, Ge, Mn, Sb, and Sn in addition to In and Ga (Cook et al., 2009).

The Taebaeksan (TBS) region in South Korea is an important metallogenic region for Zn-Pb-W-Mo-Fe-Cu (-Au-Ag) and had been a significant Zn-Pb producer accounting for 90% of the domestic production until 1994 (Yun and Einaudi, 1982; Lee et al., 2007; Choi et al., 2009). The Zn orebodies of skarn, carbonate replacement, and vein-breccia type in the TBS region are hosted in early Paleozoic carbonate sequence associated with the magmatic-hydrothermal activities of Late Cretaceous to early Paleogene (Yun and Silberman, 1979; Park et al., 1988). While chemical compositions and grades of the Zn ores from the TBS region have been studied (Yun and Einaudi, 1982; Choi et al.,

2009; Choi et al., 2010), no study has yet investigated critical elements in the ores and their economic potentials except for a preliminary study of Heo et al. (2008). Heo et al. (2008) reported In, Re, Ga, Ge, Se, Te, Y, Eu, and Sm contents in the ores from 34 metallic mines in Korea and they found relatively high In concentrations in the Gagok Zn-Pb deposit (average of 53 ppm) in the TBS region.

In our previous study on sphalerite geochemistry (Lee et al., 2019), we have shown that the sphalerite found in the TBS region contains up to 630 ppm of In and 66 ppm of Ga. Considering an average In content (15–50 ppm; Roskill, 2010) in the ores from global In-producing mines, the economic potentials of In in the numerous Zn orebodies and prospects in South Korea are worthy of further research. Furthermore, critical elements have become important due to a large demand on high-tech applications, it is essential to understand geological dis-

tributions of In and Ga, and its enrichment processes.

In this paper, we investigated the Zn orebodies and prospects in the TBS region and provide the distributions of In and Ga based on the previously studied sphalerite compositions (Lee et al., 2019). We report chemical compositions of associating or hosting lithologies for Zn ores including granitoid intrusions and sedimentary sequences, and discuss enrichment processes of In and Ga into sphalerite.

Geological Background

Taebaeksan Metallogenic Region

The TBS basin, located in the northeastern part of the Korean Peninsula, consists of the Precambrian metamorphic basement of the Yeongnam

Figure 1. (A) Geological map of the Taebaeksan metallogenic region and location of the studied deposits with their predominant types of sphalerite-bearing ore. (B) Detailed geological map of the sampling sites showing sedimentary sequences in the TBS region. The map was modified after Lee et al. (2019).

massif, sedimentary sequences composed of the early Paleozoic Joseon and late Paleozoic Pyeongan Supergroups, and granitoid intrusions of the Jurassic to early Paleogene (Fig. 1A). The Triassic Songrim orogeny and Jurassic Daebo event generated numerous folds and NNE-trending faults in the TBS region (Chough et al., 2000; Kim, 2019), which might provide the advantageous pathways for the ascending ore-forming fluids from Bulguksa granites of the late Cretaceous to early Paleogene age or the intrusion of the granite itself (Moon, 1991). The metallic deposits in the TBS region were formed by magmatic-hydrothermal activities mainly in 90–50 Ma constituting the TBS metallogenic region (Yun, 1986; Park et al., 1988; Park et al., 2013) which includes skarn, hydrothermal replacement, vein, and Carlin-like type deposits based on their proximity to the intruding magma sources (Choi et al., 2009). Most deposits and prospects are hosted in calcareous formations of the Joseon Supergroup which is generally composed of the Jangsan, Myobong, Pungchon, Hwajeol, Dongjeom, Dumugol, and Makgol Formations in ascending order. Major Zn deposits and prospects are localized to the southern part of the TBS region with higher ore grades and reserves, while the northern part is dominated by limestone and calcite marble deposits (Moon, 1991; Lee et al., 2019).

Ore Deposits and Prospects of Zn in the TBS region

Yeonhwa and Gagok

The Yeonhwa deposit had been the largest Zn-Pb producer in Korea, producing concentrates of 0.82 Mt of Zn and 0.25 Mt of Pb from 1935 to 1986 (Je and Lee, 1987). The dikes of quartz porphyry intruded the area in the direction of NS and NW with the developments of pipe-shaped skarn orebodies which are hosted in the Pungchon Formation and in minor calcareous beds of the Myobong Formation within the Joseon Supergroup (Yun and Einaudi, 1982). The K-Ar ages of sericite from ore-related quartz porphyry are reported as from 75.3 Ma to 73.6 Ma (Park et al., 1988; So et al., 1993). The Gagok deposit (also known as Yeonhwa II) was the second largest mine for Zn-Pb, producing concentrates of 0.62 Mt of Zn and 0.03 Mt of Pb from 1971 to 1987 (Yun and Einaudi, 1982; Choi et al., 2010). The Zn orebodies in Gagok show typical characteristics of skarn and carbonate replacement mineralization which are developed along the contact between the NS-trending intrusion of quartz monzonite porphyry and the Myobong-Pungchon Formations (Yun, 1979b). The mineralization age by K-Ar dating of muscovite from a skarn ore are reported as 72.6 Ma (Yun and Silberman, 1979).

Uljin and Sagok

The “Yeonhwa-Uljin mining district” is composed of Zn-Pb deposits in the eastern TBS based on their similar geology and mineralization features, and the district accounted for 80% of domestic Zn-Pb production until the 1980s (Yun, 1979a). The Uljin deposit, located in the easternmost part of the studied area, had produced 0.16 Mt of Zn-Pb concentrates from 1971 to 1986 (Choi et al., 2009). The Zn orebodies are partially distributed in the skarn zone within the Myobong-Pungchon Formations, which is associated with rhyodacite of 49.3 Ma dated by whole-rock K-Ar geochronology (Yun and Silberman, 1979). The Sagok deposit nearby the Uljin deposit was explored by the Young-

pung Mining Company who operated the Yeonhwa-Uljin mining district, but there are no reported records of the Zn-Pb productions. Skarn Zn-Pb mineralization hosted in the Pungchon Formation is dominant in the area.

Janggung

The Janggung deposit in the southern margin of the TBS region is a polymetallic deposit of Zn-Pb-Ag, Mn, V, and Fe (Lee et al., 2007). The deposit had started to exploit Mn on the surface since 1940s, but produced 0.25 Mt of Zn and 0.14 Mt of Pb concentrates during the period 1977–2001 as the Zn-Pb orebodies were discovered in underground (Lee et al., 1990; Choi et al., 2009). Lens- and pipe-shaped orebodies are hosted in the Janggung limestone Formation of the Joseon Supergroup which was structurally controlled by the Jurassic Chunyang granite (148.5 Ma). The K-Ar ages of sericite from Zn-Pb ore yielded 70.0–73.6 Ma (Lee et al., 1996).

