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厦门大学

博 士 学 位 论 文

台湾高山流域及邻近海域颗粒有机质的源汇过程

Taiwan as an example for "Source to Sink" processes of particulate organic matter in high mountainous river and surrounding marginal seas

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摘要

陆海交互界面“源到汇”沉积体系由高山环境延伸，横跨大陆边缘，最后汇入深海，是侵蚀和沉积过程发生的主要位置，构成地球沉积循环的主要组成部分，同时，也是连接地球系统长时间尺度碳循环的枢纽。在轨道时间尺度上，大气二氧化碳的含量主要取决于化石碳的氧化以及光合作用所固定碳的埋藏这二者之间的平衡。而陆海交互界面是这两个过程发生的主要场所，因此理清陆海交互界面碳的迁移转化过程对定量全球碳收支至关重要。高山岛屿（high-standing islands）的河流流域面积占地球面积不到 3%，每年向海洋输入的有机碳却占全球入海通量的 17-35%，是研究碳源汇过程理想的天然实验场。因此本文以台湾高山流域的土壤、水库悬浮物、水库沉积物、河流悬浮物、高屏陆架沉积物、高屏海底峡谷沉积物捕获器收集的沉降颗粒以及冲绳海槽柱状沉积物为载体，以稳定碳、氮同位素及 ^{14}C 为手段，从源到汇的视角较为系统地研究了有机碳的迁移转化过程及其在千年尺度上的宿命。主要结论如下：

（1）通过分析台湾 60 条高山流域的土壤垂直剖面的碳氮含量我们计算了土壤有机碳库大小，并讨论其与坡度、海拔高度等地形参数以及湿度、温度等气象数据之间的联系，此外，以碳同位素随深度的富集因子作为土壤周转时间的替代性指标，建立了土壤有机碳库周转时间与环境因子之间的关系。我们发现台湾土壤碳库大小主要受温度的影响，温度通过影响有机质降解速率进而控制碳库的大小。另一方面，我们也发现表层土壤有机质碳同位素与年均降雨量之间存在正向关系，推测是由于降雨带来了大量营养元素促进了初级生产力所导致，随后，我们发现有机碳库与年均降雨之间存在显著正相关，进一步支持了该推测。这一发现表明，尽管降雨量的增加可能使得土壤的侵蚀增强，加大土壤的碳流失量，然而由降雨促进初级生产力导致的碳增加使得总体上碳库增加，暗示随着雨量的增加，台湾土壤的碳汇能力会增强。

（2）通过对高山流域水库布放的沉积物捕获器进行一年多的观测，并结合周边土壤及水库沉积物的碳氮同位素，我们发现水库沉降颗粒物及有机质的全年输出分别有 80%和 70%是在台风期间（约占全年时间的 3%）发生，这证明了台风事件快速而有效地输出与埋藏有机质。此外，水库水体中沉降颗粒的碳同位素受控于水库季节性溶解无机碳库、季节性水体生产力，以及浮游植物群落结构的

季节性变化。而沉降颗粒氮同位素则显示水体中 NO_3^- 和 NH_4^+ 均为浮游植物的营养盐来源。此外，水库沉积物的有机质主要源于不同深度的土壤的混合，而水库自生的有机质超过 80%以上无法在沉积物中保存，大部分有机质都在水柱里被再矿化，这与大部分海洋环境的研究一致，进一步说明了不论在陆地淡水还是海洋生态系统，现代颗粒有机碳强烈的再矿化过程很可能是普遍存在的现象，并且可能是内陆水域向大气释放二氧化碳的源。

（3）通过分析土壤、河流悬浮物、河床沉积物及高屏海底峡谷沉积物捕获器的颗粒有机质的碳同位素及有机碳含量，我们发现台风期间高屏河流悬浮物、海底峡谷沉降颗粒的有机碳遵循双端元混合模型（化石有机碳及生源非化石有机碳）。模型显示台风期间河流悬浮物及海底峡谷沉降颗粒物的非化石有机碳（OC_{nf}）年龄达数千年之老，且主要来自古滑坡的侵蚀。而现代有机碳（植物碎屑）则只有在台风事件结合海底峡谷陡峭地形才能被有效传输至深海。有趣的是，我们还发现现代植物碎屑的有机碳含量及 ^{14}C 活性偏离了台风期间沉降颗粒的双端元混合模型，进一步证实了只有 OC_{nf} 年龄均一才能产生线性良好的双端元混合模型。此外，该研究还发现陆架表层沉积物碳同位素组成也遵循该双端元混合模型，暗示潮汐引起的再悬浮以及海洋有机质的吸附等生物/非生物过程对产生同位素均一、且年龄相同的 OC_{nf} 起到很大的作用，但具体机理有待验证。

