第14回ダイオキシン国際会議

— 有機塩素化合物研究の最新動向 —

グレーエム・アリンソン, 上岡真由美

[平成 7 年 1 月 10 日受理]

Current Topics in Organohalogen Research A Review of the Environmental Aspects of 'Dioxin '94'

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[Received January, 10, 1995]

Summary

Residues of polychlorinated dibenzo-p-dioxins, dibenzofurans, and biphenyls have been identified in all parts of the global ecosystem. The environmental persistence, bioaccumulation, and toxic properties of these chemicals has given rise to much concern over the last few decades, and they were the subject of the three hundred and thirty five oral and poster presentations given to "Dioxin '94 - The 14th International Symposium on Chlorinated Dioxins, PCBs and Related Compounds" held in Kyoto, Japan November 21-25, 1994. The lectures and posters presented at the National Overviews, Analytical Methods, Environmental Levels and the Polar Environment sections of the Symposium are reviewed. A full list of presentations to all twelve sections is given.

現在, 地球生態系のあらゆる場所で, 多塩素化ジベンゾ-p-ダイオキシン, ジベンゾフラン, そしてビフェニル類の残留が確認されている。これらの物質の環境における残存性, 生物濃縮, 毒性の観点において特に最近数十年にわたり重要視されており, これらに関する環境汚染問題を討議し, 最新の研究報告を交換するため, 第14回ダイオキシン国際会議 (第14回塩素化ダイオキシン, PCB, ならびに関連化合物国際シンポジウム) が, 1994年11月21日から25日まで京都において開催され, 総数335 (登録数) の口述及びポスターによる発表が行われた。シンポジウムを構成する全12部門のうち, 4部門 - 各国の規制の動向, 分析方法, 環境レベル, 極地環境の研究動向をレビューし, また, 発表全リストを掲載する。

INTRODUCTION

Environmental degradation and pollution has always accompanied human activities. However, the environmental consequences of the rapid growth in economic development and industrialization witnessed this century has become a matter of grave concern. Residues of man-made chemicals have been identified in every part of the global ecosystem. Of major concern, due to their environmental persistence, bioaccumulation, and toxicity, are the polychlorinated dibenzo-p-dioxins (PCDDs, I), the related polychlorinated dibenzofurans (PCDFs, II), and the polychlorinated biphenyls (PCBs, III)⁽¹⁻³⁾. These chemicals, which exhibit similar physical, chemical and biochemical properties, have been extensively studied over the last few decades, but the human and environmental damage caused by current environmental levels is still unclear^(4,5).

PCDDs and PCDFs are often labeled "the most toxic man-made chemicals known". They are not produced commercially but found as unwanted and undesirable by-products or impurities in a wide range of industrial products and processes *e.g.* in agrochemicals, incinerator emissions, paper processing wastes, and even car exhaust fumes. In contrast, some PCB mixtures had such acceptable and desirable physical and chemical properties that they were produced on an industrial scale. However, some of the properties that made PCBs so desirable *i.e.* high thermal stability, chemical resistance, and excellent dielectric properties, also contribute to their persistence in the environment. Environmental sources of PCBs include volatilization from landfills containing PCB-contaminated or containing wastes, sewage sludges, spills, illegal disposal and incinerator emissions and wastes⁽⁵⁾. Because of the great quantity of PCBs already in the environment, and the much larger amount still in use, the potential for environmental damage is enormous.

Exposure to PCDD/DFs gives rise to a wide range of biological effects, including chloracne, hepatic damage

and immunotoxicity. The most toxic of the PCDD/DF congeners are considered to be the 2,3,7,8-tetrachlorinated derivatives in which the chlorine atoms are located in the four lateral substitution positions⁽⁴⁾. Recent evidence has suggested that a large proportion of PCB induced toxicity may be due to the small amounts of those PCBs having chlorine atoms in the vicinal *meta* (3,5) and *para* (4) positions but without *ortho* (2) substitution. Coplanar, or non-*ortho*, PCBs such as 3,3',4,4'-T₄CB (IV), 3,3',4,4',5-P₅CB (V) and 3,3',4,4',5,5'-H₆CB (VI) exhibit the maximum amount of planar conformation and give rise to toxic responses similar to 2,3,7,8-TCDD. The large variation in toxic potential between individual members of each group, and the similarities in biological response between families, has led to the promotion of the concept of International Toxic Equivalence Factors (I-TEQs or I-TEFs) in which the toxic potential of a compound is related to that of 2,3,7,8-TCDD (2,3,7,8-TCDD I-TEQ = 1).

As well as PCDDs, PCDFs and PCBs, there are other polychlorinated organic compounds which are persistent environmental contaminants with the potential to cause adverse biological and environmental effects. As these effects have been studied and quantified, a parallel rise in environmental legislation has been seen. The growing number of national and supranational regulatory requirements has encouraged the development of sophisticated and sensitive analytical techniques capable of detecting environmental contaminants in minute quantities. These, and other topics were discussed in the three hundred and thirty five oral and poster presentations were given to "Dioxin '94 — The 14th International Symposium on Chlorinated Dioxins, PCBs and Related Compounds" held in Kyoto, Japan November 21-25, 1994. The lectures and posters presented at four of the twelve sections making up the Symposium—namely, National Overviews, Analytical Methods, Environmental Levels and the Polar Environment, are reviewed herein. Presentations given to those sections covering Sources and Formation, Emission Control, the Metabolism of PCB and Related Compounds, PCB Toxicology, Human Toxicology, Toxicology, Ecotoxicology and Yu-Cheng are not covered. However, a full list of presentations to all twelve sections is given⁽⁶⁾.

NATIONAL OVERVIEWS

The WHO/EURO activities on risk assessment and regulatory measures concerning PCDDs, PCDFs and PCBs were discussed by Ahlborg⁽⁷⁾. Their programme

aims to develop safety management strategies, evaluate, control and minimize exposures, and to assess and minimize health risks. A second round of inter-laboratory human breast milk analyses has indicated that PCDD/DF levels are not increasing in Europe⁽⁸⁾ and, therefore, has concluded that there is no need to change its recommendations concerning breast feeding. The WHO/EURO recognises that some of the planar PCBs are equally, or more, toxic than many PCDD/DFs. Consequently, a set of interim I-TEFs have been devised for these dioxin-like PCBs⁽⁹⁾. A series of inter-laboratory studies on dietary exposure to PCDDs, PCDFs and PCBs, levels of these compounds in human milk, blood, cows milk, and fish, and investigations into emissions from municipal solid waste (MSW) and municipal sewage sludge (MSS) incinerators have been undertaken. Publication of many of these results is expected in 1995.

The United States Environmental Protection Agency (EPA) recognises that although dioxins are produced in very small quantities ($\sim 14\text{kg}$ I-TEQ per year) the high toxicity of these compounds means they must be treated as significant environmental pollutants. Farland reported that the U.S. EPA first published a review of the health effects caused by dioxins in 1985⁽¹⁰⁾. In September 1994 the EPA published the first public draft of a full reassessment of the sources, levels of exposure and human health effects of dioxins. The draft report confirms the EPA's stance that dioxin is a proven animal carcinogen, a probable human carcinogen, and that exposure can result in a number of non-cancer health effects. The draft identifies wastes incineration for $\sim 95\%$ of US atmospheric emissions and confirms that the major exposure pathway for non-occupationally exposed humans is through their food. The review also identified significant data gaps and calls for further information on dioxin sources, releases, and levels in air, water, soil, human food, animal feeds, and human tissues.

Kimura discussed Japan's experience in dealing with dioxin problems⁽¹¹⁾. After reports that Japanese MSW incinerators were emitting dioxins were made by Tatsukawa⁽¹²⁾, a no-adverse-effect-through-inhalation emission level of $0.1\text{ ng / kg body weight / day}$ was recommended^(13,14). In 1990, the Ministry of Health and Welfare issued a guideline promoting dioxin emission reduction through the use of best available technologies⁽¹⁵⁾. The new target value for emissions is $0.5\text{ ng I-TEQ / m}^3$. This value is estimated to reduce Japanese emissions by $\sim 90\%$. The results of a joint Environment Agency,

Ministry of Health and Welfare and Fisheries Agency study into dioxin pollution caused by the paper and pulp industry indicated that mill effluent water and boiler emissions contain PCDD/DFs, but that air pollution and fish contamination levels around mills are similar to control areas⁽¹⁶⁾. The Environment Agency has issued guidelines which will reduce such emissions. The principal measures involve reduced use of chlorine and its eventual elimination through the introduction of alternative bleaching agents, and proper incineration controls on mill boilers.

Johansson gave an overview of the PCDD/DF, PCB and related compound contamination in Scandinavia using Sweden as the regional model⁽¹⁷⁾ while Newstead outlined the United Kingdom's use of Integrated Pollution Control to control emissions in that country⁽¹⁸⁾. However, European levels and regulatory actions are perhaps best represented by the actions taken by the Netherlands and Germany.

