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东海内陆架有机碳的源—汇过程及其沉积记录

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摘要 大陆边缘海是不同来源、不同性质有机碳沉积和埋藏的主要场所,在全球碳的生物地球化学循环过程中具有重要地位。东海内陆架接收大量陆源有机碳,并且具有较高的海洋生产力,是研究沉积有机碳来源、输运和埋藏的理想场所,已取得大量研究成果。在对相关文献进行系统整理的基础上,以沉积学的视角对前人研究成果进行了梳理,旨在为后续相关研究提供参考。全样分析(如 TOC/TN、 $\delta^{13}\text{C}$ 等)和生物标志化合物(如正构烷烃、甾醇类、木质素等)方法揭示东海内陆架有机碳的来源受沉积环境影响,由海向陆方向陆源组分整体显著增加,并具有季节性特征。东海内陆架沉积物在沿岸方向具有“夏储冬输”的宏观输运格局,该动力过程影响着陆源有机碳沿岸的输运路径和最终归宿;在东海 29° N 附近存在一个“舌形”的跨陆架输运通道,可能会存在陆源有机碳的跨陆架输运,影响深海有机碳的源—汇过程;另外,人类活动和极端气候事件也显著影响东海内陆架沉积物和有机碳的沉积过程和沉积记录,需要进一步研究。东海内陆架泥质区是陆源有机碳的重要埋藏区域,其埋藏效率受区域沉积有机碳含量和沉积速率控制,并与早期成岩过程中有机质矿化路径有关。沉积物中埋藏有机碳的地球化学特征可以用来重建长时间尺度的海平面变化、初级生产力、古海洋和古气候演化等,相关研究为理清东海内陆架地质历史时期的环境演化提供了依据。

关键词 东海内陆架;有机碳;早期成岩;跨陆架输运;沉积过程

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0 引言

大陆边缘海作为陆地与大洋的连接过渡地带,虽然只占海洋表面积的 20% 左右,却储存了海洋沉积有机碳的 80% 左右,强烈影响全球碳的生物地球化学循环^[1-4]。陆架地区往往受到河流输入陆源物质的影响,表现出陆源物质输入高、沉积速率快的特征^[5-7]。此外,河口和陆架区域还受到洋流、波浪和潮汐等水动力过程的影响(图 1),导致其沉积环境复杂,内陆架陆源沉积物和有机质的命运也不尽相同,一直是海洋沉积学研究的热点科学问题^[9-11]。深入研究内陆架区域内沉积有机碳的来源、输入、埋藏过程及其沉积记录对理解全球气候变化、有机碳埋藏与矿化以及海洋生态系统具有重要意义^[9,12-13]。

另外,有机碳作为沉积地球化学研究的常用指

标,尤其是有机碳的同位素和元素特征(例如 $\delta^{13}\text{C}$ 、TOC/TN),常用于恢复地质历史时期中的海平面变化、古环境重建、古气候和古海洋演化等^[14-16],但是这些指标在实际应用过程中往往受到多种因素影响,需要现代过程的约束。例如,TOC/TN 虽然经常被用来区分陆源和海源有机质,但是 TOC/TN 的比值会受到微生物选择性降解、输运过程中有机质氧化、沉积物早期成岩作用、近海沉积物无机氮吸附以及人类活动(如石油污染、施肥)的影响^[17-22]。海洋浮游生物生产的海源有机碳 $\delta^{13}\text{C}$ 变化范围较大,并且 $\delta^{13}\text{C}$ 值还会受到早期成岩作用的影响而使准确性降低^[23-25]。因此,对现代海洋环境有机碳来源、输运和埋藏过程的研究有助于正确解读地层记录内有机碳信息,起到“将今论古”的作用。

东海是典型的以河流输入为主导的边缘海^[26],同

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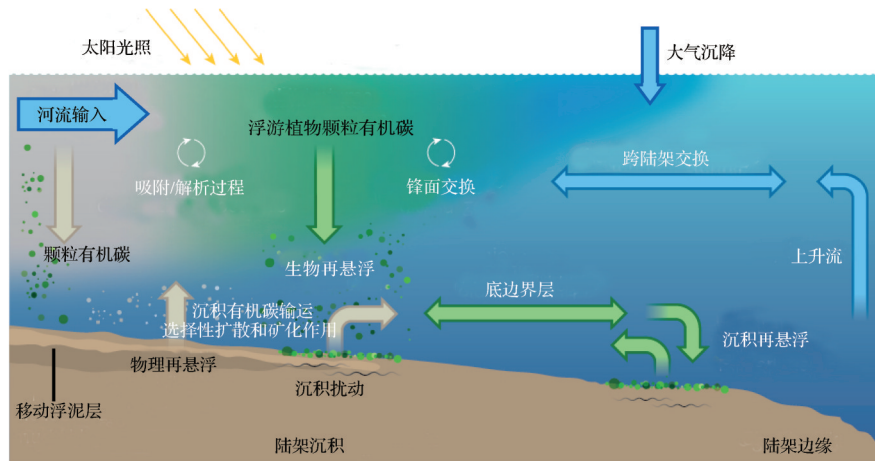


图1 边缘海沉积有机碳循环模式图(据文献[8]修改)

Fig.1 Schematic of sedimentary organic carbon cycle in marginal sea (modified from reference [8])

时也是有机质运输、沉积和碳循环最活跃的海域之一^[27-30]。以长江为主的河流携带巨量沉积物入海,使得东海内陆架沉积有机质陆源占比明显^[5,31-32]。而受西边界流和季风的影响,东海水动力条件复杂,在内陆架近岸区域形成了“夏储冬输”的宏观运输格局^[33],此外在29°N附近还存在“舌形”跨陆架输运通道^[34-36]。东海内陆架虽然具有很高的沉积速率,但有机碳保存率并不高^[6],主要原因包括沉积过程中强烈的物理扰动以及沉积后早期成岩过程中有机碳的消耗^[37-39]。综上所述,东海尤其是内陆架地区陆源输入丰富、沉积动力过程不稳定、有机质矿化过程复杂,十分适合开展边缘海有机碳沉积过程和沉积记录的研究,并且取得了大量的优秀研究成果^[30,40-45]。本文旨在梳理近年来东海内陆架沉积有机碳来源、输运和埋藏的新成果,以期为解读该地区沉积有机碳的沉积过程及沉积记录提供参考。

1 东海内陆架海洋环境背景

东海是世界上最大的边缘海之一,连接欧亚大陆板块和太平洋板块,属于典型的被动大陆边缘,其内陆架比较宽阔,平均水深60 m,宽度大约500 km^[46]。东海内陆架受长江营养丰富淡水输入以及台湾暖流入侵的影响^[47],初级生产力水平较高(108~997 mg m⁻² d⁻¹,平均值为425 mg m⁻² d⁻¹)^[48],构成沉积有机质的海洋端元。此外,东海内陆架接收大量来自河流的陆源物质(包括有机碳),构成沉积有机碳的另一个重要来源,其中长江输入是主要的物质来源之一^[41-42,44]。