Shinyemi and Imog

The Shinyemi deposit is hosted in the Makgol Formation of the Joseon Supergroup associated with the Cretaceous granitoid intrusions (Shinyemi granitoid). The skarn and carbonate replacement type orebodies in Shinyemi are divided into Fe-Mo-Zn rich western orebody and Zn-Cu-Pb rich eastern orebody (Sato et al., 1981; Seo et al., 2020). The mine had produced 56 kt of Zn and 90 t of Pb concentrates from the eastern and upper western orebodies from 1970 to 1982 (Choi et al., 2009), but has currently been exploited for magnetite ores from the lower western orebody. The Shinyemi granitoid intruded to the NNE direction at 74.7–77.2 Ma dated by K-Ar geochronology of biotite and K-feldspar (Yang, 1991), which are comparable to the mineralization age of 75.3–77.7 Ma by phlogopite K-Ar dating (Sato et al., 1981; Park et al., 1988). The Imog deposit close to Shinyemi is composed of skarn and hydrothermal vein orebodies mainly hosted in the Dumugol Formation of the Joseon Supergroup. The Imog granite, which is suggested to be a causative intrusion for the deposit, yielded K-Ar ages of 92.0–96.7 Ma on biotite (Hong, 1986; Yun, 1986). The W skarn mineralization is developed near the Imog granite while the Zn-Pb hydrothermal vein mineralization postdates in the distal part from the skarn along the NE-trending faults of the area (Im et al., 2020; Seo et al., 2020). Im et al. (2016) reported K-Ar ages of 83.6–97.5 Ma for sericite from the Zn-Pb ores.

Dongnam

The Dongnam deposit is a polymetallic deposit for Fe, Mo, Zn-Pb, Ag, and Mn. The magnetite ores were a main commodity of the mine which had been exploited until the 1960s. The Myobong and the Pungchon Formations in the area were intruded by Cretaceous granitoids of porphyritic granites, diorite, and quartz porphyry (Chang et al., 1995). The irregular pocket-shaped skarn Fe orebodies are dominated in the deposit with minor amounts of sphalerite and galena, which are developed in the contact between the Pungchon Formation and the intrusion of diorite (Chang et al., 1995; Lee et al., 2007). The phlogopite coexisting with the vuggy magnetite ore yielded a K-Ar age of 79.6 Ma (Park et al., 1988).

Wondong

The Makgol Formation in the Wondong area hosts various orebodies of Fe, Zn-Pb, and W-Mo associated with the Cretaceous intrusions of rhyolite and quartz porphyry. The carbonate replacement type Zn-Pb orebody occurs as stratiform and pipe-shape which is developed in the northwest of the early skarn Fe orebody (Hwang and Lee, 1998). Recent cross-hole survey confirmed the hidden Zn-Pb orebody in depth with an average grade of 5.50% (Lee, 2011). The quartz porphyry intrusion widely distributed in the area is cut by sheeted sub-vertical quartz veins with sericitic to argillic alteration. While the U-Pb age of zircon from quartz porphyry is 79.4 Ma, the K-Ar ages of phlogopite coexisting with magnetite ore are reported as 49.2-52 Ma (Park et al., 1988; Park et al., 2013).

Sangdong and Gurae

The Sangdong W-Mo (-Bi) deposit had been a world class W producer. The strata-bound scheelite-molybdenite orebodies in Sangdong are mainly hosted in the carbonate-rich layers which are intercalated within the Myobong Formation (e.g., M1 layer). The Sangdong deposit consists of early skarn and subsequent magmatic-hydrothermal veining stages which comprise the zonation of garnet-wollastonite skarn, pyroxene skarn, amphibole alteration, and mica alteration from periphery to center of the deposit (Fig. 2) (John, 1963; Moon, 1989; Seo et al., 2017). Minor amounts of sphalerite occur in the later-stage veins associated with muscovite alteration. The muscovite Ar-Ar age of 86.6-87.2 Ma (Seo et al., 2017) is similar to the K-Ar age of the Sangdong granite beneath the Jangsan Formation (87.5 Ma; Kim, 1986), which might be a causative intrusion for the mineralization. In the Gurae prospect near the Sangdong deposit, minor Zn-Pb-Fe-Cu (-W-Mo) orebody was explored by the geochemical survey (Yoo et al., 2016). Sphalerite-rich quartz veins are developed along the fault hosted in the intercalated carbonate-rich layer within the Myobong Formation.

Seongwoo, Songwon, and Seojin

The Seongwoo, Songwon, and Seojin are calcite marble deposits

exploiting the upper part of the Pungchon Formation. Thrust faults in the NE direction are developed in the areas of Seongwoo and Songwon (Yoo et al., 2016). While no outcrops of intrusion exist in the area, small-scale carbonate replacement Zn-Pb-Fe-Cu orebodies are associated with calcite marble along generally through the faults. The Seojin deposit show a close spatial relationship with the Imog Zn-Pb deposit and the Imog granite. Minor metallic mineralization of pyrrhotite and pyroxene skarn are developed in the contact between the upper Pungchon Formation and the Imog granite. Sphalerite occurs in quartz-sulfide vein which crosscuts the pyroxene skarn.

Petrography of Samples

We collected total 20 samples including sedimentary rock, igneous rock, and metamorphic rock near the Shinyemi, Imog, and Sangdong deposits, which are the representative hosting or associating lithologies of the Zn ores in the southern part of the TBS region (Fig. 1B). Igneous rocks were sampled from the outcrops (Imog granite) and drill cores (Shinyemi and Sangdong granites) which are the representative Cretaceous intrusions related to the mineralization in the southern TBS region. Some rock samples were obtained from skarn and hydrothermal alteration zones in the Sangdong deposit (Fig. 2) (Seo et al., 2020) to investigate an enrichment process of In and Ga during the magmatic-hydrothermal process. Summarized descriptive information of the collected samples are reported in Table S1.

Sedimentary Rocks in the Joseon Supergroup

The samples from the Jangsan Formation, which is the lowermost sequence of the Joseon Supergroup, mostly consist of quartzite with minor disseminated pyrite and fine-grained sericite (JS_JSQ01 and JS_JSQ02, respectively). The rocks from the Myobong Formation are composed of a laminated dark gray slate (JS_MB01) and a massive slate with intercalated mudstone (JS_MB02). Intercalated limestone sample (JS_MB03) from M1 layer in the Myobong Formation contains partly recrystallized calcite and is later cut by quartz veinlets.

