



海洋颗粒动力学的铀系同位素示踪

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2025-11-30 收稿, 2026-01-21 修回, 2026-02-06 接受, 2026-02-12 网络版发表

国家自然科学基金(42476030, 42076030)资助

摘要 颗粒物调控着海水中诸多元素的循环、生物可利用性以及海洋与气候变化之间的相互作用。海洋颗粒动力学涵盖了海水中颗粒物及相关元素的地球化学行为、循环过程的速率与通量以及所发生的时间尺度等信息, 是理解海洋生物地球化学循环的核心之一。铀(^{238}U)系同位素是研究海洋颗粒动力学的重要手段。本文从连续体(海湾-近海-真光层-中深层-近底层)的空间视角, 系统综述了铀系同位素示踪海洋颗粒动力学的研究, 概述了同位素示踪的原理和不同同位素适用的时空尺度, 重点探讨了观测条件和内在机制(包括物理过程、稳态-非稳态条件、通量算法和颗粒物-同位素相互作用)对同位素示踪的影响, 评述了不同水体同位素示踪研究的发展现状及面临的问题, 进而总结了适用于连续体中各区域的理想观测策略、适用同位素和计算模型, 展望了它们在示踪陆-海界面、海洋中深层和底边界层颗粒动力学的应用前景。在海洋研究走向深海的时代, 铀系同位素可为从年际-代际尺度上认识深海生物地球化学循环提供重要的技术手段和科学价值。

关键词 颗粒动力学, 铀系同位素, 生物泵, 碳循环

海洋中的颗粒物包括生源颗粒(海雪、浮游生物碎屑、粪粒)^[1]和非生源颗粒(陆源碎屑、大气尘埃、自生矿物)^[2]。颗粒物在深海碳封存^[3]、中深层生态系统能量供给^[4]和金属元素地球化学循环^[5]中起着重要作用, 并从本质上调控着海水的化学组成^[6]。海洋颗粒动力学研究海水中颗粒物的形成、聚集、分解、输运、沉降和埋藏等过程, 揭示其发生的机制、时间尺度以及相关的速率和通量等特征^[6]。海洋颗粒动力学的研究手段主要有沉积物捕获器^[7,8]和放射性同位素示踪^[9]。随着海洋科学研究向深海的挺进, 海洋内部颗粒动力学将通过链接表层海洋和深层海洋关键过程从不同时空尺度上揭示海洋的储碳能力和机制, 为深入理解海洋对全球变化的响应与反馈提供全新认识。

本文从空间连续体(海湾/近海-真光层-中深层-近底层)视角综述了应用铀(^{238}U)系同位素示踪海洋颗粒

动力学的研究进展, 旨在系统梳理海洋不同区域颗粒动力学的时间尺度、空间作用等特征, 认识铀系同位素示踪颗粒动力学的核心内涵, 不同同位素在不同海洋地带应用的优劣及面临的问题, 并提出未来发展方向。本文首先阐述铀系同位素示踪海洋颗粒动力学的基本原理, 随后分析不同海洋地带不同同位素示踪的现状, 最后提出未来的研究方向。海洋颗粒动力学与铀系同位素融合研究不仅为深化海洋不同地带生物地球化学循环研究提供新技术, 也为从不同时空尺度理解和应对气候变化及可持续发展提供科学依据, 具有重要科学价值。

1 铀系同位素示踪原理

铀-238系列同位素是以 ^{238}U 为起始衰变核素的天然放射性衰变系列核素(图1)。当前仅能对海水中半衰

引用格式: 杨伟锋, 陈敏, 蔡平河, 等. 海洋颗粒动力学的铀系同位素示踪. 科学通报

Yang W, Chen M, Cai P, et al. Particle dynamics constrained by uranium-series radionuclides in marine environments (in Chinese). Chin Sci Bull, doi: 10.1360/CSB-2025-5850

铀系	^{238}U	$\rightarrow$	^{234}Th	$\rightarrow$	^{234}U	$\rightarrow$	^{230}Th	$\rightarrow$	^{226}Ra	$\rightarrow$	^{210}Pb	$\rightarrow$	^{210}Bi	$\rightarrow$	^{210}Po
$T_{1/2}$	4.5×10^9 年		24.1天		2.4×10^5 年		7.5×10^4 年		1620年		22.3年		5.01天		138.4天
衰变类型	α		β^-		α		α		α		β^-		β^-		α
基本性质	水溶性		强颗粒活性		水溶性		强颗粒活性		水溶性		强颗粒活性		强颗粒活性		强颗粒活性
时间尺度	<5个月		<40万年				<30年		<1个月		<2年				
应用水层及示踪过程	真光层颗粒物输出		中深层颗粒物沉降				中深层颗粒物沉降		真光层颗粒物输出		真光层颗粒物输出				
	底层沉积物再悬浮						底层沉积物再悬浮				中层颗粒物再矿化				
应用区域	海湾 陆架/坡 海盆		海盆				陆架/坡 海盆		海湾 陆架		陆架/坡 海盆				

图1 海洋颗粒动力学研究中相关铀系核素的特征

Figure 1 Characteristics of U-series nuclides associated with particle dynamics in the ocean

期较长(>3 d)的核素进行定量分析,因而用于海洋颗粒动力学示踪的铀系核素主要包括 ^{230}Th 、 ^{234}Th 、 ^{210}Pb 、 ^{210}Bi 和 ^{210}Po 。 ^{230}Th 是海水中最早被分析的铀系颗粒活性核素^[10],随后 ^{234}Th 也被分析^[11]。21世纪之前通过大体积海水共沉淀法获得了有限的铀系Th同位素数据。小体积(2升)海水 ^{234}Th 共沉淀方法^[12]的出现加速了 ^{234}Th - ^{238}U 在全球海域的广泛应用^[9]。海水中 ^{210}Po 和 ^{210}Pb 的测量虽晚于Th同位素^[13],但由于其活度比 ^{234}Th 约低一个数量级,目前对海水中 ^{210}Po 和 ^{210}Pb 的相关研究显著少于 ^{234}Th 。受限于很短的半衰期及很低的含量,海水中的 ^{210}Bi 直到20世纪90年代才被分析^[14],其应用目前仅有几例报道^[15]。分析上的难度差异很大程度上影响到不同核素的研究深度及应用广度。

