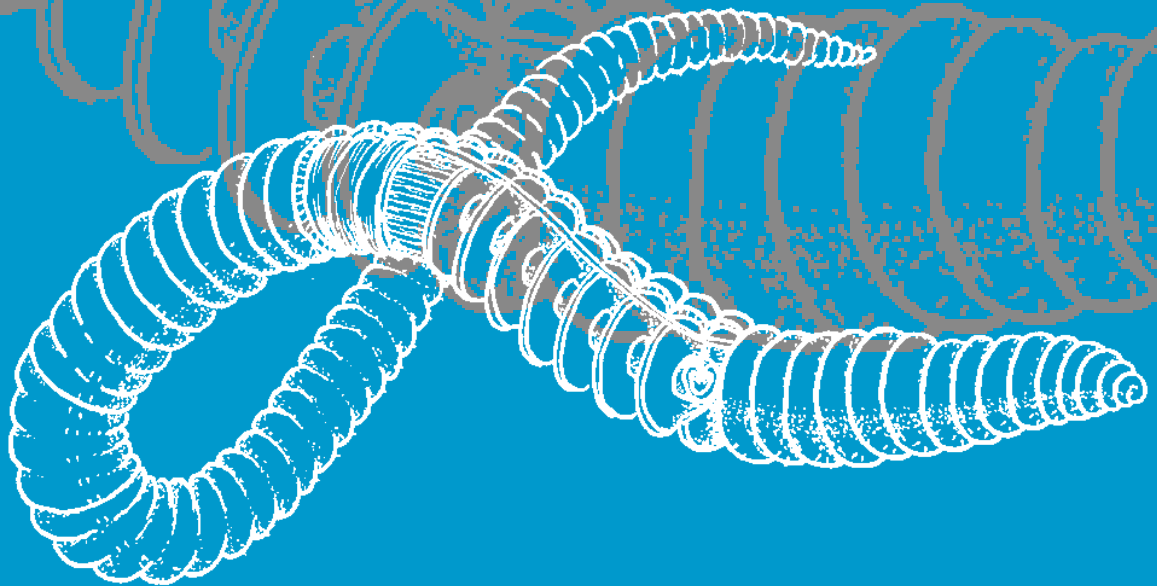


Worming your way into bioavailability

Modelling the uptake of organic
chemicals in earthworms



Tjalling Jager



Universiteit Utrecht

Institute for Risk Assessment Sciences



Worming your way into bioavailability

Modelling the uptake of organic
chemicals in earthworms

Wurm je een weg door biobeschikbaarheid

Het modelleren van de opname van
organische stoffen in regenwormen

(met een samenvatting in het Nederlands)

Proefschrift ter verkrijging van de graad van doctor aan de
Universiteit Utrecht op gezag van de Rector Magnificus, Prof. Dr.
W.H. Gispen, ingevolge het besluit van het College voor Promoties
in het openbaar te verdedigen op woensdag 11 juni 2003 des
middags te 14.30 uur

door

Dirk Tjalling Jager

geboren op 12 augustus 1969, te Purmerend

Promotor: Prof. Dr. C.J. van Leeuwen (Institute for Risk Assessment Sciences, Utrecht University)

Co-promotor: Dr. J.L.M. Hermens (Institute for Risk Assessment Sciences, Utrecht University)

Worming your way into bioavailability. Modelling the uptake of organic chemicals in earthworms / Tjalling Jager

ISBN 90-393-3384-X

The research described in this thesis was partly carried out at the National Institute for Public Health and the Environment (RIVM, Bilthoven, The Netherlands), Laboratory for Ecotoxicology, in cooperation with the Institute for Risk Assessment Sciences (IRAS, Utrecht, The Netherlands). Part of this research was commissioned by the Ministry of Housing, Spatial Planning and the Environment (VROM, The Hague, The Netherlands).

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“To sum up, as chance does not determine the manner in which objects are drawn into the burrows, and as the existence of specialized instincts for each particular case cannot be admitted, the first and most natural supposition is that worms try all methods until they at last succeed; but many appearances are opposed to such a supposition. One alternative alone is left, namely that worms, although standing low in the scale of organization, possess some degree of intelligence. This will strike every one as very improbable; but it may be doubted whether we know enough about the nervous system of the lower animals to justify our natural distrust of such a conclusion. With respect to the small size of the cerebral ganglia, we should remember what a mass of inherited knowledge, with some power of adapting means to an end, is crowded into the minute brain of a worker-ant.”

