공학석사 학위논문

A Biogeochemical Orientation Study in Mo Skarn Deposits, Jecheon District in Korea

국내 제천지역 **Mo** 스카른 광상 일대의

생지구화학탐사 적용 연구

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서울대학교 대학원

에너지시스템공학부

박 지 영

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지도교수 전 효 택

이 논문을 공학석사 학위논문으로 제출함
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서울대학교 대학원
에너지시스템공학부
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위 원 장 ______________________ (인)

부위원장 ______________________ (인)

위 원 ______________________ (인)

Abstract

A biogeochemical orientation survey was conducted in the vicinity of Mo skarn deposits in Jecheon district in Korea. The main geology of the study area is Ordovician dolostone, limestone and limeslicate rocks intruded by Jurassic biotite granite so called Jecheon granite. The skarn zones occur at the contacts of granite and limestone. Molybdenum occurs in fracture zones in skarn within the screen and in disseminated form. The skarn ore minerals are mainly of molybdenite, scheelite, galena and chalcopyrite. The control area is Gwanak mountain where the Seoul National University is located. This area mainly consists of Jurassic granite. The total samples of rocks, soils and two plant species (daimyo oak leaves/branches - *Q. dentata*, and sargent cherry leaves - *P. sargentii*) were collected from the target area and barren control area in May and June 2011, and analyzed for the analysis of Mo, Cu, Pb, Zn, Cd, As, Mn, etc. by ICP-MS. Each of three sampling lines was designed to cross over the each orebody at 30 m spacing intervals in the study area and 10 sampling points are chosen randomly in the control area. The soil samples (n=36/10, target/control) collected from the target area show higher values of Mo (<0.1~38.7 ppm) than those from the control area

(<0.1~3.2 ppm Mo). The concentration level of Mo in plants (n=108/30, target/control) from the target area (0.14~4.91 ppm in *Q. dentata* leaves, 0.01~3.19 ppm in *Q. dentata* branches and 0.15~6.56 ppm in *P. sargentii* leaves) is 4~8 times higher than that from the control area (0.08~0.47 ppm in *Q. dentata* leaves, 0.02~0.26 ppm in *Q. dentata* branches and 0.14~0.42 ppm in *P. sargentii* leaves). The biological absorption coefficient (BAC) of Mo is generally high (*Q. dentata* leaves = 1.4, *Q. dentata* branches = 0.4 and *P. sargentii* leaves = 1.2) and Mo content in soils and plants is strongly correlated. The geochemical variation patterns of Mo in plants are similar to those in soils, which suggest a corresponding Mo anomaly and enhanced contrast near the Mo orebodies. The three plant organs (*Q. dentata* leaves, branches and *P. sargentii* leaves) have high possibilities to be used as indicators for the biogeochemical prospecting of Mo.

Keywords: Biogeochemical orientation survey, Mo skarn deposits, Mo anomaly, *Q. dentata, P. sargentii*

Student Number: 2010-23338

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1. Introduction

For the past few years the rare metals have been recognized as vitamins of industry. Although the importance of the rare metals is emphasized, the supply and prices are not stable which causes the rare metal crisis. It is time to increase the significance of developing the effective exploration methods for rare metals more than ever before.

There are several exploration methods including geophysical prospecting, exploration drilling and geochemical prospecting. Geochemical exploration is able to obtain the quantitative chemical properties of naturally occurring substances including rocks, soils, stream sediments, plants and gas. In particular, the biogeochemical prospecting uses to analyze the mineral elements in plant tissues to assess the presence and nature of underlying mineralization. The plants can be viewed as a sophisticated geochemical sampling device, as yet not fully understood (Dunn, 2007 a). According to Baker and Brooks (1989), some plants easily take up large quantities of metals, others uptake metals in proportion to their quantities in the soils and the others are more likely to exclude metals.

The principle of biogeochemical prospecting is that plants can take up elements from soils or underlying bed rocks, and accumulate some elements in their organs, such as roots, branches and leaves. Accordingly, if certain plant organs contain abnormal concentrations of certain metals, they can have possibilities to be used as indicators for the geochemical prospecting (Brooks et al., 1995).

Plant samples have more points of excellence than any other sample medium, which easily provide data representing the broad area because the root can reach to bedrocks widely. In addition, the use of plants has strong possibilities to explore the mountainous areas or thick and/or transported regolith tundra areas (Jung et al., 2011 b). Moreover the biogeochemical approach to mineral prospecting is cost effective compared with any other prospecting techniques. However, there is not a one-to-one relationship between the chemistry of plants and that of soils (Dunn, 2007 a). Because of the complicated relationship between plants and soils, we need to carefully approach mineral prospecting and check a plenty of case studies so as to adjust the vegetation geochemical approaching.

Only a few case studies of biogeochemical prospecting have been carried out in Korea although there have been reported a lot of applications of vegetation exploration globally (Jung et al., 2011 b). Chung (1965, 1971) studied geobotanical indicator plants for metals, Son (1992) studied the development of biogeochemical prospecting for ore deposits in Korea. However, as the industry has become highly dependent on development of exploration methods, several recent case studies were reported. Jung (2011 a) studied biogeochemical prospecting for Au deposits, and Jeon et al. (2012) conducted an orientation survey for Fe-base metals in Korea.

In this study, a biogeochemical orientation survey was conducted in order to investigate the potential as an exploration method in the vicinity of Mo skarn deposits in the Jecheon district in Korea. The main objectives are firstly to investigate the geochemical characteristics of rocks, soils and plants, secondly to evaluate the geochemical relationship between soils and plants, also between Mo and associated elements in the soil-plant system as well, finally to determine the indicator plants for biogeochemical prospecting of Mo.

2. Theoretical Background

Vegetation geochemistry is a field of study analyzing element concentration in plant organs, observing the shape of plants and investigating characteristics of plant colony. The vegetation geochemical prospecting is founded on the underlying principle of the vegetation geochemistry. It is classified into two methods. One is the biogeochemical prospecting which involves the chemical analysis of plant tissues to obtain information about the underlying substrate (Brooks et al., 1995; Dunn, 2007 b; Jung et al., 2011 b). The other is the geobotanical prospecting whose aim is to find the mineral occurrence by observing flora and morphological botany.

The principle of biogeochemical prospecting is that plants can take up elements from soils or bed rocks and accumulate some elements in their organs, such as roots, branches and leaves (Fig. 2.1).

The biogeochemical prospecting is able to easily provide geochemical information on underground compared with the geobotanical prospecting. After advanced several analytical instruments such as ICP-MS (Inductively Coupled Plasma Mass Spectrometry) and INAA (Instrumental Neutron Activation Analysis), the biogeochemical prospecting has become more cost effective and provided accurate and precise multi-elements data. Generally the

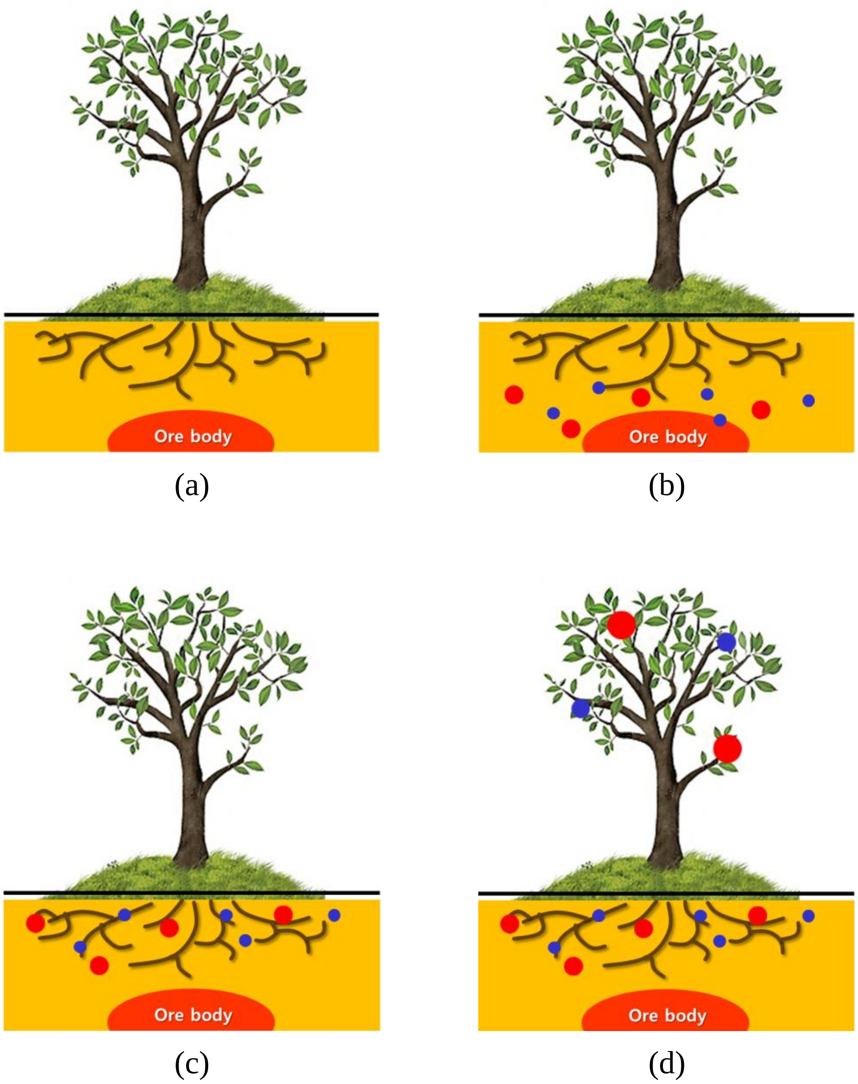


Fig. 2.1 The principle of biogeochemical prospecting.

(a) → (b) → (c) → (d)

biogeochemical prospecting is carried out in reconnaissance level and proceeds with the following steps. First of all, the preliminary survey is conducted in order to investigate the range and the depth of deposits, and the geographical distribution of plants. Secondly the target plants are set in the light of the depth of roots which are putting down deep spreading. And then in the sampling and preprocessing step, plants and soils are collected along the sampling line and have to be dried and homogenized. Finally, all of the samples are analyzed by analytical instruments and the results are interpreted.

The biogeochemical prospecting has some advantages compared to other exploration methods. Plant samples have more points of excellence than any other sample medium, which easily provide data representing the broad area because the root can reach to bedrocks widely. In addition, the use of plants has strong possibilities to explore the place which is the mountainous areas or the presence of thick and/or transported regolith tundra areas (Jung et al., 2011 b). Moreover the biogeochemical approach to mineral prospecting is cost effective compared with any other prospecting techniques such as geophysical prospecting and exploration drilling.

However, there is not a one-to-one relationship between the chemistry of plants and that of soils (Dunn, 2007 a). The plant absorption is influenced by diverse factors; plant species, age of plant organs, pH of soils, depth of the roots, drainage photosynthetic processes, daily variations in rainfall and the antagonism of the elements (Brooks, 1972).

The plants are classified into three groups according to behaviors of metal absorption (Baker and Brooks, 1989). The ‘excluder’ does not take up metals. It has a resistance to absorb metals. The ‘hyperaccumulator’ is a plant to uptake large quantities of metals easily. The hyperaccumulator plant is possible to apply to phytoremediation in contaminated site. Lastly, ‘indicator’ is able to uptake metals in proportion to their quantities in the soils. In other words, the indicator plant shows an increase of elements concentration as the element values are increased in soils. The species of indicators play a significant role in biogeochemical prospecting since the geochemical signal is reflected.

Two case studies of biogeochemical exploration for Mo are summarized in Table 2.1. First study was conducted at the Setting Net Lake Mo deposit in Canada. The second study was conducted in the Vathi area which is porphyry copper areas of sulfide mineralization, Serbo-Macedonian massif in northern Greece. In Korea, however, biogeochemical studies targeting Mo were hardly reported.

Table 2.2 is the mean background contents of trace elements in soils. And a summary of mean background contents of trace elements in plants is presented in the Table 2.3.

In addition a list which is the elemental association and associated elements useful in exploration is able to provide evidence in order to analyze data (Table 2.4).

