GREENPEACE

A RECIPE FOR DISASTER

A review of Persistent

Organic Pollutants in Food

DOWN TO ZERO

GREENPEACE

RECIPE FOR DISASTER

A review of persistent organic pollutants in food

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SUMMARY
Persistent organic pollutants (POPs) are a group of chemicals which are very resistant to natural breakdown processes and are therefore extremely stable and long-lived. POPs are not only persistent in the environment but many are also highly toxic and build up (bioaccumulate) in the tissues of animals and humans. Most do not occur in nature but are synthetic chemicals released as a result of anthropogenic activities. Vast amounts of POPs have been released into the environment and due to long-distance transport on air currents, POPs have become widespread pollutants and now represent a global contamination problem. Certain POPs have been responsible for some catastrophic effects in wildlife, ranging from interference with sexual characteristics to dramatic population losses. POPs are suspected of causing a broad range of adverse health impacts in humans and there is evidence that current levels of POPs in women in the general population of some countries is sufficient to cause subtle undesirable effects in their babies due to transfer of these contaminants across the placenta and via breast milk.

There are numerous POPs which pollute the environment. Some of those which have given rise for particular concern are persistent organochlorines. A meeting of the UNEP's governing Council in 1995 identified a list of 12 POPs as substances of clear concern in accordance with the precautionary principle. These chemicals are all organochlorines. They include dioxins and furans (PCDD/Fs), which are produced as unwanted by-products of several industrial processes including incineration and PCBs and HCB, which have several uses and are also formed as unwanted by-products. DDT, chlordane, heptachlor, aldrin, dieldrin, endrin, toxaphene and mirex, which are pesticides complete the list.

The major route of exposure to POPs in humans is through consumption of food. Most of the 12 POPs listed by UNEP are widespread pollutants and are found in foodstuffs from all over the world. Since POPs are soluble in fats (lipophilic) the highest levels are usually found in fatty foods such as meat, fish and dairy products, but POPs can also be found in vegetables, fruit and cereals. In addition to the general contamination of foods by POPs, there have also been accidents in which foodstuffs have become highly contaminated.

Greenpeace has issued this report to highlight the contamination of the human food chain by POPs. The report sources material from the published scientific literature. It identifies many instances where regulatory limits for foods are exceeded throughout the world and discusses accidents and industrial activities that have led to high level food contamination.

The Human Food Chain
Environmental pollution has led to the contamination of human food with POPs. Discharges and deposition of POPs to the aquatic environment ultimately results in fish becoming contaminated with POPs. Similarly, atmospheric deposition of POPs on soil and plants leads to their contamination and subsequent consumption by, for instance, cows, results in contamination of milk and meat. In addition, the direct application of organochlorine pesticides leads to residues on crops. Contamination of the human food chain has also occurred when contaminated wastes have been mixed with livestock feedstuffs or directly with food intended for human consumption.

Regulatory Limits for POPs in Food

In an attempt to protect public health, regulatory agencies have set permissible levels in specific foods, Maximum Residue Limits (MRLs), which should not be exceeded. For levels of organochlorines in diet, the Food and Agricultural Organisation (FAO) and World Health Organisation (WHO) have set levels which are deemed to be "safe", called Tolerable Daily Intakes (TDIs) or Acceptable Daily Intakes (ADI). These regulatory limits are set using toxicity data from studies in laboratory animals and sometimes also data from human studies. However, the process of risk assessment which is used in deriving the ADIs/TDIs, is a process that is fraught with uncertainties. Furthermore, endpoints of toxicity testing in animals may be inappropriate for detecting certain health effects, potentially leaving health effects undetected. The process of risk assessment assumes that there is a threshold dose of a chemical below which there are no health impacts and therefore "acceptable" levels of exposure are set. However, for some POPs there may be no threshold doses and usually people are exposed to mixtures of chemicals. Therefore it is unlikely that ADIs and TDIs set for organochlorines in diet are truly protective of human health.

General Trends

A review of the scientific literature revealed that in general, levels of PCBs and dioxins in food of industrialised countries are higher than in less industrialised countries due to greater past production, use and discharge of these chemicals together with present emissions. There are also far more data available documenting levels and dietary intakes of persistent organochlorines from industrialised countries. However, available data for less industrialised countries show that there are numerous instances in which levels of pesticides in food exceed permissible limits, such as levels of DDT in several foodstuffs in India and Mexico.

Highly Exposed Populations

Some populations are exposed to particularly high levels of POPs in their diet. Indigenous Peoples of the Arctic who consume a traditional diet containing sea mammals have a high intake of some organochlorines. This is because sea mammals accumulate high levels of organochlorines and the long-distance transport of POPs towards polar regions has caused contamination of the Arctic. Populations who have a high fish consumption from contaminated waters, such as the Great Lakes or parts of the Baltic Sea, have a high intake of some organochlorines. Children can have a higher intake of organochlorines than adults because of their comparatively high food intake. Moreover, the nursing infant has a particularly high intake of organochlorines because POPs are present as contaminants in breast milk. Exposure of the developing young is of great concern because the developing stages of life are most vulnerable to the toxic effects of POPs.

Dietary Intake Studies

A number of government and other scientific studies have estimated the daily intake of organochlorine contaminants resulting from diet in various countries. For intake studies of dioxins and furans and dioxin-like PCBs, data is limited to a few industrialised countries only. For organochlorine pesticides, studies are also limited in number.

Dioxins and furans and dioxin-like PCBs: WHO have set a TDI for dioxins and furans and dioxin-like PCBs of 1-4 pg TEQ kg^{-1} day^{-1}. WHO experts acknowledge that subtle effects on health from the current intake of dioxins and furans and dioxin-like PCBs might already be occurring in the general population of industrialised countries and efforts should be made to ensure that intakes are in the lower end of the TDI range. Studies show that

dietary intake in some countries is within the TDI set by WHO. However, it is of great concern that the TDI is exceeded in Spain and is exceeded by children classed as “high level consumers” in the UK. Furthermore, intake of foods containing particularly high levels of dioxins and furans and PCBs, such as seafood, could have a large impact on the TDI in all countries. It is most alarming that consumption of some fish oils as dietary supplements alone could also cause exceedance of the WHO TDI.

Children have different dietary habits and a lower body weight than adults which may cause them to have higher intakes of dioxins and furans and dioxin-like PCBs than adults. This is of concern regarding health impacts because it is a crucial time when their physical and mental capabilities are developing. Moreover, due to high levels of dioxins and furans and dioxin-like PCBs in human milk - which stems from lifetime exposure, mainly through food, - breast-fed infants have an intake which is 1-2 orders of magnitude higher than that of adults. Estimates from the Netherlands and the US clearly show that the TDI is greatly exceeded by nursing infants. Given that the infant is especially vulnerable to toxic insult from chemicals because it is a time when the body is developing, it is particularly worrying that infants have such a high intake. The application of a TDI or ADI to the breast fed infant is questionable because TDIs are designed to prevent adverse health effects over a whole lifetime exposure in a 70 kg adult. Nevertheless, given the vulnerability of the infant, it has been proposed that TDIs should be lower for infants, and therefore, current TDIs should not be exceeded by breast-fed infants. Presently, this is clearly not the case.

Total PCBs: For total PCBs, a TDI recommended by International Agencies was exceeded by the Italian diet. People of the Faroe Islands who have an average consumption of pilot whale blubber would also exceed the TDI for PCBs. This is because marine mammals accumulate high levels of organochlorine contaminants. Indigenous Peoples of the Arctic, such as Inuit from Broughton Island, Canada, who consume a traditional diet that includes sea mammals exceed the TDI for PCBs.

Organochlorine Pesticides: It is of concern that intakes of DDT are very high in India, China and Vietnam in comparison with industrialised countries in the Northern Hemisphere even though the FAO/WHO ADI was not exceeded. Moreover, the ADI was exceeded for intake of aldrin and dieldrin in India, mainly due to high levels present in dairy products. An estimated average consumption of pilot whale blubber by Faroe Islanders also caused in an intake of dieldrin which is in exceedance of the ADI. The consumption of a traditional seafood diet by Arctic Inuit was found to result in an intake of toxaphene that exceeded the ADI.

Levels of Organochlorines in Specific Foods

Organochlorines have been detected in foodstuffs worldwide. Many studies have been conducted to assess the concentration of persistent organochlorines in various food types, but data can nevertheless be considered to be limited. Most studies have been conducted in industrialised nations and data is generally more limited for less industrialised countries and is particularly sparse for Africa. Permitted levels of organochlorines in the foods (maximum residue limits, MRLs) have been set by some national authorities and by FAO/WHO for various food types, but there are no FAO/WHO MRLs for fish. There are many cases of exceedances of MRLs:

Fish: For marine fish, consumption of fatty fish from the Baltic around the south coast of Sweden resulted in significantly higher levels of dioxins and furans in the blood of consumers. Input of dioxins and furans to the marine environment from industry caused particularly high level contamination of crabs in a Norwegian fjord, in Newark Bay and New York Bight in the US, and in fish from Tokyo Bay, Japan.

For freshwater fish, studies on various rivers showed that in Spain, heptachlor epoxide exceeded the WHO/FAO MRL and in Australia, PCBs and chlordane exceeded WHO/FAO MRLs. Fish taken from the St. Lawrence River and rivers in British Colombia, Canada, had higher concentrations of dioxins and furans and dioxin-like PCBs than proposed limits due to discharges from industry. In south Taiwan, the WHO TDI for dioxins was exceeded for fish taken from a river and culture ponds as a result of industrial activities in the area.

Meat: High levels of DDT were found in meat from Thailand and the FAO/WHO MRL was exceeded for meat in Vietnam and in Mexico. For dioxins and furans, veal is the most highly contaminated meat due to a high proportion of milk in the calf diet. Second to this is beef.

Dairy: In India, a high proportion of milk samples tested were highly contaminated with DDT and HCH and some exceeded national limits. High levels of DDT were apparent in milk in Hong Kong and Argentina. Residues of aldrin and dieldrin were also reported to be high in milk from these countries and levels of heptachlor and heptachlor epoxide exceeded FAO/WHO MRLs in Hong Kong, Argentina and Mexico.

Fruit, Vegetables and Cereals: The contribution of vegetables and cereals to intakes of dioxins and furans in the diet of some countries, such as the USA, has been reported to be very small with greater than 90% of the intake coming from meat, fish and dairy products. However, a study on the Mediterranean diet in Spain reported that cereals, pulses, fruits and vegetables can have a significant contribution (45%) to PCDD/F intake. For total PCBs, intake in many countries is predominantly from meat and fish but in less industrialised, tropical countries such as Vietnam, cereals and vegetables can be the main source.

DDT residues have been found in tea and coffee. A 1994 study reported that levels in vegetables from Australia suggested recent application on crops despite the existing ban on use. Wheat stored in gunny sacks in India was found to be contaminated with DDT and HCH at levels greater than the FAO/WHO MRL. This was due to spraying of residential areas for vector control.

Time Trends

A general decline in the levels of some organochlorines in foods and in human milk has been reported in recent years. For instance, organochlorines in fish from the Great Lakes are still high although levels have generally declined in recent years. DDT levels in meat have been reported to have fallen in Australia and Canada. There have been some regional declines in PCBs in marine fish but a global decline is not expected in the next few years. DDT levels have generally decreased in marine fish but levels in areas of the tropics indicate continued input to the marine environment. In human milk, concentrations of dioxins and furans have not increased in Western countries in recent years and levels in some European countries have declined. A decreasing trend has been observed for DDT and HCB in

Europe where DDT is banned, but not in Mexico where it is still used. Levels of PCBs and chlordane in human milk do not appear to have declined in countries where levels have been monitored. These examples show that although some compounds have declined, others remain stable due to persistence and/or continued input into the environment.

Food Contamination Incidents

There have been a number of food contamination incidents involving dioxins and furans and PCBs. Some have occurred from local sources of pollution, such as incinerators, and others have occurred after waste or toxic products have been mixed with food for human or animal consumption.

Local Source Contamination

Incinerators of all types emit dioxins, furans and PCBs. There have been several instances in Europe during the 1990s in which emissions from municipal waste or hazardous waste incinerators have resulted in depositions on nearby grazing land for cows leading to contamination of milk. Incidences in the Netherlands, Austria and UK have been recorded in which levels of dioxins and furans in milk exceeded regulatory limits and resulted in bans on the sale of milk.

Reclamation of copper from cables causes the release of dioxins, furans and PCBs from burning of the PVC plastic coatings on the cables. In south Taiwan, fish from aquaculture ponds were found to be highly contaminated due to the burning of electrical cables and credit cards nearby. Fish from the area is an important local source of food and it was found to greatly exceed the WHO TDI for dioxins and furans and dioxin-like PCBs. Two accidental PVC fires in Germany in 1992 and 1996 also caused localised contamination with dioxins and furans. Subsequently, a number of bans on the sale of vegetables and animal products had to be implemented.

Accidental Contamination of the Human Food Chain

Various incidents have occurred in which cooking oil for human consumption has been contaminated by PCB oil. The “Yusho” incident occurred in Japan in 1968 and the “Yu-Cheng” incident occurred in Taiwan in 1979, both affecting around 2000 people. Increased mortality rates were recorded following the incidents and a broad spectrum of health effects were reported. Children exposed *in utero* were severely affected.

More recently there have been two incidents in which animal feeds have become contaminated. In Belgium, in early1999, PCBs oils got into animal feedstuffs. This resulted in chickens becoming highly contaminated, for example, at levels that were 10 to 5000 times levels in milk standards. Not all of the chickens were for human consumption as some were for breeding. Eggs also became contaminated and restrictions on the sale of chickens and other meats were implemented due to possible contamination. The incident had a major economic impact on the Belgian food industry with world-wide exports of food being affected. It is of concern that there was a 4-month delay between the first illness symptoms occurring in the chickens and restrictions being placed on foods resulting in unnecessary human exposure.

Use of contaminated citrus pulp pellets from Brazil in animal feed caused contamination of milk with dioxins in Germany towards the end of 1997 and beginning of 1998. Some milk had levels greater than nationally permitted. The citrus pulp was also used as animal feed in

a total of 12 EU countries. They may have caused elevated levels in milk in countries other than Germany, although data are not available. A total of 92,000 tons of citrus pulp pellets had to be disposed of. The source of the contamination in the feed was found to arise from dioxin contaminated waste lime produced as a by-product by Solvay in Brazil. The waste lime is converted into a form which is then added to citrus pulp for animal feed. Subsequent EC legislation has set a maximum limit for dioxins in citrus pulp pellets to protect European food sources from any possible further contamination. However, there are no limits for dioxins in other types of animal foodstuffs.

Conclusions

- The present review of published scientific literature on POPs in food reveals that there are large gaps in the data. Data is particularly sparse on dietary surveys of dioxins, furans and dioxin-like PCBs in less industrialised countries.

- There are many exceedances of regulatory limits (TDIs and MRLs) for POPs in food both in industrialised and less industrialised countries. This is of obvious concern and especially so given that high levels of POPs in food are likely to continue for some years. Furthermore, it is highly questionable whether TDIs are truly protective of human health. The only way that absolute protection can be guaranteed is by zero contamination of foods.

- Accidental contamination of foods, for example the Belgian chicken scandal and the Brazilian lime waste incident, can have impacts on food supplies on a global scale. The only way to discover such incidents is by continual monitoring. Presently, however, there are many gaps in monitoring and by the time contamination is detected, human exposure has usually already occurred. The only way this problem can be addressed is by prevention of contamination at source.

- Because the release of POPs into the environment is continuing, there is a potential for further severe impacts on the health of wildlife and humans. Of particular concern are effects on the developing stages of life, the unborn and nursing young. Furthermore, in addition to POPs selected by UNEP, there are many other POPs and hazardous chemicals which are a toxic to health. Given these problems, the only way to safeguard the health of future generations is to phase out the production, emissions and use of ALL POPs and OTHER HAZARDOUS SUBSTANCES and implement clean production technologies as already agreed in some regions. As a matter of global urgency, action must be taken to stop production, eliminate all discharges, emissions and losses of those chemicals prioritised for action by UNEP with a view to eliminate all POPs and prevent the marketing and commercialisation of new ones.

Table 1: Reported Instances in Which Regulatory Limits for POPs in Food are Exceeded

Country/Food	Regulatory limit that is exceeded
Total Diet Survey Spain UK (children) South Sweden (fish eaters) South Taiwan (consumption of freshwater fish)	WHO TDI for dioxins, furans and dioxin-like PCBs
Total Diet Survey Italy Faroe Islands (whale meat consumption) Arctic Inuit, Broughton Island, Canada	TDI for PCBs
Total Diet Survey India Faroe Islands (whale meat consumption) Arctic Inuit, Broughton Island, Canada	 FAO/WHO ADI for aldrin and dieldrin FAO/WHO ADI for toxaphene
Freshwater Fish Spain Australia	 FAO/WHO MRL for heptachlor and heptachlor epoxide FAO/WHO MRL for PCBs and chlordane
Meat Vietnam Mexico	FAO/WHO MRL for DDT
Dairy Hong Kong Argentina Mexico	 FAO/WHO MRL for heptachlor and heptachlor epoxide

1. INTRODUCTION

The building blocks of living organisms are organic compounds – that is chemical compounds that contain carbon and hydrogen (and in some cases other elements as well). These compounds are never indestructible and many break down relatively easily. On the other hand, man has learnt to manufacture organic compounds which are extremely difficult to break down. These chemicals are termed persistent organic pollutants (POPs).

A large number of hazardous chemicals have been, and continue to be, manufactured by the chemical industry both intentionally, as products, and unintentionally, as by-products and wastes. These hazardous substances include numerous POPs. Some of these POPs, notably the dioxins and furans, are also generated unintentionally as by-products of combustion processes.

The production and use of POPs, and generation of POPs as unintentional by-products, has led inevitably to the pollution of the environment with these substances. Because they are not easily degraded by natural processes, many persist in the environment for years. Therefore, even if release of all POPs ceased today, they would continue to pollute the environment for many years to come. Numerous POPs have become very widespread contaminants in the environment because they can be transported for thousands of kilometres on air currents, and in rivers and oceans. As a result of this long-distance transport, some POPs even contaminate remote regions such as the deep oceans, high mountain areas and even the Arctic. Indeed, they may be considered as global pollutants.

In addition to being persistent, many POPs are, by their chemical nature, highly soluble in fats (lipophilic). Consequently they have a tendency to concentrate in the fatty body tissues of living organisms and, over time, can build up (bioaccumulate) to high levels in such tissues. In some cases the levels increase (biomagnify) as one animal consumes another in the food chain so that the highest levels are present in top predator species.

Many POPs are toxic and their long-lives in living tissues may lead to adverse effects on health. Although over time POPs may be metabolised (transformed or broken down) in the body to other compounds (metabolites), some of the metabolites produced are more toxic and persistent than the original chemical. For example, the pesticides heptachlor and chlordane are respectively broken down to heptachlor epoxide and oxychlordane which are more toxic than the original chemicals.

Man-made chemicals occur in the environment and in our bodies not as single entities but as complex mixtures. We are exposed, therefore, not to individual hazardous chemicals, but to many; not to individual POPs, but to diverse mixtures. The significance of such multiple exposure remains poorly understood. Moreover, a substantial proportion of the chemicals which occur in the environment and to which we may be exposed simply cannot be identified. This further complicates the problem.

