Map of Selenium Content in Soil in Japan

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We present a new selenium (Se) map of Japan in more detail than in our previous report (*J. Health Sci.*, 42, 360–366). It contains 150 measured points, especially in the northeast of Japan. In the new sampling points, we did not find high Se levels and most of the measurements were generally at values below 1 mg/kg. In the Se map of Japan, there are two particularly high measurements, at Mt. Zao (10 mg/kg) and Tateyama Murodo Jigokudani (148 mg/kg). These two areas are the origin of a minor branch of the Abukuma River and the Jouganji River, respectively. We also analyzed the Se levels near two major copper mines, Asio and Besshi. We detected high Se content in rocks at both copper mines and an average Se level in soil from the area near the Asio copper mine. The Se level in soil at Besshi was slightly higher than that at the area near the Asio copper mine. We also measured the Se levels of some pyrites in Japan. The Se levels of most pyrites were some ten mg/kg higher than the general Se level in soil. This study provides information about the fundamental background values of Se levels in soil and rocks to correlate with Se pollution if it occurs in the future.

Key words —— Selenium, Se, soil, pyrite, Soil map, Japan

INTRODUCTION

Selenium (Se) is an element with two main characteristics: it is both toxic and is an essential part of nutrition. It was first found to be toxic in the early 20th century. It has been well documented that livestock in some regions of the U.S.A. have excess levels of Se, which causes their hooves to drop off, as reviewed by Trelease.[1] These livestock ate Se-accumulating plants, such as *Astragalus*, a kind of vetch. The Se content in these plants can be as high as 2000–5000 mg/kg.[2] This accumulation is partially dependent upon the Se content in the soil.[3] The toxicity of Se can be a problem for humans. To avoid either Se deficiency or excess in humans, the optimum intake is 50–200 μg/day.[4] Humans receive this amount from food as organic and inorganic Se (organic Se is preferable).

Se, as an essential part of nutrition was first reported in 1957 by Schwarz and Foltz, who called it factor 3.[5] Later Se was found to occur in the form selenocysteine (Sec), and glutathione peroxidase (GPx) also contains Sec at the active site. There are many Se-proteins, such as 5′ iodothyronine deiodinase, thioredoxin reductase, selenoprotein-P, and the GPx family.[6] The Sec codon is UGA which it shares with the major stop codon UGA. Sec has its tRNA and codon and is called as the 21st amino acid. We previously clarified the recognition sites on the $tRNA^{Sec}$ for Sec synthase.[7] The first step in the discrimination mechanism between the stop and Sec UGA codons was clarified.[8] However, most of the mechanism remains to be resolved.

In the preparation of a Se map of Japan, we initially reported a basic version.[9] In the previous map, the highest Se level measured was at Tateyama Murodo Jigokudani at a concentration of 148 mg/kg. The broad area of Murodo showed high Se levels. Another high Se level of 10 mg/kg was at Mt. Zao. It was suggested that the reason for the high Se level in these two areas was because they are volcanoes and therefore generate sulfur compounds. However, soils from many other volcanoes, such as Mt. Unzen and Mt. Tokachidake and ash from Mt. Pinatubo in Philippines, have low Se levels.[9] This was also confirmed by the finding of Se deficiency at volcanoes in New Zealand.[10] In addition to soil analyses, we measured the Se level in some *Astragalus* plants in Japan,[9] and did not find high Se levels in vetches, which are well known to be Se accumulators in North America.[1] We also measured the Se level in some grasses growing in high Se level areas of Murodo or Mt. Zao, and did not find any Se accumulator plants.[9] We found high Se levels near the Besshi copper mine and therefore report the Se levels in soils and rocks from the areas around the two major copper mines in Japan, Asio and Besshi. We measured the Se content in some pyrites produced in Japan, because Berzelius found Se in pyrite from his mine.

