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Approach deliberation for source identification of sedimentary organic matters via comparing freshwater lakes with multi-ecotypes

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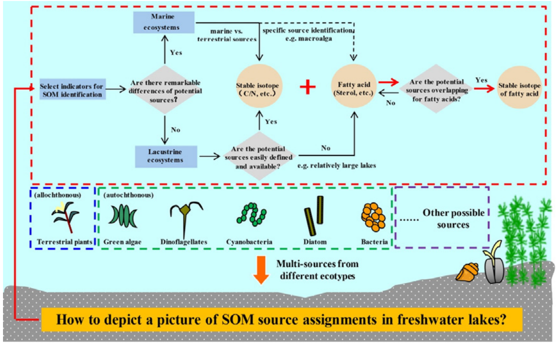
HIGHLIGHTS

- SOM sources and composition were systematically investigated in freshwater lakes.
- Multiple sources of SOM were characterized by using fatty acid biomarkers.
- Terrestrial plants derived organic carbon could well predict sediment TOC.
- Isotopic mixing model could quantitatively depict the defined vegetation sources.
- A general indicator selection procedure was proposed for SOM source identification.

GRAPHICAL ABSTRACT



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ABSTRACT

Despite of the importance of understanding the sediment quality for lacustrine management, the source evaluation of sedimentary organic matter (SOM) in freshwater lakes is still insufficient. In this study, two shallow eutrophic lakes of Lake Taihu, China and Lake Izunuma, Japan were systematically investigated. Results of fatty acid profiles demonstrated that a wide range of organic matters, varying ecotypically, was inputted into the sediments of both lakes. Interestingly, there was a strong contribution from terrestrial plants to the sediments across ecotypes, with an approximate input from bacteria, and a relatively minor input from microalgae mainly included cyanobacteria, green algae, diatom and dinoflagellates. In addition, isotopic mixing model depicted a complementary picture that a significant, but spatially variable, amount of organic matter was derived from emergent and floating-leaf plants of *Phragmites*, *Nelumbo*, *Nymphoides* and *Trapa* L in Lake Izunuma. A general indicator selection procedure for the source assignments of SOM in freshwater ecosystems was therefore proposed: fatty acids could be a valid biomarker when the potential sources are unknown or unavailable; stable isotopes could be an effective supplement approach when assessing the special or defined organic sources.

1. Introduction

Although lakes make up <2% of Earth's surface area, their sediments are well-recognized sites for the accumulation of organic carbon, in which over three times than all of the world's oceans combined

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(Molles, 2002). The sedimentary organic matter (SOM) plays an essential role in carbon and energy cycles of lakes. Therein, a critical portion provides carbon and energy sources for consumers, eventually forming the diverse food webs (Carpenter et al., 2005). Another portion is mineralized by microorganisms and releases greenhouse gases to the atmosphere (Marotta et al., 2014; Yan et al., 2017). The residuals are buried in the sediments and subsequently subjected to the diagenesis processes (Havens and Schelske, 2001; Watanabe and Kuwae, 2015). These biogeochemistry processes depend on the sources and properties of sedimentary organic matter. Therefore, the quantification of different sources is crucial for our understanding the fate of the organic matter in freshwater lakes.

It was generally accepted, so far, that lake sediments receive organic matters from autochthonous sources produced in situ by photoautotrophic phytoplankton and hydrophytes and allochthonous matters delivered by river discharge and runoff (Meyers and Ishiwatari, 1993). However, a clear picture about the extent of the aquatic versus terrestrial organic matter sources in lake sediments is still currently lacking. The terrestrial organic carbon was considered to be stored more efficiently than phytoplankton derived organic carbon in sediments, due to their susceptibility to microbial decomposition (Gudasz et al., 2015; Watanabe and Kuwae, 2015). Moreover, with great phytoplankton production by nutrient enrichment in lakes, the export of photosynthetic organisms onto the sediments (as live or dead cells) and consequently efficient storage are probably their fates with intensive and high-frequency sedimentation (Xu et al., 2014).

To date, studies of quantitatively evaluating the relative contribution of allochthonous vs. autochthonous sources to SOM pool have mostly focused on the estuarine, coastal or marine ecosystems (Woszczyk et al., 2011; Shilla and Routh, 2017). The use of sediment bulk parameters (C/N ratios, $\delta^{13}C$ and $\delta^{15}N$) and the establishment of simple models with mixing of two end members to infer SOM sources in estuarine, coastal or marine environments have a long history, as the differentiation between terrestrial and marine or between different types of plants (C3 and C4) is relatively large (Volkman et al., 2008; Rúa et al., 2017). Additionally, the use of molecular biomarkers such as fatty acids can also provide insight into the details of multiple SOM sources (e.g. microalgae, terrestrial plants and bacteria), due to their structural diversity, source specificity and relative stability (Zimmerman and Canuel, 2001; Wang et al., 2016). However, it is not evident to whether the patterns of these bulk parameters and molecular biomarkers described for estuarine, coastal or marine ecosystems are valid for source identification of lacustrine sediments. Compared with estuarine, coastal or marine ecosystems, quantitative estimation becomes uncertain in lake ecosystems as multiple organic matter sources are involved in the sediments and the differences of their stable isotope ratios are relatively small (Perdue and Koprivnjak, 2007; Li et al., 2018). Moreover, unambiguously assigning sources to molecular biomarkers is difficult, as they are rarely produced by a single class of organisms (Cook et al., 2004). Therefore, the information on SOM sources and composition inferred by these indicators in freshwater lakes is still insufficient (Xu et al., 2014, 2015b; Li et al., 2018).

