
Contaminants: Budgets and Behaviour

Case Studies

1. PCBs in coastal zones (Knut Breivik & Kevin Barrett, NILU)
2. Mercury in the Mediterranean (Torunn Berg & Kevin Barrett, NILU)

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1. PCBs (polychlorinated biphenyls) in coastal zones with emphasis on the outcomes of the ELOISE and IMPACTS projects (Knut Breivik & Kevin Barrett)

1. Introduction and outline

Polychlorinated biphenyls (PCBs) are a group of halogenated organic substances that are frequently assigned to the larger group of persistent organic pollutants (POPs). PCBs and the other POPs are of great environmental concern because they are lipophilic (bioaccumulative), occur to a significant extent in the gas phase and condensed states (semi-volatile), toxic as well as persistent.

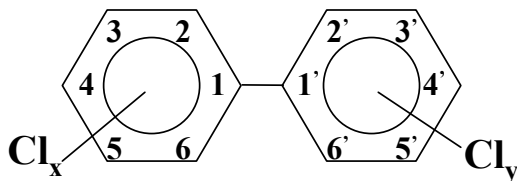


Figure 1. General chemical structure of the PCBs.

PCBs have been commercially produced for various applications since the 1930s. The known global mass production of PCBs virtually ceased in 1984, except for the production in the former Soviet Union, which ended in 1993. Worldwide, more than 1.3 million tonnes of PCBs have been intentionally produced (Breivik et al. 2002a). Dated sediment core studies have indicated that the temporal contamination pattern has mirrored the production and use of these contaminants (e.g. Christensen and Lo, 1986; Gevao et al. 1997). The environmental occurrence of these compounds in coastal zones and elsewhere are thus believed to be a direct result of the historical intentional production, consecutive use and disposal or accidental releases from products or materials containing PCBs (Breivik et al. 2002b). In addition, incidental or unwanted formation (*de novo* synthesis) is also reported to contribute to the environmental burden of PCBs (e.g. Brown et al. 1995). Even though global mass production has ended, PCBs are still frequently detected in environmental samples. This may in part be attributed to the long environmental half-lives of PCBs (Sinkkonen and Paasivirta, 2000) and in part caused by the potential long lifetime of products or materials containing PCBs (resulting in a significant time delay between production and cessation of primary emissions). The environmental concerns caused by PCBs in coastal zones are primarily a result of the affinity for PCBs to partition into the organic phases of suspended particles and bottom sediments as well as into the lipids of aquatic biota. This is problematic for several reasons. A primary issue of concern is that human exposure to PCBs tends to be significantly controlled by consumption of seafood, such as fatty fish. A second issue of long-term concern is the potential remobilisation of PCBs from contaminated sediments (Larsson, 1985), representing a potential threat for further uptake in biota for decades to come because of the long response time of sediments to loading reductions. In this document, a brief overview is

presented on key issues and research results related to PCBs in coastal zones. A particular emphasis is on results that have emerged from the ELOISE and IMPACTS projects.