Table 2.1 Case studies of biogeochemical exploration for Molybdenum.

	Case 1	Case 2
Target mineral	Mo	Mo
Sample	Rock, Soil, Plant	Soil, Plant
Region	Setting Net Lake molybdenum deposit Porphyry-type molybdenum-copper mineralization, northwestern Ontario, Canada	Porphyry copper areas Sulfide mineralization, Serbo-Macedonian massif in northern Greece
Analytic methods	-	Plasma Emission Spectroscopy
Research points	Chemical analysis of elements Comparison of concentration by distance from mineralized zone Comparison of concentration between soil and plant	Chemical analysis of elements Comparison of concentration, correlation coefficient between soil and plant
Results	Soil samples have 1~80 ppm of Mo level and 2~71 ppm in the ash of second year black spruce needles. Molybdenum uptake by coniferous vegetation is controlled mainly by Mo concentration and mobility in supporting soils.	Soil samples have 77~708 ppm of Mo level. *Minuartia verna* and *Thymus sibthorpii* should be a particularly useful biogeochemical indicator of Mo.
Reference	Wolfe (1974)	Reeves (1986)

Table 2.2 Mean background contents of trace elements in soils (Kabata-Pendias, 2010). (unit : ppm)

Element	Crustal Average	World-soil Average	Element	Crustal Average	World-soil Average
Sb	0.2	0.67	Mo	1.5	1.1
As	1.8	6.83	Nd	28	26
Ba	400	460	Ni	20	29
Be	3	1.34	Nb	20	12
Bi	0.2	0.42	Os	0.00005	-
B	15	42	Pd	0.004	0.02
Br	2	10	Pt	0.004	0.02
Cd	0.1	0.41	Pr	8.2	7
Ce	60	56.7	Re	0.0004	-
Cs	3	5.06	Rh	0.00006	-
Cl	640	300	Rb	90	68
Cr	100	59.5	Ru	0.0001	-
Co	10	11.3	Sm	4.7	4.6
Cu	55	38.9	Sc	11	11.7
Dy	3	3.6	Se	0.05	0.44
Er	2.8	2.2	Ag	0.06	0.13
Eu	1.2	1.4	Sr	375	175
F	625	321	Ta	2	1.39
Gd	5.4	3.9	Te	0.005	-
Ga	15	15.2	Tb	0.6	0.63
Ge	1.5	2.0	Tl	0.5	0.5
Au	0.004	0.003	Th	7.2	9.2
Hf	3	6.4	Tm	0.5	0.37
Ho	0.8	0.72	Sn	2.5	2.5
In	0.11	0.06	Ti	4400	7038
I	0.5	2.8	W	1.5	1.7
Ir	0.00005	-	U	2	3.0
La	30	27	V	135	129
Pb	15	27	Yb	2.2	2.6
Li	20	21	Y	33	23
Lu	0.3	0.37	Zn	70	70
Mn	900	488	Zr	165	267
Hg	0.07	0.07			

Table 2.3 Mean background contents of trace elements in plants (after Markert, 1994; *indicates modifications by Dunn, 2007 a).

Element	Units	Conc.	Element	Units	Conc.
C	%	44.5	La	ppm	0.2
O	%	42.5	Li	ppm	0.2
.H	%	6.5	Lu	ppb	3
N	%	2.5	Mn	ppm	200
K	%	1.9	Mo	ppm	0.5
Ca	%	1	Na	ppm	150
S	%	0.3	Nb	ppm	50
P	%	0.2	Nd	ppb	0.2
Mg	%	0.2	Ni	ppm	1.5
Cl	%	0.2	Os	ppm	0.0015
Si	%	0.1	Pa	ppb	?
Ag*	ppb	20	Pb	ppb	1
Al	ppm	80	Pd*	ppm	0.1
As	ppm	0.1	Po	ppb	?
Au*	ppb	0.2	Pr	ppb	50
B	ppm	40	Pt	ppb	0.005
Ba	ppm	40	Ra	ppb	?
Be	ppb	1	Rb	ppb	50
Bi	ppb	10	Re*	ppb	0.1
Br	ppm	4	Rh*	ppb	0.01
Cd	ppb	50	Ru	ppb	0.1
Ce	ppm	0.5	Sb	ppm	0.1
Co	ppm	0.2	Sc	ppb	20
Cr	ppm	1.5	Se	ppb	20
Cs	ppm	0.2	Sm	ppb	40
Cu	ppm	10	Sn	ppm	0.2
Dy	ppb	30	Sr	ppm	50
Er	ppb	20	Ta	ppb	1
Eu	ppb	8	Tb	ppb	8
F	ppm	2	Te*	ppb	20
Fe	ppm	150	Th	ppb	5
Ga	ppm	0.1	Ti	ppm	5
Gd	ppb	40	Tl*	ppb	20
Ge	ppb	10	Tm	ppb	4

Table 2.3 Continued.

Element	Units	Conc.	Element	Units	Conc.
Hf	ppb	50	U	ppb	10
Hg*	ppb	20	V	ppm	0.5
Ho	ppb	8	W	ppm	0.2
I	ppm	3	Y	ppm	0.2
In	ppb	1	Yb	ppb	20
Ir*	ppb	0.01	Zn	ppm	50

Table 2.4 Elemental association and associated elements useful in exploration (Moon et al., 2006).

Type of deposit	Major components	Associated elements
Magmatic deposits		
Chromite ores(Bushveld)	Cr	Ni, Fe, Mg
Layered magnetite(Bushveld)	Fe	V, Ti, P
Immiscible Cu-Ni-sulfide(Sudbury)	Cu, Ni, S	Pt, Co, As, Au
Pt-Ni-Cu in layered intrusion(Bushveld)	Pt, Ti, Cu	Sr, Co, S
Immiscible Fe-Ti-oxide(Allard Lake)	Fe, Ti	P
Nb-Ta carbonatite(Oka)	Nb, Ta	Na, Zr, P
Rare-metal pegmatite	Be, Li, Cs, Rb	B, U, Th, rare earths
Hydrothermal deposits		
Porphyry copper(Bingham)	Cu, S	Mo, Au, Ag, Re, As, Pb, Zn, K
Porphyry molybdenum(Climax)	Mo, S	W, Sn, F, Cu
Skarn-magnetite(Iron Springs)	Fe	Cu, Co, S

Table 2.4 Continued.

Type of deposit	Major components	Associated elements
Hydrothermal deposits		
Skarn-Cu(Yerington)	Cu, Fe, S	Au, Ag
Skarn-Pb-Zn(Hanover)	Pb, Zn, S	Cu, Co
Skarn-W-Mo-Sn(Bishop)	W, Mo, Sn	F, S, Cu, Be, Bi
Base metal veins	Pb, Zn, Cu, S	Ag, Au, As, Sb, Mn
Sn-W greisens	Sn, W	Cu, Mo, Bi, Li, Rb, Si, Cs, Re, F, B
Sn-sulfide veins	Sn, S	Cu, Pb, An, Ag, Sb
Co-Ni-Ag veins(Cobalt)	Co, Ni, Ag, S	As, Sb, Bi, U
Epithermal precious metal	Au, Ag	Sb, As, Hg, Te, Se, S, Cu
Sediment hosted precious metal(Carlin)	Au, Ag	As, Sb, Hg, W
Vein gold(Archaean)	Au	As, Sb, W
Mercury	Hg, S	Sb, As
Uranium vein in granite	U	Mo, Pb, F
Unconformity associated uranium	U	Ni, Se, Au, Pd, As
Copper in basalt(L. Superior type)	Cu	Ag, As, S
Volcanic-associated massive sulfide Cu	Cu, S	Zn, Au
Volcanic-associated massive sulfide Zn-Cu-Pb	Zn, Pb, Cu, S	Ag, Ba, Au, As
Au-As rich Fe formation	Au, As, S	Sb
Mississippi Valley Pb-Zn	Zn, Pb, S	Ba, F, Cd, Cu, Ni, Co, Hg
Mississippi Valley fluorite	F	Bs, Pb, Zn
Sandstone-type U	U	Se, Mo, V, Cu, Pb
Red bed Cu	Cu, S	Ag, Pb
Sedimentary types		
Copper shale(Kupferschiefer)	Cu, S	Ag, Zn, Pb, Co, Ni, Cd, Hg
Copper sandstone	Cu, S	Ag, Co, Ni
Calcrete U	U	V

3. Sampling and Analysis

3.1. Study area

3.1.1 Site description

The study area, the Mo skarn deposits of NMC Moland mine, is located in the middle part of the Korean peninsula, Jecheon district in Chungcheongbuk province. It lies between longitudes 128˚ 09’ E and 128˚ 16’ E and between latitudes 37 ˚ 01’ N and 37˚ 06’ N.

This study area is located in the outer zone of Jecheon basin which is situated in the joined place of Sobaek-mountains and Chiak-mountains. There are low hills and mountains. In particular, the Jecheon basin is formed from a prolonged weathering of biotite granite (called Jecheon granite). It is relevant to a mature form of land surface aspects of a geographical cycle.

Because the study area is located at high altitudes in the interior, this region falls under continental climate and shows the early first frost and freeze of the season. The average annual temperature is 10.1℃ and rainfall is concentrated in a short rainy season from June to September, and the total annual rainfall accounts for 1,284 mm. The water system meanders heavily through hills and fields since there are lots of mountaintops and the geological structure lines are not well-developed. The woods are dense with trees mainly composed of Daimyo oak, Sargent cherry, Japanese red pine and Acacia.

(a)

(b)

(c)

(d)

(e)

(f)

Fig. 3.1 Pictures of study area.

(a) NMC Moland mine (b) Mine adit (c) Old dressing plant

(d) A part of sampling location line 1 (e) Outcrop of orebody A

(f) Mine tunnel and dump of orebody B

The NMC Moland mine was enrolled in 1966 and has been developed for about 10 years since 1978. After the mine abandoned, the mining right of NMC Moland was acquired by Dongwon Corporation in 2004 and redevelopment was started in 2007. The mine property contains a proved resource of 1,315,200 tonnes with an average grade of 0.40 % MoS_2. The cutoff grade of MoS_2 is 0.10%.

3.1.2 Geological setting

Geology in the study area is composed mainly Ordovician Hongwolri dolostone and Samtaesan limestone formation from the bottom, which are intruded by Jurassic biotite granite (Jecheon granite) and Cretaceous felsic dike rocks (Table 3.1).

Table 3.1 Geologic sequence around Mo skarn deposits.

Quatarnary	Alluvial deposit	
	~ Unconformity~	
Jurassic	Ore body(Skarn)	
	Acidic dyke rock	
	Biotite Granite (Jecheon Granite)	
	- Intrusion -	
Ordovician	Limeslicate rock	Samtaesan
	Silty Limestone Limestone	
	Dolostone	Hongwolri

There are two major ore deposits, the porphyry deposit in the lower part and skarn deposit in the upper part. A skarn zone of over 600 m length is hosted in Ordovician carbonate sediments and is adjacent to both Jurassic and Cretaceous felsic intrusives. Widths of molybdenite-bearing skarn are up to 80~110 m wide and extend 600 m length. The stratification bedding is mainly N60~80E. Molybdenum occurs as veins in fracture zones and as disseminated forms within the skarn. The Mo mineralization age in the study area was estimated as about 157~179 Ma ago, which is K/Ar age of biotite occurring in Jecheon granite (Lee et al., 2007, p. 252). Around the study area, about 3 orebodies are known. The orebodies named ‘orebody A’, ‘orebody B’ and ‘orebody C’ are arranged in west to east. Each of orebodies is spaced about 50 m apart. The skarn ore minerals consist mainly of molybdenite, scheelite, galena and chalcopyrite (KORES, 2006).