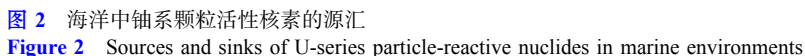
若不存在其他迁出过程,海水中铀系核素母-子体之间都应处于放射性衰变平衡,但实际情况下,海水中颗粒物对颗粒活性核素的清除往往导致很多母-子体之间存在不平衡现象^[11,16]。同位素示踪海洋颗粒动力学一般需要满足两个条件:一是核素具有强颗粒活性,即海水中这些核素容易吸附到颗粒物表面,其固-液分配系数通常大于 10^4 L kg^{-1} ^[2,17],且地球化学行为与颗粒物循环紧密相关^[18];二是核素与其母体之间存在放射性不平衡,且该不平衡与颗粒物沉降、再矿化和再悬浮等过程相关。这也造就了铀系核素示踪颗粒动力学时往往以子-母体核素对的方式出现,即 ^{230}Th - ^{234}U 、 ^{234}Th - ^{238}U 、 ^{210}Pb - ^{226}Ra 、 ^{210}Bi - ^{210}Pb 和 ^{210}Po - ^{210}Pb ^[15,19-21]。对于颗粒活性子体-水溶性母体核素对 ^{234}Th - ^{238}U 、 ^{230}Th - ^{234}U 和 ^{210}Pb - ^{226}Ra ,子体(^{234}Th 、 ^{230}Th 和 ^{210}Pb)从海

水中的快速迁出会造成其活度相对于母体亏损(子体活度小)^[22]。对于同为颗粒活性的子-母体核素对 ^{210}Bi - ^{210}Pb 和 ^{210}Po - ^{210}Pb ,子、母体都会随颗粒物沉降迁出,但由于子体半衰期远短于母体半衰期,颗粒物吸附相同数量的子母体原子数会造成更大活度的子体迁出(活度等于衰变常数与原子数之积),导致水体中剩余子体活度相对于母体亏损^[23]。

研究发现,子体的亏损程度与颗粒物沉降迁出的强度之间存在紧密关系,沉降通量越大,子体亏损程度越大^[24],这为通过子体-母体之间的不平衡程度定量颗粒物的迁出强度奠定了理论基础。定量上,一般根据海水中核素的源汇质量平衡原理(图2)来建立颗粒活性核素活度随时间的变化关系^[25,26]。若假设颗粒活性核素的迁出遵守一级动力学^[24],则有:

$$\frac{dA_d}{dt} = F_{d,in} + \lambda A_p - \lambda A_d - u \frac{\partial A_d}{\partial x} - v \frac{\partial A_d}{\partial y} - \omega \frac{\partial A_d}{\partial z} + K_x \frac{\partial^2 A_d}{\partial x^2} + K_y \frac{\partial^2 A_d}{\partial y^2} + K_z \frac{\partial^2 A_d}{\partial z^2} - F_{d,out}, \quad (1)$$

式中, A_d 和 A_p 分别表示水柱中子体和母体的活度浓度(dpm m^{-3}), λ 为子体衰变常数, u 、 v 和 ω 分别代表海水不同方向的流速(m s^{-1}), K_x 、 K_y 和 K_z 表示海水不同方向的涡动扩散系数($\text{cm}^2 \text{ s}^{-1}$),一般假定流速和扩散系数为常数, $F_{d,in}$ 代表除母体现场衰变外其他来源(如大气沉降等)的子体输入速率($\text{dpm m}^{-3} \text{ s}^{-1}$), $F_{d,out}$ 是子体的输出速率($\text{dpm m}^{-3} \text{ s}^{-1}$)。对输出速率进行深度积分就可获得输出通量。实际应用中可根据水文条件状况和核素性质来决定能否忽略海水平流和扩散的影响。



2 近岸海湾颗粒动力学示踪

Figure 1 is a graph showing the activity ratio (活度比值) versus time (时间) for four different isotope ratios. The y-axis ranges from 0.5 to 1.1. The top x-axis shows time in days (天) for ^{210}Bi , ^{234}Th , and ^{210}Po , ranging from 0 to 700. The bottom x-axis shows time in years (年) for ^{210}Pb , ranging from 0 to 150. The four curves are: $^{210}\text{Bi}/^{210}\text{Pb}$ (blue dashed line), $^{234}\text{Th}/^{238}\text{U}$ (red solid line), $^{210}\text{Po}/^{210}\text{Pb}$ (green dashed line), and $^{210}\text{Pb}/^{226}\text{Ra}$ (black solid line with 'x' markers). All curves start at 0.5 and approach 1.0 over time.

扩散)影响很大,高度的时空异质性不利于流速、扩散系数等参数的准确应用,导致难以通过质量平衡方程来获得颗粒动力学的准确信息^[31]。早期,少数研究测量了河口、海湾的 ^{234}Th ^[25,32,33],一些研究也通过稳态假设并忽略物理过程影响,尝试计算颗粒物的停留时间^[27,34,35],但铀系同位素示踪近岸海湾颗粒动力学的研究仍然较少。

3

垂向活度梯度, 结合三维流速和涡动扩散系数计算了生源硅、颗粒有机碳(particulate organic carbon, POC)和颗粒态氮的沉降通量^[31]. 然而, 这种方法需要获得高空间分辨率的平流和扩散参数, 限制了其更加广泛的应用. 对于半封闭海湾而言, 湾内与外海海水的交换通常在月-季节尺度^[36-39]. 在这一时间尺度上, 如果将整个海湾作为一个整体, 仅考虑湾口的交换, 那么湾内的平流和扩散等物理过程在宏观上并不会影响所获得的海湾整体的颗粒动力学特征. 少数研究通过整个海湾站位布设和月-季节频率的²³⁴Th采样, 将质量平衡方程中的物理项与输出项合并, 采用非稳态计算了整个海湾颗粒物的输出通量, 获得了与海湾碳汇强度一致的结果^[40,41]. 需要指出的是, 即使将海湾作为整体开展研究, 也需要布设有代表性的合适站点并开展时间序列观测, 而且这种方法无法获得湾内不同区域的颗粒动力学信息. 另外, 有研究通过近岸海湾特定站点的²¹⁰Po/²¹⁰Pb时间序列观测, 利用非稳态模型获得了颗粒动力学的时间变化特征^[42], 但其背后的地球化学机制仍有待明确.

