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**INVESTIGATIONS ON THE ENVIRONMENTAL  
SIGNIFICANCE AND THE BIOAVAILABILITY OF  
HEAVY METALS IN SOILS**

**BY**

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**In The Name of ALLAH The Compassionate, The Most Merciful**

**TO**

**MY FATHER, MOTHER AND FAMILY**

## **DECLARATION**

I hereby declare that the work reported in this Thesis is my own work unless specified and duly acknowledged by quotations

Date: 20.02.2002

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## ABSTRACT

*Heavy metals contaminated soils pose a continuing and increasing threat to human health, the increased inputs of heavy metals from the widespread disposal of industrial wastes have created an increased attention on the issue of their fate, bioavailability and environmental significance. These investigations were carried mainly to study the solid - solution phase distribution, chemical speciation and plant uptake of cadmium (Cd), lead (Pb), nickel (Ni) and zinc (Zn) in three contrasting soils amended with Cd, Pb, Ni and Zn nitrate salts at rates of 5-40 mmol/kg for incubation time varied between 3-15 month. Although heavy metal cations adsorbed to the soils at low concentrations, increasing their mass loading resulted in positive correlations with the adsorption values of these metal cations to the soil solid particles. Soil type had significant effect on the solid - solution equilibrium of the added metals. The studied metals are varied in their adsorption and desorption characteristics, Zn reported the highest solution concentration than that of Ni, Cd and Pb in the three soils while Pb was the most adsorbed metal. Metals concentration in soil solution and are positively correlated with residence time.*

*The speciation of metals in soils is essential in understanding their chemical and biological interactions in the soil environment. The solid phase speciation of Cd, Pb, Ni and Zn in soils amended with such metals at rate of 5mmol/kg was estimated by sequentially extracting the treated soils with 1M NH<sub>4</sub>OAc to dissolve the water soluble and the exchangeable metals, with 0.125 M Cu (OAc)<sub>2</sub> to dissolve complexed metals and with 1M HNO<sub>3</sub> to dissolve the precipitated and residual metals. The speciation of the added metals was greatly influenced by soil type and residence time.*

*The soil solution chemistry of heavy metal is of great importance in assessing their bioavailability and estimates their toxicity. MINTEQA2 version 3.0 used to explore the*

*speciation of the studied metals the in solution of the three soils revealed the majority of metals as the free ionic Cd<sup>2+</sup>, Pb<sup>2+</sup>, and Ni<sup>2+</sup>and Zn<sup>2+</sup>forms with varying proportions of the other different inorganic species.*

*The bioconcentration of heavy metals by plants is the major route for metal contaminants to enter the human body. Greenhouse experiments were conducted to study Cd, Pb, Ni and Zn uptake from soils amended with these metals at levels from 0-20mmol/kgsoils, using lettuce (*Lactuce sativa*) as bio-monitor. This concentration range was chosen based on the ability of lettuce to tolerate such metal rates. Significant positive correlations were obtained between the bioconcentration of Cd, Pb, Ni and Zn by plant and the metal amendments, the exchangeable chemical fraction and the residence time. The relative bioavailability of the studied heavy metals is in this order Zn > Ni> Pb>Cd.*

*The soil-plant-man exposure pathway is identified as the major exposure pathway to contaminants. The estimated plant- soil bioconcentration factor for Cd, Pb, Ni and Zn was found to be slightly elevated with the residence time of the contaminant. Thus the study concluded that amount of Cd, Pb, Ni and Zn contaminants available for human intake is subjected to gradual elevation in response to the metal mass loading and the length of time soils are exposed to the specific contaminants.*