Sangdong W-Mo deposit (main orebody)

Figure 2. Schematic cross section of the zonal patterns in the Sangdong W-Mo deposit and associated In and Ga concentrations (modified after Seo et al., 2020). Sphalerites occur in the late quartz vein in bt-mus-qtz zone. The sphalerite in the Sangdong deposit is significantly enriched in In, whereas depleted in Ga even less than associating host rocks and skarns. Abbreviations: grt = garnet, wo = wollastonite, cpx = clinopyroxene, amp = amphibole, bt = biotite, mus = muscovite.

Three limestone samples were collected from the lower and upper parts from the Pungchon Formation. The lower part of Pungchon (JS_PC01) consists of a massive dark gray limestone which partly contains oolitic texture. The upper part samples are characterized by a recrystallized white calcite of sugary texture (SJ_PC01) or a coarse-grained calcite up to a few centimeters (SD_PC01). The rocks in the Hwajeol and the Dumugol Formations are mainly composed of gray limestone. While the sample from Hwajeol contains fine-grained crystallized calcite in a massive limestone (JS_HJ01), a laminated limestone with lenticular calcite is dominant in the Dumugol sample (JS_DMG01). The Makgol Formation, which is widely distributed in the Shinyemi deposit, consists of a dolomitic limestone of light gray color (JS_MG01).

Cretaceous Intrusions

The sample of the Sangdong granite (SD_granite01) is medium-grained feldspar-rich granite consisting of K-feldspar, muscovite, quartz, plagioclase, and minor chalcopyrite and molybdenite with partial sericitic to chloritic alterations. The granites from the Shinyemi and the Imog deposits (SYM_granite01 and IM_granite01, respectively) are pinkish feldspar biotite granite which have a similar mineralogy consisting of K-feldspar, biotite, plagioclase, and quartz, but different textures are observed. While the SYM_granite01 sample is dominated by fine-grained K-feldspar and biotite, the IM_granite01 sample shows a seriate texture of coarse-grained K-feldspar and fine- to medium-grained biotite and plagioclase.

Metamorphic and Altered Rocks

The Precambrian basement sample (SBW_BS01) was collected from the Sunbawi area near the Sangdong deposit. The sample is characterized by a muscovite-rich metasediment of siltstone to sandstone origin.

The samples from the outer part of the Sangdong deposit show anhydrous skarn mineral assemblages (Fig. 2). Euhedral garnet (andradite to grossular) and interstitial wollastonite are dominant in the SD_S01 sample, and clinopyroxene and minor disseminated scheelite are observed in the SD_S02 sample. SD19 and SD16 were sampled from the inner part, which are altered by quartz veins postdating the early skarns (Fig. 2). The SD19 sample is dominated by an amphibole-magnetite alteration, and the SD16 sample shows a biotite-muscovite alteration.

Sphalerite-bearing Ores

Detailed petrography of sphalerite-bearing ores including descriptions, paragenetic sequence, and mineralogical features were reported in Lee et al. (2019). We classified the Zn-bearing ore samples into the types of 1) skarn, 2) carbonate replacement, and 3) vein-breccia based on their textures and mineralogies associating the sphalerite mineralization. While oxide minerals (e.g., magnetite and scheelite) and molybdenite are dominant in relatively high-T skarn and vein-breccia type ores, base-metal sulfides including sphalerite, arsenopyrite, galena, and chalcopyrite are common in relatively low-T carbonate replacement type ores.

Analytical Methods

Powdering of the collected rock samples was conducted at Korea Polar Research Institute (KOPRI) after petrographic analysis. Twenty fresh rock samples were ground in an agate ball mill. The whole rock powders were used for Rock-Eval Pyrolysis and for making homogeneous glass beads prior to XRF and LA-ICP-MS analysis. The details of each analytical method are as follows.

Rock-Eval Pyrolysis

Total organic carbon (TOC) and mineral carbon (MinC) contents in bulk rocks were measured by a Rock-Eval pyrolysis using Vinci Technologies Rock-Eval 6 (Lafargue et al., 1998) housed at Korea Institute of Geoscience and Mineral Resources (KIGAM). IFP 16000 was used as a standard, and the analytical reproducibility for TOC and MinC was better than 2%.

Bulk XRF and LA-ICP-MS analysis of the Fused Glasses

XRF analysis of the collected samples was conducted at KOPRI. The whole rock powders were initially weighed to determine loss-on-ignition (LOI) before heating at a furnace of 1000°C for at least 20 minutes. Dried powder of 0.6 g and flux of 6 g (lithium tetraborate; $\text{Li}_2\text{B}_4\text{O}_7$) were mixed into a platinum crucible and the mixture was fused in Katanax K2 Prime to make a glass bead. The glass beads of in-house reference materials (BCR-2, STM-2, JG-3) and a blank (only a flux) were made under the same condition to calibrate specific trace elements which might be contained in a flux or in a crucible. Produced homogeneous glass beads were loaded in X-ray fluorescence spectrometer (Malvern Panalytical Axios mAX) to acquire major elements including Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, and P as oxides.

Subsequently after the XRF analysis on the glass beads, trace element concentrations were determined by Laser Ablation (LA) ICP-MS at Korea Institute of Ocean Science & Technology (KIOST). The LA-ICP-MS consists of a 193 nm ArF Excimer laser connected to a quadrupole mass spectrometer (Agilent 7700). The ablation was conducted using a 150 μm beam diameter with a repetition rate of 5 Hz. The glass beads were loaded with NIST610 glass as an external standard. Quantification of trace elements and data reduction were performed using a SILLIS software (Guillong et al., 2008). While SiO_2 content was mostly used as an internal standard, CaO content was used for carbonate rocks which was obtained from XRF analysis. Lithium and B, which are the major elements in the flux material for glass beads, were excluded from the consideration of trace elements. Chromium, Ni, Zn, and Au were also excluded which are contained in a considerable amount in the blank bead.

Results

The complete dataset of chemical compositions of the studied samples is reported in Table S2. Trace element concentrations obtained by LA-ICP-MS are presented in an average of 3 ablated spots of each samples.

Rock-Eval Pyrolysis

TOC contents in all the studied igneous rocks are below detection limits. Sedimentary and metamorphic rocks have low TOC contents less than 0.2 wt%, and the highest TOC content of 0.15 wt% is found in the M1 layer of the Sangdong deposit. Skarn (SD_SD01, SD_S02) and altered rocks (SD16, SD19) also have low TOC contents which range from 0.02 to 0.07 wt%. MinC contents were mostly detected in carbonate-rich formations of the Joseon Supergroup ranging approximately 10 wt%. Any relationship between organic carbon contents according to stratigraphy in the TBS region is not observed.

