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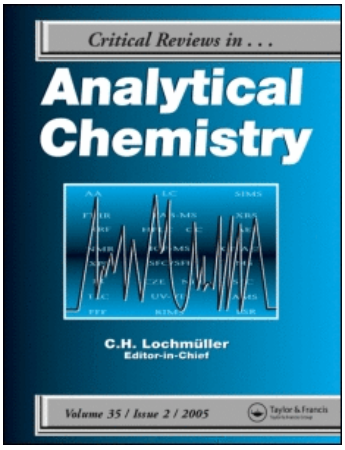

Critical Reviews in Analytical Chemistry
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713400837

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Online publication date: 03 June 2010

To cite this Article Holoubek, Ivan(1999) 'Persistent Bioaccumulative and Toxic Chemicals in Central and Eastern Europe: Levels and Risks', Critical Reviews in Analytical Chemistry, 29: 3, 179 — 185
To link to this Article: DOI: 10.1080/10408349891199374
URL: http://dx.doi.org/10.1080/10408349891199374

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Critical Reviews in Analytical Chemistry, 29(3):179–185 (1999)

Persistent Bioaccumulative and Toxic Chemicals in Central and Eastern Europe: Levels and Risks

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ABSTRACT: The article describes problems of organic substances that have persistent, bioaccumulative, and toxic characteristics likely to cause adverse human health or environmental effects. They are called PBTs (Persistent, Bioaccumulative, Toxic substances) and briefly the state of the PBTs problems in the countries of Central and Eastern Europe is described. As an example of research activities in the region, the long-term research program of Czech and other scientists so-called Project TOCOEN (Toxic Organic COmpounds in the ENvironment) is described.

KEY WORDS: persistent, bioaccumulative and toxic organic compounds, Central and Eastern European problems.

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I. INTRODUCTION

Organic substances that are persistent, bioaccumulative, and posses toxic characteristics likely to cause adverse human health or environmental effects are called PBTs (Persistent, Bioaccumulative, Toxic substances). In this context, "substance" means a single chemical species, or a number of chemical species that form a specific group by virtue of (a) having similar properties and being emitted together into the environment or (b) forming a mixture normally marketed as a single product. Depending on their mobility in the environment, PBTs could be of local, regional, or global concern.[1]

Subclass of PBTs so-called POPs (persistent organic pollutants) is group of compounds that are prone to long-range atmospheric transport and deposition.[2] The global extent of POP pollution became apparent with their detection in areas such as the Arctic, where they have never been used or produced, at levels posing risks to both wildlife[3] and humans.[4]

UN-ECE initiative that was started within the UN-ECE region (comprising Eastern and Western Europe, Canada, and the USA) in 1992 had prepared the Protocol on POPs.[2] The Protocol includes 16 POPs and the main objective of it is to control, reduce, or eliminate discharges, emissions and losses of POPs. Beside this UN-ECE initiative was started similar program of the United Nations Environment Programme in the cooperation with the International Forum for Chemical Safety.[5] This UNEP/IFCS program includes 12 POPs.

The expert groups of both international bodies call for new data needs for exposure and fate assessments.[6] Especially for data that are available for a particular region and they should obviously be used in the assessment process. These international experts strongly recommend to study the fate and distribution of the selected chemicals in different regions using compartmental mass balance models that must be verified by use of real measured data. The most serious data gap for the prediction of environmental behavior is degradation rates and their regional variability based on specific transport conditions. More data need to be collected in this area. The main topic of further research is the study of deposition/emission processes, transformation processes, and the bioavailability of POPs and PBTs in terrestrial ecosystems.[1]

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II. SOURCES OF PBT COMPOUNDS IN CENTRAL AND EASTERN EUROPEAN COUNTRIES

In the field of ambient air concentrations of PBT, compounds have little information on the state of air pollution. In the early 1990s, an emission inventory of the some POPs releasing into the atmosphere was carried out in the former Czechoslovakia[7] and the latter in the Czech Republic[8,9] and Slovakia.[10,11] It should be said that much data in this inventory in the first period (to 1995) was charged with considerably uncertainty because of the unreliability of emission factors or lack of input parameters, such as amounts of raw materials, fuel, waste, type of combustion, cleaning of combustion products, etc. Only minimum emission measurements have been realized within the Czech Republic and Slovakia and therefore published emission factors were applied if available. For these reasons, it was not possible to include some activities (emissions from fires, landfills, soil, water areas) into the inventory at all, although they may represent, mainly in the case of pesticides, PCBs, or PCDDs/Fs, decisive contribution to total emissions. At the present time, only the Czech Republic from the CEE countries realize very broad project concerning measurements of emission sources from typical sources of POPs.