长江是世界第三大河流,长约6 300 km,流域面积为1.9×10⁶ km²,在亚洲季风气候控制下,产生并携带巨量的沉积物,每年向东海输送(2~5)×10⁶ t陆源有机碳^[5,49],对流域生态环境和边缘海的海洋环境产生重大影响。此外,东海沉积物的来源还包括浙闽沿岸山地型河流(如钱塘江、甌江及闽江)和台湾河流(如浊水溪)输入的陆源物质(表1)。

前人研究表明东海内陆架沉积过程受海洋水动力条件的控制^[5,51-52]。东海内陆架具有独特而复杂的环流体系,主要有长江冲淡水(CDW)、浙闽沿岸流(ZMCC)、台湾暖流(TWC)和黑潮(KC),它们共同影响着东海沉积物的迁移、分布和沉积(图2)。高温、高盐的台湾暖流终年沿着浙闽海岸线外50~100 m等深线向东北方向流动,并沿着水下峡谷侵入长江口,年平均流速为14 cm s⁻¹^[54-55]。黑潮是太平洋西部的强边界流,具有高温、高盐、高流速的特点,起源于菲律宾以东海域,流经我国台湾东岸、东海东部,向东北

表1 东海主要输入河流概况(据文献[50]修改)

Table 1 Basic parameters of the major rivers entering the East China Sea (modified from reference [50])

河流	面积/10 ³ km ²	长度/km	最高海拔/m	流量/km ³ yr ⁻¹	TSS/Mtyr ⁻¹
长江	1 800	6 300	5 100	900	470
浙闽河流					
钱塘江	42	490	1 000	31	4.4
甌江	18	390	1 900	19	2.7
闽江	61	580	>1 000	58	2.4
台湾河流					
浊水溪	3.1	190	3 400	6.1	38

注:TSS-悬浮沉积物。

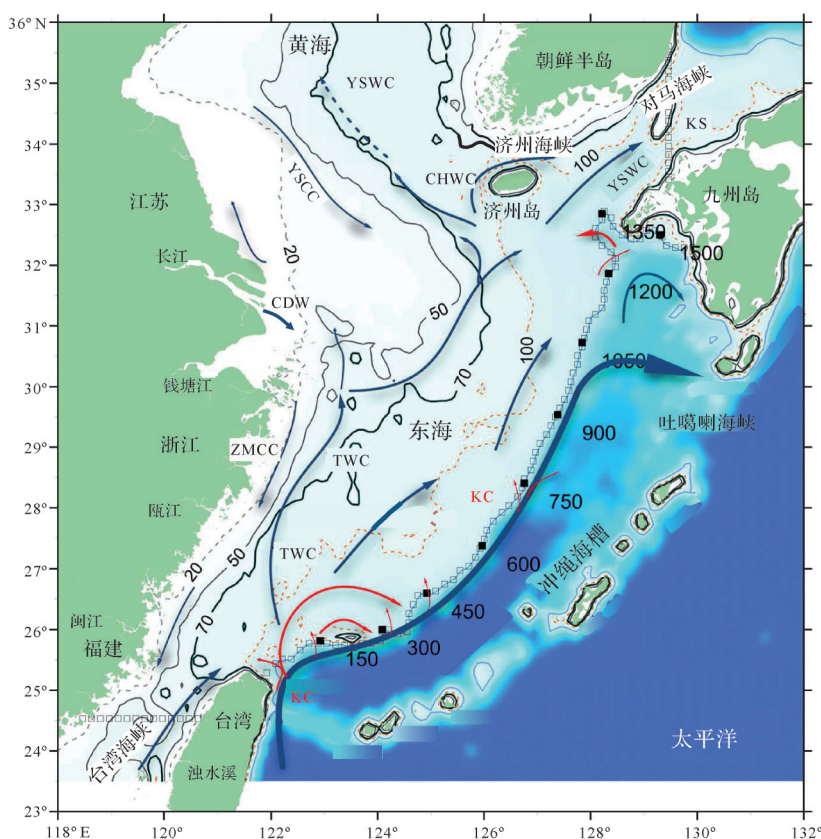


图2 东海及相邻海域洋流格局(据文献[53]修改)

YSCC:黄海沿岸流,CDW:长江冲淡水,ZMCC:浙闽沿岸流,TWC:台湾暖流,KC:黑潮,TSWC:对马暖流,CHWC:济州暖流,YSWC:黄海暖流

Fig.2 Current pattern in East China Sea and adjacent waters (modified from reference [53])

流至30°N时受到九州岛的阻隔,黑潮左侧的部分水流分离成对马暖流,其余水流东转流出东海^[56-57]。在28°N左右,黑潮次表层水可以侵入到浙闽沿岸,最远可以到达50m等深线处,进而影响东海内陆架水动力系统^[55,57]。

2 东海内陆架沉积有机碳来源

陆源与海源构成了内陆架沉积有机碳的主要来源,对于不同来源有机碳的物源追踪有多种手段,其中常用的经典指标有有机质的元素比值(TOC/TN)、稳定碳同位素($\delta^{13}\text{C}$)等^[19]。除此之外,生物标志化合物(如正构烷烃、甾醇类、木质素等)由于具有更加敏感与直观的优势,近些年来得到了广泛应用^[5,58-61]。而基于微生物四醚膜的支链和类异戊二烯四醚指标(branched and isoprenoid tetraethers, BIT)与杂环丁烷多元醇(Bacteriohopanepolyols, BHPs)则可以对边缘海表层沉积物内有机质来源进行更加精细的区分^[62-63]。

Kao *et al.*^[64]对比分析了东海内陆架和冲绳海槽南部表层沉积物中有机碳含量及其同位素组成,发现高有机碳含量区域呈现从长江口向南延伸,并由近海转向冲绳海槽南部的趋势,并且冲绳海槽南部沉积物的 $\delta^{13}\text{C}$ 值与内陆架沉积物接近,由此提出冲绳海槽南部的大部分沉积有机质可能源自东海内陆架。基于 $\delta^{13}\text{C}$ 等指标,河流、三角洲和海洋是长江口地区沉积有机碳物质来源的三个端元,并且有机碳来源会受到水动力过程的影响^[65]。蒙特卡罗模拟表明,东海内陆架超过50%的TOC来自于海洋端元,其次是三角洲端元^[66]。而利用长链烷烃和甾醇类生物标志化合物对东海内陆架表层沉积物中有机碳进行物源分析,发现长江口附近以陆源为主并随着离岸距离的增加占比减小,长江口外的上升流区及浙闽沿岸区以海源有机碳为主^[31]。Yao *et al.*^[5]利用多种指示因子来研究有机碳的来源、分布和组成,但是不同指示物呈现出了不同的分布、来源,表明了综合使用多种指示因子分析的必要性;基于支持大量生物标志化合物数据的PCA-MC模型,发现长江口及其邻

近陆架表层沉积物中,海洋、土壤和陆地有机碳的平均占比分别为35.3%,47.0%和17.6%。此外,东海内陆架有机碳分布还有明显的季节性,对比分析春、秋两季东海内陆架TOC含量、TOC/TN和 $\delta^{13}\text{C}$ 值,