## ABSTRAK

Tanah yang tercemar oleh logam berat merupakan satu ancaman yang meningkat dan berterusan terhadap kesihatan manusia. Input logam berat yang bertambah berpunca daripada pembuangan bahan buangan industri telah membangkitkan tumpuan yang lebih terhadap isu kesudahannya, keterbiosediaannya dan juga kesan terhadap sekitaran. Penyelidikan yang dijalankan bertujuan terutamanya mengkaji agihan fasa pepejal-larutan, pengspesian kimia dan pengambilan kadmium, plumbum, nikel dan zink oleh tumbuhan dalam tiga jenis tanah yang berlainan. Tanah-tanah ini telah ditambah dengan garam nitrat Cd, Pb, Ni dan Zn pada kadar 5-40 mmol/kg untuk masa inkubasi yang berlainan antara 3-15 bulan. Walaupun kation logam berat menyerap pada tanah pada kepekatan rendah, penambahan tokokan jisim menghasilkan korelasi positif dengan nilai penyerapan kation logam-logam ini. Jenis tanah mempunyai kesan yang bererti kepada keseimbangan pepejal-larutan bagi logam yang ditambah. Logam-logam yang dikaji ini mempunyai ciri-ciri penjerapan dan penyahjerapan yang berbeza dengan Zn dilaporkan membentuk kepekatan larutan yang tertinggi berbanding dengan Ni, Cd dan Pb dalam ketiga-tiga jenis tanah manakala Pb merupakan logam yang paling terjerap.

Pengspesian logam dalam tanah adalah perlu untuk pemahaman salingtindakan kimia dan biologinya dalam persekitaran tanah. Pengspesian Cd, Pb, Ni dan Zn dalam fasa pepejal bagi tanah yang ditambah dengan logam berkenaan pada kadar 5mmol/kg telah dianggarkan melalui pengestrakan tanah-tanah tadi secara urutan bermula dengan 1M NH<sub>4</sub>OAc untuk membuang logam logam yang larut dalam air serta boleh ditukarganti, dengan 0.125 M Cu(OAc)<sub>2</sub> untuk membuang logam yang telah dikomplekskan dan dengan 1 M HNO<sub>3</sub> untuk melarutkan logam yang telah mendak dan berbakri. Pengspesian logam-logam yang ditambah dipengaruhi dengan kuat oleh jenis tanah dan masa pendudukan.

Kimia larutan tanah logam-logam berat adalah sangat mustahak dalam menentukan keterbiosediaannya dan menganggarkan keracunannya. MINTEQA.2 versi 3.0 yang digunakan untuk meneroka pengspesian logam dalam larutan yang dikaji daripada 3 jenis tanah menunjukkan bahawa kebanyakkan logam wujud dalam bentuk ion  $Cd^{2+}$ ,  $Pb^{2+}$ ,  $Ni^{2+}$  dan  $Zn^{2+}$  bebas dengan kadar berbagai bagi spesies bukan organik yang lain.

Biokepekatan logam berat oleh tumbuhan merupakan laluan utama untuk logam pencemar memasuki tubuh manusia. Eksperimen rumah hijau dijalankan untuk mengkaji pengambilan Cd, Pb, Ni dan Zn dari tanah yang telah ditambah dengan logam tersebut pada kadar 0-20 mmol/kg tanah menggunakan lettuce sebagai biomonitor.

Julat kepekatan ini dipilih kerana lettuce mempunyai toleransi terhadap kadar logam ini. Korelasi positif yang bererti didapati antara pembiokepekatan Cd, Pb, Ni dan Zn oleh tumbuhan dan penokokan logam, pecahan kimia yang boleh ditukarganti dan masa pendudukan. Keterbiosediaan relatif untuk logam berat yang dikaji menurut tertib  $Zn > Ni > Pb > Cd$ .

Laluan pendedahan tanah-tumbuhan-manusia dikenalpasti sebagai laluan utama kepada pencemar. Faktor biokepekatan tumbuhan-tanah yang dianggar untuk Cd, Pb, Ni dan Zn didapati meningkat sedikit dengan masa pendudukan pencemar. Kesimpulan daripada kajian ini ialah amaun pencemar Cd, Pb, Ni dan Zn yang tersedia ada bagi pengambilan oleh manusia tertakluk kepada peningkatan sederhana sebagai rangsangan kepada tokokan jisim logam dan tempoh masa tanah didekahkan kepada pencemar spesifik.