Main sources of polycyclic aromatic hydrocarbons (PAHs) in the region are presented by electric and thermal energy production, waste incineration, road traffic, and some industrial technologies (e.g., high-temperature coal carbonation, catalytic cracking of crude oil, aluminium production). A limited amount of non-carcinogenic PAHs (naphthalene, anthracene, phenanthrene, pyrene, and carbazole) is produced industrially in the pure form (DEZA Valasske Mezirici, CR). They usually serve as starting material for the synthesis of dyes, pesticides, and pharmaceuticals.[7]

Main sources of PCDDs/Fs in the region are typical for industrial countries — they are formed as byproducts, mainly in chemical industry — production of chlorophenols and their derivatives (former Czechoslovakia, USSR, GDR), processes in which chlorinated catalysts and solvents are used; pulp and paper production — bleaching based on chlorine treatment, metallurgical production — if metal chlorides are used, magnesium production, metal scrap recycling; municipal, hospital, hazardous and industrial waste incineration; solid fossil fuel combustion (coal, wood, peat), internal-combustion engine operation — leaded petrol use with the addition of chlorinated compounds; dry distillation (dry cleaning of clothes); fires (forest, agriculture..).

Polychlorinated biphenyls (PCBs) are industrial products or byproducts formed at industrial processes.[7,10,11] In the former Czechoslovakia, PCBs were manufactured from 1959 to 1984 in a chemical plant in eastern Slovakia under the commercial name Delor. From the total 21 482 t produced, 46% was exported and the remainder was appointed for the home market of former Czechoslovakia. Owing to appropriate physical-chemical properties, PCBs were widely applied in industry, either in industry, either in closed systems (coolants and lubricants in transformers, dielectric fluids in capacitors, hydraulic fluids and heat-transfer media), or in open systems (plastificators, additives into carbonless copy paper, lubricants, inks, impregnating and paint agents, glue, wax, cement and plaster additives, lubrication of cast blocks, materials for dust separators, sealing liquids, flame retardants, immersion oils, and pesticides. Within both countries (Czech Republic and Slovakia), PCB formulations may be currently used only in the closed systems and they are gradually replaced. Currently, waste landfillings are considered to be the most relevant source of environmental pollution by PCBs in these countries. Estimated contribution of applied paint to total PCB pollution within Slovakia is about 5%, and that of industrial and municipal waste incinerators is 9%.[12]

Hexachlorobenzene (HCB) was used in the production of pyrotechnic compositions for military purposes, in synthetic rubber manufacture, in primary aluminium production, as a wood preservative and seed protectant on grain and field crops, as a chemical intermediate in dye manufacturing, and as a plasticizer for polyvinylchloride). It is generated as a byproduct in the manufacture of chlorinated solvents, some pesticides, and other chlorinated compounds that are used in metal

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smelting and electrolyzes. An important source of HCB is represented by high-temperature processes (incineration of waste, plastics, PCBs, combustion of fuel containing chlorine, metallurgical processes, including metal recycling, internal-combustion engine operation, fires).

Most of the countries in the region produce and/or formulate pesticides. The pesticide registration is a primary requirement for import, production and distribution. During the period of centralized economy in this region, the import was monopolized by the relevant state organization. By the end of the 1980s many private companies and minor distributors were involved in import and distribution of pesticides.[13,14]

Pesticide concentrations in the Danube river and its tributaries show significant differences between countries in the number and the types of pesticides analyzed. The cumulative number of analyzed pesticides was 76. Residues of only 36 pesticides and metabolites have been detected. The most frequently detected pesticides are organochlorine compounds and triazines. Only DDT and metabolites, HCH and isomers, and atrazine and metabolites were found in more than 50% of the samples.

Special attention must be given to unwanted pesticides. The problem of unwanted and expired pesticides pose the greatest danger to the natural environment and people that is brought about by chemization of agriculture in CEE countries. This problem results from many years of errors in pesticide management and especially in their distribution. Historical changes in these countries caused that together with the advent of market economy tile problem of storing expired pesticides ceased to exist. Countries still have not solve the problem of safety storage for pesticides and other chemicals classified as poisons and they have no information concerning the quantity of pesticide and chemical (for example, PCBs) waste. Many from CEE countries have no special sites dangerous materials or incinerators in which these types of chemicals could be safety burned.