1.1 The Chemicals of Concern

POPs include numerous different hazardous chemicals. A prominent and diverse group of POPs are the organohalogens, i.e. organic compounds of fluorine, chlorine, bromine and iodine. Of the halogens, chlorine has been particularly widely used by the chemical industry,

in order to manufacture organochlorine chemicals for use as pesticides, industrial chemicals, solvents, cleaning agents and plastics, particularly PVC. Indeed, PVC is the largest single use of chlorine.

All of the 12 POPs so far prioritised for action to reduce or prevent emissions under the United Nations Environment Programme (UNEP) Draft POPs Convention are organochlorine chemicals (UNEP 1995). These chemicals are described in Box 1.1

Box 1.1 POPs listed by UNEP

- Dioxins and furans: Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are commonly referred to dioxins and furans or collectively as "dioxins." There are 210 individual congeners (chemicals in the group), although some are more toxic, and some more abundant, than others. 2,3,7,8 - tetrachlorodibenzo-p-dioxin (2,3,7,8 -TCDD) is the most toxic congener, or chemical form, and is now recognised as a human carcinogen. Dioxins are produced as unintentional by-products of many manufacturing and combustion processes that use, produce or dispose of chlorine or chlorine derived chemicals. Important sources of dioxins to the environment include waste incineration, combustion of PVC in landfill fires and open burning, and many organochlorine production processes, including PVC production.

- Polychlorinated Biphenyls (PCBs): PCBs comprise of a group of 209 different congeners. Around half this number have been identified in the environment. The more highly chlorinated PCB congeners are the most persistent and account for the majority of those polluting the environment. PCBs were produced as industrial chemicals that were mainly used for insulation in electrical equipment. Production of PCBs has almost totally ceased worldwide, although there are reports of it continuing in Russia. At least one third of PCBs that have been produced are estimated to have entered the environment (Swedish EPA 1999). The other two thirds remain in old electrical equipment and in waste dumps from where they continue to leach into the environment. Although this is the major source of PCB pollution in the environment today, some PCBs are also produced as by-products of incineration and certain chemical processes involving chlorine such as PVC production.

- Hexachlorobenzene (HCB): This chemical was previously used as a fungicide for seed grain. It is also produced unintentionally as a by-product during the manufacture of chlorinated solvents, other chlorinated compounds, such as vinyl chloride, the building block of PVC, and several pesticides. It is a by-product in waste streams of chlor-alkali plants and wood preserving plants, and in fly ash and flue gas effluents from municipal waste incineration. Its major source today remains the manufacture of pesticides (Foster 1995, ATSDR 1997).

- Organochlorine Pesticides: There are eight pesticides in this category listed by UNEP. These are aldrin, dieldrin, endrin, DDT, chlordane, mirex, toxaphene and heptachlor. The majority of these are banned or restricted in many countries, although not all. For example, DDT is still widely used in developing countries particularly for mosquito control (e.g. Lopez-Carrillo et al. 1996).

Environmental and health problems caused by POPs included on the UNEP list have been recognised for some years and, as a consequence, the PCBs and many of the pesticides have been banned or have restricted use in most countries. However, POPs do not respect national boundaries, such that their continued production and use and generation as unintentional by-products in some countries adds to the global burden of these chemicals. In the case of dioxins, still produced unintentionally by many industrial and waste combustion processes as well as open burning, landfill fires and accidental fires in buildings, vehicles and warehouses throughout the globe. In some countries steps have been taken to reduce air emissions of dioxins from point sources, such as incinerators, but releases to air and soil from such facilities continue with little of no abatement. Moreover, few countries have established the material policies needed to address the chlorine-containing materials (e.g. PVC) that are, in effect, the dioxin sources during incineration as well as for diffuse sources, such as open burning and landfill fires.

The 12 UNEP POPs are only part of the problem we face. Many more persistent organic chemicals are still in widespread production and use, even in the developed world. These include for example, the pesticides technical grade hexachlorocyclohexane (HCH) and the γ-isomer of HCH, otherwise known as lindane, short chain chlorinated paraffins, organotins and polybrominated flame retardants. While the chemical industry continues to manufacture such chemicals to solve day-to-day problems, they may be creating other, long-term or even irreversible problems and compromising the ability of future generations to meet their own needs. They may also be threatening the fundamental processes which support the diversity of life itself.

Although the greatest attention to date has focused, understandably, on persistent organochlorine chemicals, the general problem of the widespread contamination of the environment with persistent chemicals extends across other chemical groups. In order to ensure protection of the environment, action must be taken to reduce and ultimately prevent emissions of all hazardous substances, particularly those which are persistent and bioaccumulative.

1.2 POPs - A Global Pollution Problem

Many POPs have become ubiquitous in the environment and can be detected at considerable levels even in remote regions such as the Arctic and Antarctic (e.g. Bidleman et al. 1993, Iwata et al. 1993). The contamination of remote regions occurs as a consequence of the long distance transport of POPs on air currents. Once in the atmosphere, POPs may be dispersed and transported across great distances on air currents before they are deposited on the earth's surface again. It is speculated that some POPs move through the atmosphere from warmer regions, where they are emitted, towards colder regions at higher latitudes. The hypothesis that explains how POPs move from warm regions to colder polar areas is known as global distillation or global fractionation. This is because once released to the environment, chemicals appear to become fractionated with latitude according to their volatility as they condense at different temperatures (Wania and Mackay 1993, Wania and Mackay 1996).

POPs are released into the environment, for example, from incinerator stacks to air, as industrial discharges to rivers, as pesticides sprayed onto crops and soil and from a variety of consumer products. Subsequent movement of POPs between air, water, soil or

vegetation depends on temperature, and on the physical and chemical properties of POPs. The global distillation hypothesis assumes that warmer temperatures favour evaporation of POPs from the Earth's surface to air, whereas cooler temperatures favour their deposition from air back onto soil, vegetation or water. The overall effect is that POPs volatilise to air in warmer climates and then condense and are deposited again on the Earth's surface in cooler climates. Researchers have suggested that POPs may migrate to the poles in a series of short hops by repeatedly undergoing the cycle of evaporation, transport and deposition (Wania and Mackay 1993). Others have suggested that the process is most likely to occur as a one-step process (Bignert et al. 1998). It has been noted that there are uncertainties about how the processes of exchange occur between air and soil/water/vegetation and that more research is needed (Addo et al. 1999).

It appears that the more volatile a chemical, the greater tendency it has to remain airborne and the faster and farther it travels on air currents towards remote polar regions. Conversely, chemicals of low volatility are unable to attain high atmospheric levels and are thus deposited close to where they are initially released. Therefore, POPs of higher volatility like α- and γ- HCH may migrate faster towards the poles than those of lower volatility like DDT (Wania and Mackay 1993, Wania and Mackay 1996).

Observations suggest that certain POPs such as HCBs and HCHs, preferentially deposit in polar latitudes, while DDT and others primarily deposit at lower latitudes (Wania and Mackay 1996). For example, a worldwide study of persistent organochlorines in tree bark found that the relatively volatile compounds HCB was distributed according to latitude, demonstrating a global distillation effect. Conversely, less volatile compounds such as endosulfan were not as effectively distilled and tended to remain in the region of use(Simonich and Hites 1995).

It is thought that POPs in polar regions mainly originate from industrial and other human activities in nearby countries. For example, studies show that sources of POPs pollution in the Arctic are most likely to come from mid-latitudes of the Northern Hemisphere such as Europe, Russia and North America (Barrie et al. 1989, Muir et al. 1997). However, the tropical countries are also responsible for spreading contamination to the polar regions, because some of these chemicals used in agriculture and public health like HCH, DDT and dieldrin are still consumed in considerable quantities in low latitude areas (Tanabe 1991).

1.3 POPs in Food

1.3.1 Transfer of POPs in the Food Chain

Environmental pollution by POPs has inevitably led to the contamination of our food with these chemicals. POPs may enter the human foodchain in different ways. Some of these routes are exemplified in figure 1.1. For instance, POPs reach the marine or aquatic environment through direct discharges to watercourses or via atmospheric deposition. Since many POPs are lipophilic, they migrate to more lipid-rich tissue in aquatic lifeforms such as fish muscle and fish liver. Through consumption of the fish, the POPs finally accumulate in man. On land, POPs may be deposited on plants directly from sprayed pesticides or from atmospheric deposition. For example, dioxins emitted from an incinerator stack will later be deposited onto the ground, such as grassland. Consumption of the contaminated grass by cows leads to contamination of milk or meat which, in turn, is consumed by humans.

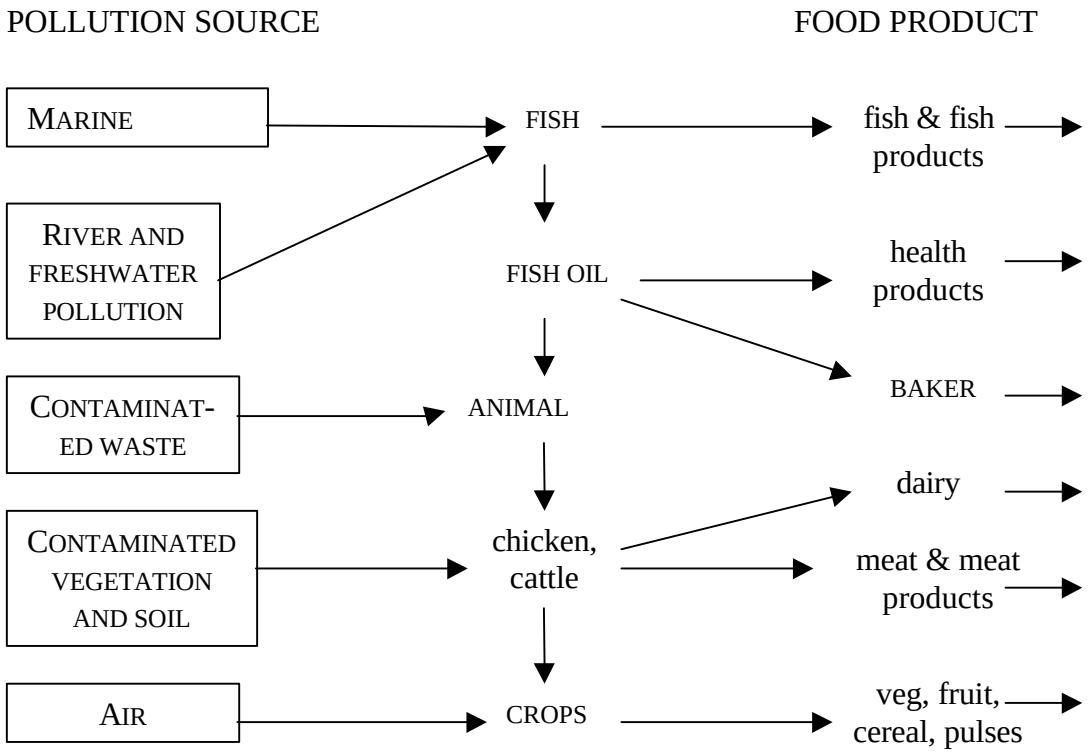
Apart from contamination of food from environmental pollution, incidences of food contamination have occurred through the mixing of contaminated waste with animal feed and nitrogen fertilisers. For example, lime waste contaminated with dioxins was mixed with citrus pulp to generate animal feed. Cows were given this feed which resulted in milk that was contaminated with dioxins and was deemed unfit for sale. In the US, a study of dioxins and metals in fertilisers that are produced with industrial by-products has been conducted. Tests on five fertilisers, all contained dioxins. Two of the samples, granular zinc fertiliser from steel mill flue dust and liming material - wood ash, had particularly high concentrations, respectively 340 and 35 ppt TEQ, which would increase the levels of dioxins in soil (Department of Ecology, State of Washington 1998).

Food is vulnerable to accidental contamination. For example, animal feed in Belgium became contaminated with PCB oil. This caused high levels of contamination in chickens and eggs (see section 4.2).

Once POPs on food are taken into the human gut, it is estimated that absorption is greater than 90%. Data on gastrointestinal absorption with breast-fed babies suggests a nearly complete absorption (Wells and de Boer 1999). As a consequence of the persistent, lipophilic, bioaccumulative properties of many POPs, and tendency of some to biomagnify within food chains, long-term exposure to relatively small concentrations of these compounds leads to the accumulation of considerable deposits in animal and human tissues. Concentrations of POPs in human milk are briefly discussed in section 1.4.

For the general population, the greatest exposure to POPs comes from consumption of food. Some exposure to POPs may also occur through inhalation and dermal contact. In Western industrialised countries, it has been estimated that 90% of the human intake of POPs is via food consumption, and particularly through consumption of fish, meat and milk products. Since POPs are typically lipophilic by nature, the highest concentrations are usually found in fatty foods. In general, higher concentrations are found therefore in fatty meats, fish and dairy than in vegetables, cereals and fruit (Wells and de Boer 1999).

HUMAN FOOD CHAIN



1.3.2 Monitoring of POPs in Food

Monitoring programmes to determine the extent of contamination by some POPs in food have been established by several governments of industrialised countries. These programmes were set up following initial scientific investigations of POPs contamination in the environment, followed by public pressure and finally political awareness. Total diet studies or the Market Basket Programmes were started by the United States Food and Drug Administration (USFDA) in 1964 and total diet studies for organochlorine pesticides and PCBs were started in Japan in 1977. Initially, studies were undertaken on PCBs in the US, Japan and Europe, but by the mid-1980s there was evidence that other organochlorines were contaminating food such as dioxins, in particular as a result of incinerators located near grazing land for cows. Concomitant with government studies on food, there was also a growth of publications by the scientific community on concentrations of POPs in food (Wells and de Boer 1999).

Political awareness of POPs in food and government monitoring programmes have led to legislation on permissible limits for POPs residues in food. In some cases, legislation has arisen directly as a result of food contamination incidents.

1.3.3 Regulatory Limits and Guidelines for the intake of POPs

In an attempt to protect public health, regulatory agencies perform risk assessments to set levels of chemical contaminants in the food supply which deemed to be ‘safe’ and therefore permissible. Depending on the regulatory body concerned, permissible levels set for the dietary intake of chemical contaminants are known by as Acceptable or Admissible Daily Intakes (ADIs), and Tolerable Daily Intakes (TDIs).

Other standards are also set by national authorities on permitted concentrations in food. For example, this includes the Maximum Residue Level (MRL) which is the concentration of a substance to be legally permitted or recognised as acceptable in or on a food, agricultural product or animal feed (Roach and Runcie 1998).

The ADI of a chemical has been defined as the daily intake of a chemical that, during a lifetime, appears to be without appreciable risk on the basis of all the facts known at that time (see Stevens et al.1993). The concept of the ADI is based on the assumption that a threshold exists below which a chemical does not cause toxicity. They are usually based on experiments on the toxicity of the chemical concerned in laboratory animals, from which a lowest observable effect level (LOAEL) or no observed adverse effect level are obtained. In general, the lowest dose value is used to determine a 'safe' level in humans. This result is then divided by a safety factor, usually the arbitrary number of 100, to account for differences in response of laboratory animals and humans, and differences in sensitivity among humans (Ostergaard and Knudsen 1996).

However, ADIs may not be truly protective of human health since they are based on many assumptions and extrapolations from laboratory data. Furthermore, endpoints in animal experiments that are used to derive toxicity data may be inappropriate for a particular chemical because they may not be sensitive enough to detect adverse health effects caused by the chemicals. This could result in higher "acceptable" levels being set than appropriate. Indeed, as more evidence of the toxicology of a chemical is accumulated, values set as acceptable limits may change. For example, WHO originally set a TDI of 10 pg TEQ kg^{-1} day^{-1} for intake of PCDD/Fs and dioxin-like PCBs in diet. This was based on liver toxicity, reproductive effects and immunotoxicity in experimental animals and using kinetic data in humans and experimental animals. However, the TDI was recently reduced to 1-4 pg TEQ kg^{-1} day^{-1} after new human and animal toxicity results became available on neurodevelopmental and endocrine effects (van Leeuwen et al. 2000).

ADIs may also not be truly protective of human health because they only consider the toxicological impact of a single chemical. In reality, however, humans are exposed to mixtures of numerous chemicals, not just single substances. Research has shown that some chemicals may have cumulative effects when mixed together. This could confound attempts to set safety limits for chemicals (Johnston et al. 1996). Finally, ADIs are based on an "acceptable" risk to human health. This does not mean "zero" risk. In this context, it is of note that with regard to endocrine-disrupting chemicals, which includes some persistent organochlorines, it has been stated by Professor vom Saal (University of Missouri), (ENDS 1997), that "*There are no safe doses of endocrine disrupters, just as there are no safe doses of carcinogens*".

ADIs have been set by the World Health Organisation (WHO) for a number of organochlorine pesticides including total DDT compounds, HCB, (aldrin + dieldrin), heptachlor, heptachlor epoxide, total chlordane and lindane (WHO 1997). ADIs have not been established for oxychlordane, *trans*-nonochlor, p,p'-DDE, p,p'-DDT, isomers of PCBs, and α and β-HCH (Quinsey et al.1996). ADIs for dioxins have been set by a number different regulatory bodies such as US EPA and WHO (US EPA 1994b, van Leeuwen et al. 2000).

The World Health Organisation (WHO) has set a TDI for humans of 1-4 pg ITEQ kg^{-1} day^{-1} (WHO 1998, van Leeuwen & Younes 1998). This includes not only the PCDD/Fs but also the dioxin-like PCBs.

Current dietary intake in industrialised countries may be in excess of this guideline. Average dioxin intakes in the EU have been estimated to be within the range of 1-3 pg I TEQ kg^{-1} day^{-1}. However, children and high level consumers of fish and fatty foods may receive significantly higher intakes than the average (King 1999). Inclusion of the dioxin-like PCBs may increase this by a factor of 2-3.

WHO experts acknowledged that subtle toxicological effects might already be occurring in the general human population and *that every effort should be made to reduce exposure to the lowest possible level.* The WHO experts also stressed that the upper range of the TDI of 4 pg ITEQ kg^{-1} day^{-1} should be considered a maximal tolerable intake on a provisional basis and that the ultimate goal is to reduce human intake levels below 1 pg ITEQ kg^{-1} day^{-1}.

In the US, a risk-specific dose, the amount estimated to cause cancer in one person in a million, is used as the basis for decision-making. The estimate for dioxins, furans and dioxin-like PCBs is 0.01 pg kg^{-1} day^{-1} (EPA 1994), considerably less than the WHO TDI and current human dietary intake.

The food consumption patterns of infants and children are different from those of adults, but no separate ADIs have yet been established for infants or children. In the present system, ADIs set for dioxins and organochlorine pesticides are designed for a whole lifetime exposure of a 70kg adult, where a mixed diet has a dilution effect on the consumption of contaminants (Quinsey et al.1996). However, such an assumption does not consider possible adverse effects of exposure at critical times during development of the foetus or child.

The application of ADIs to intake of organochlorines over the short-time period of breastfeeding is unclear (Sonawane 1995). Nevertheless, it is of great concern that current levels of some organochlorines in human milk mean that the ADI is exceeded when applied to infant intake. Since infants may be more susceptible to the toxic effects of chemicals than adults, because for example, they are undergoing rapid tissue growth and development, then it is most likely that current ADIs are higher than appropriate for infants (Quinsey et al. 1996).

1.3.4 Regulatory limits and guidelines for POPs in food

There are currently no internationally agreed maximum concentration limits for food contamination with either PCDD/Fs (dioxins) or PCBs, with the exception of EU regulations after the Belgian incident discussed below, which have been established for a limited time for a limited set of circumstances.