有些近海环境存在强烈的颗粒物水平输运, 且经常可以观测到颗粒物浓度与核素活度浓度之间存在显著线性关系^[40,42], 揭示了这些环境中颗粒活性核素的地球化学循环主要受控于颗粒物的循环, 因此可以通过颗粒活性核素的变化来定量颗粒物的输送量. 目前, 全球河流POC入海通量往往通过河流流量与POC含量的乘积来计算^[43], 颗粒态黑碳(particulate black carbon, PBC)入海通量通过POC入海通量与河流颗粒物中PBC/POC比值或泥沙通量与PBC/泥沙比值来计算^[44-46]. 这种方法得到的通量实际上代表的是径流POC或PBC到达河口的通量, 并非输送到外海的通量^[30]. 由于颗粒物在河口-陆架连续体中持续沉降, 目前还缺乏可靠的方法来定量径流颗粒组分的实际入海通量, 极大地影响了海洋中一些元素(如碳)的收支评估. 最新研究报道了应用²³⁴Th-²³⁸U量化径流PBC入海通量的一种方法, 即通过在河流主控型海湾与陆架的界面处开展潮汐尺度的高频时间序列观测, 发现海湾-外海间的²³⁴Th交换通量与潮差之间存在显著线性关系, 反映了“潮汐泵”的作用机制, 据此计算出15%~33%的径流PBC可以到达外海^[30]. 这一研究为应用²³⁴Th-²³⁸U及其他铀系核素准确定量径流颗粒组分的入海通量奠定了基础. 需指出, 颗粒物水平输运过程中存在多维度的通量(如水平扩散和垂直沉降), 目前该方法仅能

通过比较核素的产生通量和总输送通量之间的差异判断水平输送为主或是垂直沉降为主, 结合输送通量的正负值和海流方向判别颗粒物输送的方向, 若能获取颗粒物的沉降速率则可以区分不同维度的输送通量.

3 真光层颗粒动力学示踪

3.1 真光层颗粒动力学示踪概述

真光层是铀系核素示踪海洋颗粒动力学最广泛的区域(图4). 在五对铀系核素中, ²³⁴Th-²³⁸U是研究最多、应用最广的示踪剂^[9]. 这一方面得益于海水中²³⁸U与盐度之间存在的线性关系, 通过盐度可以便捷地获得²³⁸U活度^[47,48], 另一方面²³⁴Th小体积测量方法的建立^[12,49]有利于²³⁴Th数据的获取. 自1969年首次报道海水中²³⁴Th的亏损现象至20世纪90年代初, ²³⁴Th-²³⁸U的研究主要集中在²³⁴Th地球化学行为及其对总颗粒物的示踪领域, 包括²³⁴Th的颗粒活性(固-液分配)^[11,50]、清除、迁出速率及停留时间^[22,51,52]以及颗粒物的聚合、解聚速率^[19]. Coale和Bruland^[24]发现太平洋寡营养海域真光层双层结构中颗粒物沉降通量大小与²³⁴Th沉降通量对应. 1989年Epply提出应用POC储量除以²³⁴Th停留时间来计算POC输出通量(假设POC与²³⁴Th的停留时间相同)^[53]. 然而, 沉积物捕获器和²³⁴Th方法的对比研究发现, 颗粒物停留时间与²³⁴Th相同, 但POC和PN的停留时间更长^[54]. 随后, Buesseler等人^[55]提出通过真光层²³⁴Th沉降通量乘以真光层底部POC与颗粒态²³⁴Th的比值来计算真光层的POC输出通量. 虽然通过²³⁴Th停留时间或POC/²³⁴Th比值将²³⁴Th沉降通量转换为颗粒物组分沉降通量的方法并不完美, 但这些研究开启了应用²³⁴Th/²³⁸U广泛示踪颗粒循环的新纪元. 1990年启动的全球海洋通量联合研究(Joint Global Ocean Flux Study, JGOFS)计划突出了碳通量研究的需求, ²³⁴Th/²³⁸U不平衡法应势被快速推广到开阔海域真光层POC沉降通量的示踪. 后来, ²³⁰Th/²³⁴U、²¹⁰Pb/²²⁶Ra、²¹⁰Bi/²¹⁰Pb和²¹⁰Po/²¹⁰Pb不平衡法示踪颗粒物通量也是基于与²³⁴Th/²³⁸U类似的思路^[15,21,56]. 需要指出, ²³⁰Th半衰期很长, 其在海水中的含量很低, 停留时间可达几十年, 由此造成其受平流和扩散影响较大, 若不能很好地约束真光层较为活跃的物理过程, 难以通过²³⁰Th/²³⁴U获得理想的颗粒物沉降通量^[57]. 另外, 真光层的²¹⁰Pb主要来自大气沉降, ²²⁶Ra衰变产生的²¹⁰Pb贡献很小, 因而在中低纬度海域真光层中经常存在²¹⁰Pb过剩现象