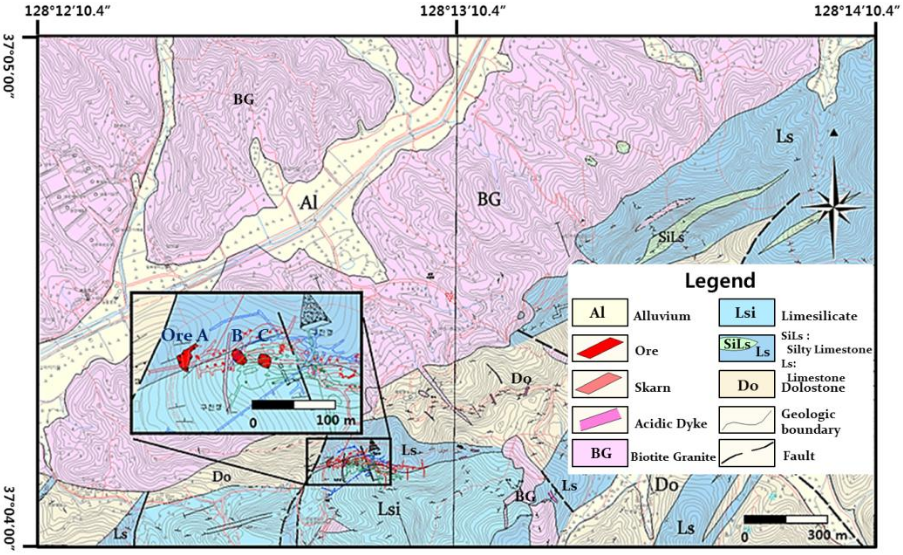
3.2 Control area

The control area, Gwanak mountain which is 632 m high, is located in the southern part of Seoul and lies in the neighboring satellite cities of Anyang and Gwacheon in Korea. The main campus of Seoul National University is located just in the northwestern corner of the mountain. This area shows under continental climate. The average annual temperature is 12.2℃, rainfall is concentrated in a short rainy season (June~September) and the seasonal climate fluctuates greatly. The total annual rainfall accounts for 1,344mm. In this area, the woods are dense with trees mainly composed of Daimyo oak, Sargent cherry, Japanese red pine and Japanese maple. Geology consists primarily of Jurassic granite and sandy soils formed through prolonged weathering.

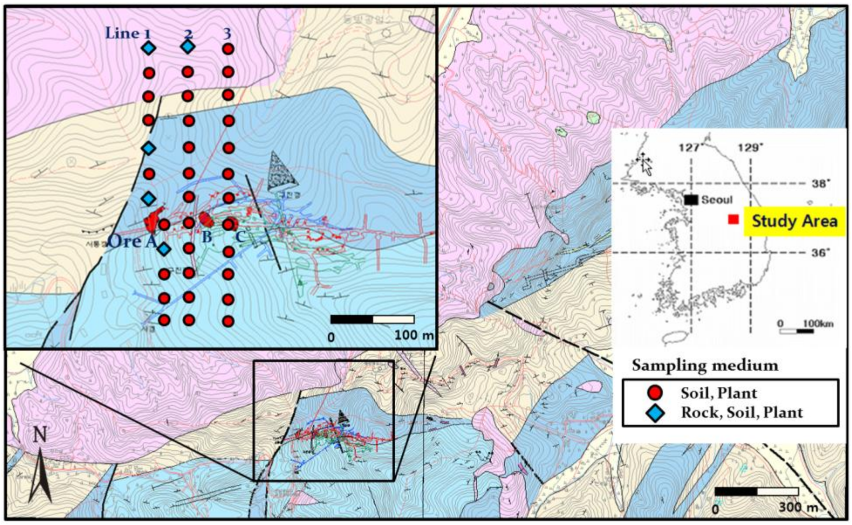
3.3 Sampling and chemical analysis

Sampling was carried out from May to June in 2011 in target (mineralized) area and barren control (nonmineralized) area. In the target area, sampling was carried out over the orebodies. Five rock samples, 36 soil samples and 108 plant samples (including three species) were collected from 3 traverse lines. The lines were designed to cross over the each orebody. Traverse line 1 is centered on the ‘orebody A’ and it is extended to north and south direction. Line 2 and Line 3 are paralleled to Line 1 and each of the lines is centered on the ‘orebody B’ and ‘orebody C’, respectively. Three lines spaced 30 or 50 m (southern part of line 1 was shifted to the east by 20 m due to the road) and sampling interval along the traverse lines is 30 m. Total length of each line is 330 m (Fig. 3.2 (b)). On the other hand, 10 points were randomly selected and 10 soil samples and 30 plant samples (including three species) were collected in the control area (Fig. 3.3).

Rock samples were collected from outcrops. Composite sampling methods were used for soils and plants sampling (Fig. 3.4). Soil samples were taken from the B horizons of approximately 15~30 cm



(a)



(b)

Fig. 3.2 Geology and sampling location map of the study area, NMC Moland mine, Jecheon, Korea.

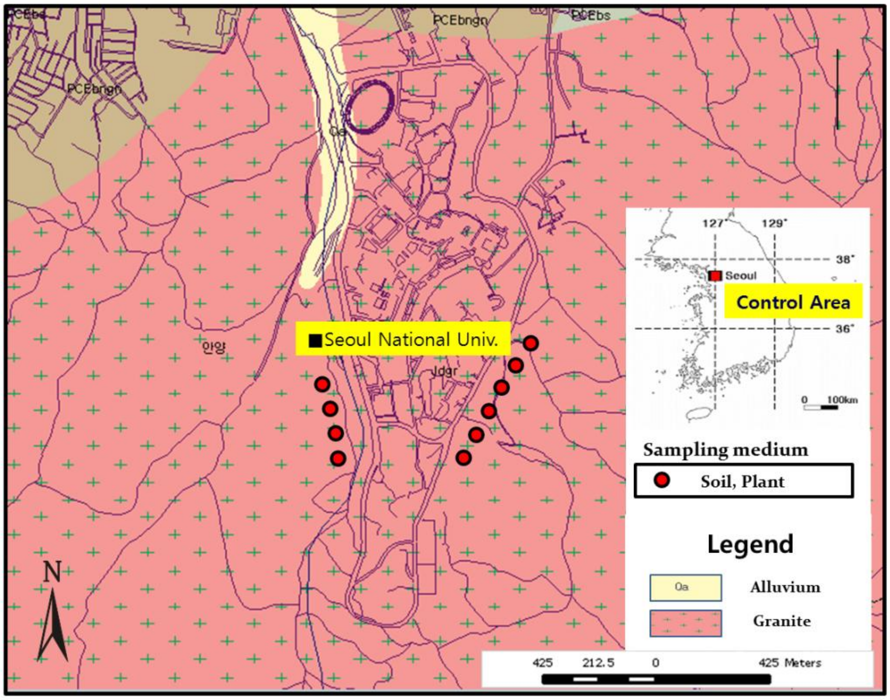


Fig. 3.3 Geology and sampling location map of the control area, Gwanak mountain, Seoul, Korea.

depth below the surface. The three plant samples in both target and control areas were daimyo oak (*Quercus dentata*) leaves and branches, and sargent cherry (*Prunus sargentii REHDER*) leaves (Fig. 3.5).

All rock and soil samples were stored and carried in polyethylene sample bags. And plant samples were transported in clean brown paper bags in order to avoid contamination. Then, each type of samples was pretreated with appropriate methods. Rock samples were washed with distilled water and dried. And then, they were crushed and pulverized with jaw crusher, and sieved to - 200 mesh (< 74 μm). Soil samples were dried at room temperature for 10 days or so and sieved to - 200 mesh. Plant samples were washed with distilled

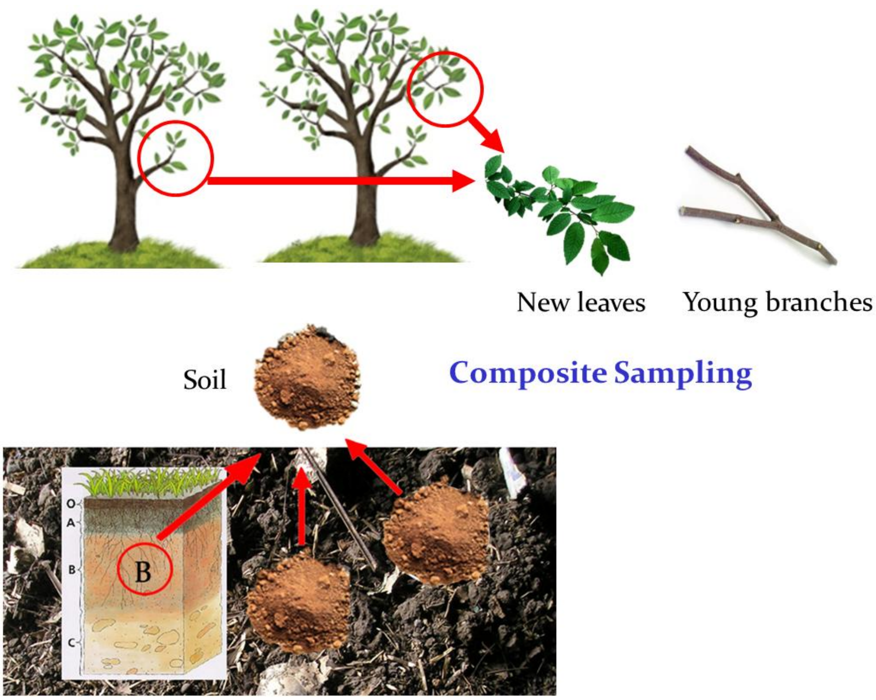


Fig. 3.4 Pictures of composite sampling for soils and plants.

water firstly so as to remove soils and dust particles, and then were dried completely at room temperature. After pretreatment, all samples were sent to a commercial laboratory, ACTLABS in Canada, for the chemical analysis of Mo and other elements by ICP-MS (Inductively Coupled Plasma Mass Spectrometry). For the rock and soil samples, 60 elements (Mo, Cu, Pb, Zn, Cd, As, Mn, Ag, Al, Ba, Be, Bi, Ca, Ce, Co, Cr, Cs, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, In, K, La, Li, Lu, Mg, Na, Nb, Nd, Ni, P, Pr, Rb, Re, S, Sb, Sc, Se, Sm, Sn, Sr, Ta, Tb, Te, Th, Tl, Ti, Tm, U, V, W, Y, Yb, Zr) were analyzed by

ICP-MS with using the 4-acid combined digestion (HF, $HClO_4$, HNO_3 and HCl).

The vegetation samples were ashed at 475℃ for a 35 hours, and acid digestion was used on the plant ash. Digested ash samples are diluted and analyzed by Perkin Elmer Sciex ELAN 6000, 6100 or 9000 ICP-MS for 59 elements (Mo, Cu, Pb, Zn, Cd, As, Mn, Ag, Al, B, Ba, Be, Bi, Ca, Ce, Co, Cr, Cs, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, In, K, La, Li, Lu, Mg, Na, Nb, Nd, Ni, Pr, Rb, Re, Sb, Sc, Se, Si, Sm, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Y, Yb, Zr).

All analysis follows the ACTLABS' quality control system; 1) a matrix blank and digested blank are each run every 35 samples, 2) two digested standards are run every 35 samples, 3) Duplicates are digested and analyzed every 14 samples, 4) Instrument is recalibrated every 70 samples. For each rock, soil, plant sample, 2, 3 and 8 duplicates are applied respectively. Microsoft Excel 2010 and MATLAB 2011 are used to interpret data, and Golden Surfer 8.0 is used to plot the spatial distribution of elements.

(a)

(b)

Fig. 3.5 Pictures of sampled plants.

(a) Daimyo oak (*Quercus dentata*)

(b) Sargent Cherry (*Prunus sargentii REHDER*)

4. Results and Discussion

4.1. Univariate statistics

A total of 60 elements for rock and soil samples, and 59 elements for plant samples were determined. We focused on the elements correlated with main skarn minerals and some geochemically associated elements (Table 2.5). Finally, seven elements of Mo, Cu, Pb, Zn, Cd, As and Mn were selected to be treated statistically.

4.1.1. Rock samples

The total number of rock samples was 5, including limestone (1), granite (2), quartz-rich granite (1) and limeslicate (1). The univariate statistical data of rock samples from target area is shown in Table 4.1. The concentration levels of Mo, Cu, Pb, Zn and As do not show any specific characteristics. Skarn orebodies of NMC Moland mine were mineralized with Mo. However, Mo value in rock samples is lower than that of average normal rocks. Other elements such as Cu, Pb, Zn and As also show lower values compared to those of average normal rocks (Levinson, 1980).