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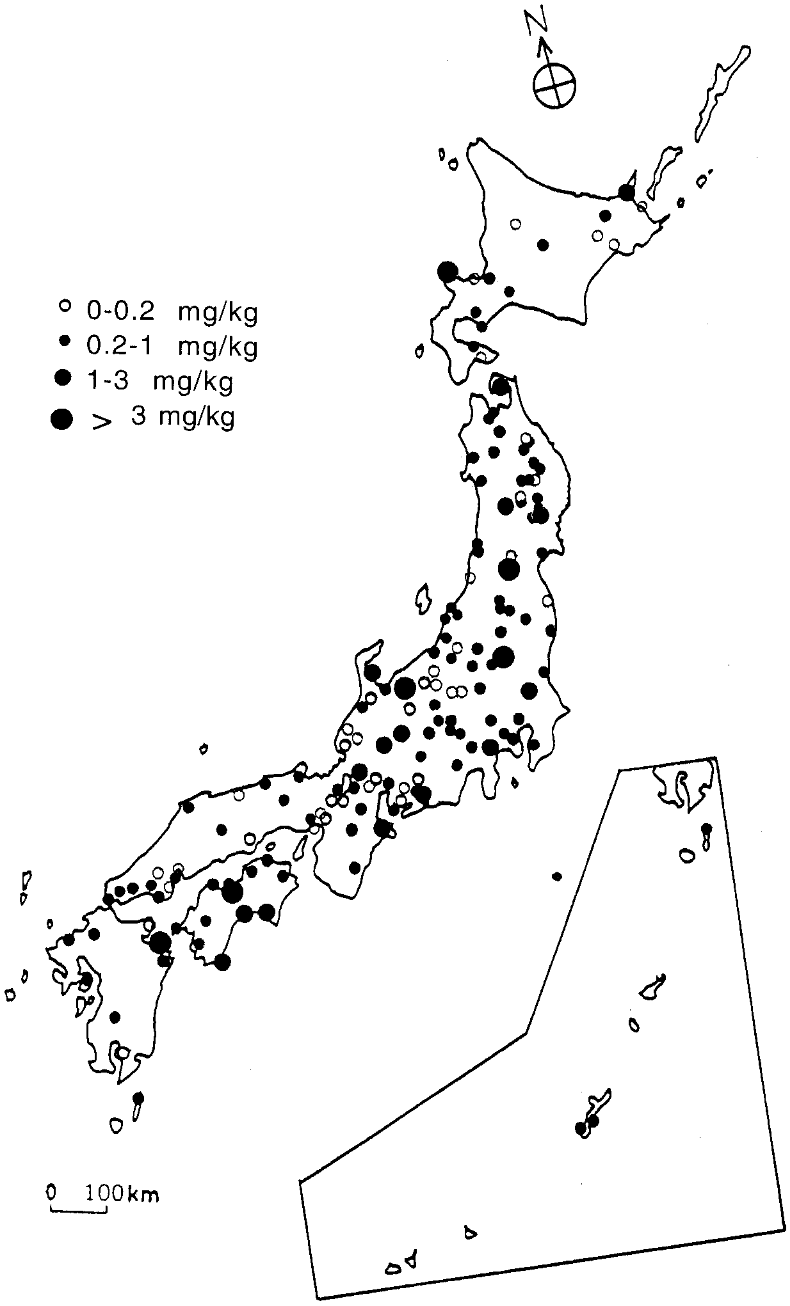

Fig. 1. Japanese Selenium Map of Surface Soil
The inset shows the southern Ryukyu Islands of Japan. Se levels are indicated in the figure by circles.