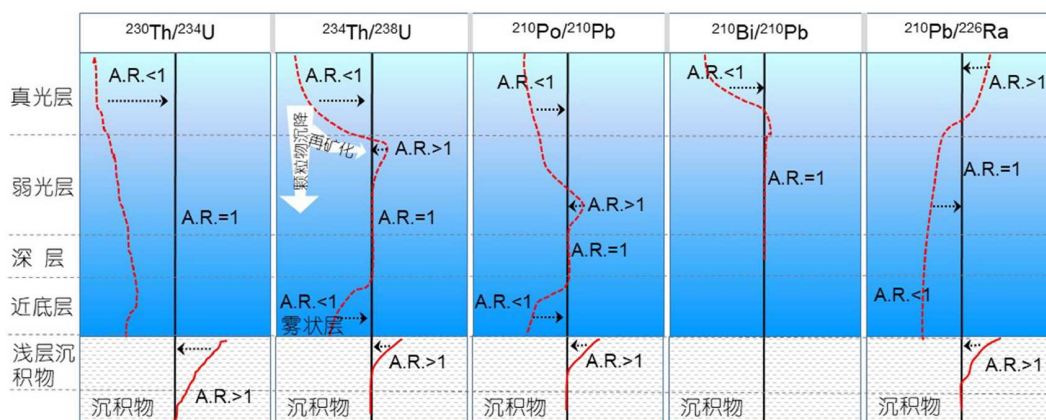


图4 海水中铀系核素对活度比值的垂直分布(A.R.表示活度比值)

Figure 4 Profiles of activity ratio of daughter to grandparent nuclides in the ocean (A.R. denotes activity ratio)

(图4). 由于大气沉降的高度时空异质性, $^{210}\text{Pb}/^{226}\text{Ra}$ 在真光层的应用需要很好地测量 ^{210}Pb 的大气沉降通量^[58,59].

3.2 物理过程对颗粒动力学示踪的影响

类似近岸海湾, 真光层同样面临物理过程影响的问题. 由于平流和扩散参数难以获得, 仅有少数研究直接或间接评估了物理过程的影响. 例如, 北大西洋平流和扩散对真光层 ^{234}Th 输出通量的影响为5%~10%^[55], 赤道太平洋为25%~35%^[60], 南海为1%~21%^[25]. 基于拉格朗日观测, 南海北部物理过程对200 m以浅 ^{234}Th 输出通量的影响低于4%^[61]. 根据模型测试, 南海南部水平及垂向的平流和扩散对 ^{234}Th 输出通量的影响与通量测定的误差接近^[26], 但南海中尺度涡对 ^{234}Th 输出通量的计算影响可达50%^[62]. 南海东北部陆坡区平流和扩散对 ^{210}Po 沉降通量的影响为17%±21%^[63]. 总体上, 外陆架和开阔海域物理过程的影响程度与核素或POC沉降通量计算的误差相当. 故对开阔海域真光层的研究而言, 忽略物理过程对核素或颗粒物沉降通量的影响比较合理, 这也是铀系核素得以广泛应用的原因. 目前 $^{210}\text{Pb}/^{226}\text{Ra}$ 和 $^{210}\text{Bi}/^{210}\text{Pb}$ 在真光层的应用很少, 尚未见有关物理过程影响的评估.

3.3 稳态条件对颗粒动力学示踪的影响

稳态假设是铀系核素示踪颗粒动力学研究中常用的条件, 即假设采样前的一段时间内真光层维持稳定状态, 核素的输入与迁出速率相等, 其活度不随时间变化. 目前大多数研究是基于某个时间点的采样, 不具备解非

稳态质量平衡方程的条件, 因而大都基于稳态假设来计算 ^{234}Th 、 ^{210}Po 、 ^{210}Pb 和 ^{210}Bi 的沉降通量^[9,15]. 稳态假设是否成立取决于核素示踪的时间尺度内颗粒物的沉降通量是否稳定. 一些研究发现, 稳态和非稳态模型计算的 ^{234}Th 停留时间差异达到3~4倍^[64,65], 水华事件甚至会导致 ^{234}Th 稳态假设得到的沉降通量完全不合理^[55]. 夏季南大洋绕极流区真光层稳态和非稳态 ^{210}Po 和 ^{210}Pb 的沉降通量相差1~2个数量级^[66]. 但是, 也有研究发现, 在颗粒物沉降通量高的海域稳态和非稳态结果之间没有明显差异^[25,67], 可能是颗粒物沉降通量达到了使 ^{234}Th 可以快速地迁出而其产生速率仅能将 ^{234}Th 活度维持在一个很低的水平, 表观上更接近稳态. 时间序列观测显示, 稳态和非稳态计算的 ^{210}Po 沉降通量之间存在显著差异^[42]. 因此, 稳态假设是否合理直接影响到铀系核素示踪结果的准确性. 对于引起非稳态的情形(如水华事件), 有必要开展时间序列观测, 以便更客观地反映颗粒动力学的特征. 目前, 尚无明确的方法用于判断海区颗粒物输出是否处于稳态. 通常, 除季节转换间期外, 开阔海域或海盆区接近稳定状态. 陆架区一般较难达到稳定状态, 但中尺度空间范围上可以近似处理. 近岸海湾受潮汐等影响, 通常不具备稳态条件, 需要开展高频观测. 另外, 满足不同核素稳态假设的时间尺度也不同. 半衰期越短的核素记录颗粒物沉降信息的时间越短, 其稳态假设越容易满足. 在颗粒物时空变化较频繁的海域(包括近岸海湾), 短半衰期的 ^{210}Bi 和 ^{234}Th 能获得较好的颗粒动力学信息, 而在颗粒物时空变化较小的水体(如开阔海域中深层), 长半衰期 ^{210}Po 、 ^{210}Pb 和 ^{230}Th 的稳态假设通常能获得较好的结果.

