中文摘要

本論文之目的在於發展預測模式，以深入瞭解水域生態系中魚介類對金屬傳輸與生物攝取之結構與機制。全文嘗試建立水域生物體內總濃度、外部/內部效應濃度與不同標的組織濃度曲線之關係。論文之研究重點在於研析金屬之生物攝取及生物有效性之動態行為及毒性的時變性。預期目標為當這些預測模式成功建立時，可較精確評估金屬對於生物體之固有危害潛勢。

本論文結合急性毒及藥理動態學模式，推導一死亡率模式，以預測九孔暴露於含鋅水域之存活率。結合藥理學中曲線下面積 (area-under-curve, AUC)的觀念與生物動力速率模式，本論文發展一預測鋅對九孔相對生物有效性之機制模式。本論文建構一14 天之實驗室暴露試驗，以求取九孔軟體組織、殼與食物龍鬚菜之生物動力參數；本論文亦建構一7 天之急性毒試驗，以求取鋅對九孔之毒性參數LC_{50} (t)值與不同時間下九孔死亡時體內鋅濃度。

研究結果顯示，當九孔暴露於含鋅水體中，其96 小時及初始之LC_{50}值分別為1.1 及1.05 mg l^{-1}。由模式預測結果得知，當九孔死亡時，其體內毒性作用區之平衡鋅濃度為198 μg g^{-1}，而當水域中鋅濃度小於1 mg l^{-1}時，九孔之死亡率皆低於50%。對於暴露實驗之吸收相，以AUC為基礎之模式模擬結果，顯示鋅對九孔軟體組織及殼之相對生物有效性分別為71.04 ± 9.71%及68.44 ± 8.29%。靈敏度分析結果則顯示，九孔之成長率及排除速率常數為影響鋅對九孔相對生物有效性最主要的因子。

本論文以不同階段之族群數量方法，研析當水域鋅濃度逐漸增加時，對九孔族群動態所產生之影響。本論文重新分析7 天急性毒及28 天慢性毒生物試驗資料，以探究九孔暴露於不同鋅濃度下之存活率與成長率。此外，本論文應用以能

量為基礎之生物學方法將鋅對九孔之繁殖率予以模式化。利用這些資料，可求得九孔於不同階段之生命速率(即存活率、成長率與繁殖率)，並運用於投射矩陣模式之參數化。最後，本論文模擬0.03 至1 mg l^{-1}之7 種鋅暴露濃度下，九孔族群數量之時變性。模擬結果顯示當鋅暴露大於0.12 mg l^{-1}時，九孔之個體成長率會減緩；而當鋅暴露濃度在0.25 mg l^{-1}以上時，會對九孔之存活造成顯著影響。當鋅暴露濃度由0 mg l^{-1} (控制組)增至1 mg l^{-1}時，九孔之族群漸進成長率λ由1.00 降至0.9968，顯示當九孔暴露於鋅濃度較高的水域時，對九孔族群固有成長率有潛在的風險。

本論文結合Michaelis-Menten (M-M)形式通量及Fick形式動態質傳通量，推導Best方程式，用以量化地模擬當淡水性吳郭魚暴露於含砷水域中，鰓組織對砷之傳輸與生物攝取機制。本論文建構15 天之吸收/排除生物試驗以檢視砷在吳郭魚鰓組織之累積情形。利用吳郭魚之生理參數與體型參數，我們計算出以擴散模式為基礎之滲透係數。我們將相關文獻中實驗資料代入M-M方程式中，以曲線擬合法求取生物親和力參數與最大生物攝取通量。結果顯示，砷之生物轉換率與水域砷濃度呈線性關係。擬合所得之生物親和力參數與最大生物攝取通量分別為3.07 mg l^{-1}及2.17 mg l^{-1} d^{-1}，表示吳郭魚鰓組織對砷之生物親和力低，但卻有相當高的結合容量。而砷穿透吳郭魚鰓組織之滲透係數由起始時之1.42 μm d^{-1}，於兩個月後達到穩定值0.82μm d^{-1}，此結果顯示在生物攝取過程中，非平衡狀態是存在且需加以考量的。

由本論文之結果可知，當預測水域生物對金屬之傳輸與生物攝取時，動力及動態過程皆需加以考量。而論文中所提出的模式，皆可有效應用於評估當生物暴露於含金屬之水域生態系時，其相關之環境風險。

關鍵詞：九孔；吳郭魚；鋅；砷；毒性；生物有效性；族群動態學；生物攝取

Abstract

The goal of this dissertation is to develop predictive models to better understand the structure and mechanism of fish/shellfish involved in transport and biouptake of metals in aquatic ecosystems. The overall paradigm is trying to establish a relationship among total, external/internal effect concentrations and target concentration profiles of metals in aquatic organisms. Emphases of the dissertation are on metal biouptake/bioavailability dynamics and time-dependent toxicity. When these approaches successfully applied, the inherent hazard potential of heavy metals can be better estimated.

