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Spectroscopic characterization of dissolved organic matter isolates from sediments and the association with phenanthrene binding affinity

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HIGHLIGHTS

- Spectroscopic properties of different dissolved organic matter isolates from sediments are compared.
- Organic sources of sediments are well reflected in alkaline extractable organic matter isolates.
- Terrestrial humic-like fluorophores are well associated with the extent of phenanthrene binding.
- Phenanthrene binding affinity is better explained by the degree of biotransformation than by aromatic content.

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ABSTRACT

In this study, selected spectroscopic characteristics of sediment organic matter (SOM) were compared and discussed with respect to their different isolation methods, the source discrimination capabilities, and the association with the extent of phenanthrene binding. A total of 16 sediments were collected from three categorized locations including a costal lake, industrial areas, and upper streams, each of which is likely influenced by the organic sources of algal production, industrial effluent, and terrestrial input, respectively. The spectroscopic properties related to aromatic structures and terrestrial humic acids were more pronounced for alkaline extractable organic matter (AEOM) isolates than for the SOM isolates based on water soluble extracts and pore water. The three categorized sampling locations were the most differentiated in the AEOM isolates, suggesting AEOM may be the most representative SOM isolates in terms of describing the chemical properties and the organic sources of SOM. Parallel factor analysis (PARAFAC) based on fluorescence excitation–emission matrix (EEM) showed that a combination of three fluorescent groups could represent all the fluorescence features of SOM. The three categorized sampling locations were well discriminated by the percent distributions of humic-like fluorescent groups of the AEOM isolates. The relative distribution of terrestrial humic-like fluorophores was well correlated with the extent of phenanthrene binding ($r = 0.571$; $p < 0.05$), suggesting that the presence of humic acids in SOM may contribute to the enhancement of binding with hydrophobic organic contaminants in sediments. Principal component analysis (PCA) further demonstrated that the extent of SOM's binding affinity might be affected by the degree of biogeochemical transformation in SOM.

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1. Introduction

Sediments are considered the sources and the sinks for nutrients and pollutants in aquatic environments, affecting their fate and the distribution. Sediment organic matter (SOM), constituting an important portion of sediments, is a heterogeneous mixture of the classes of the organic substances encompassing carbohydrates, proteins, lignins, organic acids, and various other uncharacterized compounds such as humic substances (HS). Owing to its preservative nature, SOM may provide a wealth of information on the biogeochemical changes of organic matters occurring in the sites as well as on their formation and transport (Meyers and Teranes, 2001). Early diagenetic processes on suspended solids such as biodegradation and photo-oxidation make a great contribution to the changes in the quality and the composition of SOM (Jaffé et al., 2006; Osburn et al., 2012). For many lakes and estuaries under human impact, the quality of SOM may be altered by the relative contributions of natural and anthropogenic sources (Wang et al., 2012). Natural sources of SOM can be further divided

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into autochthonous and allochthonous origins. The former is derived from carbon excretion and the leachate of algae and aquatic macrophytes, and their microbial degradation products, while the latter is typically referred to the terrigenous inputs from upstream catchments.

The fate, the transport, and the bioavailability of hydrophobic organic contaminants (HOCs) in sediments are largely affected by the quantity and the quality of SOM (Chin et al., 1997; Chefetz et al., 2000). In general, organic carbon normalized binding (or partitioning) coefficient (K_{oc}) has been used as an indicator to determine the distributions of HOCs in sediments when the total organic carbon is known (Chin et al., 1997). Many previous studies have presented close relationships between the chemical and the structural nature of dissolved organic matters (DOM) and the K_{oc} value of a specific HOC. For example, hydrophobic nature, aromatic content, and molecular weight of DOM have been suggested as the descriptors to estimate the K_{oc} values (Kopinke et al., 2001; Hur and Schlautman, 2003; Hur et al., 2013). However, it should be noted that the generally accepted relationships may be broken when the subjects are extended into a class of highly heterogeneous organic matters including SOM (Wen et al., 2007; Hur et al., 2009). Recently, fluorescence characterization of SOM has been highlighted as a simple and easy tool for estimating the K_{oc} values independent of the sediment sources (Hur and Kim, 2009).

Fluorescence EEM–PARAFAC has been widely used for assessing the quality of DOM in various aquatic systems, and for tracking DOM sources (Stedmon et al., 2003; Fellman et al., 2010). Despite the numerous applications reported to date, little attention has been paid on SOM characterization (Santín et al., 2009; Wang et al., 2013). Recently, Osburn et al. (2012) successfully quantified several fluorescence EEM–PARAFAC components from alkaline extractable organic matter from sediments. They have demonstrated that PARAFAC modeling of SOM in adjunct with DOM possibly distinguished among different organic sources, providing insight into the dynamics of the nutrient supply of the sediments to the overlying water column.