3.4 不同核素结果之间的差异

虽然 ^{234}Th 、 ^{210}Po 、 ^{210}Pb 和 ^{210}Bi 都可用于示踪真光层颗粒物的沉降通量,但不同方法的结果之间并不总是一致^[59,68]。对比研究表明,大西洋、赤道太平洋和马尾藻海由 ^{210}Po 计算的POC沉降通量高于 ^{234}Th 的结果^[69~71]。南大洋冰边缘区和西北地中海 ^{234}Th 方法得到的POC沉降通量却明显高于 ^{210}Po 方法^[56,72]。南海北部 ^{210}Bi 获得的POC沉降通量与 ^{234}Th 和 ^{210}Po 方法可比^[14,24,58,72]。 ^{234}Th 、 ^{210}Po 、 ^{210}Pb 和 ^{210}Bi 示踪的时间尺度(图1)以及受物理过程的影响程度^[61]都存在差异,尽管尚不明确这些因素在不同海域如何发挥作用,但综合这些研究仍可从不同示踪剂的结果差异获得一些启示。不同算法的影响因素不同,会导致结果之间存在差异。在停留时间算法中,核素的停留时间和颗粒物组分的储量计算需要利用真光层中所有层位的数据^[53]。这在一定程度上降低了某些质量不佳数据的影响,更适合于从宏观上约束研究区域的动力学特征。由于近岸海湾潮汐等过程导致的强烈时空变化,停留时间算法可能是适合从宏观上研究近岸海湾颗粒物输出的选择^[40,41]。比值法是基于输出界面处颗粒态核素和颗粒物一起沉降的假设而建立,由核素的沉降通量乘以比值来获得同步沉降的颗粒物组分通量^[55]。理论上比值法无需测量真光层中颗粒物组分的垂向变化,仅需获得输出界面处颗粒态核素和颗粒物组分数据波动的影响比较明显。 $\text{POC}/^{234}\text{Th}$ 比值会随时间、深度、颗粒物粒径和沉降速率等显著变化^[73]。也有报道 $^{210}\text{Po}/^{210}\text{Pb}$ 不平衡分别应用停留时间法和比值法获得的西太平洋POC沉降通量一致^[74]。因而如何选取合理的比值仍然是值得探讨的重要问题^[75]。相比,开阔海域 $\text{POC}/^{234}\text{Th}$ 比值的变化幅度明显小于近海海湾,可能更适合比值法。另外,近年来关于核素与颗粒物相互作用的研究越来越多,发现铀系颗粒活性核素对颗粒物沉降通量的示踪结果还受到颗粒物组成的影响。

3.5 颗粒物性质对核素示踪的影响

颗粒物性质(组成、粒径等)的差异本质上决定了核素与颗粒物之间的相互作用,进而影响核素示踪的结果。无论是停留时间算法还是比值法,本质上都涉及一个问题,即海水中不同核素的迁出由哪些颗粒物组分所决定?理论上,只有在核素迁出过程中起决定作

用的组分其停留时间或与核素的比值才能客观地将核素的沉降通量转换为颗粒物组分通量。现场研究发现,一些海域 ^{234}Th 与总颗粒物停留时间相同^[54],总颗粒物或POC浓度与核素(^{234}Th 或 ^{210}Po)活度之间存在显著的线性关系^[40,42],这些结果反映了总颗粒物或者POC在核素的清除过程中起主要作用,核素总体上示踪的是总颗粒物或POC的循环。也有研究显示, ^{234}Th 停留时间明显短于POC和PN的停留时间^[54],说明POC与 ^{234}Th 的行为存在较明显的差异,可能是其他颗粒组分起主要作用。南极绕极流的研究表明,POC与 ^{210}Po 有更强的亲和力,而生源硅与 ^{234}Th 和 ^{210}Pb 有更强的亲和力^[66]。模拟实验表明,颗粒物组分对 ^{210}Po 、 ^{210}Pb 和 ^{210}Bi 的吸附存在显著差异^[2,76],腐殖酸、酸性多糖和蛋白质对 ^{210}Po 有更强的亲和力,而无机颗粒物对 ^{210}Pb 和 Th 同位素具有更强的亲和力^[17,77]。另外,大分子有机物存在与否对颗粒物吸附Pb、Bi和Po有不同的影响^[78],不同的核素倾向于与不同的官能团结合^[79],富含羧基(-COOH)和脂肪族结构的磷蛋白在其中起重要作用^[80],酸性多糖在 ^{234}Th 清除中起关键作用^[81],含磷有机分子与 ^{210}Pb 有最强的亲和力,而富含氮和硫的有机分子与 ^{210}Po 亲和力更强^[82]。目前核素示踪的究竟是哪种颗粒物组分仍有待研究。颗粒物粒径的选择也会影响铀系核素示踪的颗粒物输出结果。不同粒径(0.7~210 μm)颗粒物上 $\text{POC}/^{234}\text{Th}$ 比值可在1~2个数量级范围内变化^[7]。传统观点认为,大粒径颗粒物沉降速率快,在颗粒物沉降过程中起主导作用,但近年来的研究表明慢沉降的小粒径颗粒物的贡献不容忽视^[7]。选择合适的粒径仍然是铀系同位素示踪颗粒动力学待研究的命题。

对于颗粒物特征较明显的水体,核素示踪的颗粒物组分比较清晰。比如,在水动力很强的浅水区或海底雾状层(benthic nepheloid layer, BNL),颗粒物浓度通常很高,颗粒物浓度与核素活度浓度之间有显著的相关关系^[40,42],核素能够较好地指示总颗粒物的地球化学行为。对于典型的热液羽流区, ^{234}Th 的分布与铁的分布一致^[83],其示踪的可能是含铁颗粒物。对于浮游植物类型多样的海洋真光层和颗粒物贫瘠的大多数海洋中深层,核素的清除通常由不同的颗粒物组分共同调控^[84,85],不同核素示踪的颗粒物组分仍有待明确。

4 中深层颗粒动力学示踪

真光层以下的中深层缺少初级生产过程,相对匮乏的颗粒物导致颗粒活性核素的停留时间可以长达几

年^[29], 因而半衰期较短的²¹⁰Pb和²³⁴Th在真光层以深水体通常与其母体保持放射性平衡(图4)^[15,86], 无法用于中深层颗粒物沉降的示踪。不同的是, 半衰期更长的核素与其母体之间的不平衡情形会保留更长时间(图4)。因此, 中层²¹⁰Po、中深层²³⁰Th和²¹⁰Pb仍存在相对于母体的亏损^[16,59,74,84,87], 但据此开展中深层水体颗粒物沉降的研究仍较少。

