1.2 PIXE
What can be performed by means of PIXE at NMCC?

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Abstract

The Nationwide-common utilization for PIXE at NMCC has been carrying out since April 1993, and more than 100000 samples have been analyzed up to the present. PIXE has been applied to many studies in various research fields. As a result, the samples analyzed have diversified shapes, characters and compositions. In order to cope with these diversified samples, we have been continuously developing the methods of measurement, data analysis, sample preparation and quantitative analysis. Now, one of our catchphrases is “We’ll perform quantitative analysis of any kind of samples”. Especially, the standard-free method for quantitative analysis made it possible to quantitatively analyses infinitesimal samples, powdered samples and untreated bio samples which could not be well analyzed quantitatively in the past. It also allows us to carry out quantitative analysis of untreated samples, and it saves labor and time for procedure in target preparation. Moreover, “The powdered-internal-standard” is working miracle in analyses of high-Z-matrix-powdered samples such as soil, rock, ash, dust and aerosol.

The recent topics are as follows. In in-air PIXE, a physical method of quantitative analysis was established and the samples which has been difficult to be analyzed in vacuum can be quantitatively analyzed in in-air PIXE. Moreover, standard-free methods for various samples have been developed in in-air PIXE, and it becomes possible to quantitatively analyze the samples such as a drop of oil and living plants which have been difficult to be quantitatively analyzed by traditional methods. In in-vacuum PIXE, the standard-free method for organs taken from patients and experimental animals has been established and it becomes possible to quantitatively analyze very small organ samples of less than 1 mg. These methods have already been applied to many practical studies.

Keywords: PIXE, Quantitative analysis, Standard-free, Internal standard, External standard, Living plant, Organ, Oil, Measuring system, In-air, Spectrum analysis, Small quantity, Clinical samples, Experimental animals, Environmental studies, Human exposure, Toxic elements, Powdered samples, High-Z, Untreated samples, Absorption curve, Efficiency
Characteristic features of ores and mineral assemblage of Silurian-Devonian volcanogenic massive sulfide deposits in South and Middle Urals, Russia

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Abstract

The volcanogenic massive sulfide (VMS) deposits in South and Middle Urals are divided into Cyprus, Bessi, Baimak-, and Ural-types volcanogenic massive sulfide deposits. The aim of study is to clarify characteristic features of ores of the VMS deposits (Yaman-Kasy, Molodezhnoe, Dusa and Safyanovka deposits) in South and Middle Urals based on data of minerals assemblage and chemical compositions of ores. The chemical compositions of ores of Ural- and Baimak-type VMS deposits were characterized by high Fe-Cu and low Pb contents except Zn-Pb-rich ores of the Safyanovka deposit. The Yaman-Kasy, Molodezhnoe and Dusa volcanogenic massive sulfide deposits have similar characteristic, however the Safyanovka deposit is thought to be different type from the Yaman-Kasy, Molodezhnoe and Dusa deposits.
Geochemical characteristics of gold-bearing ores of Boliden deposit, Skellefte district, Sweden

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Abstract

The Au-bearing Boliden massive sulfide deposit located in the eastern part of the Skellefte district in northern Sweden was formed in a volcanic arc setting at 1.89-1.85 Ga and well known for its high gold grades (average 15 ppm). Chemical characteristics of ores from Boliden deposit were examined in this study based on the data of mineral assemblage and chemical compositions of ore determined by AAS, PIXE and ICP-MS. The ores of the Boliden deposit are divided into two types, arsenopyrite-rich ore consisting mainly of recrystallized euhedral to subhedral arsenopyrite, pyrite, electrum and rutile and sulfosalt-rich pyrite-chalcopyrite-sphalerite ores consisting of pyrite, pyrrhotite, sphalerite, chalcopryite, tetrahedrite, bournonite, laitakarite and native bismuth which cut the arsenopyrite ores. TiO₂ and Au contents of the ores show a positive correlation. Rutile is also associated with acidic alteration zone composed of quartz-andalusite-kaolinite of the Boliden deposit. Data of the mineral assemblage and chemical composition of ores suggest that the gold mineralization was caused by acidic hydrothermal activity.
1. Introduction

The Au-bearing Boliden massive sulfide deposit, which was formed in a volcanic arc setting at 1.89-1.85 Ga, is well known for its high gold grades (average 15 ppm) and is characterized by complex mineral assemblages of ore minerals. Earlier studies considered the massive sulfide deposits in the Skellefte district as metamorphosed equivalents of Kuroko-type ores (Rickard and Zweifel, 1975; Vivallo, 1987). In contrast, recent structural and geochemical investigations indicate that the Boliden deposit may be a high-sulfidation-type epithermal sub-seafloor deposit formed by infiltration and replacement (Allen et al., 1996; Weihed et al., 1996). Particle induced X-ray emission (PIXE) can be used to analyze minute samples smaller than a milligram, not only simultaneously detects elements from Na to U in a short time but also detects major-to-trace elements at the concentration level of ppm (Vaggellia et al., 2003; Saitoh et al., 2002). We examined chemical composition of ores from the Boliden deposit by PIXE and ICP-MS to know the chemical characteristics of the ores based on the relationship between Au concentration and other elements in the ores.

2 Outline of geology

The Boliden deposits are located in the eastern part of the Skellefte district in northern Sweden which has been mined for base and precious metals in the early 1920s (Allen et al., 1996) (Fig. 1). The deposit has mined 8.3 million tons of ore including 128 t of Au, 411 t of Ag, 566,000 t of As, 118,000 t of Cu, and 2.1 Mt of S. The massive sulfide orebodies of the Boliden deposit occur as a set of elongate lenses, obliquely cutting the host rocks structurally. The lenses trend east-west with a roughly vertical dip (Wagner and Jonsson, 2001). Alteration of the Boliden deposit can be classified into two types: sericite-kaolinite-andalusite zone and chlorite zone. The sericite-kaolinite-andalusite zone is surrounded by the chlorite zone. The orebody occurs in the sericite-kaolinite-andalusite alteration zone. The samples used in this study are of upper (Borehole 24) and middle (Borehole 64) positions of orebody.