Table 4.1 Univariate statistics of elements in rock samples. (unit: ppm)

		Mo	Cu	Pb	Zn	As
Target area	Limestone (n=1) [a]	1	2.6	1.3	nd	5.1
	Granite (n=2)	1	2.6	1.3	nd	5.1
	Quartz-rich Granite (n=1)	0.8	1.5	2.1	9.6	4.4
	Limesilicate (n=1)	nd	1.7	2.7	20.5	1.7
Detection limit		0.1	0.2	0.5	0.2	0.1
Average normal rocks (Granite, Limestone) [b]		1~3	10~15	8~20	25~40	1.5~2.5

nd : not detected.

[a] n = number of samples.

[b] Average normal rocks (Granite, Limestone) (Levinson, 1980).

4.1.2 Soil samples

The total contents of Mo, Cu, Pb, Zn, Cd, As and Mn in soil are shown in Table 4.2, and box plots of element concentrations in soil are presented in Fig. 4.1. The mean Mo concentration in soil from target area (5.5 ppm Mo) is 5 times higher than that in the control area (1.1 ppm Mo) and also in average normal soil (Kabata-Pendias, 2010). One of the soil samples in target area yields very high anomalous Mo value of 38.7 ppm. In addition, the mean concentration of Mn in soil from target area (1365 ppm Mn) is higher than that from the control area (479 ppm Mn). Regarding other elements such as Cu, Pb, Zn, Cd and As, the mean concentrations in target area do not show enhanced geochemical contrast compared to those in the control area. However, the maximum metal contents from some sampling locations are slightly high (80.4 ppm Cu, 294 ppm Pb, 377 ppm Zn and 62.8 ppm As) compared with those in the control area (43.7 ppm Cu, 180 ppm Pb, 225 ppm Zn and 14 ppm As). In particular, the mean values of Pb and Zn in soil samples from control area show higher values than average normal soil. The concentrations of Pb and Zn from target area indicate relatively 2~3 times high values compared with normal soils.

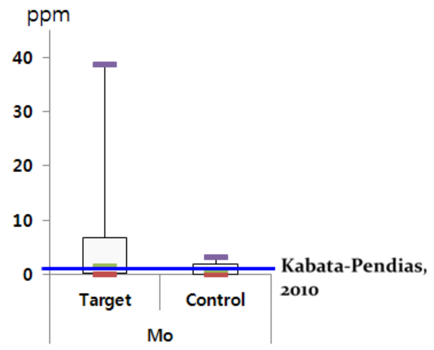
Table 4.2 Univariate statistics of elements in soil samples. (unit: ppm)

		Mo	Cu	Pb	Zn	Cd	As	Mn
Target area (n=36)[a]	**Range**	nd~38.7	5.4~80.4	27.2~294	56.7~377	nd~0.9	3.3~62.8	276~3520
	Mean	5.5	29.6	73.5	173.3	0.3	11.0	1365
Control area (n=10)	**Range**	nd~3.2	21.9~43.7	57.9~180	115~225	0.1~0.3	7.2~14	297~976
	Mean	1.1	34.5	97.6	157.7	0.2	10.2	479
Detection limit		0.1	0.2	0.5	0.2	0.1	0.1	1
Average normal soils [b]		1.1	38.9	27	70	0.41	6.83	488

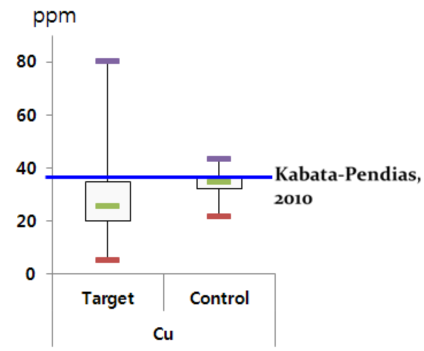
nd : not detected.

[a] n = number of samples.

[b] Average normal soils (Kabata-Pendias, 2010).



(a)



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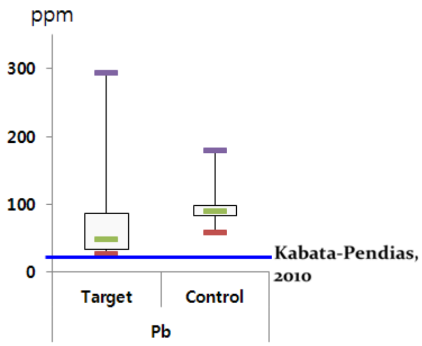

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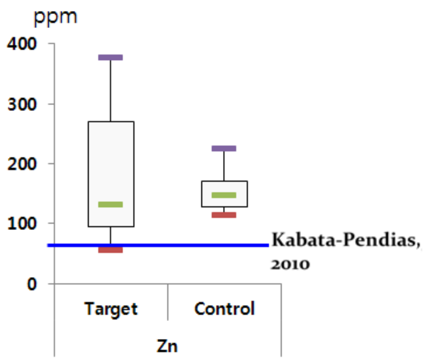

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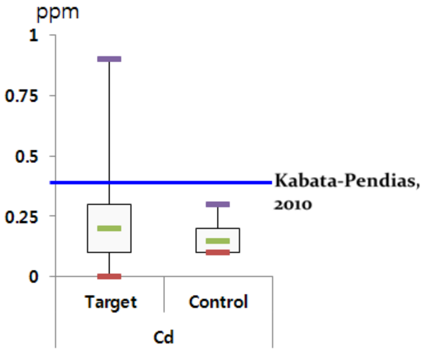

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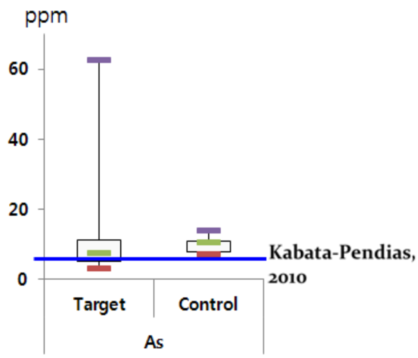
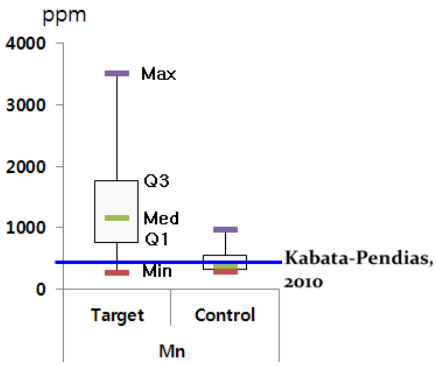


(f)

Fig. 4.1 Box plots of element concentrations in soil samples.

(a) Mo, (b) Cu, (c) Pb, (d) Zn, (e) Cd, (f) As, (g) Mn



(g)

Fig. 4.1 Continued.

4.1.3. Plant samples

Three kinds of plant samples including *Q. dentata* leaves and branches, and *P. sargentii* leaves were collected, and a total of 108 samples from target area and 30 samples from control area were analyzed. Univariate statistics of Mo, Cu, Pb, Zn, Cd, As and Mn are summarized in Table 4.3, and boxplots of element contents for each sample are shown in Fig. 4.2.

The mean concentration of Mo in the plant is highest in *Q. dentata* leaves (0.86 ppm Mo), next in *P. sargentii* leaves (0.83 ppm Mo) and then in *Q. dentata* branches (0.53 ppm Mo). The concentration level of Mo in *Q. dentata* leaves and branches, and *P. sargentii* leaves is within the range of the reference plants (Kabata-pendias, 2010).

However, Mo concentration from the target area (0.14~4.91 ppm Mo in *Q. dentata* leaves, 0.01~3.19 ppm Mo in *Q. dentata* branches and 0.15~6.56 ppm Mo in *P. sargentii* leaves) is 4~8 times higher than that from the control area (0.08~0.47 ppm Mo in *Q. dentata* leaves, 0.02~0.26 ppm Mo in *Q. dentata* branches and 0.14~0.42 ppm Mo in *P. sargentii* leaves). On the other hand, concentrations of other elements including Cu, Pb, Zn, Cd, As and Mn in plant samples are not significantly different at the 95% significance level.

Boxplots of Mo and other elements shows the geochemical between samples from the target area and from the control area. Molybdenum concentration in leaves and branches of *Q. dentata*, and in leaves of *P. sargentii* from the target area is extremely higher than that from the control area. In boxplots of other elements, plant samples from target area do not show any significant contrast compared with those from the control area and with values of reference plants.

From the above results, it is possible that Mo exists in a bioavailable form compared with those to be easily absorbed by plants. All plant samples such as *Q. dentata* leaves and branches, and *P. sargentii* leaves seem to be possible as Mo accumulators.

Table 4.3 Univariate statistics of elements in plant samples. (unit: ppm)

			Mo	Cu	Pb	Zn	Cd	As	Mn
Target Area	Q. dentata leaves (n=36) [a]	range	0.14~4.91	5.23~9.73	0.65~1.58	15.4~25.3	0.03~0.14	0.20~0.44	26.8~887.0
		mean	0.86	6.98	1.10	18.7	0.07	0.30	373.1
	Q. dentata branches (n=36)	range	0.01~3.19	2.88~5.51	0.58~6.88	9.3~21.4	0.06~0.20	0.19~1.65	32.7~440.0
		mean	0.53	3.86	1.92	13.9	0.12	0.88	217.3
	P. sargentii leaves (n=36)	range	0.15~6.56	4.16~8.54	0.68~3.47	9.5~22.8	0.02~0.08	nd~0.59	20.9~668.6
		mean	0.83	5.46	1.52	16.4	0.05	0.22	327.4

nd : not detected.

[a] n = number of samples.

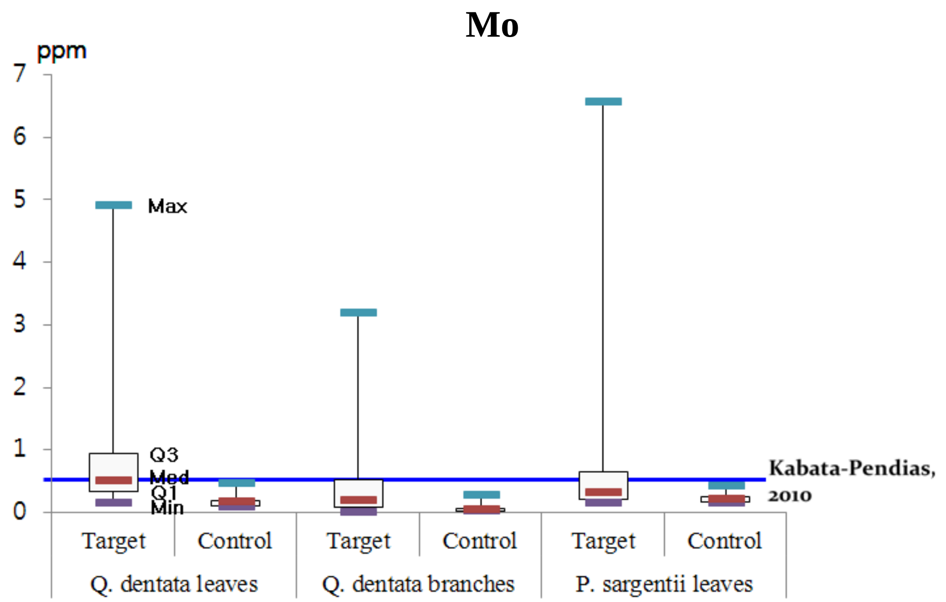
4.3 Continued. (unit: ppm)

			Mo	Cu	Pb	Zn	Cd	As	Mn
Control Area	**Q. dentata leaves (n=10)** [a]	**range**	0.08~0.47	6.45~12.37	1.12~2.81	20.8~37.3	0.04~0.12	nd	198.9~798.3
		mean	0.18	8.23	1.96	26.3	0.08	nd	426.1
	Q. dentata branches (n=10)	**range**	0.02~0.26	2.90~5.30	0.96~4.50	10.4~47.4	0.06~0.19	0.42~1.37	76.2~449.5
		mean	0.07	4.00	2.48	20.2	0.13	0.75	251.0
	P. sargentii leaves (n=10)	**range**	0.14~0.42	5.98~11.56	2.87~7.67	19.2~55.2	0.02~0.12	nd~1.66	147.4~1690.6
		mean	0.23	8.11	4.78	33.4	0.05	0.85	651.7
Detection limit			0.1	0.2	0.1	1	0.01	1	0.1
Data for Reference plant [b]			0.2~0.9	5~20	1.5~2.4	12~47	0.07~0.46	0.5~80(ppb)	17~334

