

# Phytoremediation of arsenic by aquatic floating macrophytes

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## 英文要約

The phytoremediation of arsenic by aquatic macrophytes *Spirodela polyrhiza* L. and *Salvinia natans* L. was investigated. Plants were grown hydroponically on MS culture solution. Arsenic uptake was higher from phosphate-deficient solution, when the source was arsenate. Arsenic uptake was not affected by phosphate, when the source was DMAA.

Arsenic and phosphate uptake was negatively correlated ( $p < 0.001$ ), and arsenic and iron uptake was positively correlated ( $p < 0.05$ ), when the initial source was arsenate. Arsenic uptake from DMAA solution was neither affected by phosphate nor correlated with iron uptake ( $p > 0.05$ ). A substantial amount of arsenate and arsenite was adsorbed on Fe-plaques of plant surfaces. The concentrations of As(V) and As(III) on plant surfaces were about 43% and 58% higher after Fe-plaque induction than without Fe-plaque induction, respectively. Fe-plaque induction did not affect MMAA and DMAA uptake and adsorption. The aquatic macrophytes might use physiological and adsorptive uptake mechanisms for arsenic uptake and a different mechanism for MMAA and DMAA uptake.

The EDTA increased about 4-6% of arsenic uptake, when the source was arsenate or arsenite, while the uptake was not affected by EDTA, when the source was MMAA and DMAA. It might be because the co-precipitation of iron occurs only with inorganic arsenic.

## 本文

Phytoremediation, a plant based green technology becomes promising to remediate environmental pollution due to some unavoidable limitations of traditional technologies. In the present study, we investigated the uptake of arsenate (As(V)) and dimethylarsinate (DMAA) by widely grown aquatic macrophytes *Spirodela polyrhiza* L. and *Salvinia natans* L. to determine their arsenic uptake efficiency from water, and to determine the influence of phosphate and iron ions in selective uptake of arsenic species by the aquatic macrophytes. Plants were grown hydroponically on standard Murashige and Skoog (MS) culture solutions for 6 days. Compared to the control treatment, the aquatic macrophytes accumulated significantly higher amount of arsenic from phosphate-deficient solution, when the source was arsenate. However, arsenic uptake by the macrophytes was not affected by phosphate, when the source was DMAA. The *Spirodela polyrhiza* L. accumulated 51-fold arsenic from arsenate solution compared to that from DMAA solution with initial concentrations of 4.0 M and 0.02 M of arsenic and phosphate, respectively.

Arsenic and phosphate uptake into the aquatic macrophytes was negatively correlated ( $p < 0.001$ ), when initial source was arsenate while there was no correlation between arsenic and phosphate uptake ( $p > 0.05$ ), when initial source was DMAA. The results indicate similar sorption mechanisms of  $\text{AsO}_4^{3-}$  and  $\text{PO}_4^{3-}$  for uptake carrier. Similarly, significant correlation ( $p < 0.05$ ) was observed between arsenic and iron uptake in tissues of aquatic macrophytes, when initial source was arsenate while the correlation was not significant ( $p > 0.05$ ), when initial source was DMAA. About 56% of the total arsenic was distributed in tissues of the plants compared to 44% in citrate-bicarbonate-EDTA (CBE)-extract (Fe-plaque) of plant surfaces, when the plants were grown on arsenate solution. Arsenic uptake into the aquatic plants from DMAA solution was neither affected by phosphate concentrations nor correlated with iron uptake ( $p > 0.05$ ). The results suggest that adsorption of arsenate on Fe-plaques of the plant surfaces contributes to the arsenic uptake greatly. Thus, arsenate uptake in aquatic plants might occur through the phosphate uptake pathway and by physico-chemical adsorption on Fe-plaques of plant surfaces as well. The plants use different mechanisms for DMAA uptake.

The *Spirodela polyrhiza* uptakes higher amount of arsenic, when the source was inorganic arsenic species (arsenate or arsenite) while the uptake was negligible, when the source was methylarsenic species (MMAA or DMAA). The concentrations of As(V) and As(III) in CBE-extracts of the macrophyte grown after Fe-plaque induction were about 43% and 58% higher than those plants grown without Fe-plaque induction, respectively. The results suggest that a substantial amount of arsenate and arsenite was accumulated by *Spirodela polyrhiza* L. through the physico-chemical adsorption on Fe-plaques of plant surfaces. The Fe-plaque induction on plant surfaces did not affect MMAA and DMAA contents in tissues and CBE-extracts of the macrophyte.

The influence of ethylenediaminetetraacetic acid (EDTA) on the arsenic species uptake by *Spirodela polyrhiza* L. was investigated. The EDTA increased arsenic uptake, when the source was arsenate or arsenite while the uptake was not affected by EDTA, when the source was MMAA and DMAA. It was estimated that about 4-6% of arsenate or arsenite was desorbed/mobilized from Fe-plaques of plant surfaces by EDTA while the desorption/mobilization of organoarsenic species (MMAA and DMAA) did not occur by the addition of EDTA. It might be because the co-precipitation of iron occurs only with inorganic arsenic, especially the arsenate. Phosphate uptake was not affected by EDTA though its concentration in CBE-extract was much higher than that of plant tissues. Iron uptake into *Spirodela polyrhiza* L. increased significantly ( $p > 0.05$ ) while its concentration in CBE-extract decreased by EDTA. The higher As(inorganic)/Fe ratios in plant tissue compared to CBE-extract indicate that the increase of inorganic arsenic uptake into the plant by EDTA is related to iron uptake.

## 学位論文審査結果の要旨

モハマド アジズ ラハマン氏より提出された学位論文に関して、各審査委員が個別に審査した後、平成 20 年 7 月 10 日に審査員による予備審査会を実施するとともに、平成 20 年 8 月 6 日に口頭発表会と論文審査委員会を開催し、以下のように判定した。

ヒ素汚染が深刻なバングラディッシュの湿地帯では、コスト面から有効な浄化法に限られる一方、自然サイクルを利用した環境修復技術としてファイトレメディエーションが注目を集めている。本研究では、同技術に基づく浄化法の確立を目的として、現地より採取した浮遊性水生植物の中からヒ素を選択的に高濃縮する種を探索するとともに、その取り込み挙動の詳細を検討した。一連の研究において、生体内へのメチルヒ素の吸収量は無機ヒ素よりも著しく少ないことを確認し、ヒ素はメチル化されると phytoavailability (植物可用性) が減少することを明らかにした。また、5 価無機ヒ素の根細胞への取り込みに関しては、化学的性質が類似するリン酸と拮抗することを示した。更に、植物細胞におけるヒ素の化学形態別分析法を確立するとともに、同法を適用して根近傍におけるヒ素の化学種分布を観測し、根表面に形成された鉄粒子層が細胞内へのヒ素の取り込みを阻害することを見いだした。

以上、本研究は、浮遊性水生植物のヒ素取り込み挙動やヒ素化学種の影響を解き明かしたものであり、湿地帯におけるヒ素汚染の浄化に有用な知見を提供する研究として博士(学術)の学位に値するものと判断した。