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厦门大学

硕 士 学 位 论 文

阿拉伯海岩心氮同位素记录及其对末次冰期以来氮循环过程的指示

Nitrogen Isotopic Records in Sediment Cores and Their Implications to Nitrogen Cycle History of the Arabian Sea since the Late Glacial

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摘要

海洋氮循环与众多元素生物地球化学循环过程、海洋生产力及全球气候变迁联系紧密。阿拉伯海作为全球三大缺氧区之一，其反硝化作用对海洋氮移除有着显著贡献，是古海洋氮循环研究的热点区域。本论文运用元素分析－同位素质谱联用(EA-IRMS)技术测定采自阿拉伯海东南部陆坡区的重力岩心 SK177/11 中不同组分碳氮含量及其稳定同位素组成，结合阿拉伯海已发表的氮同位素资料，旨在(1)了解颗粒有机物从生产、沉降、到埋藏过程中氮同位素信号的传递、转化及含义；(2)分析固氮和反硝化强度在冰期－间冰期的相对变化及调控机制；(3)探讨阿拉伯海南北部的氮循环过程空间差异与演化历史。

岩心 SK177/11 记录显示近 3.5 万年(35 ky BP, kilo year before present)来总氮(TN, Total Nitrogen)含量变化范围为 0.23~0.75%，其中有机氮占有率大于 80%，其次为酸可溶出氮(LN, Leachable Nitrogen, 0~20%)和结合态氨(FA, Fixed Ammonium, < 4%)。有机氮同位素值与总氮同位素值非常一致，介于 4.7~7.1‰之间。δ^{15}N-FA 值变化范围为 2.5~7.6‰，然其占有率小，对 δ^{15}N-TN 影响极小。LN 占有率最高虽达~20%，但 LN 来自有机氮的降解转化，仍属于原始 TN 的一部分。故 TN 可代表初始有机氮，δ^{15}N-TN 反映有机颗粒埋藏的初始信号。

自 3.5 万年前以来总有机碳(TOC, Total Organic Carbon)含量为 2~6%，与 TN 的变化趋势十分一致。δ^{13}C-TOC 的值-21.5~-18.5‰似乎都落在海源有机物 δ^{13}C 端元值-22~-18‰范围内，但 TOC/TN 摩尔数比为 7.5~13.6，偏离 Redfield 比值，表明该有机物可能为海源有机物和陆源性 C3、C4 植物来源有机物的混合物。运用三端元混合模型计算得出海源有机碳占 TOC 比例为 48~90%，海源有机氮占 TN 比例为 82~98%；而冰期 C4 植物相对 C3 贡献量大，冰消期则 C3 比例更高。根据氮同位素质量守恒原理，推算出海源 δ^{15}N 值与 δ^{15}N-TN 趋势一致，前者略高于后者，二者平均差值为 0.35±0.17‰。综上所述，SK177/11 的 δ^{15}N-TN 基本可以反映海源有机氮 δ^{15}N。

阿拉伯海以往的资料显示北部真光层底部约 100 m 处 δ^{15}N-NO_3^-值与沉积物捕获器的沉降颗粒 δ^{15}N-PN 最为相近，暗示该层为沉降颗粒营养盐来源。而沉积物捕获器对应站位的表层沉积物 δ^{15}N-TN 总是显著高于沉降颗粒 δ^{15}N-PN，表明

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有机氮同位素信号在沉积后发生同位素正偏移。搜集报道的大面积范围的表层沉积物 δ^{15}N-TN 数据，发现沉积物 δ^{15}N-TN 与底深之间存在良好且相异的线性关系(北部 $\delta^{15}N = 0.55\ (\pm 0.08) \times 10^{-3} \times Depth + 8.12\ (\pm 0.18)$，$R^2 = 0.40$，n = 78，$p < 0.0001$；南部 $\delta^{15}N = 0.76\ (\pm 0.14) \times 10^{-3} \times Depth + 6.03\ (\pm 0.33)$，$R^2 = 0.66$，n = 17，$p < 0.0001$)，即 δ^{15}N-TN 随底深增大而变大，可能与有机质暴氧时间有关。斜率 *k* 在北部较低，表明缺氧较严重的北部海区的底深效应弱于相对有氧的南部。利用 *k* 对底深效应引起的 δ^{15}N-TN 值偏差进行修正(即将底深统一设定为 100 m)后，北部表层沉积物 δ^{15}N-TN 的范围集中在 6~9‰之间，与真光层底部 $\delta^{15}N\text{-}NO_3^-$ (7~9‰)较一致，表明底深效应修正后，沉积物总氮同位素信号可反映真光层底部氮同位素源。