Liem gave MSW and effluent emission figures for the Netherlands⁽¹⁹⁾. An MSW emissions guideline of $0.1\text{ ng I-TEQ / m}^3$ has now become law. Emissions to Dutch surface waters are estimated to be 3g I-TEQ / yr . The median dietary intake for adults in the Netherlands is $1\text{ pg I-TEQ / kg body weight / day}$. If co-planar PCBs are included this figure rises to $2.4\text{ pg I-TEQ / kg body weight / day}$. Background levels in top soil are $1\text{--}20\text{ ng I-TEQ / kg dry soil}$, air concentrations are $\sim 24\text{ fg I-TEQ / m}^3$, and airborne deposition is $8\text{ ng I-TEQ / m}^2\text{ / yr}$. Dutch Rhine sediments have been found to contain an average of $220\text{ ng I-TEQ / kg dry sediment}$. Higher levels are found near known old sources.

Basler discussed the regulatory measures in Germany⁽²⁰⁾. The 1990 Ordinance on Combustion Plants prescribes a limit value for dioxins in exhaust gases of $0.1\text{ ng I-TEQ / m}^3$, which is predicted to produce a one hundred-fold reduction in emissions to $\sim 4\text{ g I-TEQ / yr}$. The ban on chlorinated and brominated hydrocarbon scavengers in leaded petrol, and restrictions on the use of pentachlorophenol (PCP) herbicides and PCBs since 1989 has reduced German environmental inputs by more than 5 kg I-TEQ / yr . The 1992 Ordinance on Sewage Sludge introduced limit values for sewage sludges used as fertilisers. The first report on the Joint Working Group on Dioxins set a target value of $< 5\text{ ng I-TEQ / kg dry soil}$ ⁽²¹⁾. Unrestricted agricultural cultivation is allowed when levels are between $5\text{--}40\text{ ng I-TEQ / kg dry soil}$, but action is recommended should higher levels be

found. Similar guidelines were imposed for milk and dairy products. The desired, long term target is < 0.9 pg I-TEQ / g milk fat. Levels above 3.0 pg I-TEQ / g milk fat will trigger a cessation of milk/dairy product supply to the public and a change of land use. Limit values for PCDD/DFs and PBrDD/DFs in products not already covered by specific legislation are being formulated.

ANALYTICAL METHODS

The widespread availability of accurate and precise analysis has led to ever greater demands being placed on public and private laboratories. In general, the main purpose of most analyses is to indicate the actual levels of pollution / contamination and / or whether regulatory requirements are being met. Although much of the actual analytical measurement has now been automated, considerable resources and efforts are expended in the pre-measurement steps i.e. sample collection, preservation, and clean up. There is a recognised need to develop and introduce new and improved techniques that will reduce the level of resources devoted to routine analysis and assessment of chemical pollution, while maintaining high analytical sensitivity and the ability to satisfy regulatory requirements.

Interlaboratory calibration studies and standard, or certified, reference materials (SRMs / CRMs) are used to validate analytical methods and results. Rappe presented the results from an international intercalibration study on PCDD/DFs in fly ash extracts⁽²²⁾. Six Japanese, one American, one Finnish and one Swedish laboratory took part in the study. All participating laboratories showed good agreement in reporting I-TEQs (r. s. d = 25%). The statistical variation within laboratories was larger than the differences between them. Rappe remarked that the interlaboratory calibration study was useful in evaluating the results produced by different laboratories and should be repeated incorporating a study of non-*ortho* PCBs.

de Boer and van der Meer presented the results of the combined International Council for the Exploration of the Sea, the Intergovernmental Oceanographic Commission, and the Oslo and Paris Commissions (ICES/IOC/OSPARCOM) study on PCBs in seals, fish and marine sediment⁽²³⁾. Seventy five laboratories in eighteen countries participated in the study. The major conclusions were that the stepwise design of the study led to improved participant performance, that principal compo-

nent analysis (PCA) is a powerful evaluation tool, long term analytical variation would be improved by reference materials, a large percentage of marine laboratories now agrees on the determination of PCBs 118, 138 and 153 in marine sediment, and that further study of PCBs in lean fish are required.

That CRM matrix and analyte concentrations should be representative of the samples to be analysed is accepted wisdom^(24,25). Morita introduced a new series of CRMs (NIES CRM No.'s 1-10) containing all the PCDD/DF congeners that occur in incineration processes and which should, therefore, be extremely useful for chemical speciation and trace organic analysis of incinerator fly ash⁽²⁶⁾.

Chambers introduced and described the NIST SRMs No.'s 1945 (Whale Blubber), 1974 (Organics in Mussel Tissue) and 1941a (Organics in Marine Sediments)⁽²⁷⁾. SRM 1945 is a frozen whale blubber certified for twenty-six PCB congeners and fifteen pesticides. SRM 1974 is a frozen mussel tissue homogenate with certified values for nine polynuclear aromatics (PAHs). SRM 1941a is an homogenised marine sediment with certified values for a range of polychlorinated hydrocarbons (pCHCs). Chambers observed that these SRMs are homogenous for many organic contaminants with similar physical and chemical properties to PCDD/DFs. Therefore, if these SRMs were also certified for PCDDs and PCDFs, they would be very useful for many of the delegates in marine organism and sediment research.

Accurate and precise analysis of trace organic compounds often requires the use of internal standards. Lujik remarked that the analysis of brominated and mixed halogenated PCDD/DFs is often complicated by the lack of labeled standards. He discussed the synthesis of $^{13}\text{C}_{12}$ -labeled mixed halogenated PXDDs ($\text{X} = \text{Cl}, \text{Br}$) by direct bromination of dibenzo-p-dioxin standards, and partial exchange of bromine in PBrDD by CuCl_2/HCl in a gas-solid phase reaction on a model fly ash system⁽²⁸⁾. Yields were 40-135%. In a similar vein McLachlan described the photochemical synthesis of $^{13}\text{C}_{12}$ -labeled mono- and di- BrDD, and mono, di, and tri BrDF from commercially available $^{13}\text{C}_{12}$ -labeled 237-T₃BrDD and 2378-T₄BrDF, respectively⁽²⁹⁾. Homolytic cleavage of the Ar-Br bond is followed by the abstraction of a hydrogen atom from the solvent. In mixed brominated / chlorinated aromatic compounds the bromine atom is exclusively replaced. Yields were 50-80%.

Sample collection, preparation and clean-up, and con-

gener separation and identification are important considerations in environmental analysis. For some matrices, such as snow or water, the physical and chemical properties of organochlorine contaminants results in very low levels of contamination. Matsumura described the development of a pre-concentration system for PCDD/DFs in sea water⁽³⁰⁾. Essentially, large volumes of sea water are pumped rapidly through a series of replaceable cartridge filters and a resin column. This combination traps both truly dissolved and particulate-bound PCDD/DFs. These are determined by subsequent extraction of the filters / column and analysis of the resulting solution. Determination at the femto gram / litre levels is possible.

Determination of PCDD/DFs and PCBs in human milk samples can be difficult due to extremely low contamination levels, and the high sensitivity and selectivity required of the analytical method. Yamamoto described a rapid clean-up method for milk samples involving a sequential three column separation of the sample matrix⁽³¹⁾. After saponification, a multi-layer silica gel column was used to remove fatty acids, the PCDD/DFs and co-planar PCBs were then separated from other organochlorine compounds on an activated carbon column, and finally isolated after neutral alumina column chromatography.

The advantages of supercritical fluid extraction (SFE) in overcoming some of the disadvantages of conventional liquid extraction were described by Akgerman⁽³²⁾ and Kawamoto⁽³³⁾. These advantages include the ability of supercritical fluids (SCFs) to dissolve organic contaminants from soils, and also the potential to vary the SCF solvent power by small changes in operational parameters. Carbon dioxide is the favourite 'solvent' since it is non-toxic, environmentally acceptable, non-inflammable and does not introduce safety hazards during use.

A critical step in the determination of co-planar PCB congeners is the separation of these congeners from the bulk of the PCBs. Generally, only a small fraction of the sample ends up being presented to any GC-MS system. However, on-line coupled LC-GC techniques present a larger proportion of the sample to the analytical system. Grimvall introduced a coupled LC-GC-MS system for the determination of 0.5-1 pg / g PCB 126 and 169 in human blood plasma⁽³⁴⁾. Haglund described how two dimensional HPLC (LC-LC) was coupled to a GC-ECD and used to separate indicator congeners of mono- and non-*ortho* PCBs and PCDFs from other, interfering compounds⁽³⁵⁾. It was claimed that the data produced was comparable

with classical GC-HRMS.