Major and Trace Elements

SiO₂ and Na₂O+K₂O contents in igneous rocks range from 66.9 to 75.4 wt% and from 7.1 to 8.2 wt%, respectively. The Shinyemi granite has the highest silica and alkali contents followed by the Sangdong and the Imog granite. Al₂O₃ contents in the igneous rocks are 13.0–14.3 wt% which indicates that all the studied igneous rocks are per-aluminous granites. Most of carbonate-rich formations of the Joseon Supergroup has LOI of about 40 wt%, and the CaO content is the

highest in the Pungchon Formation (48.4–52.6 wt%). The Myobong Formation (JS_MB01, JS_MB02) contains high Al₂O₃ (16.7–19.4 wt%) and Fe₂O₃ (7.7–12.1 wt%) contents compared to other sedimentary sequences.

Indium was generally below the detection limits (about 0.2 ppm) by LA-ICP-MS analysis on the fused glasses, whereas Ga was detected in most of the studied rock samples. Indium concentrations were low in the Imog granite (1.2 ppm) and in the Pungchon and Makgol Formations (0.2 ppm). Skarn samples from the Sangdong W-Mo deposit contain relatively high In concentrations ranging from 4.5 to 10.3 ppm compared to the samples from alteration zone ranging from 1.5 to 2.5 ppm (Fig. 2). Gallium was found to be one to ten ppm in the studied rocks. Gallium concentrations in sedimentary rocks in the Joseon Supergroup are highly variable. The highest Ga concentrations up to 24.1 ppm were found in the Myobong Formation while the rocks from the Pungchon Formation were below detection limits (about 0.7 ppm). The granites in the region show a comparable range of Ga concentrations (13.4–17.1 ppm) while showing the highest in the Sangdong granite.

Vanadium concentrations are relatively high in the Imog granite (43–46 ppm) and in slate-rich layer of the Myobong Formation (122–380 ppm) among the analyzed rock samples. Cobalt was found to be

Table 1. Stratigraphic column and In and Ga concentrations in lithology in the Taebaeksan metallogenic region (modified after Moon et al., 1989; Seo et al., 2020)

Stratigraphy & ages of lithology	Intrusion and Formation (thickness in meter)	General lithology	Bulk In and Ga concentrations in intrusions and host rocks (this study; ppm)	In and Ga concentrations in sphalerite (Lee et al., 2019; ppm)
late Cretaceous intrusion	Imog granite	K-feldspar & biotite rich seriate holocrystalline granite	In = 1, Ga = 13–14	
late Cretaceous intrusion	Shinyemi granite	K-feldspar & biotite rich fine-grained granite or quartz porphyry	In = bdl, Ga = 15	
late Cretaceous intrusion	Sangdong granite	K-feldspar & muscovite rich equigranular granite	In = bdl, Ga = 17	
Pyeongang Supergroup (Carboniferous-early Triassic)		Various lithologies including shale, sandstone, conglomerate and limestone		
	Makgol (250–300)	Dark gray limestone and dolomitic limestone	In = 0.2, Ga = 1	Shinyemi (In = 2–630, Ga = 1–18), Wondong (In = 7–40, Ga = 1–2)
	Dumugol (200–250)	Interlayered carbonate-shale, vermicular limestone and sandy shale	In = bdl, Ga = 3	Imog (In = 0.1–0.4, Ga = 24–61)
	Dongjeom (20–30)	Dark gray quartzite		
	Hwajeol (100–150)	Interlayered carbonate-shale, vermicular limestone and sandy shale	In = bdl, Ga = 5	
		Upper part: milky-white limestone or marble	In = 0.2, Ga = bdl (calcite marble)	Seongwoo (In = 0.1–11, Ga = 2–66), Songwon (In = 36–38, Ga = 7), Seojin (In = 170–240, Ga = 1–2)
		Middle part: light gray dolomitic limestone		
		Lower part: light gray to dark gray limestone	In = bdl, Ga = bdl	Yeonhwa (In = 58–130, Ga = 4–28), Gagok (In = 9–49, Ga = 0.7–6), Uljin (In = 79–83, Ga = 2–4), Sagok (In = 12–13, Ga = 1), Dongnam (In = 1–32, Ga = 1–2)
	Myobong (80–150)	Clay-rich slaty shale with thin carbonate-rich layers or lenses	In = bdl, Ga = 20–24 (slate), In = bdl, Ga = 2 (M1 carbonate layer), In = 2–10, Ga = 9–74 (skarn and alteration)	Sangdong (In = 310–330, Ga = 3–4), Gurae (In = 7–8, Ga = 1)
	Jangsan (150–200)	Light to dark gray quartzite	In = bdl, Ga = 3–4	
Metamorphic basement (Precambrian)		Mica-rich metasedimentary rock (dominantly schist) and granitic gneiss	In = bdl, Ga = 9	

high in the Makgol Formation up to 100 ppm. Germanium was not detected in all rocks except for the skarn rocks in the Sangdong deposit (26–61 ppm). Molybdenum appeared in trace amounts in the Sangdong granite (1.7 ppm) and the lower part of the Pungchon Formation (0.6 ppm). Tungsten was detected in all the lithologies except the Hwajeol Formation, and the highest concentration was found in the Sangdong granite (44 ppm). Tungsten concentrations are generally around 30 ppm in the sedimentary sequences of the Joseon Supergroup.

Indium and Ga concentrations in all studied lithologies are reported in Table 1, and we compare them with the concentrations in sphalerites associated or hosted in the lithologies. The sphalerites were inclusion-free without the texture of “chalcopyrite disease” or micro-inclusions (Lee et al., 2019). Although the rocks contain relatively low concentrations of In and Ga, In and Ga concentrations in sphalerites show varying ranges from 0.1 to 630 ppm and from 0.7 to 66 ppm, respectively. The average In concentrations in sphalerites were found to be high in the order of the ore types of vein-breccia (86 ppm), skarn (56 ppm), and carbonate replacement (9 ppm). Gallium was the highest in the carbonate replacement (20 ppm) followed by the vein-breccia (13 ppm) and the skarn (5 ppm) types (Lee et al., 2019).