（4）通过化学湿式氧化法将冲绳海槽中部钻孔过去 30ka 的沉积物中不同碳氮组分分离出来，我们发现键结在黏土矿物里的无机氮含量占到沉积物总氮的 38%，同位素数据则显示这部分黏土矿物键结的无机氮记录了陆地上降雨的信号。如此高的陆源无机氮的输入进一步揭示了传统沉积物有机质 C/N 在物源追踪方面的应用需考虑矿物键结氮的影响，尤其在中国边缘海这种以伊利石等 2:1 型矿物为主要黏土矿物的环境。而经过强氧化剂处理后残余的碳占沉积物总有机碳的 28-51%，这很可能来自于化石有机碳的贡献，这与前人在南冲绳海槽估算的化石碳含量吻合。此外残余碳的含量与沉积物中黏土矿物的含量成正比，这暗示着黏土矿物可能对有机质保存扮演十分重要的角色。

综上，温度通过影响有机质降解，而降雨通过影响生产力，两者共同控制了高山流域土壤有机碳库大小，而水库的观测则证实了大部分水体有机质及陆源植物碎屑在传输过程中被大量降解，而在海底峡谷的输送过程研究表明只有陡峭地

形结合台风事件才能有效将植物碎屑往深海输送从而保存，最后本文还发现黏土矿物有利于有机质的保存。

关键词：陆海界面；高山河流；碳同位素；有机碳；源汇过程

Abstract

The sedimentary “source to sink” system in land-to-ocean boundary is extended from the high mountain, across the continental margin, and eventually dumps into the deep sea. It is the primary locations for the erosion and deposition processes take place, which constitutes the sedimentary cycle of the earth and is also connected to the earth system for a long-term carbon cycle. At geological time scale, the concentration of atmospheric carbon dioxide was regulated mainly by the balance between the oxidation of fossil carbons and the burial of photosynthesis fixed carbon. Therefore, to better constrain the global carbon budget, it is very important to clarify the carbon erosion, transfer and burial processes. The high-standing islands in Western Pacific, occupied less than 3% of the Earth's area, but contribute 17 - 35% of annual global organic carbon flux to the ocean, thus being an ideal natural laboratory for studying carbon source and sink processes. Therefore, in this thesis, we collected the samples from the suspended sediment in the small mountainous river of Taiwan, the sinking sediments and on-floor sediment of reservoir, soil profiles, the surface shelf sediments, submarine canyon sinking sediment, and a piston core in Okinawa Trough. Stable carbon, nitrogen isotope and ^{14}C of these materials were analyzed to study the erosion, transformation, and bury processes of organic carbon from source to sink perspective. The main conclusions are as follows:

(1) By analyzing the 60 vertical profiles of soil carbon content and its isotope compositions, we calculated the soil organic carbon stock, and use the vertical carbon isotope enrichment factor as the proxy for the soil turnover rate. Combined with the topographical and meteorological data, we find that the soil carbon stock in Taiwan is mainly affected by temperature. That is, the higher temperature enhances the degradation rate of organic matter, leading to decline of carbon pool size. On the other hand, we also found that there was a positive relationship between the carbon isotope of the surface soil organic matter and the mean annual rainfall, presumably due to the large amount of nutrients (P and Cu) brought by the rainfall, which led to the increase in primary productivity. And there is a significant positive correlation between mean

annual rainfall and carbon stock further support this inference. These findings suggest that, despite the increased rainfall might accelerate the carbon loss via enhance of soil erosion, the brought nutrient alleviate the nutrient limitation and thus promote the primary productivity, leading to an increase in carbon stocks. Thus, we speculate that as the rainfall increases under global warming scenario, Taiwan soil's carbon sink capacity will increase.