When using HPLC systems, the physical and chemical properties of the species under investigation may limit or define the choice of column that can be used. Atuma discussed the use of a porous graphite HPLC column for the separation and isolation of PCBs in fish⁽³⁶⁾. Compared to an activated charcoal column the porous graphite column required less solvent and offered faster PCB separation. Tanaka reported the use of electron-donor and electron-acceptor stationary phases for the separation of all di- and tri-chlorinated dioxins, as well as a number of higher chlorinated congeners, by reversed phase LC⁽³⁷⁾. The stationary phases also permitted isomer identification. Of the columns investigated, 2- (1-pyrenyl)ethylsilyl phases preferentially retained PCDDs with the more symmetrical chlorine substitution, while 2- (nitrophenyl) ethylsilyl and 3- (p-nitrophenoxy) propylsilyl phases showed preference for PCDDs with the more proximal chlorine substituents. Naikwadi described the development of main-chain liquid crystal polymer (MCLCP) stationary phases for use in GC capillary columns⁽³⁸⁾. The general structure of the MCLCP consists of alternating dimethyl siloxane and liquid crystal units. The maximum operating temperature of the columns is around 300°C. Fused silica capillary columns with MCLCP showed superior selectivity for the separation of 2378-substituted PCDDs. Complete separation of OCDD from OCDF could also be achieved.

Although one of the favoured methods for dioxin analysis is high resolution GC-MS, not every laboratory or researcher has access to such systems. Therefore, other analytical methods are always in demand. Patterson reported that isomer differentiation of PCDD isomer pairs was possible by using a combination of three chromatographic techniques (GC, HPLC and micellar electrokinetic chromatography) and three spectroscopic techniques (FT-IR, ¹³C and ¹H NMR)⁽³⁹⁾. Isomer-pair structural assignments showed an excellent correspondence in terms of isomer pair percentages. The high quality correspondence of the independently determined assignments substantially increases the confidence level in isomer identification.

Zimmermann presented the first resonance-enhanced laser mass spectrometry results for a PCDF⁽⁴⁰⁾. This technique combines resonance-enhanced multi-photon ionisation with lasers and time-of-flight mass spectrometry. As a two-dimensional analytical method the technique shows high selectivity and sensitivity, which

may lead to many on-line / in-process applications. A knowledge of the appropriate molecular UV spectroscopic and ionisation potential data is required. This data can be obtained by molecular orbital calculations. Zimmermann compared ionisation potentials of chlorinated benzenes, dibenzo-p-dioxin, 2378-TCDD and OCDD calculated by seven semi-empirical quantum-mechanical methods- MINDO3, MNDO, AM1, PM3, INDO, CNDO and ZINDO/1⁽⁴¹⁾. All methods tended to produce high values. However, if experimental data is available for at least one compound in a class a correction factor can be introduced. Zimmermann commented that he found that his experimental data was best reproduced by the AM1 and PM3 programs.

A system for the rapid screening for 2378-TCDD using a competitive enzyme immunoassay (EIA) was described by Harrison⁽⁴²⁾. The system is specific for 2378-TCDD and related congeners. Sample preparation can be automated, and allows immunoassay screening of soils, serum, water and many other matrices. Concentration factors and endpoint conditions are flexible. The analysis is performed by the addition of the prepared sample extract to antibody coated microwells. After incubation with a competitor conjugate (capable of binding specifically to unoccupied antibody sites), an enzyme substrate is added for colour development. The more intense the colour the LESS dioxin is present in the sample. Detection of 10 ppb 2378-TCDD in soil by direct analysis of crude extracts is possible. The system offers significant improvements in speed, sample throughput and cost compared to GC-MS.

ENVIRONMENTAL LEVELS

The keynote speech in this section came from Rappe and Kjeller who discussed the historical trends and budget calculations for environmental PCDDs and PCDFs in Sweden⁽⁸¹⁾. Their main conclusions were that natural formation of PCDD/DFs occurs, but combustion was the primary source of PCDD/DFs found in historical samples, that 19th Century contamination most likely resulted in contaminated foodstuffs and concomitant exposure of humans, that post-1940 samples are influenced by the HpCDD and OCDD found in the commercial pentachlorophenol used at that time, combustion is the primary source of lower chlorinated PCDD/DFs, and that environmental levels peaked in the late 1960s and are now decreasing, but have yet to reach historical levels.

The results of the Swedish Dioxin Survey were pre-

sented by de Wit^(82,83). The Swedish Environmental Protection Agency conducted this five year survey (1988-93) to determine the levels of PCDD/DFs in the Swedish environment. The effects of geographical distribution, bioaccumulation, bio-magnification time trends, as well as species, age, size and sex on body concentrations were studied. A considerable number of species were analysed- herring, pike, burbot, whitefish, Arctic char, salmon, trout, plaice, mackerel, cod, crab, lobster, mussels, guillemot, white-tailed sea eagle, osprey, wolf, fox, moose, reindeer, badger, otter, rabbit, grey, common, harp and ringed seals, and porpoise. The collection sites represented hot spots, areas with diffuse pollution, less industrialised areas and remote areas. Generally speaking, PCDD/DF concentrations in aquatic ecosystems are much higher than those found in terrestrial animals. Organisms from the Baltic Sea have the highest levels. The low PCDD/DF levels found in seals and porpoise indicate that these species do not biomagnify these substances. However, the levels in fish eating birds such as guillemot, sea eagle and osprey are much greater than the fish species they prey on, and do seem to suggest they biomagnify PCDD/DF and co-planar PCBs. The highest concentrations were found in adult osprey from Southern Sweden. Concentrations in guillemot eggs and pike collected between 1969 and 1992 have slowly decreased over this time period, indicating that the actions taken to ban PCB, pentachlorophenol and 2,4,5-T use has led to reduced inputs of PCDD/DFs and PCBs to the environment.

One major theme that emerged at 'DIOXIN '94' was the use of statistical analysis to compare PCDD/PCDF/PCB congener profiles between samples and sampling sites. Geographically separated wildlife populations show different contamination patterns. Statistical analysis can help illuminate similarities and differences that otherwise would be obscured by the sheer volume of data. Of the statistical methods used principal component analysis (PCA) was very much *in vogue*.

Yamamoto used to PCA to characterise the similarities and differences in PCDD/DFs found in urban and remote Japanese aquatic sediments⁽⁸⁰⁾. Levels found in Japanese aquatic sediments were within the ranges reported for other industrialised countries. Concentrations in Tokyo Bay and Osaka Bay mainly included products of combustion processes, indicating the contribution of the large, heavily industrialised areas behind the bays. These urban sediments had higher levels than more remote areas. Lake sediment PCDD/DFs were mainly de-

rived from the former use of PCP in the associated hydrological basins, while river sediments from northern Japan were characterised by pollution with pesticide derived dioxins i.e. 1368 and 1379-TCDD.

Fiedler and Rappe presented a two part evaluation of a southern Mississippi river system suspected of being contaminated by PCDD/DFs emitted by a pulp mill^(84,85). Using cluster analysis, PCA and multivariate data analysis (MDA) sediment and soil samples from above and below the pulp mill were compared with soil data from recently flooded areas, targeted sediment with a known dioxin contamination history, sewage sludges, and effluent from the plant itself. None of the statistical methods identified the pulp mill as the source of the PCDD/DF found in the river drainage.

The levels of PCBs in sediments, amphipods, isopods, zoo- and phyto-plankton, mysids, perch, four horned sculpin, herring, salmon, salmon trout and white-fish samples collected from four sites in the Gulf of Bothnia were also subjected to a chemometric comparison by van Bavel⁽¹⁴⁰⁾. Their conclusions were that levels of PCBs in sediment and amphipods differs throughout the Gulf of Bothnia, with highest levels seen in the south; that there are large seasonal fluctuations in PCB levels; and that PCA revealed a significant difference in the PCB congener pattern between the sub-Arctic sampling site and the other three, more southerly regions.

In 1988 there was an epidemic of European seal disease in the North Sea. The PCB congener patterns found in dead harbour seals collected from three geographically separate regions were compared by PCA and linear discriminant analysis by Storr-Hanssen⁽⁸⁶⁾. The congener patterns were characteristic for each animal and, by implication, for each area. To gain a better understanding of the underlying processes behind these results, groups of captive female harbour seals were fed with fish from two different locations: either plaice from the Wadden Sea or mackerel from the Atlantic. PCB levels and congener patterns were determined in the fish and seals. It was possible to discriminate between the seals fed Wadden Sea fish, and those fed Atlantic fish. There was also a clear difference between the PCB congener patterns found in seals and those found in the fish.

Of perennial concern is the human exposure to PCBs, PCDDs and PCDFs through the food chain, especially via high fat foods such as human milk, cow's milk and dairy products. van Cleuvenbergen produced data on PCDD/DF levels in human milk in Flanders, Belgium⁽⁸⁷⁾.

