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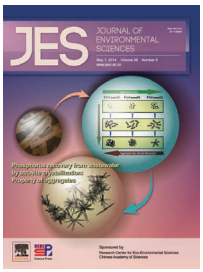

A review of environmental characteristics and effects of low-molecular weight organic acids in the surface ecosystem

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ABSTRACT

Low molecular weight organic acids (LMWOAs) are prevalent on the earth's surface. They are vital intermediate products during metabolic pathways of organic matter and participate in the tricarboxylic acid cycle during life activities. Photochemical reactions are pivotal for LMWOAs' origination and play a large role in determining their diversity and their ultimate fate. Within the long time that organic matter is preserved in sediments, it can be decomposed and converted to release organic and inorganic pollutants as well as C, N, and P nutrients, which are of potential ecological risk in causing secondary pollution to lake water. The sediment pool is a comprehensive and complex compartment closely associated with overlying water by various biochemical processes, during which LMWOAs play critical roles to transport and transform elements. This article elucidates geochemical behaviors of LMWOAs in the surface environment in details, taking natural water, soil, and aerosol as examples, focusing on reviewing research developments on sources and characteristics, migration and mineralization of LMWOAs and relevant environmental effects. Simultaneously, this review article depicts the categories and contents of LMWOAs or their contribution to DOC in environmental media, and evaluates their importance during organic matter early diagenesis. Through concluding and discussing the conversion mechanisms and influencing factors, the next research orientations on LMWOAs in lake ecosystems are determined, mainly concerning relationships with hydrochemical parameters and microorganisms, and interactions with pollutants. This will enrich the knowledge on organic matter degradation and related environmental effects, and help reconstruct a theoretical framework for organic compound succession and influencing factors, providing basic data for lake eutrophication and ecological risk assessment, conducive to better control over water pollution and proper management of water quality.

Introduction

Dissolved organic matter (DOM) mineralization consumes a large amount of oxygen and releases C, N, P, and S, thus seriously deteriorating water quality and promoting water eutrophication. Low molecular weight organic acids (LMWOAs), which could be produced and consumed in this complicated ecological and physiological process, and are utilized as an index of water quality evolution (Stinley et al., 2004; Petra et al., 2004), are ubiquitous water soluble organic compounds that have been detected in a variety of environmental samples (Brinkmann et al., 2003; Gogou and Stephanou, 2004; Tedetti et al., 2006). LMWOAs regulate heavy metals and toxic organic compounds in terms of their ecological toxicology and environmental migration, through adsorption and chelation/complexation with them (Elkhatib et al., 2007; Gao et al., 2010). As important carriers of materials and energy in the biosphere,

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LMWOAs are of great significance during these processes, being consumed by microorganisms and converted to become part of the organisms to increase biomass. Conversely, they are also released from these organisms' own excretion and macromolecular organic matter's decomposition (Kleerebezem et al., 1999). Bottom sediments strongly influence the eutrophication and biodiversity of lakes, and preserve the historical records of environmental change. Simultaneously, sediment provides an important field for organic matter activities, such as the biogeochemical cycle of carbon, nitrogen, phosphorus and the fate of pollutants. Many organic compounds are produced and consumed with nutrient (N and P) regeneration during sediment particulate organic matter mineralization (Wu et al., 2008). During sediment diagenetic processes, the molecular composition of organic matter changes, and these changes follow some general patterns leading to the definition of diagenetic indicators, the amino acids whose relative abundance and composition have been used as the basis of evaluating diagenesis (Cowie, 1994). However, studies have suggested that LMWOAs function similarly to indicate sediment early diagenesis. Moreover, LMWOAs are useful to elucidate photochemical effects and corresponding biological effects in lake waters (Zagarese et al., 2001; Jonsson et al., 2007). Therefore, the components and characteristic dynamic features of LMWOAs, whether in lake water or in sediments, became the main emphasis of their discussion. Therefore, the influence of LMWOAs' biogeochemical processes on water environment chemistry, as well as their sources, characteristics and migration-transformation, and other environmental effects will also be expounded comprehensively in this paper.