3 Mineral assemblages of ores

The two different types of ore can be distinguished in the Boliden deposit: arsenopyrite-rich and
sulfosalt-rich pyrite-chalcopyrite-sphalerite ores. The occurrence mode of the sulfosalt-rich pyrite-chalcopyrite-sphalerite ores cutting arsenopyrite ores suggests that the arsenopyrite-rich ores were formed prior to the formation of the sulfosalt-rich ores (Figs. 2B and 2C). The arsenopyrite ores are generally fine-grained (Figs. 2A and 2B). The ores consist mainly of recrystallized euhedral to subhedral arsenopyrite, pyrite, electrum and rutile (Fig. 2A, Table1). The mineral assemblages of sulfosalts-rich ores consist of pyrite, pyrrhotite, sphalerite, chalcopyrite, tetrahedrite, bouronite, laitakarite and native bismuth (Fig. 2D). The gangue minerals are quartz, biotite, muscovite, andalusite, kaolinite, sericite, and chlorite.

![Fig.2. Photomicrographs in reflected light showing representative textural relationship for arsenopyrite-rich ores and sulfosalts-rich ores, Boliden deposit. A. Anhedral rutile crystal enclosing electrum in fine-grained and partially recrystallized arsenopyrite ore. B. Typical sulfosalt-rich vein consisting of chalcopyrite and pyrrhotite crosscutting fine-grained massive arsenopyrite containing numerous silicate and sulphide inclusions. C. Pyrite porphyroblasts coexisting with sphalerite and chalcopyrite in sulfosalts-rich vein. D. The intergrowth of tetrahedrite, bismuth and selenium-galena formed the veinlet occur in massive chalcopyrite.](image)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Type of ore</th>
<th>pyrrhotite</th>
<th>pyrite</th>
<th>Arsenopyrite</th>
<th>chalcopyrite</th>
<th>sphalerite</th>
<th>electrum</th>
<th>rutile</th>
<th>quartz</th>
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<tr>
<td>Boliden 24, 99.71-99.73 m</td>
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<td>+++</td>
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<td>++</td>
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<tr>
<td>Boliden 24, 138.00 m</td>
<td>py-asp-rich</td>
<td>+++</td>
<td>+</td>
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<td>+</td>
<td>+</td>
<td>+</td>
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<tr>
<td>Boliden 64, 14.07-14.09 m</td>
<td>asp-rich</td>
<td>+++</td>
<td>+</td>
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<td>+</td>
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<tr>
<td>Boliden 64, 21.80-21.83 m</td>
<td>py-cp-rich</td>
<td>+</td>
<td>+++</td>
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<td>++</td>
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<td>++</td>
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</table>

+++ abundant, ++ moderate, +a few
4 Methods

The rock and ore samples were prepared using PIXE developed by Sera et al., (1999), where the bulk sample was ground into fine powder in an agate mortar. Thin targets were prepared by taking about 0.1 mg of the powder and putting it on a 0.40 $\mu$m thick polypropylene film, dropping roughly 1$\mu$l of 10% collodion solution diluted with ethyl alcohol on it to fix the powder, and drying the film through a oven, then keeping the film into the polypropylene bottle. Chemical composition of the samples was determined by PIXE at Nishina Memorial Cyclotron Center (NMCC), Japan Radioisotope Association, using 2.9 MeV protons from a baby cyclotron. Beam currents, the accumulated charge and the typical measuring time were 20-40 nA, 11-19 $\mu$C, and 5-10 min, respectively. X-ray spectra were analyzed using the SAPIX program.

Quantitative data were obtained by the combination of data determined by PIXE and Cu and Zn contents of corresponding samples analyzed by AAS. The ore samples examined by PIXE are also analyzed by ICP-MS described by Satoh et al. (1999).

5 Results and Discussion

The analytical results by PIXE and ICP-MS are summarized in Table 2. Ores of the Boliden deposit consist mainly of arsenopyrite. Therefore, Cu, Pb and Zn contents of ores in the Boliden deposit are low and range from 446-19600 ppm, 429-1590 ppm, 28-27200 ppm, respectively. On the other hand, arsenic content of the ores is high for the arsenopyrite-rich ores (maximum content of 249000 ppm), TiO$_2$, Ag, Au and Bi contents are also high in the arsenopyrite-rich ores (4940 ppm TiO$_2$, 55.0 ppm Ag, 1730 ppm Sb, 36.4ppm Au and 762 ppm Bi). TiO$_2$ and Au measured by PIXE and ICP-MS shows a positive correlation (Fig. 3). The electrum containing Hg were observed in a rutile crystal in the arsenopyrite-rich ores. The mode of occurrence of electrum accords with the data of TiO$_2$ and Au contents of the ores. Rutile is also present in altered rocks composed of quartz, andalusite and kaolinite and is thought to be formed by recrystallization of the acidic altered rocks surrounding lenticular orebodies of the Boliden deposit. The positive correlation between TiO$_2$ and Au suggests that gold mineralization was caused by acidic hydrothermal activity.

<table>
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<tr>
<th>Sample</th>
<th>TiO$_2$</th>
<th>Mn</th>
<th>Co</th>
<th>Cu</th>
<th>Zn</th>
<th>Ga</th>
<th>As</th>
<th>Sr</th>
<th>Mo</th>
<th>Ag</th>
<th>Cd</th>
<th>In</th>
<th>Au</th>
<th>Pb</th>
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<td>129</td>
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<td>0.37</td>
<td>2132</td>
<td>4.77</td>
<td>171</td>
<td>14.70</td>
<td>0.31</td>
<td>429</td>
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<td>Boliden 24,</td>
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<td>184</td>
<td>820</td>
<td>3000</td>
<td>1</td>
<td>6920</td>
<td>1.75</td>
<td>0</td>
<td>8.51</td>
<td>13.5</td>
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<td>13.6</td>
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<td>138.00 m</td>
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<td>Boliden 64,</td>
<td>4940</td>
<td>18</td>
<td>480</td>
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<td>645</td>
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<td>Boliden 64,</td>
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TiO$_2$, Pb and As contents were determined by the combination PIXE and AAS, concentration of other elements were determined by ICP-MS. unit: ppm.
6 Conclusion

The geochemical characteristics of ores from the Boliden deposit examined on the basis of mineral assemblage and chemical compositions of the ores. The two different ore types can be distinguished in the Boliden deposit: arsenopyrite-rich and sulfosalt-rich ores. The arsenopyrite-rich ores consist mainly of recrystallized euhedral to subhedral arsenopyrite, pyrite, electrum and rutile. The mineral assemblage of sulfosalt ores consist of pyrite, pyrrhotite, sphalerite, chalcopyrite, tetrahedrite, bournonite, laitakarite and native bismuth. The positive correlation between TiO$_2$ and Au contents suggests that gold mineralization was caused by acidic hydrothermal activity.

