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Chemotaxonomy of plants by resistant macromolecular analysis in charred mesofossils from the Cretaceous Futaba Group

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Resistant macromolecules composing living plant tissues tend to be preserved through degradation and diagenesis, hence constitute major parts of fossil plants or sedimentary plant-derived organic matter. And their monomer compositions vary widely among different plant taxa, organs and growth stages. Thus, analysis of such macromolecule may serve as new technique for paleobotanical evaluation distinctive from classical paleobotnical studies depends on morphological preservation of fossils. However, there have been few studies of the macromolecules, especially in ancient geological samples such as the Paleozoic and Mesozoic. In the present study, we analyzed plant fossils from the Cretaceous strata in Japan to examine chemotaxonomic characteristics of fossil macromolecules.

Charred mesofossils of angiosperms and gymnosperms were separated from carbonaceous sand stone of the Cretaceous Ashizawa Formation, Futaba Group. These mesofossils include fruit fossils of *Hironoia fusiformis* and *Archaefagacea futabensis*, a flower fossil of *Esgueiria futabensis*, leaf fossil of *Juniperus*, a stem fossil of *Epfedra* and some fossils of fruits, seeds and woods. Powdered fossil samples were extracted with methanol and dichloromethane, and were subsequently refluxed under 110 ℃ to remove free compounds completely. The residues were hydrolyzed by KOH / methanol under 110 ℃. These released compounds were analyzed by GC-MS. Additionally, multivariable analysis were calculated using SPSS software. We used hierarchical clustering to group fossils with similar lipid distributions among species or organs.

n-alkanes, branched isoprenoids, sterans, hopanes, and aromatic hydrocarbons were mainly present in solvent extract fraction. Aromatic hydrocarbons contained various higher plant derived diterpenoid and triterpenoid derivatives. These compounds are commonly considered as chemotaxonomic markers of gymnosperms and angiosperms respectively. Unexpectedly, triterpenoid derivatives were detected from gymnosperm fossils abundantly, indicating that free lipids may have moved in the coal bed, thus these lipids are not suitable for chemotaxonomic use in this study. On the other hand, as main hydrolyzed products (ester-bound molecular units) from all fossils, C6-C28 *n*-alkanoic acids and C8-C28 *n*-alkanols were detected. Multivariable analysis were calculated in lipid distribution for these released alkyl lipids from each fossils. Cluster analysis revealed a recognizable pattern in released alkyl lipid distribution. All five fossils of woody tissue were present in a cluster that excluded non-woody tissues. Additionally, exclusive of Juniperus fossil, the lipid signatures were similar among angiosperms or gymnosperms. From these results, we propose that it is likely to be realized that paleolipidomics-like detailed chemotaxonomy of fossil plants by making a comprehensive evaluation for various lipid components involve bond alkyl lipids.

Keywords: chemotaxonomy, alkyl lipid, plant fossil, Cretaceous, resistant macromolecule, multivariable analysis