MATERIALS AND METHODS

Analyses of Se were carried out according to the previous methods,[9] as follows. Samples of the surface soil (at a depth of 0–20 cm) were collected at the points indicated in Fig. 1. We used original soil, not cultivated soil. Visible organic materials were removed from the samples and fine grains less than 0.1 mm^3 (rock samples were grained and used) were used for the measurements. The soil was dried for 2 days at 50°C. The amount of Se in each 5-mg dried sample was measured in triplicate by fluorometry with diaminonaphthalene (DAN, a product of Aldrich).[11] Mixed acids composed of concentrated sulfuric acid 30 μl and 60% perchloric acid 20 μl were added to each sample, which was then heated at 200°C for 30 min. Then, 6M HCl 0.25 ml was added, and the solution was allowed to stand for 30 min at 100°C to reduce Se (VI) to Se (IV). Fifty percent formic acid (0.25 ml) and stabilizing solution (0.5 ml) containing EDTA 4.7 mg and hydroxylamine–HCl 12.5 mg were added, and the mixture was adjusted to about pH 2 with 7 M ammonia 0.5 ml. DAN solution 0.25 ml (0.5 g/l of 0.1 M HCl) was washed twice with cyclohexane and then added to the mixture and heated at 50°C for 20 min. After heating, cyclohexane 1 ml was added and mixed. The cyclohexane layer was measured at 360 nm (excitation wavelength) and at 520 nm (emission wavelength). The amount of Se was calculated from the mean value of three determinations using a standard curve based on authentic Se. The standard inorganic Se used was a product of Merck for atomic absorption spectroscopy. To normalize the Se levels in soil samples, we also measured the Se level in a standard soil sample (JLK-1 Lake sediment, supplied by the Geological Survey of Japan). Some pyrite samples were generous gifts from Dr. Haruyuki Yutaka of the Geological Survey of Japan, Yatabe, Ibaragi, Japan.

RESULTS AND DISCUSSION

After the previous study,[9] in which we reported 70 sampling points in constructing a Se map of Japan, we accumulated an additional localities 80 (all 150 sampling points and Se levels at each point are shown in Table 1) and those findings are plotted in Fig. 1. The present map contains some sampling points in the Tohoku (northeast) region of Japan and the San-In area. In the previous map, we showed that the highest Se levels were at Murodo Jigokudani of Mt. Tateyama, Mt. Zao, and the area near the Besshi copper mine (the level of Murodo Jigokudani was 148 mg/kg). We did not find plants containing high Se levels in the Murodo Jigokudani area.[9] In the present study, we did not find high levels of Se but only normal levels between 0 to 1 mg/kg. Many localities in the northeast region except for Mt. Zao had normal Se levels, as shown in Fig. 1. Areas with slightly elevated Se content were from Mt. Tateyama to the Mino district of Gifu prefecture, and the south area of Shikoku island. This Se map of Japan may play a role in providing background values if Se pollution occurs.

In the previous study, we found high Se content in soil obtained near the region of the Besshi copper mine. Therefore we measured the Se levels in areas near two major old copper mines, Asio and Besshi, as shown in Tables 2 and 3, respectively. These two areas were famous for sulfur pollution when actively producing copper. Table 2 shows the Se levels in soil and rocks obtained from the Asio copper mine, such as copper stones and soil in caves. Rocks from the copper mine had high Se levels of 4.5 mg/kg and one copper ore sample contained 2.2 mg/kg. However, soil samples contained average Se levels. Previously, this area was polluted by sulfur compounds when copper was produced, but rocks that produced copper did not contain high Se levels, as selenite and/or elemental Se. Analyses of the Se level of Asio showed higher Se content in the samples obtained from the Nikko area, suggesting that this was due to the proximity of Asio copper mine. However, the Se levels at the Asio copper mine were low. These findings clarified that there is no Se pollution in the Asio area.

Table 3 shows Se levels in samples obtained from the area of the Besshi copper mine, which is no longer in production and has been converted into the Besshi Minetopea Museum. We obtained some samples from Minetopia and measured the Se levels. In contrast to the findings in shown Table 2 from the Asio copper mine, some soil obtained from the area of the Besshi mine contained high levels of Se from 1.0–4.5 mg/kg, as well as high levels in copper rocks (8.7 mg/kg). This suggested that the area near the Besshi copper mine may be slightly polluted by Se as well as sulfur compounds.

