Alina Kabata-Pendias

Arun B. Mukherjee

Trace Elements from Soil to Human

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With 26 Figures and 209 Tables

Springer

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Preface

The understanding of fundamental principles and phenomena that control the transfer of trace elements in soil-plant-human chain can contribute to the protection of the environment and human health. Many books, articles, and reports have already described both fundamental and/or detail problems related to these topics.

The intention of the authors is to provide up-to-date and selected interdisciplinary data for the concise presentation of existing knowledge on trace element transfer in the food chain, from soil to human. To accomplish this, the inclusion of appropriate data has been necessary. This book inevitably leaves publications of many investigators' uncited. The authors regret that such approach was necessary.

This volume is composed of two parts. Part I – *Biogeochemistry of the Human Environment* – presents fundamental information on biogeochemical properties of environmental compartments (soil, water, air, plants, humans) concerning trace elements. Part II – *Biogeochemistry of Trace Elements* – provides detailed data of the behavior and the occurrence of trace elements in the environment. There is a close relationship in the biogeochemical behavior between elements and their position in the Periodic System. Therefore, the format of this book follows the elemental sequences of the contemporary Periodic Table. The book provides data on the production usage, and on the occurrence of trace elements in soils, waters, air, plants, and humans (animals). Environmental stress, and biological functions of these elements are widely discussed.

The authors hope that information, presented in this book, will encourage young scientists to undertake further studies for better understanding of all factors that influence cycling of trace elements in a given ecosystem, and to develop the most effective methods for the effective remediation of contaminated sites. Finally, this knowledge will improve the assessment of health and ecological risk. The authors will be satisfied if this book fires the imagination of some readers and encourages them to study the biogeochemistry of trace elements in greater depth.

Alina Kabata-Pendias, Arun B. Mukherjee
January 2007

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The easy access to references enabled us to use a versatile database with the assistance of the library staff at: The Central Agricultural Library (CBR) in Puławy, The Library of the Polish Geological Institute in Warsaw, and The Viikki Science Library of the Helsinki University in Helsinki.

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January 2007

Past to Future

Inscription on the Sybilla's Temple
erected 1801 in Puławy, Poland

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Presentation of Data

Basic units of the International System (SI Unit) are used in this book. Mean contents refer to arithmetic mean value, unless otherwise specified.

The concentrations of a trace element in soil, plant, animal, and human samples are based on the total content by weight of the element in air-dried or oven-dried (at 40 °C) material. Otherwise it is indicated as follows:

AW – ash weight
FW – fresh weight or wet weight
BW – body weight

All data are given for topsoils, unless otherwise indicated. If not identified, a content of an element in environmental samples is given as, so called "total", i.e., measured in totally digested sample or measured directly in a sample.

Units

Mt – million metric tons (10^3 kt)
kt – thousand metric tons (10^3 t)
t – metric ton (10^3 kg)
kg – kilogram (10^3 g)
g – gram (10^{-3} kg)
mg – milligram (10^{-3} g)
µg – microgram (10^{-3} mg)
ng – nanogram (10^{-3} µg)
pg – picogram (10^{-3} ng)
fg – femtogram (10^{-3} pg)
pm – picometer (10^{-12} m); 1 Å = 100 pm
ha – hectare (10 000 m^2)
l – liter (1 dm^3)
m^3 – cubic meter (10^3 dm^3)
Bq – becquerel
mBq – millibecquerel
Ci – curie; 1 Ci = 3.7×10^{10} Bq
mCi – millicurie
nCi – nanocurie
pCi – picocurie

Symbols

°C – temperature in Celsius degrees
cmol(+) kg^{-1} – centimoles of positively charged ions (cations) in 1 kg of sample (formerly meq / 100 g; 1 cmol(+) kg^{-1} = 1 meq / 100 g)
Eh – redox potential (volts, millivolts)
pH – negative logarithm, base 10, of hydrogen ion concentration (activity)
M – molar concentration of solution of an element (compound)
$t_{1/2}$ – half life-time of an radionuclide
K_d – dissociation constant: product of the activities of cations and anions divided by the activity of the unionized electrolyte

The Periodic Table of Elements

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 * La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 ** Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt									

* Lanthanide series	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
** Actinide series	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

The symbols given in thin letters indicate the elements not occurring naturally in the environment.