The average I-TEQ (fat basis) was ~ 35 pg / g. This dioxin burden agrees well with that documented from other West European countries. Dioxins and furans contributed almost equally to the I-TEQ level. A tendency towards higher values at urban locations was observed. There was some suggestion that the PCDD/DF levels in that part of Europe, which have tended to be the highest world-wide, are decreasing slowly.

It is generally accepted that dioxin contamination of the environment increased significantly with the advent of industrialisation and intensive agriculture^(4,5). Baldasari presented PCB contamination data for milk and cheese samples from the Valassina region of northern Italy⁽⁸⁸⁾. In that area, cattle are allowed to graze freely in the high mountain meadows during the summer, then fed fodder during the winter. All milk and cheese samples showed similar PCB congener profiles. PCB levels in both milk and cheese samples were consistently higher than other areas of Italy, and, interestingly, higher in the summer than in the winter, despite the fact that the cattle were presumed to be feeding on low contamination level sites. A specific pollution source of PCB existing near the Valassina farms was proposed as a reason for this variation.

Some remote areas are contaminated even though there are no obvious local sources. Long range atmospheric transport is a suggested mechanism, and sediment core sampling is being used to confirm this theory. Kjeller and Rappe presented data from a sediment core taken from the Baltic Sea⁽⁸⁹⁾. The sediment is of an accumulation type and has stratified into lamina discs, each disc representing ~ 30 years. PCDD/DFs were detected in all sediment discs. The deepest and oldest discs representing the late 19th century have a full range of PCDD/DF congeners. Deposition from ~ 1880 – ~ 1960 shows only low contamination levels, with only a marginal increase over that time period. This is suggestive of contamination from combustion sources only. Contamination levels increased rapidly after this time, indicative of the increased common usage of PCDD/DF contaminated products.

An alternative route to historical trend data might be to use sediment from an undisturbed area. The open ocean floor would seem ideal site. Mass accumulation is slow but essentially at a constant rate undisturbed by tides or waves. Matsumura and Kato collected a sample from the ocean floor 4030 m below sea level⁽⁹⁰⁾. The sediment core was cut into slices. The results from the sur-

face slice showed concentrations of the tetra, penta, hexa, hepta and octa PCDD and PCDF congeners of 1.6, 5, 19, 39, 42 and 14, 7, 9, 33, 20 pg / g, respectively.

The concentrations and regional differences in the levels and congener patterns of PCDDs, PCDFs and PCBs in Fukuoka Prefecture, Japan river and coastal sediments were investigated by Ohsaki⁽⁹¹⁾, who concluded that congener profiles in sediments were different from those observed in nearby soils, that the PCDD, PCDFs and PCBs in river and coastal sediments may have been derived from agricultural sources, and the levels of these chemicals varied with sampling site in the order of river-lower reach > coast > river, upper reach.

Remediation of PCDD, PCDF or PCB contaminated sites can be very expensive and often requires extreme measures. Biological degradation offers a more 'environmentally friendly' way of decontaminating such sites. Takada reported the degradation of PCDD/DFs by the ligninolytic fungus *Phanerochaete sordida* YK-624⁽⁹²⁾. 2378-TCDD and higher chlorinated congeners were degraded substantially over a fourteen day period. Rates of degradation were promoted by adding glucose and oxygen.

POLAR ENVIRONMENT

Of the many chlorinated hydrocarbon (CHC) compounds released into the environment over the years, polychlorinated camphenes (PCCs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), DDT, PCBs and PCDD/DFs have been identified in the air, snow, ocean water and the biota of the Arctic ecosystem⁽¹³⁰⁻³⁴⁾. CHCs are resistant to environmental degradation by chemical and biological processes, and have long environmental half-lives even in temperate climates. This process may be slowed considerably in the harsher polar regions.

Norstrom remarked that these chemicals have sufficiently high vapour pressures and Henry's Law constants to permit global distribution in the troposphere via cycling between the atmosphere and land or surface waters⁽¹³⁵⁾. The atmospheric residence time of CHCs is several months. The Arctic, therefore, may be acting as a "cold-finger" sink for CHCs, leading to global distillation from warmer to colder environments. Of the CHCs found in the Arctic, HCHs and HCB (0.58 and 0.19 ng / m³) dominate in the air, with DDTs, chlordane and PCBs an order of magnitude lower. HCHs are still the dominant compound in the snow and surf (1-2 and 4-5 ng / l cf. 2-3 ng / l in the Bering Sea and < 2 ng / l in the mid-

Pacific Ocean). The importance of DDTs and PCBs relative to other CHCs increases through the food chain from water to zooplankton to amphipods to cod to beluga. Biomagnification of these compounds is estimated to be 10⁷ - 10⁹. Seals do not biomagnify HCHs, HCB or PCCs, while polar bears mainly accumulate PCBs and oxychlordane. The logarithm of the concentration of total chlordane, total PCBs and CB-153 was linearly correlated to trophic level. Susceptible compounds within the chlordane and PCB classes are metabolised by polar bears, seals, walrus and birds resulting in lower biomagnification factors.

Oehme determined the levels of PCDDs, PCDFs, PCBs, and pesticides in the brains and blubber of harp seals (*Phoca groenlandica*) caught east of Greenland in the West Ice region⁽¹³⁶⁾. Congener specific analysis was undertaken and gave a 2378 I-TEQ of 3.9 - 10.4 pg / g for PCDD/DFs: 2378-TCDD, 123678-HxCDD, OCDD, and 2378-TCDF levels were 0.5-1.2, 1.4-9.4, 1.3-6.6 and 3.6-10.4 pg / g, respectively. The non-ortho PCBs' I-TEQs were 4-10 times higher than that of the PCDD/DFs at 16-96 pg / g ; 33'44'-TeCB, 33'44'5-PeCB and 33'44'55'-HxCB levels were 91-410, 130-472 and 24-942 pg / g, respectively. DDT and DDT-metabolite levels were an order of magnitude lower. Oehme also commented on the similarities, differences, and relative concentration patterns of CHCs in the air and biota of the two polar regions⁽¹³⁷⁾. Much lower levels are found in the Antarctic. This was attributed to less industry and less intensive agriculture in the Southern Hemisphere combining with meteorological and ocean current conditions to produce less favourable conditions for long range transport. When compared to the Arctic, HCH and HCB are approximately 5 times lower in Antarctic air, with γ -HCH the dominant isomer. The levels and congener patterns of higher chlorinated PCBs were comparable, but dioxin and dibenzofuran comparisons were difficult because of poor data presentation in the published literature and species variation. The congener patterns in the Antarctic differ from those found at the North Pole, perhaps indicating different emission patterns, food habits or species variation in the different hemispheres. An Antarctic 2378-TCDD I-TEQ of 2 pg / g was given.

Polischuk has been studying the relationship between PCB concentration, body burden, and percent body fat in female polar bears (*Ursus maritimus*) inhabiting the western Hudson Bay area of Canada⁽¹³⁸⁾. These bears can lose up to 90% of their body weight during their

winter fasting period. Samples of hip adipose tissue were taken between 1992-1994 from a number of bears at the beginning and end of their fasting periods. The samples showed that PCBs 99, 153, 138/163, 180 and 170 accounted for greater than 95% of the total PCB congeners. There was no correlation between PCB body burden with an animal's total mass of fat (kg fat) or the percentage of body mass that this represented (% fat). However, sequential sampling of individual bears indicated that female polar bears may decrease their PCB body burden during pregnancy and the early stages of lactation, rais-

ing fears for the health and welfare of the neonates and cubs.

Paasivirta measured levels of PCC, γ -HCH, α -HCH, HCBz, DDT and PCBs in Arctic Cod and Salmon collected in the Teno river area of the North Cape, and in the Simo River area of the Baltic Sea⁽¹³⁹⁾. The most abundant contaminants were chlorohydrocarbons. Of these only DDT showed a significant linear regression over the last decade. Most of the dioxin-type load in Arctic fish was due to the PCBs.