In 1817, Berzelius found Se as red precipitates at the bottom of a lead chamber used in the process to produce sulfuric acid from sulfite gas by burning pyrite from his mine. Pure pyrite is FeS_2, but it is possible that impure pyrite contains some Se. Therefore we measured the Se content in pyrite from Japan, generously supplied by the Geological Survey of Japan, as shown in Table 4. The maximum Se level in Table 4 is 330 mg/kg in pyrite obtained from Goto Island, Nagasaki prefecture. With the exception of pyrite samples from Hitachi and Chichibu, the Se levels in other pyrite samples were some ten mg/kg. Table 4 also shows the findings copper rock from Ikuno in Hyogo prefecture and Kiwa-cho in Mie prefecture, Japan.

Keshan, from which the name of the Se deficiency syndrome Keshan disease came, is located in the northeast area of China (the eastern terminal of the Eurasian continent).[12] This disease occurs due to the low Se levels in the soil and crops. The Japanese islands separated from the Eurasian continent in the ancient past. Therefore it is possible that soil from Japan has low Se levels. However, as shown in this study, Se levels in Japan are generally nor-

Table 1. Sampling Points in Figure 1 and Se Levels

Point	Se (mg/kg dry weight)	Point	Se (mg/kg dry weight)
Sharigun Kiyosatocho, Hokkaido pref.	0.33	Inawasiroko, Fukushima pref.	0.59
Siretoko-toge, Hokkaido pref.	1.24	Tsukuba City Hachimandai, Ibaragi pref.	1.13
Shari-jinja, Hokkaido pref.	0.25	Katsuta Station, Ibaragi pref.	0.42
Nemuro Notsukemisaki, Hokkaido pref.	0.16	Nikko Nishisando, Tochigi pref.	1.52
Akanko, Hokkaido pref.	0.17	Nikko Honden, Tochigi pref.	0.81
Kushiro-sitsugen, Hokkaido pref.	0.17	Nikko Okusha, Tochigi pref.	4.90
Tokachidake, Hokkaido pref.	0.36	Chuzenjiko Hutarasan-Chuguji, Tochigi pref.	0.82
Asahikawa City, Hokkaido pref.	0.15	Kegonnotaki, Tochigi pref.	0.52
Otaru City, Hokkaido pref.	0.05	Tsudo Station, Tochigi pref.	0.66
Sapporo City Hokudai, Hokkaido pref.	0.87	Osegahara-Yamanohana, Gunma pref.	0.29
Sapporo Hitsujigaoka, Hokkaido pref.	0.27	Takasaki Castle, Gunma pref.	0.22
Chitose Air Port, Hokkaido pref.	0.31	Mizugami City Tone River, Gunma pref.	0.19
Muroran City Hachimangu, Hokkaido pref.	0.30	Kisarazu City Academia Park, Chiba pref.	0.25
Touyako Nakajima, Hokkaido pref.	0.39	Urawa City Otakubo, Saitama pref.	0.93
Hakodate Goryoukaku Hokkaido pref.	0.34	Bunkyoku Hongo Sanshiroike, Tokyo pref.	0.74
Shakotan-misaki Hokkaido pref.	5.29	Akasaka Hie-jinja, Tokyo pref.	0.61
Oirase, Aomori pref.	0.75	Yushima Seido, Tokyo pref.	0.69
Aomori City Uto-jinja, Aomori pref.	0.56	Machida City, Tokyo pref.	0.43
