Impact of Biogeochemical Cycling of Phytoplankton-Produced Lipid Compounds in Ocean Systems on Paleoceanographic Records Buried in Sediments

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Deciphering historic records buried in ancient marine sediments is critically important for understanding the past and current changes in environmental and climate conditions of the earth. Many molecular approaches, including phytoplankton-produced lipid compounds and their stable isotopic compositions, have been developed to conduct this mission [1-4]. Applications of these approaches are based on a presumption that once these chemical and isotopic signals are generated by phytoplankton in surface water, they remain intact until ultimate burial in sediments. However, most (>95%) phytoplankton-produced organic compounds are recycled by biological and biogeochemical processes in water and surface sediments [5-8]. Therefore, a logical question has been raised on whether or how these molecular signals are altered during transport/recycling processes.

The first example is the record of paleo-surface water temperature using alkenone-based index Uk-37° [9,10]. Although the variability in the Uk-37° index is largely dependent on the temperature in surface water at which the algae grow, other factors have been recognized to affect the index. For example, cell growth rate or growth stage may play a role along with temperature in controlling the unsaturation degree of alkenones [11]. Alkenones are disproportionately synthesized by Prymnesiophyceae cells either as membrane compounds or as metabolic energy storage compounds during different growth stages [12,13], leading to potential deviations (up to 3°C) in the estimated temperature [14-17]. Grazing activities of zooplankton and benthic fauna seem to have little impact on alkenone degradation [18-20], but microbial processes play a dominating role in alkenone degradation [21,22]. Contrasting effects of microbial degradation of alkenones on the Uk-37° index have been observed in different systems or under different environmental conditions, which are likely due to involvement of different microorganisms [23], different degrading reactions [24], and variable redox conditions [25].

The second example is to determine paleo-CO₂ level using alkenone δ¹³C signals, based on a presumption that isotopic composition of one single alkenone compound preserved in sediments is linked to carbon isotopic fractionation (ε) of phytoplanktonic organic matter while a simple linear relationship exists between CO₂ concentration and the (ε) [26,27]. This simple linear relationship was challenged by a finding that cell growth rate also affects isotopic fractionation of phytoplanktonic organic matter and the (ε) is strongly related to the ratio of growth rate to CO₂ concentration (μ/C) rather than [CO₂] alone [28,29]. More studies [30-33] have documented that nutrient-limitation (either nitrate or phosphate) and Fe-limitation control the cell growth rate and thus affect isotopic fractionation. On the other hand, many field observations and laboratory experiments [34-36] have repeatedly shown strong evidence on isotopic depletion (negative shift) of alkenones during transport/recycling. Interpretations on isotopic alterations of alkenones during transport/recycling have been controversial and various hypothesized mechanisms have not been clarified. There are two major problems for interpretations of negative isotopic alteration: (1) isotopic fractionation of compounds during degradation cannot cause a negative alteration due to a kinetic factor; and (2) dilution effect of isotopic fractionation (even for positive alteration) can minimize the isotopic alteration for lipid compounds with a longer (e.g. >C20) chain length. Probably, isotopic alteration of organic compounds during degradation is not necessarily caused by kinetic fractionation. Instead, combined impacts of heterogeneity in isotopic signals within molecular and cellular structures, which is related to cell physiological state, and selective degradation may lead to an alternative mechanism to change isotopic signals of lipid compounds.

The third example is application of lipid biomarker hydrogen isotopic composition in determining historic changes in water hydrological cycle. Some studies [37,38] found that δD values of sterols and alkenones were well correlated with water D/H ratio as well as salinity, which can be used to reconstruct ancient records of influx of fresh water from rivers into oceans under varying climatic conditions. Like carbon isotopic signals, compound-specific hydrogen isotopic compositions of lipid biomarkers are affected by cell growth rate, induced by nutrient-limitation [38,39] further revealed that sterols from the nitrogen limited culture were significantly enriched in deuterium relative to the nitrogen-replete culture while fatty acids from both cultures had similar δD values. The difference in D/H fractionation between sterols and fatty acids was attributed to different synthesis pathways for acetylgenic (linear) and isoprenoid (branched) compounds [39]. There is evidence [40] showing that δD values of different lipid compounds are differentially altered during degradation processes.

Although much progress has been made at understanding how physiology of phytoplankton and environmental conditions affect isotopic fractionation of organic matter (and organic compounds) during photosynthesis, significant uncertainties still exist concerning the effect of biogeochemical cycling on isotopic composition of individual lipid compounds [41]. The following lists several significant points that need to be further considered: (1) effect of cell physiological state on generation of chemical and isotopic signals; (2) variability in proportions of lipid compounds in different cellular compartments along cell growth stages; (3) heterogeneity in chemical and isotopic compositions within different molecular and cellular structures; (4) relative labilities of various compounds bound in different cellular

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structures during biogeochemical cycling; and (5) relative importance of cell respiration, animal’s grazing, and microbial decomposition on stability of signals.

References