Levels and Trends of Dioxins, PCBs, and Other POPs in Abiotic Compartments

Richard J. Wenning and Linda B. Martello

Abstract Studies reporting on levels of polychlorinated dibenzo-p-dioxins (PCDDs), dibenzofurans (PCDFs), polychlorinated biphenyls (PCB), and other persistent organic pollutants (POPs) in the air, soil, sediment, and surface water are available from the 1970s to the present. While typically focused regionally, these studies provide important information on the evolution of the distribution and occurrence worldwide of environmental contamination attributed predominantly to human activity. This chapter summarizes monitoring work conducted during the past four decades that have contributed to our understanding of levels and trends in the abiotic environment. This includes a summary of available environmental data from the past two decades describing the current understanding of background conditions and global cycling of POPs. Data are summarized at a continental level for Africa, Asia/Pacific, Europe, North America and South America, and the polar regions. The results confirm the early views of the preeminent scientist in this field, Dr. Otto Hutzinger, who first suggested a "pulse" of highly persistent compounds entering the environment beginning in the 1930s and 1940s, peaking in the 1960s and 1970s, and gradually declining to the present time. This trend, however, does not yet apply to emerging POPs or to some regions of the world where long-range transport processes and monitoring work are evident only more recently. Overall, the distributions and levels of POPs continue to behave as predicted by Dr. Hutzinger; environmental levels of classical industrial chlorinated compounds are generally higher in the northern hemisphere than in the southern hemisphere, and levels are declining worldwide with the possible exception of PCBs and in remote locations where global cycling is finally extending its influence.

R.J. Wenning (✉)
Ramboll Environ, 136 Commercial Street, Suite 400, Portland, ME 04101, USA
e-mail: rjwenning@ramboll.com

L.B. Martello
Ramboll Environ, 2200 Powell Street, Emeryville, CA 94608, USA
e-mail: lmartello@ramboll.com

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Dr. Hutzinger's pioneering research over the past half-century laid the foundation for ongoing improvements in laboratory analysis and monitoring and interpretation of complex data sets, which continue to close the knowledge gap and improve our understanding of POPs in the air, soil, sediment, and water around the world.

Keywords Abiotic compartments, Air, Marine environment, Persistent organic pollutants, POPs, Sediment, Soil, Surface water, Terrestrial environment

Contents

1 Introduction

In 1984, Dr. Otto Hutzinger observed that the interaction of chemicals with biological organisms was as old as life itself, but that chemical pollution was human interference with natural chemical cycles and the release of man-made, unnatural compounds [1]. According to Dr. Hutzinger, the challenges posed by air pollution, water pollution, occupational exposure, pollution from agricultural practices, and contamination of food were best understood from a historical perspective, which could aid in the rational and prudent evaluation of present-day pollution problems.

Over 20 years from 1984 to 2004, Tickner et al. [2] concluded from a review of sustainable chemistry practices, which Dr. Hutzinger encouraged many years ahead of its time, that data collection on chemical risks and phaseouts of the most egregious chemicals alone would not promote the cultural and institutional changes needed to ensure the design and implementation of safer chemicals, processes, and products in the future. While society has benefited greatly from the commercial use of a growing class of persistent organic chemicals, some of those benefits have come at great cost to the quality of the environment.

For example, despite our efforts to restrict the commercial use of polychlorinated biphenyls (PCBs) and reduce incidental formation of polychlorinated dibenzo-p-dioxins (PCDDs or dioxins) and polychlorinated dibenzofurans (PCDFs or furans) in various combustion and chemical manufacturing activities, the contamination of soil and sediment and transient levels in ambient air and surface water continues to the present day. There is conflicting evidence from human and biota monitoring studies about whether current levels are indicative of a continued decrease from the generally higher levels observed in the 1970s and 1980s or reflect a plateau associated with either continued (albeit low) inputs or redistribution and recycling in the environment [3].

Our understanding of POPs in the abiotic environment emerged from environmental chemistry research and monitoring of polychlorinated biphenyls (PCBs), dioxins, and certain chlorinated pesticides widely used or accidentally released in the 1950s to 1970s. The 419 theoretically possible individual PCB, PCDD, and PCDF congeners have a range of physicochemical characteristics [4–6], which profoundly affect their persistence and environmental distribution, as well as bioaccumulation potential [7, 8]. PCDD/PCDFs were never produced intentionally for chemical manufacturing or marketable purposes. Dioxins were generated from many different combustion processes wherein a source of chlorine and ringed compounds was heated to a certain temperature range [9]. Dioxins also occurred as unwanted by-products in chemical manufacturing processes and various chlorinated chemical formulations. Among the best recognized sources of dioxins are chemical processes associated with the manufacture of chlorinated phenols such as 2,4,5-trichlorophenol and pentachlorophenol, pesticides such as Agent Orange, and antibacterial formulations such as hexachlorophene [9].

The leadership of environmental scientists such as Dr. Hutzinger and others over the past few decades has led to many important advances in our understanding of long-range transport and global cycling of man-made pollutants in the abiotic environment. Their leadership also has guided several improvements in sampling and laboratory analytical methods supporting environmental monitoring and advanced our understanding of the implications to human health and ecology. These and other scientific advancements have prepared us well for the new challenges posed by emerging persistent chemicals [10].

The purpose of this chapter is to build on monitoring work conducted from about the 1970s, when scientists first began to raise concerns about the persistence and toxicity of so-called “classical” industrial chlorinated chemicals appearing in the environment, namely, the PCDDs, PCDFs, PCBs, and a few chlorinated pesticides such as DDT and chlordane. Monitoring and laboratory work evolved significantly from the mid-1980s through leadership and technical advancements by Dr. Hutzinger and others. A closer inspection of current levels and trends and understanding of background conditions and global cycling of POPs are provided from reflection on these earlier decades and closer examination of available environmental data from the past 10 years. Data are summarized at regional and continental levels for Africa, Asia/Pacific, Europe, North America and South America, and the polar regions.

Collectively, the information from nearly 50 years of monitoring and analysis allows us to draw conclusions about levels and trends in air and the terrestrial (i.e., soils) and aquatic (i.e., sediments and surface water) environments around the world, as well as forecast likely levels in the future. This effort draws heavily from environmental surveys conducted by research organizations that have been actively involved in POPs monitoring and research activities in several countries, including the polar region. In many respects, this work is an update nearly 20 years later of the temporal trend analysis reported by Alcock and Jones [11], which provided a similar perspective in the mid-1990s on levels in different abiotic environmental compartments.

2 Fate and Transport Characteristics

The generalized environmental processes by which persistent man-made chemicals such as the dioxins, furans, PCBs, chlorinated pesticides, and other persistent contaminants move through the environment are reasonably well known [1, 12]. A global distribution model has been proposed to explain the accumulation of these substances in environmental compartments at higher latitudes [13]. Global distillation theory is based on physical–chemical POP properties, sources to the environment, and the earth's climatic conditions (Fig. 1). The physical and chemical properties that control the behavior of POPs are their low vapor pressures, low solubility in water, and preferences for binding to organic matrices. POPs are man-made organic compounds that, to a varying degree, resist photolytic, biological, and chemical degradation. POPs are also semi-volatile, enabling migration across long distances in the atmosphere before deposition.