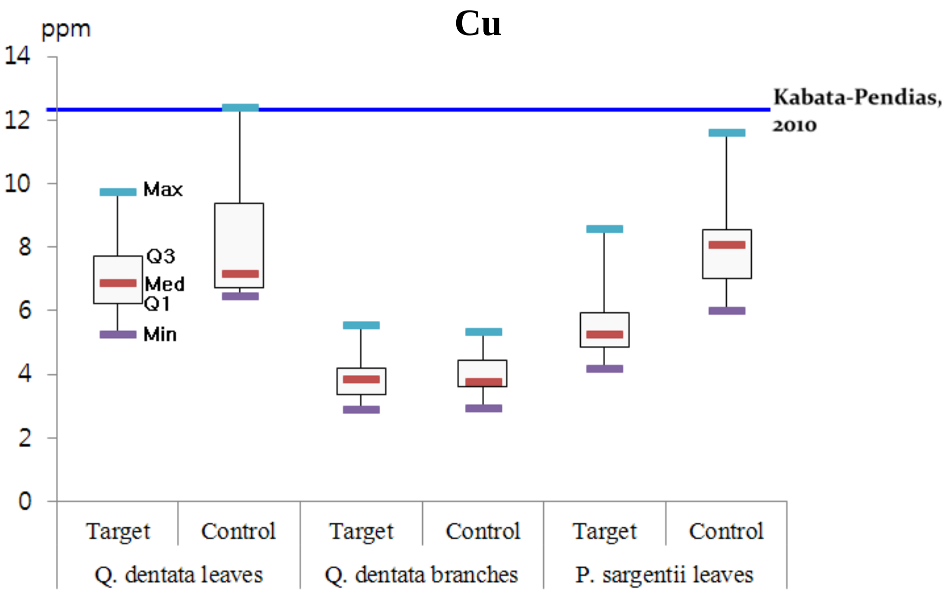
nd : not detected.

[a] n = number of samples.

[b] Average normal plants (Kabata-Pendias, 2010).

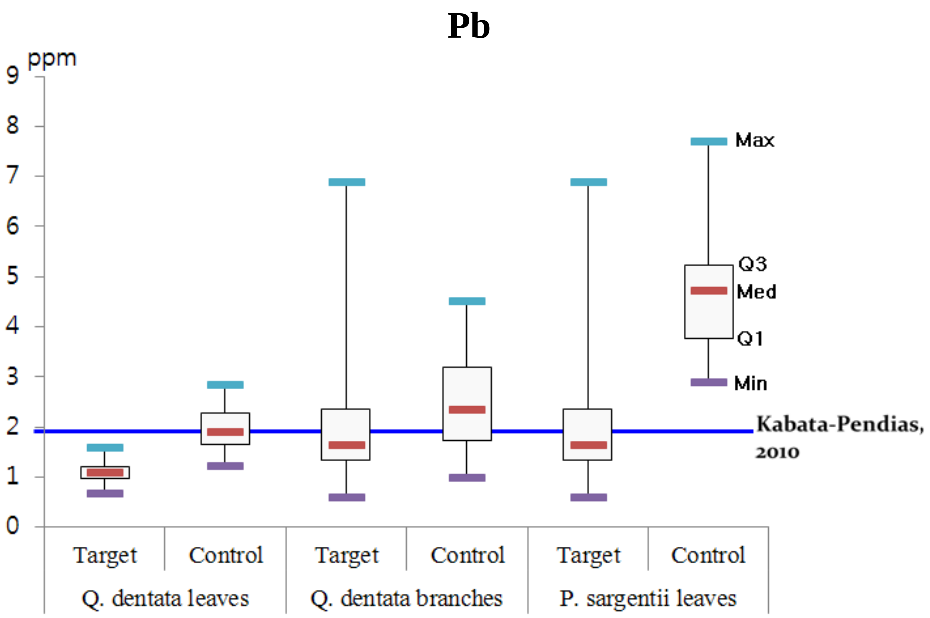


(a)



(b)

Fig. 4.2 Boxplots of element concentrations in plant samples.
(a) Mo (b) Cu (c) Pb (d) Zn (e) Cd (f) As (g) Mn



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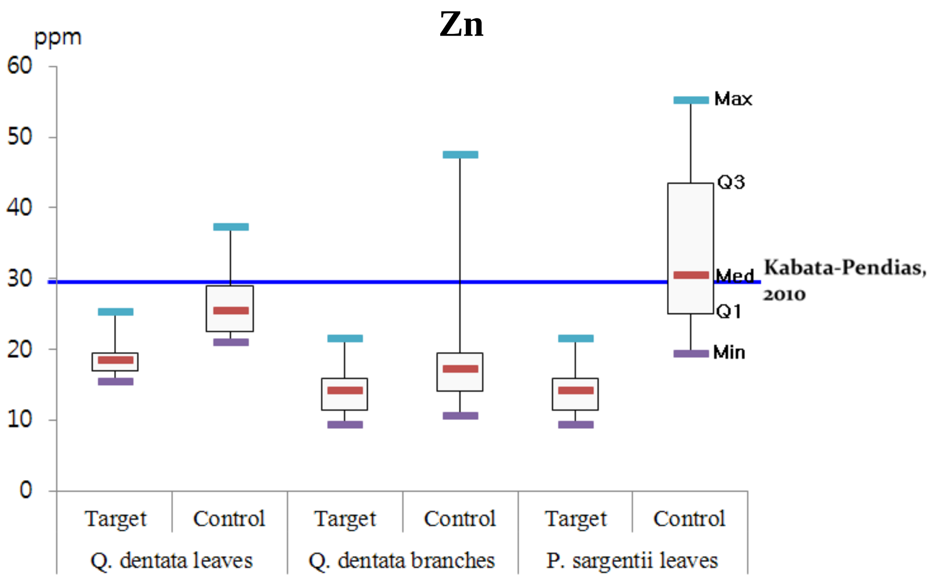
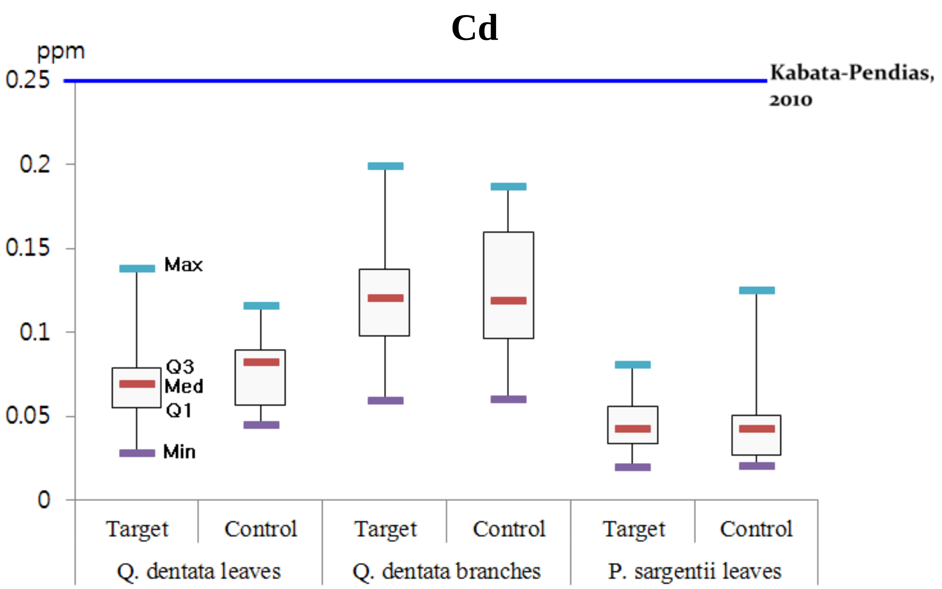


(d)

Fig. 4.2 Continued.



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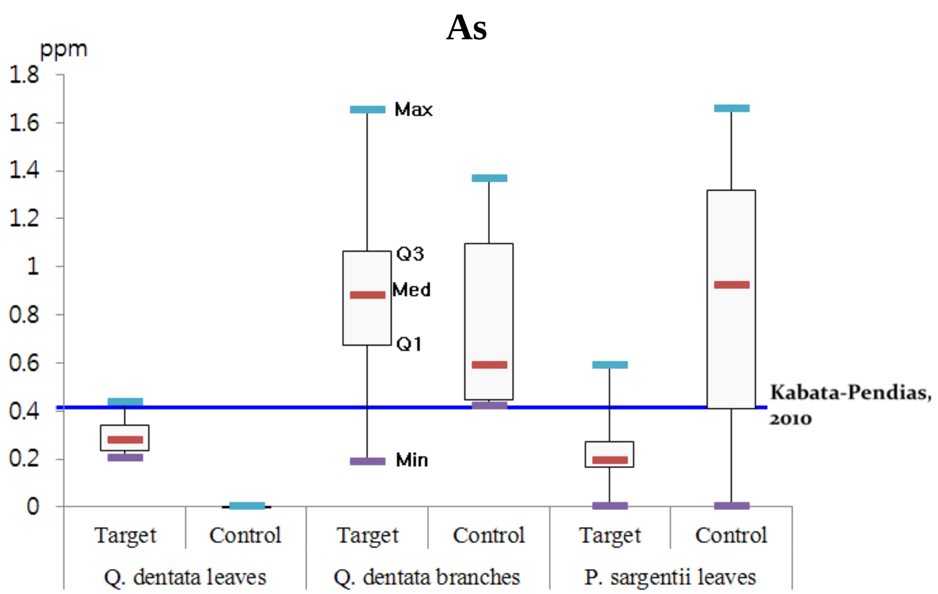
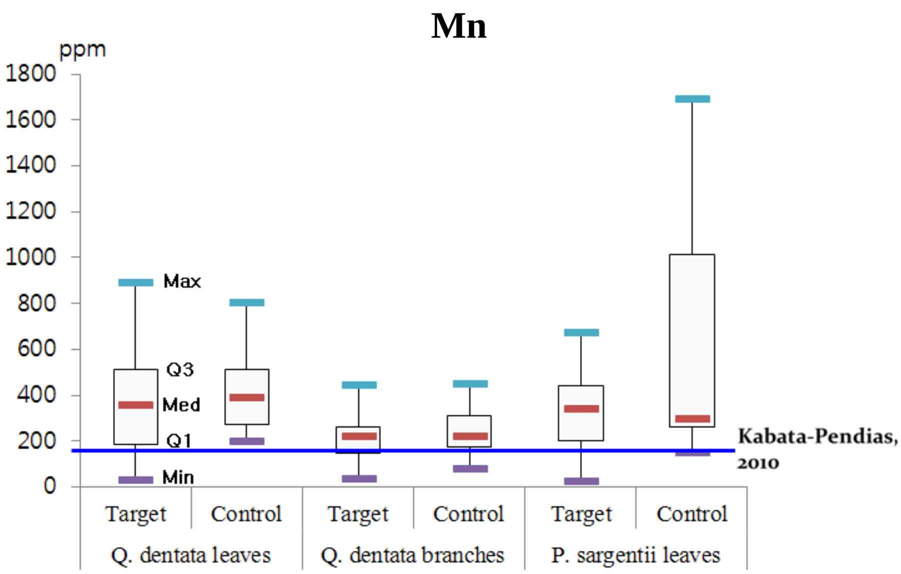


(f)

Fig. 4.2 Continued.



(g)

Fig. 4.2 Continued.