Towadako Towada-jinja, Aomori pref.	0.84	Kiyose City, Tokyo pref.	0.69
Sannaimaruyama, Aomori pref.	0.26	Tokyo University Komaba Campus, Tokyo pref.	1.06
Chusonji Konjikido, Iwate pref.	1.25	Shibuy-ku Saruraku-iseki, Tokyo pref.	1.93
Morioka Castle, Iwate pref.	0.09	Yokohama Aoba Park, Kanagawa pref.	0.53
Yudamachi Kinshuko, Iwate pref.	0.37	Kamakura Kenchoji, Kanagawa pref.	0.74
Sangi-jinja, Iwate pref.	0.84	Hakonemachi Owakudani Red Soil, Kanagawa pref.	1.62
Sawauchimura Gingakougen, Iwate pref.	0.14	Odawara Castle, Kanagawa pref.	0.36
Mizusawa Kumano-jinja, Iwate pref.	0.45	Nakakomagun Showamachi, Yamanashi pref.	0.50
Iwatemachi Numamiya-jinja, Iwate pref.	0.68	Obuchizawa Station, Yamanashi Pref.	0.61
Ichinoseki Genbikei, Iwate pref.	0.25	Minamikomagun Shimobe-onsen, Yamanashi pref.	0.46
Kitakami Yasuragi, Iwate pref.	0.94	Nagano City Zenkoji Park, Nagano pref.	0.14
Ninohe Kindaichi-onsen, Iwate pref.	0.16	Iiyama Station, Nagano pref.	0
Ninohe Yakushido, Iwate pref.	0.29	Karuizawa Station, Nagano pref.	0
Kitakamidanso, Iwate pref.	0.04	Komoro City Kaikoen, Nagano pref.	0.14
Jouhojimachi, Iwate pref.	0.31	Hakuba Happo, Nagano pref.	0.14
Tamayamamura Himegamiyama, Iwate pref.	0.60	Shinano Omachi, Nagano pref.	0.15
Tazawako Sengantoge, Iwate pref.	0.49	Yatsugatake Tsuboniwa, Nagano pref.	0.08
Zaou Okama, Miyagi pref.	10.0	Yatsugatake Amigasadake, Nagano pref.	2.42
Zaou Kattadake, Miyagi pref.	22.3	Kurumayama Kogen, Nagano pref.	0.26
Matsushima, Miyagi pref.	1.76	Shirakabako, Nagano pref.	0.33
Shiogama City Susonodacho, Miyagi pref.	0.42	Kisohukushima Station, Nagano pref.	0.27
Higashinosiro City, Akita pref.	0.72	Kisokomagatake Senjojiki, Nagano pref.	0.27
Akita City Senshu Park, Akita pref.	0.34	Kiso-Ontake, Nagano pref.	0.81
Odate Station, Akita pref.	0.29	Niigata City, Niigata pref.	0.59
Yokote City Yokotekoen, Akita pref.	1.51	Murakami Station, Niigata pref.	0.19
Kawanishicho, Yamagata pref.	0.59	Kitakanbaragun Yasudamachi, Niigata pref.	0.87
Yamadera, Yamagata pref.	0.20	Santogun Koshijimachi, Niigata pref.	0.24
Sakata City Yagumo-jinja, Yamagata pref.	0.26	Teradomari, Niigata pref.	0.24
Tsuruoka City, Yamagata pref.	0.51	Yuzawa Naeba, Niigata pref.	0.48
Aizukogen, Fukushima pref.	0.27	Tsunan, Niigata pref.	0.14
Iwaki City Taira Castle, Fukushima pref.	0.41	Nakakubikigun Ogatacho, Niigata pref.	0.31
Haranomachi City, Fukushima pref.	0.19	Toyama Station, Toyama pref.	0.33
Aizuwakamatsu Tsurugajo, Fukushima pref.	0.31	Oyabe City, Toyama pref.	0.08
Koriyama City Atago-jinja, Fukushima pref.	0.40	Tateyama Station, Toyama pref.	0.93
Kitagata Station, Fukushima pref.	0.27	Kanazawa Station, Ishikawa pref.	0.22