In the atmosphere, POPs generally exist in both the gaseous phase and bound to particles, depending upon the environmental conditions [14]; two particularly important partitioning variables are air temperature and total suspended particle loading [15]. For dioxins and similar chlorinated compounds, there is a continual exchange between the particle and vapor phase and during the summer months, and when temperatures are high, the less chlorinated compounds tend to be found

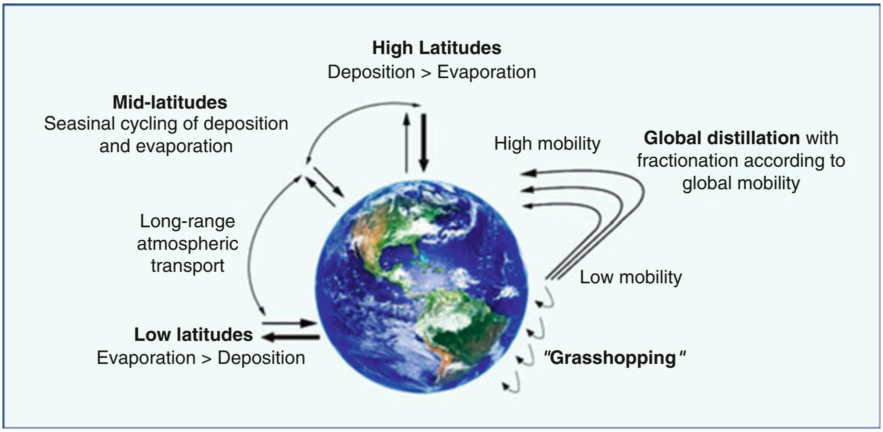

Fig. 1 The global distillation theory has been proposed to explain the distribution of POPs worldwide and particularly their occurrence in remote regions around the world where human activity is minimal (adapted from European Union Science Education Multimedia repository; http://www.eusem.com/main/CE/SIP_C2_bg)

predominantly in the vapor phase; in the winter months, dioxins and other compounds are split between the particulate and vapor phases [16]. In the polar region, snowpack can also influence atmospheric behavior [17, 18]. Similarly, seasonal differences have been noted in the exchange of POPs across the air–water/soil interface [19].

The future is somewhat uncertain, however; some suggest that indirect consequences of climate change such as shifts in agriculture and urbanization are more likely to influence future contaminant distributions than direct climate change [20]. Others suggest changing weather patterns and hydrological conditions will be more important than human activity, particularly for polar and remote regions [21, 22]. Modeling to predict fate and cycling in the North Sea also indicates that climate change may have a negligible influence on the fate and transport of PCB 152 and HCH [23] into the twenty-first century.

The main pathway by which POPs move from the atmosphere to the terrestrial environment is deposition to soil, vegetation, and surface waters by wet and dry processes. Vapor phase and particle-phase dry deposition are important for deposition to soil, though organic matter content is important to long-term retention in the soil matrix [24, 25]. Small amounts can be returned to the atmosphere by the resuspension of previously deposited material or re-volatilization in the case of lower halogenated substances [26]. It is well understood that POPs accumulate in most soil types [24], even clay soils devoid of significant organic content [27]. Half-lives for degradation of dioxins, PCBs, and most persistent organics in soil are generally measured in years [28], and both aerobic and anaerobic degradation of higher to lower chlorinated congeners remain uncertain [29, 30].

Air–vegetation exchange of POPs is an important process controlling the entry of POPs into terrestrial food chains and the air–vegetation transfer influenced by several physiochemical and environmental factors and plant characteristics [31]. Levels in vegetation tend to be surficial and typically reflect recent atmospheric deposition since most vegetation is exposed for relatively short periods of time, with new growth replacing old and crops being harvested. For agricultural leaf crops, the main source of contamination is direct deposition from the atmosphere and soil splash and for root crops soil contamination and binding to the lipids in cell walls [32]. However, the significance of root uptake of POPs from the soil requires further investigation, as there appear to be large differences between plant species [32, 33]. Grazing animals are exposed to POPs by ingesting contaminated pasture crops, whereby these substances are found to accumulate primarily in the fatty tissues and milk [34].

In the aquatic environment, the major inputs of POPs to surface water bodies are via atmospheric deposition and direct inputs from industrial effluent and soil runoff. It is well established that POPs partition rapidly to organic matter and accumulate in sediments, sometimes permanently but often times temporarily and thereby becoming a future source of contamination elsewhere or affecting aquatic organisms and human health through the diet [35, 36]. Dissolution in surface water is generally not a significant pathway, though concerns about rising temperatures could alter partitioning of POPs in sediments and biota [37]. POPs accumulate in aquatic biota as a result of the ingestion of contaminated organic matter and through the movement of substances through food chains. The concentrations of persistent substances in fish tissue is generally found to increase up the food chain as a result of the progressive ingestion of contaminated prey [38], although the processes by which this occurs are not well quantified, at present, in some deep water, tropical, or remote ecosystems [39, 40].

3 Background Levels

Considerable work has been conducted over several decades to understand background levels in the environment, predicated on the assumption that dioxins and other POPs have both a natural and anthropogenic origin. There is little debate that the majority of POPs are from anthropogenic origins in the northern hemisphere [41]. In practice, specifically in the regulatory community, the term background requires clarification if research and environmental regulation is to distinguish between the natural occurrence of some POPs and their distribution in the environment from diffuse man-made sources, both of which are characterized by large area extents, low concentrations, and the lack of point sources.

Increasingly in the context of monitoring POPs, terms such as background, background soils, and background content or concentration typically refer to widespread pollution caused by long-range transport or recycling of persistent substances alternately trapped and released from different soil and water compartments [20, 42]. An intuitive approach to allocate background locations is chosen by Rotard et al. [43]. A more recent study shows that the understanding of background condition is only possible if there is a sufficiently large data set to distinguish with statistical confidence differences between background diffuse pollution and local or regional contamination [44].

Several studies, however, suggest that several chlorinated and brominated compounds, including certain dioxins and PBDEs, can be formed by natural processes [45, 46], thereby supporting the theory of background levels in air, sediments, soil, and surface water in different parts of the world. Of particular scientific interest is the presence of dioxins found in sedimentary kaolin clay in North and South America, Europe, the South Pacific, and Asia [27, 47]. Schmitz et al. [48] confirm Horii et al. [47] observations that dioxin levels are higher in tertiary, primary nonsedimentary kaolin (ball) clays than in secondary, sedimentary kaolinitic and lignitic clays, suggesting an as yet unknown geologic enrichment process occurs in the environment. The available evidence remains somewhat inconclusive, and scientists involved in this work do not extend the theory to PCBs, chlorinated pesticides, the vast majority of halogenated compounds, and emerging POPs, which are generally assumed to be entirely of human origin.