Discussion

Distributions of In and Ga in the TBS sphalerites

Histograms of In and Ga concentrations in sphalerites from the various deposits in the TBS region are respectively illustrated in Figs. 3 and 4 (modified after Lee et al., 2019). High In concentrations up to

630 ppm are in the sphalerites in the Yeonhwa, Gagok, Uljin, and Shinyemi deposits where Zn ores had been produced as a main commodity. The average In concentration of these deposits is 100 ppm, which is higher than those of global In-producing mines (15–50 ppm; Roskill, 2010). Indium in the TBS region, therefore, can potentially be an economic in the near future, if the Zn reserves in the aforementioned deposits are secured.

Indium concentrations are variable within the same deposit depending on its ore types (Fig. 3 and Table 1). In the Shinyemi deposit, skarn and vein-breccia type ores contain respective averages of 570 ppm and 57 ppm, whereas carbonate replacement ores have an average of 2.5 ppm. The features are thus indicating that we require a further detailed chemical analysis for the specific ore type of sphalerites, which is mainly exploited in the mines. Indium was also enriched in the Sangdong W-Mo deposit (average of 320 ppm) and Seojin calcite marble deposit (average of 200 ppm), but the economic Zn orebodies are not discovered yet in the areas.

About 90% of global Ga has currently been produced by processing bauxite ores (Rongguo et al., 2016) which are reported to contain Ga about an average of 57 ppm (Schulte and Foley, 2014). The rest of Ga is produced through smelting of Zn ores from sediment-hosted deposits including MVT (Mississippi Valley-type; average of 58 ppm) and SedEx (Sedimentary Exhalative; average of 26 ppm) (Schulte and Foley, 2014; Foley et al., 2017). Among the major Zn deposits in this study, recognizable Ga concentrations are only found in the Imog and Janggung deposits (Fig. 4) with an average of about 40 ppm indicating an economic potential for Ga in the TBS region is limited.

Figure 3. Histograms showing In concentrations in inclusion-free sphalerites analyzed from studied deposits. Data are from Lee et al. (2019).

Figure 4. Histograms showing Ga concentrations in inclusion-free sphalerites analyzed from studied deposits. Data are from Lee et al. (2019).

Concentrations of In and Ga in Intrusions and Sedimentary Sequences

The In contents in granite and carbonate rock are generally low ranging from ten to 500 ppb (Schwarz-Schampera and Herzig, 2002 and references therein). Indium concentrations in the Cretaceous intrusions and the carbonate rocks in the southern TBS region associating the Zn deposits and the Zn prospects are found to be below detection limits (generally less than 0.2 ppm; Table S2), while the In concentrations in sphalerites are considerably higher compared to the associating lithologies (Table 1). Lead isotope ratios in the TBS sphalerite suggest that the Zn ores in the TBS region are strongly associated with a hydrothermal activity caused by the Cretaceous granitoid intrusions (Lee et al., 2019). The presence and enrichment of In in the sphalerite indicate apparent hydrothermal affinities during the In mineralization (Audétat and Zhang, 2019) and suggest that In is significantly concentrated by Cretaceous magmatic-hydrothermal processes in the TBS region.

Indium is reported to be relatively incompatible during granite fractionation (Simmons et al., 2017). Indium-rich sphalerites commonly occur in magmatic-hydrothermal Sn-polymetallic deposits distributed in Japan, China, and Bolivia (Ishihara et al., 2011), and Murakami and Ishihara (2013) suggested that an oxidized magmatic source is essential for the In enrichment. The Cretaceous intrusions in the TBS region are commonly fractionated I-type and magnetite-series granitoids (Choi et al., 2019; Seo et al., 2020), which might be favorable to the In enrichment in sphalerite.

Gallium contents in the rocks in the continental crust are much higher than In, reported to be 16–35 ppm in granite, average of 4 ppm in carbonate rocks, and average of 19 ppm in shale (Burton and Culkin,

1978; Mielke, 1979), and the those reported Ga contents are comparable to the rocks in the TBS region (Table 1). Aluminum in aluminosilicate minerals can be easily substituted by Ga due to their similar geochemical affinities (Burton et al., 1959). Except for in the skarn and altered rocks in the Sangdong W-Mo deposit, Ga concentrations showed a strong positive correlation with Al ($r = 0.98$; inset of Fig. 5). While Al/Ga ratios in the associating igneous and sedimentary rocks tend to remain constant about 4000, the hydrothermally affected lithologies such as skarn and altered rocks (e.g., Sangdong W-Mo deposit) show lower and highly variable Al/Ga ratios (from 220 to 3200; Fig. 5). This trend of Al/Ga ratios suggests that Ga can also be remobilized and enriched by magmatic-hydrothermal activities.

Despite an incompatible affinity of Ga during fractionation of magma (Breiter et al., 2013; Simmons et al., 2017), Ga in sphalerite is significantly depleted (2.9 ppm) compared to In (320 ppm) in the Sangdong W-Mo deposit (Fig. 2) where an extensive batholith-scale fractionation of magma caused an enrichment of incompatible element such as W and Mo (Seo et al., 2020). In general, Ga concentrations in sphalerites are generally low compared to In in the TBS region (Table 1). Here we further discuss this distinctive enrichment processes of In and Ga into sphalerite in the following texts.

Mineralization Temperatures and Associating Mineral Assemblages of Sphalerite

In the TBS region, sphalerites in the carbonate replacement type or paragenetically late-stage ores generally contain low In (up to 40 ppm) and high Ga (up to 66 ppm) (Lee et al., 2019). Indium tends to be enriched in sphalerite from the relatively high temperature deposits associated

Figure 5. Plot of Al concentrations versus Al/Ga ratios in rocks from the Taebaeksan metallogenic region. Skarn and altered rocks in the Sangdong deposit show inconsistent trend of Al/Ga ratios compared to the unaltered lithologies, which might suggest an enrichment of Ga by a magmatic-hydrothermal fluid.

with a magmatic-hydrothermal activity such as a skarn type, while Ga is abundant in the lower temperature deposits such a MVT type (Cook et al., 2009; Frenzel et al., 2016b). This apparent temperature-dependent substituting affinities of In and Ga during the Zn mineralization have been applied for a sphalerite geothermometry (Frenzel et al., 2016a).

Mineral assemblages in the ores in general are temperature-dependent (Sullivan, 1957). Indium and Ga concentrations in the sphalerites show a relationship with their associating minerals (Fig. 6). Gallium-rich sphalerites commonly coexist with arsenopyrite and galena, whereas those sphalerites are depleted in In. Arsenopyrite and galena in the TBS

Figure 6. Mineral assemblages found in sphalerite-bearing ores from the Taebaeksan metallogenic region. The ores were arranged at increasing In and Ga concentrations in sphalerite. Data are from Lee et al. (2019). Dominant = ●; Common = ○; Rare = ◦.