Zhou *et al.*^[32]发现春季平均TOC含量高于秋季,而TOC/TN和 $\delta^{13}\text{C}$ 值相差不大;以 $\delta^{13}\text{C}$ 为标志物作二元(陆源与海源)模型分析,发现东海内陆架表层沉积物中0~34.6%的沉积有机质为陆源(图3)。

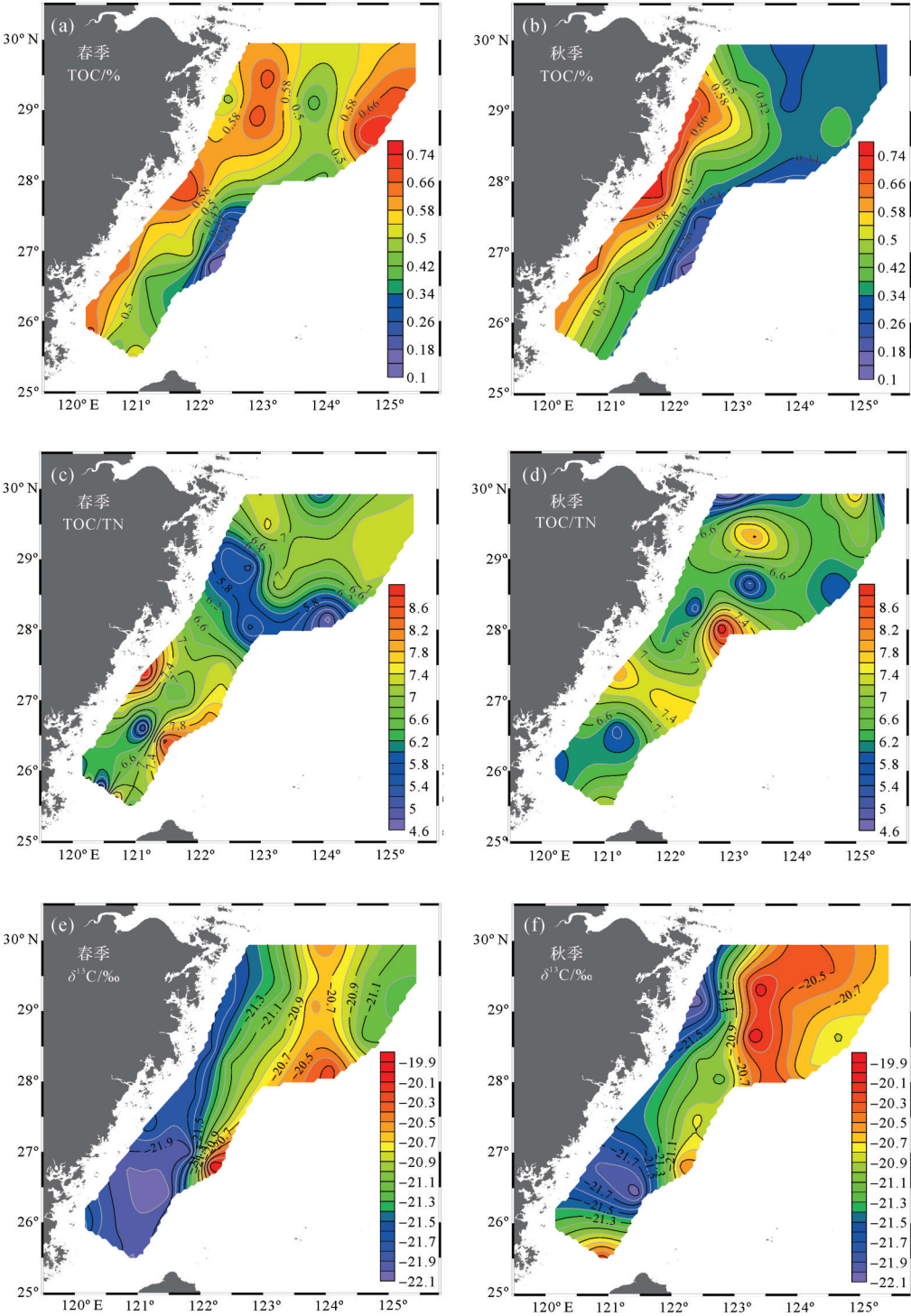


图3 东海内陆架TOC含量(a)(b)、TOC/TN(c)(d)和 $\delta^{13}\text{C}$ 值(e)(f)春、秋分布图(据文献[32]修改)

Fig.3 Distribution maps of TOC content, TOC/TN and $\delta^{13}\text{C}$ on the inner shelf of the East China Sea in Spring and Autumn (modified from reference [32])

如前所述,利用全样分析的方法判断有机碳来源和比例难免存在偏差,而相较而言,生物标志化合物方法具有明显优势,可以更准确地区分有机碳物源信息。但是生物标志化合物方法也存在自身的弊端,如正构烷烃会受到浮游生物中丰富的硅藻、颗石藻以及细菌输入和细菌改造作用的影响,使其具有不明显的奇偶优势,混淆其物源指示意义^[67];甾醇化合物的差异虽然能够指示物源,但是沉降过程中甾醇的细菌加氢还原作用也可以产生甾醇,由此会影响其准确性^[68-69]。因此,利用多重示踪方法和多端元模型可以更加准确细致地甄别有机质来源。

3 陆源有机碳的沉积和输运过程

陆源有机质在边缘海的散布过程是有选择性的,受水动力条件的影响,并主要受颗粒物粒径、密度、比表面积等因素控制,即粒径细、密度低的颗粒物可能会随着波浪和沿岸流等迁移输运到距离较远的海域,而粗颗粒、高密度的颗粒物则会优先沉积在河口附近^[70-72]。而在这一过程中,有机质还会受到波浪和海流的动力分选作用,不同来源、年龄和活性的物质受侵蚀、搬运和再悬浮的影响而发生分选或分异,这个过程也使得吸附在颗粒上的不同来源和性质的有机碳重新分配(图4),进而影响沉积有机碳在边缘海的分布与埋藏^[70-71,74-75]。

3.1 沿岸沉积输运过程

长江口及邻近的东海内陆架是世界上最大的大河三角洲前缘河口之一,其复杂的水动力环境直接导致此处复杂的有机碳输运过程。长江输送来的陆源有机碳在长江冲淡水、沿岸流、风暴以及上升流等因素的共同作用下,形成了沿浙闽沿岸向南的输送路径,只有很少一部分从河口向北或离岸输运^[52,76]。长江三角洲地区物质输运具有明显的季节性特征:夏季,受夏季风影响,沿岸流减弱,北向的黑潮水系增强,同时长江达到汛期,长江冲淡水增强,在这些条件的共同作用下长江输入的有机质大部分优先堆积在123°E以西的长江口区^[33,77];冬季,受冬季风影响,已经沉积的物质发生再悬浮,悬浮物在浙闽沿岸流的驱动下向东南输运,此时东北流向的台湾暖流仍旧起到了隔挡作用,将陆源物质输运限制在了浙闽沿岸^[33,52,76-77]。由此,在中国东海形成了“夏储冬输”的宏观输运格局^[33],例如,冬季,增强的中国沿岸流将源自长江的细颗粒沉积物带入底层,与海岸线平行地向南输送物质,形成浑浊状态^[52]。长江口泥质区和浙闽沿岸的沉积有机碳具有明显 $\delta^{13}\text{C}$ 亏损和较高的TOC/TN值,表明这些地区沉积有机碳中陆源输入的显著贡献^[6]。来自内陆架和水下三角洲的细颗粒沉积物受到风和潮汐作用发生再悬浮,并受到水动力作用通过底层雾状层的夹带发生扩散,其中年龄较老的有机碳更容易发生较长距离的沿岸输运^[78]。