4.2. Relationship between soil and plant

The BAC (biological absorption coefficient) means the ratio of metal concentration in plant to that in soil.

$$BAC = C_p/C_s$$

C_p : concentration of an element in plant

C_s : concentration of the same element in the substrate

The BAC of each plant sample is shown in Table 4.4. The BAC of Mo in *Q. dentata* leaves and branches, and *P. sargentii* leaves shows generally high value (*Q. dentata* leaves 1.4, *Q. dentata* branches 0.4 and *P. sargentii* leaves 1.2). Also the BAC of Cu, Zn, Cd and Mn is considerably high. However, other elements such as Pb and As present the low BAC values probably due to the low mobility and less amounts of elements in plants. From these results, *Q. dentata* leaves and *P. sargentii* leaves could be considered as good accumulators as well as good indicators of Mo mineralization. The correlations between element concentrations in soils and those in plants are summarized in Fig. 4.3. In particular, Mo in soil has a strong positive correlation with all three plant samples compared with other elements.

4.3. Bivariate analysis

Molybdenum in soil and three plant samples does not show any significant correlations with other elements (Fig. 4.4 to Fig. 4.7). Determined elements of Cu, Pb and Zn and other geochemically associated elements (Table 2.4) were checked for the correlations with Mo. However, complicated mechanism of plant metabolism and barrier uptake systems in the plant may influence the lack of significant correlation between Mo and other metal concentrations in plant samples (Dunn, 2007 a; Jung et al., 2011 b).

Table 4.4 BAC value of each plant sample in the target area.

		Q. dentata leaves (n=36) [a]	**Q. dentata branches (n=36)**	**P. sargentii leaves (n=36)**
Mo	Range	0.015 ~ 5.84.	0.009 ~ 1.962	0.008 ~ 6.465
	Mean	1.4	0.4	1.2
Cu	Range	0.072 ~ 1.301	0.038 ~ 0.713	0.068 ~ 1.024
	Mean	0.3	0.2	0.2
Pb	Range	0.003 ~ 0.058	0.006~0.166	0.003~0.068
	Mean	0.02	0.04	0.03
Zn	Range	0.044 ~ 0.324	0.026 ~ 0.285	0.036 ~ 0.389
	Mean	0.1	0.1	0.1
Cd	Range	0 ~ 1.199	0 ~ 1.982	0 ~ 0.709
	Mean	0.3	0.6	0.2
As	Range	0.004 ~ 0.123	0.019 ~ 0.401	0.002 ~ 0.117
	Mean	0.05	0.1	0.04
Mn	Range	0.013 ~ 1.9	0.016 ~ 1.228	0.018 ~ 2.265
	Mean	0.4	0.2	0.4

[a] n = number of samples.

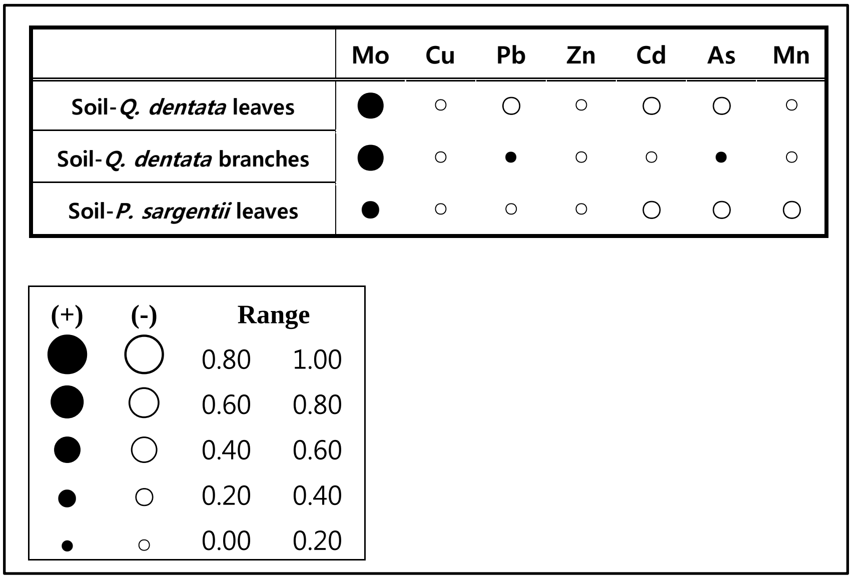


Fig. 4.3 Correlations of elements between in soil and plant samples.

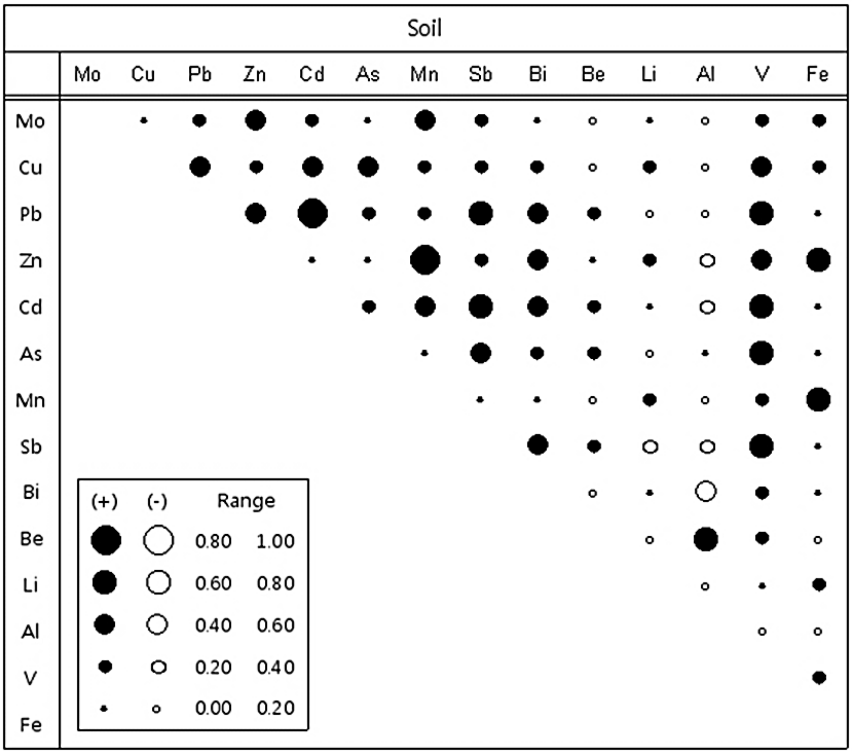


Fig. 4.4 Correlations of element pairs in soils.

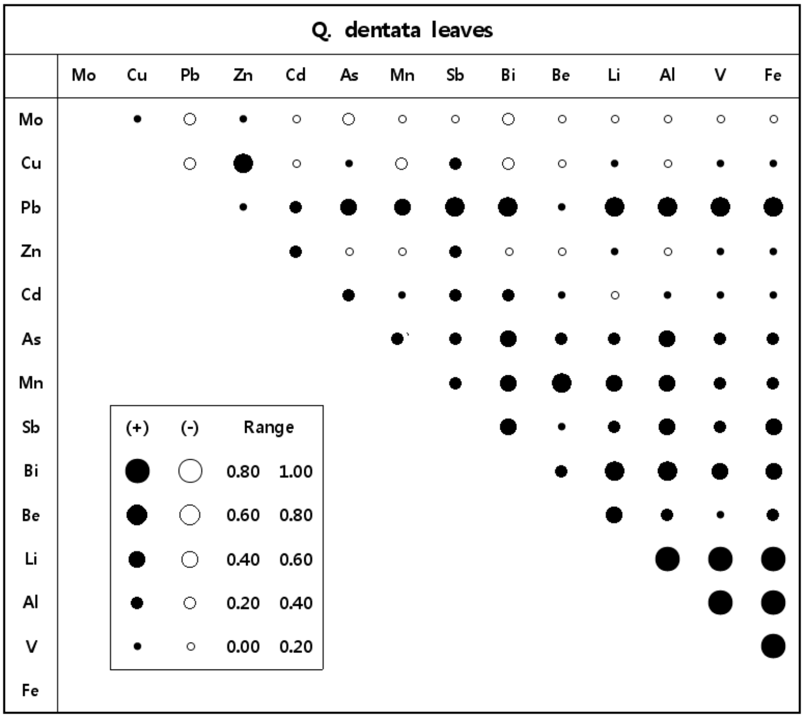


Fig. 4.5 Correlations of element pairs in *Q. dentata* leaves.

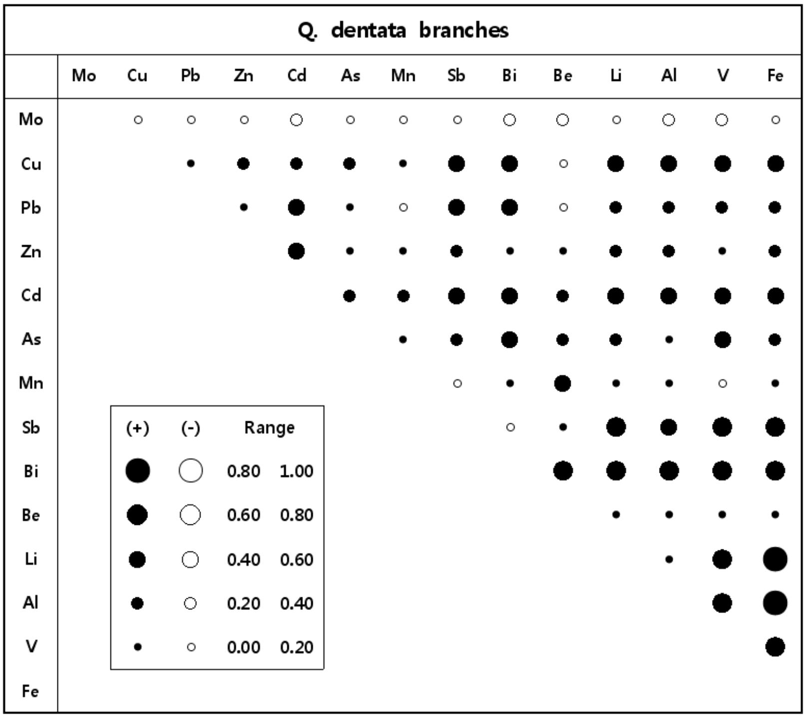


Fig. 4.6 Correlations of element pairs in *Q. dentata* branches.

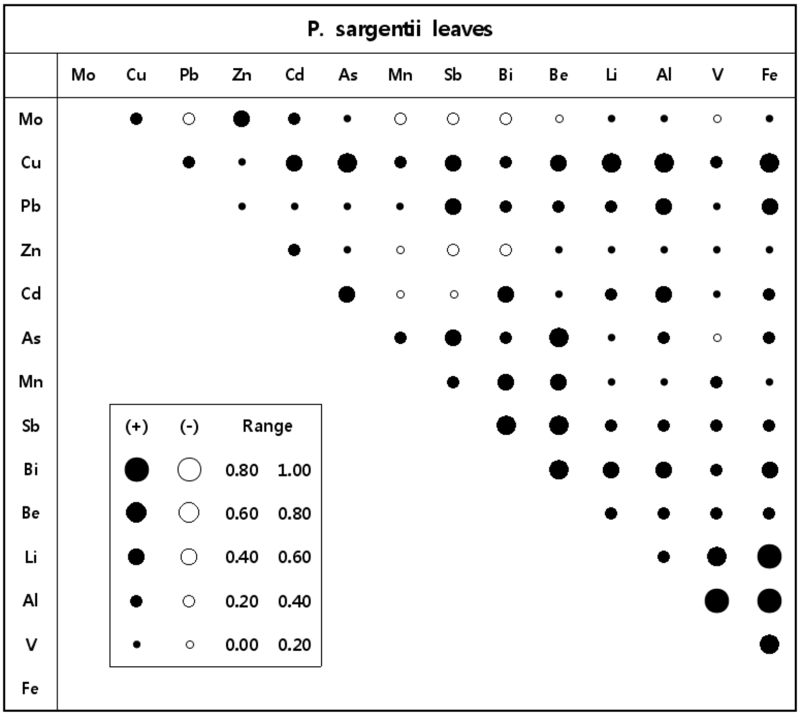
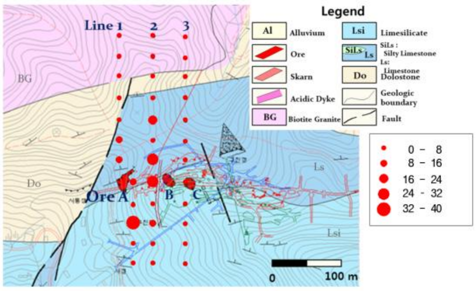


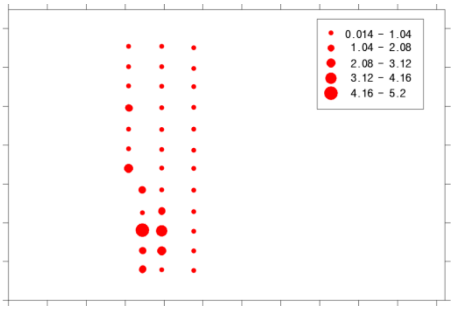
Fig. 4.7 Correlations of element pairs in *P. sargentii* leaves.