1 Source, characteristics of LMWOAs and the influencing factors

Water-soluble organic compounds include complex constituents, and not only consist of dissociative macromolecules such as enzymes, aminosaccharides, polyphenols and humic acid, but also contain dissociative low molecular weight organic acids, amino acids and saccharides. Natural fresh waters contain a wide array of low molecular weight organic molecules together with more complex organic constituents such as humic substances and detritus (Drozd et al., 1997). The origin of LMWOAs is considered to be microbial decomposition of various plant materials, and some are even used as identifiers for certain genera of decomposers (Berdie et al., 1995).

Low molecular weight organic acids and related polar compounds are ubiquitous water-soluble compounds that have been detected in a variety of environmental samples, including aerosols, rainwaters, ice-cores, marine sediments, freshwaters, and soils as illustrated in **Table 1**. LMWOAs also participate in many biological processes. They are important intermediates in the tricarboxylic acid and glyoxylate cycles, in catabolism and anabolism of amino acids. In anoxic marine sediments, LMWOAs act as important active intermediates between large biomolecules and the ultimate remineralization products CH_4 and CO_2 during their metabolic activities. Through HPLC detection, the carboxylic acids have been found to be mainly formate and acetate all year round, with the concentration rising from the interface to an asymptotic value of 150–200 μmol/L at 30-cm depth. However, during summer pooling of fermentation products as described above, the identified portion of ΣCOOH rose to 80%–90% and was composed mostly of acetate and propionate (Albert and Martens, 1997). This information can be useful to determine carboxylic acids and their role in the anaerobic carbon cycle. The organic acids from Lake Aydat, a small eutrophic lake located in the French Massif Central, have been analyzed to determine their possible sources and their fate during early diagenesis. Researchers found that the H^+-labile components, assumed to be mostly of bacterial origin, disappeared rapidly with depth. In contrast, notable OH^--labile compounds, mostly derived from algae and terrestrial plants, survived in the deepest samples analyzed (Stefanova and Disnar, 2000). C_2–C_5 dicarboxylic acids and malic, glyoxylic, and pyruvic acids were determined using capillary electrophoresis. Oxalic acid dominated in these acids, with seasonal average concentrations of 107–412 ng/m^3, malic and succinic acids were comparable mutually in different seasons within the concentration range from 20–60 ng/m^3, glutaric acid was less at an average of 10 ng/m^3, and the total organic acids in $PM_{2.5}$ accounted for 3%–15% of the organic carbon. These organic acids in the atmosphere were mainly formed through secondary photo-reactions, as well as vehicular emissions and coal combustion (Huang et al., 2005). The organic acids which contribute to the acidity in rain have been discussed mainly in terms of their sources and transformation mechanisms. The formic and acetic acid in rainfall ranged from 1.5 to 30 and 1.5 to 20 mol/L, organic acids originating from natural, anthropogenic sources and chemical transformation of organic matter in aerosol (Yu, 2000; Sigha-Nkamdjou et al., 2003; Kawamura et al., 2007). In addition, soil and ants' release and oceanic aerosol discharge all contributed to organic acids in the atmosphere (Khare et al., 1997; Kesselmeier and Bode, 1997; Li et al., 2000).

A homologous series of α,ω-dicarboxylic acids (C_2–C_{10}), ω-oxocarboxylic acids (C_2–C_9), pyruvic acid, and α-dicarbonyls (C_2–C_3) were measured in Greenland ice core samples using capillary gas chromatography and mass spectrometry. Concentration ranges of the total diacids, ω-oxoacids, and α-dicarbonyls were 3.1–32.5 ng/g, 0.13–2.8 ng/g, and 0.09–1.7 ng/g, respectively. Relative abundances of diacid carbon contents in total organic carbon