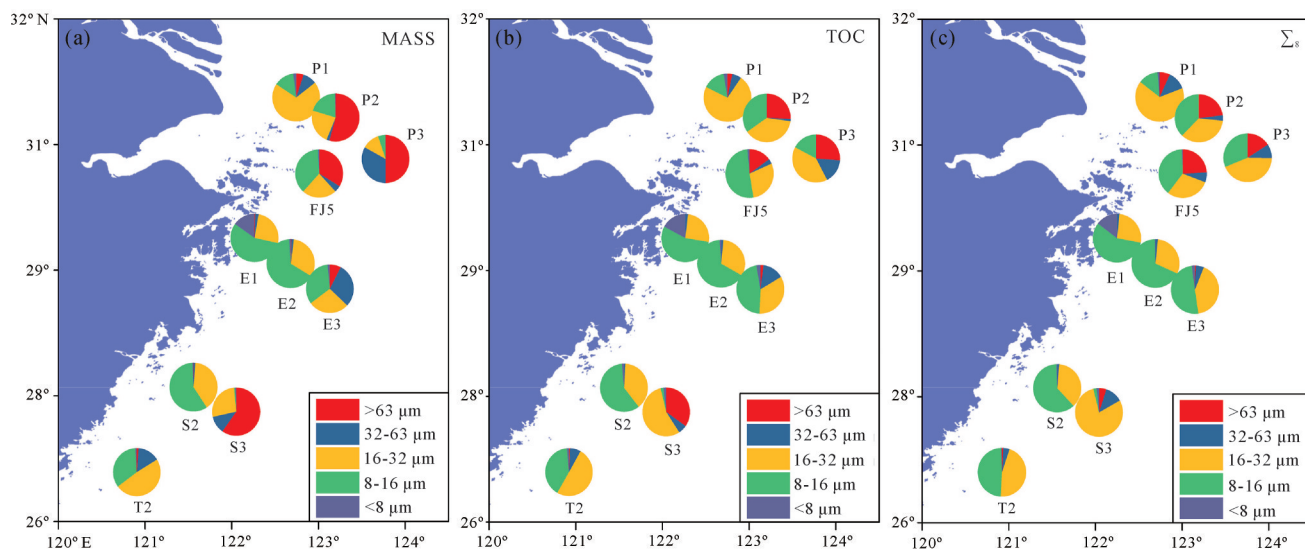


图4 沉积物粒度(a)、TOC(b)和 $\Sigma 8$ 木质素酚(c)不同组分所占比例分布图(据文献[73]修改)

Fig.4 Distribution of (a)different components of sediment size; (b) TOC;and(c) $\Sigma 8$ lignin phenol(modified from reference [73])

3.2 跨陆架沉积输运过程

跨陆架输运在海洋生态和物质循环中起着重要作用,它也被认为是陆源沉积物从源到汇输运的关键过程之一^[79]。在中国东海 30° N 以南的区域,盐分、微量元素和悬浮沉积物的分布表明存在大规模的跨陆架输运^[56,80-81]。Yuan *et al.*^[34]基于叶绿素 a 浓度卫星图像的分析,发现东海大陆架上存在物质的跨陆架输运,跨陆架穿透锋面呈舌形,它在近岸地区具有较宽的根系,并向海变窄(图 5)。跨陆架穿透锋面是指从诸如温度、盐度、叶绿素 a 浓度等海洋特性的平均锋面直接和大范围地入侵,穿透距离通常超过 50 km^[82]。郭志刚等^[33]对东海北部悬浮体分布研究发现,冬季在东海北部有部分陆架悬浮体输送入冲绳海槽,其输送的主要位置在 P-N 断面以北、32° N 断面以南区域,而夏季基本不存在这一跨陆架输运。Zhang *et al.*^[35]通过分析表层沉积物粒径和碎屑矿物成分(图 5),再次验证了舌形区域的存在,其轴线位于 29° N 附近,从海岸延伸到 124° 40' E 或更远,这个舌形区域的沉积物富含非常细的粉砂和粘土,有利于结合颗粒有机碳,因此可能伴随着有机碳的跨陆架输运。最近,对外陆架—斜坡—冲绳海槽表层沉积物的物源分析显示,斜坡和

冲绳海槽的陆源有机碳大部分是从东海内陆架输运而来,进一步证实了陆源有机碳跨陆架输运的存在^[83]。因此,陆源有机碳除了在内陆架沉积埋藏,还会通过跨陆架输运的方式进入到更深的海域,其中相关的输运机制和沉积动力过程亟待进一步研究。

3.3 人类活动与极端气候对有机碳沉积输运过程的影响

东海内陆架有机碳沉积输运过程除了受浪潮流等自然因素的影响外,人类活动(如河流水库建设、流域土地利用变化、海域富营养化等)的干扰强度也越来越大,对大陆边缘海有机碳循环产生了重要影响。其中,三峡大坝的兴建使得河流中下游悬浮泥沙含量和通量显著下降,颗粒有机碳(POC)含量从 1952—1986 年的 $10.9 \pm 2.6 \text{ Mt yr}^{-1}$ 下降到 2003—2008 年的 $1.9 \pm 1.0 \text{ Mt yr}^{-1}$ ^[84],并且河流流量的降低使得黑潮与台湾暖流的入侵加强,局部物理化学生态环境发生深刻改变,对区域有机碳循环产生了深远影响^[47,85-88]。Wang *et al.*^[89]对比三峡大坝蓄水前后有机碳含量和来源,发现 2018 年陆源有机碳沉积通量较 2006 年骤然减少近一半,三峡大坝的建设显著影响着东海有机碳的输运过程与沉积模式。