Figure 7. Lens-shaped Zn-Pb orebody replacing calcite marble in the Seongwoo calcite marble deposit and distributions of In and Ga concentrations in the ore. Sphalerites are sequentially associated with arsenopyrite, pyrrhotite, and chalcocopyrite. Data are from Lee et al. (2019). Abbreviations: sph = sphalerite, gn = galena, apy = arsenopyrite, po = pyrrhotite, cpy = chalcocopyrite.

sphalerite-bearing ores are mainly associated in relatively later stage of paragenetic sequence which are possibly somewhat decreased hydrothermal temperature (Lee et al., 2019). This trend suggest that the mineral assemblages of sphalerite could be a possible indicator of In and Ga enrichments in the sulfide ores.

The lens-shaped ore prospect occurs in the Seongwoo calcite marble deposit show a distinctive spatio-temporal mineralization pattern from the ore center to the periphery near wall rock (Fig. 7). While the center is closely associated with arsenopyrite, the periphery is rich in sphalerite and galena. Even within a section of 15 cm intervals, In concentration in sphalerite of the center (about 8.7 ppm) is higher than that of the periphery (about 0.1 ppm), whereas Ga is higher in the periphery (about 66 ppm) showing a difference of 60 ppm compared to the sphalerite in the center (about 6.2 ppm). Although a further study is necessary to identify mineralization temperatures of those sphalerites, it is clear that the enrichments of In and Ga are highly selective depending on the mineralization temperatures.

Enrichment processes of In and Ga

The incorporations of In(III) and Ga(III) in sphalerite are facilitated by a coupled substitution with some monovalent cations including Cu^+ and Ag^+ (e.g., $2\text{Zn}^{2+} \leftrightarrow \text{Cu}^+ + \text{In}^{3+}$). In the TBS region, Cu was measured as the most abundant trace element in sphalerite and showed a positive correlation with In and Ga (Lee et al., 2019). Indium is reported to form chloride complexes such as InCl_3 in relatively high temperature hydrothermal fluids up to 350°C (Seward et al., 2000), while Ga prefers to comprise a hydroxide complex such as $\text{Ga}(\text{OH})_3$ (Wood and Samson, 2006). Chemical reactions for the In- and Ga-bearing sphalerite precipitations from hydrothermal solution could be as follows:

and

where the In-Cu and the Ga-Cu couples are incorporated into the sphalerite.

The reactions for the In- and Ga-bearing sphalerite precipitation can be promoted by a low acidity condition in the ore solution, which is advantageous in carbonate rock hosting Zn ores in the TBS region. Saline fluid is favorable to form an In-chloride complex for mobilization and enrichment of In. Audétat and Zhang (2019) reported that hypersaline brines in natural magmatic-hydrothermal fluids contain In ranging from 0.2 to 20 ppm, while Ga is low ranging up to 3 ppm. A formation of hydroxide (OH), formed by a water dissociation, is highly temperature-dependent (Sweeton et al., 1974). The ionization constant of water ($\text{p}K_w$) is generally low in the range of 200-300°C (lowest at 250°C) and increases when temperature decreases (Bandura and Lvov, 2006), which indicates that Ga hydroxides might be more stable below 250°C. Since the reactions 1 and 2 are redox-independent, contents of reducing materials such as a sedimentary organic C (e.g., TOC content; Table S2) in the hosting lithologies might not be a decisive factor for In and Ga enrichments.

Conclusions

In this study we investigated the geological distributions of In and Ga and discussed their enrichment processes in the TBS metallogenic region. While associating sedimentary and igneous lithologies in the TBS region have very low In contents, we found high In concentrations up to 630 ppm in sphalerite. This suggests a significant In enrichment by regional magmatic-hydrothermal processes. Average concentration of In is about 100 ppm in the Yeonhwa, Gagok, Uljin, and Shinyemi

deposits where had been the major Zn producers in Korea. It suggests a potential of economic In orebodies in the region, which requires a further investigation of the Zn reserves and ore grade in the prospects for securing In resources. Enrichment of In in the TBS sphalerite might be closely associated with a magmatic-hydrothermal ore-forming process in the region, the high temperature and high salinity fluids might promote a formation of In-chloride complex.

Gallium concentrations in sphalerites (up to 66 ppm) in the TBS region are not significant, and its concentrations are comparable to the associating lithologies such as the granitoid intrusions (13–17 ppm) and unaltered sedimentary sequences (up to 24 ppm). While Ga can be enriched by a magmatic-hydrothermal fluid as well, the degree of Ga enrichment in sphalerite is not as high as In. Since a Ga enrichment might be related to a low temperature hydrothermal process due to its hydroxide complexation, we conclude that Ga might not be promising in the TBS region. Detailed future studies on the mineral assemblages of sphalerite-bearing ore and fluid inclusion microthermometry would further clarify the enrichment process and would provide useful tools for an exploration of In and Ga in the prospects.

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Table S1. Summarized details of rock samples collected from the Taebaeksan metallogenic province