Altogether, it can easily be inferred that the chemical and the structural information on SOM derived from fluorescence EEM–PARAFAC could also be used to estimate the extent of HOC binding. To our knowledge, however, there has been no study to explore the potential use of the EEM–PARAFAC for the prediction of the extent of HOC binding. In this study, we collected 16 sediment samples from a costal lake and the surrounding areas, which were further categorized into three different locations including a coastal lake, streams at industrial areas, and upper streams at rural areas. The three categorized sampling locations can be characterized by their major organic sources of in situ primary production of algae, human and industrial activities, and terrestrial input from the catchments covered by forest and agricultural areas, respectively. This study aimed (1) to compare selected spectroscopic properties of SOM and the extent of HOC binding with respect to different SOM isolation methods and three categorized sampling locations, and (2) to suggest the descriptors for the extent of HOC binding primarily based on fluorescence EEM–PARAFAC.

2. Experimental

2.1. Study area and sample collection

The study area is an artificial coastal lake, the Shiwha lake (N37°17′, E126°15′), and the upstream catchments, which are located on the west coast of Korea (Fig. 1). The northern part of the upstream catchments is occupied by an industrial complex, while the upstream main channel or the eastern part of the upstream catchments of the lake is mostly covered by rural and forest areas (Fig. 1). The artificial lake was initially developed in 1994 by the construction of a dike in the boundary between the coast and the open sea. A tidal power plant was constructed in the middle of the dike afterwards so that seawater circulated into the lake.

Bottom sediments were collected using a grab sampler (Ekman dredge) in June, 2013, from 16 different sites, which were further categorized into three sampling locations – lake (L1–L7), streams at industrial areas (I1–I5), and streams at rural areas (S1–S4). The samples were kept in vacuum sealed plastic bags and stored in a closed ice box while they were transported from the field to the laboratory for analysis.

2.2. Extraction of SOM from sediments

Different extraction procedures were made to obtain three types of SOM isolates from the collected sediments, which include pore water DOM (PDOM), water soluble organic matter (WSOM), and alkaline extractable organic matter (AEOM). WSOM refers to an organic fraction of SOM pool that is easily mobilized into the underlying water column. AEOM includes a majority of the HS

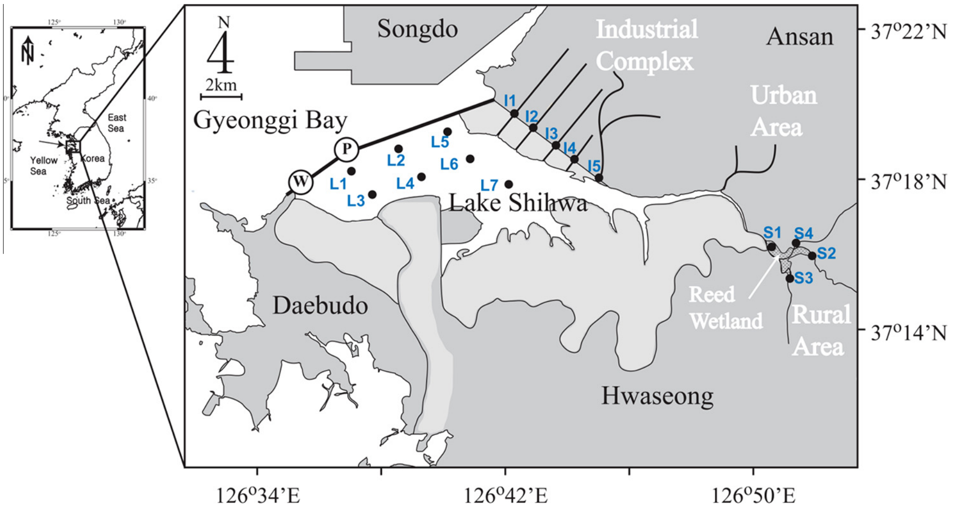

Fig. 1. Map of the lake Shiwha and the surrounding areas. The sediment sampling locations are indicated. The dark grey and the light grey colored letters indicate the geographical names and land uses of the upper catchments, respectively. The letters "W" and "P" indicate the locations of water gate and tidal power plant, respectively. The map is adopted from Phong et al. (2014).

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