Table 1. Continued

Point	Se (mg/kg dry weight)	Point	Se (mg/kg dry weight)
Wajima City Sosogi Kaigan, Ishikawa pref.	0.08	Ikuno Pyrite, Hyogo pref.	0.31
Hakui City Kitataisha, Ishikawa pref.	1.01	Sonikogen, Nara pref.	0.27
Wakura-onsen, Ishikawa pref.	1.78	Todaiji, Nara pref.	0.37
Oyama-jinja, Ishikawa pref.	1.00	Horyuji, Nara pref.	0.30
Sakaicho, Fukui pref.	0.18	Asukamura Ishibutai, Nara pref.	0.22
Maruoka Castle, Fukui pref.	0.23	Nachitaisha, Wakayama pref.	0.22
Eiheiji, Fukui pref.	0.04	Sihgu City Kumano-Hayatama-jinja, Wakayama pref.	0.49
Awara-onsen, Fukui pref.	0.24	Kurayoshi Imazaike, Tottori pref.	0.15
Fukui City Chuo Park, Fukui pref.	0.07	Tottori City, Tottori pref.	0.38
Nenohara-kougen, Gifu pref.	0.79	Izumotaisha, Shimane pref.	0.58
Gujohachiman Atago-jinja, Gifu pref.	1.75	Muikamachi Churengawa, Shimane pref.	0.10
Neodani, Gifu pref.	2.11	Okayama City Ohkubo, Okayama pref.	0.06
Nakatsugawa Kamienasan-jinja, Gifu pref.	0.46	Niimi City Senya, Okayama pref.	0.23
Ena City, Gifu pref.	0.08	Otake City Kamei Castle, Hiroshima pref.	0.34
Ibukiyama, Gifu pref.	1.15	Hiroshima City Hijiyama, Hiroshima pref.	0.10
Kamitakaramura Shinhotaka, Gifu pref.	0.18	Itsukushima-jinja, Hiroshima pref.	0.43
Gifu City Mitahora, Gifu pref.	0.45	Hikari City Kanbaien, Yamaguchi pref.	0.57
Kosakacho Nigorigo-onsen, Gifu pref.	1.78	Shimonoseki City Daigakucho, Yamaguchi pref.	0.40
Fujisan, Shizuoka pref.	0.26	Yamaguchi City Yoshida, Yamaguchi pref.	0.74
Hamanako Tsuzuki-jinja, Shizuoka pref.	1.11	Mine City Atsuho, Yamaguchi pref.	0.97
Hamanako Kosaicho, Shizuoka pref.	1.22	Tonogun Kanocho, Yamaguchi pref.	0.24
Nagoya City University, Aichi pref.	0.31	Kugagun Wagicho, Yamaguchi pref.	0.15
Kasugai City, Aichi pref.	0.11	Tokushima City Kawauchimachi, Tokushima pref.	0.31
Amagun Sayacho, Aichi pref.	0.12	Takamatsu City Ritsurinkoen, Kagawa pref.	0.53
Toyota City Takatori-jinja, Aichi pref.	0.18	Kotohira-jingu, Kagawa pref.	0.37
Kitasidaragun Tomiyamamura, Aichi pref.	0.11	Yashima, Kagawa pref.	0.45
Gamagori City, Aichi pref.	0.19	Niihama Station, Ehime pref.	0.89
Sidaragun Horaicho, Aichi pref.	0.26	Satamisaki Misaki, Ehime pref.	0.34
Chitahanto Toyohama, Aichi pref.	0.80	Uchiko, Ehime pref.	0.28
Fujiwaradake, Mie pref.	0	Uwajima City, Ehime pref.	0.25
Suzukasanmyaku Buheitoge, Mie pref.	0.25	Kochi City Keraotsu, Kochi pref.	1.03
Isejingu Gegu, Mie pref.	1.24	Tosashimizu City, Kochi pref.	1.09
Seta River, Shiga pref.	0.11	Akigun, Kochi pref.	1.66
Nishi-otsu, Shiga pref.	0.35	Dazaihu Tohuro, Fukuoka pref.	0.48
Tagacho, Shiga pref.	0.55	Higashimatsuuragun Hamatamacho, Saga pref.	0.66
Eigenji, Shiga pref.	0.21	Unzen-onsen, Nagasaki pref.	0.12
Kiyomidzudera, Kyoto pref.	0.19	Saganoseki, Ooita pref.	4.20
Yosagun Mineyamacho, Kyoto pref.	0.50	Toana, Ooita pref.	0.41
Kyoto Hushimiku Hujinomori-jinja, Kyoto pref.	0.15	Kunisakihanto, Ooita pref.	0.18
Hieizan, Kyoto pref.	0.46	Sakurajima, Kagoshima pref.	0.18
Hirakata City, Osaka pref.	0.31	Tanegashima Nishiomote City, Kagoshima pref.	0.65
Asahi-ku Yodogawa-kasenshiki, Osaka pref.	0.04	Hishikari-kozan, Kagoshima pref.	0.70
Senri, Osaka pref.	0.16	Gushikami, Okinawa pref.	0.21
Rokkosan Arashino-oka, Hyogo pref.	0.28	Urazae City, Okinawa pref.	0.32
Arima-onsen, Hyogo pref.	0.90	Ginowan City, Okinawa pref.	0.52
Kobe Port-island, Hyogo pref.	0.31	Yomitan, Okinawa pref.	0.56