Charles Darwin (1881)

The formation of vegetable mould, through the action of worms, with observations on their habits.

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1

General Introduction

ABSTRACT — Soil pollution is a substantial problem in many countries. Total concentrations in soil are poor measures for uptake and toxicity, and therefore, to what degree the contaminants are bioavailable requires study. Earthworms are abundant in many soils and are appropriate model organisms because they live in close contact with the soil, have a thin, permeable cuticle, and consume large amounts of soil. Furthermore, these organisms contribute to soil fertility and are on the diet of many birds, mammals, and invertebrate predators. In this thesis, it is investigated how organic chemicals are taken up in earthworms, and to what extent. Furthermore, models are developed to predict accumulation in earthworms through various routes of exposure. This chapter introduces the problems and origins of soil pollution, and also the animal group that is the focus of this thesis: the earthworms. A brief exposé on earthworm ecology is provided, including the classification in different ecological groups and basic physiology. This knowledge is helpful in interpreting the results from bioaccumulation studies as presented in this thesis. Further, the leading theory on uptake of organic chemicals (equilibrium partitioning) is introduced, although a more thorough discussion is postponed to later chapters. This chapter concludes with an outline of this thesis and a definition of the most important terminology.

ORIGINS OF SOIL POLLUTION

Soil pollution is, strictly speaking, as old as the soil itself. However, before human interference, the pollution used to be restricted to specific areas, such as locations with superficial metal ores, sites of volcanic activity, and tar pits. Human activities have changed this pattern over the last few thousand years. Even though heavy metals are naturally occurring compounds, metal pollution has become more prominent, particularly since roman times, due to mining and smelting activities. Several organic soil pollutants, like polycyclic aromatic hydrocarbons (PAHs), are also naturally occurring chemicals, originating from incomplete combustion (*e.g.* as a result of forest fires). However, with the start of the twentieth century, we have witnessed a dramatic increase in soil levels, coinciding with an extensive increase in the use of fossil fuels [36]. Several of these PAHs are carcinogenic, which makes them particularly interesting from a human health perspective. In the beginning of the twentieth century, the industrial production of organic chemicals started for various applications. As an example, polychlorinated biphenyls (PCBs) were produced for electrical transformers and capacitors, but these compounds turned out to be not only toxic, but also extremely persistent in the environment. These compounds travel all the way around the globe, and considerable levels of PCBs and other organochlorine chemicals are, for example, detected in the fat of polar bears [60]. Currently, the emissions of PCBs are declining, but large numbers of new industrial chemicals are produced each year: between 300 and 350 new chemicals are notified in Europe annually¹. To name just a few categories: surfactants (*e.g.* for use in washing powders), photochemicals, intermediates (chemicals used in production of other chemicals), paints and dyes, polymers (and chemicals involved in polymerisation and polymer processing), and flame retardants. With the declining emissions of PCBs, the problems with persistent and toxic chemicals have not become a thing of the past; also among the currently used chemicals, there are suspect groups. For instance, perfluorinated surfactants have recently created a lot of interest. These compounds are used in lubricants, paints and fire-fighting foams, and are highly persistent in the environment, are widely distributed across the globe, and accumulate in the food chain [28,37,56].

These industrial chemicals can reach the soil through various routes: directly (dumping, leakage and spilling), atmospheric emission followed by wet and dry deposition, and via sewage sludge (see Fig. 1). In many countries, emissions to surface water are treated in sewage treatment works. Hydrophobic chemicals that are poorly degraded end up in the sewage sludge and in several countries, this sludge is applied as fertiliser on agricultural soil; a process that can lead to substantial contamination of agricultural soils [21]. In the Netherlands, sewage sludge from municipal plants is no longer applied in agriculture since 1995. However, from the industrial treatment plants, a part of the sludge is used in agriculture (13%), including a small fraction of sludge that is derived from chemical industries [11]. In contrast, nearly 50% of the sludge that is produced in German municipal plants is used in agriculture, usually after a treatment step [78]. This application of sewage sludge has led to contamination of the soil with heavy metals, PCBs, dioxins and furans. In recent years, the levels of some of these compounds are declining, owing to effective legislation, but a new class of chemicals, the polybrominated diphenyl ethers (PBDE), are being increasingly dispersed in the soil environment through this route [54]. Other chemical inputs into the soil result from the dredging of regional waters to ensure

¹ See <http://ecb.jrc.it/new-chemicals>

sufficient water depth for drainage and navigation. These sediments are placed on soil where they may contribute to the pollution, because sediments act like a sink for air-transported chemicals like PAHs [27]. The risks of dredge materials should however not be exaggerated, as placing sediments on land may also enhance biodegradation of these compounds [26].