The challenges associated with the determination of background environmental levels from monitoring conducted in rural or remote areas are well highlighted by work conducted on the dioxins. A summary of PCDD/PCDF levels in rural or

Table 1 Total PCDD/PCDFs and dioxin-like PCBs (dl-PCB) in surface soil from rural or remote regions

Country/region	Soil environment	Date	*N*	Σ PCDD/PCDFs (pg g^{-1} dw)	Σ dl-PCBs (pg g^{-1} dw)	PCDD/PCDF TEQ (pg TEQ/g)	PCDD/PCDF + dl-PCB TEQ (pg TEQ/g)	TEQ scheme	Reference
Arctic (Ny-Ålesund)	Tundra soil	2008	20	9.97 (3.55–16.6)	–	0.33 (0.16–0.62)	–	WHO 1998	[49]
Antarctic (Fildes Peninsula)	Tundra soil	2007–2008	15	2.18 (0.49–6.72)	–	0.02 (ND–0.06)	–	WHO 1998	[49]
Australia	Outback soil	2003	19	890 (0.31–15,000)	5.1 (ND–66)	0.38 (0.00056–5.0)	0.38 (0.00068–5.2)	WHO 1998	[50–52]
Austria	Forest soil	1993	25	319 (106–2676)	–	4 (1.6–31.0)	–		[53]
Central Europe Alps	Spruce forest, humus layer	2004	31	313.84 (115.7–758)	1,591 (439–3,266)	4.44 (1.37–10.81)	6.76 (116.4–15.73)	WHO 1998	[54]
China (Zhangmu-Nyalam, Tibetan Plateau)	Grassland and forest soil	2011	9	26.22 (2.43–73.28)	–	0.37 (0.06–0.65)	–	WHO 1998	[49]
Italy (NW Lombardy region)	Agrarian soil	2011–2012	10	–	–	2.13 (0.38–5.27)	2.87 (0.43–5.49)	WHO 2005	[55]
Norway	Woodland soil	1998	21	560 (15–4,100)	–	10 (0.2–78)	–	WHO 1998	[56]
The USA	Background, rural soil	1985–2011	Data from 14 studies	–	–	1.1–7.1 (0.1–22.9)	–	WHO 2006	[57]

Data presented as mean (range). The Σ PCDD/PCDFs data represent the mean and range for reported measurements of all dioxin and furan congeners remote soils typifies the wide range of so-called background conditions reported worldwide (Table 1). A detailed review of dioxins prepared by the US EPA [58] concluded that the background level in US soils was approximately 8 parts per trillion (ppt) of total dioxin equivalents (TEQ). USEPA's findings were based on 95 soil samples collected throughout the continental USA. More recent studies corroborate USEPA's findings, although data quality challenges continue to hamper clear understanding of natural occurrence and long-range transport from human sources and deposition in remote areas. Summarizing the available background soil data, Urban et al. (2014) concluded there was substantial variability in how soil dioxin data are presented in the literature (e.g., raw vs. summary data, congener vs. TEQ concentration, and number of congeners included in derivation of total level or TEQ). According to Urban et al. (2014), the reinterpretation of available data indicates that background levels in urban/suburban soils are higher and more variable than in rural soils: ranging 0.1 to 186 ng/kg TEQ and 0.1 to 22.9 ng/kg TEQ, respectively. Since much of the available data does not include dioxin-like PCBs, background TEQ levels in soil may be underestimated.

In Canada, 4 ng TEQ kg^{-1} (using WHO TEFs) is considered representative of the mean background concentration of PCDD/PCDFs in Canadian soils [59]. This value, however, may not accurately reflect the ambient background concentration of PCDD/PCDFs in soils elsewhere in Canada. For example, ambient PCDD/PCDF concentrations in soils collected from remote northern sites that were at least 20 km from human activity ranged from non-detectable to 0.000009 ng TEQ kg^{-1} (or 9 fg TEQ kg^{-1}) [59]. In general, scientists believe that natural background levels in extreme polar regions for all POPs are zero, and low (less than 1) ng/g soil levels are ambient levels reflecting either direct human activity or evidence of global cycling [60]. Meijer et al. [61] and Aichner et al. [62] describe how mechanisms such as vegetation and fate in different soil horizons likely influence soil levels of POPs.

Fewer and smaller data sets describing background soil conditions are available outside of North America [63]. According to Müller et al. [50–52], background concentrations of dioxin-like chemicals in Australian soils (0.54 to 3.8 pg TEQ g^{-1} dw) are, on average, among the lowest reported in any industrialized country; interestingly, soil from 1925 contained detectable concentrations of PCDDs, PCDFs, and PCBs at levels higher than in soil from the 1930s and 1940s. Buckland et al. [64] reported background dioxin concentrations of PCDD/PCDF expressed as I-TEQ ranging from about 0.17 to 1.99 pg g^{-1} dw in seven forest soils and five grassland soils collected in remote or pristine locations in New Zealand. The New Zealand results are somewhat similar to those reported in Australia, if results are expressed on a TEQ basis. In Germany, Rotard et al. [43] reported background concentration PCDD/PCDF in different cultivation types (forest, grassland, and plowland) ranged from about 10 to 110 pg I-TEQ g^{-1} dw. In Korea, Im et al. [65, 66] found 0.2 pg TEQ g^{-1} dw in mountaintop soil. In Brazil, Braga et al. [67] reported a concentration of 0.04 pg TEQ g^{-1} dw (PCDD/PCDF) in forest soil considered pristine. Work in the UK, Norway, and Europe indicates that background levels tend to decline from south to north in soils and levels are higher in forests than grasslands [68, 69].

In the air, data from Cleverly et al. [70] continue to reflect current understanding. Cleverly et al. [70] reported preliminary results of air monitoring at 17 rural stations and eight national parks in the USA conducted four times during calendar year 2000. Two of the 17 stations were located in suburban Washington DC and San Francisco, CA, to provide an indication of levels in more populated areas. All of the 2,3,7,8-substituted dioxin congeners were detected in ambient air at the 15 rural stations; dioxins were detected in 70% of the air samples. Excluding PCB 169, PCBs also were detected in all air samples. There was a 28-fold range in dioxin-TEQ annual average air concentrations at the rural sites, ranging between 2.5 and 58.3 fg m^3. PCB-TEQ air concentrations ranged from 0.2 to 9.9 fg m^3, an approximately 50-fold range. Passive air sampling in Europe corroborates much of the observational data in the USA regarding regional differences attributable to urban and remote locations [71].

Work conducted to date on background levels of dioxins in soil and air demonstrates how background may change from area to area within and between regions. Although global averages are of general use, no specific global background levels, for example, in soils, can be defined for environmental management or health assessment purposes; at best, regional or local operational estimates can be made, though with caveats [72].