Glossary (Abbreviations, Acronyms)

AAAc	ammonium acetic acid
AAAcEDTA	mixture of ammonium acetic acid with Na_2EDTA
AAP	acid available particulate
AD	Alzheimer disease
ADI	acceptable daily intake
AMAP	Arctic Monitoring and Assessment Programme
A/N	ratio of anthropogenic to natural sources
AROMIS	Assessment and Reduction of Heavy Metal Inputs into Agro-Ecosystems
ASB	alkaline-stabilize biosolids
ASM	artisanal and small-scale gold mining
ATSDR	Agency for Toxic Substances and Disease Registry
AHM	Asian herbal medicines
BAC	biological absorption coefficient
BAT	biological tolerance values at workplace
BC	Before Christ
BCF	biological concentration factor
BIM	biologically induced minerals
B-x	blood level of a given element
Cc	continental crust concentration of element
CEC	cation exchangeable capacity
CLPP	community level physiological profile
Clarke	Clarke's data for mean values for chemical elements in a given geological material
COPR	chromite-ore processing residue
Cw	dissolved concentration of element in water
Cw/Cc	ratio of element concentration in water to its content in the continental crust
DBT	dibutyltin
DDI	daily dietary intake
DM-X	dimethylated element
DMT	dimethyltin
DNA	de(s)oxyribonucleic acid, carrier of genetic information

DOC	dissolved organic carbon
DOM	dissolved organic matter
DTPA	diethylenetriaminepentaacetic acid
DU	depleted uranium
EC	European Commission
EDTA	ethylenediaminetetraacetic acid
EF_c	enrichment factor-ratio of element concentration in air to its content in the Earth's crust, normalized to the reference element (Al)
EPA	Environmental Protection Agency
ESADDI	estimated safe and adequate daily dietary intake
FA	fly ash
FAO	Food and Agriculture Organization of the United Nation
FDA	Food and Drug Administration
GIS	geographical information system
GEMS	Global Environment Monitoring System
GERM	Geochemical Environmental Reference Methods
HA	humic acids
HELCOM	Helsinki Commission
HI	hazardous index
HDL	high density lipoproteins
HRE	heavy rare earth
IHC	interactive health communication
IAEA	International Atomic Energy Agency
IARC	International Agency for Research on Cancer
IBA	index of bioaccumulation
ICRP	International Commission on Radiological Protection
IDD	Iodine Deficiency Disorders
IHC	Interactive Health Communication,
IHR	International Health Regulation
IPCS	International Program on Chemical Safety
IUPAC	International Union of Pure and Applied Chemistry
IUR	International Union of Radioecology
JECFA	Joint FAO/WHO Expert Committee on Food Additives
KD	Keshan disease
KBD	Kashin-Beck disease
LDL_{50}	lethal dose, the simple dose of an element which causes the death of 50% of a population in a specific period of time
LDL_0	the lowest lethal dose

LDH	low density lipoproteins
LOAEL	lowest-observed-adverse-effect level
LRE	light rare earth
LTI	lowest threshold intake
MAC	maximum allowable concentration
MAK	maximum concentration of a chemical substance on air at the workplace (after German)
MBT	monobutyltin
MCL	maximum concentration level
MCRA	Monte Carlo Risk Assessment
Me-X	methylated metal
MF	modifying factors
ML	maximum level
MPL	maximum permissible limit
MMT	methylocyclopentadienyl manganese tricarbonyl
MM-X	monomethylated metal
MND	motor neuron disease
MPC	maximum permissible concentration
MSW	municipal solid waste
MTD	maximum tolerable dose
NAS	National Academy of Sciences
NIOSH	National Institute for Occupational Safety and Health
NOAEL	no-observed-adverse-effect level
NOEC	no-observed-effect concentration
OEL	occupational exposure limit
OECD	Organization for Economic Cooperation and Development
OTC	organo-tin compound
OSHA	Occupational Safety and Health Administration (in USA)
PbB	lead in blood
PCBs	polychlorinated biphenyls
PGM	platinum group metal
PM	particulate matter (e.g., PM10 = particle 10 μm in diameter)
PT_{50}	phytotoxicity threshold corresponding to 50% growth retardation
PTDI	provisional tolerable daily intake
PTWI	provisional tolerable weekly intake
PMTD	provisional maximum tolerable daily intake
RDI	recommended daily intake
RDA	recommended dietary allowance
REE	rare earth element
RfD	reference dose
RNA	ribonucleic acid, structural element of the cytoplasm and cell nucleus