はじめに

環境破壊と汚染は、常に人間の行動に伴っているが、今世紀の急速な経済発展と産業化の結果、非常に重大な問題となった。人の手によってつくられた化学物質の残留物は、今や地球生態系のあらゆるところで発見されている。これら全ての化学物質のうち、多塩素化ジベンゾ-p-ダイオキシン (PCDDs, I), それに関連した多塩素化ジベンゾフラン (PCDFs, II), そして多塩素化ビフェニル (PCBs, III) は、その環境における残存性、生物濃縮、毒性の観点において特に重要視されている⁽¹⁻³⁾。これらの化学物質は、類似の物理的、化学的、生化学的性質を持ち、最近数十年にわたって熱心に研究されているが、実際の環境中の濃度がヒトや環境破壊にどのように影響しているかははっきりしていない^(4,5)。

PCDDs と PCDFs (総称して、ダイオキシンとよぶ) は、「ヒトがつくった化学物質のなかで最も毒性の高いもの」として知られている。これらは、工業的に合成されたものではなく、不要な望ましくない副産物もしくは不純物として、広範囲に製品中もしくは製造過程で発見された。例えば、農業化学薬品やゴミ焼却炉、パルプ・紙精製過程廃棄物、さらに自動車の排気からも発見されている。ダイオキシンとは対照的に、PCB は、工業用途に物理的、化学的に望ましいものもあり、工業的に生産されてきた。しかしながら、一部の PCB は、高温において安定で、化学的に抵抗性があり、すぐれた不伝導性を持っており、それが工業用途にふさわしいのみならず、環境での残存性が高いことにもなる。環境中の PCB 汚染源は、PCB 汚染廃棄物または含有廃棄物、汚泥、漏洩、

不法投棄、または焼却炉からの排出、廃棄物が捨てられたゴミ投棄場からの揮散などである⁽⁵⁾。大量の PCB がすでに環境中に存在し、また今なお大量に使用されているため、環境破壊の可能性ははかりしれない。

PCDD/DFs を暴露すると、クロルアクネ、肝臓障害、免疫毒性などの広範囲な生物学的影響を引き起こす。PCDD/DFs のうち最も毒性が高いと考えられているのは、2,3,7,8-テトラクロロ誘導体で、塩素原子が 2, 3 位と 7, 8 位に対称に結合している⁽⁴⁾。PCB が引き起こす毒性の原因のほとんどは、オルト(2)位でなく、隣接しているメタ(3, 5)位とパラ(4)位に塩素原子を持つ、少量の PCB によるものの可能性が最近の研究で明らかになっている。このようなコプラナーまたはノンオルト型の PCB のうち最も量が多いのは、33'44'-T4CB (IV), 33'44'5-P5CB (V), 33'44'55'-H6CB (VI) で、2378-TCDD と類似の毒性を示す。それぞれのグループの個々の物質で非常に異なった毒性を持ち、また、グループ間で生物学的に類似した影響を持つので、これら化合物の毒性と 2378-TCDD (2378-TCDD I-TEQ=1) の毒性とを関連づけ、国際毒性等価ファクター(I-TEQs または I-TEFs) の概念を導入することとなった。

他の多塩素化有機化合物も、PCDDs, PCDFs, PCB と同様、環境中に残存し、生物学的にも環境に対しても有害な影響を及ぼす可能性がある。これらの有害な影響が研究され定量されるにつれ、環境政策もこれらの研究に平行して行われてきた。国内や国境を越えた政策的対応への要求は、微量な環境汚染の定量を可能にするすぐれた感度の良い分析技術を開発するのに貢献した。こういった経緯を含み、今回の京都で11月21日から25日まで

行われた第14回ダイオキシン国際会議（第14回塩素化ダイオキシン、PCB、ならびに関連化合物国際シンポジウム）において、口述ならびにポスター発表が行われた。335の発表は、12の各部門—分析方法、発生源制御、環境レベル、発生源と生成、極地環境、各国の規制の動向、PCBと関連物質の代謝、PCB毒性、ヒト毒性、毒性、生態毒性、油症—に分かれてシンポジウムを構成している。このうち、各国の規制の動向、分析方法、環境レベル、極地環境の4部門についてレビューし、さらに12部門の全発表リストを掲載する⁽⁶⁾。

各国の規制の動向

スウェーデンのアールボルグは、WHO/EURO（世界保健機構ヨーロッパ支部）のPCDDs、PCDFs、PCBsのリスクアセスメントと規制対策に関する活動について発表した⁽⁷⁾。このプログラムの主な目的は、安全管理対策を開発し、汚染の評価、制御、最小化すること、そして健康へのリスクを評価し最小化することである。研究所間での共同研究により得られた二度目の母乳分析の結果によると、PCDD/DFsのレベルはヨーロッパにおいて増加しておらず⁽⁸⁾、結論として、授乳を禁止する必要はないということである。WHO/EUROは、コプラナー型PCBも多くのPCDD/DFsと同様か、もしくはそれ以上に毒性があることを確認している。そのため、これらのダイオキシン類似PCBのための暫時的なI-TEFsが再検討された⁽⁹⁾。一連の研究所間での共同調査で、PCDDs、PCDFs、PCBsの食餌による暴露、ヒトの母乳、血液、また、牛乳、魚中に含まれる濃度、固形廃棄物（MSW）や下水汚泥（MSS）の焼却炉からの排出に関する研究が行われ、その結果が多くの文献として1995年に発表される予定である。

米国環境保護庁では、ダイオキシンは非常に少量（～14kgTEQ/年）しか生産されていないけれども、その毒性の高さから、重要な環境汚染物質として扱わなければならないとしている。ファーランドは、1985年米国環境保護庁が、除草剤の2,3,5-T中のダイオキシンを最初に扱ったと報告している⁽¹⁰⁾。1994年9月米国環境保護庁は、ダイオキシンのヒトへの健康影響、汚染源と暴露レベルについてはじめて完全評価を行いその草案を発表した。この草案は、米国環境保護庁のダイオキシンに対する扱いを明らかにし、ダイオキシンが動物に発ガン性を持ち、ヒトへもその可能性があること、またダイオキシンへの暴露によってガン以外の多くの健康影響もあることを確認した。草案によると、アメリカにおけるダイオキシンの大気への排出の95%までが廃棄物焼却によるものであるとし、また職業的にはダイオキシンに暴露しないヒトの主な汚染源は食物からによるものであることを認めている。また、データの顕著な欠如、特にダイオキ

シン汚染源、ダイオキシン排出、空気中、水中、土壌、ヒトの食物、及び人体組織中のダイオキシンレベルのデータが不足していることも認めている。

キムラは、日本におけるダイオキシン問題に関する経験を述べた⁽¹¹⁾。タツカワによる日本のゴミ（MSW）焼却炉のダイオキシン排出の報告⁽¹²⁾の後、0.1ng/kg体重/日が吸入による悪影響をもたらさないレベルであると報告された^(13,14)。1990年厚生省は、最も適当な技術を用いて、ダイオキシン排出レベルを減少させるガイドラインを発表した⁽¹⁵⁾。新しい排出レベルは、0.5ngTEQ/Nm³で、これにより約90%までに排出レベルを減少させる。紙パルプ産業によって引き起こされるとされているダイオキシン汚染の調査が、環境庁、厚生省、農水省合同で行われた。その結果によると、製紙工場からの廃液と、ボイラーからの排気は、PCDD/DF類を含んでいたが、工場付近の大気汚染と魚類の汚染レベルは、制御地域と同様の値であった⁽¹⁶⁾。環境庁は、これらの排出レベルを削減するガイドラインを発表した。このガイドラインによると、塩素使用の削減、漂白剤の代替物導入による削減、及び製紙工場ボイラーの適切な焼却方法の適用が述べられている。

ヨハンソンは、スウェーデンを地域モデルに、スカンジナビアにおけるPCDD/DFs、PCBとその関連化合物の汚染についてレビューし⁽¹⁷⁾、一方ニューステッドはイギリスにおける排出規制である統合汚染規制（ICP）の実行について概略を述べた⁽¹⁸⁾。しかしながら、オランダとドイツが行った規制は、ヨーロッパにおける最良の例かもしれない。

リエムは、オランダの廃棄物（MSW）と排出に関する数字を提示した⁽¹⁹⁾。現在、廃棄物排出ガイドラインの0.1ng I-TEQ/m³は法律となった。表層水への排出はおよそ3g I-TEQ/年と推定される。オランダにおける成人食物摂取のメディアン値は1pg I-TEQ/kg体重/日である。もしコプラナーPCBがこの数値に含まれるとすると、2.4pg I-TEQ/kg体重/日に増加する。表層土壌のバックグラウンド値は、1～20ng I-TEQ/kg乾燥土壌で、空気中濃度は約24fg I-TEQ/m³、そして大気輸送汚染は8ng I-TEQ/m²/年であった。オランダ・ライン川堆積土壌中の平均値は220 ng I-TEQ/kg乾燥堆積土壌であることが判明したが、より高い値が、かつて知られた汚染源の近くで見つかっている。

パスラーは、ドイツにおける規制対策について述べた⁽²⁰⁾。燃焼施設に関する1990年法令は、排ガス中のダイオキシンの限度濃度を0.1ng I-TEQ/m³に定めており、約4g I-TEQ/年まで排出減少を見込んでいる。1989年以降、有鉛ガソリン中の塩化及び臭化炭化水素浄化剤の禁止、ペンタクロロフェノール（PCP）除草剤とPCBの使用禁止は、ドイツの環境汚染を5kg I-TEQ/年