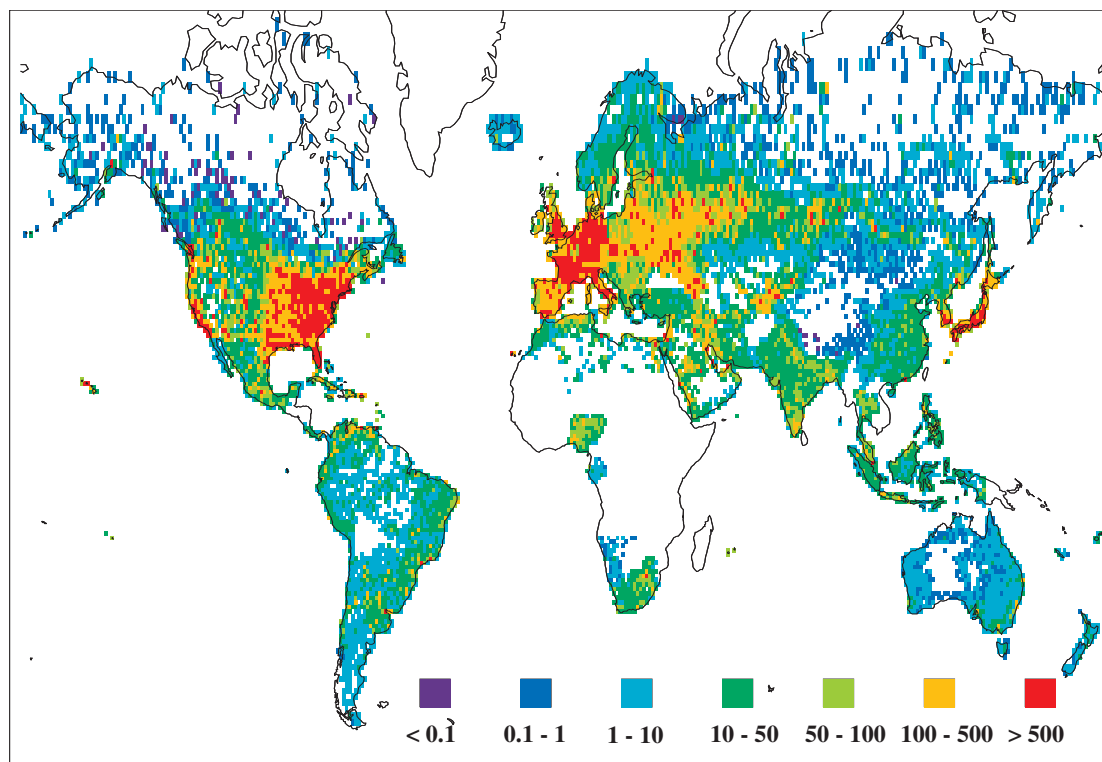


Figure 2. *Estimated cumulative usage of PCBs in tonnes* (Breivik et al. 2002a).

2. Sources and pathways of PCBs to the coastal environment

It is of considerable interest to understand contemporary sources and pathways of PCBs to the coastal environment. Although there is still limited quantitative understanding of the relative importance of the sources and pathways into the aquatic environment, it is generally considered that atmospheric inputs are of key significance on a regional scale, whilst direct discharges may be of key significance on the local scale (e.g. Maldonado and Bayona, 2002). Sources and pathways of PCBs to the aquatic environment have been key issues within the Popcycling-Baltic (Wania et al. 1998; Haugen et al. 1999; Pacyna et al. 1999), AIRWIN (García-Flor et al 2004), EROS-21 (Maldonado and Bayona, 2002) and ADIOS projects (Mandalakis et al. 2003; Mandalakis and Stephanou, 2004) with particular emphasis on atmospheric behaviour and fate. Haugen et al (1999) studied processes affecting the occurrence of PCBs in air at a coastal site in Norway from long-term monitoring data, whilst Mandalakis et al. (2003) conducted pioneering field studies on the degradation of PCBs in air at a remote site of the eastern Mediterranean. Additional field studies have focussed on the transfer of PCBs from the atmosphere to the aquatic environment (Mandalakis and Stephanou, 2004). Finally, theoretical approaches used to describe air-sea exchange of such compounds have been reviewed (Wania et al. 1998).

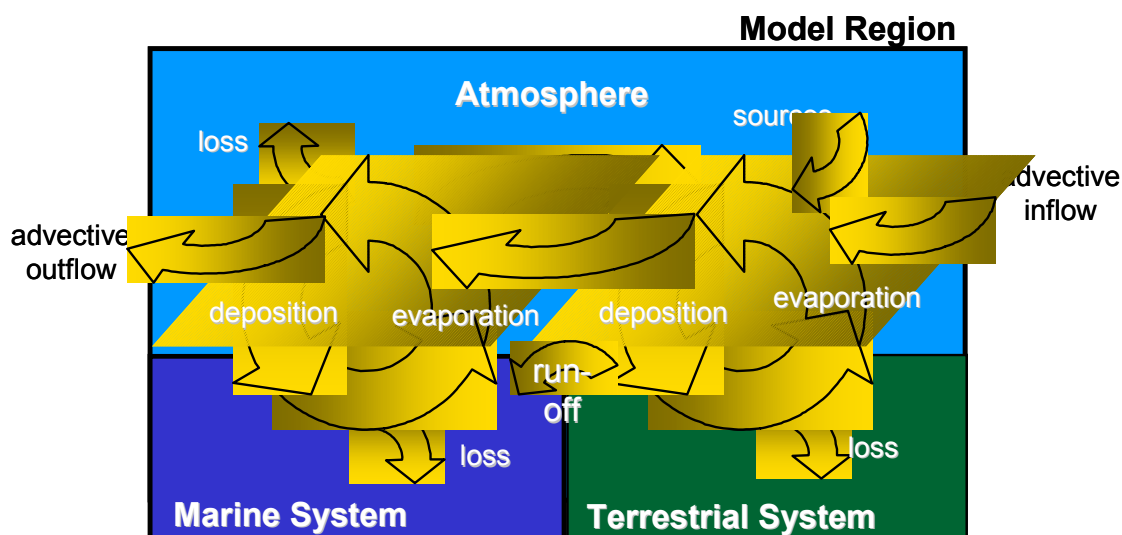


Figure 3. The POPCYCLING-Baltic project aimed to quantify the pathways of POPs including PCBs from the terrestrial environment to the marine environment via atmosphere and rivers (Wania et al. 2000).