## خلاصة البحث

لقد أكدت الدراسات البيئية السابقة أن التربة الملوثة بالعناصر الثقيلة مثل خطرًا دائمًا على صحة الإنسان. وإن زيادة وجود المعادن الثقيلة الناتجة من المخلفات الصناعية قد أدت إلى زيادة الاهتمام بأثرها البيئي والحيوي. قد أجريت هذه البحوث على ثلاثة أصناف مختلفة من التربة. تمت معالجتها باملاح التربات لعناصر الكالسيوم، والرصاص، والنikel، والخارصين. بمعدلات ٥ - ٤٠ مليمول / كجم لفترات زمنية تراوحت بين ٣ - ١٥ شهرًا. وقد هدفت البحوث لدراسة توزيع العناصر في التربة في طوريها الصلب والمحلول، التصنيف الكيميائي والأثر الحيوي للمعادن الأربعه الأنف ذكرها.

بالرغم من أن امتراز التربة لكاتيونات المعادن الثقيلة المدروسة قد تم بتركيز بسيط، إلا أن زيادة تركيزها قد أدت إلى زيادة طردية في قيم وحجم الامتراز. وقد توصلت الدراسة إلى أن نوعية التربة تؤثر تأثيراً واضحًا على التوازن الكيميائي ( محلول - صلب ) للمعادن المضافة إليها.

وقد تفاوتت خصائص عملية الامتراز والعملية العكسية لها من معدن إلى آخر. حيث توصلت الدراسة إلى أن عنصر الخارصين له تركيز أعلى من تركيزات النikel، والكادميوم، والرصاص في محلول التربة بينما ثبت أن الرصاص هو أكثر العناصر امترازاً وأن تركيز المعادن له علاقة طردية مع الفترة الزمنية التي تمت فيها معالجة التربة.

كما أن لوجود كيميات المعادن الثقيلة في محلول التربة أهمية كبيرة في تقدير أثراها الحيوي وحساب درجة سوميتها. وقد تم استخدام البرنامج الحاسوبي (*MINTEQA2 Version 3.0*). حيث تمت تغذية هذا البرنامج بالمعلومات الخاصة بالظروف الشيرمودايناميكية الخاصة بكل تربة، وذلك بغرض استبيان المكونات غير العضوية في محلولها.

وقد كانت خلاصة هذا الاستبيان أن معظم المكونات الكيميائية للمعادن المدروسة في محلول التربة موجودة على صورتها الأيونية مضافاً إليها نسب متفاوتة لمكونات أخرى غير عضوية تختلف نسبتها من معدن إلى آخر. ومن المعروف أن التركيز الحيوي للمعادن الثقيلة في النبات هو المعيار الرئيسي لهذه المعادن إلى جسم الإنسان.

ولدراسة التركيز الحيوي للمعادن المذكورة فقد قامت الدراسة بإجراء تجارب بستانية حيث وضعت التربة الملوثة داخل القصاري. وقد استخدمت أربعة مستويات للتلوث ( ٥ - ٢٠ / كجم )

لكل معدن على حدة. واستخدام فيها نبات الخص بحكم طبيعته الحيوية. وقد خلصت تجربة القصارى إلى أن الاتاحة الحيوية النسبية للمعادن الثقيلة موضع الدراسة كانت كما يلى:  
الخارصين أكبر من النيكل أكبر من الرصاص أكبر من الكادميوم.  
(خارصين < نيكل < رصاص < كادميوم)

إن الملوثات تصل إلى الإنسان عن طريق الممر الحيوي (ترية — نبات — إنسان). وقد زاد عامل التركيز الحيوي من التربة إلى النبات للمعادن المذكورة زيادة طفيفة عند زيادة الفترة الزمنية لوجود هذه المكونات داخل التربة.

لذلك توصلت هذه الدراسة إلى أن استنتاج كميات ملوثات الكادميوم / الرصاص / النيكل / والخارصين المتاحة للأخذ بواسطة الإنسان معرضة إلى الزيادة التدريجية بفعل زيادة تركيز الملوث المعين وطول فترة بقائه داخل التربة.

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