あまりまで減少させた。下水汚泥に関する1992年法令は、肥料として用いられる下水汚泥のための制限値を儲けた。ダイオキシンに関する合同調査グループの最初の報告は、目標値を $< 5 \text{ ng I-TEQ/kg 乾燥土壌}$ に設定した⁽²¹⁾。値が、 $5 \text{ ng I-TEQ/kg 乾燥土壌}$ であれば、制限なく農業耕作が許されているが、それより高い値が検出されれば、農業の制限が望まれる。同様のガイドラインがミルクや乳製品に対して課せられた。理想的な長期目標は、 $< 0.9 \text{ pg I-TEQ/g milk fat}$ である。3.0pg I-TEQ/g milk fatを超える値は、牛乳、乳製品の一般供給の中止や土地利用の変化を招くであろう。まだ特別な法規制のないPCDD/DFsとPBrDD/DFsの制限値は、今法制化されつつあるところである。

分析法

正確で精度の高い分析がどこでも出来るようになって、公的及び私的研究機関への研究要請がますます高くなってきている。一般にほとんどの分析目的は法的要請に関わらず実際の汚染濃度を調べることである。実際、測定のおほとんどは、今や自動化されてきているが、測定に至る段階—サンプル収集、サンプルの保管、クリーンアップ—においてなおかなりの研究資源と努力が費やされている。化学物質汚染の定期的分析とその評価に際する資源を節約するために、最新の及び改善された技術を開発導入することの必要性が認められる一方、高度な分析精度を維持して法的要請を満足させなければならない。

研究所間のクロスチェック調査及び標準リファレンス物質 (SRMs または CRMs) が、分析方法とその結果を評定するために用いられている。ラッペらは、「フライアッシュ」抽出物中のPCDD/DFs調査で国際間クロスチェックを行い結果を発表した⁽²²⁾。日本から6つの研究所、アメリカ、フィンランド、スウェーデンから各1ヶ所の研究所がこの調査に参加した。すべての参加研究所は、I-TEQs 値において満足する結果を得た (RSD25%)。研究所間の差異は、研究所内のデータのばらつきよりも小さかった。ラッペは、研究所間クロスチェックは異なった研究所によって出される研究結果を評価するのに有用で、ノンオルトPCBの研究においても同様なクロスチェックが繰り返されるべきだと述べている。

デ・ボーアとヴァン・デル・ミーアは、アザラシ類、魚類、そして海洋底質におけるPCBに関する3つの研究組織の調査結果をまとめて発表した⁽²³⁾。18ヶ国75研究機関がこの調査に参加している。主な結論は、この調査において逐次調整が行われ、参加研究機関の研究姿勢をより改善させたこと、PCA (主要コンポーネント分析) が有力な評価ツールであること、長期分析によるばらつきがリファレンス物質によって改善され得ること、今や

大部分の海洋研究所が、海洋底質中のPCB118, 138, 153の同定に合意していること、そして脂肪分の少ない魚類のPCB研究が将来的に望まれていることである。

CRMのマトリックスと分析対象物質の濃度は、分析対象サンプルを正しく代表しているべきであるという考え方は、広く受け入れられている^(24, 25)。モリタは、燃焼過程で生じる全てのダイオキシン同族体を含んでおり、フライアッシュの物質特定と有機物分析トレースに非常に有用な、CRM新シリーズ (NIES CRM No.1-10) を紹介している⁽²⁶⁾。

チャンバースは、NIST SRM No.1945 (鯨脂)、No.1974 (貝組織有機物)、No.1941a (海洋底質中有機物) を紹介した⁽²⁷⁾。SRM1975は26種のPCB同族体と15種の農薬が同定された冷凍鯨脂である。SRM1945は、9種の多核芳香族 (PAHs) が確定されている均質冷凍貝組織、SRM1941aは一定の多塩素化炭化水素 (pCHCs) が確定されている均質海洋底質である。チャンバースは、SRMはPCDD/DFsと同様の物理的・化学的性質を持つ多くの有機汚染物質と同質であり、もし海洋哺乳動物、貝類、海洋底質にPCDDsとPCDFsが有効であれば、多くの海洋生物や底質調査にこれらのSRMが非常に有用であると述べている。

正確で精度の高い微量有機化合物の分析は、しばしば内標の使用が求められる。リュイックは、臭化物及び混合ハロゲンのダイオキシンの分析はしばしば放射性元素でラベルされた標準物質の不足から困難になっていると述べた。彼は、ジベンゾ-p-ダイオキシンの標品を直接臭素化した混合ハロゲン化ダイオキシンの¹³Cラベル物質の合成とモデルフライアッシュシステムにおいて気-固相反応中CuCl₂/HClで多臭素化ジベンゾダイオキシンの部分塩素化について発表した⁽²⁸⁾。収量は、40-135%であった。類似の方法で、マクラクランは商品として入手可能な¹³Cラベル237-T3BrDDから、¹³Cラベルモノ-及びジ-BrDDの、そして2378-T4BrDFから、モノ-、ジ-、トリ-BrDFの光化学合成について述べた⁽²⁹⁾。芳香族炭素と臭素の均等開裂のあと、溶媒から水素原子が抽出される。臭素化物を塩素化物混合炭化水素化合物中、臭素のみが置換される。収量は、50-80%。

サンプル収集、その調整とクリーンアップ、同族体の分離と同定は、環境分析において重要視される。雪や水のようなマトリックスは、その物理的、化学的性質から有機塩素化合物汚染の濃度は極めて低いものとなる。マツムラは、海水中のダイオキシン類のための予備濃縮システムの開発について述べた⁽³⁰⁾。本質的には、大量の海水を急速に交換可能なカートリッジフィルターと樹脂カラムにポンプで送り込むものである。この組み合わせは、完全に溶解したダイオキシンと粒子結合したダイオキシンのどちらもトラップする。その後フィルター又は

カラムから抽出し、得られた溶液を分析して同定する。 fg/l レベルの同定が可能である。

母乳サンプル中のダイオキシン類とPCB類の同定は、極度に低濃度であるため、高感度、高選択能が分析方法に要求されるので困難であろう。ヤマモトらは母乳の迅速クリーンアップ法について述べている⁽³¹⁾。この方法は、サンプルマトリックスの3本連続カラムによる分離法で、けん化後、多相シリカカラムによって脂肪酸を取り除く。このあと、活性炭素シリカカラムで他の有機塩素化合物からダイオキシン類及びコプラナーPCBを分離し、最終的に中性アルミナカラムクロマトグラフィーで分離するものである。

アクガーマン⁽³²⁾とカワモト⁽³³⁾は、従来の液相抽出における不便さを解決するための、超臨界液抽出(SFE)の利点について述べている。利点とは、超臨界液が土壌からの有機汚染物を溶解し、また、操作パラメーターを少し変更するだけで超臨界液の溶解能を変えられることである。二酸化炭素は無害で環境に影響が少なく、不燃性で使用中安全性に問題がないので、望ましい「溶媒」である。

コプラナーPCBの同族体物質を同定するのに重要なのは、大量のPCBからこれらの同族体を分離することである。一般に、GC/MSにはほんの少量のサンプルの分画のみ、最終的に送り込む。しかしながら、オンラインでつながったLC-GCシステムは、分析システムにより多くのサンプルを提供する。グリーンヴァールは、ヒト血漿中PCB126とPCB169の0.5–1 pg/g を同定するのに、オンラインLC-GC/MSを導入した⁽³⁴⁾。ハグルンドは、二次元HPLC(LC-LC)がどのようにGC-ECDと結合しどのようにモノー、ノンオルト-PCBとPCDFの指標同族体を他の妨害物質から分離するかについて述べた⁽³⁵⁾。得られたデータは、従来のGC/HRMSに匹敵すると主張している。

HPLCを用いるとき、調査対象の種の物理的、化学的性質によって用いることのできるカラムの選択が限られることがある。アツマは、魚中のPCBの分離に対し多孔グラファイトHPLCカラムの使用について述べた⁽³⁶⁾。活性炭素カラムと比較して、多孔グラファイトカラムは溶媒の使用を減らし、PCB分離をより速くする。タナカは、電子供与体と電子受容体が結合した固定相カラムを用いて全ての二塩化、三塩化ダイオキシン、及び多数の多塩素化同族体の分離を逆相液体クロマトグラフィーによって行った⁽³⁷⁾。固定相は異性体の同定もおこなえる。研究されたカラムのうち、1-(1-ピレニル)エチルシリル相は、より対称位にある塩素の置換に伴い、PCDDの保持がよく、また、2-(ニトロフェニル)エチルシリルと3-(p-ニトロフェノキシ)プロピルシリル相は、より基部に近い塩素の置換に伴ったPCDDをより好む。ナイ

クワディは、主鎖液晶ポリマー(MCLCP)の固定相の、ガスクロマトグラフィー(GC)キャピラリーカラムへの使用の開発について述べた⁽³⁸⁾。MCLCPは、一般にジメチルシロキサンと液晶とによって交互に構成されている。このカラムの最高許容温度は約300℃である。MCLCPを用いた溶融シリカキャピラリーカラムは、特に2378-置換PCDDに対しすぐれた分離能を示した。OCDFからの完全なOCDD分離もまた可能である。