The chemicals that are used industrially or in consumer products do not deliberately end up in the soil. Another important group of toxic pollutants are, however, directly and intentionally emitted: the chemicals used in agriculture to deal with unwanted weeds and pest organisms (herbicides, pesticides, fungicides, also euphemistically called “plant-protection products”). The well-known insecticide DDT was developed in 1939¹, and widely used after the second world war until the 1960's. It was not only used in agriculture, but also for vector control against malaria, and as contact poison against head louse. However, the long-lasting effectiveness that made this chemical so popular, also led to serious problems. DDT (and its metabolites) are highly persistent and accumulate in food chains, leading to severe effects on bird populations, especially birds of prey [34,81]. In response to these problems, DDT was banned in most western countries, although it is still used in third-world countries to combat malaria (and its production thus continues). However, 20 years after the last application, levels in orchard soils are still high enough to pose a threat to birds feeding on earthworms [33]. In the Netherlands, banning took place in 1973, but in a large number of soil samples taken between 1993 and 1995, the levels still exceeded the quality objective [32]. For agricultural locations, all of the top soil samples exceeded the quality objective, by up to a factor of 63. Similar conclusions were drawn for other notorious pesticides like dieldrin (banned in 1980). Clearly, we are still seeing the results of the agricultural applications of these chemicals in the soil, decades after their use has ceased.

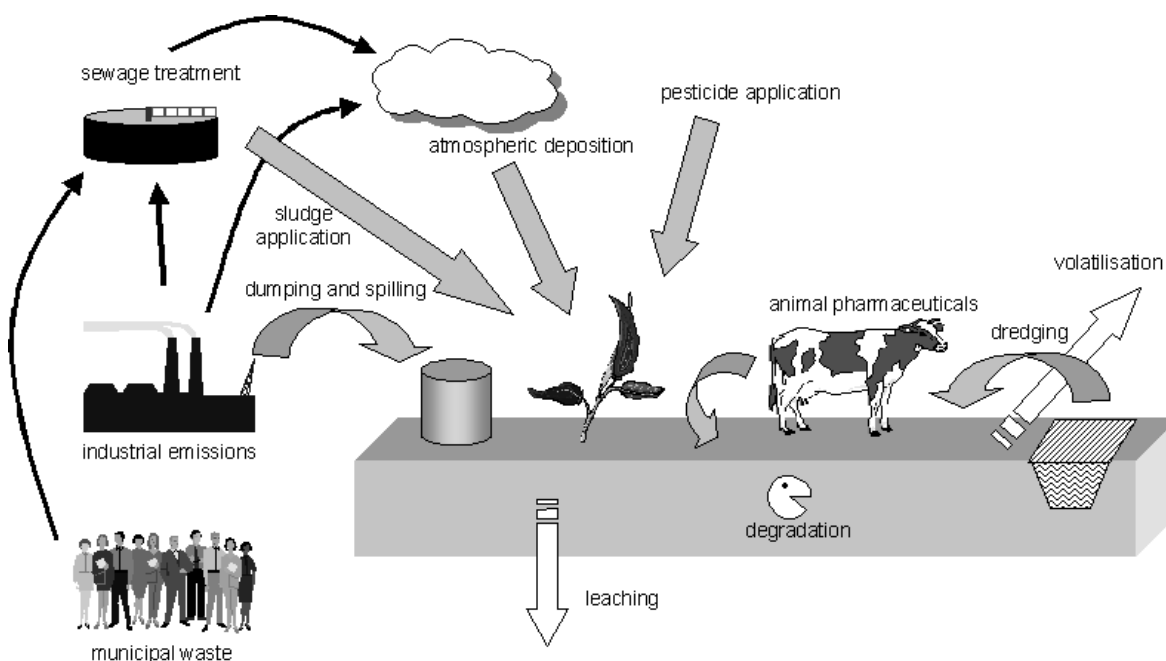


Figure 1. Major sources of soil pollution and removal processes.

¹ The Swiss chemist Paul Muller who invented DDT, received for this feat the Nobel price for medicine in 1948.