References


Gold mineralization of Q prospect at Chatree deposit, central Thailand

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Abstract

The Chatree gold deposit is classified as a low sulfidation epithermal deposit. Gold mineralization at Q prospect occurs as veins and stockworks in andesitic volcanic-clastic and volcano-sedimentary rocks. Gold mineralization of the deposit is divided into four stages; Stage I: precious and base metal mineralization, microcrystalline quartz with minor dendritic pyrite, chalcopyrite, K-feldspar and calcite; Stage II: barren coarse-grained comb quartz; Stage III: barren medium-grained mosaic quartz with minor pyrite; Stage IV: precious and base metal mineralization (stockwork), fine to coarse-grained quartz showing flamboyant texture, K-feldspar, calcite, fine-grained pyrite and chalcopyrite. Stages I and IV are characterized by dendritic and flamboyant textures suggesting rapid crystal growth, higher gold, $K_2O$ and CaO contents. Stages I and IV are thought to be formed under highly supersaturated hydrothermal solutions caused by boiling based on the texture and mineral assemblage of ores.
1 Introduction

The Chatree deposit is located about 300km north of Bangkok (Fig. 1). The deposit is in the central part of the Loei-Phechabun-Nakhon Nayok Permo-Triassic volcanic belt (LPN). This belt extends from western Lao along the western edge of Khorat plateau to the Cambodian central plain (Bunopas and Vella, 1983, 1992; Charusiri et al., 2002). The Chatree gold deposit consists of several prospect areas. The deposit has an estimated total gold production of about 5 million ounces and 35 million ounces of silver, with an average ore grade 1.07 g/t of gold and 7.5 g/t of silver (personal communication). Gold occurs in quartz-carbonate-sulfide veins and stockworks in a sequence of volcanic rocks (Jame and Cumming, 2007; Salam et al., 2006).

The Hishikari epithermal Au-Ag deposit is located in Japan. It is one of the richest gold veins in the world. Gold reserves are estimated to be 150 metric tons at an average grade of 80 g/t with ore produced at a rate of 350~400 metric ton/day (Ibaraki et al., 1991). The veins in Hishikari deposit are mainly quartz, adularia and smectite showing banded structure. In comparison with Q prospect at the Chatree and the Hishikari deposit, the characteristics of width of veins, ore grade and mineral assemblage of Q prospect are different from those of typical epithermal quartz veins such as the Hishikari deposit.

The aim of this study is to clarify confirm that the gold mineralization of Q prospect is epithermal-type mineralization based on data of textures of quartz and chemical compositions of ores.

2 Outline of geology and ore deposit

Geology of the Chatree deposit is composed of oceanic and continental arc complexes that developed before and during the suturing of the Shan-Thai and Indochina Terranes (Charusiri et al., 2002). The geology of LPN volcanic belt including of the Chatree deposit is composed of conglomerate, sandstone, shale and limestone dominate the district-scale geology of thick Carboniferous to Early Permian.
sedimentary sequences. The Chatree deposit occurs in coherent andesitic to rhyolitic lavas, which is overlain by fine-grained volcanioclastics, epiclastic siltstone and mudstone (Fig. 2) (Corbett, 2006; Cumming et al., 2008). Alteration in the regional scale is associated with volcanic centers and major NE-SW fault systems (Corbett, 2006; Crossing, 2006). Alteration of southern part of the Chatree deposit is propylitic alteration (chlorite±epidote-calcite-pyrite), while alteration of northern part of the deposit is argillic and phyllic alteration (sericite-illite-quartz-pyrite) (Cumming, 2006).

The radiometric age of gold mineralization of the Chatree deposit is estimated to be 250.9 ± 0.8 Ma. The age is close to the emplacement of the andesitic volcanics (Early Triassic) (Zaw et al., 2007; Salam et al., 2007 and 2008). Granodiorite intruded in the southern part of the Chatree deposit. An age of granodiorite is estimated to be 244 Ma suggesting the age of post mineralization in the area (Zaw et al., 2007; Salam et al, 2007 and 2008). Dikes are cutting all of rock units.

3 Vein characteristic and Petrography

The area studied in Q prospect is one of the major prospects of the Chatree deposit (Fig. 2). There are many veins at Q prospect. The veins are orientated along NW-SE, NE-SW and N-S direction with moderate to steeply dipping. Veins mineralized also occur between strata consisting of volcanic-sedimentary and andesitic volcanic-clastic rocks. Gold occurs in veins and stockworks with minor amount breccia of the andesite volcanic-clastics and volcanic-sedimentary rocks. Gold-bearing veins are composed of mainly quartz, adularia, chlorite, carbonate and sulfide minerals.

Mineralization stages at Q prospect were examined on the basis of cross-cutting relationships of veins (Fig. 3). The stages are divided into four stages:

Fig. 2 Geologic map of the Chatree deposit (partly modified from Salam et al., 2004).
Stage I: light-dark grey quartz veins, veinlets and breccias containing precious and base metal mineralization. The constituent minerals are a large amount of microcrystalline quartz with small amounts of pyrite (dendritic pyrite), chalcopyrite, K-feldspar and calcite.

Stage II: a large amount of barren coarse-grained comb quartz with a small amount of coarse-grained pyrite.

Stage III: a large amount of barren medium-grained mosaic quartz with small amounts of pyrite and chlorite.