Several national governments have set guidelines for the maximum acceptable concentrations of dioxins in food, predominantly milk. These are summarised in Table 1.1 (see also Table 3.10).

Table 1.1: National Guidelines for the Maximum Acceptable Concentrations of Dioxins in Food

Country	Concentration (pg ITEQ g^{-1} milk fat)	Notes and references
Belgium	5	de Fre & Wevers 1998
Germany	<0.9	Long term goal
	3.0	Intervention value I; milk should not be sold directly to the public but mixed with milk from other farms, and measures should be initiated to ameliorate the source
	5.0	Intervention value II; milk above this value should not be marketed at all (Thiesen et al. 1997)
Netherlands	6	Liem et al. 1990 & 1991
UK	16.6	relates to the total ITEQ for both PCBs and PCDD/Fs (MAFF 1998)
Austria	35	Set by Federal Health Ministry; Federal Environment Agency recommends using the Dutch limits (Riss 1993)

In addition to the milk guidelines, Germany also has an intervention value of 10 pg ITEQ g^{-1} dry weight (1pg ITEQ g^{-1} wet weight) for leafy vegetables (Thiesen et al. 1997) and a tiered system for addressing soil contamination of agricultural soil designed to prevent unacceptable food contamination. The target value is < 5 pg ITEQ g^{-1} dry soil; between 5 – 40 pg ITEQ g^{-1} there may be unrestricted food cultivation but certain practices such as grazing may be avoided if dioxins are detected in elevated concentrations in foodstuffs. Above 40 pg ITEQ g^{-1}, the recommendation is that land be used for defined agricultural practices, such as growing plants that do not take up dioxins (Basler 1994).

Two of the food contamination incidents described in this report resulted in EC legislation on PCBs and dioxins. However, in both these cases, the legislation is very specific and so will not protect against future incidents of this kind.

The dioxin and PCB contamination of animal feed in Belgium during 1999 lead to several EC Decisions which initially restricted sales, and subsequently, specified limits of PCBs in Belgian poultry, eggs, pork, milk and products derived from them. The most recent Decisions (EC 1999a and EC 2000) outline current controls, which include a maximum concentration of PCBs (as a total of 7 congeners) of 200 ng g^{-1} fat for fresh poultry meat and derived products, fresh pig meat and derived products.

The incorporation of contaminated lime from a Brazilian factory into citrus pulp pellets used for cattle feed in Europe caused a significant dioxin contamination incident, which is discussed elsewhere. It resulted in an EC Directive limiting the dioxin content of citrus pulp in animal feed to 500 pg ITEQ kg^{-1} (98/60/EC, EC 1998). This limit was retained in 1999 when the original Directive was superseded by Directive 1999/29/EC (EC 1999b) and remains unchanged in the proposed Directive which would replace Directive 1999/29/EC (EC 1999c). However, this limit refers specifically to citrus pulp and does not apply to other types of animal feed. It also does not contain any limits for PCBs in feedstuffs, despite the severe disruption of the European food industry which occurred when PCBs became were added to feedstuffs in Belgium in 1999.

European and US legislation define materials containing or contaminated with more than 50 mg kg^{-1} of PCBs as hazardous waste (EC 1996, Rogan 1995) as does the Basel Convention, which is a global convention controlling the international trade in hazardous wastes (UNEP 1998). For limits and guidelines for organochlorine pesticides in various food products see table 3.6, 3.9 and 3.15.

1.4 Levels of POPs in Human Tissues

Most of the 12 POPs listed by the UNEP are widespread pollutants and they have been found to commonly occur in human blood, milk and adipose tissues worldwide. A previous Greenpeace report discussed levels of POPs in human tissues with particular reference to human milk (Allsopp et al. 1998). The report noted for PCDD/Fs and dioxin-like PCBs, that the highest levels in human milk were generally found in industrialised countries. This is because these compounds are by-products of many industrial processes including incineration. The report also commented that in general, levels of organochlorine pesticides in human milk were highest in countries where these chemicals are still in use, notably in less industrialised countries. For example, by far the highest levels of DDTs in human milk were evident in Asia, notably India, Africa and South America due to the continued use of DDT in agriculture and/or sanitation campaigns for vector control.

An exception to the highest levels of organochlorine pesticides occurring near places of use is for Indigenous Peoples of the Arctic. Particularly high concentrations of HCB, DDT and chlordane were found in blood of women from Arctic regions who consumed a traditional diet that included sea mammals. PCB levels were also elevated. High levels of organochlorines are present in some populations of Arctic Indigenous Peoples because sea mammals, which are consumed as part of a traditional diet, are contaminated by high levels of some persistent organochlorines. This is due to the long-distance transport of POPs towards Arctic regions and because marine mammals accumulate particularly high levels of POPs in their tissues.

Studies on the levels of PCDD/Fs in human milk show that levels in industrialised countries have not increased in recent years and have decreased in a few European countries. Data shows that PCBs have remained stable in recent years. Levels of DDT have declined in some countries where it is banned but not in countries where it is still used such as Mexico. Interestingly, studies in Australia indicate a decline in DDT levels in human milk between the mid 1970s and early 1990's but one study shows levels of DDE have increased in recent years (Miller et al. 1999). A review of data on chlordane in 1991 concluded that levels of chlordane in people are not declining.

1.5 Toxicity of POPs

Organochlorine pesticides have had some devastating health impacts in wildlife and have caused massive population crashes. They have also been associated with a wide range of health effects in humans including carcinogenic effects. The adverse effects of organochlorine pesticides and PCDD/Fs on health have been reviewed in other Greenpeace reports (Allsopp et al. 1999, Allsopp et al. 1997, Allsopp et al. 1995).

Dioxins and dioxin-like compounds

The term “dioxins” is the commonly used name, though not the correct chemical nomenclature, for a class of chemicals known as polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). The number of chlorine atoms in these compounds varies between 1 and 8, resulting in a possible 75 different PCDDs and 135 PCDFs. Another group of chemicals, the Polychlorinated Biphenyls (PCBs), constitute a group of 209 congeners. Certain PCBs have been found to exert similar toxicity to 2,3,7,8-TCDD, and are known as dioxin-like PCBs.

There is considerable evidence that chemicals which cause dioxin-like toxicity exert many of their toxic effects by binding to a receptor in cells known as the Aryl Hydrocarbon (Ah) receptor. This common mechanism of toxicity has lead to the development of the concept of Toxic Equivalents (TEQs) for complex mixtures of dioxins.

Over the years, there have been a number of TEQ systems developed and all work in the same way. Using various toxicological measures obtained from laboratory experiments (e.g. Ah receptor binding and enzyme induction), each congener's toxicity is assessed relative to the toxicity of 2,3,7,8-TCDD and recorded as the congener's Toxicity Equivalence Factor (TEF). The TEQ of a sample containing a mixture of dioxins is obtained by multiplying the individual concentration of each congener by its TEF and summing the results.

Despite its undoubted utility, the TEF system represents a considerable simplification of the real world situation and may not accurately describe the full range of toxicological effects of dioxins, furans and PCBs. TEFs will require adjustment as toxicological data improves.

The World Health Organisation (WHO) has developed the newest and most sophisticated TEFs: these, for the first time, acknowledge interspecies differences and present different TEFs for mammals, birds and fish. In time, they will supersede the International TEQs (ITEQs) which have been used almost universally throughout the 1990s. The ITEFs and the WHO human/mammal TEFs are given in table 1.2.

TABLE 1.2 Current Toxicity Equivalent Factor (TEF) Schemes for PCDD/Fs

Compound	WHO 1998 Human/mammal	I-TEF
2378-TCDD	1	1
12378-PeCDD	1	0.5
123478-HxCDD	0.1	0.1
123678-HxCDD	0.1	0.1
123789-HxCDD	0.1	0.1
1234678-HpCDD	0.01	0.01
OCDD	0.0001	0.001
2378-TCDF	0.1	0.1
12378-PeCDF	0.05	0.05
23478-PeCDF	0.5	0.5
123478-HxCDF	0.1	0.1
123678-HxCDF	0.1	0.1
123789-HxCDF	0.1	0.1
234678-HxCDF	0.1	0.1

1234678-HpCDF	0.01	0.01
1234789-HpCDF	0.01	0.01
OCDF	0.0001	0.001

Source: Maisel & Hunt (1990), van den Berg et al. (1998).

Dioxin-like PCBs

Persistent compounds which also bioaccumulate and cause dioxin-like effects such as enzyme induction may qualify for inclusion in the TEF scheme (van den Berg et al. 1998, van Birgelen 1998). Certain of the PCBs have long been recognised as having dioxin-like toxicity. The most widely used PCB TEF scheme is the WHO/IPCS scheme (Alhborg et al. 1994). However, in 1998, an international WHO consultation group updated the TEFs, giving different factors for humans/mammals, fish and for birds. The WHO/IPCS and the 1998 WHO human/mammal PCB TEF schemes are given in table 1.3.

The WHO emphasised the importance of including PCBs in the TEF scheme for dioxins with the comment that:

> *"Use of TCDD alone as the only measure of exposure to dioxin-like PCDDs, PCDFs and PCBs severely underestimates the risk to humans from exposure to these classes of compounds. Thus, the TEF approach is recommended for expressing the daily intake in humans of PCDDs, PCDFs, non-ortho PCBs and mono-ortho PCBs in units of TCDD equivalents (TEQs) for comparison to the tolerable daily intake (TDI) of TCDD."* (WHO 1998a)

However, it must be remembered that the growing reliance upon the assessment of toxicity of PCBs through the TEQ may fail to predict total PCB toxicity accurately. There are also significant toxic effects (for example mammalian neurotoxicity) which are expressed through non-dioxin mechanisms, and these effects must also be considered when considering PCB toxicity..

In the present report, concentrations of PCDD/Fs and dioxin-like PCBs in food are presented as International TEQs.

Table 1.3 Different TEF schemes for dioxin-like PCBs.

IUPAC no	WHO 1998 humans/mammals	WHO/IPCS 1994
81	0.0001	
77	0.0001	0.0005
126	0.1	0.1
169	0.01	0.01
105	0.0001	0.0001
114	0.0005	0.0005
118	0.0001	0.0001
123	0.0001	0.0001
156	0.0005	0.0005
157	0.0005	0.0005
167	0.00001	0.00001
189	0.0001	0.0001
170		0.0001
180		0.00001

Source: Data are taken from Ahlborg et al. (1994) and van den Berg et al. (1998).

1.5.1 Relevance of Animal Studies to Human Dioxin Toxicity

The relevance of animal testing data to the elucidation of dioxin toxicity to humans was examined by Grassman et al. (1998);

> "*The adverse effects of dioxins are well established based on studies of experimental animal models and highly exposed human populations. From these investigations, the current view of dioxins as potent toxicants capable of producing a multitude of diverse biologic effects has emerged*."

They summarised the relevance of animal models to human health end-points as follows:

- *The reproductive, developmental, immunologic, and carcinogenic responses to dioxins seen in humans also occur in animal models;*
- *The preponderance of biochemical effects induced by dioxins in both animals and humans are mediated by the AhR;*
- *Animal dosing regimens can be varied to examine the range of exposures encountered in human populations;*
- *Dose metrics based on internal dose (tissue dose and body burden) can be used to compare responses across species as these parameters take into account species differences in clearance rates;*
- *The biochemical responses to dioxins in animal models show qualitative and quantitative similarity to those observed in humans;*

and concluded that,

"*the biochemical responses in human and animals, biologic responses to dioxins are qualitatively and often quantitatively similar. Exposure to dioxins has been implicated in a wide range of human health effects related to reproduction, immune function, growth and development, and cancer*."

1.6 Problems Comparing Levels of Organochlorines in Food Between Different Countries

Many studies have been published on the concentrations of organochlorines in food. However, there are several problems which can hinder comparisons between studies. These problems arise from differences in the scientific methods which are used to measure the concentrations of contaminants. For example, differences between studies may occur in sample collection, storage, preparation, the method used for chemical analysis, mathematical analyses and data interpretation. A difference in the sensitivity of an analytical method for instance could affect whether or not a compound is detected.

In the present report, comparisons of organochlorine concentrations in foods from different countries are made. Although differences in methodologies used by different studies makes direct comparison difficult, the presentation of contaminant concentrations in food can at least give an indication of levels. Information is given as to whether regulatory food standards are exceeded or not.

2. STUDIES ON DIETARY INTAKE OF ORGANOCHLORINES

Food is known to be a major source of PCDD/Fs and other persistent organochlorines in the general population (Hall 1992; Jimenez, Hernandez et al. 1996a). Consequently, studies have been conducted to investigate whether levels of organochlorines in food lie within permitted safety limits designated by national and international authorities. Studies have also been conducted to monitor whether a decrease in the use of organochlorines has resulted in a concurrent decrease in levels in food.

Average daily intake (ADI) and tolerable daily Intake (TDI) are standards that have been set as regulatory guides for food intake (see Section 1.3). Using these standards, many studies have been undertaken which have compared the relevant ADI or TDI to an estimated daily intake of an organochlorine contaminant in a normal diet. To do this, most studies derive levels of organochlorine contaminants in various foodstuffs that are selected to be representative of the normal diet in a given country. Analysis of different food samples is carried out and then an "average" concentration in foods is estimated. By combining these "average" concentrations with food consumption data, a tentative estimate for the average dietary intake of contaminants by the population may be obtained (Jimenez, Hernandez et al. 1996a). In some cases, however, somewhat different methodologies have been adopted, although the basic aim of studies remains to determine the intakes of contaminants in diet. (Schecter, Startin et al. 1994). Schecter (1994) has noted that the large variation of PCDD/F levels found in food samples in a US study highlights the difficulty in estimating intake for a large population.

Studies have been conducted on intake of PCDD/Fs and PCBs in diet and on intake of some organochlorine pesticides.

2.1. Dietary Intake of PCDD/Fs and PCBs

Table 2.1 shows estimated daily dietary intakes of PCDD/Fs and PCBs in various countries. No studies were located on less industrialised nations in the scientific literature and data are limited to industrialised countries only. In some cases, studies have reported intakes of PCDD/Fs and others have also included dioxin-like PCBs. It is important to note that the dioxin-like PCBs can contribute significantly to the daily intake of TEQs. For instance, in Norway, dioxin-like PCBs contributed more than PCDD/Fs to intake (Becher, Eriksen et al. 1998), and in the Netherlands, dioxin-like PCBs represented more than 50% of the daily intake (Theelen, Liem et al. 1993).

The current TDI established by WHO for PCDD/Fs plus dioxin-like PCBs is 1-4 pg TEQs kg^{-1} body weight day^{-1}, (pg TEQ kg^{-1} bw d^{-1}) (van Leeuwen et al. 2000). Most of the estimated daily intakes for different countries lie within this range. However, a Norwegian study estimated daily intakes of PCDD/Fs and PCBs to be 2.28-3.16 pg TEQs kg^{-1} bw d^{-1}, but pointed out that high level intake of food with high concentrations of PCDD/Fs and PCBs, such as seafood from contaminated areas, can have a large impact on the intake of TEQs (Becher, Eriksen et al. 1998). Furthermore, the WHO TDI is exceeded in Spain, the upper bound daily intake being 4.5 pg TEQ kg^{-1} bw d^{-1} (Jimenez, Hernandez et al. 1996a), and is exceeded by school children designated to be high level consumers in the UK (MAFF 1997c). Indeed, it has been noted in a US study by Schecter (1994) that the dietary habits and lower body weight of children may cause them to have markedly higher intakes of

PCDD/Fs than adults at the crucial time when their physical and mental capabilities are developing. A dietary exposure study in France for 1998 showed that the average intake of children (mean 3.31 pg TEQ kg^{-1} bw d^{-1}), was greater than the average intake for men and women (respectively 1.78 and 2.17 TEQ kg^{-1} bw d^{-1}), (European Commission DG Environment 1999).

The application of the TDI to breast-fed infants is questionable, although given the vulnerability of the early formative stages of life to toxic chemicals, it has been stated that TDIs should be lower for breast-fed infants (see section 1.3.4). The intake of PCDD/Fs and dioxin-like PCBs is 1-2 orders of magnitude greater than adult intakes. In the Netherlands, it was estimated that breast-fed children were exposed to 200 TEQ kg^{-1} day^{-1} (see Wells and de Boer 1999).

In comparison with other countries, the estimated dietary intake in New Zealand was lower (Buckland, Scobie et al. 1998). This is likely to be a reflection of the low level of industrialisation in New Zealand relative to Europe and North America.

The consumption of high amounts of fatty fish from contaminated waters can lead to a very high dietary intake of PCDD/Fs and dioxin-like PCBs. For instance Baltic Sea Fishermen were found to have an intake of 10.3 Nordic TEQ kg^{-1} bw d^{-1} . This is far higher than the average Swede, estimated to have an intake of 0.83-0.92 Nordic TEQ kg^{-1} bw d^{-1} (European Commission DG Environment 1999).

A study on the Italian diet reported a mean daily intake of total PCBs of 3.72 μg $person^{-1}$ day^{-1} (or 0.062 μg kg^{-1} d^{-1} for a 60 kg person) which exceeds the minimum risk level recommended by International Agencies (0.02 μg kg^{-1} d^{-1}), (Zuccato, Calvarese et al. 1999). However, the intake of dioxin-like PCBs (0.57 pg TEQ kg^{-1} d^{-1}) did not exceed the WHO TDI of 1-4 pg TEQ kg d^{-1}. Even so, was noted that occasional exposure to highly toxic PCB congeners may cause this limit to be exceeded.

In the Arctic, populations who consume a traditional diet which includes consumption of sea mammals may be exposed to high levels of PCBs. This is because PCBs build up to particularly high levels in the tissues and blubber of sea mammals such as whales, seals and walrus. A study of dietary intake of organochlorines from the traditional diet of Arctic Inuit from Broughton Island, Canada, in 1987/8 found that the many consumers exceeded the tolerable daily intake for PCBs (Kinloch, Kuhnlein et al. 1992). Although the health risks of this consumption are uncertain, it has been noted that limiting the intake of traditional foods could not be recommended, in part, because available alternative imported foods are nutritionally inferior. In addition, as well as conferring nutritional benefits the consumption of a traditional diet has important cultural and social benefits.

In the Faroe Islands, Denmark, pilot whales are consumed by many of the Islanders. Due to high levels of heavy metal contaminants in the whales, the Faroes Food and Environmental Institute has already warned that pilot whale flesh and blubber should not be eaten more than once a week and that kidney and liver should not be eaten at all. A study of levels of organochlorines in pilot whale blubber from the Faroe Islands found high levels of PCBs and other organochlorines. It was estimated that at an average consumption level (49 g per week of blubber) the intake of PCBs was 2.06 μg kg^{-1} bw d^{-1}. This exceeds the US Food and Drugs Administration tolerable daily intake for PCBs of 1 μg kg^{-1} bw d^{-1}. Therefore,

even at the limited intake recommended by the Faroes Food and Environmental Institute, the tolerable daily intake for PCBs is exceeded (Simmonds et al. 1994).