Sample number	Name and stratigraphy of the sampled rocks	Sampling location (GPS coordinates)	Description
SD_granite01	Sangdong granite (drill core)	37°9'7"N / 128°50'3"E	K-feldspar-rich muscovite granite recovered from drillcore in the Sangdong W-Mo deposit
SD_S01	M1 layer in Myobong Formation (underground)	37°9'7"N / 128°50'3"E	garnet-wollastonite-rich skarn in M1 layer in the Sangdong W-Mo deposit
SD_S02	M1 layer in Myobong Formation (underground)	37°9'7"N / 128°50'3"E	clinopyroxene-rich skarn in M1 carbonate layer in the Sangdong W-Mo deposit
SD19	M1 layer in Myobong Formation (underground)	37°9'7"N / 128°50'3"E	amphibole-magnetite rich alteration in M1 carbonate layer in the Sangdong W-Mo deposit
SD16	M1 layer in Myobong Formation (underground)	37°9'7"N / 128°50'3"E	mica-rich alteration in M1 carbonate layer in the Sangdong W-Mo deposit
SD_PC01	upper part of Pungchon Formation (drill core)	37°8'53"N / 128°49'42"E	megacryst of calcite (1-2 cm) in marble in the Sangdong high-Ca marble deposit
JS_PC01	lower part of Pungchon Formation (outcrop)	37°8'50"N / 128°49'58"E	partly oolitic and partly recrystallized dark gray limestone
JS_MB01	Myobong Formation (outcrop)	37°9'28"N / 128°48'6"E	laminated dark gray and fine-grained slate
JS_MB02	Myobong Formation (outcrop)	37°8'44"N / 128°50'35"E	massive slate to mudstone with quartz and muscovite grain
JS_MB03	M1 layer in Myobong Formation (outcrop)	37°9'30"N / 128°48'6"E	partly recrystallized limestone with quartz grain, wavy thin quartz veins (~1 mm) crosscut
JS_JSQ01	Jangsan Formation (outcrop)	37°9'12"N / 128°48'22"E	laminated quartzite with disseminated pyrite
JS_JSQ02	Jangsan Formation (outcrop)	37°8'43"N / 128°50'30"E	quartzite with crossbedding with sericite
SBW_BS01	Precambrian metamorphic basement (outcrop)	37°7'54"N / 128°48'31"E	meta-sandstone to meta-siltstone with muscovite in the Sunbawi area
IM_granite01	Imog granite (outcrop)	37°9'40"N / 128°40'9"E	K-feldspar rich seriate biotite granite
IM_granite02	Imog granite (outcrop)	37°9'43"N / 128°40'51"E	K-feldspar rich seriate biotite granite
SYM_granite01	Shinyemi granite (drill core)	37°11'34"N / 128°40'7"E	K-feldspar rich fine-grained biotite granite recovered from drillcore in the Shinyemi Fe-Zn-Pb deposit
JS_MG01	Makgol Formation (outcrop)	37°11'39"N / 128°40'35"E	light gray dolomitic limestone
JS_DM01	Dumugol Formation (outcrop)	37°10'13"N / 128°39'12"E	lenticular laminated gray limestone
JS_HJ01	Hwajeol Formation (outcrop)	37°10'1"N / 128°39'36"E	massive and fine-grained calcite crystallized limestone
SJ_PC01	upper part of Pungchon Formation (drill core)	37°10'24"N / 128°40'42"E	sugary grained calcite (~0.2 cm) marble in the Seojin high-Ca marble deposit

Table S2. Complete dataset of chemical compositions of rock samples from the Taebaeksan metallogenic province

Sample number	Rock-Eval (wt%)		XRF (wt%)											
	TOC	mineral C	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	LOI	Sum
SD_granite01	NA	NA	71.6	0.2	13.0	1.6	0.1	0.3	1.2	2.8	5.0	0.0	3.4	99.1
SD_S01	0.07	0.60	40.6	0.0	0.6	13.6	0.4	0.8	34.9	0.1	1.2	0.1	7.2	99.5
SD_S02	0.02	0.03	44.7	0.0	0.4	23.2	1.4	0.8	26.2	0.1	0.0	0.0	0.0	97.0
SD19	0.02	0.04	38.5	0.1	3.7	44.0	0.5	1.3	6.9	0.4	0.5	0.1	0.8	96.8
SD16	0.02	0.06	65.1	0.9	11.0	8.2	0.2	2.7	4.9	0.6	2.0	0.3	3.1	98.9
SD_PC01	0.04	9.66	3.4	0.0	0.0	0.0	0.0	0.2	52.1	0.0	0.1	0.0	43.3	99.2
JS_PC01	0.13	9.92	4.5	0.0	0.4	0.4	0.1	3.0	48.4	0.1	0.1	0.1	42.8	99.8
JS_MB01	0.02	0.01	57.5	0.9	19.4	7.7	0.1	2.6	0.2	0.5	5.9	0.1	3.9	98.7
JS_MB02	0.06	0.04	50.7	3.1	16.7	12.1	0.2	3.1	2.5	0.4	5.7	0.2	3.9	98.7
JS_MB03	0.15	9.56	14.8	0.1	1.4	1.6	0.4	0.8	42.7	0.1	0.4	0.2	37.2	99.8
JS_JSQ01	0.02	0.01	90.8	0.1	2.8	2.7	0.0	0.1	0.3	0.0	0.8	0.1	2.2	99.8
JS_JSQ02	0.02	0.01	91.8	0.2	4.0	0.5	0.0	0.1	0.2	0.1	1.1	0.0	2.0	100.0
SBW_BS01	0.06	0.43	75.4	0.2	10.1	2.5	0.1	0.7	2.0	2.8	1.3	0.1	4.9	100.1
IM_granite01	NA	NA	70.1	0.4	14.2	3.4	0.1	1.3	2.3	2.8	3.6	0.1	2.3	100.6
IM_granite02	NA	NA	66.9	0.4	14.3	3.2	0.1	1.3	2.7	2.8	4.3	0.1	2.0	98.1
SYM_granite01	NA	NA	75.4	0.1	13.0	1.3	0.1	0.2	0.6	3.2	5.0	0.0	1.9	100.8
JS_MG01	0.10	12.24	2.8	0.0	0.2	0.4	0.0	21.5	28.6	0.1	0.1	0.0	46.7	100.3
JS_DM01	0.13	9.92	7.7	0.1	1.2	0.4	0.0	1.8	46.2	0.0	0.8	0.0	40.8	99.1
JS_HJ01	0.08	8.80	14.9	0.1	3.3	1.4	0.1	1.5	42.0	0.1	1.5	0.0	34.1	99.0
SJ_PC01	0.02	10.11	2.7	0.0	0.0	0.0	0.0	0.4	52.6	0.0	0.0	0.0	43.5	99.3