Many of pesticides from UN-ECE list of POPs never been used in many countries from this region (aldrin, chlordane, mirex, heptachlor, toxaphen). These were banned, and restricted in several countries from a region similar in some other countries of the world.

Because of the position of human beings on the top of the food chain, high concentrations of these lipophilic and persistent compounds are often found in their tissues and excretions such as adipose tissue and milk.[15–17] It has been estimated that more than 90% of organochlorines body burden in the general population occurs via diet. Thus, the xenobiotics found are proportional to the dietary intake, that is, fish, animal fat, dairy products, cereals, and vegetables, etc.

With regard to females, two additional routes for reduction of the organochlorine body "pool" can take place: elimination through the placenta and excretion with the milk. These clearance mechanisms may pose a particular risk for developing foetus and breast-fed infants. Elevated levels of organochlorines compared to other European countries indicated high exposure of young women in the former Czechoslovakia.[15,18]

The first survey in the field of PBT compounds carried out in the former Czechoslovakia at the beginning of the 1960s was initiated by concerns over serious contamination of humans caused by widespread use of DDT. Besides measurement of DDTs in the human adipose tissue, later studies paid attention also to hexachlorobenzene (HCB), hexachlorocyclohexanes isomers (HCHs), and at the 1980s to polychlorinated biphenyls (PCBs). A successive decrease in the concentrations of DDT, HCHs, and HCB in human adipose tissue was observed on connection with the governmental regulations to their withdrawal from use.

However, considerable differences in residue levels determined in the human fat from different part of Czech Republic have been found frequently. Moreover, in some regions the generally recognized decrease is not so pronounced due to the serious contamination of the foodstuffs produced and distributed within particular area.

The trend results indicate that actual PCB concentrations remain in human adipose fat high even after several decades banning on their production.

III. PROJECT TOCOEN

The TOCOEN Project (**T**oxic **O**rganic **CO**mpounds in the **EN**vironment) is a long-term

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environmental research project involving many Czech and Slovak universities, research institutions, and various companies and at the present time collaboration with some universities and institutions from other countries.[15,16]

The TOCOEN Project had its inception in 1988. The basic goal of the project was a detailed understanding of the fate of selected organic pollutants in the environment. This includes input of these pollutants to various parts of environment (through emissions), their transport in compartments and between them, their transformations (photochemical, chemical, thermal, biochemical), their biological effects (dose exposure analysis), modeling of these processes, and risk assessment, management, and prognosis of contamination development.

Certain groups of POPs, such as polycyclic aromatic hydrocarbons (PAHs), chlorinated pesticides (Cl-PEST), polychlorinated biphenyls (PCBs), dibenzo-*p*-dioxins (PCDDs), and dibenzofurans (PCDFs), were selected as model compounds at the beginning of the project. Currently, other groups of organics such as chlorinated benzenes, phenols, certain other types of pesticides, chlorinated aliphatic hydrocarbons, and other chlorinated aromatic compounds are also being studied intensively. All the model compounds are determined in the important parts of environment: air, atmospheric deposition, surface waters, sediments, soils, aquatic, and terrestrial biota.

Biological indicators such as soil biota and plants have been used to monitor regional patterns of air pollution, as well as atmospheric deposition of pollutants transferred over long distances. Analyses of earthworms made it possible to constitute an effective monitoring system in the study of pollutant fluxes to terrestrial ecosystems. Earthworms have been used widely for studies on the bioaccumulation and chronic toxicity of PAHs, PCBs, chlorophenols, and 2,3,7,8-TCDD. Similarly, mosses, conifer needles, and lichens serve as valuable indicators of contamination in TOCOEN ecotoxicological studies.

The first TOCOEN period (1988 to 1993) was focused on basic and preliminary measurements and monitoring. During this period, sampling and analytical design was optimized, and monitoring provided the first information about POP contamination of important regions in the Czech and Slovak Republic. Monitoring activities were focused on five types of model sites:

- Background sites: (GEMS — Global Environmental Monitoring System — observatory Kosetice, south Bohemia).
- Heavily polluted areas — the combination of industrial and urban pollution.
- Landscape with typical agricultural production.
- River catchment areas.
- known sources of particular compounds, including:
  1. PAHs (DEZA Valašké Meziriči, a chemical factory producing aromatic and polyaromatic compounds, phthalates, and carbon black; Coal and gas fuel company, Vresova),
  2. PCBs, PCDDs/Fs (Municipal waste incinerators: Brno, Bratislava; industrial waste incinerator: LIAZ Mnichovo Hradište; Technoplast Chroryne; oil refinery: OSTRAMO Ostrava).

