学校编码：10384 密级______

学号：33120120153566

厦门大学

博 士 学 位 论 文

根系分泌物中类黄酮对白骨壤根际镉生物地球化学行为影响的研究

Influence of Root Secretion Flavonoids on the Biogeochemistry Behavior of Cadmium in *Avicennia marina* (Forsk.) Vierh Rhizosphere

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论文提交日期： 2015 年 4 月
论文答辩时间： 2015 年 5 月

2015 年 6 月

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目录

Content

中文摘要

红树林生态系统主要分布于热带和亚热带的海岸潮间带，其具有独特的生态学和生物学特性，为污染物的吸附治理提供了最佳场所。然而，近年来，随着工业化、城市化和地区经济的快速发展，导致大量生活污水和工业废水流向红树林湿地，使得红树林生态系统面临越来越严重的重金属污染，从而使得滨海湿地面临着日益严峻的重金属污染。进入红树林湿地的污染物最先在红树植物根际区进行一系列形态转化等过程，而后被植物吸收累积。因此，红树林根际区重金属化学行为已经成为近年来污染生态学的研究热点。

类黄酮是植物体内广泛存在的一类重要次生代谢产物，是一种常见的根系分泌物，对植物缓解胁迫、抵御逆境有着重要的生理生态作用。目前对红树林生态系统中类黄酮的研究主要集中在植物体内定量和结构定性、化感作用、生物地球化学循环等方面，但关于类黄酮对红树林沉积物中重金属的生物地球化学行为的影响研究仍显不足。

本研究以福建省九龙江口红树林湿地沉积物为研究对象，以我国东南沿海红树林的先锋树种——白骨壤（*Avicennia marina* (Forsk.) Vierh）为试材，研究了红树林湿地生态系统中类黄酮对重金属镉的生物地球化学行为的影响。通过研究类黄酮对白骨壤根际、非根际中酸可挥发性硫（AVS）、活性固相二价铁和酸可提取态镉的迁移转化的影响，探讨类黄酮对沉积物中镉的活化机制与释放作用；结合白骨壤根系吸收镉的动力学特征对类黄酮的响应研究，认识类黄酮对重金属镉进入植物体过程中对植物的耐受性影响及转运、保护机制。重金属镉由植物体外的根际沉积环境向植物根表铁膜迁移直至被根系吸收的过程中受类黄酮影响下发生形态转化与迁移，以及根系在类黄酮影响下镉的吸收饱和与吸收速率变化，对阐明重金属镉在红树林生态系统中的生物地球化学行为有重要意义；为红树林湿地的保护与修复提供科学、有效的途径，丰富红树林湿地生态学研究内容。

研究结果表明：

类黄酮和重金属镉的增加均明显改变了沉积物的氧化还原环境，导致氧化还原电位（Eh）降低。加之类黄酮自身的还原能力导致白骨壤根际、非根际中

的活性固相二价铁的含量明显增加，促进了植物根系对铁的吸收，缓解了铁对红树植物生长的限制作用。根际中部分二价铁离子被氧化沉淀，使根表铁膜加厚，非根际中活性固相二价铁显著高于根际。根系活动使根际、非根际产生氧梯度差和Eh梯度差，造成重金属镉在根际、非根际的形态赋存差异；加之根系分泌物的酸化、增溶及螯合作用，均增加了根际中镉的浓度。类黄酮能使红树林沉积物中的镉得到活化，造成根际、非根际沉积物中弱酸提取态镉（Acid extractable Cd）的含量显著增加，增加了镉的活性，根系活动使得根际沉积物中镉含量与非根际差异更为明显。但是根表铁膜作为根系表面的屏障，其加厚增加了重金属离子进入植物体的难度，同时二价铁离子与镉离子之间存在明显的竞争关系，从而减少了进入植物体内的重金属；类黄酮使镉在根表铁膜的累积明显减少，阻碍了白骨壤幼苗对镉的吸收与转运，根系中镉的累积降低，导致镉在植物体内的累积明显降低。类黄酮打破了红树林沉积物中原有的“库-源”平衡，对红树林沉积物中镉的释放与解毒起到至关重要的作用；同时植物体对镉的吸收，并未因类黄酮对镉的活化而增加，反之减少，此结果对红树林的保护及修复有极其重要的意义。

本文利用经典米氏酶动力学模型研究了在重金属镉进入植物根系的过程中类黄酮对根系吸收的动力学影响。研究发现米氏动力学模型能够很好的描述白骨壤根系在类黄酮作用下对镉离子的吸收。与对照组相比，*Km*（半饱和常数）和*Vmax*（最大吸收速率）在类黄酮作用下均升高，证明了类黄酮能够增强植物体对镉的耐受性。但是，类黄酮对植物根细胞壁吸收镉离子没有显著影响。当受到呼吸抑制剂CCCP（Carbonylcyanidem-chlorophenyl-hydrazone）处理时，得到了同样的结果，证明类黄酮对白骨壤根系镉吸收的跨膜运输起到重要作用，但是不会影响其质外体运输。当白骨壤根系受到Ca^{2+}离子通道抑制剂$LaCl_3$处理时，施加类黄酮的处理组中*Km*和*Vmax*均高于未受类黄酮处理的组，而低于对照组，说明钙离子通道不是白骨壤根系吸收镉的唯一跨膜运输途径。因此类黄酮利用Ca^{2+}通道和其他离子运输途径，通过跨细胞质膜的共质体运输增加了红树植物白骨壤吸收镉的速率与饱和浓度，增强了植物对重金属镉的耐受能力，能有效减少重金属对植物的毒害作用。