Stage IV: Ores of stage IV shows network structure composed of mainly quartz and calcite. The stage IV is characterized by a precious and base metal mineralization. The constituent minerals are fine to coarse-grained quartz with small amounts of K-feldspar and calcite. Clasts of stages I (dark grey quartz), II and III are included in network as breccias.

Fig. 3 Paragenetic sequence ore and gangue minerals of Q prospect at the Chatree deposit.

<table>
<thead>
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<th>II</th>
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<td>Chalcopyrite</td>
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<tr>
<td>Tetrahedrite</td>
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<td>Sphalerite</td>
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<th>Gangue minerals</th>
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<td>K-feldspar</td>
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<td>Illite</td>
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Fig. 4 Photomicrographs (crossed nicols) showing quartz texture from stages I to IV. a: Microcrystalline quartz in stage I. b: Comb quartz (stage II) cutting fine-grained quartz (stage I). c: Mosaic texture of medium-grained quartz. d: Flamboyant texture of quartz in stage IV.
Gain size of quartz from stages I to III changes from fine-grained to coarse-grained according to progress of mineralization stages (Fig. 4). Stage I is also composed of a large amount of fine-grained pyrite. Quartz in stages II and III is medium to coarse-grained quartz crystals. Stage IV consists of quartz fine-grained to coarse-grained with large amount of pyrite, chalcopyrite and sphalerite. Electrum is abundant in aggregates of fine-grained quartz and inclusion in pyrite in stages I and IV.

4 Geochemistry

Twenty-six samples were analyzed by PIXE at NMCC and ICP-MS at Akita University to clarify geochemical characteristics of gold mineralization of Q prospect. The chemical contents of K$_2$O, CaO, Au, Cu and Zn components are shown in Table 1. The variations of Au, K$_2$O and CaO contents accord with the variations of abundance of electrum (Au, Ag), adularia (KAl$_3$Si$_3$O$_8$) and calcite (CaCO$_3$) (Fig 5). The fact that the positive correlated among Au, K$_2$O and CaO contents suggests that electrum precipitated with adularia and calcite. Fine-grained quartz in stages I and IV also has high Au content. Some samples in Stage I contain electrum and dendritic pyrite. The mineral assemblage and texture suggest precipitation of ore minerals from supersaturated hydrothermal solution. Based on the presence of brecciated structure and flamboyant texture in quartz network veins of stage IV and the possibility that the flamboyant texture was formed by boiling of hydrothermal solution (Dong et al., 1995), precipitation of gold in quartz veins of Q prospect thought to be caused by boiling. These characteristics are similar to those of typical epithermal gold-bearing quartz veins such as the Hishikari deposit. The gold mineralization of Q prospect is thought to be one of style of epithermal gold mineralization.

<table>
<thead>
<tr>
<th>ppm</th>
<th>Vg-1eu</th>
<th>Vg-1ed</th>
<th>Vg-1Lg</th>
<th>Vg-1IIw</th>
<th>Qgs-3II</th>
<th>Q-III</th>
<th>Hbx-2IV</th>
<th>Hbx-3IV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au*</td>
<td>1.0</td>
<td>1.9</td>
<td>2.4</td>
<td>1.9</td>
<td>0.6</td>
<td>0.5</td>
<td>0.6</td>
<td>1.5</td>
</tr>
<tr>
<td>Cu*</td>
<td>35.6</td>
<td>28.9</td>
<td>28.6</td>
<td>11.9</td>
<td>11.0</td>
<td>20.4</td>
<td>6.9</td>
<td>23.7</td>
</tr>
<tr>
<td>Zn*</td>
<td>11.1</td>
<td>23.8</td>
<td>190.9</td>
<td>68.9</td>
<td>29.1</td>
<td>15.4</td>
<td>11.2</td>
<td>30.0</td>
</tr>
<tr>
<td>K$_2$O*</td>
<td>693C</td>
<td>4380</td>
<td>19100</td>
<td>6020</td>
<td>474</td>
<td>74.0</td>
<td>163</td>
<td>1630C</td>
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<tr>
<td>CaO**</td>
<td>271C</td>
<td>168</td>
<td>11500</td>
<td>16000</td>
<td>2302</td>
<td>204</td>
<td>916</td>
<td>4830C</td>
</tr>
</tbody>
</table>

*: ICP-MS  **: PIXE

Table 1 Chemical compositions of gold-bearing quartz from stages I to IV by PIXE and ICP-MS analyses.

![Fig. 5 Diagram showing variations of K$_2$O, CaO, Cu, Zn and Au contents from stages I to IV.](image)
5 Conclusions

Gold-bearing quartz veins in Q prospect of the Chatree deposit are divided into four stages. The gold-bearing quartz veins are composed mainly of quartz, adularia, chlorite, calcite and sulfide minerals. The grain size of quartz changes from fine to coarse-grained crystals according to progress of mineralization stages. Stages I and IV are composed of mainly fine-grained quartz with a large amount of fine-grained pyrite. In the Q prospect, quartz-adularia-calcite may be precipitated with gold in the stage I and IV. K_2O and CaO contents of gold-bearing veins also show a weak positive correlation with Au, Cu, Zn in stages I and IV. Gold mineralization of Q prospect at the Chatree deposit is one of epithermal-type mineralization and the gold mineralization could be caused by boiling.

References


Geochemical characteristics of concentrations of major and minor elements of main four rivers flowing through Akita plain

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Abstract

The purpose of this study was to clarify the characteristics of chemical compositions of river water of Kamishinjyou, Asahi, Taihei and Iwami Rivers flowing through Akita Plain. Concentrations of major and minor elements and suspended materials with particle size over 0.45 µm in the water were measured using ion chromatography and the PIXE method. Major chemical concentrations increase by about two or three times from the upstream region to the Sea of Japan coast in each river. These increases are thought to be caused by sea salt. As for minor components, Si, Fe, Mn and Zn were detected, and Si and Fe concentrations were high in the downstream regions of Kamishinjyou and Taihei Rivers and low in Asahi River. In the main suspended materials with particle size over 0.45 µm, concentrations of Si were high in all three rivers and the concentrations of Al was high in the downstream region of Taihei River. The concentration was high in areas of siltstone and sandstone in the downstream region of kamishinjyou and Taihei Rivers. In Asahi River, there is a possibility that groundwater sprang up in the upstream region.
Comparison of data analyzed by PIXE with data by XRF in chemical composition on soil samples collected in Mt.Iwaki