ダイオキシンに最も適した分析方法は、高分解能GC/MSであるが、全ての研究室や研究者がこのシステムを使用できるわけではないため、別の分析方法が常に要求される。バターソンは、PCDD異性体ペアの分離は、三つのクロマトグラフィー技術(GC, HPLC, ミセル動電クロマトグラフィー)と三つの分光法技術(FT-IR, ^{13}C NMR, ^1H NMR)の組み合わせによって可能であると述べた⁽³⁹⁾。異性体ペアの構造決定は、異性体ペアのパーセンテージに応じてすぐれた一致を示した。個々に決定された構造がよく一致することは、異性体同定の信頼性を実質的に増すことになる。

チンマーマンは、PCDFに関するはじめての共鳴レーザーマッススペクトルの結果を発表した⁽⁴⁰⁾。この技術はレーザーによる共鳴マルチフォトンの分子化と飛行時間型マッススペクトルとを組み合わせたものである。二次元分析法としてこの技術は高選択性と高感度を示し、多くのオンライン及びインプロセスアプリケーションを可能にし得る。適した分子UV分光法と分子化能データの知識が必要である。このデータは、分子軌道計算によって求めることが可能である。チンマーマンは、塩素化ベンゼン類、ジベンゾ-p-ダイオキシン、2378-TCDD、OCDDの分子化能を、計算された七つの半経験的量子力学的法(MIND3, MNDO, AM1, PM3, INDO, CNDO, ZINDO/1)と比較した⁽⁴¹⁾。どの方法も有意義であろう。しかし、もし実験データが少なくとも一つのクラス中一つの化合物に対し入手可能であれば、修正係数が導かれる。実験データは、AM1とPM3プログラムによって最適化された。

ハリソンは、競合酵素免疫アッセイ(EIA)を用いた2378-TCDDの迅速スクリーニングシステムについて述べた⁽⁴²⁾。このシステムは、2378-TCDDと関連同族体に特異的である。サンプル調整は自動化でき、土壌、血漿、水等、多くのマトリックスの免疫アッセイスクリーニングが可能である。濃度条件や最終状態は、自由度が高い。この分析は、抗体でコートされたセルに、用意したサンプル抽出物を加えることによって行われる。競争共役体(抗体のふさがっていない側と特異的に結合することのできる)とインキュベーション後、酵素基質が色の展開のために加えられる。色が濃いほど、ダイオキシンの量は少ない。天然抽出の直接分析によって土壌中の

2378-TCDD 10ppb の検出が可能である。このシステムは、スピード、サンプル量、コストの点で GC/MS と比べて格段の進歩がある。

環境レベル

このセクションの基調講演は、ラッペとクジェラーによってスウェーデンにおける環境中 PCDDs と PCDFs の歴史的経緯と蓄積量(推定)計算について述べられた⁽⁸¹⁾。主だった結論は、自然界での PCDD/DFs の形成はおり得るが、しかし、歴史的サンプル中に発見された PCDD/Fs の元々の発生源は自然燃焼であること、19世紀の汚染は、ほとんどが汚染食物とそれに伴いヒトの暴露として帰着すること、1940年以降のサンプルは当時使用された一般用ペンタクロロフェノール中に発見された HpCDD と OCDD に影響を受けていること、燃焼は、低塩素化 PCDD/DFs の第一次汚染源であること、そして環境中レベルは1960年代後半にピークとなり現在は減少しているが、まだ歴史的レベルにまで到達していないことである。

スウェーデンにおけるダイオキシン調査の結果は、デ・ウィットによって発表された^(82,83)。スウェーデン環境保護局は、1988-93年の5年間にわたってスウェーデンの環境中の PCDD/DFs のレベルを同定する調査を行った。調査は地理的分布、生体濃縮、生体拡散の時間的流れの他、生物種、年齢、大きさ、性別と体内濃度との関係についてなされた。相当数の生物種において分析が行われたーニシン、カワマス、カワミンタイ、マスの一種、北極イワナ、サケ、マス、ヒラメ、サバ、タラ、カニ、ロブスター、貝類、ウミガラス、オジロワシ、ミサゴ、オオカミ、キツネ、アメリカヘラジカ、トナカイ、アナグマ、カワウソ、ウサギ、アザラシ類4種、イルカなど。サンプル収集が行われたのは、汚染地域、拡大汚染地域、工業化の少ない地域、そして遠隔地である。一般的に PCDD/DFs の海洋生態系における濃度は、地上動物よりずっと高い。バルティック海の生物体は、最も高い濃度を示す。アザラシ類は、低濃度の PCDD/DFs 値を示し、イルカ類は、これらの物質を生体濃縮しないことを示した。しかしながら、魚を餌とする鳥、ウミガラス、ウミワシ、ミサゴは、餌となる魚そのものよりずっと高い値を示し、これらの鳥は、PCDD/DFs とコプラナー PCB の生体濃縮をすることを示しているようである。最も高濃度を示したのは、南スウェーデンの成長ミサゴであった。1969年から1992年の間に収集された、ウミガラスの卵とカワマス中の濃度は、この期間にゆっくり減少し、PCB、ペンタクロロフェノール、2,4,5-T の使用を禁止したことにより、PCDD/DFs と PCB の環境への移行を減少させたことを示している。

今回のダイオキシン学会で特に目立った話題は、サン

ブルとサンプリングの場所ととの間の PCDD /PCDF/PCB 同族体プロファイルと比較するための統計分析手法の使用である。地理的に隔離された地域の野生生物母体は、異なった環境汚染パターンを示す。統計学的分析は、さもなくば大量のデータで覆い隠されがちな、類似性や相違性を明らかにするのに役立つ。用いられた統計手法の中で、PCA は最も人気であった。

ヤマモトは、PCA を日本の都市部及び遠隔地の水圏底質中にみられる PCDD/DFs の相似性と相違性の特徴づけに用いている⁽⁸⁰⁾。日本の水圏底質中レベルは、他の先進工業国において発表されている範囲内のレベルであった。東京湾と大阪湾の濃度は主に燃焼過程で生じる物質を含み湾岸の巨大密集工業地域からの影響を示している。これら都市部の底質は、遠隔地より高い値を示した。湖沼底質の PCDD/DFs は、主に、関連のある陸水学的流域中の、以前使用された PCP に由来し、一方北日本の河川底質は、農業由来の1368及び1379-TCDD ダイオキシンによる汚染が特徴づけられる。

フィードラーとラッペは、パルプ工場から排出された PCDD/DFs の汚染のおそれがある南ミシシッピの河川系のデータの評価を2つのパートにそれぞれ発表した^(84,85)。クラスター分析、PCA、多変量統計解析(MDA)を用い、底質と土壌のサンプルをパルプ工場(製紙工場)の上流及び下流からサンプルを集め、最近洪水をおこした地域からの土壌データ、歴史的にダイオキシン汚染が知られている堆積地、下水汚泥、工場からの直接廃液と比較した。どの統計手法も、パルプ工場が河川排水流域中にみつかった PCDD/DFs の汚染源であることを示さなかった。

ファン・バベルは、ボスニア湾内の4カ所から収集された以下のサンプルー底質、端脚類動物、等脚類動物、動物プランクトン、植物プランクトン、アミ類、スズキ、ハゼ類、ニシン、サケ、マス類のサンプル中の PCB レベルの計量化学的比較を行ったとしている⁽¹⁴⁰⁾。結果、底質と端脚類動物中の PCB レベルはボスニア湾の場所によって異なり、最も高濃度を示したのは南側で、また PCB レベルは季節によって大きく変動し、さらに PCA の統計手法によると、PCB 同族体パターンは、亜北極サンプリングサイトと他の三カ所のより南よりの地域との間に有意な差異が認められることを明らかにした。

1988年、北海のヨーロッパアザラシの病気が流行した。ストールハンセンは、地理的に離れた3カ所から集められたアザラシの死体からみつかった PCB 同族体パターンを、PCA 統計分析と一次判別式分析とで比較した⁽⁸⁶⁾。同族体パターンは、それぞれの動物に特徴的であるが、地域によってもその傾向がみられる。これらの結果の背後にある根元的な過程をよりよく理解するには、メスアザラシの一群を捕獲し、二つの異なった場所からの魚ー

ワデン海のヒラメと大西洋のさばーを餌として与えた。アザラシと魚中に見つかったPCB同族体パターンにも明らかな違いがあった。

長期にわたり憂慮すべき問題は、食物連鎖—特に母乳、牛乳、酪農製品のような高脂肪食を通してヒトに暴露するPCB、PCDD、PCDFである。ファン・クルーヴェンベルゲンらは、ベルギー・フランダース地方の母乳中ダイオキシン濃度レベルのデータを発表した⁽⁸⁷⁾。平均TEQ (fat basis) は、約35pg/gであった。このダイオキシン量は、他の西ヨーロッパの国々の報告値とよく一致する。PCDDとPCDFは、TEQ値がほとんど同等で、また、都市部でより高い値を示す傾向が見られた。ヨーロッパの、世界的にも最も高い値を示していたダイオキシン濃度は、ゆっくり減少しているといういくつかの示唆も見られた。