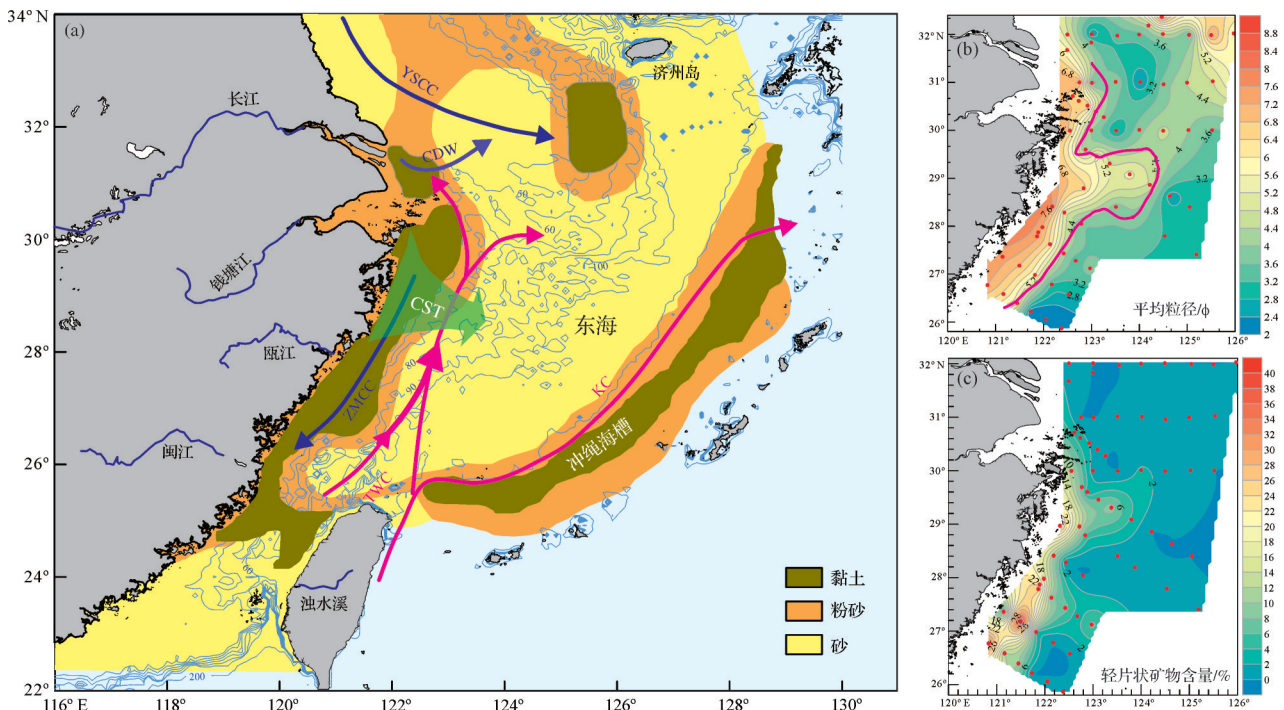


图5 沉积物粒径和矿物特征揭示的东海跨陆架输运(据文献[35]修改)(CST: Cross-Shelf Transport, 跨陆架输运)

Fig.5 Cross-shelf sediment transport in the East China Sea indicated by grainsize and mineral characteristics

(modified from reference[35])

除此之外,极端气候事件(如台风、洪水等)也是影响东海内陆架碳循环非常重要的因素。西北太平洋是热带气旋高发地之一,台风期间边缘海沉积物的输运状态发生巨大改变^[90-91],水体悬沙含量和输运率可数倍甚至10倍于平静天气,海底表层沉积物也因此发生再悬浮^[92-93],进而影响沉积物的埋藏和沉积过程。例如,2013年“潭美”登陆后闽江口溶解有机碳(DOC)和颗粒有机碳(POC)含量相比登陆前分别增加了169.3%和138.3%,对区域有机碳循环产生了深刻影响^[94]。

4 有机碳埋藏与再矿化

大陆边缘海虽然只占全球海洋面积的20%,但占全球海洋初级生产力的30%以上,占全球有机碳埋藏总量的87%,是碳埋藏的主要场所^[2,8,95]。沉积有机碳埋藏能力的大小与区域沉积有机碳的含量和沉积速率有关,具体可以通过以下方程式获得^[95]:

$$OCAR = (C \times LSR \times \rho \times (1 - \Phi))$$

有机碳堆积速率 (Organic carbon accumulation rates, 简称 OCAR) 单位为 $\text{g C cm}^{-2} \text{ yr}^{-1}$, C 为沉积物中的 TOC 含量 (%), ρ 为沉积物干密度 (g cm^{-3}), Φ 为孔隙率, LSR 为线性沉积速率 (cm yr^{-1}), 通常由 ^{210}Pb 测得。由于不同区域沉积物 TOC 含量和沉积速率不同, 有机碳的埋藏速率也有很大的差异 (图 6)。除此之外还可以利用生物标志化合物 (如木质素、叶绿素 a 及其降解产物等) 定性研究不同区域的有机质埋藏特点^[13]。

东海内陆架接收了来自长江的大量有机质,同时

具有适宜的水动力条件,是一个重要的碳埋藏热点区域^[7,96]。Deng *et al.*^[97]利用表层沉积物 TOC 含量、沉积速率以及干密度资料,研究分析了东海陆架不同区域 OCAR,估算得到东海陆架年平均 OCAR 为 $14.7 \text{ g C cm}^{-2} \text{ yr}^{-1}$,其中长江三角洲附近 OCAR 最高,超过 $200 \text{ g C cm}^{-2} \text{ yr}^{-1}$,离岸和向南方向逐渐降低;济州岛南面的泥质沉积区和台湾东北方向的东海陆架坡折处也具有较高的 OCAR ($>10 \text{ g C cm}^{-2} \text{ yr}^{-1}$),而中陆架砂质沉积区的 OCAR 较低 ($<6 \text{ g C cm}^{-2} \text{ yr}^{-1}$),陆架上有有机碳堆积可能主要受控于河流输入和黑潮入侵。东海陆架沉积物中有机碳的埋藏通量为每年 $7.4 \times 10^6 \text{ t}^{[97]}$,其中海源有机碳的埋藏量为 $5.5 \times 10^6 \text{ t}$,占东海浮游生物固碳量的 5.4%^[96]。Sun *et al.*^[7]对东海内陆架泥质区估算的 OCAR 范围为 $33.8 \sim 54.7 \text{ g C cm}^{-2} \text{ yr}^{-1}$ (平均 $41.2 \text{ g C cm}^{-2} \text{ yr}^{-1}$),这些值高于其他边缘海(如阿拉伯海、路易斯安那内陆架及波罗的海)的估计值,表明东海内陆架泥质区可能是沉积有机碳的汇。

潮汐、风暴、波流等物理过程、河流流量变化、底栖生物扰动等多种因素使得边缘海具有动荡的沉积动力环境,沉积物频繁地发生再悬浮和再改造,将已经沉积下来并被新的沉积物覆盖的有机碳再次暴露在氧化或次氧化环境而发生再矿化分解^[12,98]。在微生物作用下,海源有机碳降解的同时会由于“激发效应”的存在而促进难降解的陆源有机碳的降解,最终只有30%左右的陆源有机碳和10%左右的海源有机碳能被埋藏在底层沉积物中^[12,98-100]。长江口与亚马逊河口、巴布亚湾弗莱河口及密西西比河口等具有移动泥(mobile mud)区的大河河口相似(图6),都具有高沉积速率和低有机碳保存效率的特征^[6]。

