

## Editorials

# Dioxin – Contemporary and Future Challenges of Historical Legacies

Dedicated to Prof. Dr. Otto Hutzinger, the founder of the DIOXIN Conference Series

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This Editorial describes background, aims and scope of a new series in ESPR entitled 'Case Studies on Dioxin and POP Contaminated Sites – Contemporary and Future Relevance and Challenges'. The Editors are Roland Weber, Mats Tysklind and Caroline Gaus. Table 2 presents a summary of the cases discussed in this series and a comprehensive review on these and other studies will open the series with a following issue of ESPR.

### The beginning of the chlorine industry and Dioxin history

It has long been recognised that significant PCDDs/PCDFs (Dioxins) formation during industrial processes commenced in the early twentieth century with the chloro alkali process and the subsequent high volume production of organochlorines (Table 1). It has only recently been recognised that high levels of Dioxin contamination may be linked to industrial activities taking place prior to the 1900s. This discovery, which was reported at the DIOXIN 2007 conference in Tokyo, demonstrated that Dioxins, particularly PCDFs in the range of 1–10 kg TEQ, were formed in a Leblanc Soda factory which operated from 1848 to 1893 in Germany (Balzer et al. 2007). Approximately 10 to 15 similar factories were operating during this time in Germany alone. The Leblanc Soda process can be considered the birth of the chemical industry in the late eighteenth century at the recycling of the HCl generated in the Leblanc process to produce chlorine and chlorinated chalk (see Table 1) and the start of the chlorine industry. From the very beginning of the chlorine industry, Dioxins were important unintentionally produced by-products, resulting in long-term challenges for research and regulatory bodies to address (for an overview of chlorine industry issues see Stringer & Johnston 2001).

### Dioxin today and the Stockholm Convention

More than a century later, the Stockholm Convention on Persistent Organic Pollutants (POPs) addresses Dioxins and Dioxin-like PCBs (all PCBs) on a global scale within the initial 'dirty dozen' (Stockholm Convention (SC) 2001, [www.pops.int](http://www.pops.int)). This international treaty promises the eventual elimination of Dioxin sources, thereby hopefully introducing the last chapter of the Dioxin history. Within the recent Stockholm Convention activities, a number of national and regional Dioxin inventories have been established

(Fiedler 2007) to provide an overview of contemporary releases and to facilitate priority setting within the National Implementation Plans. These inventories estimate that the key Dioxin emission sources today are open burning (waste and biomass), waste incineration, and the metal industry (sinter plants, secondary metal industry). However, the ongoing Dioxin legacy of the chlorine industry, related to the production, accidents, use and attempted destruction of chlorinated chemicals (see Table 1; UNEP Chemicals 2005, Masunaga et al 2001, Weber & Masunaga 2005, Weber 2007a), are not yet adequately addressed within these emission estimates. This may lead to the impression that the elimination of Dioxin sources can be achieved solely via addressing contemporary inventoried releases and that historical sources are no longer relevant.

### Contemporary versus historic dioxin emissions

The past formation of Dioxins during historical activities of the chlorine industry represents an ongoing challenge. This is best illustrated by comprehensive historical inventories compiled for Japan and Sweden, where Dioxin contamination from past pesticide use was estimated at 460 kg TEQ for Japan (Masunaga et al. 2001, Weber and Masunaga 2005) and from wood treatment for Sweden to 200 kg TEQ (Swedish EPA 2005). This exceeds the estimated TEQ emissions of contemporary PCDD/F releases from a total of 55 countries (approx. 20 kg TEQ/year (Fiedler 2007)) by ap-

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**Table 1:** Key historic events in the history of Dioxins and Dioxin-like compounds