We developed a mortality model, by coupling an acute toxicity model and a pharmacodynamic model, to predict survival of abalone (*Haliotis diversicolor supertexta*) exposed to waterborne zinc (Zn). We developed a mechanistic model based on the pharmacological area-under-curve (AUC) concept associated with a biokinetic rate model to predict relative bioavailable Zn to abalone. A laboratory 14-day exposure experiment was conducted to obtain biokinetic parameters for soft tissue and shell of abalone and their food source, red alga *Gracilaria tenuistipitata* var. *liui*. A 7-day acute toxicity test was conducted to obtain $LC_{50}(t)$ and time course of lethal body burden of Zn in abalone.

Results of the mortality model demonstrate that 96-h LC_{50} and incipient LC_{50} for *H. diversicolor supertexta* exposed to Zn are 1.1 and 1.05 mg l^{-1}, respectively. Our predictions show that equilibrium lethal body burden at site of action is about 198 μg g^{-1}, whereas the mortalities never reach 50% when *H. diversicolor supertexta* exposed to Zn is ≤ 1 mg l^{-1}. In addition, the AUC-based model demonstrates that during uptake phase of the exposure experiment, estimated relative bioavailable Zn to soft tissue and shell of abalone were 71.04 ± 9.71% and 68.44 ± 8.29%, respectively. Sensitivity

analysis indicates that relative bioavailable Zn to abalone are greatly affected by growth rate and depuration rate constants of abalone.

We performed the stage-classified demographic method to investigate the effects of increased waterborne Zn concentrations on the population dynamics of abalone *Haliotis diversicolor supertexta*. We reanalyzed the results of a 7-d acute and a 28-d chronic toxicity bioassays to examine the survival and growth performances when exposing abalone to different levels of zinc stresses. An energy-based biological approach was adopted to model the effects of zinc on fecundity. These data provided stage-specific schedules of vital rates that were used to parameterize a projection matrix model for abalone. Simulations were carried out to produce temporal population abundance changes under seven exposure concentrations ranged from 0.03 to 1 mg l^{-1} Zn. Model manipulations indicate that a reduction of individual growth rate is observed at an exposed Zn concentration greater than 0.12 mg l^{-1}, whereas the significant influence of survivorship is occurred until the Zn concentration reached 0.25 mg l^{-1}. The asymptotic population growth rate decreases from λ = 1.00 for the control group to λ = 0.9968 for abalone population exposed to 1 mg l^{-1} Zn, indicating a potential risk of population intrinsic growth rates for abalone exposed to higher levels of waterborne Zn.

We coupled the Michaelis-Menten (M-M) type flux and the Fick's type of dynamic mass transfer flux to arrive at the Best equation to quantitatively model the transport and biouptake mechanism of the gills of freshwater tilapia (*Oreochromis mossambicus*) exposed to waterborne arsenic (As). A 15-day uptake/depuration after two months, indicating the non-equilibrium aspects of biouptake processes is involved. bioassay was conducted to examine the accumulation kinetics of As in tilapia gills. A diffusion-based permeability was calculated using the physiological and

allometric-related parameters. The bioaffinity parameter and the limiting uptake flux in M-M equation were acquired by fitting the experimental values from published literature. A linear relationship between As bioconversion rate and As concentration in ambient water was obtained. The fitted bioaffinity parameter and limiting uptake flux were 3.07 mg l^{-1} and 2.17 mg l^{-1} d^{-1}, respectively, suggesting a low As binding affinity of tilapia gills, yet a relative high binding capacity was obtained. The As permeability through tilapia gills membrane decreases from 1.42 μm d^{-1} to a steady-state value of 0.82 μm d^{-1}.

Our studies suggest that we have to take into account both kinetic and dynamic processes in predicting the transport and biouptake of metals to aquatic animals. The proposed models have potential applications in assessing the environmental risks associated with aquatic ecosystems of metal exposure.

Keywords: Abalone; Tilapia; Zinc; Arsenic; Toxicity; Bioavailability; Population dynamics; Biouptake