4.4. Spatial distribution of elements

Spatial distribution of Mo and Mn in soil and plant samples is shown in Fig. 4.8 and Fig. 4.9, respectively. From Fig. 4.8 (a), Mo in soil samples is enriched near the orebodies. Accordingly, the high value of Mo in plants is shown at the southern part of the orebodies (Fig. 4.8 (b), (c)). In other words, Mo concentration of *Q. dentata* leaves and branches show very similar distribution trends with that in soil. Especially, Mo level of *Q. dentata* leaves and branches exceeds 3~4 ppm Mo near the orebodies. However, *P. sargentii* leaves from traverse line 2 do not reflect the anomaly of the soil samples. The possible reasons could be due to the different leaf size. In general,



(a)



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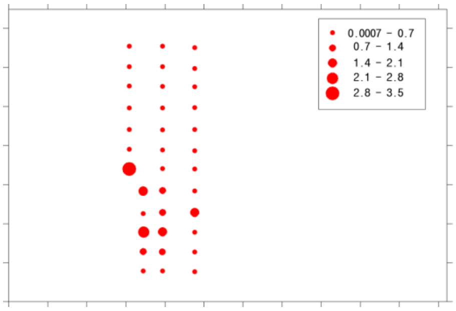

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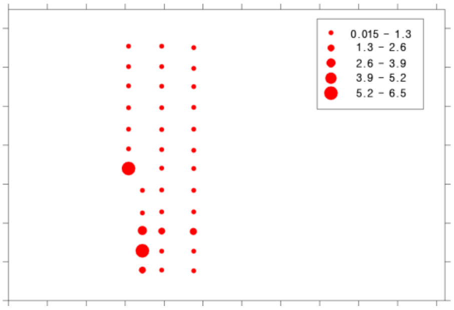


(d)

Fig. 4.8 Spatial distribution of Mo by sampling medium.
(a) Soil (b) *Q. dentata* leaves
(c) *Q. dentata* branches (d) *P. sargentii* leaves

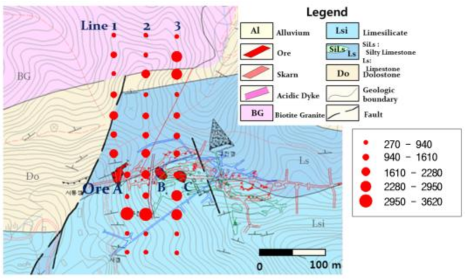
broadleaf trees are more easily uptake the elements compared to needleleaf trees. Among broadleaf trees, plants with large leaf size faithfully reflect the anomalous value of elements in soil. In traverse line 3, Mo concentrations in soil and plants are generally lower than those in other two traverse lines.

Spatial distribution of Mn in soil and plant samples is presented in Fig. 4.9. The manganese content in soil samples shows significant anomaly at some sampling points regardless of orebodies. In three plant samples, however, the concentration of Mn is relatively high near the orebodies and in the area of biotite granite and silty limestone. Moreover, the pattern of Mn concentration is similar in plant samples along the each traverse line.

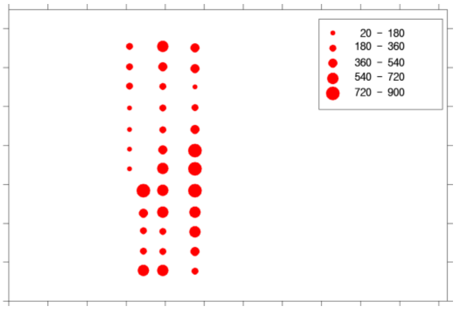
The patterns of other elements (Cu, Pb, Zn, Cd and As) in soils and plants do not represent similarities between soils and plants.

4.5. Geochemical variation

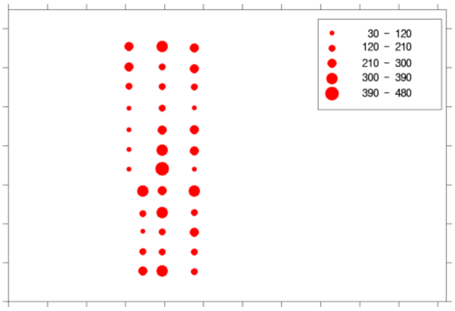
Geochemical variations of Mo and Mn along the three sampling traverse lines in soil and plant samples are plotted in Fig. 4.10 and Fig. 4.11. Molybdenum concentration is high at the point of S60 (60 m south of orebody) and another point of N30 (30 m north of orebody) in soil samples from traverse line 1. Peaks of Mo in plant samples are very similar to the result of the soils (Fig. 4.10 (a)). Also, Mo value is high near the orebody (within the point of N90~S60) except for the



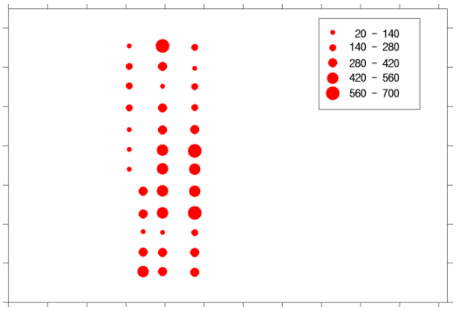
(a)



(b)



(c)



(d)

Fig. 4.9 Spatial distribution of Mn by sampling medium.
(a) Soil (b) *Q. dentata* leaves
(c) *Q. dentata* branches (d) *P. sargentii* leaves

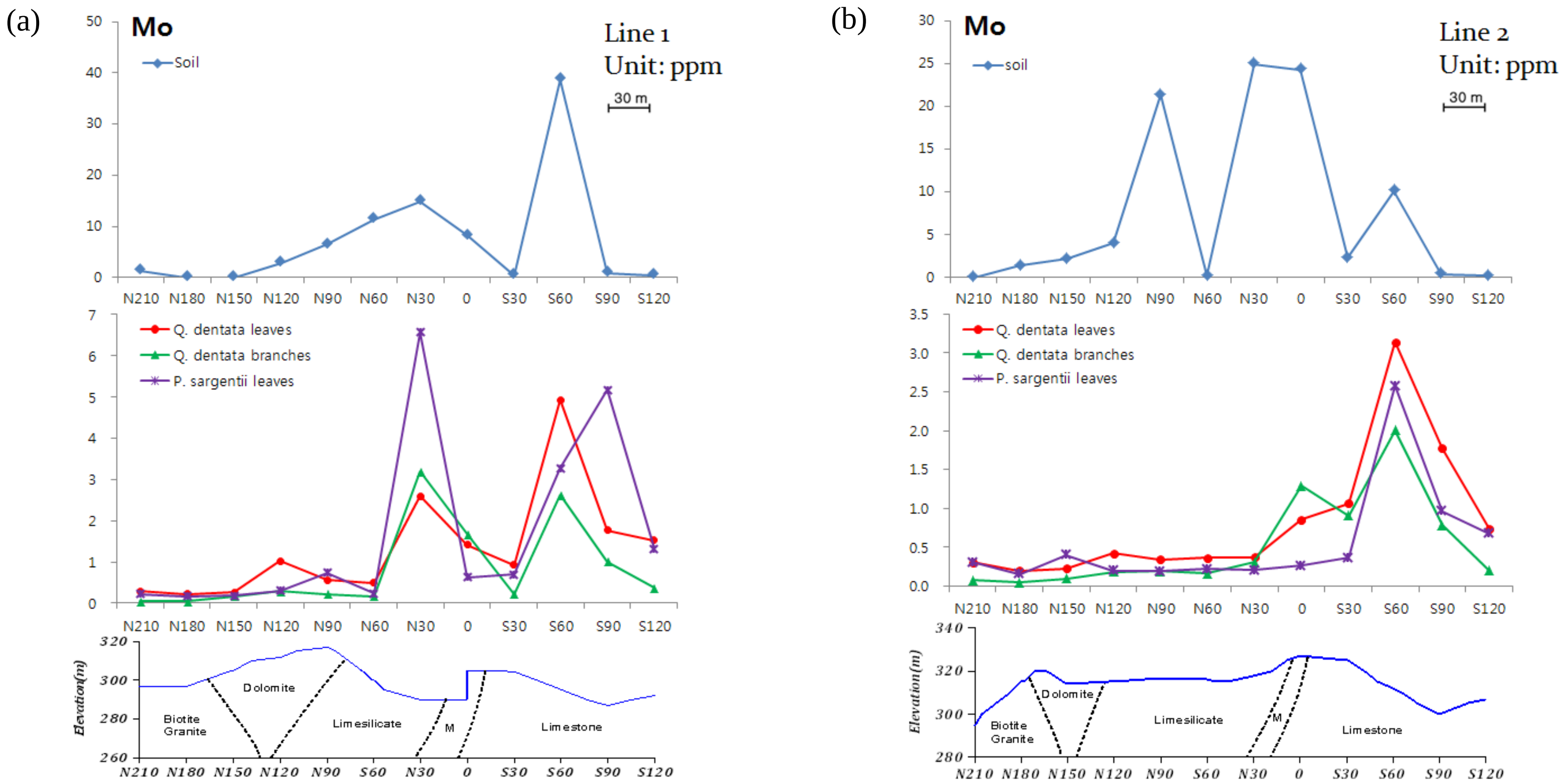


Fig. 4.10 Geochemical variation of Mo in soils and plants. (a) Line 1 (b) Line 2 (c) Line 3

(c)

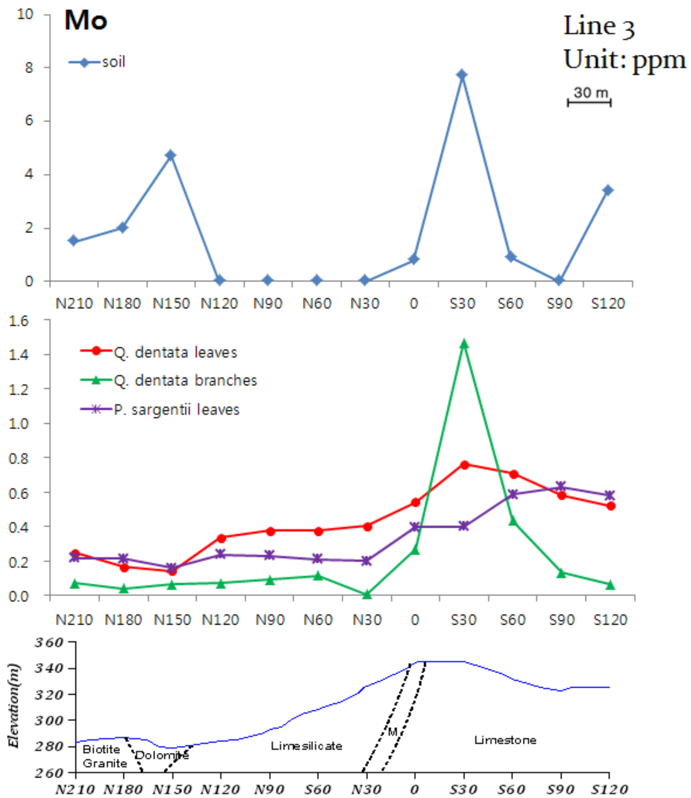


Fig. 4.10 Continued.