It has been reported that a number of studies on PCDD/Fs in food from several countries found that meat, milk and fish products account for about 95% of the human general population intake of PCDD/Fs (Schecter, Startin et al. 1994). However, recent research in Spain found that the high consumption of vegetable and cereals included in the Mediterranean diet contributed significantly to the daily intake of PCDD/Fs (Schuhmacher, Franco et al. 1997; Domingo, Schuhmacher et al. 1999). For instance the total TEQ intake for meat, fish, eggs, fat and oils, milk and dairy products was 117 pg TEQ day^{-1}, but when vegetables, pulses, cereals and fruits were included, the total TEQ rose to 210 pg TEQ day^{-1}. Similar results were reported in an Italian study. The authors noted that food groups such as vegetables, fruits, and cereals should, therefore, not be excluded to estimate the total dietary intake of PCDD/Fs by general populations, especially in countries and/or regions in which their consumption's are notable (Domingo, Schuhmacher et al. 1999).

For intake of total PCBs, studies have shown that the contribution of different foods to dietary intake varies between different countries. In Germany dairy products were the predominant source of PCBs, in Japan and Finland fish, in Canada meat and in Vietnam and other tropical less industrialised countries cereals and vegetables (Kannan, Tanabe et al. 1992a).

Research has shown that daily intakes of PCDD/Fs and PCBs have decreased considerably in recent years. For instance, in the UK, the daily intake of PCDD/Fs plus PCBs for adults fell from 6.8 pg TEQ kg^{-1} bw d^{-1} in 1982 to 2.4 pg TEQ kg^{-1} bw d^{-1} in 1992 (MAFF 1997c).

Table 2.1 Estimated Daily Intakes of PCDD/Fs and dioxin-like PCBs (pg TEQ kg^{-1} body weight day^{-1}) from Dietary Intake Studies in Various Countries

Location/Date	PCDD/F	PCDD/F + PCBs	Reference
Canada (Ontario) (1986)	1.52		Birmingham, Thorpe et al. 1989
Germany (1993-1995)	0.88		Malisch 1998b
Germany (1994/5)	0.18 - 1.7		Schrey, Mackrodt et al. 1995
Italy (1999)		0.57 (PCB only)	Zuccato, Calvarese et al. 1999
Italy (Venice)	0.25 - 2.13		Zanotto, Alcock et al. 1999
Japan (1987)	1.3		Ono, Kashina et al. 1987
Japan (1997)		2.7	Hori, Iida et al. 1999
Netherlands (1990-1996)		1.2 - 2.4	Cuijpers, Bremmer et al. 1998
Norway (1998)		2.28 - 3.16	Becher, Eriksen et al. 1998
New Zealand (1998)		0.33 (adult male 80 kg in weight) 0.76 (adolescent male)	Buckland, Scobie et al. 1998
Russia, Republic Baskortostan (1996)	1.15 - 2.31		Maystrenko, Kruglov et al. 1998
Spain (Catalonia) (1996)	3.5		Schuhmacher, Franco et al. 1997
Spain (Madrid), (1995)	2.4	4.5	Jimenez, Hernandez et al. 1996b
UK (1992)		2.4 (average consumers) 4.2 (high level consumers) 2.8 (school children) 4.6 (school children - high level consumers)	MAFF 1997c
USA (1995)	0.52 - 2.57	1.16 - 3.57	Schecter, Cramer et al. 1996

Footnotes: Where the date of sampling is not given in a study, the date is recorded as the date of publication. Where a study presents dietary intake in pg TEQ day^{-1}, the daily intake per kg body weight per day is calculated by assuming an adult of body weight 60 kg.

2.2. Dietary Intake of Organochlorine Pesticides

A number of studies have been reported on daily intakes of organochlorine pesticides from diets in several countries as shown in table 2.2. However, data is limited since in many countries, studies have not been performed. In addition, some organochlorine pesticides are not analysed frequently in reports such as heptachlor, chlordane and toxaphene. Of the organochlorine pesticides, most data is available for intake of DDTs.

ADIs for dietary intakes of DDT, HCH, aldrin and dieldrin and HCB are presented in table 2.2. For all of the countries presented in table 2.2, together with a study on intake by women and children in Germany (Petzold, Schäfer et al. 1999), none of the ADIs are exceeded for any of the organochlorines by dietary intake with the exception of India. Most of the dietary intakes were very low in comparison to ADIs. However, in India the ADI for aldrin and dieldrin is exceeded. Intakes of DDT and HCH are also extremely high in comparison with other countries (Kannan, Tanabe et al. 1992b). Dairy products contributed highly to intakes of HCH (70%), DDT (87%) and aldrin and dieldrin (87%) in India. The higher intake of these chemicals in India can be attributed to their use in agriculture and vector control. In China and Vietnam, intake of DDT was also high in comparison with other countries and intake of HCH is also somewhat higher (Chen and Gao 1993), (Kannan, Tanabe et al. 1992a). In Vietnam, the primary route of exposure to DDTs was through fish and shellfish whereas exposure to HCHs was predominately via cereals and vegetables.

In the Arctic, consumption of a traditional seafood diet which includes sea mammals by Indigenous Peoples can result in a high intake of organochlorines. A study of dietary intake by Arctic Inuit of Broughton Island, Canada, found that many consumers exceeded the tolerable daily intake of toxaphene (Kinloch, Kuhnlein et al. 1992). Another study on dietary exposure in the Dene/Métis Indigenous Peoples of the western Northwest Territories, Canada was conducted. The diet of these people consists mainly of herbivorous animals and fish with little consumption of sea mammals. The study found that dietary exposure to organochlorine contaminants was relatively low (Berti, Receveur et al. 1998).

In the Faroe Islands, flesh and blubber from pilot whales is consumed by many of the Islanders. A study on organochlorines in the blubber of pilot whale from the Islands revealed that at an estimated average consumption (49 g $week^{-1}$ of blubber), the mean daily intake calculated for dieldrin (0.3 μg kg^{-1} d^{-1}) exceeded the WHO/FAO ADI of 0.1 μg kg^{-1} d^{-1}. The Faroes food and Environmental Institute had previously recommended consumption of no more than one meal per week of whale flesh and blubber because of high levels of heavy metals in this food, but at this rate of consumption for blubber, the ADI for dieldrin is exceeded (Simmonds and Johnston 1994).

Most of the studies reported in table 2.2 noted that dietary intake of organochlorine pesticides has decreased substantially in comparison to intake in the 1970s/80s. For instance, an approximately 50 % decrease in DDT intake was reported between the mid-1970s and 1990 in Sweden (Vaz 1995) and China (Chen and Gao 1993), and significant declines were also reported in Spain, Slovakia (Prachar, Uhnák et al. 1996) and the Netherlands (Brussard, van Dokkum et al. 1996). HCH has been reported to decrease from the mid-1970s to around 1990 by 2-fold in Slovakia (Prachar, Uhnák et al. 1996), 10-fold in Spain (Herrera, Ariño et al. 1996) and 30-fold in China (Chen and Gao 1993). An appreciable decline in HCB was reported in Slovakia (Prachar, Uhnák et al. 1996), Sweden (Vaz 1995) and the Netherlands (Brussard, van Dokkum et al. 1996), and a general decline in dieldrin was reported in Sweden.

An exception to declining dietary intakes of DDT and HCH can be seen in India. Here, comparison of dietary intake data from 1989 with that of the early 1980s shows that consumption of DDT and HCH in the diet has increased. This is due to increased use of DDT and HCH throughout this time period (Kannan, Tanabe et al. 1992b).

Few studies reported on intakes of heptachlor in the diet. A study in Taiwan reported a daily intake of 2.147 μg person^{-1} day^{-1} heptachlor and 0.702 μg person^{-1} day^{-1} of heptachlor epoxide (Doong and Lee 1999). These intakes are below the ADI recommended by FAO/WHO of 30 μg person^{-1} day^{-1}, see (Kannan, Tanabe et al. 1994). Pork and fish contributed largely to this intake although significant residues of these compounds were also detected in cereals. In Australia, the estimated daily intake of heptachlor and heptachlor epoxide for persons aged 25-34 was 1.2 μg person^{-1} day^{-1} (Kannan, Tanabe et al. 1994), and in India the daily intake of heptachlor was 0.07 μg person^{-1} day^{-1} (Kannan, Tanabe et al. 1992b).

Table 2.2 Estimated Mean Total Dietary Intake of Organochlorines (μg person^{-1} day^{-1}) in Various Countries

Location/Date	Total DDTs ADI 1200μg/day	Total HCHs ADI (α+β+γ) = 840 μg/day	Aldrin +Dieldrin ADI 6 μg/day	HCB ADI 1.03 μg/day	Reference
Australia	1.95	-	-	-	see Chen and Gao 1993
Australia (1990/2)	5.8	1.6	4.0	0.28	Kannan, Tanabe et al. 1994
Republic of Belarus	2 - 5	-	-	-	Barkatina, Pertsovsky et al. 1999
China (1990)	20.47	5.04	-	-	Chen and Gao 1993
India (1989)	48	155	19	0.13	Kannan, Tanabe et al. 1992b
Japan (1992/3)	1.42	0.56	0.09 (dieldrin)	-	Nakagawa, Hirakawa et al. 1995
Netherlands (1988-9)	1.0	-	-	0.2	Brussard, van Dokkum et al. 1996
Slovakia (1995)	5.75	-	-	0.225	Prachar, Uhnák et al. 1996
Spain (1987-90)	1.22	2.92	0.48 (dieldrin)	1.03	Herrera, Ariño et al. 1996
Switzerland (1981-83)	1.7	2.5	0.9 (dieldrin)	1.1	see Herrera, Ariño et al. 1996
Taiwan (1999)	0.541	-	0.061	-	Doong and Lee 1999
USA (1990)	1.56	0.12	0.1 (dieldrin)	0.03	see Herrera, Ariño et al. 1996
Vietnam (1990/1)	19	5.4	0.55	0.1	Kannan, Tanabe et al. 1992b

3. CONCENTRATIONS OF ORGANOCHLORINES IN VARIOUS FOOD TYPES

3.1. Fish

3.1.1. Marine Fish

Levels of organochlorines detected in fish muscle from marine fish world-wide are presented in table 3.1. Direct comparison of results between studies is made difficult by differences between laboratories in analytical techniques and the fact that different species of fish are being compared. However, the data are presented to give an indication of organochlorine levels in fish from different regions.

In general, levels of organochlorines in marine fish are higher in the Northern Hemisphere than the Southern Hemisphere. Fish from tropical Southeast Asia were found to have lower levels than those in fish of temperate regions. Data presented in table 3.1 shows that levels are also generally lower than levels in cold waters of the Northern Hemisphere. This is different to levels of some organochlorines in air and seawater which are higher in the tropics than the mid-latitudes. The reason that fish from tropical latitudes accumulate lower concentrations of organochlorine compounds than fish from temperate regions may be explained by the shorter residence time of these compounds in the tropical environment. For instance, DDT, HCH, chlordanes and HCB are likely to rapidly volatilise in the tropical environment. Also, higher temperatures in the tropics could enhance the elimination rate of chemicals in fish (due to the influence of temperature on respiratory requirements of the fish), as the biological half-lives of compounds such as DDT are shorter at high temperature. A survey of organochlorine levels in fish in tropical Southeast Asia observed that DDT was the predominant organochlorine and levels of other organochlorines were relatively low.

In addition to analysing concentrations of organochlorines in fish, studies have also determined levels in mussels in different areas of the world in a number of Mussel Watch schemes. These studies in mussels have been conducted to check contamination levels in the interests of public safety. In addition, mussels are used as bioindicators to assess the state of marine pollution by toxic contaminants (Tanabe 1994). Mussel Watch programs have been conducted in the US (O'Connor 1998), France (Claisse 1989) and Southeast Asia (Tanabe 1994). Results of these studies are not directly comparable because those from Southeast Asia are presented on a wet weight basis whereas those from US and France are on a dry weight basis. Mussels and other shellfish have also been monitored in many other countries to assess contamination levels of organochlorines.

Table 3.1 Range (in Parentheses) and Mean Concentrations of Organochlorines (ng g^{-1} (ppb) wet weight) in Marine Fish

Location	number of samples	PCBs	DDTs	HCHs	Aldrin and Dieldrin	Chlordanes	HCB	Reference
West Svalbard (Arctic)	52 (Arctic cod)	0.6	0.46	0.18	-	-	-	AMAP 1998
Pechora Sea and South Novaya Zemlya (Arctic)	57 (Arctic cod)	0.54	0.22	0.22	-	-		AMAP 1998
Cumberland Sound, Baffin Island, Canada	10 (Turbot, Greenland Halibut)	165	129	15.1	24.4 (dieldrin)	127		AMAP 1998
East Beaufort sea, Banks Island, Canada	10 (Turbot, Greenland Halibut)	202	128	13.6	24.4 (dieldrin)	115		AMAP 1998
Barents Sea, Norway	25 (Atlantic cod – liver)	392 (158-685)	114 (67-201)	12 (9-16)	-	75 (39-181)	-	AMAP 1998
Faeroe Islands	5x5 (Atlantic cod -liver)	63 (52-68)	46 (42-50)	5 (4-5)	-	25 (22-30)	-	AMAP 1998
Iceland	5x5 (Atlantic cod – liver)	76 (63-109)	71 (60-98)	9 (7-9)	-	49 (40-57)	-	AMAP 1998
Swedish coastal waters	49	-	means 0.42-98	means 0.14-8.4			means 1.13-5.3	Atuma, Linder et al. 1996
UK coastal waters	Dab – liver		means <12-220		means 4 - 49			CEFAS 1998
Mediterran-ean, Spain	6 (Red mullet)		20.72	-	-	-	0.68	Sanchez, Sole et al. 1993
Mediterran-ean, Morocco	15 *Mullus barbatus*	-	17.4	3.95 (lindane)	2.83	-	0.62	El Hraiki, El Alami et al. 1994
New Jersey, USA	-	-	37.75-168.32	-	-	9.08-163.96	-	Kennish and Ruppel 1996b; Kennish and Ruppel 1996a
Hong Kong			means 7.94 - 75.63	-	-	-	-	Dickman and Leung 1998
Cambodia		0.36 (0.05-1.2)	8.1 (0.51-25)	0.08 (0.01-0.22)		0.11 (0.03-0.04)	0.09 (<0.01-0.32)	Monirith, Nakata et al. 1999
India	48	3.5 (0.38-110)	15 (0.86-140)	28 (0.48-380)	3.1 (<0.1-15)	2.4 (<0.01-30)	0.07 (<0.01-0.55)	Kannan, Tanabe et al. 1995
Thailand	17	1.6 (0.8-2.7)	6.2 (0.48-19)	0.82 (0.22-1.8)	3.7 (0.97-9.6)	2.6 (0.1-15)	0.24 (0.01-2.1)	Kannan, Tanabe et al. 1995
Vietnam	19	10 (3.1-24)	26 (3.9-76)	1.8 (0.58-4)	0.29 (<0.1-1.1)	0.11 (<0.01-0.35)	0.05 (0.01-0.31)	Kannan, Tanabe et al. 1995
Indonesia	5	2.6 (2-3.8)	28 (0.66-76)	0.73 (0.06-1.4)	1.2 (<0.1-2.3)	0.45 (0.24-0.69)	0.05 (0.01-0.18)	Kannan, Tanabe et al. 1995
Papua New Guinea	13	7.5 (0.8-16)	0.43 (0.07-1.4)	0.57 (0.18-1.6)	1.3 (0.1-3.0)	0.37 (<0.01-2.1)	0.03 (<0.01-0.05)	Kannan, Tanabe et al. 1995
Solomon Islands	10	3.6 (0.66-15)	4.8 (0.91-24)	0.53 (0.23-1.9)	0.32 (0.1-1.6)	0.57 (0.11-1.6)	0.02 (0.01-0.06)	Kannan, Tanabe et al. 1995
Australia	37	55 (0.22-720)	22 (0.14-230)	0.34 (<0.01-2.1)	10 (0.12-55)	51 (0.06-720)	4.2 (<0.01-60)	Kannan, Tanabe et al. 1995

Table 3.2 Concentrations (ng g^{-1} wet weight) of Organochlorine Residues in Mussels in Southeast Asia.

Location	PCBs	DDTs	Chlordanes	HCHs	HCB	Reference
Hong Kong (1986)	49-330	50-520		53-100		Phillips 1989
Thailand (1994/5)	<0.01-20	1.2-38	0.25-5.9	<0.01-0.33	<0.01-0.12	Kan-atireklap, Tanabe et al. 1997
South India (1988/9)	0.66-7.1	2.8-40		4.3-16		Ramesh et al. 1990
Philippines (1994-1997)	0.69-36	0.19-4.2	0.15-9.5	<0.01-0.19	<0.01-0.04	Prudente, Ichihashi et al. 1999

PCBs

Many studies besides those given in table 3.1 are published in the scientific literature on levels of PCBs in fish. These studies are not given because differences in PCB analysis between studies makes comparison difficult.

A general downward trend in the levels of PCBs has been observed for fish from the Belgian Continental Shelf, inland waters of the Netherlands and the North Sea although some higher chlorinated PCB congeners have remained constant in the latter (Roose, Cooreman et al. 1998). No change was recorded for PCB levels in fish from the Arctic and Baltic (Paasivirta, Koistinen et al. 1994). Indeed, it has been noted that despite a few regional declines in PCB levels, a global decline is not expected in the next few years (Roose, Cooreman et al. 1998). In Sweden, it was estimated that intake of cod liver could result in an intake equivalent to the total guideline tolerable dose. Consequently, the sale of turbot liver and cod liver has been banned in Sweden for almost two decades (Vaz 1995).

PCB levels in one study on Mediterranean fish reported that levels of PCBs were lower here than other regions of the Northern Hemisphere (Sanchez, Sole et al. 1993). PCB levels in fish from tropical Southeast Asian countries were considered to be low (range 0.38 to 110 ng g^{-1} wet weight) and by comparison, levels in Australia and in cold waters of the Northern hemisphere are higher. Amongst the tropical Southeast Asian countries levels of PCBs in Vietnamese fish were slightly higher. This may be due to the use of PCBs in electrical equipment imported from Australia up to the mid-1980s and possibly from weapons used during the second Indo-Chinese war (1961-71). The lowest levels among Southeast Asian countries were reported for Cambodia (Monirith, Nakata et al. 1999).

In Australia, levels of PCBs in fish are high (range 0.22 to 720 ng g^{-1} wet weight). The wide range of values indicates that there are specific sites of contamination in Australia. Like Australia, fresh inputs of PCBs around the coasts of New Zealand have also been shown to occur. The high PCB levels in Australian fish are comparable to those found in fish from Japan, another industrialised nation. Given that PCBs are still present in old electrical equipment in Australia it is assumed that continued inputs of PCBs into the environment will continue in the future (Kannan, Tanabe et al. 1995).
Table 3.2 shows levels of PCBs in green mussels collected from coastal waters of Thailand, India, the Philippines and Hong Kong. As in tropical fish, levels of PCBs are generally low. Higher levels within countries are associated with industrial areas, for instance, relatively higher levels within the Philippines were found for mussels taken from Manila Bay (Prudente, Ichihashi et al. 1999). In the US, annual monitoring of levels of PCBs in mussels

between 1986 and 1996 revealed that levels of PCBs had essentially remained unchanged during this time period (O'Connor 1998).