尽管²¹⁰Pb和²³⁴Th通常不能用于示踪中深层水体颗粒物的沉降, 但少数研究发现, 紧邻真光层底部的中层水体有时存在²¹⁰Pb和²³⁴Th相对于母体过剩的现象^[15,88], 归因于真光层POC的快速沉降及其在中层水体的再矿化。这种亏损可用来示踪中层水体POC的再矿化通量^[15,89]。另外, 在一些受特殊过程影响的中深层水体也可以观测到²³⁴Th的亏损, 如在西南印度洋及大西洋热液羽状流中均发现了²³⁴Th的明显亏损^[83,90,91], 据此获得的POC沉降通量证实了热液羽状流中铁锰颗粒吸附海水中DOC加速碳沉降的机制^[91]。在南海东北部和墨西哥湾也发现了跨陆架/坡颗粒物的水平输送引起的中层²³⁴Th亏损^[61,92]。这些中深层的²³⁴Th亏损可用于示踪相关特定过程的颗粒动力学。

地球化学海洋断面研究(Geochemical Ocean Section Study, GEOSECS)计划实施期间获得了各大洋²¹⁰Po、²¹⁰Pb、²²⁶Ra和²³⁰Th在中深层的分布特征, 并应用²¹⁰Po/²¹⁰Pb和²¹⁰Pb/²²⁶Ra不平衡定量了海洋内部²¹⁰Po和²¹⁰Pb的停留时间^[16,18], 但直至20世纪90年代, 海洋中²¹⁰Po和²¹⁰Pb的大多数研究都集中在它们的地球化学行为和停留时间方面。1990年末至21世纪初, 伴随着²³⁴Th/²³⁸U对真光层POC沉降通量的广泛报道, 学者逐步开始关注真光层以下POC的沉降与再矿化, ²¹⁰Po/²¹⁰Pb对颗粒物组分沉降通量的示踪逐步得到发展^[56,66,70]。限于海水及颗粒物中²¹⁰Po和²¹⁰Pb的分析周期长达1~2年, 目前应用²¹⁰Po/²¹⁰Pb示踪中深层颗粒物沉降及再矿化的研究仍不多。我国学者联合应用²¹⁰Po亏损和黑碳示踪了南海东北部中层水中不同来源POC的沉降通量, 并获得了马丁曲线指数^[63]。基于²¹⁰Po/²¹⁰Pb不平衡计算了南海和南大洋中层水体POC的沉降与再矿化通量^[89]以及西太平洋的POC沉降通量^[74]。中层²¹⁰Po的亏损也揭示了南海西部和北部以及亚北极白令海广泛存在陆架/坡颗粒物跨陆架向海盆中深层的输送^[21,93,94]。21世纪初开始的痕量元素及其同位素海洋生物地球化学循环研究(An International Study of the Marine Biogeochemical Cycles of Trace Elements and

Isotopes, GEOTRACES)计划在各大洋开展了全水柱更高分辨率的²¹⁰Po、²¹⁰Pb和²³⁰Th等铀系核素分析, 将会推动铀系同位素示踪颗粒动力学的发展。

相较于真光层, 中深层受风、光和生物活动等影响较小, 其环境一般相对稳定。同位素活度随时间的变化较小, 稳态假设在中深层更易满足。比如, 南海中深层(1000~3500 m)²¹⁰Po稳态和非稳态条件计算出的沉降通量无显著差异, 1000 m处的沉降通量仍然表现出与真光层相似的季节变化, 但2000~3500 m的通量则没有明显季节变化^[95]。另外, 中深层平流和扩散作用比上层海洋更弱, 但这并不意味着物理过程对中深层²¹⁰Po和²¹⁰Pb沉降通量的影响更小。传统的箱式模型采用水柱积分算法, 虽然计算的是中深层界面的²¹⁰Po和²¹⁰Pb沉降通量, 但依然包含了真光层输出的核素通量。比如, 南海东北部陆坡区150 m和1000 m平流对²¹⁰Po沉降通量的影响分别为17%±21%和23%±12%^[63]。为此, 有学者提出了中深层²¹⁰Po的一维逆向模型^[96,97], 通过数学方法获得物理过程综合作用下颗粒物的沉降速率, 一定程度上避免了测量平流和扩散参数的难题。基于此方法可以获得中深层水体中较高分辨率的颗粒物沉降速率, 再通过沉降速率与颗粒物组分的乘积计算沉降通量^[98]。然而, 在一些过程复杂、核素分布规律性差的海域, 逆向模型不一定能获得理想的结果。海洋内部应用²¹⁰Pb/²²⁶Ra示踪颗粒物循环的报道更少, 但中深层²¹⁰Pb的亏损普遍存在^[29]。有研究发现, 太平洋和南海沉积物捕获器获得的中深层POC沉降通量与²¹⁰Pb的沉降通量之间存在显著的线性正相关^[99,100], 为²¹⁰Pb/²²⁶Ra不平衡示踪中深层颗粒物组分的沉降通量奠定了基础。少数探索性研究揭示了中深层一些过程对²¹⁰Po和²¹⁰Pb应用的影响。在陆架-海盆相互作用强烈的海域, 陆架沉积物再悬浮后的横向输送会加剧中深层²¹⁰Po和²¹⁰Pb的亏损程度^[21], 综合颗粒物浓度增大的现象, 可以定量横向输送的贡献^[93]。有机质组分在沉降过程中的再矿化会弱化²¹⁰Po和²¹⁰Pb的亏损甚至使²¹⁰Po过剩, 但准确定量再矿化通量仍待解决^[89]。有研究发现²¹⁰Pb存在边界清除^[29], 该过程对中深层²¹⁰Pb应用的影响仍无法评估。鉴于不少真光层中应用的核素示踪手段无法在中深层使用, 进一步开发²¹⁰Po/²¹⁰Pb和²¹⁰Pb/²²⁶Ra在中深层的示踪应用颇具价值。少数研究应用²³⁰Th/²³⁴U不平衡示踪了中层颗粒物沉降通量, 结果与²³⁰Th/²³⁴U-²²⁸Ra/²²⁸Th双示踪剂方法结果存在明显差异, 揭示了²³⁰Th/²³⁴U示踪颗粒动力学需要二维模型更好地约束