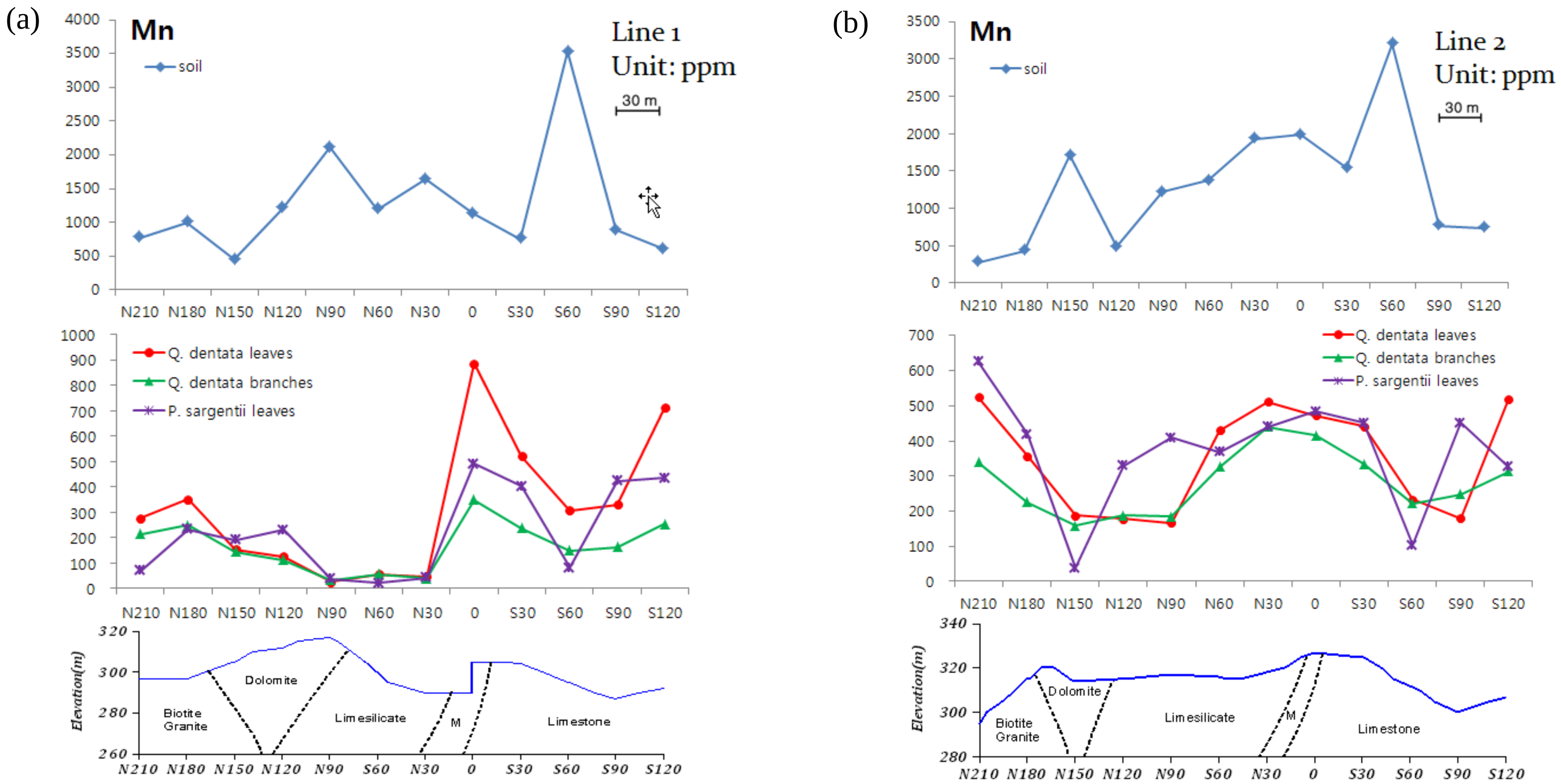


Fig. 4.11 Geochemical variation of Mn in soils and plants. (a) Line 1 (b) Line 2 (c) Line 3

(c)

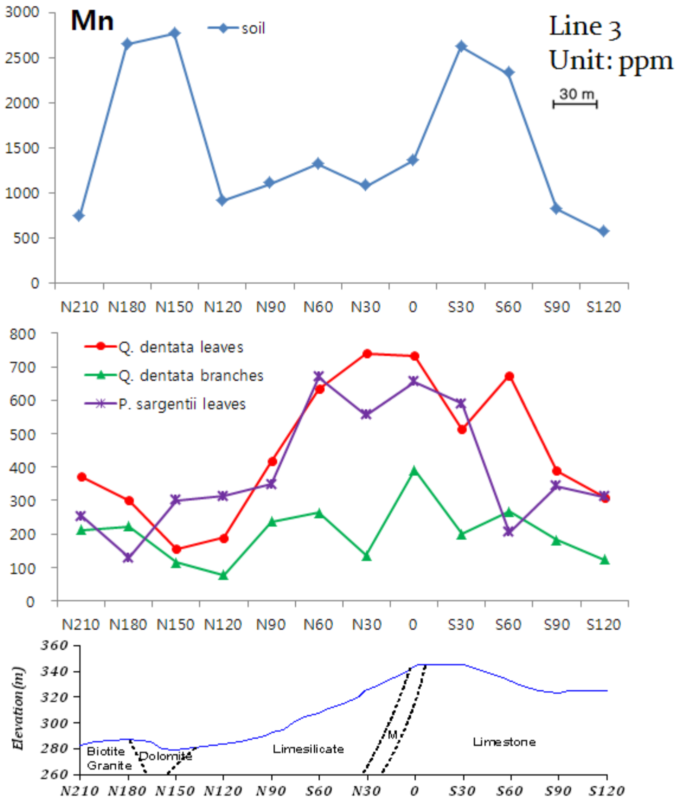


Fig. 4.11 Continued.

point of N60 in line 2. Accordingly three plant samples show high peaks within the points of 0 to S120. This geochemical variation of the plants makes a pattern to move slightly to the south from the orebody compared to that in the soils (Fig. 4.10 (b)). In line 3, Mo has a similar geochemical variation pattern in soils and plants. Especially Mo peaks of soils and *Q. dantata* branches show well coincidence (Fig. 4.10 (c)). There is no specific tendency of Mn content between soils and orebody. However, three plant samples show similar trends relatively. High Mn concentration is shown near each orebody. And then, concentration decreases gradually while moving to both of the ends.

5. Conclusions

A biogeochemical orientation study was conducted in Mo skarn deposits, Jecheon district in Korea. The main objective of this study was to investigate the concentration level and migration of elements in rock-soil-plant system and to evaluate the applicability of biogeochemical prospecting for mineral resources.

1) Rock samples from Mo skarn deposits do not show anomalous value of elements compared with average normal rocks. Soil samples (n=36/10, target/control) collected from the study area show higher values of Mo (<0.1~38.7 ppm) and Mn (276~3520 ppm) compared to those from the control area (<0.1~3.2 ppm Mo and 297~976 ppm Mn). The mean concentration of Mo in all of the plant samples (*Q. dentata* leaves and branches, and *P. sargentii* leaves) is 4 to 8 times higher than that from the control area. In target area, range of Mo concentration in *Q. dentata* leaves and branches is 0.14 to 4.91 ppm and 0.01 to 3.19 ppm, respectively, and that in *P. sargentii* leaves is 0.15 to 6.56 ppm. However, there is no specific other elements contrast in plants.

2) The BAC (Biological Absorption Coefficients) of Mo in plant organ is high in the order of *Q. dentata* leaves (1.4), *P. sargentii* leaves (1.2) and *Q. dentata* branches (0.4). Also the BAC of Cu, Zn, Cd and Mn is relatively high (0.1~0.6). However, Pb and As

show low BAC values (0.02~0.1). In addition, Mo content in soils and plants is strongly correlated compared to other elements, whereas, Mo in soils and plants does not show any significant correlations with other elements.

3) Molybdenum concentration in soils and plants is spatially well correlated with Mo skarn orebodies. Meanwhile, there is no specific tendency of Mn content between soils and orebodies. However, distribution of Mn in three plant samples shows spatially good correlation with Mo skarn orebodies.

4) The geochemical variation patterns of Mo in plants are generally similar to those in soils, which suggest a corresponding Mo anomaly and enhanced contrast near the Mo orebodies. The three plant organs have high possibilities to be used as indicators for the biogeochemical prospecting of Mo.

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초 록

연구지역인 NMC Moland 광산은 행정구역상 충청북도 제천시 금성면에 위치하고 있는 스카른 타입 광상으로 휘수연석과 같은 Mo 광물이 산출된다. 주요 지질은 제천화강암이라고 불리는 쥬라기의 화강편마암이 관입한 오오도비스기의 백운암, 석회암, 석회규산염암들로 구성되어있다. 스카른은 화강암과 석회암의 경계면에 부존하고 있으며 스카른 내 파쇄대에서 Mo이 판상, 산점상으로 배태되어 있다. 주요 광물은 휘수연석, 회중석, 방연석, 황동광 등이 있다. 대조지역으로 선정된 곳은 서울대학교 본 캠퍼스가 위치하고 있는 관악산이며 지질은 주로 쥬라기의 화강암으로 이루어져있다. 시료는 2011년 5월과 6월 사이에 암석, 토양 및 식물을 대상으로 채취하였으며, 식물은 우세종인 떡갈나무(*Quercus dentata*, daimyo oak)의 잎과 가지, 산벚나무(*Prunus sargentii REHDER*, Sargent Cherry)의 잎을 채취하였다. 채취한 시료들은 ICP-MS으로 Mo, Cu, Pb, Zn, Cd, As, Mn 등의 원소를 대상으로 분석하였다. 연구지역의 시료채취선은 각 광체를 수직으로 지나도록 남북방향으로 총 3개를 잡았으며 각 시료채취선에서 30 m 간격으로 총 암석 5개(임의의 지점), 토양 36개 식물 108개의 시료를 채취하였다. 대조지역에서는 임의로 10개의 시료채취점을 선택하여 토양 10개, 식물 30개의 시료를 채취하였다.

연구지역의 암석, 토양, 식물시료 내 Mo, Cu, Pb, Zn, Cd, As, Mn을 대상으로 분석해본 결과, 암석시료의 경우 연구지역의 시료에서 특별히 눈에 띄는 지구화학적 특징은 보이지 않았다. 토양시료는 연구지역에서 Mo의 함량(0.1~38.7 ppm)이 대조지역에서의 Mo의

함량(0.1~3.2 ppm) 보다 높았다. 연구지역의 식물 시료 내 존재하는 Mo의 평균함량은 떡갈나무 잎(0.86 ppm; 0.14~4.91 ppm), 산벚나무 잎(0.83 ppm; 0.15~6.56 ppm), 떡갈나무 가지(0.53 ppm; 0.01~3.19 ppm)의 순서로 그 함량이 높았다. 이 값은 대조지역에서의 떡갈나무 잎(0.18 ppm; 0.08~0.47 ppm), 산벚나무 잎(0.23 ppm; 0.14~0.42 ppm), 떡갈나무 가지(0.07 ppm; 0.02~0.26 ppm)에 비하여 4배에서 8배 가량 높은 수치였다. 생물학적 흡수계수(BAC)의 결과는 Mo의 경우 떡갈나무 잎에서 1.4, 가지에서 0.4, 산벚나무 잎에서 1.2의 수치를 보여 세 식물, 특히 떡갈나무 잎과 산벚나무 잎이 높은 BAC 값을 나타냈다. 또한 Mo 원소는 토양과 식물 사이의 상관성이 다른 원소에 비하여 매우 높은 것으로 밝혀졌다. 더불어 공간적인 분포(spatial distribution)와 지구화학적 변동(geochemical variation)으로부터 토양과 식물내의 Mo의 함량의 변화는 Mo 이상대를 잘 반영하고 있음을 알 수 있었다.

결과를 종합하여 보았을 때 Mo 원소는 식물에 의해 잘 흡수될 수 있는 형태로 존재할 가능성이 높으며, 기관내의 원소 함량이 토양 내의 원소 함량을 잘 반영하는 지시식물인 indicator로서 Mo을 대상으로 생지구화학탐사시에 활용될 가능성이 있다.

주요어: 생지구화학탐사, Mo 스카른 광상,
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