4 Levels in the Air

Atmospheric transport has long been recognized as a major mechanism for dispersion of dioxins, PCBs, and other POPs around the world and, as such, has been a particularly important focus of investigation for decades [24, 25]. Early work on atmospheric transport and the influence of highly populated regions on levels in the air initially focused on the US Great Lake region [73, 74, 171], UK [75, 76], and Europe [61, 77] (Fig. 2).

At present, the Global Atmospheric Passive Sampling (GAPS) Network is the only global-scale air monitoring and surveillance program for legacy POPs and new priority chemicals. The program involves periodic deployment of polyurethane foam (PUF) disk passive air samplers to approximately 60 locations around the world and testing for dioxins, organochlorine pesticides, PCBs, PBDEs, and emerging candidate POPs [79, 80]. Led by researchers from Environment Canada, the monitoring is intended to provide spatial and temporal (including seasonal) air concentration data useful to developing emission estimates, validating predictions from fate and transport models, and risk management decision-making [168].

GAPS results from PUF samplers deployed in 2002 for 2–7 months [79] and in 2005 for four consecutive 3-month periods [81] reveal similar results. Pozo et al. [81] reported that annual geometric mean concentrations in the air were

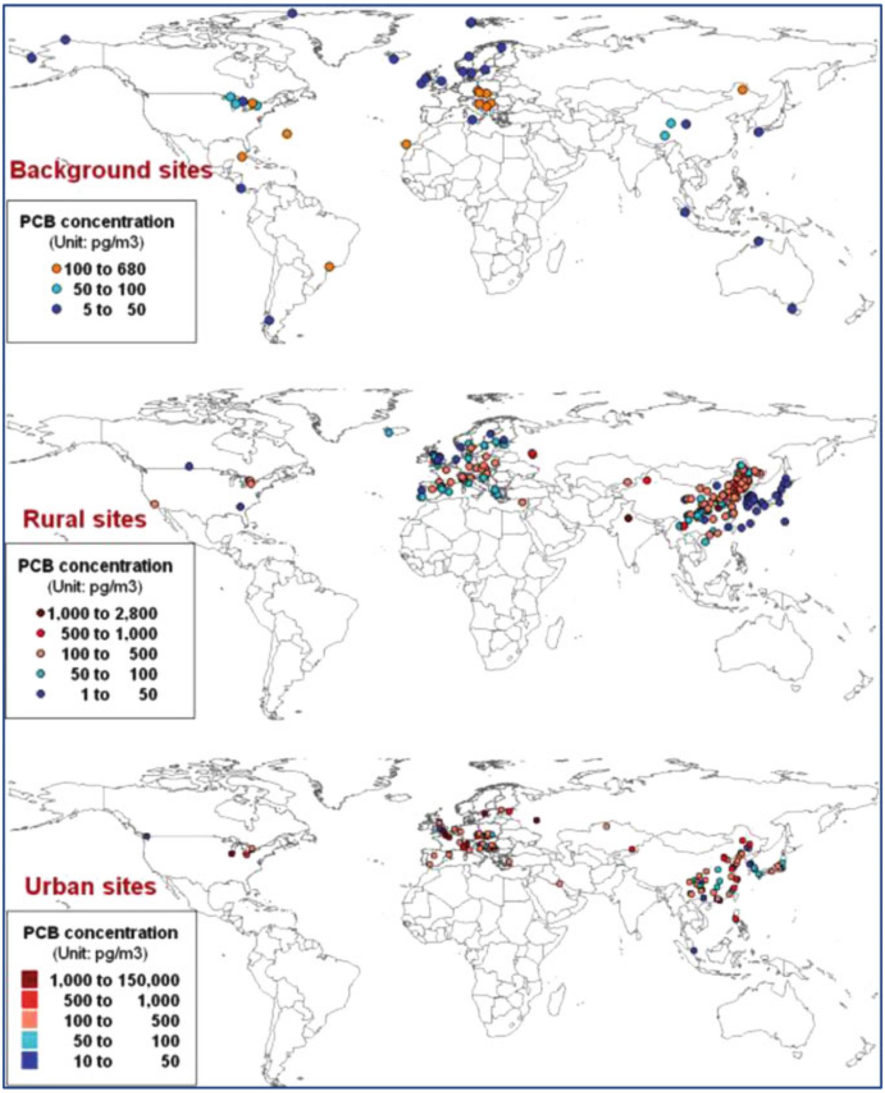

Fig. 2 Concentrations (pg/m^3) of ∑PCBs in ambient air reported at remote locations (*top*), rural sites (*middle*), and urban locations (*bottom*) around the world (adapted from [78])

highest for endosulfan (geometric mean, 82 pg/m^3) and PCBs (geometric mean, 26 pg/m^3); other chemicals regularly detected included α- and γ-hexachlorocyclohexane (HCH), chlordanes, heptachlor, heptachlor epoxide, dieldrin, *p*,*p*′-DDE, and PBDEs. Monitoring results from 2002 to 2009 did not reveal seasonal patterns on a global basis, although trends and seasonal patterns were evident for some substances. Endosulfans, for example, exhibit strong seasonality with highest concentrations during summer months at or near agricultural areas;

highest concentrations of POPs generally occur in the midlatitudes of the northern hemisphere; and PCB global emission estimates correlate well with the highest concentrations in developed and industrialized regions.

At the global scale, long-term monitoring data suggesting POP levels that may be declining overall [82] is optimistic and remains largely inconclusive for most scientists. Concerns about climate change and evidence of increases in the levels of several traditional and emerging POPs in marine food webs have prompted monitoring work in the Arctic in the past decade [83–85]. Air monitoring in the Arctic [86–89] and results from the third phase (2003 to 2011 monitoring) of Canada's Northern Contaminants Program (NCP) provide some evidence of declining trends for some but not for all POPs [90]. Bidleman et al. [83] found annual differences in the partially degraded fractions (enantiomer fraction) of HCH and chlordanes measured from 1994 to 2000 at the Alert, Canada, monitoring station, suggesting different emission sources contributing to atmospheric concentrations in the warm versus cold seasons.

POP contaminant trends are not apparent, however, in several developing countries where economic pressures may compromise some environmental regulations [91]. For much of the world, information is incomplete at the regional scale, and work is just beginning to assess spatial and temporal changes in the air [92, 93]. Work underway to train laboratories in passive air sampling for POPs in West, East, and South Africa, Latin America and the Caribbean, and Pacific Islands [94] will greatly help to fill this important knowledge gap. Initial ambient air results from this effort and reported by Bogdal et al. [95] suggest ∑PCBs are approximately four times higher in Africa (median 84 pg/m^3) than in the Pacific Islands or Latin America; ∑DDT is nearly six times higher in the Pacific Islands (306 pg/m^3) than in Africa and barely detectable in Latin America; and ∑PCDD/PCDF TEQs are slightly higher in Latin America (74 fg WHO98 TEQ/m^3) than in Africa and nearly 30 times higher than in the Pacific Islands (Fig. 3). E-waste and ship dismantling in Africa, malarial control programs in the Pacific Islands, and intensive urbanization in Latin America may be important factors contributing to the highest levels of PCBs, DDTs, and PCDD/PCDFs, respectively, observed in these regions.