DDT

DDT continues to pollute the marine environment even where it has been banned for several years due to its persistence and continued inputs from, for example, non-point source run off, tributary loads, dredged spoils and ocean dumped sewage sludges. In areas of Southeast Asia and South America the use of DDT continues with the consequent input into the marine environment. DDT compounds accumulate in bottom sediments of the seafloor and this is a major factor contributing to the continued contamination of fish inhabiting coastal ecosystems (Kennish and Ruppel 1996b). In recent years, studies have noted a decline in levels of DDT in fish from some regions (Kannan, Tanabe et al. 1995; Atuma, Linder et al. 1996; Kennish and Ruppel 1996b), but it is nevertheless, a persistent contaminant of fish world-wide.

Table 3.1 shows that levels of DDT were higher in fish from the Northern Hemisphere than those from the Southern Hemisphere (Kannan, Tanabe et al. 1995). Levels are consistently high from temperate regions in the northern Hemisphere to Arctic waters. Studies on contamination of fish in Sweden have reported that fish from the Baltic are more highly contaminated than from other Swedish waters (Atuma, Linder et al. 1996).

In the US, high levels of DDT in fish were reported in New Jersey between 1988-91, and bans on the sale of striped bass were in place between 1988-91 together with advisories on limited consumption of other fish (Kennish and Ruppel 1996b). Fish and shellfish from coastal waters of Morocco in the Mediterranean had higher levels of DDT than other organochlorines although the concentrations in this region were considered to be relatively low (El Hraiki, El Alami et al. 1994). In Southeast Asia, levels of DDT are lower by comparison with other areas, but are still considered to be elevated, suggesting the continued use of DDT in these regions. In Cambodia for example, levels in fish are low compared to other Southeast Asian countries, but even here, elevated levels in one region, Sihanouk Ville, imply recent input to the marine environment (Monirith, Nakata et al. 1999).

In the US, a study of levels of contaminants in mussels from coastal and estuarine waters in 1996 reported an annual median level of total DDT of 27.9 ng g^{-1} dry weight (O'Connor 1998). A general downward trend in the levels of DDT was observed from 1986 to 1996, the annual median concentration in 1986 being 37.07 ng g^{-1}. By contrast, levels of DDT in mussels from the Gulf of Mexico were higher (Sericano, Wade et al. 1990). For instance, total DDT in 1988 was 80 ng g^{-1} dry weight, range 2.4-1400 ng g^{-1}. DDTs were found in every sample analysed with a wide range of concentrations, indicating their continued availability to oysters. A review of historical data nevertheless implied that there was a declining trend in DDT levels. In France, levels of total DDT in mussels were similar in to the Gulf of Mexico in some areas (Claisse 1989). With the exception of the Seine estuary, levels were <100 ng g^{-1} dry weight along the French coastline in the English Channel and the Atlantic down to the Loire. However, they were more contaminated in the south, reaching a peak in the Arcachon basin (mean 233.8, range 24.3-1015 ng g^{-1}). Levels along the Mediterranean coast were also quite high (mean 151.9, range 7.4-733 ng g^{-1}). In the South West Baltic Sea, levels of DDT in mussels were lower (Lee, Kruse et al. 1996). Mean levels from different sites in 1990/1 ranged from 5.1 to 88.3 ng g^{-1} dry weight.

Levels of DDT found in green mussels in tropical Southeast Asia (see table 3.2) were within the range of levels found in the US and similar to levels in tropical fish. An exception is Hong Kong where levels found in mussels in 1986 were notably high. It is not known whether such high levels are presently incurred in this region. In Thailand, past studies show that levels of DDT appear to have fallen in recent years and this may reflect the ban on its agricultural use in 1983. Nevertheless, mussel samples taken in 1995 in Thailand had considerable DDT levels in some areas and may indicate significant current sources in these locations due to DDT use for malaria vector control (Kan-atireklap, Tanabe et al. 1997). The lowest levels of DDT in green mussels were reported for the Philippines. Here, the use of organochlorines for agricultural purposes was halted in 1977 and usage of DDT for malaria control was phased out in 1992 (Prudente, Ichihashi et al. 1999).

HCHs

Data presented in table 3.1 shows that levels of HCH in fish are consistently higher in fish from cold waters of the Northern Hemisphere than fish from Southeast Asia and Oceania. The exception was India where higher levels of HCH reflect the continued use of HCH in this country. HCH has, however, also been used in other Southeast Asian countries at least until the 1980s. The low level of HCH in fish tropical Southeast Asian countries is likely to be due to the rapid vaporisation of this compound in tropical latitudes (Kannan, Tanabe et al. 1995).

Within Southeast Asia and Oceania, levels of different isomers of HCH differed among countries. α-HCH was the predominant isomer in fish from India and Thailand, and β-HCH was predominant in Vietnam, Papua New Guinea and Australia. The predominance of α-HCH in India and Thailand suggests the use of technical grade HCH in these countries. In other countries, Indonesia and the Solomon Islands, fish contained higher amounts of γ-HCH which suggests the use of lindane in these areas. Indeed, reports have indicated the use of lindane in Indonesia, the Solomon Islands and Vietnam (Kannan, Tanabe et al. 1995).

No data were available on levels of HCHs from Mussel Watch schemes in the US, France and South America. In UK coastal waters, γ-HCH (lindane) was detectable in the common mussel at low concentrations (0.25-5.5 ng g^{-1} wet weight), (Marine Pollution Monitoring Management Group 1998). In Southeast Asia, as in tropical fish, levels of HCH in green mussels (see table X.2) were low except for India. In Thailand, γ-HCH was the most prevalent HCH isomer in some locations and this may suggest the continuing use of lindane in Thailand (Kan-atireklap, Tanabe et al. 1997).

Chlordane

Chlordane was once used mainly by industrialised countries of the Northern Hemisphere, but its use has spread southwards to tropical regions in recent years. Table 3.1 shows that levels of chlordane were generally higher in fish from the Northern Hemisphere than tropical regions of the Southern Hemisphere. Chlordane was detectable in herring from the northern Baltic (Strandberg, van Bavel et al. 1997), and a German study found that chlordane residues were present in those fish which had a high to moderate fat content (Karl, Lehmann et al. 1998).

The highest chlordane levels were observed for the US, a country where chlordane was widely used although since 1988 it has been banned (Kennish and Ruppel 1996a). Chlordane has been reported to be the predominant organochlorine pesticide in the aquatic environment in the USA, Japan and New Zealand. In Australia, levels of chlordane in fish from several regions of were comparable, except for Sydney where levels were higher, and suggest the widespread use of chlordane in this country. Levels of chlordane in Southeast Asian fish are considered to be low (Kannan, Tanabe et al. 1994). The low accumulation of chlordanes in tropical Southeast Asian countries is likely due to the volatile nature of chlordanes and their subsequent long-distance transport towards colder regions.

In the US, the annual median concentration of chlordane in mussels was reported to be 7.97 ppb dry weight (O'Connor 1998). A decreasing trend in levels was reported to have occurred since 1986, the median level at this time being 14.7 ppb dry weight. In tropical Southeast Asia, levels of chlordane in green mussels were low. A study in the Philippines noted that levels of chlordane were higher in mussels near urban locations compared to rural locations. This suggests that chlordane may be used partly against termites in more populated areas, similar to previous uses in Japan (Prudente, Ichihashi et al. 1999).

HCB, Aldrin and Dieldrin

Data on levels of HCB, aldrin and dieldrin in fish were more limited than for other organochlorine chemicals. HCB levels are considered to be low in fish from tropical Southeast Asian countries and were somewhat higher in Australia. HCB levels were also considered to be low in green mussels from Thailand (Ruangwises, Ruangwises et al. 1994; Kan-atireklap, Tanabe et al. 1997) and the Philippines (Prudente, Ichihashi et al. 1999). The presence of HCB in marine fishes may originate from pesticide formulations which contain HCB as an impurity, from incineration of municipal and industrial wastes and from various chlorination processes in which HCB is a by-product. HCB is likely to rapidly volatilise in tropical regions and hence, like other semi-volatile chemicals such as DDT and HCHs, be short-lived in tropical waters.

Levels of aldrin and dieldrin in fish from Southeast Asia and Oceania were considered to be low even though these compounds have been used in recent years to control crop pests in India, Pakistan, Thailand, Malaysia, Indonesia and the Philippines and to control termites in Australia (Kannan, Tanabe et al. 1995). A study of levels of organochlorines in benthic (bottom-dwelling) invertebrates from coastal areas of Thailand and Malaysia found that levels of dieldrin were higher in Jeram, Malaysia, than Ao Ban Don and Pattani Bay, Thailand (Everaarts, Bano et al. 1991). Another study in Thailand found that levels of aldrin (0.2-0.92) and dieldrin (0.08-0.47 ng/g wet weight) in green mussels from the Gulf of Thailand were within the range of levels found in fish from tropical Southeast Asia (Ruangwises, Ruangwises et al. 1994). These chemicals have been extensively used for termite control in Thailand, aldrin being used in greater amounts than dieldrin.

Dieldrin was reported to be detectable in mussels in UK coastal waters (Marine Pollution Monitoring Management Group 1998) and US coastal waters (O'Connor 1998). It was also found in herring from the northern Baltic (Strandberg, van Bavel et al. 1997).

Toxaphene

Toxaphene is a major organochlorine contaminant in both freshwater and marine fish and is present at concentrations similar to, or higher than, those of PCBs (Muir, Kidd et al. 1997). Data on levels in fish are however quite limited. Toxaphene has been found in North Sea fish (see Wells and de Boer 1999). A study of fish from Germany detected toxaphene in the majority of samples tested. The highest concentrations were found in fish with high to moderate fat content such as halibut, herring, redfish and mackerel (Alder, Beck et al. 1995). The study concluded that such fish can accumulate considerable amounts of toxaphene in the edible part. Indeed, it has been noted that the main route of intake of toxaphene in western Europe is expected to be through fish consumption, although little is known about toxaphene levels in other foods (Wells and de Boer 1999).

In arctic waters, toxaphene was a prominent organochlorine in Atlantic cod liver from northern Norway. Levels were similar to those found in the same species from the Baltic Sea during the 1980s (AMAP 1998).

PCDD/Fs

In marine and freshwater fish samples from all over the world, the predominant PCDD/F isomers are the toxic 2,3,7,8-substituted congeners. This includes 2,3,7,8-tetraCDD (TCDD) or dioxin, the most toxic of the congeners. Other PCDD/F congeners are also present (Rappe, Bergqvist et al. 1991). Consumption of fish and shellfish from contaminated areas may be an important source of human exposure to PCBs and PCDD/Fs. Due to the toxic nature of these compounds, high levels in fish and shellfish have resulted in the closure of fishing grounds. For instance, fishing areas in Howe Sound, northwest of Vancouver, Canada were closed in 1988 due to high concentrations of PCDD/Fs in crabs, prawns and shrimp. Some of the fishing areas were reopened in 1995 after levels in fish had fallen due to process changes implemented by nearby pulp mills to eliminate the production of PCDD/Fs (Starodub, Miller et al. 1995).

In an attempt to lessen levels of PCDD/Fs in shellfish from contaminated waters so they may be consumed by humans, experiments have been carried out in which oysters have been transplanted from contaminated to non-contaminated areas and left for some time. However, a study revealed that after contaminated oysters were transplanted to a cleaner area for 50 days, they still contained significant levels of PCDD/Fs (Gardinali, Sericano et al. 1995).

Levels of PCDD/Fs in Arctic waters have been found to be low in comparison to other areas. For instance, Atlantic cod near to Russeviknesset had levels of 0.02-0.04 pg/g TEQ wet weight (or ppt). Relatively low concentrations were found in mussels in the vicinity of a smelter in northern Norway (range from 0.61 pg/g wet weight near the smelter to 0.2 pg/g at a more distant site), (AMAP 1998). Another Norwegian study assessed levels of PCDD/Fs in herring from the Norwegian coast and North Sea and reported levels of 2.15 pg TEQ/g wet weight (Biseth, Oehme et al. 1990).

A Finnish study detected an average concentration of PCDD/Fs in Baltic herring of 0.94 pg/g wet weight (Vartiainen and Hallikainen 1992). Another study found levels in Baltic herring of about 1 pg TEQ /g wet weight, and in the Gulf of Finland, concentrations were clearly higher (3.6 pg TEQ /g), (Korhonen and Vartiainen 1997). Based on results of a scientific study, the Finnish Food Board have recommended that herring which is over 3 years old (greater than 17 cm in length) should not be consumed because the TDI for

PCDD/Fs could be exceeded. Herring older/larger than the recommendation may contain 10-20 pg TEQ /g (Hallikainen 1997). A study in the southern part of the Baltic Sea also found relatively low levels of PCDD/Fs in fish and shellfish (Falandysz, Florek et al. 1997). The study suggested that PCDD/F contamination in fish in this area was probably from atmospheric input and there was no indication of important local sources of emission in the western part of the Gulf of Gdansk. However, higher levels of PCDD/Fs have been found in Baltic herring and salmon from the south coast of Sweden. Samples of herring had concentrations of 8-18 pg TEQ (Nordic) whole fish as compared to 2-3 pg TEQ/g in herring from the less polluted waters of the west coast of Sweden. Wild salmon had concentrations of 30-90 pg TEQ/g. A study on fish consumers from southern Sweden found they had significantly higher levels of PCDD/Fs in their blood than non-consumers. The study concluded that fish is a major source of exposure to PCDD/Fs in the fish-eating population around the Baltic Sea (Svensson, Nilsson et al. 1990). A later study also found that levels of DDT, DDE and PCBs were associated with fish consumption such that higher levels were found in blood of high fish consumers from southeast Sweden (Asplund, Svensson et al. 1994). The study concluded that fish from the Baltic Sea was a major source of exposure to these compounds.

Levels of PCDD/Fs in fish and shellfish from sites near to industrialised countries can be high. Striped bass, crabs and lobsters collected in Newark Bay and the New York Bight, USA were highly contaminated (Rappe, Bergqvist et al. 1991). High PCDD/F levels in fish most likely originated from their feeding in a nearby polluted river and in the shellfish from a nearby ocean waste disposal site for sewage sludge, fly ash and dredge spoils. Levels of TCDD in crab meat were about 100 ppt but a value exceeding 6000 ppt was found in a sample of crab hepatopancreas. This is thought to be the highest level ever reported in food from the aquatic environment. It is of note that a number of American ethnic groups as well as other ethnic groups around the world such as some Scandinavians, eat the hepatopancreas of crabs in addition to the meat. The study on PCDD/Fs in fish from Newark Bay and New York Bight commented that the wide range of congeners present in besides TCDD in the fish increases the relative risk associated with the consumption of these animals.

A study on human intake of PCDD/Fs from contaminated crabs was conducted on the Frierfjord in southern Norway (Becher, Johansen et al. 1995; Johansen, Alexander et al. 1996) A magnesium factory has discharged PCBs and PCDD/Fs into the area for the past 35 years and levels in marine biota exceed those from diffuse polluted areas by a factor of 2 to 40. Crabs are a popular food item for some local residents. The study revealed that consumption of crabs correlated with human blood concentration of PCDD/Fs such that people who had a high intake a crabs had significantly (5-fold) higher blood levels. In addition, the PCDD/F profile in the blood of people who had a high intake of crab was changed to resemble the PCDD/F profile in the crabs. This evidence shows that consumption of contaminated crabs in this area is an important source of exposure to PCDD/Fs. Almost all the individuals in the high-intake group exceeded the tolerable weekly intake (TWI) of 35 pg TEQ kg^{-1} body weight $week^{-1}$ proposed by a Nordic Expert Group and a few exceeded the WHO TWI of 70 pg TEQ kg^{-1} body weight $week^{-1}$.

A study on levels of PCDD/Fs in the edible portion of 6 species of fish from Tokyo Bay, Japan detected concentrations ranging from 6.5 - 106 pg/g wet weight or 0.32 - 2.07 pg TEQ/g wet weight. Shellfish and crabs had considerably higher levels (178.57 - 1025.77

pg/g wet weight or 2.56 to 3.56 pg TEQ/g wet weight). Based on the values for fish, if 100g per day were consumed, which is common in Japan, the daily intake would be 0.53 - 3.4 pg TEQ kg^{-1} body weight day^{-1} (Masunaga, Kim et al. 1997). This alone, without considering additions from PCBs in the fish or PCDD/Fs from other food stuffs, is within the range of the WHO TDI (1 - 4 pg TEQ kg^{-1} bw day^{-1}) for PCDD/Fs and dioxins-like PCBs in food.

Another study on PCDD/Fs in shellfishes taken from seas around Japan and Korea showed that levels from both countries are in the same range, suggesting a similar situation for both countries (Hashimoto et al. 1998). For example, levels of total PCDDs and PCDFs in shellfish from Tokyo Bay, Osaka Bay and Kii Channel in 1993 ranged from 25 to 97 pg/g wet weight and levels in oysters from several locations in Korea in 1996/7 ranged from 7.5 to 37.0 pg/g wet weight. Levels of PCDD/Fs for shellfish from Korea were higher in industrial areas compared to rural areas which could be due to the impact of industrialisation. Nevertheless, PCDD/Fs were also present in shellfish from more rural areas which implied that long distance transport of dioxins was also contributing to pollution in these marine animals. Congener profiles suggested that combustion sources, for example incineration, were possibly responsible for the occurrence of PCDD/Fs in the shellfish from both rural and urban areas of Korea. Similarly, a previous study in Japan on dioxins in blue mussels from Osaka Bay in 1987 indicated that municipal incinerators may be the main contaminant source of dioxins in these shellfish (Miyata et al. 1987a, 1987b). A study on foods in the US found relatively high levels in Spanish mackerel from the Gulf of Mexico (0.72 pg TEQ/g) but lower levels in mullet (0.027-0.089 pg TEQ/g), (Cooper, Fiedler et al. 1995).

A study in Australia on mussels from five sites along the Ninety Mile Beach, Victoria in 1992/4 detected PCDD/Fs in these shellfish (al. 1995). The direct discharge of industrial, domestic and paper mill effluent into the area Haynes et was not found to affect the levels of dioxins in mussels. It was therefore concluded that the discharge of effluent has negligible impact on the levels of dioxins in mussels.

3.1.2 Freshwater Fish

No surveys of levels of organochlorines in freshwater fish covering more than one country were located in the scientific literature. A comparison of levels of organochlorines in freshwater fish world-wide is therefore limited to compiling information from studies in individual countries. This has the disadvantage of comparisons being made between many different fish species. The concentrations of organochlorines in fish can be partly dependent on the species, for instance some fatty fish accumulate higher levels of organochlorines than other fish. Published studies often investigate particularly contaminated areas or very remote areas, both of which may not be representative of other areas in a country. In this context, together with the limited number of studies available in the scientific literature, it is difficult to portray an accurate comparison of organochlorine levels in fish between different countries.

It is clear, however, that levels of organochlorines in fish exceed recommended limits in many places. In addition, some population groups are highly exposed to organochlorines in fish because they have a high fish intake. For instance, native peoples in the Pacific Northwest of the US and Canada have a higher fish consumption than the general

population. Fish consumption among recreational anglers is variable but a small fraction of them eat large amounts of fish (Ebert, Price et al. 1996). A study on anglers in Wisconsin found that increased fish consumption correlated with increased PCB concentrations in their blood (Sonzogni, Maack et al. 1991).