岩心 SK177/11 及阿拉伯海多数报道岩心记录都显示由冰期向间冰期过渡阶段，δ^{15}N-TN 值明显增大，与另外两大缺氧区(东赤道北太平洋 ETNP 和东赤道南太平洋 ETSP)的情况相似。分气候阶段拟合底深效应方程，δ^{15}N-TN 值经修正后显示在冰期阶段，南北的 δ^{15}N-TN 并无差异，且接近全球海洋 $\delta^{15}N\text{-}NO_3^-$均值 4.5~6‰，但南、北部差异自冰消期以来总体增大，并有扩大趋势，表明阿拉伯海冰期氮循环受全球氮循环驱动，而全新世和现代则可能多受局部过程影响。尤其北部岩心 δ^{15}N-TN 在晚全新世持续增大，与南部和 ETNP 及 ETSP 在晚全新世呈现的下降趋势相反，似乎暗示北部逐渐增强的反硝化引发了南部海域较强的固氮作用，致使次表层 $\delta^{15}N\text{-}NO_3^-$差异逐渐扩大，即固氮与反硝化作用存在的空间耦合关系。造成以上现象的原因可能是水流交换在冷暖期格局不同，同时影响了氧气供应及不同氮磷比的营养盐的分布，从而引发固氮与反硝化作用之间的反馈响应。

关键词： 氮循环；阿拉伯海；沉积物氮同位素；固氮作用；反硝化作用

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Abstract

Nitrogen cycle in the ocean is tightly related to the biogeochemical cycle of various elements, marine primary productivity and climate changes. As one of the three largest oxygen deficient zones (ODZs) in the world, Arabian Sea (AS), where denitrification contributes remarkablely to the nitrogen losses, is a hotspot for the past nitrogen cycle study. In this study, a sediment core (SK177/11) was collected from slope of southeastern AS. Core SK177/11 was subsampled and analysed for C and N contents of different components as well as their stable C and N isotopes through Elemental Analyzer-Isotopic Ratio Mass Spectrometer (EA-IRMS). This study compiled results from the above analysis with $\delta^{15}N$ records from previous studies in AS, and aims to (1) understand the transfer and transformation of the nitrogen isotope signal during the production, sinking and burial of organic particles, and its implication; (2) constrain the role of N_2 fixation and denitrification since the Late Glacial and its regulation mechanism; and (3) unveil the spatial difference and evolution of nitrogen cycle in the southern and northern parts of AS.

Total nitrogen (TN) content in core SK177/11 had a range of 0.23~0.75% since 35 ky BP (kilo year before present), in which organic nitrogen shared more than 80%, and 0~20% was leachable nitrogen (LN), and less than 4% was fixed ammonium (FA). Values of $\delta^{15}N$ of organic nitrogen were consistent with the $\delta^{15}N$-TN, ranging from 4.7‰ to 7.1‰. Values of $\delta^{15}N$-FA were within 2.5~7.6‰. Considering the low proportion of FA in TN, FA had little effect on $\delta^{15}N$-TN. Despite the relatively high ratios of LN/TN, LN was the degradation product of organic nitrogen. TN therefore was part of the original TN. Thus, the burial TN maintained the initial N isotopic signals.