SeCys selenocysteine
SEFcrust soil enrichment factor in relation to crust mean concentrations (ratio of mean soil content to crustal content)
SeGSH selenoglutathione
SeMC selenomethylocysteine
SeMet selenomethionine
SOD superoxide dismutase
SETAC Society of Environmental and Toxicological Chemistry
SOM soil organic matter
SPTF soil-plant transfer factor

TAV trigger action value
TBT tributyltin
TF transfer factor (ratio of an element content in plant to its concentration in soil)
TDI total dietary intake
TDS Total Diet Study
TI tolerable intake
TIC trace inorganic contaminants
TLV threshold limit value
TUIL tolerable upper intake level
TWA time weighted average

UF uncertainty factors
UL tolerable upper intake level
UNEP United Nations Environmental Program
UNESCO United Nations Educational, Scientific and Cultural Organization
UPAC International Union of Pure and Applied Chemistry
USEPA US Environmental Protection Agency
USGS United States Geological Survey

WHO World Health Organization
W/S worms/soil concentration ratio

Introduction

The quality of human life depends on the chemical composition of food and of the surroundings. Recent improvements and new methods in analytical chemistry and increasing fields of environmental investigations have added substantially to our knowledge of the biogeochemistry of trace elements. In the last three decades there has been a real "explosion" of research data and various publications on occurrence and behavior of almost all trace elements including both elements of known and unknown physiological functions in organisms. In order to realize the vast significance of the biogeochemistry of trace elements, it is essential to gather the knowledge acquired over this period into one comprehensive compilation.

Soil is not homogenous, and the microscale heterogeneity creates a real problem in representative sampling. Also the variability in sampling procedure from plants and other organisms is a subject of concern and this has made the assessment and evaluation of some data almost impossible. Therefore, quantitative comparisons of analytical data for soils, plants and human/animal tissues have often been difficult.

Different chemical preparations of samples (e.g., HNO_3 microwave decomposition, ashing with aqua regia, total digestion) as well as different instrumental methods used for the determination of elements (e.g., ICP-MS, ICP-OES, F-AAS, ETA-AAS) have an influence on final results. Luckily, analytical quality assurance and the use of reference materials have decreased uncertainties of analytical data. Therefore, each measurement of trace elements builds up a database and contributes to a better understanding of their overall distribution and behavior in given media and in the total environment.

A better understanding of the biogeochemical processes that control trace element cycling and comprehensive dataset on the abundance of trace elements in abiotic and biotic environmental compartments may be a key to better management of trace elements in the environment that is prerequisite to sustainable land use and, presumably, to diminish health risks due to trace inorganic pollutants.

The term "trace elements" has never been defined precisely. It has been used in geochemistry for chemical elements that occur in the Earth's crust in amounts less than 0.1% (1 000 mg kg^{-1}) and also in biological sciences, for elements at similar concentrations. Therefore some elements that are "trace" in biological materials are not "trace" in terrestrial ones (i.e., iron). The term "trace elements" is related to their abundance and includes elements of various chemical properties: metals and metalloids.

Common trace cations descriptors are "trace metals" and/or "heavy metals". The trace metalloids are simple "trace elements". The other terms: "micronutrients", "essential elements", and "toxic elements" are related to their physiological functions and are rather confusing since their effects on organisms and health depend upon concentrations. All these terms are inadequate, and a great deal of confusion has occurred in the literature where authors have been imprecise in their use of these terms. Especially the term "heavy metals" has recently become a subject of a broad discussion that emphasizes its non-precise definition. Duffus (2002) has written: "Over the past two decades, the term 'heavy metals' has been widely used... and related to chemical hazards. It is often used as a group name for metals and semimetals (metalloids) that have been associated with contamination and potential toxicity or ecotoxicity." This term is based on various criteria (i.e., atomic weight, atomic number, density, chemical properties etc.). Thus, the inconsistent use of the term "heavy metals" reflects inconsistency in the scientific literature. This term has never been defined by any authoritative body, such as IUPAC.