3. Environmental cycling and fate of PCBs in the coastal environment

Various aspects regarding the environmental cycling and fate of PCBs in the coastal environment have been studied within several projects (ADIOS, EROS-21, Popcycling-Baltic). These studies include both process-oriented field studies (Maldonado and Bayona, 2002), monitoring approaches (García-Flor et al. 2004) as well as modelling studies (Sinkkonen and Paasivirta, 2000; Wania et al. 2000; 2001). Maldonado and Bayona (2002) studied the sources, fate and transformation of PCBs in the north-western Black Sea. They found that PCBs introduced to surface waters tend to be taken up by phytoplankton, with consecutive rapid sinking in sorbed state and degradation or solubilization along the water column (Maldonado and Bayona, 2002). Wania et al. (2001) developed a multi-compartmental model to describe the fate of PCBs in the Baltic Sea. This initial model was later to be further developed and improved as the major outcome of the Popcycling-Baltic project (The Popcycling-Baltic model: Wania et al. 2000). The POPCYCLING-Baltic model is believed to provide invaluable insight into the complex set of interactions that determine the overall fate of POPs in the Baltic Sea drainage basin, but which are inaccessible to measurements.

4. Closing remarks

It is recognized that the IMPACTS and ELOISE projects that addressed PCBs in coastal zones are very diverse in terms of key objectives. A simple overview of relevant projects (ADIOS, AIRWIN, BIOCET, EROS-21, FAMIZ, POPCYCLING-BALTIC) is therefore included in an Appendix to highlight the unique features of each project. Contact details and websites of selected projects are furthermore included for which more detailed information can be obtained.

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Appendix

ADIOS (IMPACTS)

The key incentive of the ADIOS project was to assess the impact of atmospheric deposition on the open Mediterranean Sea with respect to natural processes and anthropogenic activities. Organic pollutants were among the target compounds considered in the project. ADIOS did not only include studies related to the basin-wide characterisation of atmospheric deposition, but also the determination of anthropogenic compounds in the ecosystem of the open Mediterranean Sea. Methodologies used included environmental (air, water, sediments) and ecotoxicological monitoring, laboratory experiments, remote sensing and modelling.

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AIRWIN (IMPACTS)

The AIRWIN project aimed to characterize processes occurring in the sea surface microlayer (SML) and their relevance to global change and effects on the marine environment and its living resources. Key objectives included; (i) To define the role of the SML in the entry and loss of organic and inorganic pollutants in the marine environment, (ii) To identify those pollutants which are preferentially transferred to humans through marine food webs, (iii) To identify and to isolate micro-organisms that could be used for bioremediation and as cosmetic products to improve human's health, (iv) To provide potential bioindicators of atmospheric pollution. Examples of fieldwork carried out within the project with regards to organic contaminants demonstrated the enrichment of such substances (e.g. PCBs, PAHs, etc.) in the SML, as an organic carbon driven process, both in the dissolved and particulate phases. In this respect, the trophic status of the coastal waters will determine the partitioning of xenobiotics between the particulate, colloidal and truly dissolved phases, which has important implications in assessing the corresponding bioavailability. The fieldwork, in combination with the modelling task, has also evidenced that coastal regions are characterized by important net volatilization fluxes of POPs such as PAHs and PCBs. Conversely, net absorption fluxes are observed at open sea.

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BIOCET (IMPACTS)

The BIOCET project aimed to quantify and model the transport pathways and impact on reproduction of bioaccumulation of persistent organic pollutants (POPs) in four species small cetaceans (harbour porpoises and dolphins) in European Atlantic waters. The geographical scope of the project encompassed the Irish, Scottish (UK) and Dutch coasts, as well as the Atlantic coasts of France and Spain. The organic contaminants studied included the PCBs and DDE as representatives of the 'classical' POPs as well as selected brominated flame retardants (PBDEs).

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EROS-21 (ELOISE)

The key objective of EROS-21 was to develop an integrated framework of coupled biogeochemical models on the basis of field and laboratory studies, in order to provide a management tool for eutrophication reduction scenarios in the Black Sea. This project

additionally included research on the sources, distribution and water column processes of selected POPs (PAHs, PCBs and selected organochlorine pesticides) in the study area.