Total organic carbon (TOC) content ranged from 2% to 6%, with a trend similar to TN since 35 ky BP. It seemed that values of $\delta^{13}C$-TOC (−21.5~−18.5‰) were consistent with the $\delta^{13}C$ of marine organic matter end-member (−22~−18‰). However, molar ratio of TOC/TN was 7.5~13.6, drifting away from the Redfield ratio, implying a mixture of marine organic matter and terrestrial C3 and C4 plants. Based on a three end-member mixing model, the proportion of marine organic carbon in TOC was 48~90% and marine organic nitrogen in TN was 82~98%. Contributions of C3 and C4 plants to TOC were different in each climate stage, showing relatively high contribution from C4 plants during the glacial but higher from C3 plants than from C4 plants during the deglacial. Furthermore, based on nitrogen isotopic mass balance, the calculated $\delta^{15}N$ of marine organic nitrogen was slightly higher (0.35±0.17‰) than $\delta^{15}N$-TN. However, both of them had similar down-core trends. In summary, $\delta^{15}N$-TN of core SK177/11 can be indicative of the $\delta^{15}N$ of marine organic nitrogen.

The available data showed that values of $\delta^{15}N$-NO_3^- at the bottom (~100 m) of euphotic zone were similar to $\delta^{15}N$ of sinking particles ($\delta^{15}N$-PN). It was likely that the source of nutrients for organic matter was largely derived from the depth of around 100 m. Values of $\delta^{15}N$-PN were obviously higher than $\delta^{15}N$-TN of surface sediments in surrounding areas, implying a positive shift of $\delta^{15}N$ in organic nitrogen at the surface of sediment. From previously published data in wide area of AS, we found obvious, but disparate linear relationships between the $\delta^{15}N$-TN of surface sediments and bottom depth in northern and southern AS (for northern AS, $\delta^{15}N$ = 0.55 (±0.08) × 10^{-3} × Depth + 8.12 (±0.18), R^2 = 0.40, n = 78, $p < 0.0001$; for southern AS, $\delta^{15}N$ = 0.76 (±0.14) × 10^{-3} × Depth + 6.03 (±0.33), R^2 = 0.66, n = 17, $p < 0.0001$). It meaned that values of $\delta^{15}N$-TN increased with the deepening of the bottom depth, probably as a result of oxygen exposure time of the organic matter. The bottom-depth effect, reflected by the slope (*k*), seemed less effective in northern AS, where oxygen deficiency in water column was more intensive. As corrected from the bottom-depth effect by *k* (fixed all bottom depths at the value of 100 m), most of the $\delta^{15}N$-TN values of surface sediments in northern AS were in the range of 6~9‰, much closer to the $\delta^{15}N$-NO_3^- (7~9‰) in bottom layer of euphotic zone. As a result, the nitrogen isotopic signal in sediments could be representative of the value of $\delta^{15}N$-NO_3^- at the bottom depth of euphotic zone.

Similar to other intense ODZs (e.g. Eastern Tropical North Pacific: ETNP, and Eastern Tropical South Pacific: ETSP), time series of $\delta^{15}N$-TN in core SK177/11 and

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published $\delta^{15}N$ records of other cores from AS displayed a sharp increase during the deglacial period. The $\delta^{15}N$-TN values in the whole AS during the glacial, corrected by bottom-depth effects of different climate stages, were close to mean $\delta^{15}N$-NO_3^- values (4.5~6‰) in the world deep ocean. However, the offsets of $\delta^{15}N$-NO_3^- between southern and northern AS expanded since the last deglaciation, which indicated that nitrogen cycle in AS was driven by global nitrogen cycle during the glacial, while it was more likely to be influenced by local processes during the Holocene and in modern time. In particular, $\delta^{15}N$-TN values of northern cores continue to rise during the late Holocene; however, an inverse trend was found in southern AS cores and in ETNP and ETSP. It seemed that the intensified denitrification in northern AS stimulated the increased N_2 fixation in southern AS, resulting in the greater differences of $\delta^{15}N$-NO_3^- in subsurface between this two parts. That was in support of the view that spatial coupling of nitrogen inputs and losses in the ocean. We inferred that the phenomena mentioned above might be attributed to altered patterns of water current during warm and cold periods. This could affect the supply of oxygen and the distribution of nutrients, and thus initiate the feedback between N_2 fixation and denitrification.

Key words: Nitrogen cycle; Arabian Sea; Sedimentary nitrogen isotope; N_2 fixation; Denitrification

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