Other terms that need to be defined are related to chemical speciation and fractionation of elements. Thanks to new developments in analytical instrumentation and methodology, the identification and measurement of element species in a particular system is possible. In an attempt to end the confusion regarding the usage of the term speciation, three IUPAC Divisions collaborated to consider the issue (Templeton et al. 2000). Their definitions for the recommended use of term speciation are following:

- *Chemical species.* Chemical elements: specific form of an element defined as to isotopic composition, electronic or oxidation state, and/or complex or molecular structure
- *Speciation analysis.* Analytical chemistry: analytical activities of identifying and/or measuring the quantities of one or more individual chemical species in a sample
- *Speciation of an element, speciation.* Distribution of an element among defined chemical species in a system
- *Fractionation.* Process of classification of an analyte or a group of analytes from a certain sample according to physical or chemical properties

Part I Biogeochemistry of the Human Environment

Chapter I-1 The Biosphere

Chapter I-2 Soils

Chapter I-3 Waters

Chapter I-4 Air

Chapter I-5 Plants

Chapter I-6 Humans

The Biosphere

The biosphere, also called the ecosphere, is the natural environment of living organisms and is the complex biological epidermis of the Earth whose dimensions are not precisely defined. It consists of the surficial part of the lithosphere, a lower part of the atmosphere, and the hydrosphere. Several ecosystems have been developed within the biosphere. Each ecosystem is a fundamental division of the total environment consisting of living organisms in a given area and having a *balanced* cycling of chemical elements and energy flow.

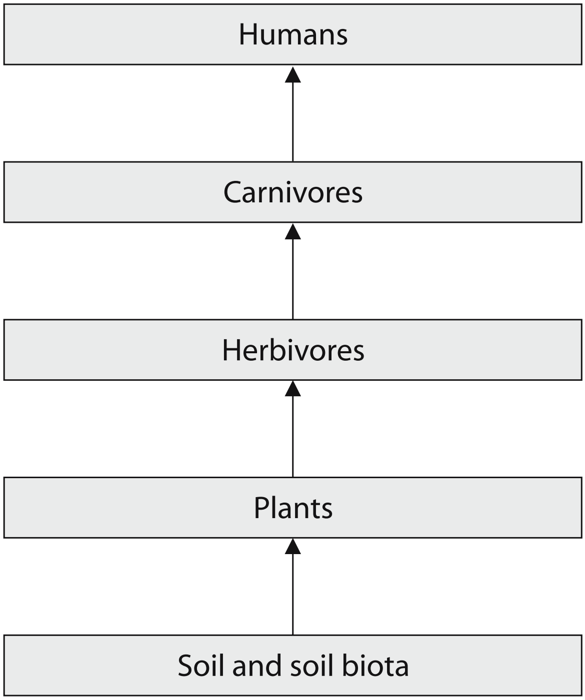
Among the principal resources of which man disposes, are terrestrial ecosystems consisting of soil and water, and associated animal and plant life. Ecosystems are functional environmental units, having *balanced* cycles of chemical elements, organic materials and energy flow. There is a homeostatic interrelationship between the non-living media (abiotic compartments) and the living organisms (biotic compartments). However, a significant part of the ecosystems has already been considerably modified by humans, and these processes will continue.

The energy for life is derived from the radiant energy of the sun, which drives the chemical reaction of photosynthesis. The other sources of energy, e.g., geothermal, gravitation, and electrical, are of negligible importance in the total energy flow, but may determine specific conditions of some ecosystems.

Organisms have adjusted during the course of evolution and life to the chemistry of their environment and have developed their biochemistry in close connection to the composition of the natural environment. These phenomena have been easily observed, mainly in microorganisms and plant populations that have evolved tolerance to high concentrations of trace elements either in natural geochemical provinces, or under man-induced conditions.

Most of the chemical elements for life on the land are supplied mainly from the soil overlying the surficial lithosphere (Fig. I-1.1). Although mechanisms of biological selection of chemical elements allow plants to control, to a certain extent, their chemical composition, this barrier is somewhat limited in respect to trace elements. Therefore concentrations of trace elements in plants are often positively correlated with the abundance of these elements in growth media. This creates several problems for plants, animals and humans associated either with deficiency or with excess. Thus, questions of how and how much of an element is taken up by organisms have been hot topics of research in recent decades. Usually the quantitative differences between essential amounts and biological excesses of trace elements are very small. A proper balance between trace and major elements plays a significant role in biochemical processes.

Fig. I-1.1.
The transfer of chemical elements in schematic terrestrial trophic chain



The bioavailability of these elements is variable and is controlled by specific properties of abiotic and biotic media as well as by physical and chemical properties of a given element.