Table 3.3 presents data from different studies on levels of organochlorines detected in freshwater fish from various countries. Concentrations of organochlorines in fish from arctic or sub-arctic regions of Finland (Paasivirta, Koistinen et al. 1994), Russia and Canada (AMAP 1998) are somewhat lower than other regions of the world. By comparison, levels of PCBs and DDTs in a river in the Baltic region are far higher and it has been noted that levels of POPs in salmon in this region may cause ecotoxic damage (Paasivirta, Koistinen et al. 1994). Lower concentrations of toxaphene, HCB and chlordanes measured in Finnish Arctic and Baltic Salmon indicate that these compounds are dispersed in long-distance transport processes more rapidly than the other organochlorines monitored in the study (Paasivirta, Koistinen et al. 1994). Studies on remote lakes in the Pyrenees, Spain (Sanchez, Sole et al. 1993) and Canada (Swackhamer and Hites 1988) found considerable levels of contamination in fish even though the only source of organochlorines to these lakes was atmospheric transport.

In rivers of the Catalonia region in Spain, most observed levels of organochlorines in fish fell within those recommended for human consumption (López-Martín, Ruizolmo et al. 1995). However, heptachlor epoxide concentrations often exceeded limits, in particular in the lower reaches of the River Ebro and River Ter. The study commented that PCB and DDE concentrations fell within the range of levels previously reported in other areas of western Europe. An exception to this was rivers in southern Italy which were reported to have higher organochlorine pesticide levels than those reported for Spain, and an equally wide distribution of heptachlor epoxide levels. A study on fish farms in Leon, Spain, in 1993, found almost all samples taken were contaminated by organochlorines (Sahagún, Terán et al. 1997). The most frequently detected compounds were lindane, heptachlor epoxide and DDT. The highest DDT concentrations in brain tissue at four fish farms ranged from 628-1533ppb. The study noted that organochlorines in farmed trout had declined since 1987.

The highest levels of organochlorines recorded from various studies presented in table 3.3 were in fish from the Lake Michigan, USA. Despite high concentrations of organochlorines persisting in Lake Michigan fish in the 1990s, levels have declined from levels during the 1970s and 1980s. A study on toxaphene in fish from the Great Lakes in 1992 also found that levels had declined since 1982 in all Lakes apart from Lake Superior where concentrations remained the same (Glassmeyer, Myers et al. 1996).

Levels of chlordane in freshwater fish from New Jersey, US were quite high (Kennish and Ruppel 1996a). It was noted that during the 1986-1987 sampling period that the mean concentration of chlordane in carp exceeded the FDA action level of 300 g kg^{-1} wet weight.

It was recommended that regular monitoring of chlordane levels in finfish and in shellfish should continue in the region to reduce the public's risk of chlordane contamination. A study of levels of organochlorines in wild and pond-raised crayfish in Louisiana found relatively low contamination in these animals (Madden, Finerty et al. 1989).

In Australia, a study on the Georges and Cooks rivers showed that they were highly contaminated with PCBs and DDTs (Roach and Runcie 1998). DDT was not above the National Food Authority Maximum Residue Limit (MRL), but at some locations, PCBs and chlordane were significantly greater than the MRL. The study commented that fishers who catch sea mullet, yellowfin bream, silver biddy or pink eyed mullet from the contaminated areas samples in the study may exceed provisional tolerable weekly intake and may have an increased risk of inducing various human health effects associated with these substances. At Coff's Creek on the north coast of New South Wales, testing of fish between 1986 and 1989 revealed high levels of dieldrin in the fish (183-473 ng/g), (McDougall and Bettmann 1990). Most of the fish tested had levels above the MRL (100 ng/g). In New Zealand, little information exists on organochlorine contaminants in the aquatic ecosystem. A study on freshwater mussels from the Waikato River found that mussel tissue concentrations did not exceed levels considered safe for human consumption , and levels were low compared with freshwater mussel data for industrially contaminated freshwater sites in other countries (Hickey, Buckland et al. 1997).

There are several instances recorded in the scientific literature which indicate that recommendations have been given by local authorities which either ban or advise fishermen to consume less fish from contaminated areas. For instance, in Newark Bay, US, advisories have been given to guide citizens on safe consumption practices for certain fish and crabs from the area. However, the success of advisory information being taken on by the public may sometimes be questionable. A recent survey of fishermen in the area found that around 60% of them had heard of the advisories. Unfortunately, most did not believe them or were unconcerned about health effects from eating contaminated species (Pflugh, Lurig et al. 1999). At Akwesasne on the St. Lawrence River, a study has been undertaken on fish consumption in a Native American Mohawk community. Fish from the river are contaminated by PCBs and Mohawk women were found to consume more fish than other women in the area. Advisories on potential health effects from fish consumption have been issued. It was found that there has been a general decrease of fish consumption by Mohawk mothers, most likely as a result of local health advisories. However, changes in traditional lifestyles which have occurred as a result of environmental contamination, have detrimentally affected the community socially, culturally, and economically. Also, fish is high in protein and substitution in the diet with nutritionally less desirable foods may lead to other health impacts (Fitzgerald, Hwang et al. 1995).

PCDD/Fs and PCBs

Studies on levels of PCDD/Fs in fish from different countries of the world appear to be limited in number. Table 3.4 gives levels of PCDD/Fs, and in some cases PCBs, in fish from different countries published in the scientific literature. A study in Finland observed quite low levels of PCDD/Fs in various species of fish from the Kymijoki River (Korhonen and Vartiainen 1997). Levels in salmon were greater those in other fish due to the higher fat content of these fish. Levels in fish liver (up to 122 TEQ pg/g) and spawn (up to 22 TEQ pg/g) were much higher than in fish muscle, again due to a higher fat content. In South Germany, most of the freshwater fish that is consumed is produced in fish cultures. A study on levels of PCDD/Fs in rainbow trout and carp found they were low (Mayer 1995b).

In the Netherlands, a landfill in the Volgermeerpolder, a grazing land area, just north of Amsterdam was found to be heavily contaminated with dioxins in the early 1980s as a result of massive chemical waste dumping. Tests in 1994, revealed that eel from the area still have

similarly high concentrations (91.9 pg TEQ/g wet weight) to those found in 1981/4 and eel consumption from the region has been banned (Heide, van der Oost et al. 1995).

The St. Lawrence river in Canada is known to be contaminated with a range of persistent organochlorines derived from the Great Lakes and from several industrial and municipal effluents along the river. Table 3.4 shows that levels of PCDD/Fs and PCBs in some fish from the river were very high. Levels in Chinook Salmon were 10 times higher than proposed acceptable levels for human consumption by the Ministry of Environment of Quebec and Ontario and levels in eel were 5 times higher (Brochu, Hamelin et al. 1995).

A study on fish collected from Lake Ontario between 1989 and 1994 found levels of TCDD in fish were variable but mainly between 10 and 20 ppt (Reiner, MacPherson et al. 1995). The study suggested that from the available limited, levels in fish appear to be decreasing though at a slower rate than during the 1980s. High levels of PCDD/Fs have been measured in fish from the Fraser and Thompson Rivers in British Colombia that are exposed to effluent from bleached kraft pulp mills. One study estimated that individuals who consume one pound of mountain whitefish per month from some areas could potentially exceed the USA TDI for PCDD/Fs (Law and Gudaitis 1994). Research on farm raised catfish from Mississippi, Alabama, and Arkansas in the Southeast US found significant levels of PCDD/Fs and PCBs in the fish (9.5-43 TEQ pg/g lipid). The source of the high contamination appeared to come from the catfish feed (Cooper, Bergek et al. 1996).

Milkfish taken from culture ponds in southern Taiwan near to open air waste incineration sites were found to have very high levels of coplanar PCBs (Lu, Miyata et al. 1995a). Total TEQs in these fish and fish from a market ranged from 0.195 to 153 pg/g. The study noted that the highest level detected in fish from the culture ponds was equivalent to 278 times greater than the average (0.55 pg/g) of Japanese market fish samples. The culture milkfish in this area of southern Taiwan is an important source of protein for residents. Some of the fish would greatly exceed the WHO TDI for PCDD/Fs plus PCBs. Fish from the Er-Jen river in Taiwan were also found to contain significant levels of PCDD/Fs and coplanar PCBs (Ling, Soong et al. 1995). The study calculated a daily intake value of 28 to 450 pg kg^{-1} day^{-1} for consumption of fish from the river. This greatly exceeds the WHO TDI value of 1-4 pg kg^{-1} day^{-1}. In Japan, a study a fish collected from Lake Kasumigaura found that levels of PCDD/Fs in the fish were relatively low (0.83-2.64 TEQ pg/g), (Kim, Sakurai et al. 1995). Levels were close to those reported for coastal fish in Japan.

Table 3.3 Mean Concentrations of Organochlorines (ng g^{-1} (ppb) wet weight) in Freshwater Fish

Location	number of samples/species (where given)	PCBs	DDTs	HCHs	Aldrin and Dieldrin	Chlordane	HCB	Reference
Finland								
Teno River (Arctic) 1990	15 (Salmon)	15.3	8.4	-	-	4.5	1.3	Paasivirta, Koistinen et al. 1994
Simo River (Baltic) 1988-92	73 (Salmon)	241.1	299.2	-	-	17.0	7.83	
Russia	1 (Pidschian)	2.5	0.86	0.39	-	0.66	-	AMAP 1998
Ob River								
Canada		2.17	1.00	1.97	0.1	1.62	-	AMAP 1998
Cambell	4 (Broad							

Lake, and,	whitefish) 4							
Travaillant Lake, Mackenzie Delta		1.86	0.31	0.46	0.08	0.67		
Spain								
rivers of Catalonia 1990-92		181	81	21	-	-	-	López-Martín, Ruizolmo et al. 1995
USA								
Lake Michigan, Wisconsin (1990)	29 lake trout	2440	1830	-	130	320	-	Miller, Kassulke et al. 1993
Camden and, New Jersey	1 (carp)	-	-	-	-	275	-	Kennish and Ruppel 1996a
Northeast New Jersey	1 (carp)	-	-	-	-	149		
Egypt								
Maryut Lake (1987)	*Tilipia zillii*	21.9	39.6	-	7.8	-	-	see Roach and Runcie 1998
Kenya								
Athi River (1987)	Catfish	-	31-41.7	-	-	-	-	see Roach and Runcie 1998

South Africa								
Wilderness lakes (1987)	Kob	-	10	-	-	-	-	see Roach and Runcie 1998
	Groovy mullet	-	40-50	-	-	-	-	
Australia								
Prospect Creek and	11 (*Gerres subfasciatus)*	5514	912	-	nd	62	nd	Roach and Runcie 1998
Homebush Bay, Georges/Cooks Rivers	11 (*Mugil cephalus)*	3782	1159	-	97	87	25	
Richmond River	Flathead	97.1	2.0	-	3.7	1.4	-	see Roach and Runcie 1998

Table 3.4 Concentrations of PCDD/Fs (TEQ ppt) in Fish Muscle/Edible Portions of Fish from Various Countries

Location and Year	Number of Samples and Species	Total PCDD/F ITEQ pg g^{-1} (ppt) wet weight	Reference
Finland	various species	0.3 - 7.1	Korhonen and Vartiainen 1997
Kymijoki River			
South Germany	rainbow trout	0.34	Mayer 1995b
Fish farms	carp	0.73	
Canada		(all values include	Brochu, Hamelin et al. 1995
St Lawrence River	14 Brown trout	PCBs)	
	8 Rainbow trout	23.5	
	6 Copper Redhorse	8.6	
	7 Chinook Salmon	12.4	
	American eel	185	
		75.7	
SE USA	Catfish		
fish farm		9.5-43 pg/g lipid	
Taiwan			Lu, Miyata et al. 1995a
Wan-Li, Culture Pond (1993)	13 Milkfish	0.195 - 153 (includes PCBs)	
Japan			Kim, Sakurai et al. 1995
Lake Kasumigaura (1994)	shrimp, pond smelt, piscivorous chub	0.83 - 2.64	

3.1.3 Fish Oils

Fish oils are an important commodity, accounting for around 2% of the total world annual production of oils and fats currently estimated at 80 million tonnes. The largest use of fish oils, principally in Europe, is in the partially hydrogenated form in the baking industry although it is also used in a variety of other applications (Jacobs, Johnston et al. 1997). An important component of the market is commanded by food grade and pharmaceutical grade nutritional supplements, estimated at around £50 million in the UK alone in 1992.

Fish oil in addition is used as a raw material and ingredient in foodstuffs intended for human consumption such as margarine and ice cream and is also added to animal feedstuffs. Total UK production and use of fish oils in the UK in 1992 was estimated at 108.6 kilotonnes.

Analysis of fish oils has revealed that they are significantly contaminated with a variety of persistent organic pollutants (Jacobs, Santillo et al. 1998). The data in Table 3.5 were obtained from a selection of pharmaceutical, food and industrial grade fish oils. The pharmaceutical and food grade oils were obtained directly from retail outlets in a number of countries and included both fish liver oils and whole fish-body oils.

The results obtained from these analyses taken together with recommended consumption figures indicate that fish oils could contribute a significant proportion of the daily intake of PCBs when taken as dietary supplements. They also have the potential to contribute to daily intakes through their incorporation into other food products. Although this contribution is difficult to quantify, in the case of animal feeds, this can be measured in terms of the residues present in meat and dairy products.

There is also a possible contribution by fish oils of persistent organic pollutants to the residues found in fish produced in aquaculture operations. Sample 30 in Table 3.5 below was a salmon oil marketed in the UK and originating from aquaculture operations in Norwegian waters. The extremely high residues of PCBs found in this sample are thought to have arisen as a result of industrial grade fish oils present in the feed and from the practice of feeding discarded portions of processed fish back into the fish rearing pens.

Table 3.5: Concentrations of organochlorine contaminants (μg l^{-1}) in fish-oils obtained from various sources.

Sample	Origin	Grade	Type	HCB	a-HCH	g-HCH	p,p'-DDE	p,p'-TDE	p,p'-DDT	Total DI	?PCBs
1	Australia	F	FO	-	-	-	10	-	-	10	-
2	Australia	F	CLE	3	4	-	9	-	-	9	10
3	Australia	F	CL	19	13	-	91	122	-	213	369
4	Australia	F	CL	14	18	5	91	155	-	246	159
5	Australia	F	CL	-	-	-	4	-	-	4	-
6	Austria	P	FO	-	-	-	39	-	-	39	17
7	Austria	P	FO	-	-	-	5	-	-	5	-
8	Austria	P	FO	-	-	-	-	-	-	-	-
9	Austria	P	FO	-	-	-	-	-	-	-	6
10	Belgium	F	FO	7	23	4	13	-	-	13	22
11	Belgium	F	FO	15	10	4	97	332	492	921	805
12	Belgium	F	FO	5	-	-	28	-	-	28	40
13	Belgium	F	FO	3	-	-	26	-	-	-	228
14	Brazil	P	FO	19	7	-	141	182	-	323	346
15	Canada	F	CL	-	-	-	33	68	-	101	109
16	Canada	F	CL	-	-	-	18	-	-	18	43
17	France	F	FO	-	-	4	-	-	-	-	-
18	Germany	U	HO	35	63	13	106	117	-	223	525
19	Japan	F	FO	-	6	-	62	-	33	95	313
20	Japan	F	FO	-	-	-	2	-	-	2	-
21	NL	F	FO	4	-	-	15	-	-	15	73
22	NZ	F	CL	45	28	11	254	357	133	744	619
23	NZ	F	CL	-	-	-	14	-	-	14	-
24	Norway	P	FO	-	-	-	45	29	74	148	570
25	Norway	P	FO	-	-	-	30	19	14	63	440

26	Norway	P	CL	-	-	-	2	-	-	2	-
27	Spain	F	CL	18	93	-	59	10	-	69	261
28	Spain	F	CL	-	-	-	22	6	12	40	212
29	UK	F	CL	21	9	3	56	44	19	119	990
30	UK	P	SO	46	8	9	87	53	-	140	1132
31	UK	F	CL	-	-	-	28	27	7	62	428
32	UK	P	MLC	-	-	-	4	-	-	4	10
33	UK	F	HL	-	-	-	3	-	-	3	37
34	UK	F	CL	10	12	3	60	48	29	137	1055
35	UK	P	CL	-	7	-	6	-	-	6	14
36	UK	F	CL	10	12	-	60	47	31	138	1050
37	UK	P	MLC	-	-	-	-	-	-	-	-
38	UK	P	CFO	-	-	-	35	23	-	58	915
39	UK	F	LO	-	-	13	-	-	-	-	-
40	USA	F	CL	39	7	6	161	219	-	380	717
41	Iceland	U	FO	14	36	3	11	-	7	18	366
42	Germany	U	?	21	33	20	56	51	30	137	939
43	Germany	U	RF	24	18	-	33	37	29	99	1106
44	Germany	U	SE	8	14	11	14	23	-	37	463
45	UK	V	CL	10	26	-	9	-	-	9	183
LIMIT OF DETECTION				3	3	2	1	4	5	N/A	5

Footnotes: P: pharmaceutical grade; F: food grade; U: oil for industrial applications V: veterinary grade. (-): not detected. N/A: detection limit not applicable (Total DDTs calculated as sum of DDT, DDE & TDE); FO: Fish oil; CL: Cod liver oil; CLE: cod liver oil emulsion; SE: Sandeel; RF: Redfish Oil (Probably No: 2 cod liver oil); MLC: Marine Lipid Concentrate; HL: Halibut liver oil; CFO: Fish oil concentrate; SO: Salmon oil; LO: Linseed oil. Descriptions are those given by the manufacturer/supplier.

The results of these analyses were broadly confirmed by analyses conducted by the UK Ministry of Agriculture Fisheries and Food (MAFF 1997b) which considered both PCBs and the polychlorinated dioxins and dibenzofurans. This study confirmed earlier results showing these oils to have relatively high concentrations of persistent organic pollutants.

When expressed as TEQs the (MAFF 1997b) data showed the combined concentrations of dioxins and PCBs in cod liver oils to range between 7.4 and 33 ng TEQ kg^{-1} of oil in 1994 and between 18 and 41 ng TEQ kg^{-1} oil in the 1996 survey. From the 1996 survey data, the maximum estimated intakes of dioxins and PCBs were in the range of 6.6-9.1 pg TEQ kg^{-1} body weight day^{-1} for adults and schoolchildren, 12-16 pg kg^{-1} body weight day^{-1} for toddlers and 11 pg TEQ kg^{-1} body weight day^{-1} for breast fed infants. This suggested that the consumption of some bottled fish oils could result in excedance of the then TDI of 10 pg TEQ kg^{-1} body weight day^{-1} in some sectors of the population. Subsequently the TDI has been revised by the WHO to 1-4 pg TEQ per kg body weight per day. Hence the potential for excedance of the TDI has increased significantly.

Typical intakes were also estimated accounting for variation between different lots of oil with a concentration of 31 ng TEQ kg^{-1} of oil. In this case the contributions fell in the range of 4.8-6.5 pg TEQ kg^{-1} body weight day^{-1} for adults and schoolchildren, 8.6-11.6 pg TEQ kg^{-1} body weight day^{-1} and 8.2 pg TEQ kg^{-1} body weight day^{-1} for breast fed infants. This is clearly a substantial potential contribution to dietary intake of these chemicals and in combination with other dietary components could be expected to contribute to the excedance of the TDI.

3.2 Meat

A number of studies have been undertaken, predominantly in western Europe and North America, to determine whether residues of persistent organic pollutants in meat and meat products are present at concentrations likely to be deleterious to human health. Figure 3.1 is a diagrammatic representation of the pathways by which the PCDD/Fs and PCB compounds move from their source into meat, although the pathway is relevant to the majority of the 12 UNEP POPs. Aerial deposition, whether through the particle or the vapour phase, is the main route by which these compounds move from source onto vegetation and onto soil. Vegetation can become contaminated while growing in the open or post-harvest, during storage. Once consumed the compounds may then move across the gastro-intestinal mucosa into the tissues of the animal. A summary of the residues found in meat from various countries is given in Table 3.7.