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Abstract

Chemical contents of soil samples were analyzed by Particle Induced X-ray Emission analysis (PIXE) and X-ray fluorescence spectrometry (XRF). A comparison of data by PIXE with by XRF is discussed in composition for soil samples collected on the soil surface at the bottom of the old volcanic crater in Mt. Iwaki, Japan. Between these two methods, serious differences were observed in analytical concentrations for Al, Si, Na and Mg. Because the 100% of total content in oxides may prove analytical accuracy for each samples, PIXE data for these elements were corrected with standard sample, NIES pond sediment.
Influence of nutrient materials on the absorption of various minerals

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Abstract

[Introduction] Although percutaneous endoscopic gastrostomy (PEG) feeding is widely used as a convenient method for long-term nutritional support, administration of liquid nutrients is often accompanied by complications such as vomiting or diarrhea. Vomit caused by gastro-esophageal reflux (GER) may cause critical conditions such as aspiration pneumonia. To avoid this complication, semi-solidified enteral nutrition formulations are used in hospital- and home-based care. However, the influence of nutrient materials on the absorption of various minerals has not been fully understood.

[Samples and Methods] In this study, we subjected the patients, who were replaced with PEG, in the following three groups. Namely, the liquid nutrient materials administrated group (17 cases), the low viscosity semi-solidification nutrient materials administrated group (17 cases), and the high viscosity semi-solidity nutritional supplement administrated group (18 cases). Blood samples were collected from the patients on the day of PEG and one and two weeks after PEG. We measured the contents of Fe, Cu, Zn, Se, Ca, Mn and Mg in these sera by a PIXE method and analyzed the changes of these minerals by repeated measures ANOVA.
[Results] The results demonstrated that serum Fe concentration in the liquid group was increased one week after PEG but those in other two groups were decreased. Significant difference in serum Fe concentration was found in the different kinds of nutrient materials (F=2.956, p <0.05). Serum Cu concentration was significantly deceased with time, but the difference between three groups was not recognized (F=5.146, p <0.01). The Zn concentration in the liquid group trended to be increased, but that in the high viscosity group was decreased, although they were not statistically significant. We did not observe the significant changes in the other elements in this study.
PIXE analysis of mothers’ and infants’ hairs collected at medical checkups held in Fukuoka city

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Abstract

This is the third report of our project to examine relationships between atopic dermatitis and hair minerals of infants and mothers. The hair minerals are measured using the proton-induced x-ray emission (PIXE) method. The second report published last year described the characteristics the hair minerals of mothers and children and some differences of them between one month and ten months after birth. This study examined the accuracy of the PIXE measurements by statistically analyzing two independent measurements obtained from different hairs of the same person.

The project is expected to proceed as follows:
1) The results of the PIXE measurements will be reported to the participation doctors.
2) Association analysis of the amount of minerals, dining habit, and clinical performance.
3) Statistical analysis for causal relationships between mineral deficiency/excess and atopy/allergy conditions.
4) Redefine the participating mother/child population as a new cohort to study the effects of environment, dining and minerals on their health conditions.
5) Cooperation with the “Birth cohort project” by the Ministry of Environment.
6) Cooperation of the birth cohort study by Norway
An attempt for evaluation of air dusts by PIXE analysis of

*Parmotrema tinctorum*

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Abstract

Dust pollution in the air has been examined along the way from Futakuchi valley through Akiu hot spring to metropolitan Sendai. We used elemental abundances in *Parmotrema tinctorum* as pollution indicators. In order to determine the elemental abundances in the lichen we used PIXE analysis.

Each elemental group shows respective special variations: most considerable fluctuations have been recognized for alkaline elements. As for alkaline earth elements, they also fluctuate much, but not so much as the alkaline elements. Transition metals show special variation to similar extents as the alkaline earth elements. Non-metallic elements and light metal shows respective variation along the sampling route. Although silicon is one of the dominant elements as an elemental pollution, calcium, iron, magnesium and potassium abundances in *Parmotrema tinctorum* are much more than silicon abundance.

Assuming that the elemental abundances can be applicable to estimate dust pollution in the air, natural dust and agricultural dust as well as industrial dust can be classified. Sulfide mineral pollutants do not play an important role in the air pollution of the investigated area. Among three dust group, we found significant positive co-variation between sulfur and phosphorus: this might have derived from agricultural fertilizer. Iron and manganese co-vary within small abundances of manganese, but the reason of this co-variation is not clear. Abundance relations among many elements imply that the major dust might have derived from natural sand, natural soil and natural rock powders. None of them displays some kinds of elemental co-variation. It should be emphasized that a remarkably good co-variation between nickel and chromium has been found: this cannot be produced in natural. Such good co-relation must have derived from industrial dust. After all, we would like to point out that air in even countryside area is polluted enough. There is not so much difference between municipal air pollution and countryside air pollution.
Determination of trace elements in pancreases and testes
of Zn-deficient mice

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Abstract

Eight-week-old male mice of ICR strain were divided into two groups; one was fed with zinc deficient diet (<1 μg/g Zn), the other with control diet (30 μg/g Zn). After 1 week of treatment periods, their pancreases and testes were removed. Sodium dodecyl sulphate-polyacrylamide gel electrophoresis (SDS-PAGE) and two-dimensional electrophoresis (2-DE) were performed for cytosolic fraction. After electrophoresis, the gel was cut into protein spots and subjected to PIXE analysis.
Engine lubricating oil analysis using in-air PIXE

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Abstract

Elemental particles contained in automobile exhaust particles stem from the elements in the automobile engine lubricating oil, and may become a nucleus of atmospheric particles. Therefore, investigation of elements in automobile engine lubricating oil became our major consideration relating to chemical speciation of particles in the atmosphere. Detection of elements in automobile engine lubricating oil is one of the challenging subjects in environmental research. Consequently, we tried to elemental quantitative analysis by in-air PIXE for an engine lubricating oil. Experimental result, the determination limit by the analysis method is 50 ppm for light elements (internal standard: Sc 100 ppm). For mass number elements larger than Ar (internal standard: Y100 ppm), the determination limit is 10 ppm. Thus, the problem for the quantitative analysis was able to be clarified.
Relationship between radioactive isotopes and metals contained in the air suspended

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Abstract

Relationships between stable elements and radioactive isotopes were explored for understanding the behavior of aerosol around Nagasaki-City. In the report, we presented relationships between the amounts of radioactive isotopes Pb-210 and Be-7 obtained from gamma spectrometry and the stable elements obtained from PIXE analysis, and considered conceivable behavior.