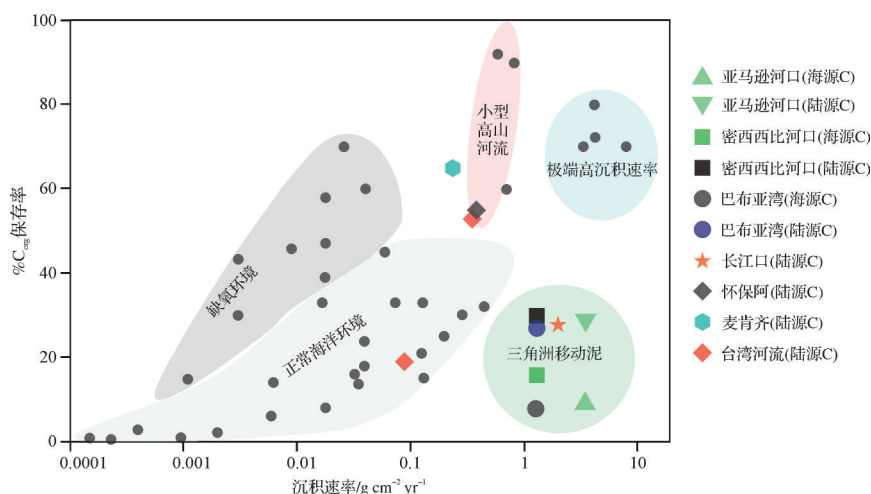


图6 不同沉积环境中有机碳的埋藏保存效率(据文献[18]修改)

Fig.6 Buried and preserved efficiency of organic carbon in different sedimentary environments(modified from reference [18])

在早期成岩过程中,有机碳矿化无疑是一个十分重要的过程,这一过程使得有机质经由再矿化转化成了无机形式,构成碳生物地球化学循环中的重要一环^[13,51,101-102]。有机质再矿化又包含了若干过程,在稳态环境中,按照吉布斯自由能大小,有机质矿化先后经过有氧呼吸、反硝化、锰铁氧化物还原、硫酸盐还原和 CO_2 还原等过程,构成沉积物剖面上理想的氧化还原序列^[102-103](图7)。在边缘海有机质矿化过程中,硫酸盐还原是有有机质矿化最主要的途径,可以占陆架边缘海沉积环境中有机质矿化的一半以上^[105-106]。此外,异化铁还原在东海内陆架泥质沉积物的成岩过程中也非常重要^[37]。但是在现实环境中,沉积物经常受到再悬浮、生物扰动、事件沉积等因素的影响,使得氧化还原带相互重叠或缺失,形成非稳态的沉积成岩环境^[107-109]。

目前常用的有机碳再矿化速率测定方法有两种,分别基于 O_2 消耗速率和 CO_2 产生速率^[98]。通过测定沉积物—水界面溶解氧含量,计算从上覆水向沉积物的 O_2 扩散通量,将其作为有机碳氧化所消耗的 O_2 速率,然后按照 $\text{C}_{106}:\text{O}_{138}$ 的比例将其转化成 CO_2 的产生速率,并以此来代表有机碳的再矿化速率^[110-111]。但是这种方法更适合静态沉积的大洋区域,对于水动力活跃的边缘海不合适^[13]。反之,通过培养的方法,测定沉积物样品 ΣCO_2 (DIC)的生产速率,可以用于研究边缘海有机碳的再矿化速率,甚至可以通过测定沉积物间隙水中 CO_2 的 $\delta^{13}\text{C}$ 进行物源分析,从而

区分不同物源有机碳的再矿化速率^[13]。Yao *et al.*^[28]研究发现长江口及浙闽泥质区沉积有机碳保存效率相比热带地区很低,长江口沉积物的全柱样(0~24 cm)孔隙水 ΣCO_2 的净反应速率为 $5.1 \text{ mmol m}^{-2} \text{ d}^{-1}$,上15 cm沉积物的净反应速率为 $9.3 \text{ mmol m}^{-2} \text{ d}^{-1}$,下层的消耗速率为 $4.2 \text{ mmol m}^{-2} \text{ d}^{-1}$,近似于主要阳离子的消耗速率,这意味着有机碳再矿化产生的 CO_2 推动了碳酸盐和粘土矿物的自生,促进了反风化作用^[112]。边缘海移动泥在有机碳再矿化方面也发挥了独特的作用,被称为有机质的“焚烧炉”^[113]。

除了原位观测和培养的方法可以示踪有机质矿化之外,沉积物环境磁学的信号也可以指示有机质矿化过程和成岩路径。有机质矿化过程伴随着磁性矿物的演化(图7)^[38-39],在铁氧化物还原阶段,亚铁磁性的氧化物(如磁铁矿)在还原环境中发生溶解,在异化铁还原菌的作用下溶解释放的 Fe^{3+} 被还原成 Fe^{2+} ^[37-38];在硫酸盐还原阶段, Fe^{2+} 与 H_2S 反应形成铁硫化物,其中在水溶液中最先出现的是四方硫铁矿,后又形成不稳定的磁黄铁矿和胶黄铁矿,最终形成还原条件下最稳定的黄铁矿^[38,114-115],以上演化过程反映了异化铁和硫酸盐还原过程对有机质的消耗。Kao *et al.*^[114]根据主要的磁性矿物组合(胶黄铁矿、磁黄铁矿和磁铁矿为主)对台湾西南部两个沉积序列划分了不同的地层带,发现胶黄铁矿和磁黄铁矿与细粒沉积物密切相关,而磁铁矿在粗粒沉积物中更为丰富,说明细粒物质有利于有机质参与的硫酸盐

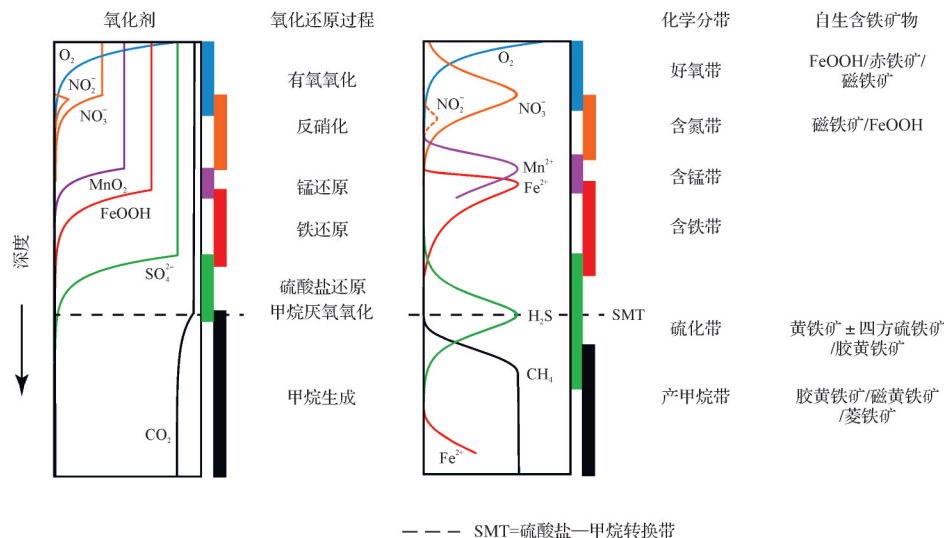


图7 理想状态下有机质矿化序列及其形成的氧化还原带和相关的自生矿物(据文献[104]修改)

Fig.7 Ideal sequence of organic matter mineralization, redox zone and related authigenic minerals

(modified from reference [104])