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FAMIZ (IMPACTS)

FAMIZ aimed to study the food web uptake of POP in the Barents Sea. The main incentive for carrying out the project relied on the observation that the ratio between the concentration of POPs in higher organisms and the concentration of POPs in seawater appeared to be higher in the Arctic than in other marine ecosystems. The objectives of FAMIZ were to understand this phenomenon by developing mechanistic models of the introduction of POPs into food webs in the highly productive marginal ice zone of Barents Sea.

The studies were directed towards two main hypothetical mechanisms: (1) Effects of microhabitats - locally elevated exposure to POPs, in particular to ice-algae microhabitats caused by ice rafted sediment, brine formation and/or melting deposited snow; and (2) Effects of food webs - more efficient POP accumulation due to slower growth, higher lipid content and longer life cycles of plankton and sympagic organisms. The project included laboratory and field studies as well as model development.

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POPCYCLING-BALTIC (ELOISE)

The overall goal of the project POPCYCLING-BALTIC was to develop a comprehensive, multicompartmental model to study the fate and behaviour of POPs in the Baltic Sea environment. Specific objectives of the project were to (1) understand and quantify the major sources, pathways and loss processes of the contaminants, (2) establish a link between contaminant loadings and environmental concentrations (3) estimate the time response to loading reductions (4) interpret spatial patterns and temporal trends of the contamination. The project also included the development of emission inventories for POPs (HCHs, PCBs, DDT, HCB, PAHs, PCDD/Fs) across Europe, as well as a database on environmental levels of selected POPs in various environmental compartments of the Baltic Sea drainage basin.

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Mercury in the Mediterranean

(Torunn Berg & Kevin Barrett, NILU)

1. Background

Mercury occurs naturally as elemental mercury and as organic and inorganic species. A large fraction of mercury is strongly bound to sediments and organic matter, and thus unavailable to organisms. However, microorganisms can transfer inorganic mercury to methylmercury which is fat soluble and are able to pass cell membranes, accumulates in animals, and biomagnifies in the food web.

Scientists have found particularly high concentrations of mercury in the in the Mediterranean tuna compared to similar species in most parts of the Atlantic, as well as in smaller species such as anchovy and sardines, and also in local marine birds and their eggs. An investigation of fishermen and their families in coastal villages on the north Tyrrhenian Sea found a correlation between the number of seafood meals and the mercury levels from hair samples. The process of biomagnification in the Mediterranean Basin is evident (<http://www.westonaprice.org/envtoxins/mercury.html>).

The biomagnification is probably due to the natural presence of mercury from volcanic activity. About half of world mercury resources are located in the Mediterranean area. Mining in the area has increased the release of mercury into the environment. Cinneabar, which has been known for 3000 years, represent the principal ore containing mercury. The world most abundant deposits are located in the Mediterranean region, and mercury from sites such as Almaden (Spain), Idrja (Slovenia) and Monte Amiata (Italy) has been exploited since ancient times for gold and silver extraction. A significant increase in mercury emission to the atmosphere occurred during the industrial revolution due to fossil fuel combustion and other human activities. Main antropogenic sources today are processing of mineral resources at high temperatures, such as combustion of fossil fuels, roasting and smelting of ores, kiln operations in cement industry, as well as incineration of wastes and production of certain chemicals. (Pacyna et al. 2001; Pirrone et al. 2003).

The following document present major findings of four ELOISE projects (MAMCS, MOE) and the MED-OCEANOR programme funded by the Italian National Research Council (CNR) to support part of field investigations of the MAMCS project. The projects provide(d) major results of these projects on different patterns involved in the dynamics of atmospheric pollutants in the Marine Boundary Layer (MBL) of e.g. the Mediterranean Sea, chemical and physical mechanisms in the lower atmosphere and sea water, atmospheric transport and deposition as well as gaseous exchanges at the air–water interface with changing meteorological conditions, and sampling and analytical techniques used to determine ambient levels of trace contaminants in the air and water samples.

2. Cycling of mercury in the Marine Boundary layer

A lot of work has been carried out in order to contribute to the assessment of the biogeochemical cycling of mercury in the Mediterranean region.

Mercury evasion derived from measurements of dissolved gaseous mercury (DGM) as well as speciated air measurements including reactive gaseous mercury (RGM) total particulate mercury (Hg-P) and gaseous elemental mercury (TGM) were performed at the Mediterranean Sea during three cruises. Using the DGM data and an empirical gas-exchange model (Wanninkhof, 1992) the data suggest that about 66 tonnes of mercury are released to the atmosphere from the Mediterranean Sea during the summer (Gårdfeldt et al., 2003). This emission is considerable in comparison to European antropogenic emissions and should

thus be taken into account in regional atmospheric modelling and assessment. Hence, it was found that Mediterranean Sea constitutes sources of airborne elemental mercury influencing the European domain.