Thirteen elements were significantly detected from the PIXE analysis. Amount of the 9 elements, nss-Br, Al, Se, nss-S, Fe, Zn, nss-Sr, Rb and Pb were correlated with Pb-210 radioactivity and varied seasonally, where nss indicates non-sea-salt. The other 4 elements, Cu, nss-Mg, Ni and Na showed no seasonal variation. It suggests that large part of the 9 elements may come from the continent with the small grains of aerosol and the 4 elements may come from neighborhood with the relatively large grains.
Chemical characterization of atmospheric aerosols at Phimai, Thailand

II. Contribution of fossil fuel combustion, biomass burning, and soil dust

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Abstract
An intensive field program was performed to measure atmospheric aerosols at the Observatory for Atmospheric Research, in Phimai, Thailand during July 2007-June 2008. According to a backward trajectory analysis, the surface wind pattern in the dry season was northeasterly from middle October 2007 to middle March 2008, and then shifted southerly from middle March to early May. For the other period, southwesterly monsoon was prevailed in the wet season. A relationship between elemental carbon (EC) and non sea-salt sulfate (nss-SO₄²⁻) in fine particles revealed that the polluted air masses rich in (NH₄)₂SO₄ emitted from east Asia was transported to Phimai, while in the latter period of dry season, aerosols rich in EC emitted from biomass burning in Indochina were dominant. These source-receptor relationships which
were analyzed in the previous report were supported by the detailed analysis using Pb and Zn in fine particles in the dry season. For soil dust, Zr in coarse particles was a good indicator of soil dust in atmospheric aerosols in the latter dry season, because it could be re-suspended into the atmosphere from surface soils in Indochina after the surface soil became dry. Furthermore, the relationship between K and Fe in fine and coarse particles strongly suggests that the soil dust could be also released into the atmosphere by the strong thermal plume generated by biomass burning in the latter dry season.
Composition characteristics of size-resolved airborne particles in Himeji city

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Abstract
In order to shed light on the effect of airborne particles (particulate matter: PM) on human health, we carried out size-resolved sampling of PM in Himeji City, and elemental and ionic composition analyses of the PM sample. Size-resolved PM was collected using a 3-stage NLAS impactor (Tokyo Dylec Co., Ltd.; particle cut size at sampling stages was 10, 2.5 and 1.0 μm for a flow rate of 3 L/min) with a 1 week sampling interval, and the PM sampling was began in November, 2009. As a result, the composition characteristics of size-resolved airborne particles were able to be clarified.
Analysis of cooling water samples in accelerator facilities

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Abstract

It is very important that the cooling water system for accelerator components such as magnets and targets is maintained safely in high energy accelerator facilities. Dissolution of metallic elements and radioactive nuclides into the cooling water may be much enhanced in high dose radiation fields in high energy accelerator facilities. In this work, the concentration of the metallic elements in the cooling water collected just after the beam stoppage was determined in order to clarify the dissolution behavior of the metallic elements from the cooling water system in radiation fields. The measurement was carried out in an electron linear accelerator facility. The PIXE method was attempted to be applied to the cooling water measurement.
Clay body analysis of "Uda pot" in the mid-Tumulus period by PIXE

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Abstract

It aims to presume cultural exchanges of an ancient movement and the trade, etc. by making comparative study analyzing PIXE of the earthenware excavated in the Takisawa-village and another region and clarifying the elementary composition. In this promotion, the earthenware at the tumulus period from [oogamadate] ruins in the Takisawa-village to the Tokai region was excavated, and it was assumed to be a subjectivity to judge the earthenware was from Tokai by comparing the clay body element analyses of the Takisawa-village excavation earthenware and the Tokai region earthenware.
Method development for identification of geographical origin of Japanese apples by PIXE analysis

-Characteristic of elements in soils from apple producing regions-

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Abstract

We studied characteristic of elements in soil samples from apple producing regions to develop method for identification of geographical origin of Japanese apples by PIXE analysis. Soils of Japanese 'Fuji' apples were examined in this study. Sampling sites were selected from Kazuno and Yokote city that were apple producing regions in Akita Pref. in Japan. Soil samples were collected from upper (0-20 cm) and lower (20-40 cm) layers in their sites. Twenty four elements were respectively determined from Kazuno and Yokote samples. The following elements were respectively determined from Kazuno and Yokote samples: “Na, Mg, Al, Si, K, Ca, Ti, V, Fe, Cl, Cr, Mn, Cu, Zn, Ga, As, Br, Rb, Sr, Y, Zr, Nb, Pb”; “Na, Mg, Al, Si, K, Ca, Ti, Fe, Cl, Cr, Mn, Cu, Zn, Ga, As, Br, Rb, Sr, Zr, Nb, Mo, Ba, Pb”. Both soil samples in Kazuno and Yokote were rich in “Na, Mg, Al, Si, K, Ca, Ti, Fe”. Concentration ranges of those elements were three orders of magnitude higher than those of other elements (>=1000 μg/g-dry). Soil samples in Kazuno were poor in “S, Cl, V, Cr, Mn, Cu, Zn, Ga, As, Br, Rb, Sr, Y, Zr, Nb”. Regarding soil samples in Yokote, they were poor in “S, Cl, Cr, Mn, Cu, Zn, Ga, As, Br, Rb, Sr, Zr, Nb, Mo, Ba, Pb”. Concentration ranges of those elements were three orders of magnitude lower than those of other elements (<1000 μg/g-dry). “V and Y” and “Mo and Ba” were respectively determined from Kazuno and Yokote samples alone.
PIXE analysis of trace element levels in feather samples of birds of prey

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Abstract

The purpose of this research is to observe the effects of harmful trace elements in birds of prey.