The biochemical functions of essential trace elements are already known. A great number of trace elements are known to have a biological role, often as cofactors or part of cofactor in enzymes and as structural elements in proteins. Some of them also are used in several processes of electron transfer. Non-essential elements seem to be involved in vital processes but their biochemical functions are not yet understood. The essentiality of other trace elements, possible at very minor concentrations, may be discovered in the future. Most of trace elements that are essential to humans are also essential to plants. Unfortunately, contents of most elements that may be harmful to humans and animals are not toxic to plants. This has created an increased transfer of some elements in the food chain.

The survival of mankind is a story of food. Both, lack of food and bad quality of food have created throughout the centuries serious problems for people. Nowadays it is calculated that over 3 billion people worldwide suffer from either deficiency or toxicity of some trace elements.

Here is a place to remind Paracelsus' (1538) statement:

> All substances are poisonous, there are none which is not a poison; the right dose is what differentiates a poison from a remedy.

The anthroposphere. Many ecosystems have been considerably modified by humans and therefore it has become necessary to distinguish the anthroposphere – the sphere of man's settlement and activity. The anthroposphere does not represent a separate sphere, but may be applied to any part of the biosphere that has been changed under an influence of technical civilization.

While geological, geochemical and biological alterations of the lithosphere have been very slow, changes introduced or stimulated by humans have been accumulated extremely quickly in recent decades of the past century. Anthropogenic changes, associated mainly with chemical pollution, lead most often to a degradation of the natural human environment. Among all chemical pollutants, trace elements are of a special ecological, biological and health significance.

The production of energy and the consumption of natural resources are the main source of trace elements as contaminants. However, agricultural activities and especially application of sewage sludge, manure, mineral fertilizers (NPK), and pesticides also contribute significantly to the trace metal status of agroecosystems.

Bowen (1979) has suggested that when the rate of mining of a given element exceeds the natural rate of its cycling by a factor of ten or more, the element should be considered a potential pollutant. Thus, the potentially most hazardous trace metals to the biosphere may be: Ag, Au, Cd, Hg, Pb, Sb, Sn, Te, W. Also those elements that are essential to plants and humans, such as: Cr, Cu, Mn, and Zn, may be released, in some regions, in excessive amounts.

Soils

Soil is not only a part of the ecosystem but also occupies a basic role for humans, because the survival of man is tied to the maintenance of its productivity. Soil functions as a filtering, buffering, storage, and transformation system protect against the effects of trace element pollution. Soil is effective in these functions only as long as its capacity for cation exchange and its biological activity are preserved. The frequent association of trace element pollution with acid deposition (mainly S, NO_x, and HF) greatly complicates the overall effects in the environments.

Soil is the main source of trace elements for plants both as micronutrients and as pollutants. It is also a direct source of these elements to humans due to soil ingestion affected by "pica-soil", geophagia, dust inhalation, and absorption through skin.

The soil-plant transfer of trace elements is a part of chemical element cycling in nature. It is a very complex process governed by several factors, both natural and affected by humans. Thus, the prediction of trace element uptake by plants from a given growth medium should be based on several biotic and abiotic parameters that control their behavior in soil.

Soils contain trace elements of various origins: *(i) lithogenic* – inherited from the lithosphere (parent material), *(ii) pedogenic* – from lithogenic sources but forms changed due to soil-forming processes, and *(iii) anthropogenic* – elements deposited onto and/or into soils as results of human's activities. Soil processes and anthropogenic factors control the behavior of all these elements (Table I-2.1). It has been assumed that the behavior of trace elements in soils and in consequence their phytoavailability differ as to their origin. Several recent reports have indicated that regardless of the forms of the anthropogenic trace metals, their availability to plants is significantly higher than those of natural origin (Kabata-Pendias and Pendias 2001).

Soils of several regions of the world (and especially of Europe) have been and will be in the future subjected to mineral fertilization, pesticide application, waste disposal, and industrial pollution. All these human activities affect both chemical and physical soil properties, and will lead to changes in the behavior of trace elements in soils. The impact of soil acidification, alkalization, salinity and losses of SOM on the uptake of trace elements by vegetation, particularly by crop plants, have already become serious issues for the environment and for human health.

Weathering. Weathering, the basic soil-forming process is a complex set of interactions between the lithosphere, atmosphere, and hydrosphere that occur in the biosphere and are powered by solar energy. It can be described chemically as the processes of