还原反应的进行。Zheng *et al.*^[39]对东海内陆架 MD06-3040 岩芯中的磁性矿物分析发现,从岩芯顶部到 3.5 m 亚铁磁性(磁铁矿)逐渐减少,可能受到异化铁还原的影响;在 8.45~9 m 处磁性矿物粒径变粗、磁性矿物含量下降,指示硫酸盐—甲烷厌氧氧化过程生成的硫化环境。有机质矿化过程会影响沉积物中磁性矿物成岩作用(图 7),尤其是会受沉积速率的影响^[115-117]。例如,东海内陆架泥质区内 MD06-3042 岩芯沉积物受到突变沉积速率的影响,形成了两层 SMT^[117],揭示沉积速率是控制有机质矿化和成岩过程的重要因素。

5 长时间尺度的沉积有机碳记录

有机碳作为地球化学研究的常用指标,特别是全样的 $\delta^{13}\text{C}$ 、TOC/TN 以及单分子组成和同位素特征常用于指示海平面变化、古环境重建、恢复海洋生产力和季风演化过程等多方面研究^[14-16,19]。古海平面的恢复通常是通过生物学和物理指标(如花粉,硅藻,有孔虫,颗粒大小等)相结合来实现的^[118-119]。例如,时小军等^[120]利用海南岛东部琼海的珊瑚礁记录恢复了中晚全新世高海平面,在距今 5 500~5 200 年前的海平面可能比如今海平面高出 2.0~2.2 m。但是在某些情况下,沉积物中微化石可能很少或不存在,此时有机碳的地化指标 $\delta^{13}\text{C}$ 、TOC/TN 便成为更好的选择^[15]。由于光合作用类型的不同,使得植物分为 C_3 、 C_4 和 CAM 三类群并具有不同的碳同位素组成,进而导致生物链及其衍生物碳同位素的不同,这就赋予了碳同位素重建某一地区或某一时期古环境的能力^[121]。Goslin *et al.*^[122]利用地化指标 $\delta^{13}\text{C}$ 、TOC、TN 在法国布列尼塔沼泽恢复的全新世海平面位置与使用传统古生物化石获得的位置高度吻合。对东海内陆架 EC2005 岩芯的研究也发现利用有机碳 $\delta^{13}\text{C}$ 和 TOC/TN 可以很好地区分环境的演化(图 8)^[123]。东海内陆架在 16.4 ka 以来从陆地环境转变到海陆过渡环境,并最终形成现代内陆架泥质沉积环境。但综合分析 $\delta^{13}\text{C}$ 、TOC/TN 在沿海环境(如河口、潟湖、海湾和隔离盆地)古环境重建中的应用,发现部分示例 $\delta^{13}\text{C}$ 、TOC/TN 值可能会受到浮游生物、有机质分解等因素的影响^[15],因此,通过多种方法综合重构古环境会更加准确。

海水中有有机碳的通量与初级生产力之间呈正相关,因此沉积物中有机碳的累积率被认为是表层海

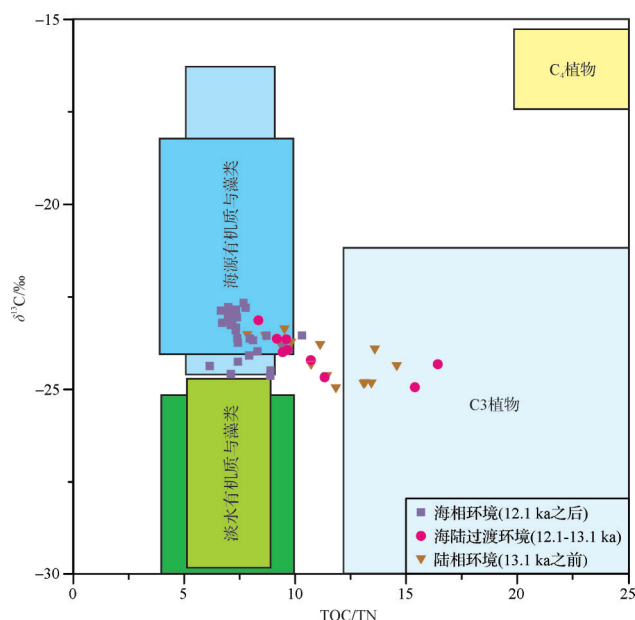


图 8 东海内陆架 EC2005 岩芯不同时期有机质来源 (据文献[123]修改)

Fig.8 Source of organic matter in core EC2005 during different stages (modified from reference[123])

水中初级生产力的直接标志^[124-125]。除此之外,有机碳还可以用来恢复季风演化过程,有机碳含量及堆积速率会受到季风的控制,其变化可以指示季风的强度^[126]。蔡德陵等^[124]利用东海现代沉积物中有机碳稳定同位素组成得到了海源有机碳与古生产力指标的相关关系。Hu *et al.*^[16]分析了东海内陆架的柱样(图 9, THB-2, 120.51° E, 25.87° N),发现 3.6~0.8 ka 河流输入有机质百分比随东亚夏季风的强度一起变化,而在最近 800 年季风气候和人类活动的共同作用控制着长江河流有机碳在东海内陆架的运输和埋葬。东海内陆架泥质区有机质矿化的硫酸盐还原过程可以导致自生黄铁矿的形成,并且伴随着黄铁矿的再氧化生成了自生石膏,说明有机碳埋藏过程伴随着 C-S-Fe 循环^[51]。因此,利用有机碳及相关生物地球化学的沉积信号不仅可以恢复长时间尺度(地质历史时期)东海内陆架沉积环境的演化,还能进一步阐明东海沉积物内元素地球化学循环过程,可以开展多指标、跨学科的研究,提供边缘海有机碳研究的新视角。