Year	Key events
1791 (till approx. 1900)	Leblanc Soda productions and adjacent recycling of waste HCl for production of chlorine and chlorine chalk
1872	Synthesis of OCDD by Merz and Weith
1890s	Start of chloro alkali production in Germany (Griesheim 1890, Bitterfeld 1893, Ludwigshafen 1897, Rheinfelden 1898)
1897/1899	Severe acne first described by Von Betman in 1897 in chloro alkali workers. Herxheimer named the illness 'Chloracne' (1899) since he thought that it was caused by chlorine itself
1900	Start of chloro alkali production in the US (Heribert Dow, Michigan)
1918	The first report on chloracne from production of chlorinated naphthalene (PERNA)
From 1920s onwards	Along with the production of chlorinated aromatic compounds, the workers in the production and application frequently developed chloracne and liver damages. In particular the liver damages resulted in death in some cases
1929	Start of PCB production (PCB restricted 1970s; banned 2004 with Stockholm Convention)
1936	Start of PCP production (PCP restricted in many countries in the 1980s)
1940–1945	2,4-D and 2,4,5-T discovered as warfare agent by UK and US military research. Plan to use the chemicals for destruction of part of the rice harvest in the war against Japan
1949–1976	2,4,5-trichlorophenol and 2,4,5-T production: contamination of workers from day-to-day production to severe accidents: Monsanto (1949, Nitro/USA), Boehringer (1953, Ingelheim/Germany) BASF (1953, Ludwigshafen/Germany), Böhlinger (1954, Hamburg/Germany), Bayer (1954, Leverkusen/Germany), Rhone-Poulance (1956 Pont de Claix/France), Hooker Chemicals (1956 Niagara Falls/USA), Thomson Hayword (1959, New Jersey/USA), Diamond Shamrock (1960, USA), Philips Duphar (1963, Amsterdam/Netherlands), Dow Chemicals (1964, Michigan/USA), Unknown (1964, UDSSR), Spolana Neratovice (1964–69; Czech Republic); Coalite and Chemicals (1968 & 1970, Derbyshire, UK), Unknown (1970, Japan), Chemie Linz (1972/73, Linz/Austria), ICMESA/Hofmann La Roche (1976 Seveso/Italy)
1957	Discovery of 2,3,7,8-TCDD as cause of chloracne by Sorge in 2,4,5-T production in Hamburg
1962	Rachel Carlson publishes 'Silent spring' and reaches the public and politicians
1963–1970	US Agent Orange is sprayed in the Viet Nam war and contaminated large areas in South East Asia
1968	Yusho oil disease (PCB/PCDF) in rice oil in Japan
1976	Seveso accident via explosion of 2,4,5-TCP production at ICMRSA contaminating a part of the city and creating public awareness on Dioxins. Most of the 2,4,5-T was phased out during the 80 <sup>th</sup>
1977	Olie and Hutzinger discovered PCDD/PCDF in municipal waste incineration emissions
1979	Yu-cheng oil disease (PCB/PCDF) in rice oil in Taiwan
1980	Evacuation of love canal (US)
1980	First Dioxin Conference in Rome/Italy initiated by Professor Otto Hutzinger. Meanwhile 28 Dioxin Conferences were held (see <a href="http://www.dioxin20xx.org">www.dioxin20xx.org</a> )
1983	Evacuation of times beach (US)
1983–1985	General population in industrial countries found to be contaminated with PCDD/PCDF
1984	Closure of 2,4,5-T and HCH factory in Hamburg due to elevated PCDD/PCDF contamination in production residues
1986	PCDD/PCDF detected in pulp and paper mill effluents
1988	First USEPA reassessment on dioxins
1991	Emission limits of PCDD/PCDF for municipal waste incinerators in Germany
1991	Second USEPA reassessment started
1994	USEPA draft report on Dioxins
1998	Global concept for TEF factors for PCDD, PCDF and PCB (WHO)
1998	Dioxin/PCB food crises in Belgium
2001	First historic PCDD/F inventory of a country (Masunaga for Japan)
2001	Stockholm Convention on reduction of Persistent Organic Pollutants (POPs) adopted. PCDD, PCDF and PCB listed within the dirty dozen
2004	Stockholm Convention entered into force
2004	Poisoning of the Ukraine president by 2,3,7,8-TCDD
2005	Limits for Dioxin and dioxin-like PCBs in food and feed in the European Union
2005	PBDD, PBDF and brominated-chlorinated PXDD/PXDF suggested for evaluation for the TEF concept (WHO)

proximately 20 fold and 10 fold, respectively. Similarly, this contemporary PCDD/F emission inventory of 20 kg TEQ/year can be compared to other historic Dioxin emissions, such as, for example, the release of approximately 377 kg TEQ from a single former HCH and 2,4,5-T producing company in Hamburg (the only factory of this type where a complete historical inventory has been compiled (University of Bayreuth 1995). Further, Dioxin emissions of 130 to 366 kg TEQ was estimated to have occurred via spraying of defoliants in the Vietnam war (Stellman 2003, Young 2008, Young et al. 2008) and the release of up to 30 kg TEQ was estimated to have occurred by the Seveso accident in 1976 from the 2,4,5-T<sub>3</sub>CP production company ICMESA (Mocarelli 2001). The latter was the last reported accident of a long list of disasters initiated by the production of 2,4,5-TCP and 2,4,5-T (see Table 1). The contamination of parts of Seveso city and inhabitants initiated public discussion on and awareness of Dioxins and triggered the European Union Directive on the control of major-accident hazards (Seveso II) (Council Directive 96/82/EC 1996).