In China, which arguably has the world's most notable air pollution, few surveys of the concentrations of POPs in the atmosphere on a national scale have been carried out so far, and, therefore, chemistry data concerning the overall contamination status of atmospheric POPs in China are absent. Zhao et al. [96] have proposed the use of tree bark as a passive sampling medium to understand regions acting as possible sources and sinks and global cycling because tree bark accumulates both gas-phase and particle-phase POPs simultaneously from the surrounding air [97] and reflects time-integrated overall air pollution levels [98]. By examining the spatial distribution of 18 polycyclic aromatic hydrocarbons (Σ18PAHs), five organic chlorinated pesticides (Σ5OCPs), ten polychlorinated biphenyls (Σ10PCBs), and 17 brominated flame retardants (Σ17BFRs) in 163 bark samples from 68 locations across mainland China, Zhao et al. [96] could demonstrate that

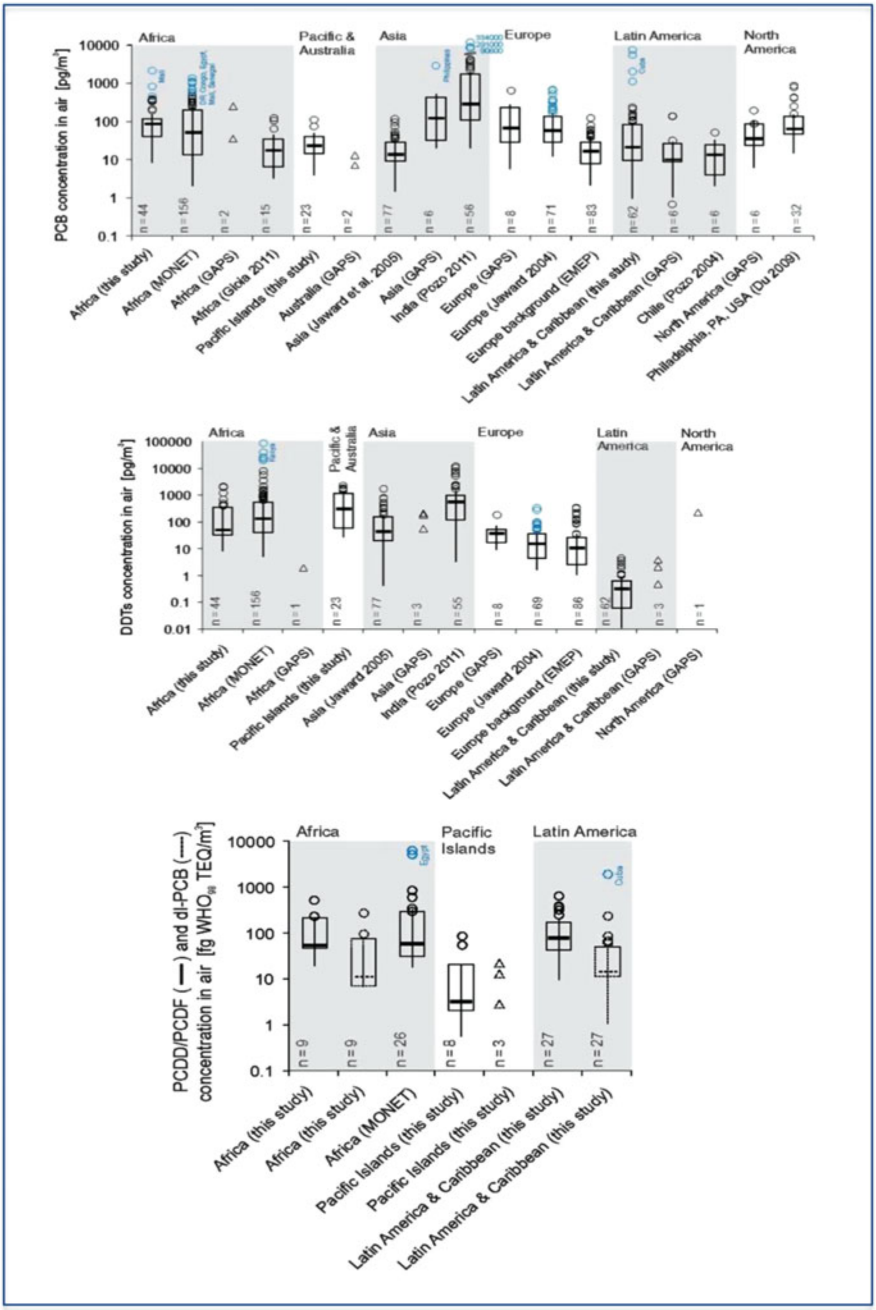

Fig. 3 Results of passive air sampling for $\sum$PCBs (pg/m^3), $\sum$DDTs (including *o,p′* and *p,p′* congeners of DDT, DDD, and DDE; pg/m^3), and $\sum$PCDD/Fs (fg WHOTEQ/m^3) in Africa, Pacific Islands, and in Latin America and the Caribbean (adapted from [95])

environmental contamination by atmospheric POPs was more serious in eastern and middle China than in western China.

Kalantzi et al. [99] argued that passive air sampling and the use of surrogate measures of air concentrations are sensitive to local, regional, and global-scale spatial and temporal atmospheric trends for many POPs and proposed butter and possibly other dairy products as useful monitoring surrogates. For most surrogate materials, confounding factors limit the usefulness of the data for monitoring or to predict long-term trends. Using dairy products as tool for air monitoring, for example, it has been shown that climatic factors and livestock management practices likely influence air–milk fat transfer processes [100–102]. Other surrogates such as vegetation also have limitations. For example, the collection and analysis of vegetation samples, such as conifer needles, lichens, and tree bark [103–105], may be hampered by specie differences, changes to predominant weather patterns, and other confounding factors that make data interpretation difficult [106, 107]. These and other factors confounding the use of either biotic or abiotic monitoring surrogates highlight the importance of assessing the spatial and temporal variability in long-term trend analysis [102, 108].

5 Levels in Terrestrial Environments

Generally, soil studies conducted during the 1960s to 1990s focused primarily on inputs from hazardous waste sites, industrial activities, and municipal sources. Scientific attention largely shifted beginning in the 2000s to understanding environmental fate and global distribution with increasing recognition that levels of dioxins, PCBs, and other POPs were different in urban, rural, and remote locations [11], as well as the advent of national POP inventories and concerns with both chemical persistence and endocrine disruption [109].