環境中のダイオキシン汚染濃度が、産業化と集約的農業の出現に伴って増加したことは、一般的に認められている^(4,5)。バルダサリは北イタリアのヴァルダシナ地域からのミルクとチーズのサンプルに関するPCB汚染のデータを発表した⁽⁸⁸⁾。この地域では、夏の間、畜牛は高地の牧草地で自由に草を食べており、また冬の間は家畜飼料を与えられている。全てのミルクとチーズのサンプルは同様のPCB同族体パターンを示した。PCB濃度レベルはミルクもチーズもどちらのサンプルもイタリアの他の地域と比べて一貫してより高い値を示し、興味深いことに、低濃度汚染と考えられている地域の餌を与えられているにも関わらず、冬よりも夏の値の方がより高い。ヴァルダシナ農地付近に存在するPCB汚染源は、この変化のため特定された。

地域内に明確な汚染源が見られないにも関わらず汚染している遠隔地域もある。長距離大気間輸送は、提案された一つのメカニズムで、底質コアサンプリングは、この理論を確立するのに用いられている。クジェラーとラッペは、バルティック海の底質コアのデータを発表した⁽⁸⁹⁾。この底質は薄層の蓄積型で30年ごとに各層を成している。PCDD/DFsは、全ての層から検出された。19世紀後半の一番深くて古い層は、全てのPCDD/DFs同族体を含んでいた。約1880年から約1960年の間の堆積層の汚染レベルは、非常に低レベルを示し、この期間にわたってほとんど増加を示しておらず、汚染源と考えられるのが燃焼のみであることを示唆している。この後の時代では汚染レベルが急激に増加し、PCDD/DFs含有生成物の一般的使用が増加したことを示す。

別の歴史的経緯のデータを知るため、ヒトによって侵害されていない地域の底質を用いる必要もあり、大海の海底は、それに適しているであろう。蓄積量は少ないが、本来、潮や波による影響が少ない。マツムラとカトウは、大洋海底から海拔4030m地点のサンプルを24cm回収し

⁽⁹⁰⁾、底質コアは薄片にカットされた。このサンプルの表面から、tetra, penta, hexa, hepta, octa-PCDD同族体が、1.6, 5, 19, 39, 42, PCDF同族体がそれぞれ、14, 7, 9, 33, 20pg/g検出された。

オオサキは、福岡県の河川、海岸底質中のPCDDs, PCDFs, PCBsのレベルと同族体パターンの、濃度と地域差について調査を行った⁽⁹¹⁾。底質の同族体プロフィールは、その付近の土壌とは異なり、河川と沿岸の底質中のPCDDs, PCDFs, PCBsは、農業による汚染源に起因する可能性があり、またこれらの化学物質のレベルは河川のサンプリングの場所によって、河川下流>海岸>河川上流のように異なっている。

PCDDs, PCDFs, PCBs汚染の回復は、非常に高価で極端な処置が必要な場合も多い。生物的分解は、汚染の回復に、より環境に優しい方法を提供するであろう。タケダは、PCDD/DFsのリグニン真菌*Phanerochaete sordida* YK-624による分解について報告した⁽⁹²⁾。2378-TCDDと五塩化以上の同族体は14日間で十分分解された。分解率は、グルコースと酸素の付加で加速化された。

極地汚染

何年にもわたって大量の塩素化炭化水素 (CHC) 化合物が環境中に放出され、多塩化カンフェン (PCCs)、ヘキサクロシクロヘキサン (HCHs)、ヘキサクロベンゼン (HCB)、DDT、PCB、PCDD/PCDFsは、空気、雪、海水、そして北極の生態系生物相にまで発見されるようになってきた⁽¹³⁰⁻⁴⁾。CHCは化学的にも生物学的にも環境中分解しにくく、温和な気候中ですら長い環境中半減期を持っている。より厳しい気候の極地地域においてその分解プロセスが減速することは想像できよう。

ノールストロームは、これらの化学物質が十分に高い蒸気圧とヘンリー係数を持ち、大気と地面、水面との間の循環を介して、地球規模の対流圏中に広がるができることを示唆した⁽¹³⁵⁾。CHCの大気中残存期間のは、数カ月であるため、北極はCHCにとって暖かいところから冷たい環境へ地球規模に蒸留させる「コールド・フィンガー・トラップ」の巣窟のようなものであろう。北極にCHCが検出されたが、HCHとHCB (0.58, 0.19ng/m³) はDDTとともに空气中に優位を占め、クロルデンとPCBはそれよりワンランク低レベルである。HCHは、雪や海表面に最も高濃度 (1-2, 4-5 ng/L, 参考: ベーリング海 2-3 ng/L, 太平洋中部沖 < 2 ng/L) の化合物である。他のCHC化合物より比較的重要なDDTとPCBは、食物連鎖を通して水から動物プランクトン、端脚類動物、タラ、シロチョウザメに至って増加する。これらの化合物の生物濃縮は、107から109と推定される。アザラシ類はHCH, HCB, PCBとも生物

濃縮しないが、北極グマは主にPCBとオキシクロルデンを濃縮する。全クロルデン濃度の対数をとると、全PCBとCB-153は食物連鎖のレベルと直線的な相関関係がみられた。クロルデンとPCB類のような影響を受けやすい化合物は、北極グマ、アザラシ類、セイウチ、鳥類に代謝され、低生物濃縮要因となる。

オーメは、ウエストアイス地域の東グリーンランドで捕獲したハーブアザラシ (*Phoca groenlandica*) の脳と脂肪中のPCDDs, PCDFs, PCBsと農薬のレベルについて同定を行った⁽¹³⁶⁾。同族体特定の分析がおこなわれ、PCDD/DFs換算2378 I-TEQ 3.9-10.4pg/gで、2378-TCDD, 123678-Hx-CDD, OCDD, 2378-TCDFはそれぞれ0.5-1.2, 1.4-9.4, 1.3-6.6, 3.6-10.4pg/gであった。ノンオルトPCBのI-TEQ値はPCDD/DFsのI-TEQ値より4-10倍高く16-96pg/gで、33'44'-TeCB, 33'44'5'-PeCCB, 33'44'55'-HxCBはそれぞれ91-410, 130-472, 24-942pg/gであった。DDTとDDT代謝物のレベルは、一桁低いことがわかった。また、オーメは、南極、北極両地域の空气中、生物中のCHCについてその相似性、相違性、比較濃度パターンについても述べた⁽¹³⁷⁾。南極においては、かなり低濃度であることが判明し、南半球が産業的にも集約農業的にも遅れていることが貢献しており、さらに気象学的及び大洋海流の状況が長距離輸送を難しくしていることと相まっている。北極と比較すると、HCHとHBHは主な異性体 γ -HCHとともに、南極大気において約5倍低い。多塩素化PCBのレベルと同族体パターンは、両地域とも匹敵するが、ダイオキシンとジベンゾフランは、南極での生物種の多さ

とともに公にはデータ不足で比較が困難であった。南極での同族体パターンは、北極のそれとは異なっており、おそらく両半球での排気パターン、食慣習、生物種の違いを示唆するものであろう。南極の2378-TCDD I-TEQは2pg/gであった。

ポリシュックは、カナダ西ハドソン湾地域に生息するメス北極グマ (*Ursus maritimus*) について、そのPCB濃度と身体負荷、体脂肪パーセントとの関係を研究している⁽¹³⁸⁾。これらの北極グマは、冬の断食中に90%まで体重を落とすことが可能である。1992-1994年にかけて、断食時期の始めと終わりに、多数のクマから臀部脂肪組織のサンプルを収集した。サンプルは、PCB99, 153, 138/163, 180, 170が、PCB同族体全体の98%を占めた。動物個体の全脂肪量 (kg fat) か、もしくは体重あたりの脂肪パーセンテージ (% fat) と、PCB体負荷との間には、相関関係は見られなかった。しかしながら、個々のクマごとに経緯を追うと、メス北極グマは妊娠中と授乳の初期にはPCBの体負荷が減少し、新生児とコグマの健康状態への悪影響が増加することが示唆された。

パーヴィルタは、ノース・ケープのテノ川地域とバルティック海のシモ川地域に生息する北極タラとサケにおけるPCC, γ -HCH, α -HCH, HCBz, DDT, PCBのレベルを測定した⁽¹³⁹⁾。最も汚染が激しかったのは、塩素化炭化水素類だった。これらの中で、DDTのみがこの10年で明らかに直線的に減少している。北極魚のうち最もダイオキシンタイプ物質による汚染は、PCBに起因するものであった。

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