关键词：红树林；镉；类黄酮；根际

Abstract

Mangrove ecosystem is widely distributed in tropical and subtropical coastal intertidal zone, and it provides the best places for adsorption of pollutants and governance of environment because of its unique biological and ecological characteristics. However, a large amount of domestic and industrial sewage discharge into mangrove with rapid development of urbanization, industrialization and regional economy in recent years, which posed mangrove wetlands facing increasingly serious heavy metal pollution, almost exceeded mangrove system self-purification capacity and caused coastal wetlands facing increasingly severe metal pollution and organic pollution. At present, interiorly wide studies have been reported on heavy metals pollution in mangrove. Contaminant existing in mangrove wetlands is taken up and enriched after a series of species transformation in the rhizosphere. Therefore, substances cycle in the rhizophere has been a research hotspot for pollution ecology.

Flavonoids are one kind of important secondary metabolites and widely distributed in plants, which has an important role in alleviating the stress and against adversity in physiology and ecology. At present, the researches of flavonoids focused on flavonoids quantify and qualitative structure in plants, allelopathy, biogeochemical cycle, but one of a very few research about the influence of flavonoids on the biogeochemistry of heavy metals in mangrove sediments was found.

In this paper, the mangrove wetland sediments from Jiulong River mangrove natural reserve (24°24′N, 117°55′E), Fujian, China, as the research object and Chinese southeast coastal mangroves pioneer species - *Avicennia marina* as test materials were researched for the biogeochemical behavior change of Cd when flavonoids were provided in mangrove ecosystem. The role of flavonoids to Cd biogeochemistry behavior in sediments was assessed through researching the effects of flavonoids on the species distribution and migration of Cd. Through study of the influence of flavonoid amendment on concentration-dependent kinetics of Cd absorption in *A. Marina* roots, we identify the mechanism of heavy metal Cd access into plant roots. We have studied the behavior of Cd migration from rhizosphere to plant roots and the concentration-dependent kinetics of Cd access into roots and exposed the biogeochemistry behavior of heavy metal Cd when flavonoids were employed in mangrove wetlands ecosystem. We provided scientific and effective ways for mangrove remediation and protection,

and provided theoretical foundation for further study and understanding of mangrove.

The results have shown that: providing flavonoids and Cd changed the redox environment in sediments and resulted in oxidation reduction potential (Eh) reducing. Additionally, reducing capacity of flavonoids led to reactive solid-phase Fe (II) increasing clearly, which facilitated absorption of Fe in plants, and remitted the limit of Fe for mangrove plants growth. For the absorption of roots and roots activity, e.g. radial oxygen loss (ROL), part of Fe (II) is precipitated in iron plaque, which make reactive solid-phase Fe (II) higher in the non-rhizosphere than that in the rhizosphere. The increase of iron plaque played a role of "barrier", impeding the access of heavy metals into roots. The competition occured between Fe^{2+} and Cd^{2+}, and the accumulation of Cd decreased in roots and iron plaque as response to flavonoids exposure, which prevented the uptake of Cd in *A. Marina* seedlings, and then resulted in a reducing concentration of Cd in plants. Flavonoids played an important role in protection of mangrove plants. Phenols facilitated iron accumulation in iron plaque and Fe (II) producing in sediments which was favorable to Fe absorption in plant roots. It alleviated the limit of Fe for mangrove plant growth. Cd was activated as response to phenol treatment then that increased activity of Cd in mangrove sediments, especially in rhizosphere. This result implies that phenols disturb the "source-sink" balance of Cd in the mangrove sediments. Thus it appears that phenol treatment resulted in increase of Cd activity, but the concentration of Cd clearly decreased greatly in the plants themselves, accounting for Cd accumulation decreasing in iron plaque, and competition between Fe^{2+} and Cd^{2+} impeding Cd uptake in *A. marina* seedling roots. We can summarize that phenols play a critical role in detoxification of Cd in mangroves which is an arresting discovery for the biogeochemical behavior of Cd turning it into a "source" when phenols were added, and it has a great significance for Cd release and soil remediation in mangrove.

The role of flavonoids on the process of plant absorption heavy metal Cd was researched. A Michaelis-Menten kinetic model consisting of saturation and linear components has represented transmembrane transport in roots and apoplastic transport in root cell walls, which has been widely used in concentration-dependent kinetics of

heavy metal. It was suitable to describe Cd^{2+} absorption in mangrove plant *A. Marina* roots under the influence or non-influence of flavonoids. Compared to the control, *Km* and *Vmax* was higher under the influence of flavonoids, which demonstrated that flavonoids enhanced the tolerance of plant to cadmium; nevertheless， flavonoids showed no influence on the uptake of Cd^{2+} in root cell walls. The same result was achieved in roots treated by CCCP. These results showed that flavonoids act on symplasm transport when roots take up the heavy metal Cd^{2+}, but do not affect apoplastic transport. In the roots treated with ion transport inhibitor ($LaCl_3$), *Km* and *Vmax* was higher compared with the group without flavonoid amendments, and lower than the control, which demonstrated that the Ca^{2+}-channel was not the unique means of symplasm transport for Cd^{2+} absorption in *A. Marina* roots. More research is needed for other channels of Cd^{2+} absorption beyond the Ca^{2+}-channel. Thus flavonoids increased the saturation concentration by symplastic transport, enhanced the tolerance for Cd in plant and favoured the protection of plant.

Key words: Mangrove; Cadmium; Flavonoids; Rhizosphere