Several notable studies conducted during the past two decades provide a good understanding of spatial and temporal changes in soil levels of POPs worldwide. In general, ΣPCDD/PCDF TEQ, ΣPCB, and ΣPBDE levels in soils in North America, China, and Europe are 10–100 times higher than soil levels in Australia/New Zealand, Africa, and Latin America [12]. Further, it is widely acknowledged that the dioxins, PCBs, and several other legacy POPs are highest in heavily populated regions of the world [12]; for example, the distribution of PCBs reported by Li et al. [78] and shown in Fig. 4 is a typical global profile. Soil levels, however, are highly variable within countries and regions and influenced largely by the intensity of human activity (Fig. 3). Holoubek et al. [110], for example, examined 18 years of time trend POPs data collected in the southern Czech Republic since 1988 and reported significant variability in occurrence and distribution of selected groups of persistent pollutants in soil and sediment.

For comparison, the lowest ∑PCDD/PCDF concentrations reported worldwide in soil are from Jia et al. (2014) on the Fildes Peninsula, Antarctica, (0.015 pg I-TEQ/g), Arctic soil (0.33 pg I-TEQ/g), and Tibetan Plateau (0.37 pg I-TEQ/g).

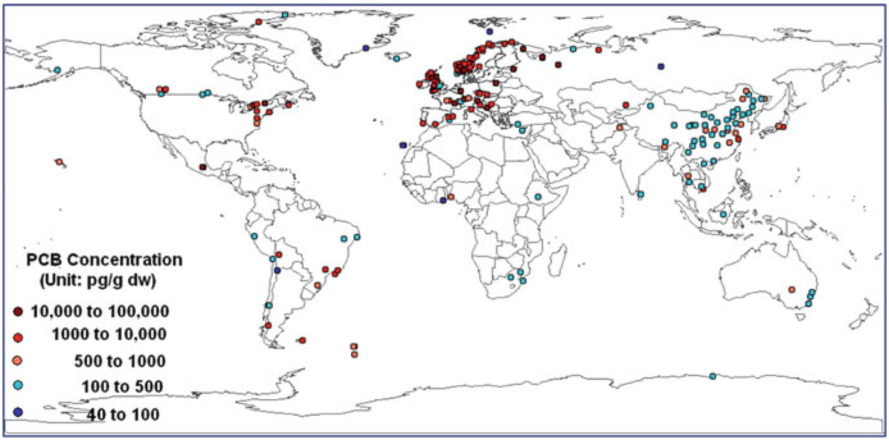

Fig. 4 Concentrations (pg/g dw) of ∑PCBs in soil around the world (adapted from [78])

These results are consistent with 1-year model simulations of atmospheric dioxin deposition reported by Booth et al. [111] and comparable to results of ambient air sampling conducted by Piazza et al. [112]. Studies conducted in high alpine regions such as the Italian Alps, Himalayas, Rocky Mountains, and Peruvian Andes monitoring several environmental compartments (air, soil, sediment, surface water, and foliage) simultaneously to understand the driving forces determining the distribution patters of POPs generally report similar observations [113].

Data describing POP levels in Africa are limited, but available data showing increasing levels of PCBs and other persistent contaminants can be related to growing industrialization and poor waste disposal practices [114]. The use of pesticides in Africa is likely a contributing factor as well [115, 116]. Studies on chlorinated pesticide occurrence in Botswana, for example, suggest that the residence time of OCPs in arid subtropics is very short. In the Okavango Delta [117], the low uptake capacity of the environment is likely due to the high temperatures and the low organic matter content of the soils. The few reported studies focus on a limited number of sampling sites for short periods of time; therefore, it is difficult to obtain information on the spatial variability of OCPs, identify their temporal trends, or gain an understanding of the fate of OCPs in low-latitude environments.

6 Levels in Aquatic Environments

While the relative importance of atmospheric and hydrospheric transport in controlling the global distribution of specific POPs remains uncertain, it is generally recognized that these two mechanisms are the predominant explanations for long-range environmental transport and global cycling of PCBs and most POPs. For ionic perfluorinated contaminants (PFCs) such as carboxylated and sulfonated perfluoroalkyl acids, for example, negligible vapor pressure, water solubility, and moderate sorption to solids predict ionic PFC accumulation in surface waters; therefore, the environmental distribution of PFCs is likely governed by hydrodynamics [118]. For the dioxins, however, traditional physical processes involving transport of sediments and windblown particulate are widely accepted as the primary dispersive mechanisms.

The summary of PCDD/PCDF and PCB levels in sediments shown in Table 2 typifies the features of global distribution reported worldwide. Perhaps the largest influence of POPs on human health is their widespread occurrence in different marine and freshwater environments throughout the world [142]. Levels in freshwater environments tend to be higher than marine environments owing largely to oceans and estuaries behaving as sinks for coastal and inland sources. In addition to the legacy POPs, Loos et al. [143] found several emerging chemicals as among the chemicals of most concern in 100 European rivers from 27 European countries, where water samples tested positively for 35 pharmaceuticals, pesticides, PFOS, PFOA, benzotriazoles, synthetic and natural hormones, and endocrine disrupting chemicals.

China is believed to have some of the highest contamination levels in the aquatic environment because of rapid industrialization over the past 30–50 years. Surface water, sediment pore water, and sediments in the Pearl River Delta, Minjiang River estuary, Zhujiang River, Yellow River, and elsewhere both inland and in coastal areas have been shown to contain high levels of OCPs, PCBs, PBDEs, dioxins, and other POPs [144–147]. The spatial distribution characteristic of contamination in the whole country is generally believe to be southeast > central > northwest [148].

It is generally recognized that contamination is less evident in the Southern Hemisphere, though the situation may change in South America and Africa with further industrialization. Temporal trends for both PBDEs and HBCD in Asia are unclear currently, and e-waste recycling has raised concerns that this activity will lead to increased levels, particularly in Asia and Africa [149]. Klánová et al. (2008) reported soil concentrations ranged between 0.51 and 1.82 ng g^{-1} for seven indicator PCB congeners, between 0.49 and 1.34 ng g^{-1} for HCH congeners, between 0.51 and 3.68 ng g^{-1} for Σ DDT (i.e., *p,p*-DDT, DDE, and DDD), and between 34.9 and 171 ng g^{-1} for Σ16PAHs. Sediment levels from 0.32 to 0.83 ng g^{-1} were found for ΣPCBs, from 0.14 to 0.76 ng g^{-1} for HCHs, from 0.19 to 1.15 ng g^{-1} for ΣDDT, and from 1.4 to 205 ng g^{-1} for ΣPAHs. A prevalence of low-mass PAHs, less chlorinated PCBs, and more volatile chemicals indicates that the long-range atmospheric transport from populated areas of Africa, South America, and Australia is the most probable contamination source for the solid matrices in James Ross Island.