The aim of this study is to challengingly test the validity of the aforementioned bulk parameters and molecular biomarkers in quantification of the relative contribution of allochthonous vs. autochthonous organic matter to SOM pool in freshwater lakes. For this purpose, two Asian freshwater lakes of Lake Taihu, China and Lake Izunuma, Japan with different ecological characteristics were systematically investigated. Their SOM sources were evaluated and compared by fatty acid biomarkers. In addition, the vegetation contributions to SOM pool in Lake Izunuma were determined by stable isotope ratios. We hypothesized that fatty acids can be a valid indicator when the potential sources of SOM in freshwater lakes are unknown or unavailable. When the potential SOM sources are determined, stable isotopes can be a complementary approach. The findings in this study provide a general indicator selection procedure for clarifying SOM accumulation processes and are benefit for properly managing or ameliorating the quality of sediments in freshwater lakes.

2. Materials and methods

2.1. Study sites

Lake Taihu, located in southeastern part of the Yangtze Delta, is a shallow (an average depth of 1.9 m) and large (an area of 2338 km^2) freshwater lake in China (Qin et al., 2010). Owing to decades of wastewater discharge, industrial pollution, and over-application of chemical fertilizers around the basin, the lake is currently subjected to hypereutrophic problem with annual heavy cyanobacteria blooms in western and northern parts of Lake Taihu. In the present study, a total of 12 sampling sites, averagely distributed in three areas (Ecotype 1, the northern region; Ecotype 2, the southern region and Ecotype 3, the lake center), were selected in consideration of different ecological characteristics of Lake Taihu. The accurate positions of all sampling sites recorded by global position system (GPS) were shown in Table 1.

Lake Izunuma, located in a cropland in northern Miyagi, is a shallow (an average depth of 0.76 m) and small (an area of 3.69 km^2) lake in Japan (Yasuno et al., 2012). Owing to decades of nutrient discharge, the lake has been transformed into its present eutrophic state, with about 80% of phytoplankton is green algae. On the other hand, the lake is also subjected to paludification because it is wholly covered by emergent and floating-leaf plants (Xu et al., 2014). The same sampling strategy with Lake Taihu is adopted for Lake Izunuma, with accurate positions of all sampling sites shown in Table 1.

2.2. Sample collection

Sample collection was conducted in June–July 2011 for Lake Izunuma and November 2011 for Lake Taihu. Triplicate surface sediment samples (0–2 cm) from each of three sites were combined and homogenized before being stored in the dark at <4 °C in the field, using the cylindrical PVC coring tubes coupling with a grab sampler. In Lake Izunuma, according to its ecological characteristics, triplicate samples of the potential vegetation including *Phragmites*, *Nelumbo*, *Nymphoides* and *Trapa* L. were collected. After being rapidly transported to the laboratory, vegetation samples were washed with distilled water for removing attached particulate organic matter. Especially, the samples of *Nelumbo* were cut and separated into leaf, stem and root. Lastly, all of the samples were freeze-dried for 24 h and stored at −40 °C for further analysis.

2.3. Sample analysis

2.3.1. Total organic carbon

Sediment samples from each location were ground (<5 μm) and treated with the addition of 5 ml 1 M HCl to remove carbonates for determining TOC contents. Thereafter, samples were dried to a constant weight (60 °C) and analyzed by TOC analyzer (SSM-5000A, Shimadzu).

2.3.2. Fatty acids

Lipids of all sediment samples were extracted and esterificated to fatty acids methyl esters (FAMEs) according to one-step method (Abudulkadir and Tsuchiya, 2008). Then, the FAMEs were separated and quantified using a gas chromatograph (GC-2014, Shimadzu) equipped with a split/splitless injector and a flame ionization detector (FID). The gas chromatograph was fitted with a Select FAME (100 m, 0.25 mm i.d., 0.20 mm) capillary column. The GC oven was programmed as follows: 150 °C (hold 5 min) to 230 °C (hold 10 min) at 4 °C/min and last hold at 250 °C for 20 min. The FAMEs assignments were made by comparison of retention times with those derivatives of authentic standards (Supelco, Aldrich, Sigma). The percentages and concentrations of individual fatty acids were calculated according to the descriptions of