物理过程的影响^[20,57]。但研究发现,很多海域 ^{230}Th 随深度而增加,通过可逆交换模型预测其分布特征可获得颗粒物的沉降速率,为研究中深层海水颗粒动力学提供了关键参数^[50,101]。由于中深层颗粒物浓度通常很低,颗粒活性核素的清除强度较小,获得的颗粒物沉降通量等参数误差一般较大,可通过减小测量误差和提高采样的空间分辨率获得更好的结果。

5 近底层颗粒动力学的示踪

一般情况下,近底层水中的颗粒物浓度很低,颗粒活性核素的停留时间较长,除 ^{230}Th 和 ^{210}Pb 外, ^{234}Th 、 ^{210}Bi 和 ^{210}Po 都会与其母体达到放射性平衡。然而,一些海域近底层存在BNL,即沉积物再悬浮形成的高颗粒物浓度水体^[102]。有研究发现,BNL之上海水中的 ^{234}Th 与 ^{238}U 达到了平衡,而BNL中的 ^{234}Th 则明显亏损,证明沉积物再悬浮造成了近底层的 $^{234}\text{Th}/^{238}\text{U}$ 不平衡^[103,104]。另外,沉积物中 ^{234}Th 相对于 ^{238}U 过剩,且过剩程度与近底层水中 ^{234}Th 的亏损程度相近,说明沉积物中的过剩 ^{234}Th 来自近底层水而非上覆水柱^[103,105]。目前,在南海北部、南大洋、西北冰洋和地中海均观测到沉积物再悬浮形成的异常 $^{234}\text{Th}/^{238}\text{U}$ ^[106,107]和 $^{210}\text{Po}/^{210}\text{Pb}$ 信号^[108], ^{234}Th 和 ^{210}Pb 也被用于定量东海沉积物的再悬浮^[27]。BNL也会导致 ^{230}Th 的含量低于可逆交换模型预测,反之,该现象也可以指示近底层活跃的颗粒动力学过程^[109]。过去40年的研究结果表明,北大西洋西部、南大西洋阿根廷海盆、南大洋部分海域和南非外缘广泛存在BNL,而太平洋大部分海域BNL很弱或不存在^[110]。最新的研究证实了东太平洋深海采矿形成的沉积物再悬浮及扩散会导致底层水中 ^{234}Th 的强烈清除^[111],为铀系颗粒活性核素广泛示踪BNL提供了支持。沉积物再悬浮一定程度上影响到海洋深层溶解有机质的来源^[112]和有机碳在海洋中的输运^[113]。随着技术的进步,铀系颗粒活性核素在近底层生物地球化学研究中将会发挥更大作用。

由于近底层海水中加强的颗粒物清除过程,在上层沉积物中常常会观测到过剩的 ^{230}Th 、 ^{210}Pb 以及 ^{234}Th ,这为铀系颗粒活性核素示踪沉积动力学过程提供了条件。通常,沉积物中过剩 ^{234}Th 可用于约束沉积物中的生物扰动强度^[114],近海、陆架/坡沉积物中过剩 ^{210}Pb 可用于示踪沉积物的沉积速率^[115],深海沉积物中过剩 ^{210}Pb 可计算生物扰动强度^[116],沉积物中过剩 ^{230}Th 则可以确定沉积速率和年代学^[117]。

6 总结与展望

近半个世纪以来,海水及颗粒物中铀系颗粒活性核素的分析方法不断完善。分析 ^{234}Th 和 ^{210}Po 的海水体积已由20世纪60年代的~20升减小至几升,提高了数据的可获得性,但测定仍然以 β 计数法(^{234}Th)和 α 能谱法(^{210}Po)为主,液闪法虽有报道,但较高的检测限并没有得到推广。 ^{210}Pb 分析仍然通过其生长的子体 ^{210}Po 或 ^{210}Bi 测定,通过 ^{210}Po 测定对海水体积要求较低,但等待 ^{210}Po 生长的时间较长(>4个月),通 ^{210}Bi 测定等待时间较短(~1个月),但需要更大的海水体积(>20 L)。 ^{226}Ra 、 ^{230}Th 和 ^{238}U 早期通过能谱法测定需要大体积海水,目前已经实现了质谱测量,测量所需海水体积已大幅减小且误差更小。随着技术的发展,有望有更多的铀系同位素通过质谱分析。

从研究关注的重点看,铀系颗粒活性核素从以核素地球化学行为为主逐步转变至不同海洋环境中颗粒动力学的示踪,铀系核素为从不同时空尺度理解海洋生物地球化学过程作出了重要贡献。比较而言, $^{234}\text{Th}/^{238}\text{U}$ 的研究和应用最为广泛,主要应用于上层海洋和近底边界层的颗粒动力学研究。 ^{210}Po 、 ^{210}Pb 和 ^{230}Th 近20年来得到越来越多的关注,也是正在实施的GEO-TRACES计划涵盖的同位素,主要应用于上层、中层和深层海洋的颗粒动力学过程。 ^{210}Bi - ^{210}Pb 的应用研究刚起步。 ^{210}Pb - ^{226}Ra 虽然较早获得一些数据,但其在深海区的应用研究还很少,随着海洋科学研究向深海的拓展,其应用预计将越来越多。综合已有研究成果,河口-外海、近岸/海湾、真光层、中深层和近底层等不同水体颗粒动力学研究中建议采用的最适宜铀系核素及其应用方案如图5所示。

目前,铀系核素示踪颗粒动力学也面临诸多待解决的问题和发展机遇,主要包括:(1)除 ^{234}Th - ^{238}U 和 ^{230}Th - ^{234}U 外,其他核素(^{210}Bi 、 ^{210}Po 、 ^{210}Pb)的分析仍需较长时间或采集大体积海水,不利于其广泛应用,有必要发展小体积高灵敏度的分析方法;(2)河口-海湾-近岸连续体颗粒物组分的源汇及其入海通量是地球化学领域的难题之一,铀系核素为该问题的解决提供了可行的手段,但其适用条件和环境仍需进一步的研究;(3)中深层海洋颗粒物及相关元素循环(如碳封存、铁收支)是当前海洋化学领域关注的焦点之一,但研究手段匮乏, $^{210}\text{Po}/^{210}\text{Pb}$ 和 $^{210}\text{Pb}/^{226}\text{Ra}$ 的示踪时间尺度恰好匹配,可用于开展中深层高时空分辨率的颗粒动力学研究;(4)近年来我国深潜器的海底作业揭示了