### The contemporary relevance of Dioxin legacies

Even today, few historical PCDD/F emissions have been inventoried or estimated. But the cases that are known illustrate that the key historic Dioxin sources far exceed the releases from all contemporary sources. Since PCDD/Fs stored in soil/sediment and waste reservoirs persist over decades or even centuries, these legacies represent a large proportion of the global Dioxin contamination today. Due to strict regulations or banning of the production of most hazardous chlori-

nated products (e.g. 2,4,5-T, PCP, CNP, PCB, HCH), changes in industrial processes (e.g. chloro alkali production) and improvements to hazardous waste incineration, the majority of these historical Dioxin sources are reduced or eliminated in many countries. However, as Dioxins persist in the environment, and contaminated soil/sediment can represent a secondary source to the food chain, these legacies will often remain of contemporary and future relevance. This has been recognised by the US EPA, who suggest that Dioxins from contaminated sites/hot spots will soon become the major source of contemporary pollution (US EPA 1994). It must also be recognised that intensive research, monitoring and regulation of Dioxins and other POPs to date have been almost entirely restricted to the relatively few highly industrialised countries of the Northern Hemisphere, and many of the 'historical' POPs (at least pesticides and PCBs) still represent current sources in developing or transition countries.

### Dioxin and POPs contaminated sites – A new series in ESPR

In order to increase our knowledge and understanding on Dioxin and other POP legacies, a special session on 'Contaminated sites – Cases, remediation, risk and policy' was held at the DIOXIN conferences in 2006 and 2007 (Weber 2007b) and will be continued at DIOXIN 2008 in Birmingham ([www.dioxin2008.org](http://www.dioxin2008.org)). In the first two sessions in 2006/2007, a total of about 50 short papers were presented. Selected cases of these sessions and some additional studies will be published in a new series of ESPR under the title 'Case Studies on Dioxin and POP Contaminated Sites – Contemporary and Future Relevance and Challenges' (Table 2). The

**Table 2:** Overview on the contaminated site cases which will be presented in detail within the new series of ESPR (the cases in this table are compiled according to the chronology of the 'chlorine cycle' and/or the origin of contamination)

Case Location (company)	Contamination source	Main contaminants	Specific issues of the case	Total TEQ <sup>a</sup> (approx.)	Approx. cost: remediation or securing (US \$)
<b>Contaminated sites from production of chlorine and organochlorines</b>					
Germany Lampertheim	Leblanc Soda production & associated chlorine production (1840s to 1893)	Heavy metals PCDD/PCDF, PAH	Oldest type of industrial PCDD/F contaminated megasite; start of chlorine production; remediation of housing estates	1–0 kg TEQ	100 million (securing)
Germany Rheinfelden (Griesheim Elektron – Dynamit Nobel – Hüls)	Chloro alkali process (1898–1985) PCP production (1972–1986)	PCDD/PCDF, PCN, AOX, Heavy metals	Migration of residues due to construction activities and resulting contamination of larger areas of the inner city; historic investigation and well managed remediation by city	8.5 kg (Chloro alkali) + 7.7 kg (PCP residues)	35 million (securing and remediation)
Sweden	Chloro alkali processes	Hg, PCDD/PCDF, PCN, AOX	Contaminated sediments and contaminated soil	1–2.5 kg	Ongoing
Finland Kymijoki River	Chlorophenol production (Ky5: TetraCP, PCP, TriCP 1940–1984) Chloro alkali process	Mercury, PCDD/PCDF Chlorinated Diphenylethers (PBDE)	Contamination of river and Baltic Sea; mobility of contaminants in river sediments; comprehensive risk assessment	28 kg	Phase of evaluation
Germany Hamburg (Böhringer)	HCH production/recycling 2,4,5-T production (1951–1984)	PCDD/PCDF, HCH	Inventory of pollutants in soil of pesticide production site; ground water contamination; movement of PCDD/PCDF with DINAPLs; closure of drinking water well	377 kg	Ca. 200 million (securing) (+ more for landfills)
Russian Federation; Ufa (Khimprom plant)	2,4,5-T production (1965–76), 2,4,5-Trichlorophenol (till 1987) 2,4-D production Chloro alkali production	PCDD/PCDF, Chlorinated organics	Industrial PCDD/PCDF contaminated megasites; contamination of larger areas of the inner city	10–200 kg	Phase of evaluation (estimation 850 million)
Australia Homebush bay (Union Carbide)	2,4-D; 2,4,5-T production Chloro alkali production	PCDD/PCDF DDT, HCB	Contamination of Sydney harbour area; stop of fisheries (since 2005); exposure of fishermen to elevated PCDD/F levels	N.E.Y	Ongoing