Recent studies in Chile and Colombia, for example, report levels of BFRs (up to 2.43 and 143 ng g^{-1} dw of PBDEs in Chile and Colombia, respectively) and UV-F (non-detect to 2.96 and 54.4 ng g^{-1} dw in Chile and Colombia, respectively) in the low range of published data from other regions of the world [167]. Miglioranza et al. [150], providing the first systemic data on levels of OCPs, PCBs, and PBDEs in soils, sediments, SPM, and surface water along the Rio Negro basin

Table 2 PCDD/PCDFs and dioxin-like PCBs (dl-PCBs) in surface sediment reported from around the world

Country/region	Sediment environment	Date	*N*	Σ PCDD/PCDFs (pg g^{-1} dw)	Σ dl-PCBs (pg g^{-1} dw)	Σ PCBs (pg g^{-1} dw)	PCDD/ PCDF TEQ (pg TEQ/g)	PCDD/PCDF + dl-PCB TEQ (pg TEQ/g)	TEQ scheme	Reference
Africa										
South Africa (central region)	Freshwater	2006	7	42.8 (1.4–183)	0.20 (120–1,800)	–	0.26 (0.08–0.79)	0.46 (0.12–1.4)	WHO 2005	[119]
Morocco (Nador lagoon; Moulay Bousselham lagoon)	Marine	2006-2007	4	3,682 (960–3,683)	–	–	0.01 (0.004–0.04)	–	WHO 2005	[120]
Morocco (Ports of Tangier, Larache, and Kenitra)	Marine	2003-2007	4	75 (12.7–164)	–	–	0.86 (0.04–2.7)	–	WHO 2005	[120]
Kuwait (rural area)	Marine	NR	2	2.1* (0.4–3.8)	–	–	0.2	–	WHO 2005	[121]
Kuwait (industrial area)	Marine	NR	10	130.25* (12.5–131.5)	–	–	1.72 (0.2–4)	–	WHO 2005	[121]
Asia/Pacific										
Australia	Freshwater	2003	33	490* (ND–3,500)	160 (ND–1,300)	–	0.32 (0.0005–2.2)	0.41 (0.002–2.9)	WHO 1998	[50–52]
Australia	Marine	2003	12	460* (33–2500)	79 (0.018–440)	–	0.45 (ND–3.2)	0.53 (0.0000018–3.9)	WHO 1998	[50–52]
Australia	Estuarine	2003	30	14,000* (7.6–110,000)	3,200 (ND–28,000)	–	30 (0.0038–510)	32 (0.0038–520)	WHO 1998	[50–52]
China (Yellow River)	Freshwater	2010	13	2.1–19.8*	1.11–9.9	2.5 1.12–9.9	0.8–0.55	0.08–0.57	WHO 2005	[122]
China (Yangtze River)	Freshwater	2010	13	21.8* 6.1–84.9	1.8–24.1	1.79–27.07	0.09–0.47	0.13–0.29	WHO 2005	[122]

South Korea (Yellow Sea)	Marine	2008	12	35.3* (ND–142.1)	–	–	–	–	WHO 2005	[123]
China (Yellow Sea)	Marine	2008	28	10.35* (ND–62.4)	–	–	–	–	WHO 2005	[123]
China (Haihe River estuary and Dagu River)	Estuarine	2003	13	48,000* (151–557)	–	23,006 (775–1046)	20–975	21–996	WHO 2005	[124]
China (Kaifaqu and Yongdingxin River estuaries)	Estuarine	2003	2	291* (210–372)	–	1,045.5 (922–1,169)	1.7–2.7	1.8–2.8	WHO 2005	[124]
China (Changjiang River estuary)	Estuarine	2003–2004	10	170* (26–374)	–	–	0.8 (0.4–1.4)	–	WHO 2005	[125]
Hong Kong (Pearl River estuary)	Estuarine	NR	16	6,040* (4,439–9,404)	–	–	12.5 (10.8–16.4)	–	WHO 2005	Müller et al. (2002)
South Korea	Marine	1992	11	102–6493	–	–	1–76	–	WHO 1998	[65, 66]
South Korea (industrial bays)	Marine	2000-2002	122	216–755	–	–	1.2–7.2	1.3–10.8	WHO 2005	[126]
Japan, Toyano Lagoon	Estuarine	2000-2003	5	18,762 (370–54,000)	–	–	0.5–76.0	–	WHO 2005	[127]
Japan, Tokyo Bay	Marine	2000-2003	9	–	–	–	3.1–49	3.3–52.0	WHO 2005	[127]
Vietnam, coastal lagoons	Estuarine	2002, 2006	11	871* (197–2919)	–	–	1.85 (0.25–5.2)	–	WHO 2005	[128]
Vietnam, Saigon River estuary	Estuarine	2004-2005	12	650 (250–1800)	1,665 (18–8.400)	287–294	–	4.1 (0.73–17)	WHO 2005	[129]

(continued)

Table 2 (continued)

Country/region	Sediment environment	Date	*N*	Σ PCDD/PCDFs (pg g^{-1} dw)	Σ dl-PCBs (pg g^{-1} dw)	Σ PCBs (pg g^{-1} dw)	PCDD/ PCDF TEQ (pg TEQ/g)	PCDD/PCDF + dl-PCB TEQ (pg TEQ/g)	TEQ scheme	Reference
Europe										
Finland (Gulf of Finland)	Estuarine	1998-2003	44	15,300 (1,030–52,900)	–	–	83 (13–216)	–	WHO 2005	[130]
Italy (Venice Lagoon)	Estuarine	1996-1998	22	12.3* (1.9–34)	–	–	1.11 (0.28–2.15)	–	WHO 1998	[131]
Mediterranean Sea	Marine	2003-2004	6	413* (102–680)	–	–	3.7 (1.3–5.6)	–	WHO 1998	[132]
Portugal (Lima, Ria de Aveiro, Mondego, Tejo, Sado, Mira, and Ria Formosa estuaries)	Estuarine	2011	31	4.6–634.6*	–	16.0–11,278.8	0.1–5.3	0.1–11.6	WHO 2005	[133]
Spain (Catalonia coast)	Marine	2000	18 (PCDD/ PCDF and dl-PCB) (45 (PCB)	1,790* (60–8,140)	7,750 (270–35,600)	53,500 (1,100–311,000)	8.88 (0.4–39.24)	12. 92 (0.43–42.76)	WHO 1998	[134]
Spain (Cantabria coast)	Marine	2006	6	1.7* (0.15–3.99)	396 (139–691)	2,493 (558–4,656)	0.1 (0.003–0.164)	0.3 (0.08–0.52)	WHO 1998	[135]
The UK (Tees, Thames, and Firth of Forth estuaries)	Estuarine	NR	35	–	–	–	0.41–18.3	3.42–18.3	WHO 1998	[136]