NA = not analyzed

Table S2. Continued

Sample number	LA-ICP-MS (ppm)																																								
	Be	Sc	V	Co	Cu	Ga	Ge	As	Rb	Sr	Y	Zr	Nb	Mo	Ag	Cd	In	Sn	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	W	Pb	Bi	Th	U
SD_granite01	2.9	21	5.5	6.0	23	17	<21	<17	230	110	17	130	12	1.7	1.0	<4.2	<0.1	4.6	6.5	620	26	53	5.4	19	3.6	0.7	3.1	0.5	3.0	0.7	1.8	0.3	2.2	0.3	3.7	1.2	44	14	4.9	12	3.9
SD_S01	1.5	5.2	4.7	6.7	60	14	61	<21	37	14	14	5.3	0.6	13	<0.5	<2.9	10	1200	0.6	<0.7	6.3	19	2.1	9.7	2.2	0.6	2.5	0.4	2.0	0.4	1.3	0.2	<0.2	0.2	<0.1	<0.1	170	2.1	180	2.3	1.0
SD_S02	7.0	5.9	2.7	24	3.7	9.0	26	<19	<0.9	0.9	3.4	4.7	0.7	8.1	2.7	2.7	4.5	36	<0.4	1.5	1.0	2.6	0.4	2.0	0.5	0.3	0.6	0.2	0.3	0.2	0.3	<0.1	<0.2	0.1	<0.1	0.1	570	1.9	3.2	0.8	<0.0
SD19	19	5.4	24	40	170	74	<33	38	12	28	23	16	3.6	50	6.2	<4.5	2.5	120	3.4	72	3.4	12	1.4	8.8	2.1	0.6	2.7	0.5	3.2	0.7	2.3	0.3	2.0	0.3	0.4	<0.2	6400	2.9	1800	2.9	1.3
SD16	6.6	16	89	43	74	18	<42	<31	290	160	51	370	14	9.0	<0.6	18	1.5	19	48	640	63	140	14	58	12	2.7	11	1.7	9.5	2.0	5.9	1.1	6.3	1.0	11	2.0	480	3.1	5.0	25	4.6
SD_PC01	0.3	<1.0	<0.4	5.8	81	<0.7	<19	<14	2.0	160	1.1	0.5	0.1	<0.2	<0.3	<1.9	<0.1	<2.0	<0.3	0.5	0.9	2.7	0.3	1.2	0.2	0.0	0.3	0.1	<0.1	0.1	<0.2	<0.0	<0.2	<0.0	<0.1	<0.0	31	1.2	<0.4	0.1	<0.0
JS_PC01	<0.3	1.4	1.8	6.6	12	<0.6	<19	<15	2.5	280	6.6	3.9	0.4	0.6	<0.3	<2.6	<0.1	<2.0	1.0	16	3.7	8.0	0.9	3.9	1.1	0.2	0.9	0.1	1.0	0.2	0.7	0.1	0.5	0.1	<0.1	<0.0	29	5.0	<0.3	1.1	0.4
JS_MB01	3.5	24	120	17	7.9	24	<36	<27	240	24	30	160	16	<0.5	0.5	<4.9	<0.2	5.8	21	490	46	87	9.1	35	5.9	0.7	5.5	1.0	5.6	1.1	3.5	0.5	3.6	0.5	5.0	1.3	8.8	5.3	<0.6	21	3.2
JS_MB02	2.3	37	380	27	<2.8	20	<34	<25	250	670	38	220	19	<0.5	<0.5	<4.6	<0.2	4.2	52	2500	30	58	6.5	28	6.0	1.3	6.4	1.1	6.8	1.4	4.4	0.6	4.2	0.7	6.1	1.3	33	36	<0.6	10	2.6
JS_MB03	0.7	3.9	14	<0.4	3.3	2.0	<28	<22	14	330	32	48	1.8	<0.5	<0.5	<4.2	<0.2	<3.1	2.0	54	17	40	4.4	19	4.3	1.1	5.2	0.8	5.0	1.0	2.9	0.4	2.7	0.3	1.3	0.2	<0.7	5.4	<0.5	3.8	1.3
JS_JSQ01	<2.1	7.7	8.9	5.4	96	2.5	<42	41	45	75	2.8	86	3.1	<0.7	<0.6	<5.9	<0.2	8.7	1.1	44	15	31	3.1	11	1.7	<0.5	1.2	0.1	<0.6	<0.0	0.3	<0.1	0.4	0.1	2.5	0.2	35	2.1	<0.8	9.6	0.7
JS_JSQ02	<1.5	8.3	4.4	6.4	6.1	4.2	<29	<22	23	75	9.8	140	4.7	<0.5	1.1	<4.0	<0.2	<2.9	0.9	250	20	41	3.9	14	2.4	0.4	1.8	0.3	1.7	0.2	1.0	0.1	0.9	0.1	3.8	0.3	32	2.0	<0.6	11	0.7
SBW_BS01	<2.3	11	23	5.2	8.9	9.0	<47	<35	95	130	8.7	100	5.0	<0.8	<0.7	<6.5	<0.3	10	4.1	220	22	41	4.4	17	2.8	0.5	2.3	0.3	1.8	0.2	0.8	<0.1	0.9	0.1	2.8	0.2	9.3	3.1	4.3	6.2	1.4
IM_granite01	2.0	22	46	4.3	<2.4	14	<35	<27	95	290	17	150	9.9	<0.6	3.5	22	1.2	<4.8	2.4	510	56	100	10	36	5.7	0.8	4.4	0.5	2.9	0.6	1.6	0.2	1.6	0.3	4.3	0.8	<0.8	10	<0.7	19	2.8
IM_granite02	<1.5	20	43	10	<2.6	13	<37	<29	120	310	14	120	8.6	<0.7	<0.4	<6.8	<0.2	<5.1	4.2	880	21	41	4.4	17	3.3	0.9	3.0	0.4	2.5	0.5	1.5	0.2	1.5	0.2	3.5	0.8	32	10	<0.7	11	2.8
SYM_granite01	2.5	18	4.7	5.9	3.8	15	<24	<19	150	110	21	78	13	<0.5	<0.2	<5.1	<0.1	<3.8	2.4	280	39	81	8.1	28	4.7	0.4	4.2	0.6	3.5	0.7	2.2	0.3	2.5	0.3	2.8	1.1	27	13	<0.5	15	5.6
JS_MG01	<0.3	<1.0	0.9	97	71	0.8	<17	<13	1.7	190	1.6	2.7	0.5	<0.3	<0.3	<2.4	0.2	<1.9	0.3	18	2.2	5.0	0.5	2.0	0.5	0.2	0.4	0.1	0.3	0.1	0.2	0.0	0.1	0.1	<0.0	0.1	28	3.2	0.4	0.3	1.1
JS_DM01	0.8	1.9	4.6	7.1	4.1	3.1	<18	<14	8.0	290	2.7	11	1.5	<0.3	<0.3	<2.6	<0.1	<2.0	<0.2	35	3.8	8.1	0.8	3.2	0.7	0.2	0.6	0.1	0.5	0.1	0.3	0.0	0.2	0.0	0.2	0.1	32	2.4	<0.3	1.6	0.8
JS_HJ01	0.7	5.6	17	3.0	3.6	4.7	<23	<19	33	360	5.3	25	3.3	<0.4	<0.4	<3.5	<0.1	<2.6	3.5	84	9.2	16	1.8	7.3	1.4	0.3	1.2	0.2	1.0	0.2	0.6	0.1	0.4	0.1	0.7	0.2	<0.7	2.7	<0.4	2.2	0.7
SJ_PC01	0.6	<1.0	0.6	6.0	<2.0	<0.7	<19	<14	0.9	210	0.7	0.6	0.2	<0.2	1.8	10	0.2	2.1	0.5	1.8	0.7	1.6	0.4	0.7	0.2	0.4	0.2	0.1	0.1	0.1	<0.2	<0.0	<0.2	0.0	<0.1	<0.0	32	1.5	16	0.1	<0.0