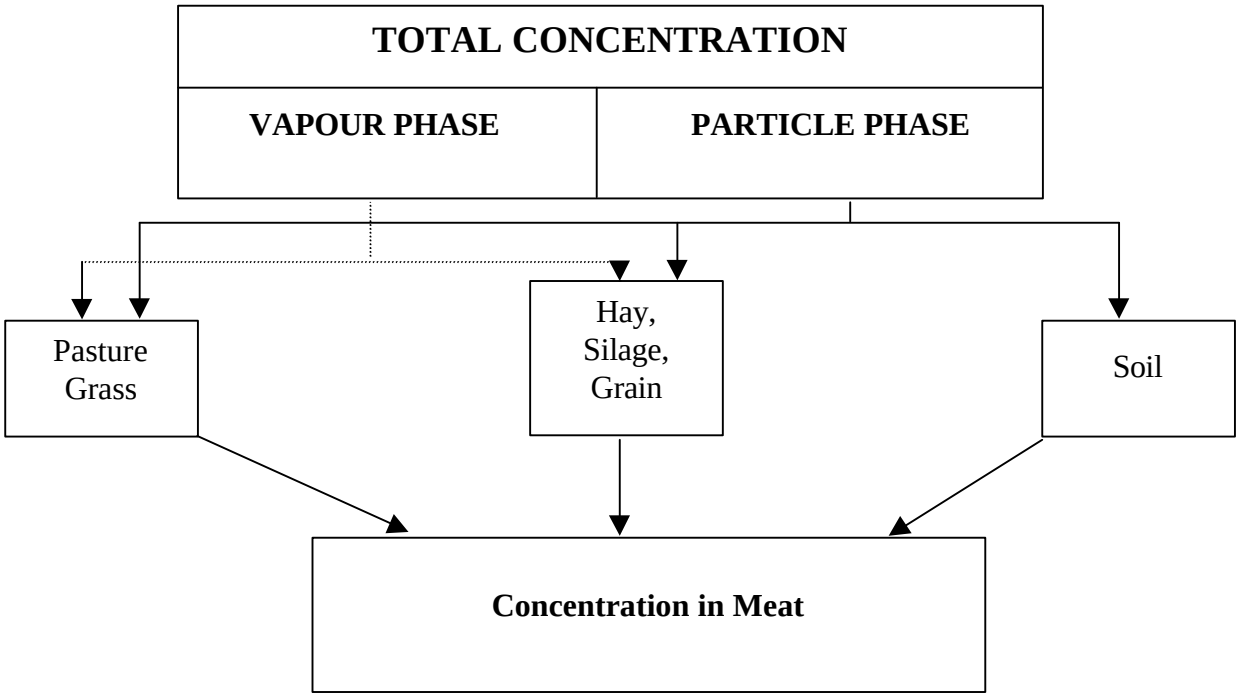


Fig 3.1: Model for the movement of PCDD/Fs and PCBs from source to meat (adapted from Lorber, Cleverly et al. 1994).

One of the main human and animal health concerns with regard to persistent organic compounds is their long depuration time (i.e. the time it takes for the compounds to be metabolised and/or excreted from the tissues). Chronic exposure to organochlorines through food is a global phenomenon for animals as well as humans and residues can build over a lifetime. There are, however, exceptions. The excretion of lipophilic compounds through mammalian milk can result in significantly increased residues in very young or recently weaned animals (Ariño, Herrera et al. 1993b). The consumption of milk fed veal, and of young lamb, may, therefore, result in increased human exposure when compared with the consumption of older carcasses. Concomitantly, lactating animals may exhibit lower tissue residues of organochlorines than males or non-lactating females (Ariño, Herrera et al. 1993b).

Table 3.6: FAO/WHO MRLs (µg kg^{-1} fat) for Meat

Chemical	Meat (generic)	Poultry
Aldrin & Dieldrin	200	500
Chlordane	50	500
DDT	5000	-
Endrin	100	1000
Heptachlor	200	200
Lindane	2000	700

Source: Joint FAO/WHO Food Standards Programme Codex Alimentarius Commission 1993.

Table 3.7: Organochlorine Residues in meat (μg kg^{-1} fat) in Various Countries.

Location	Chemicals						References
	PCBs	DDTs	HCHs	Aldrin & Dieldrin	HCB	Hepta-chlors	
Australia	11[(a)]				<0.0 1-37		Wells and de Boer 1999
		52	7.2	2.9-42	1.6	1.1-26	Kannan, Tanabe et al. 1994
India	<10-40						Wells and de Boer 1999
	210	1000	2200	130-240	4.8	4.0	Kannan, Tanabe et al. 1992b
Indonesia	2.9-4.5						Wells and de Boer 1999
Netherlands	1.6-25						Wells and de Boer 1999
Papua New Guinea	45-124						Wells and de Boer 1999
Thailand	2.3-12						Wells and de Boer 1999
South Korea	2.2-3.7						Wells and de Boer 1999
Solomon Islands	5.2-17						Wells and de Boer 1999
United Kingdom	0.27-0.96						Wells and de Boer 1999
Vietnam	480	3200	280	8.7-41	2.4	1.3-8.8	Kannan, Tanabe et al. 1992a

As meat is rarely eaten raw, some studies have attempted to determine whether cooking or curing can reduce organochlorine residues. The results of these studies seem to indicate that cured meat (involving fermentation or microbiological action) may have lower residues of lindane and α-HCH than uncured meat, but that these residues do not diminish appreciably with normal cooking (Ariño, Herrera et al. 1993a). It has also been suggested that there might be a considerable reduction of PCDD/Fs in meat during cooking but that this was primarily accounted for through fat loss (Thorpe, Kelly et al. 1999). In contrast, smoked meat may be highly contaminated with PCDD/Fs at the surface. Some smoked meat products are regional specialities (e.g. black smoked ham in southern Germany) and consuming large amounts of these products may lead to a significant increase of dietary PCDD/F intake (Mayer 1998).

As organochlorine compounds are highly lipophilic, fatty animal foods are one of the prime routes of exposure for the human population to POPs used in farming as crop sprays, or for vector control against such diseases as malaria. People who consume large amounts of meat and dairy produce are, therefore, likely to be exposed to higher levels of these pollutants than other consumers (Waliszewski, Pardio et al. 1997b). Although many countries have now decided to control the use of organochlorine compounds either by following the FAO/WHO guidelines or by the instigation of national controls, there continue to be cases where these guidelines are exceeded or ignored.

Table 3.8: PCDD/Fs and PCB I-TEQ values (pg g^{-1} fat) found in Meat in Various Countries.

Location	Meat	References
Canada	0.19-0.56	Wells and de Boer 1999
Germany	1.2-7.2[(a)]	Wells and de Boer 1999
	0.3-2.7	Wesp, Rippen et al. 1996
	0.9-2.6[(a)]	Wells and de Boer 1999
Italy	0.001-0.07	Zanotto, Alcock et al. 1999
Netherlands	0.43-14	Wells and de Boer 1999
Norway	0.33	Wells and de Boer 1999
Russia	0.2-5	Wells and de Boer 1999
Sweden	0.76-0.86	Wells and de Boer 1999
United Kingdom	0.21	Wells and de Boer 1999
USA	0.29-0.89	Wells and de Boer 1999
	0.53-1.10	Wells and de Boer 1999

DDT

According to Waliszewski, Pardio et al. (1997) Mexico is the leading consumer of DDT in Latin America, with approximately 3,000 tons yr^{-1} being used in that country's anti-malaria campaign alone. DDT and/or its metabolites were detected in 100% of meat samples analysed. Mean total DDTs amounted to 2,545 μg kg^{-1} (Waliszewski, Pardio et al. 1997b). The latest recommendations from the Codex Alimentarius Commission have introduced a reduced MRL of 500 μg kg^{-1} fat (FAO/WHO 1998). The Mexican meat samples, therefore, exhibited a mean concentration in excess of 5 times the Codex MRL.

Similarly in Thailand, residues of DDT were found in livestock samples, despite that country's ban on DDT which came into effect in 1983. Residues in chicken and pork fat were higher than expected. Of greater concern was the high proportion of p-p'-DDT in the meat, suggesting a current source of contamination. It was considered that illegal spraying of cattle sheds for malaria control could be the source (Tanabe, Kannan et al. 1991). Even so, residues did not exceed the WHO/FAO MRL of 500 μg kg^{-1} fat (FAO/WHO 1998). In nearby Vietnam residues of DDT in meat were greatly in excess of the current Codex Alimentarius MRL (500 μg kg^{-1} fat) (FAO/WHO 1998) (See Table 3.7). Details of production and consumption of DDT in Vietnam were not known at the time of the study, but the fact that the majority of the residues were p-p'-DDT rather than the p-p'-DDE metabolite suggested continued usage. High livestock contamination would point to the use of DDT for spraying cattle sheds (Kannan, Tanabe et al. 1992a).

Although figures for pesticide residues in game animals are not given in the tables, some data are available. A study undertaken in Germany in the 1980s that examined pesticide residues in pheasant, partridges and hares taken from an agricultural region of Germany, with no nearby heavy industry, found detectable levels of DDT in the fat from all the pheasants (mean concentration 160 μg kg^{-1}), most of the hares (mean concentration 668 μg kg^{-1}) and approximately 50% of partridges (mean concentration 527 μg kg^{-1}). The highest residues of DDT were found in woodcock (2165 μg kg^{-1}) a mean concentration greatly in excess of the then FAO/WHO MRL of 1000 μg kg^{-1}. As woodcock is a migratory species, however, it was not possible to determine the source of the contamination (Brunn, Berlich et al. 1985).

The Canadian province, Ontario, has been running a monitoring scheme for organochlorine residues in meat and eggs since 1969 and has found declining levels of DDT, TDE and DDE in samples tested since that time. By 1990 only DDE could be detected (Frank, Braun et al. 1990b).

DDT concentrations in lamb body fat from both Sydney and Perth in Australia were 79-82 $\mu g\ kg^{-1}$. DDT was used in large quantities in Western Australia to control pests in the 1970s. Its use was banned in 1981 and concentrations in Australian biota have been declining slowly since that time. This is despite the fact that in recent years DDT has been used for restricted agricultural purposes in some Australian states (Kannan, Tanabe et al. 1994).

HCB

Little has been reported in the scientific literature about the presence and concentrations of HCB found in meat. Comparatively, however, residues in meat products and in particular chicken from Spain were very much higher than those found in Australia, India, Thailand and Vietnam (by approximately an order of magnitude) (See Table 3.7). Hexachlorobenzene was detected in 47% of bovine meat samples tested in the Veracruz region of Mexico (Waliszewski, Pardio et al. 1997a). There are, at present, no FAO/WHO MRLs reported for this compound (FAO/WHO 1998).

Lindane and Other HCHs

Lindane residues in the fat taken from German game animals have been reported as 52 $\mu g\ kg^{-1}$ fat for partridges, 155 $\mu g\ kg^{-1}$ fat for pheasant and 121 $\mu g\ kg^{-1}$ fat for hares (Brunn, Berlich et al. 1985).

The total HCH burden found in 165 meat samples taken in Veracruz, Mexico had a mean of 0.728 $\mu g\ g^{-1}$ which the author reported as being above the FAO/WHO guideline value (Waliszewski, Pardio et al. 1997a). That it should be the mean value that exceeds the FAO/WHO figure is of particular concern, indicating widespread contamination of bovine meat in this region of Mexico (See Table 3.7).

PCDD/Fs and PCBs

The US Environmental Protection Agency have reported that consumption of animal products is an important pathway for human exposure to dioxins and related compounds because of the potential exposure caused by deposition of these compounds on pasture and forage crops (Fries, Feil et al. 1996) (See Fig 3.1). Animals may also be exposed to these compounds when they come into contact with wood treated with PCPs. Such wood was used extensively in the United States until use of pentachlorophenol was restricted in 1980 and may have been the cause of increased PCDD/F exposure in some individuals (Fries, Feil et al. 1996).

A study undertaken in the United States of America that reported on the concentrations of coplanar polychlorinated biphenyls (coplanar PCBs) in bovine meat found that, with the exception of PCB77, all other PCBs tested for (105, 118, 126, 156, 157, 169) were present in more than 85% of the meat analysed (See Table 3.8). PCB77 was present in only 19% of samples (Winters, Cleverly et al. 1996). The study concluded that the concentrations of PCBs found were comparable to those found in a similar study in Finland (Winters, Cleverly et al. 1996).

Although the concentrations of the majority of organochlorines have been declining in Sweden during the past 10 years, PCB153 has not fallen in line with the other compounds tested. In some cases residues of this compound were found at concentrations over 100 μg kg^{-1} in fat. Why this should be the case is uncertain, although it has been suggested that incidences of high PCB levels in beef may be due to contamination of feed in silos coated with PCB-containing material. Leakage of PCB-contaminated hydraulic oil from old equipment into feed during processing has also been suggested as a source of contamination (Glynn, Wernroth et al. 1998).

In general, pork contains the lowest levels of PCDD/Fs, probably because animals are slaughtered at a relatively early age (approximately 6 months) and have a large quantity of fatty tissue which will dilute the PCDD/F concentrations. In contrast veal tends to exhibit higher PCDD/F residues than does beef as veal calves usually consume a high proportion of milk in their diet (Malisch, Gleadle et al. 1999). Horsemeat, however, seems to follow the more general trend in which older herbivorous animals exhibit the highest body burdens (Malisch, Gleadle et al. 1999).

A German study investigated this ranking of dioxin contamination further and published the following increasing order of contamination for meat types:

Pork < Meat products < Poultry < Beef (Hecht and Blüthgen 1998).

This ranking of contamination is similar to findings from a study in Australia for PCBs. The residual wet weight concentrations of PCBs in beef, pork, lamb and chicken were comparable. But when concentrations in fat were calculated the highest concentrations were found in beef (mean 160 μg kg^{-1}) followed by lamb (95 μg kg^{-1}), pork (67 μg kg^{-1}) and chicken (34 μg kg^{-1}) (Kannan, Tanabe et al. 1994). Both compounds, therefore, appear to have their highest concentrations in beef.

Hecht (1998) also found that there was a regional difference in contamination between north and south Germany, with tissue concentrations in animals from the south of Germany being less contaminated than those from the north. In Sweden similar regional differences in contamination of beef was found, with residues from the south of Sweden being higher than those in animals from the north of the country. It was suggested that this might be due to the greater industrialisation found in the south of the country (Glynn, Wernroth et al. 1998). This explanation would be equally valid for the findings of the German study, where industrialisation and contamination are both greater in the north of the country. Overall intake of PCDD/Fs did not, however, appear to be different between the north and the south of the country (Wesp, Rippen et al. 1996).

In Thailand PCB residues were present in all foodstuffs examined. Highest concentrations among the items chosen were found in meat (range 2.3-12 μg kg^{-1}) and, in contrast to the findings of the German and Australian studies mentioned earlier, pork exhibited the highest concentrations (Tanabe, Kannan et al. 1991).

3.3 Dairy And Eggs

3.3.1 Dairy

According to Hippelein, 1996, milk and dairy produce contribute approximately 30-45% of human exposure to PCDD/Fs and 27% to PCBs. However, the concentrations may be highly variable depending on the country of origin of the produce. The main portion of this exposure travels through the route air⇒feed⇒cow⇒milk⇒man, although other sources may contribute to overall contamination (including binder twine treated with contaminated oil, wood silos treated with pentachlorophenol, contaminated soil or sewage sludge).

Table 3.9 FAO/WHO MRLs (μg^{kg-1}) for Milk and Eggs.

Chemical	Milk	Milk products with fat >2% (value expressed as μg^{kg-1} fat)	Eggs
Aldrin & Dieldrin	6	150	100
Chlordane	2	50	20
DDT	20	100	100
Endrin	0.8[a]	20	200[a]
Heptachlor	6	150	50
HCB	No MRL established or revoked	No MRL established or revoked	No MRL established or revoked
Lindane	10	250	100

Source: FAO/WHO 1998. (a) FAO/WHO Codex Alimentarius 1993.

Table 3.10 TDIs and MRLs for PCDD/Fs in milk set in different countries.

Country/Agency	*TDI/TEQ* pg I-TEQ kg^{-1} bw/day	MRL Milk
WHO/EURO	10	
Canada	10	
Germany	1[a]	3
	1-10[b]	5
	>10[c]	
Japan	1	
Netherlands	1	6
Nordic Council	5	
Switzerland	10	
Sweden	5	
United Kingdom	10	17.5
USFDA	0.0572	
USEPA	0.006	

Footnotes: [a]Precautionary limit; [b]range of risk; [c]intervention.
Source: Adapted from Wells and de Boer 1999.

Mixtures of a number of organochlorines are often found in a single food sample. This has been the case in Mexico where two areas formerly used for cotton growing were later turned to pasture for dairy herds. Cotton production had been associated with large scale pesticide use and resistance to these pesticides eventually led to the cessation of cotton growing. In these regions the production of milk and other dairy products has become an important economic activity (Albert 1990). Cheese from these areas contained residues of twelve different chlorinated pesticides including α-, β- and γ-HCH and p-p'-DDT and its

metabolites and in some cases samples were severely contaminated (Albert 1990). A similar situation pertained in Argentina, where a survey of butter samples contained organochlorine pesticide residues in 100% of samples analysed (n=146). 23% of samples contained at least five pesticides, 20% four. 2% of samples contained residues of more than 8 different organochlorine pesticides (Lenardon, Maitre de Hevia et al. 1994).

Table 3.11: Organochlorine Residues in Milk (μg kg^{-1} fat) in Various Countries.

Location	Chemicals							References
	PCBs	DDTs	HCHs	Aldrin & Dieldrin	Chlordane	HCB	Hepta-chlors	
Argentina		990	8.5		23.2	6.8	54.9	Maitre, Delasierra et al. 1994
			42.1					Maitre, Delasierra et al. 1994
Canada	0.1-0.8							Wells and de Boer 1999
China		2.8	2.9					Chen and Gao 1993
Egypt		7.52[a]	11[a]					Dogheim, Mohamed et al. 1999
Hong Kong		100	100	90		90	90	Wong and Lee 1997
India		150						Kannan, Tanabe et al. 1992b
		42-382	166					Mukherjee and Gopal 1993
		<50	22-166					Mukherjee and Gopal 1993
	<10[a]							Wells and de Boer 1999
Italy	46-83[b]							Baldassarri, Bocca et al. 1994
	4.7-15[c]							Baldassarri, Bocca et al. 1994
Mexico		159	94			14		Waliszewski, Pardio et al. 1997b
Slovakia		362				4		Prachar, Veningerova et al. 1995
Spain		56	43	26		19		Herrera, Ariño et al. 1996
United Kingdom	0.27-0.31[a]							Wells and de Boer 1999
Yugoslavia	1100[b]							Wells and de Boer 1999
	60[c]							Wells and de Boer 1999
	11[c]							Wells and de Boer 1999

Table 3.12: Organochlorine Residues in Butter (μg kg^{-1} fat) in Various Countries.