6 总结与展望

东海内陆架独特的沉积体系,特别是泥质沉积区沉积物的源—汇过程受到越来越多的关注,与细

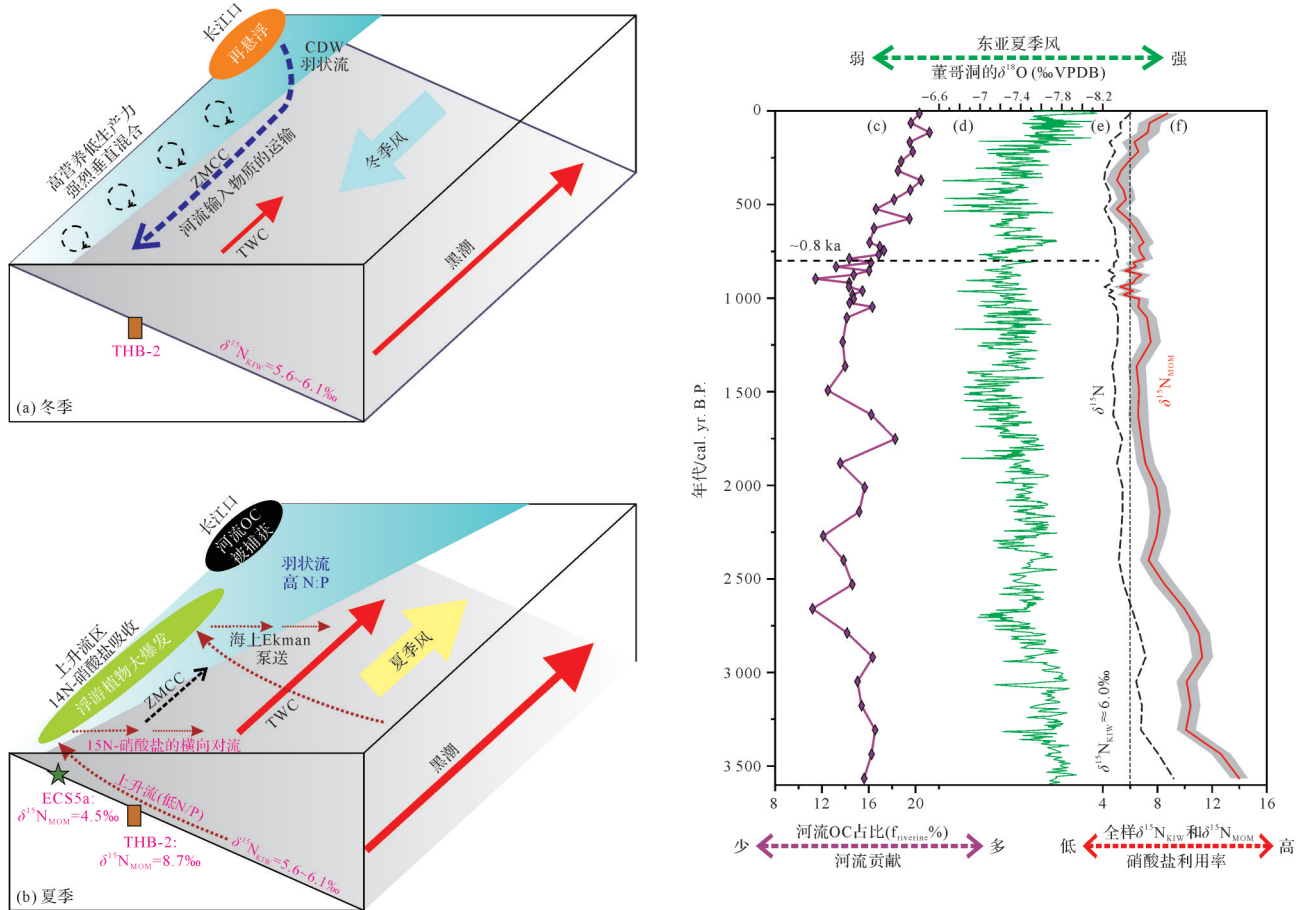


图9 东海西北部冬季(a)和夏季(b)水动力与养分示意图, (c) THB-2 中河流有机质占比, (d) 董哥洞石笋 $\delta^{18}\text{O}$ 记录, 上升流强度控制的 (e) $\delta^{15}\text{N}$ 和 (d) $\delta^{15}\text{N}_{\text{MOM}}$ 硝酸盐利用率 (据文献[16]修改)

Fig.9 Schematic of hydrodynamics and nutrients:(a) in Winter, and (b) in Summer in the northwestern ECS; (c) proportion of organic matter in rivers in THB-2; (d) $\delta^{18}\text{O}$ record of Dongge cave stalactites; (e) $\delta^{15}\text{N}$; and (f) $\delta^{15}\text{N}_{\text{MOM}}$ nitrate utilization rate (modified from reference[16])

粒沉积物相关的C-S-Fe生物地球化学过程也被日益重视。本文尝试用沉积学的角度梳理东海内陆架有机碳物源、输运、埋藏等方面的研究进展。总体来看, 目前对于不同来源、不同类型沉积有机碳沉积过程和沉积记录已有较好的研究基础, 但在如下方面的工作还有待进一步深入:

(1) 目前关于东海内陆架有机碳来源虽然已有大量成果, 但其中大多只进行了陆源与海源的划分, 而对于不同端元的进一步识别(比如陆源有机碳又可以分为植物和土壤来源)相对较少, 需借助新的技术手段, 开展更精细的研究。

(2) 目前对于东海沉积有机碳的跨陆架输运, 更多的是从物源角度证明了跨陆架输运的存在, 而对于跨陆架输运是否存在季节性及相关沉积动力学机制仍不甚清楚, 需要加强现代沉积动力过程的观测与模拟工作。

(3) 人类活动(例如大坝建设、土地利用、水体富营养化)明显影响东海内陆架有机碳的沉积过程, 进而影响碳循环及海洋生态系统, 需要进一步的研究。伴随着极端气候事件的频发, 台风和洪水等极端气候事件对有机碳的沉积过程和沉积记录影响的研究亟待加强。

(4) 非稳态环境内有机碳再矿化过程, 以及相关的自生矿物(例如自生黄铁矿和碳酸盐矿物)及反向风化作用(例如粘土矿物的生成)等生物地球化学过程, 需要开展跨学科的综合研究。

(5) 有机碳相关的指标在古环境重建等方面具有很好的应用前景, 但目前在东海内陆架及相邻海域的研究还相对偏少。同时, 研究长时间尺度上的内陆架有机碳对沉积环境的响应, 能为预测未来气候变化对内陆架有机碳归宿的影响提供地质依据。

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Source-to-Sink Process of Organic Carbon on the Inner Shelf of the East China Sea and its Sedimentary Records

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Abstract: Continental marginal seas are the main sites for deposition and burial of organic carbon from different sources with different properties, and they play an important role in the global biogeochemical carbon cycle. The inner shelf of the East China Sea (ECS) accepts a large amount of terrestrial organic carbon with high primary productivity and is an ideal site for the study of the source, transport and burial of sedimentary organic carbon. This comprehensive analysis of the literature gives a brief review of related sedimentological studies, with the aim of providing references for future research. Bulk analyses (TOC/TN, $\delta^{13}\text{C}$, etc.) and biomarkers (*n*-alkanes, sterols, lignin, etc.), among other methods, have indicated that the source of organic carbon in this location is affected by the sedimentary environment. The terrestrial components increase significantly landward from the sea and have seasonal characteristics. The inner shelf sediments of the ECS have a macroscopic transport characteristic best described as "storing in Summer and transporting in Winter" along the coast, and this dynamic process affects the coastal transport path and final destination of terrestrial organic carbon. There exists a "tongue-shaped" zone of transported terrigenous organic carbon near latitude 29°N in the ECS, which may be accompanied by cross-shelf transport of terrestrial organic carbon, affecting the source-sink process of deep-sea organic carbon. In addition, human activity and extreme climatic events have also significantly affected the records of the deposition processes of the sediment and organic carbon; such factors need further study. The inner shelf of the ECS is an important area of buried terrigenous organic carbon. Its burial efficiency has been influenced by the source, the content and the sedimentation rate, and is related to the mineralization path of the organic matter during the early diagenetic process. The geochemical characteristics of the organic carbon contained in the sediments are also useful for reconstructing long-term sea-level change, primary productivity, paleoceanography and paleoclimate evolution, thus providing a basis for clarifying the environmental evolution of the ECS inner shelf during its geological history.

Key words: inner shelf of the East China Sea; organic carbon; early diagenesis; cross-shelf transport; deposition process