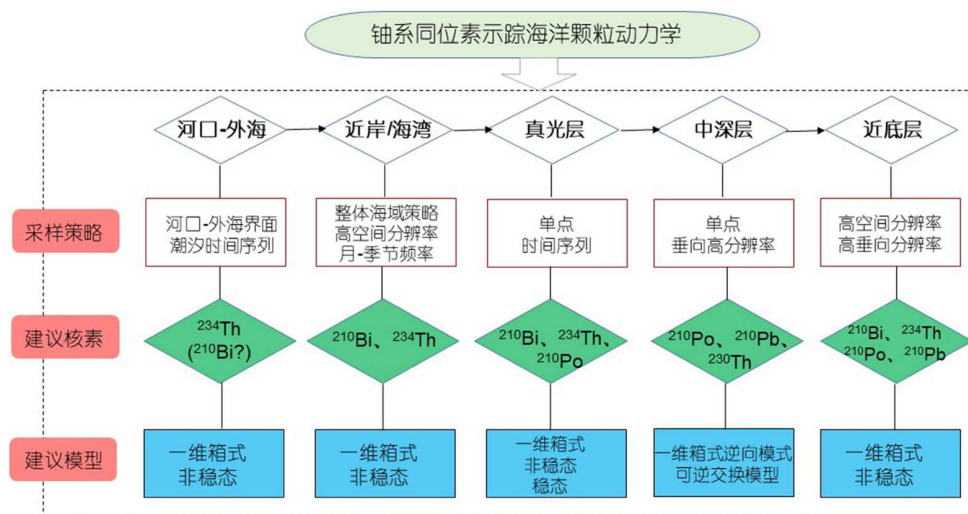


图5 不同海洋环境中颗粒动力学研究建议采用的铀系核素及应用方案

Figure 5 Proposed strategies for investigating particle dynamics in various oceanic regimes using U-series nuclides

海底冷泉、热液、泥火山等活跃而相对独立的系统，其生物地球化学过程不同于传统的真光层，铀系颗粒

活性核素的应用可为这些系统及底边界层的生物地球化学过程提供新的认识。

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Summary for “海洋颗粒动力学的铀系同位素示踪”

Particle dynamics constrained by uranium-series radionuclides in marine environments

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Particulate matter largely governs the geochemical cycling and bioavailability of many elements by regulating their removal and regeneration, thereby affecting the interactions between the oceans and climate change. Particle dynamics encompasses the fundamental geochemical behavior of particulate matter and its associated elements, resident timescales, (re) cycling rates, and sinking fluxes of particulate components, thereby constituting a crucial component of marine biogeochemistry. As global change continues to accelerate changes in the ocean surface, understanding the responses and carbon capacity of the ocean interior has become imperative. Particles, which act as a quick linkage between the ocean surface and ocean interior, are key to understanding changes in the magnitude of carbon dioxide sequestration by the ocean. Uranium-series radionuclides are gaining importance because of their dominant role in tracking particle dynamics in the ocean.

This review provides a comprehensive synthesis of the applications of U-series radionuclides for constraining particle dynamics within the land–seabed continuum (i.e., bay–coast–euphotic zone–meso-/bathypelagic zone–bottom layer). The basic principle of radionuclides in tracing particle cycling and timescales is presented, with emphasis on the influence of observational conditions and intrinsic mechanisms on the applications of U-series nuclides, including physical processes, steady/non-steady conditions, algorithms, and the interaction between particles and radionuclides. Additionally, the challenges faced by U-series radionuclides in constraining particle dynamics, particularly within the ocean interior, are summarized. The study proposes a preferred strategy and model for constraining particle dynamics in terms of different oceanic regimes, and presents the questions that must be addressed to improve particle dynamics.

One of the major advances realized within the bay–coast continuum is the quantitative evaluation of particulate organic carbon sinking on a bay-scale in strong dynamic shallow waters based on monthly seasonal sampling, which significantly resolves the quantitative constraint of particle settling in technique-deficient environments and extends the application scope of U-series nuclides. The other is the accurate calculation of the actual riverine particulate components in coastal seas, which enables researchers to revisit the budgets for organic carbon and other elements in the coastal and shelf waters.

Reports on the euphotic zone allow the assessment of the influence of observational strategies and assumptions on the applications of U-series nuclides. Physically, single sampling is generally acceptable assuming steady-state conditions. By contrast, time-series observations are required for bloom events, upwelling, and scenarios with highly variable processes over short timescales. From an algorithmic perspective, the residence time method appears to be appropriate for regimes characterized by highly heterogeneous environments, whereas the ratio method seems to be better suited for steady environments.

Understanding of particle dynamics has been very limited in the ocean interior, i.e., from the mesopelagic zone to the seafloor, owing to the lack of techniques. However, in recent years, studies have provided insights into particle dynamics in the mesopelagic and bottom waters, establishing a foundation for examining the sinking and regeneration of organic carbon and carbon storage in the deep ocean. The application of U-series nuclides to constrain resuspension provides an approach for (quasi) quantitatively evaluating the material exchange between the seawater and sediment.

In conclusion, understanding of particle dynamics has significantly improved owing to the use of U-series nuclides in the ocean. Progress in shallow, meso- and bathypelagic, and bottom waters provides opportunities to utilize U-series nuclides for the investigation of particle dynamics in changing environments by using limited techniques and/or approaches, especially in bay/coastal areas and the ocean interior. By emphasizing particulate cycling over different timescales in the ocean interior, we can more effectively evaluate the response of the ocean to climate change and the role of the ocean in sequestering carbon dioxide.

particle dynamics, U-series nuclides, biological pump, carbon cycling

doi: [10.1360/CSB-2025-5850](https://doi.org/10.1360/CSB-2025-5850)