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Case Location (company)	Contamination source	Main contaminants	Specific issues of the case	Total TEQ <sup>a</sup> (approx.)	Approx. cost: remediation or securing (US \$)
Italy Seveso (ICMESA)	2,4,5-Trichlorophenol production	2,3,7,8-TCDD	30 year Seveso accident	30 kg	Securing
Germany Bitterfeld (Kombinat Bitterfeld)	Production of chlorinated organics, chloro alkali process magnesium production, inorganic chemicals	HCH, Chlorinated organics	Comprehensive modelling of groundwater for risk assessment and remediation strategy	N.E.Y	Ongoing
Australia Sydney (Orica)	Production of chlorinated solvents; carbon tetrachloride, perchlorethylene (PER), ethylene dichloride (EDC), vinylchloride (VCM)	Unintentionally produced hexachlorobenzene (HCB), hexachlorobutadien (HCBD), trichloroethylene (TCE)	Unintentionally produced POPs waste from production of chlorinated organics; POPs waste export; shallow and deep groundwater plumes; challenges with encapsulated waste	20,000 t HCB waste	Ongoing
Global HCH	HCH waste isomers HCH recycling	HCH isomers PCDD/PCDF	Global status; largest POPs legacy; compounds in POPs reviewing process	tons	Ongoing
Brazil Sao Paolo (Solvay)	Chloro alkali production EDC, VCM, PVC	PCDD/PCDF, mercury	PCDD/PCDF contaminated lime resulting in contamination of citrus pulp sold as feed in the European market	N.E.Y	Evaluation
Switzerland Bonfol, Kölligen (Novartis; Syngenta Clariant, Ciba, Roche)	Chemical landfills (1961–1976 and 1977–1986) from production of pesticides, pigments, dyes, plastics and pharmaceuticals	Organics (Chlorinated and non-chlorinated), PCDD/PCDF	Complete remediation of chemical landfills; drinking water contamination; dioxin inventory from production	Several 10s–100s kg	Bonfol 280 million Kölligen 450 million Full remediation
Netherlands (Philipps Dufar, Akzo, Shell)	Chemical landfills production of organochlorines (waste 2,4,5-T production, pesticides). Mixed with household waste	Chlorinated organics PCDD/PCDF	Long term monitoring strategy; funds for cleaning of leachats if site starts to leach	N.E.Y	Monitoring
Germany Selected rivers	Historically contaminated sediments (mainly industrial sources)	PCDD/PCDF, PCB, Dioxin-like activity, POPs	Remobilisation of contaminants by flooding events and construction measures	N.E.Y	
<b>Contaminated sites from application of organochlorines</b>					
Vietnam/South East Asia (most 2,4,5-T producers)	(Agent Orange)	PCDD/PCDF 2,4-D, 2,4,5-D	Impact on large area; contaminated hot spots	366 kg sprayed	Ongoing
Japan Rice Fields	Pesticide application (PCP, Chlornitrofen (CNP))	PCDD/PCDF	Large area of contaminated soil; run-off over decades; contaminated sediments; initial remediation strategy	460 kg	Ongoing
Sweden	Chloro phenol based wood preservatives	PCDD/PCDF, PCDE	Movement of dioxins as colloids; 400 to 500 contaminated sites; largest inventoried dioxin source in Sweden	5–50 kg in soil; 200 kg applied to wood	Ongoing
<b>Contaminated sites from waste recycling, destruction and disposal</b>					
Japan Tejima Island	Dumping of shredder residues (car shredder, electronic waste, industrial sludge)	Heavy metals PCB, PCDD/PCDF	Contaminated site from deposition of mainly hazardous municipal waste; complete remediation.	1.6 kg	450 million
China Guiju	E.-waste (mainly exported from US) manual recycling with lowest technology standards	Heavy metals PCDD/PCDF, PBDD/PBDF	Contaminated site from recycling activities; chlorinated and brominated PXDD/PXDF; bio-assay approach to determine total dioxin like toxicity	N.E.Y	Not started
United Kingdom Bolsover (Calcite Chemicals)	Industrial waste incinerator Production/destruction chlorophenols	PCDD/PCDF	Dioxin contaminated site from industrial waste incineration (chlorophenols and others); contamination river, land, milk	N.E.Y	
Japan Nose-Osaka	Municipal waste incinerator	PCDD/PCDF	Dioxin contaminated site from municipal waste incineration	N.E.Y	Ongoing 19 millions
<b>Contaminated sites from manufacturing processes in metal industry/inorganic chemical</b>					
Germany Rastatt (FRASA, Fahlbusch)	Cables, secondary metal (copper, zinc, silver) fly ash recycling, E-waste	Heavy metals PCDD/PCDF Brominated contaminants	Demolition of contaminated buildings, remediation and securing neighbouring buildings; groundwater contamination	N.E.Y	Ongoing 70 million (remediation and securing)
Various locations	Manufacturing Titaniumoxide (TiO <sub>2</sub> )	PCDD/PCDF	Formation of PCDD/PCDF during manufacturing of inorganic chemicals; contamination of river	N.E.Y	Evaluation