North America										
Trinidad, Port of Spain Harbor	Marine	2007	6	638 (403–1,157)	–	–	3.49 (2.16–6.27)	–	WHO 2005	[137]
The USA (Houston Ship Channel)	Marine	2002-2003	83	4,186* (2,737–6,502)	–	–	25.3 (17.6–32.6)	–	WHO 2005	[138]
The USA (Pensacola Bay)	Marine	NR	53	1,502* 19.8–60,825	838.1 (21.3–23,453)	–	4.6 (0.2–129.3)	6.7 (0.3–145.5)	WHO 2005	[139]
The USA (Detroit)	Freshwater	1998	35	348* (68.7–1,420)	(1,338–87,300)	–	20 (3–62)	22 (3–69)	WHO 1998	[140]
The USA (Florida panhandle)	Marine	2002	32	–	–	–	0.51–77.5	–	USEPA1989	[141]

Data presented as mean (range). The Σ PCDD/PCDFs data represent the mean and range for reported measurements of all dioxin and furan congeners; an asterisk (*) indicates the mean and range represent the sum of the seventeen 2378-substituted congeners. NR indicates the sampling date was not reported by the authors in Patagonia Argentina, reported levels spatially distributed consistent with historical intense pesticide application, the urban and industrial discharges, and the presence of hydropower dams. Verhaert et al. [151] reported similar results in the Congo River basin, where, in general, levels of PCBs, PBDEs, and OCPs in different environmental compartments were low compared to other studies around the world.

Understanding POP levels in surface water is far more unpredictable and uncertain than in soil and sediment because of dynamic and ever-changing source inputs, suspended organic matter levels, and climatic conditions that typify the hydrosphere. Periodic monitoring of PCDD/PCDFs in sediments over several years in German rivers and in San Francisco Bay, California, highlights how environmental conditions can improve or degrade quickly and almost randomly (Table 3). Similar observations are often noted for other POPs in surface waters; historical sampling results provide, at best, a temporal snapshot of regional conditions that may not be representative of current conditions.

Furthermore, debate continues on surface water sampling methodologies. Menzies et al. [154], for example, has called attention to sampling methodology as possible reason for differences in marine surface water levels reported around the world, suggesting that even in so-called "surface" samples, the differences may be attributable, at least in part, as much to sampling protocol as to proximity to sources and long-range transport mechanisms. Sampling the sea water microlayer comprising the top few centimeters of the water column, Menzies et al. [154] found six different categories of POPs, chlorobenzenes, hexachlorocyclohexanes, chlordane-related compounds, organochlorine pesticides and other cyclodiene pesticides, DDT and metabolites, and polychlorinated biphenyls, in sea water collected between 1997 and 2001 near shore coastal marine locations and oceanic islands, atolls and reefs in the western Caribbean, Pacific coasts of Central and South America, and the tropical South Pacific. The concentrations observed were generally low compared to other regions in the northern hemisphere, ranging from <1 ng L^{-1} to 18.45 ng L^{-1}. Other studies also report higher concentrations of organic compounds by focusing on the sea-surface slick as compared to more traditional water sampling reported in other studies conducted in the same region [155–157]. Comparing sea-surface microlayer (SML) and seawater samples collected from Singapore's coastal marine environment and analyzed for selected chlorinated pesticides and PCBs, Wurl and Obbard [157] reported that concentration ranges of $\sum$HCH, $\sum$DDT, and $\sum$PCB in subsurface (1 m depth) seawater were 0.4–27 (mean 4.0), 0.01–0.6 (mean 0.1), and 0.05–1.8 ng L^{-1} (mean 0.5 ng L^{-1}), respectively. In the SML, the concentration ranges of $\sum$HCH, $\sum$DDT, and $\sum$PCB were 0.6–65 (mean 9.9), 0.01–0.7 (mean 0.2), and 0.07–12 ng L^{-1} (mean 1.3 ng L^{-1}), respectively.

Regional differences may also play a role in shaping global trends. According to Verhaert et al. [151], some researchers have suggested that the environmental fate of POPs in tropical ecosystems is different from temperate and cold ecosystems, because of the prevailing high temperatures and heavy rainfall [158] and higher leaching and volatilization [159]. Some suggest that tropical regions also may act as a sink since removal processes (microbial transformation and chemical hydrolysis)

Table 3 Periodic monitoring of PCDD/PCDF concentrations (pg I-TEQ/g) in surface waters in Germany and California, USA

Location	Date	Range or maximum level	Reference
Germany			
Elbe River, Elbemeßstationen	1990	36–167	[152]
Elbe River, Elbe-Messstationen	1994	339	
Elbe River, Hamburger Hafen	1992	1,500	
Elbe River, Hamburger Hafen	2001	113	
Spittelwasser	1992	1,500	
Spittelwasser	2001	83,000	
Saale/GÜSA-Messstellen	1994	57,6	
Bode/Unterlauf ab Staßfurt	1995	1,170	
California, USA (pg TEQ/L)			
Sacramento River	January 2002	0.029	[153]
	July 2002	0.048	
	January 2003	0.025	
	August 2003	0.032	
Yerba Buena Island	January 2002	0.046	
	July 2002	0.071	
	January 2003	0.026	
	August 2003	0.057	
Dumbarton Bridge	January 2002	0.259	
	July 2002	0.073	
	January 2003	0.079	
	August 2003	0.041	

The Σ PCDD/PCDFs data are reported using WHO 1998 TEFs for the sum of 17,2378-substituted congeners

may be faster compared to temperate and Arctic regions [160–163]. Recent studies indicate POPs are mobile in the tropical rainforest soils due to fast litter turnover (leading to rapid POP transfer to the subsoil) and leaching rates exceeding degradation rates especially for hydrophobic substances; these results suggest higher overall storage capacity of tropic soils in comparison to colder environments [164].

7 Summary and Conclusions

Our scientific knowledge and understanding of environmental levels have improved considerably since the 1990s. Lohmann et al. [165] point to several challenges that have not yet been overcome appreciably to the present time. Scientists would agree with the need for a “global mass balance,” which combines knowledge of source emission rates with the quantification of environmental reservoirs and final sink fluxes. However, accurate emission rates remain a challenge for legacy and emerging POPs. To date, it continues to be much easier to account for inventories of POPs (e.g., their presence in soils or sediments), but we have more difficulties in estimating historical emissions, removal fluxes through reactions, and the presence of POPs in remote locations, such as deep oceans and the polar regions.

Nizzetto et al. [166] also point to knowledge gaps and the need for better understanding of past, current, and future trends of POPs in the environment, which requires accounting for both primary emissions and reemissions to the atmosphere from reservoirs in the global environment. These reservoirs, which include soils, vegetation, biota, water bodies, and sediments, confound our understanding of environmental trends and global cycling. Improvements in sampling and analytical methodologies, monitoring program, and environmental models will continue to close the knowledge gap and improve our understanding of how human activities impact our environment. Dr. Hutzinger was at the forefront of this effort throughout his scientific career. It is left to future generations of scientists to follow in his footsteps and travel beyond.

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