The feathers of Osprey \textit{Pandion haliaetus}, Black-tailed Gull \textit{Larus crassirostris} and Ural Owl \textit{Strix uralensis} were analyzed for 5 harmful trace elements (Cr, As, Cd, Hg, Pb) using PIXE at NMCC. The concentration of Hg and Pb were detected from all samples. Range of those elements' concentrations were as follows: (Hg 4.3—34.7\textmu g/g, Pb 3.4—27.6\textmu g/g). It seemed that the difference of Hg concentrations was not caused by the difference of food habits among birds of prey. Cr and As were not detected from most of samples. Cd was not detected from all samples.
Investigative studies on water contamination in Bangladesh
– Primary treatment of water samples at the sampling site –

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Abstract
Arsenic concentration in 13 well waters, 9 pond waters, 10 agricultural waters and a coconut juice taken in Comilla district, Bangladesh, where the problem of arsenic pollution is the most severe, was investigated. High-level arsenic is detected even in the well water which has been kept drinking by the people. Relatively high arsenic concentration was detected for some pond and farm waters even though the sampling was performed just after the rainy season and the waters were expected to be highly diluted. Clear relationship was observed in elemental compositions between the pond water and the coconut juice collected at the edge of the water. These results are expected to become the basic information for evaluating the risk of individual food such as cultured fishes, shrimps and farm products, and for controlling total intakes of arsenic. In order to solve the problem of transportation of water samples internationally, a simple method of target preparation performed at the sampling cite was established and its validity was confirmed. All targets were prepared at the sampling cites in this study on the basis of this method.

Keywords: PIXE, Arsenic, Poisoning, Contamination, Well water, Pond water, Bangladesh, Keratosis, Leucomelanosis
Improvement in digestion method using nitric acid for PIXE analysis of plant samples containing opal-A

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Abstract

Si is found in plants at concentrations ranging from 0.1 to 10 %, which is equal to or even higher than that of several macronutrients. There are some general trends in silicon accumulation that monocots and dicots tend to be high and poor Si accumulators, respectively. *Eleocharis acicularis*, a monocot, has recently been reported as a Si accumulator. PIXE is one of the convenient methods to analyses the total concentrations of heavy metals in plant. However, *E. acicularis* contains opal-A within the plant cells, which is difficult to be totally dissolved by using nitric acid. Therefore, it is necessary to improve the digestion method using nitric acid for PIXE analysis of plant samples containing opal-A. In this study, the effect of microwave heating times and leaving time after heating was investigated. When the samples were heated twice, the concentrations of heavy metal by PIXE are lower than those by ICP-MS. However, when the were heated three times and left one day after heating, metal concentration difference between PIXE and ICP-MS became small. The result of the present study suggested that it is necessary to improve the digestion method for plant samples containing opal-A.
**Difference between arsenate and arsenite in toxicity**

**to Fe nutrition in barley grown hydroponically**

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**Abstract**

The symptoms of Arsenic (As) toxicity was severe in the plants treated with 67 µM Arsenite and 670 µM arsenate. It was inferred that severity of toxicity in arsenite is more than 10 times of that of arsenate.

Release of Mugineic acid (MA) in roots was statistically similar among the plants treated with arsenate in all of the sampling days within 3 weeks. In the plants treated with 67 µM arsenite did not release MA in each sampling day within 3 weeks. The MA concentration of roots of the plants was measured after the treatment for 2 weeks. The MA concentration in the roots of the plants treated with 6.7 µM arsenate was significantly lower as compared with control, but that of the plants with 67 µM arsenate was similar to control. In the plants treated with 67 µM arsenite did not produce MA in the roots.

The plants treated with arsenate increased Apoplastic-As according to the increase of As concentration in the medium. Moreover, the ratio of As/Fe was 0.98 in the plants treated with 670 µM arsenate; subsequently, 0.88 in the plants with 67 µM arsenate. On the other hand, the ratio of As/Fe of the plants treated with arsenite was highest with the value, 0.185, at the plants with 6.7 µM. These results suggested that arsenate may adsorb to the root surface with Fe in the form of FeAsO₄. It is possible that Fe might be converted to unavailable or immobilized form by being complexed with As.
Multi-element analysis of PIXE for lichen samples from Shirakami-Sanchi, Akita
-Utilities of lichens as bioindicators of environmental monitoring-

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Abstract

Utilities of lichens as bioindicators of environmental monitoring were examined by PIXE analysis. Several lichens sampled from Shirakami-Sanchi, Akita. They were dried by air and milled by an agate mortar. Powdered lichens were sifted and suspended into water containing Triton X-100 as a surface-active agent and polyvinylalcohol as a binder agent. The suspensions were filtered on Nuclepore Track-Etch Membrane (pore size : 10 μm) and the membrane targets were bombarded by 3 MeV Proton from NMCC cyclotron. 5 Kinds of lichen were analyzed by PIXE and concentration for 26 elements were determined by PIXE analysis. Determined elements were major, minor and trace elements in biological materials and contained several essential elements such as Fe, Zn and Cu. And also this PIXE analysis could measure some toxic elements, Hg and Pb. Relative standard deviation for each lichen sample from 3 analytical targets were 1-13% for traces of Hg and Pb. It was shown that the uniformity of the target was excellent. The sample preparation for lichens is simple and this PIXE analysis is able to measure so handily multi-elements, that it is useful for the environmental evaluation by lichens.
The dynamics of liposomal anti-tumor agent

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Abstract

The aim of this experiment was to confirm that the platinum reaches to the regional lymph node (submandibular lymph node) or not when the liposomal cisplatin was injected to the mouse tongue.

The liposomal cisplatin was injected in the mouse tongue under anesthesia. After 24 hours, the tongue and the both side of submandibular lymph nodes were extracted, and then they were desiccated in the dryer at the 200 degree centigrade overnight. The tongues were treated by the chemical ashing method using nitric acid adding with Indium as the internal standard. The submandibular lymph nodes were treated by the standard-free method using the PIXE.