<sup>a</sup> The estimated contemporary total PCDD/PCDF release from 55 inventoried countries amounted to ca. 20 kg TEQ/year (Fiedler 2007)

N.E.Y: Not evaluated yet

key aims of this series are to present an overview on the types of contaminated sites, to evaluate the contemporary relevance of these legacies and to document evaluation strategies and remediation concepts. Most of the cases presented in this series highlight the contemporary and future relevance of POP legacies and stress that the question of whether these sites represent a future risk can only be answered by detailed site-specific evaluation.

Another aim of this series is to communicate experiences from industrialised countries to governments and other stakeholders in developing/transition countries which are operating or establishing chlorine (bromine/fluorine) industries – often without established waste management plans – and to point out that securing and/or remediation of environmental contamination are costly, generally in the order of tens or hundreds of millions of dollars (see Table 2). These costs probably exceed the historic profits or even the historic sales of many of the polluting companies!

#### **Contributions of the scientific community – From the dirty POPs dozen to the challenge of chemical mixtures**

The Stockholm Convention opened the opportunity to eliminate Dioxin sources on a global scale. In most industrialised countries where strict bans/regulation and monitoring of PCDD/F emissions were enforced over the past decades, human exposure to PCDD/PCDF has already declined significantly. This has been facilitated by the scientific community, from discovering the problems, providing an understanding on the complex pathways and processes of these compounds including their toxicity, to finding solutions that allow reduction of emissions and their monitoring. This development to the modern diverse Dioxin science was greatly facilitated by the annual DIOXIN Conference series which was initiated in 1980 by Otto Hutzinger and is now attended by around 1000 experts each year.

Over the past 10 years, the focus of the DIOXIN Conference broadened to cover a wider range of persistent chlorinated, brominated and fluorinated organic pollutants, and the conference has effectively developed into a comprehensive POPs Symposium (Weber 2007b). This reflects the increasingly effective control of Dioxin sources and associated decreases in contamination, together with the growing concern of the scientific community regarding a wider range of chemical contaminants and especially their potential connection to adverse human health effects such as allergies and cancer rates (Clapp et al 2005). The key concern of Dr. Linda Birnbaum (USEPA) in the closing session of DIOXIN 2007 was the challenges of chemical mixtures with regard to environmental and human health concerns. Today, the daily chemical 'cocktail' consists of thousands of contemporarily produced chemicals in addition to the remainder of historical POPs such as Dioxins. Therefore, a key task for the scientific community is to provide the understanding necessary to address contamination legacies, a strategic approach to address contemporary produced chemicals and their combined arising issues. The Initiative for an International Panel on Chemical Pollution (IPCP, [www.ipcp.ch](http://www.ipcp.ch)) is an important step in this direction (Scheringer et al. 2006).

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