mal except for a few certain areas, such as Murodo Jigokudani of Mt. Tateyama and some pyrite rocks. One study reported acute Se toxicity after swallowing gun blue, a lubricant solution.[13] The symptoms were the sudden onset of profuse, watery diarrhea and vomiting.[13] We measured Se levels in soil and rocks by fluorometry using DAN as described in Materials and Methods. The Se compounds in the

Table 2. Se Content in the Soil and Rocks from the Area near the Asio Copper Mine

Sampling point	Se (mg/kg dry weight)
Oziri Beach of Chuzenji Lake, Tochigi pref.	0.68
Chuguji of Chuzenji Lake, Tochigi pref.	0.82
Kegon Falls, Tochigi pref.	0.52
Tudou Station, Tochigi pref.	0.66
Asio copper rock 1, Tochigi pref.	0.26
Asio copper rock 2, Tochigi pref.	4.50
Wall of copper mine, Tochigi pref.	2.21
Iron rust in copper mine caves, Tochigi pref.	0.25
Natural $CuSO_4$ crystals in caves, Tochigi pref.	0.51
Asio Pottery, Tochigi pref.	0.79

Table 3. Se Content in the Soil from the Area near the Besshi Copper Mine

Sampling point	Se (mg/kg dry weight)
Bus stop at Besshi Minetopia, Ehime pref.	2.16
Entrance of Besshi Minetopia, Ehime pref.	4.54
Soil at exit of cave, Besshi Minetopia, Ehime pref.	1.02
Rock at exit of cave, Besshi Minetopia, Ehime pref.	0.24
Rock as a souvenir of Besshi Minetopia, Ehime pref.	8.69
Soil from a gutter, Besshi Minetopia, Ehime pref.	0.37
Niihama Station, Ehime pref.	0.89
Mitsuzaki of Sata Cape, Ehime pref.	0.34

Table 4. Se Content in Pyrite and Copper Rocks from Japan

Sampling point	Se (mg/kg dry weight)
Pyrites	
Kamikita mine, Aomori pref.	22.4
Osarizawa mine, Akita pref.	31.7
Sayama mine, Akita pref.	28.5
Awanoyu mine, Yamagata pref.	74.3
Hitachi mine, Ibaraki pref.	6.4
Chichibu mine, Saitama pref.	5.6
Konuki mine, Niigata pref.	17.7
Harimichi mine, Nara pref.	24.0
Kanbe mine, Nara pref.	19.4
Goto mine, Nagasaki pref.	330
Peru (from the Museum of the University of Wisconsin)	61.4
Copper rocks	
Besshi	8.69
Asio	4.50
Ikuno, Hyogo pref.	0.31
Kiwa-cho, Mie pref.	47.6

soil and rocks are hydrolyzed in mixed acids. However, insoluble Se compounds in soil have previously been reported.[14] Therefore it is possible that insoluble Se in mixed acids was not measured and the levels reported for some areas may be lower than the total Se value.

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