(第3号様式)(Form No. 3)

学位論文要旨
Dissertation Summary

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論　文　名 (Dissertation Title)：Lichen-substratum interactions at abandoned mine sites in southwest Japan

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Abstract

Heavy metal pollution is a worldwide problem that must be solved. Many organisms have been investigated for use in the assessment, monitoring, and/or remediation of heavy metal pollution. The use of native organisms in a polluted area is an approach that could contribute to a reduction in costs and secondary environmental impacts. Because several fruticose lichens occur even in areas highly polluted by heavy metals, such as abandoned mine sites, lichens may meet the requirements for use in the assessment, monitoring, and/or remediation of polluted sites.

In statistical analyses of lichens, correlations have been identified between the accumulation capacity of lichens and the heavy metal concentrations of the substrata. However, the use of lichens in practical applications as bioindicators or biomarkers of heavy metal pollution in soils has not been proposed in previous studies. Therefore, several fruticose lichens, including *Stereocaulon exutum* and several *Cladonia* Spp., from contaminated abandoned mine sites and the associated control areas, and the corresponding substrata were comprehensively investigated (1) to determine the behavior of heavy metals during the weathering of slag as affected by abiotic processes and by biotic processes mediated by *Stereocaulon exutum*, (2) to determine the distribution of the heavy metals in the thalli of *S. exutum*, and (3) to determine the correlations between the Cu, Zn, As, and Pb concentrations of lichens and those of the corresponding substrata in areas polluted with heavy metals and the associated control areas. As a final step,

distribution maps of Cu, Zn, As, and Pb in the study areas were constructed using by the average concentrations in the soils and the lichen thalli, and with these maps, the use and practical application of lichens as biomarkers for the assessment and monitoring of heavy metal pollution of soil were evaluated.

The surface of a waste dump is composed of primarily tailings and slag fragments. The slag consists primarily of willemite, fayalite, and/or magnetite with a silicate glassy matrix and contains matte drops, which are Cu-, Zn-, Pb-metals, -alloys, and -sulfides. The heavy metal-rich phases are altered during the weathering process in the following order: willemite > matte drops >> fayalite. The willemite and matte drops are ultimately converted to Fe-hydroxides during the weathering process. In addition to abiotic weathering, the heavy metals are dissolved during biotic weathering processes by substances from the lichens and hyphal penetration. The heavy metals tend to concentrate in the lichen thalli during lichen-mediated weathering processes, which implies that the dissolved heavy metals are absorbed into the lichen thalli. Absorbed Cu and Zn are distributed within the cells of hyphae, whereas iron and arsenic are distributed on the surface of hyphae. At the sites of Fe and As concentrations on the surface of hyphae, amorphous or low crystalline materials occur.

Based on previous studies, cations in lichen thalli are distributed into four fractions, i.e., the intercellular and surface, ion exchange site, intracellular, and residual fractions. Although the form of the ions was not identified in this study, the distribution of elements in the hyphal cells indicated the possible absorption of ions into the cytoplasm from external solutions or through ion exchange sites. For the Fe and As concentrated on the surface of hyphae as amorphous or low crystalline materials, this distribution could be explained by elemental precipitation or the formation of compounds during the evaporation of external solutions.

The concentrations of heavy metals of lichens in areas with heavy metal pollution were higher than those of lichens in the control areas. The concentrations of Cu, Zn, As, and Pb of the thalli were positively correlated with those of the corresponding substrata for a wide range of metal concentrations. Thus, lichens can be used as biomarkers of heavy metal pollution in soils. However, to determine the use of lichens for practical applications, more than statistical analyses are required. Consequently, the practical application was evaluated by using distribution maps of the average heavy metal concentrations of lichens and the corresponding substrata.

At the scale of one study area, the distributions of average heavy metal concentrations of lichens and those of the corresponding substrata were broadly similar. Although the maps for the distribution of Pb varies widely, the maps for the distribution of Cu, Zn, and As in lichens had very similar distributions to those of the corresponding substrata at the scale of all study sites in southwest Japan. Therefore, in the present study, a large-scale analysis of lichens with many samples successfully detected the distribution of Cu, Zn, and As pollution in soil. Based on the correlations and the distribution maps, lichens were good biomarkers for heavy metal pollution of soil, and therefore, the interactions between lichens and substrata become very important and of interest in environmental studies. Compared with soil analyses, the analysis of lichens has several advantages in the assessment and monitoring of soil pollution by heavy metals. To increase the sensitivity of pollution detection, the variability of the data can be reduced by increasing the number of samples. However, for soil samples, larger volumes are required than those of lichen samples to reduce the variability of the data. Lichens absorb the elements in surface and pore waters dissolved from the substrata, and therefore, the analysis is not directly affected by mineral composition, grain size, solubility of elements in metal-rich minerals as occurs with soil. Moreover, the analysis of lichens is more applicable than that of soils for monitoring long-term changes in soil pollution. As a result, analysis of lichens may decrease the cost and secondary environmental impacts of the assessment and monitoring of soil pollution, which are important advantages for application in practical use. In conclusion, based on this study, *Stereocaulon exutum*, *Cladonia humilis*, *C. ramulosa*, *C. krempelhuberi*, *C. crispata*, *C. scabriuscula*, *C. macilenta*, *C. rangiferina*, and *C. trassii* can be used in practical applications for biomonitoring and risk assessment of Cu, Zn, and As pollution of soils. Moreover, lichens occur in various environments and therefore can have broad utility as biomarkers of heavy metal pollution worldwide.