The large size of liposome remained in the tongue more than the small size of liposome. The enough quantity of platinum was accumulated in the regional lymph nodes (over 2.6μg/g). The large size (about 800 nmφ) of liposome was very useful for the sustained release agent.

We believe that this method is useful for control to the side effects caused by the administration of anti-tumor agent.
Investigation of anticancer drug concentration on cancer cell to enhance therapeutic effect of particle radiotherapy

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Abstract

Concentrations of the chemotherapeutic agent, cis-diamminedichloro-platinum(II) (CDDP) in a solid murine tumor were investigated by submilli- and micro-PIXE analyses as well as a conventional PIXE analysis to study enhancement of antitumor effects caused by radiotherapy combined with CDDP treatment. The conventional PIXE analysis showed that the platinum concentrations of the tumors treated with CDDP at a single dose of 10 mg/kg were approximately 1.6ppm 1 hour after the injection, 2.0 ppm after 6 hours. The platinum distributions in the tissue or cells of the treated tumors could not be evaluated in the present submilli- and micro-PIXE analyses since the platinum concentrations might be nearly equal to or lower than their detection limits.
Antitumor effect of radiosensitive microcapsules
under subcutaneous injection

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Abstract
Since 2004, we reported the use of liquid-core microcapsules for anticancer drug targeting. However, rupturing of microcapsules via radiation was lower than we expected. The more efficient rupturing of microcapsules by radiation was needed. In this study, we tested whether radiation-induced O\textsubscript{2} from H\textsubscript{2}O\textsubscript{2} facilitate the rupturing of microcapsules, or not.
The capsules were generated by spraying a mixture of 2.0% hyaluronic acid, 2.0% alginate, supplemented with 0.2 mmol carboplatin and 3 \% H\textsubscript{2}O\textsubscript{2} on mixture of 0.5 mol/L CaCl\textsubscript{2} andFeCl\textsubscript{2}. Resulting microcapsules were irradiated by \textsuperscript{60}Co \gamma ray at doses ranging from 0.5 to 2.5Gy. The released carboplatin was detected and quantified by particle-induced X-ray emission.
The antitumor effect was measured by growth delay. The strength of adverse effect was measured basing on fuzzy hair, loss of body weight and death. The radiation-induced O\textsubscript{2} from H\textsubscript{2}O\textsubscript{2} significantly increased rupturing of microcapsules. Those increased the intratumoral concentration and antitumor effect of carboplatin; however they were not significant. Our microcapsules should be more improved to increase radiation-induced rupturing.
The analyses of bystander effect induced by low-dose radiation in glioma cell

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Abstract

Recently, it was considered that the cell lethal effect by low dose radiation was due to bystander effect. Cells irradiated low dose radiation secreted something liquid factor that induced lethal effect by signal transduction. So far, we suggested that radiation induced bystander effect is closely relative with sphingomyelinase. To analyze mechanism between activation of sphingomyelinase and induction of bystander effect, in this study we investigated divalent metal that are necessary for sphingomyelinase activation using PIXE analysis and mechanism of lethal effect by sphingomyelinase. Extracellular zinc value by radiation (0.1Gy) decreased for 5 min and increased for 15 min after irradiation. On the other hand, fluctuation of extracellular zinc value by treatment of sphingomyelinase inhibitor was suppressed. NO and DNA damage were induced in the bystander cells, that were non-irradiated cells culturing with medium of irradiated cells. These results indicate sphingomyelinase is activated by binding zinc element, sphingomyelinase itself function as bystander factor and induce cell death.
PIXE analysis of fluoride and trace elements in shark teeth

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Abstract

We have measured the concentration of fluoride and other elements simultaneously in shark teeth using PIXE. 17 samples taken from 10 species of shark. As a result, 18 elements were detected. Particularly, Ca, Cl, Mg, Na, P, Sr, Zn, and F showed high concentrations. There is a high statistical correlation between Ca and P in shark teeth. However, there is a low statistical correlation between Ca and F. Judging from shark teeth is composed of Apatite, it is possible to consent to the fact. We have 100 samples of Shark teeth and are planning on reporting the findings of a study with larger samples in the near future.
Origin and transportation course of heavy metal elements in the particulate matter (PM) at the Hachimantai mountain range in northern Japan

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Abstract

Particulate matter (PM) was collected at the Hachimantai mountain range in northern Japan. In the present study, the origin and transportation course of the heavy metal elements were discussed for PM_{fine} and PM_{coarse}, determined by using PIXE (Particle Induced X-ray Emission) and back trajectory analyses. The result shows that the PM_{fine} emitted mainly from artificial sources, compared with the PM_{coarse} from natural sources, since the Enrichment Factor (EF) value and S/K ratio of PM_{fine} was one order higher than that of PM_{coarse}. The origin of Pb in PM at Akita Hachimantai mountain range has both of the anthropogenic and the metal refining origins, using the analysis of Pb/Br. Furthermore, from result of back trajectory analysis when the air mass was transported over the Japanese Islands, the air mass was mainly passed over the large-scale industrial area in Japan. The origin of As and Se were mainly gasoline and coal combustion, and were transported from Chinese continent and/or Korea peninsula.
Quantitative analysis of untreated oil samples in in-air PIXE

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Abstract

The method of quantitative analysis of oil samples in in-air PIXE has been developed on the basis of the standard-free method. The components of the continuous X-rays originated from air and a backing film can be exactly subtracted by normalizing with the yields of Ar K-α X-ray peak. The method was developed using three kinds of standard oil samples and its accuracy and reproducibility were confirmed by comparing the results with those obtained by the internal-standard method using single-element standard oils. Validity of the method for practical oil samples was confirmed by applying the method to various kinds of oils such as engine, machine and a few kinds of edible oils. It was confirmed that the method is valid in a case where arbitral element is designated as an index element depending on the composition of the oil sample. Moreover, targets can be prepared by anyone within ten seconds only by putting a drop of oils onto a backing film. It is expected that the method will become a powerful tool for the studies in atmospheric environmental sciences.

Keywords: Oil, In-air PIXE, Standard-free, Quantitative analysis, Mobile oil