(2) We investigated the seasonal variation of sinking sediment flux and associated variations of carbon and nitrogen isotope composition by deploying sediment traps in 20m and 70m depth at Feitsui Reservoir (FTR), northeastern Taiwan. During observation periods (Nov. 2004-Nov. 2005), seven typhoon events were recorded. Allochthonous source material (three soil profiles in upper river reach of FTR) as well as ultimate sediment deposits (five gravity sediment cores) was also collected to evaluate the organic carbon and nitrogen cycling within reservoir from source to sink perspective. We found more than 80% of annual sediment flux was exported during typhoon events, agreeing with previous studies on river side which proposed that episodic events dominate the sediment transfer in Taiwan. The carbon and nitrogen contents and their isotopic composition ($\delta^{13}C_{20m\text{-}POC}$ and $\delta^{15}N_{20m\text{-}PN}$) in non-typhoon period displayed evident seasonal variation, which was governed by reservoir internal biogeochemical processes. In particularly, $\delta^{13}C_{20m\text{-}POC}$ in nontyphoon period was regulated by seasonal variations of temperature, dissolved inorganic carbon pool size, and seasonal community structure alteration; while the $\delta^{15}N_{20m\text{-}PN}$ temporal variation patterns might reflect the phytoplankton uptake multi-nitrogen sources (both ammonium and nitrate). Binary mixing model ($\delta^{13}C$ versus 1/OC) suggests the organic carbon preserved in FTR was result from mixing of different depth soil, and the reservoir autochthonous summer primary primary signal cannot be found in sediments. Collectively, our results imply that the oxidation of *in-situ* primary production might be an important route for CO2 gas evasion in inland water.

(3) A sediment trap was deployed at 610 m deep in the Gaoping submarine canyon at western Pacific during summer 2008, during which a typhoon event

(Kalmaegi) was recorded. Basing on cross relations of total organic carbon (TOC), isotopic compositions ($\delta^{13}C$, ^{14}C) and nitrogen to carbon ratios (N/C) among newly measured and previously reported source materials, i.e., mountain soils, river suspensions, river sediments, shelf sediments and sediment trap samples, we deciphered particulate organic carbon sources and quantified the content of fossil organic carbon (OC_f) and biospheric non-fossil (OC_{nf}) for typhoon and non-typhoon transports. The ages of OC_{nf} components in trap samples during typhoon and non-typhoon periods were similar; both pre-aged and uniformly pre-mixed. During typhoon, canyon was more connected to river and the OC_{nf} in canyon trap was likely sourced from organics buried in ancient landslides. The plant debris cannot be found in trap except in the hyperpycnal layer, suggesting hyperpycnal flow was capable of entrain plant debris while segregation had occurred during non-hyperpycnal periods. Extreme events coupled with canyon system created an efficient way for deep sea burial of freshly produced organic-rich material. During non-typhoon, canyon is more connective to shelf, where wave and tide benefit reworking, thus, allow abiotic and biotic processes to prepare isotopically uniform and aged OC_{nf} for canyon transport. Results shed lights on the ephemeral behavior of organics transport in a canyon system at active margin boundary.

(4) We use KOH-KOBr to separate operationally defined total organic matter into oxidizable (labile) and residual fractions for content and isotope measurements. In a sediment core in the Okinawa Trough, significant amounts of carbon and nitrogen existed in the residual phase, in which the C/N ratio was ~9 resembling most documented sedimentary bulk C/N ratios in the China marginal seas. Such similarity creates a pseudo-C/N interrupting the application of bulk C/N. The residual carbon, though composition unknown, it displayed a $\delta^{13}C$ range (-22.7 to -18.9‰, mean -20.7‰) similar to black carbon (-24.0 to -22.8‰) in East China Sea surface sediments. After removing residual fraction, we found the temporal pattern of $\delta^{13}C_{LOC}$ in labile fraction (LOC) was more variable but broadly agreed with the atmospheric pCO_2-induced changes in marine endmember $\delta^{13}C$. Thus, we suggested adding pCO_2-induced endmember modulation into two-endmember mixing model for

paleo-environment reconstruction. Meanwhile, the residual nitrogen revealed an intimate association with illite content suggesting its terrestrial origin. Additionally, $\delta^{15}N$ in residual fraction likely carried the climate imprint from land. Further studies are required to explore the controlling factors for carbon and nitrogen isotopic speciation and to retrieve the information locked in the residual fraction.

Key words: Source to sink; land-ocean boundary; high mountain rivers; organic carbon; carbon isotope

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