The vertical profiles of DGM measured in the Mediterranean basin revealed that the concentration generally increases with depth (Ferrara *et al.* 2003). This finding indicates that DGM is produced not only in surface water as a consequence of photo-induced reactions, but also in deep water as a consequence of bacterial and/or geochemical activity. Volatile forms of mercury diffuse from the deep to the surface waters and then partially pass to the atmosphere. DGM concentrations and mercury evasion in surface water in the Western Mediterranean basin were lower than that observed in the Eastern basin. The maximum was found in the Strait of Sicily. The observed differences between the two basins may be explained by different biochemical scenarios. (Ferrara *et al.* 2003, Gårdfeldt *et al.* 2003).

Measurements performed during summer and early spring season at off shore as well as coastal sites consistently demonstrated that maximum solar radiation at midday coincided with maximum DGM concentration. During nights DGM concentrations were generally lower compared to the concentrations measured during daylight. Hence, a diurnal variation has been demonstrated in the off shore waters as well as the coastal sites investigated during the cruises (Andersson *et al.*, 2004).

Results from MAMCS and MOE on simultaneous measurements on mercury in Europe, showed that observed concentrations of total gaseous mercury (TGM), particulate mercury (TPM), and reactive gaseous mercury (RGM) were generally slightly higher in the Mediterranean region than in Northwest Europe (Wängberg *et al.* 2001).

The RGM formation in the Marine Boundary Layer (Sprovieri *et al.* 2002; Hedgecock *et al.* 2002; Hedgecock and Pirrone, 2001) is:

- driving the deposition processes of RGM to the sea
- affecting the cycling/exchange of Hg species between the atmosphere and the seawater
- the key to all chemical processes in gaseous, aqueous and heterogeneous phases that may drive the formation of oxidized mercury and transform elemental Hg to oxidized Hg

Also physical processes are very important with reference to the gas-particle partitioning of gaseous mercury with suspended particulate matter in the atmosphere. Obviously this process depends very much by meteorology (temperature, relative humidity, radiation) and particle composition. Enriched particles with EC are certainly the main factor that brings gaseous elemental mercury onto the particle and therefore is deposited faster to the surface.

Ongoing research is performed in order to evaluate the relative contributions of all these mechanisms in the overall mercury cycle and the relative contribution of natural vs. anthropogenic vs. recycling mechanisms. Indeed the emission of natural sources is a crucial step in the assessment of the global cycle in the Mediterranean sea basin. The Hg emission from the top water microlayer to the atmosphere and emissions from contaminated soil as well as from volcanoes and other geothermal activities are very important. See also Special issues in Atmospheric Environment published in 2001 and 2003.

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Appendix

BEEP: Biological Effects of Environmental Pollution in Marine Coastal Ecosystems

Goal: How biological markers may be used to determine the incidence of chemical contamination on marine organisms in monitoring programs

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Web-site: <http://beep.lptc.u-bordeaux.fr/DOCS/WP/WP0.htm>

MAMCS: Mediterranean Atmospheric mercury cycle system

The overall goal of the MAMCS project is to improve our understanding of the mechanisms which influence the dynamics of atmospheric mercury in the Mediterranean Sea region. MAMCS will also contribute to the improvement of policies and strategies for reduction of the risks associated with environmental pollution by mercury in EU member countries, particularly in Southern Europe. The Mediterranean Sea Region is characterized by a geological anomaly (it contains 65% of the world cinnabar (HgS) deposits). It also has higher seasonal temperature and levels of solar irradiation than areas previously modeled; factors which are believed to be behind many of the processes involved in the biogeochemical mercury cycle.

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MOE: Mercury Species over Europe

The general objective of MOE is to identify and quantify sources of atmospheric mercury (Hg) species focusing on production and fluxes of methylmercury (MeHg). The research addresses the relative importance of the atmospheric deposition of MeHg, in comparison to methylation/demethylation processes in terrestrial and aquatic ecosystems, in heavily impacted regions in Northern and Central Europe.

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MERCYMS: An Integrated Approach To Assess The Mercury Cycle Into The Mediterranean Basin

The overall goal of MERCYMS is to fill the gaps firstly specified and develop a Practical Working Tool (PWT) for a wide spectrum of users (i.e., policy makers, environmental planners) that would allow to assess for different environmental conditions and socio-economic scenarios, the relationship between the atmospheric input of mercury and its compounds to the Mediterranean Sea and the formation/production of the most toxic forms of mercury (i.e., MMHg, DMHg) in the marine system which have a significant impact on human health and the environment.

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