A very important part of the studies performed focused on the presence of POPs in vegetation, soil biota, and soil, enabling some conclusions on mechanisms of deposition to be drawn. Bio-indicators of changes in stressed soil environments represent one of the newly established research activities. During the last 3 years, the TOCOEN Project contributed to the following four problems areas:

1. Survey of soil contamination in the Czech Republic through monitoring type of projects[21–23]
2. Biomonitoring of stressed soils[24–26]
3. Study of diversity and activity of soil microbial communities in terrestrial ecosystems stressed by heterogenous mixtures of POPs[24–26]
4. The development of methods for ecological risk assessment in contaminated soils[28]

The accumulation of PBTs compounds in soil and sediments is a potential risk for the future, potential "Chemical Time Bombs".[28] For example,

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sediments act as a sink for many PBTs that enter aquatic ecosystems. Contaminated sediments can be a source of contaminants to aquatic organisms even long time after the contamination of the water body has stopped. In many regions, freshwater sediments were found to be a major continental reservoir of these harmful organic compounds. Project TOCOEN is focused to study of sediments from the beginning of research activities in 1988. The sampling network is formed from TOCOEN model sites (the surroundings of model sources of PBTs) and the most important tributaries of rivers Morava and Danube (the river Morava is the main river in Moravia and one of the most important tributaries of the Danube).[29,30]

The sediments were also collected from river Dřeunice, tributary of river Morava. This sampling area is located in East Moravia in the surroundings of town Zlín, sediments were collected in sampling period 1995 to 1998.

In this area of region Zlín, east part of the Czech Republic, a model case study as the newest part of Project TOCOEN is realized. This long-term research project BETWEEN (The Relationships BETWEEN Environmental Levels of Pollutants and Their Biological Effects) is focused to the identification of ecological risks based on study of real environmental mixtures of persistent environmental pollutants (PBTs) and the long-term effects on ecosystems. Project includes very wide range of chemical and ecotoxicological laboratory and field methods and compares their results.

It is difficult to provide direct conclusive proof of a causative relationship between environmental levels of specific PBTs and adverse impacts on a wildlife population. Linkages between contaminant exposure and effects may nevertheless be identified by evaluating all available study data and applying a multiple statistical analysis, PCA, etc.

These topics are studied from molecular and cell levels to ecosystem. The project has three levels of basic approaches:

1. Hazard identification vs. ecotoxicological properties of environmental compartments
2. Hazard identification and assessment in the field without previous knowledge about the stress factors involved
3. Risk assessment focused on sites (area) with known influence of stress factors

On the molecular and cell level, the effects of potential environmental pollutants on cell proliferation, differentiation, apoptosis, and risk/safety assessment of their role in tumor promotion are studied. Ecosystem levels include the study of the effects of anthropogenic and natural hazards on the population and communities in aquatic and terrestric ecosystems (study of biodiversity — aquatic toxicology, *in vitro* tests of toxicity, biochemical markers *in vivo* in fish liver, study of parasites).

This study of potential harmful effects that used very wide laboratory and field battery of tests is focused to environmental/ecological risk assessment of various types of environmental mixtures of pollutants. The project is realized in present time in the Czech Republic as a example of "from molecular and cell levels to ecosystem type study," and the results are used for development of methodology of environmental and ecological risk assessment.

IV. CONCLUSIONS

Further research of PBTs compounds will be focused at determining and correlating the key physical, chemical, and biological properties of chemicals, the research of new and unexpected organic compounds, the development of new test methods for study of long-term effects. Further studies are recommended on deposition/emission process, transformation process, and bioavailability of PBTs in terrestrial ecosystems; on the phytotoxic effects of PBTs and their effects on soil microbial populations and soil fauna; on transport processes, persistence, fate and effects of PBTs in aquatic media; on the effects of mixtures of chemicals similar to those actually encountered in the environment; on the study of mechanism-based biomarkers of effect; on effect monitoring for PBTs making use of existing accidentally high exposed populations in comparison with those exposed to the highest background levels.

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