Location	Chemicals							References
	PCBs	DDTs	HCHs	Aldrin & Dieldrin	Chlordane	HCB	Hepta-chlors	
India		1600		160				Kannan, Tanabe et al. 1992b
Mexico		49	94			14	35	Waliszewski, Pardio et al. 1997b
Slovakia		10				4		Prachar, Veningerova et al. 1995
Thailand	38	24	69	67		2.2	3.2	Tanabe, Kannan et al. 1991
Vietnam	17	7.2	49	<0.1-2.2		5.0	8.0	Kannan, Tanabe et al. 1992a

Table 3.13: Organochlorine Residues in Cheese (μg kg^{-1} fat) in Various Countries.

Location	Chemicals							References
	PCBs	DDTs	HCHs	Aldrin & Dieldrin	Chlordane	HCB	Hepta-chlors	
India		345						Kannan, Tanabe et al. 1992a
Mexico		1420	140			50		Albert 1990

Aldrin and Dieldrin

There is little data in the literature on aldrin and dieldrin in dairy products. Concentrations of these compounds were, however, noticeable (mean 14 $\mu g\, kg^{-1}$ wet weight) in a study conducted in Australia (Kannan, Tanabe et al. 1994). The maximum concentration of dieldrin was found in a sample of butter from Hobart, Tasmania (26 $\mu g\ kg^{-1}$). According to the author this amounted to approximately 10% of the MRL for this substance in Australia (200 $\mu g\ kg^{-1}$). The current FAO/WHO guidelines (FAO/WHO 1998) recommend a lower concentration, 150 $\mu g\ kg^{-1}$ total concentration in dairy fat for aldrin and dieldrin, although this is still significantly higher than the residue reported. Aldrin and dieldrin are still used for sub-floor treatment for termites in some Australian states and their use has been correlated with the incidence of high residues of these compounds in human breast milk. Besides their use in termite control, aldrin and dieldrin were also used in agriculture in Australia in large quantities during the 1960s. This use has left considerable residues in soil that have subsequently accumulated in forage crops grown on them (Kannan, Tanabe et al. 1994).

3.4% of butter samples (n=146) analysed in Argentina exhibited residues in excess of guideline concentrations for aldrin/dieldrin (Lenardon, Maitre de Hevia et al. 1994). Aldrin and Dieldrin residues were particularly high in milk from Hong Kong and Argentina (See Table 3.11).

DDT

A study undertaken in Slovakia (Prachar, Veningerova et al. 1995) discussed the contamination of dairy produce from that region. Highest residues were of p-p'-DDE and p-p'-DDT (See Table 3.11) and were ascribed to the use of imported cattle food. Residues have, however, been falling in Slovakia during the last 20 years (Prachar, Uhnák et al. 1996).

Cheese from two regions of Mexico were heavily contaminated with DDT and its metabolites after dairy production began on land previously used for growing cotton (Albert 1990). The total DDT residue was calculated at 1420 $\mu g\ kg^{-1}$ in samples from one region. The study reported that these residues were much greater than the advisory level for cheese set by WHO of 1000 $\mu g\ kg^{-1}$. The reported maximum residue limit for this substance in cheese has since been reduced (See Table 3.13) to 50 $\mu g\ kg^{-1}$ fat.

DDT was detected in buffalo milk samples from an area in Egypt which contains the largest pesticide manufacturing facility in that country. The study discovered that buffalo milk from this area contained levels of DDT close to the maximum permitted concentration (Dogheim, Alla et al. 1996).

A study conducted by the Indian Council of Medical Research examined 2,205 samples of milk from both rural and urban areas of India. DDT was detected in more than 80% of samples, and 37% of samples had DDT concentrations exceeding the maximum prescribed by the Indian Ministry of Health and Family Welfare (Kalra, Kaur et al. 1999). The overall average value for all samples was 75 $\mu g\ kg^{-1}$ which was reported as being greater than the corresponding values from most other countries (Kalra, Kaur et al. 1999). In a study

investigating milk supplies in the Delhi area pesticide residues were detected in 100% of samples. DDT residues ranged from 104-220 μg kg^{-1} (whole milk) (See Table 3.11).

Increasing DDT residues were found in milk supplies in Hong Kong over the period 1985-1995, when mean residue figures rose from 10 μg kg^{-1} fat to 50 μg kg^{-1} fat (which is the current FAO/WHO MRL for milk fat). This has been attributed to the closure of a number of dairies in Hong Kong and the increase of imported milk supplies from mainland China (Wong and Lee 1997).

Highest DDT residues were found in milk from Argentina, butter from India and cheese from Mexico which in all cases exceeded the WHO/FAO MRL (See Table 3.9).

HCB

Bratislava in Slovakia is considered a hot-spot for environmental contamination by HCB, although residues found in butter from that area were not significantly higher than from similarly industrialised regions in Germany and the Czech Republic (in the region of 7-14 μg kg^{-1} fat) (Prachar, Veningerova et al. 1995). No FAO/WHO MRLs exist for this compound (See Table 3.9). In Hong Kong surveys of HCB and other pesticides in milk have found an increase in HCB residues in milk during the period 1985-1995. As with increased DDT residues, the increase in HCB concentrations in milk seems to be the result of a change in milk sources in Hong Kong over that period from local dairy industries to imported milk from China (Wong and Lee 1997).

In Australia, HCB was used as a seed dressing fungicide against bunt fungi (*Telletia* spp.), but was also produced as a by-product in various chlorination processes and in combustion of industrial products. HCB was a major contaminant in Australia during the early 1970s, but was officially banned from agricultural use in 1972. It has, nonetheless, been reported that large amounts of waste HCB remain stored at several sites. The concentrations of HCB in foodstuffs was, however, among the lowest of the various chemicals analysed (Kannan, Tanabe et al. 1994).

Heptachlor and Heptachlor Epoxide

Heptachlor and its epoxide concentrations were generally low in most foodstuffs in Australia, with the exception of dairy products. Like aldrin and dieldrin, heptachlor is used in Australia for the control of termites in buildings, against beetles and crickets on golf courses and on lawns (Kannan, Tanabe et al. 1994). Total heptachlors were 54.9 μg kg^{-1} in milk from Argentina and heptachlor epoxide was reported at 90 μg kg^{-1} in Hong Kong (See Table 3.11) (Maitre, Delasierra et al. 1994; Wong and Lee 1997). Milk in Mexico also exhibited high residues of heptachlor (See Table 3.11), although the reported value was significantly lower than (approximately half) the value of that for milk in Argentina. All these residues are an order of magnitude greater than the MRL for heptachlor and its epoxide of 6.0 μg kg^{-1} (FAO/WHO 1998). These values contrasted strongly with those found in southern Asia. In Thailand and Vietnam heptachlor and heptachlor epoxide residues were consistently below 5 μg kg^{-1} (Tanabe, Kannan et al. 1991; Kannan, Tanabe et al. 1992a).

In a survey of butter samples in Argentina 5.9% of samples analysed (n=146) exceeded safety limits for heptachlor (Lenardon, Maitre de Hevia et al. 1994).

Lindane and Other HCHs

A study undertaken in Veracruz, Mexico found that α-HCB residues were detectable in 92.0% of milk samples tested, at a mean concentration of 15 μg kg^{-1}. 63.3% of milk samples contained β-HCH at an average concentration of 49 μg/ kg^{-1}. Lindane (γ-HCH) was detected in 80% of samples at a mean concentration of 30 μg kg^{-1}. Total HCHs in milk were 94 μg kg^{-1}, close to the FAO/WHO tolerance limit in place at the time of 100 μg kg^{-1} (Waliszewski, Pardio et al. 1997a). This MRL has since been reduced to 10 μg kg^{-1} (FAO/WHO 1998). The presence of these compounds in dairy products was thought to be as a result of lindane being used in Veracruz as a pesticide of choice for livestock vector control programmes, and for the control of agricultural pests (Waliszewski, Pardio et al. 1997a).

The manufacture and use of technical grade HCH has been banned in India since April 1997. Nonetheless in a country-wide survey undertaken by the Indian Council of Medical Research, more than 80% of milk samples contained detectable residues of these compounds. α-, β-, γ-, and δ-HCH residues were found in excess of the Indian Ministry of Health and Family Welfare guidelines in 21, 42, 28 and 4% of samples analysed respectively (Kalra, Kaur et al. 1999). There were regional differences in the extent of this contamination. The proportion of samples with residues of α-HCH above the legal limit was 52% in Andhra Pradesh and Uttar Pradesh against 6% in Himachal Pradesh, Haryana and Punjab. β-HCH and γ-HCH were much less frequently detected than α-HCH. Contamination of milk was ascribed to administration of contaminated feed to the animals concerned, although the spraying of cattle sheds with HCH (and DDT) for control of mosquitoes could also be a factor (Kalra, Kaur et al. 1999).

1.4% of butter samples analysed in Argentina (n=146) contained residues of HCH in excess of recommended limits (Lenardon, Maitre de Hevia et al. 1994).
In Australia concentrations of HCH were relatively high in butter (0.11-0.32 μg kg^{-1} wet weight). Like DDT, HCH was used as an acaricide to control external parasites infesting cattle and the use of HCH in cattle dips used to be common in that country (Kannan, Tanabe et al. 1994). This compound was used extensively for the control of cane grubs and beetles in the soils of Queensland and New South Wales. α-HCH was the predominant isomer in dairy products, in contrast to other types of foodstuffs analysed where all isomers were present in roughly equal measure (Kannan, Tanabe et al. 1994).

PCDD/Fs and PCBs

The excretion of PCBs and PCDD/Fs via milk is the main elimination pathway of these compounds in cows (Baldassarri, Bocca et al. 1994). Levels of PCBs in dairy products and particularly in cow's milk have been reported for a number of countries, although there is considerable variation in the values reported (Krokos, Creaser et al. 1996).

From May 1989-March 1990 milk from dairy farms located near to municipal waste incinerators (MSWs) in the Netherlands was analysed for PCDD/Fs. Findings of up to 13.5 pg TEQ g^{-1} (fat) in some samples taken from farms close to the MSWs led to the decision by the Dutch government to instigate an upper limit for dioxin levels in milk and milk products of 6 pg TEQ g^{-1} (fat) (Liem, Hoogerbrugge et al. 1990). Furthermore, milk from farms near to metal reclamation plants contained relatively more furans than dioxins when

compared with milk from farms in the vicinity of MSWs (Liem, Hoogerbrugge et al. 1990). Milk and milk products are the major route for human dioxin exposure in the Netherlands (Theelen, Liem et al. 1993). Studies undertaken in Switzerland have also shown that milk from cows grazing near such incinerators contains higher residues of dioxins and furans than from cows grazing on unexposed areas. This would become of particular concern for producers of high fat dairy produce such as cheese, butter, chocolate and ice cream who obtain their raw milk from cows grazing near an incinerator. Cows grazing close to incinerators in the Netherlands and in the Tyrol in Austria had up to 13.5 pg TEQ g^{-1} and 8.6 pg TEQ g^{-1} respectively. Higher concentrations (13.5-37.0 pg TEQ g^{-1}) were reported in milk from cows grazing near to a metal reclamation plant in Germany. Lower dioxin emissions from newer incinerators were, however, also reported (0.8 pg TEQ g^{-1}) (MAFF 1997d).

The Ministry of Agriculture, Fisheries and Food (MAFF) in the United Kingdom first began surveying cow's milk for dioxins in 1989 and found that concentrations were usually in the range 1.1-1.5 pg TEQ g^{-1} milk fat in milk from rural farms and 2.0-7.1 pg TEQ g^{-1} milk fat in farms situated closer to urban and industrial areas. This work brought to light a case of extreme contamination where dioxin residues in excess of 40 pg TEQ g^{-1} were found in milk from two farms near Bolsover in Derbyshire (See also Section 4.7). These residues were greatly in excess of the Maximum Tolerable Concentration for dioxins of 17.5 pg TEQ g^{-1} fat set by the UK MAFF and the Department of Health, and the milk from these farms was withdrawn from sale. Milk from other farms in the area exhibited dioxin concentrations similar to those found in rural areas (1.1-7.1 pg TEQ g^{-1}) (See Table 3.14). By 1994 only one farm was still producing milk with dioxin concentrations over the MTC set by MAFF/DoH and residues from other farms had declined over a relatively short period. This suggested that the contamination was not on-going, but caused by a particular incident (Harrison, Gem et al. 1994).

Residues of PCDD/Fs in dairy products from North-Rhein-Wesphalia in Germany fell between the years 1990 and 1994 by approximately 25% from 1.35 μg kg^{-1} I-TEQ fat to 1.02 μg kg^{-1} I-TEQ fat (Fürst and Wilmers 1995). In addition the range of contamination fell between these years from a maximum in 1990 of 2.62 μg kg^{-1} I-TEQ fat to 1.75 μg kg^{-1} I-TEQ fat. A similar study conducted in Bavaria, in southern Germany, indicated that PCDD/F concentrations in milk from that region fell within the same range (Mayer 1995a).

A monitoring exercise in France analysed a number of milk, butter, cheese and cream samples in order to determine the background I-TEQ for PCDD/Fs. The average value (See Table 3.14) of 1.33 μg kg^{-1} I-TEQ fat indicated low background contamination of these food items which compared favourably with similar exercises undertaken in Germany and the Netherlands. In these countries background contamination of dairy products averaged 5 and 6 μg kg^{-1} I-TEQ fat respectively (Defour, Fraisse et al. 1997).

Butter samples from the most highly populated regions of Egypt were analysed for PCDD/Fs and exhibited a wide range of contamination. Samples from lower Egypt (i.e. the area encompassing Cairo and points north through the Nile delta to the sea) were most heavily contaminated and nearly all samples exceed 5 pg TEQ g^{-1} fat. The mean of all samples was 7.69 pg TEQ g^{-1} fat (See Table 3.14). Butter from other regions of the country such as El-Fayoum and Kom Ombo was not so highly contaminated, however, and

none of the samples from these regions contained residues above 3 pg I-TEQ g^{-1} fat (Malisch and Magdi 1996).

Milk samples from 12 dairy farms in Spain were analysed along with 23 samples of pasteurised cow's milk for PCDD/Fs. Farms in areas without specific dioxin sources such as incinerators produced milk with background levels of PCDD/Fs in the range 1.3-2.47 pg TEQ g^{-1} fat. This contrasted with the higher concentrations of PCDD/Fs found in milk produced near waste incinerators, chemical factories and metal reclamation facilities which, although more highly contaminated, did not exceed 6 pg TEQ g^{-1} but were in the range 0.67-3.80 pg TEQ g^{-1} (See Table 3.14) (Ramos, Eljarrat et al. 1996; Ramos, Eljarrat et al. 1997).

Although production of PCBs ceased in Slovakia in 1984, there continue to be incidents of high residues in soil caused through accidental spillage, or premature closure of PCB manufacturing facilities. In addition silo coatings containing PCBs led to the contamination of some milk supplies. PCB residues have declined since these coatings were removed (Prachar, Veningerova et al. 1995).

A study in the United Kingdom investigated PCB contamination of bulk milk supplies and found that total PCB concentrations (the sum of 37 different congeners) ranged from 3.4-16.4 μg kg^{-1} milk fat, with a mean concentration of 8.4 μg kg^{-1}. The results of this study indicated that these concentrations would typically contribute 11% of the average daily total PCB intake for individuals in the UK. This contribution would increase to 30% when exposure through other dairy products such as cheese and butter is taken into account. The inclusion of the TEF assigned to PCBs by this study would typically increase the TEQ rating of cows' milk by approximately 40% over that attributed to PCDD/Fs alone (Sewart and Jones 1996).

Table 3.14: PCDD/Fs and PCB I-TEQ values (pg g^{-1} fat) found in Milk in Various Countries.

Location	Contamination Type	Milk	References
Austria	Urban	20.1-69.5	Wells and de Boer 1999
		1.0-2.1	Wells and de Boer 1999
Belgium	Urban	2.7-5.1	Wells and de Boer 1999
	Rural	2.7-3.9	Wells and de Boer 1999
Canada		0.07	Wells and de Boer 1999
Denmark		2.6	Wells and de Boer 1999
France	Background	1.91	Defour, Fraisse et al. 1997
		0.99-4.24	Ramos, Eljarrat et al. 1997
	Background	1.74	Fraisse, Schnepp et al. 1996
Germany		5.1-9.0	Hippelein, Kaupp et al. 1996
		3.9-5.2	Hippelein, Kaupp et al. 1996
	Rural	2.32	Wells and de Boer 1999
	Urban	1.9-6.5	Wells and de Boer 1999
	Urban	5-26	Wells and de Boer 1999
	Rural	0.8-3.2	Wells and de Boer 1999
	Rural	0.6-1.4	Ramos, Eljarrat et al. 1997
	Incinerator	5.0-5.7	Ramos, Eljarrat et al. 1997
	Background	0.6-1.54	Ramos, Eljarrat et al. 1997
	Background	1.0	Wesp, Rippen et al. 1996
	Contaminated	0.48-5.62	Ramos, Eljarrat et al. 1997
Italy		0.15-0.9	Zanotto, Alcock et al. 1999
Netherlands	Background	0.7-2.5	Liem, Hoogerbrugge et al. 1990
	Background	0.38	Cuijpers, Bremmer et al. 1998
	Rural	0.7-2.0	Wells and de Boer 1999

	Rural	1.58	Ramos, Eljarrat et al. 1997
	Urban	1.2-13.5	Wells and de Boer 1999
	Nr MSW	5.1-13.5	Liem, Hoogerbrugge et al. 1990
New Zealand		0.18-0.22	Wells and de Boer 1999
Norway		0.11	Wells and de Boer 1999
Russia		0.28-5.0	Wells and de Boer 1999
Spain	Background	0.73	Ramos, Eljarrat et al. 1997
	Background	2.02	Domingo, Schuhmacher et al. 1999
	Rural	0.67-2.47	Ramos, Eljarrat et al. 1997
	Incinerator	1.09-3.5	Ramos, Eljarrat et al. 1997
Sweden		0.44-1.4	Wells and de Boer 1999
Switzerland		0.7-3.28	Wells and de Boer 1999
	Industrial	2.02-4.85	Ramos, Eljarrat et al. 1997
United Kingdom		0.08[(a)]	Krokos, Creaser et al. 1996
		1.1-7.1	Harrison, Gem et al. 1994
	Incinerator	3.0-7.1	Ramos, Eljarrat et al. 1997
		0.06	Wells and de Boer 1999
		0.08	Wells and de Boer 1999
United States		0.42-0.81	Fiedler, Lau et al. 1996
		0.42-0.81	Cooper, Fiedler et al. 1995

3.3.2 Eggs

The US Government (Food and Drug Administration) began a survey of some food items, including eggs, after discovering elevated residues of 2,3,7,8-TCDD (the most toxic PCDD congener) in chickens. This contamination was traced to a flowing agent, ball clay, which was included in animal feed for a number of farms. Whole chicken egg 2,3,7,8-TCDD concentrations from ball-clay affected farms averaged at least 50 times higher than detection limits reported from other regions of the US species (Hayward, Nortrup et al. 1999). The discovery of this contamination through routine monitoring suggests that such monitoring of food items continues to be of value.

Eggs may also contribute a considerable fraction of daily exposure to POPs from food. Unless the chicken food itself is contaminated, the best indicator of potential contamination in eggs is their housing, with wire-caged chickens producing eggs with considerably lower residues than chickens with access to soil (Fürst, Fürst et al. 1993).

Egg consumption can be a significant contribution to the daily intake of PCDD/Fs. In Germany 5% of exposure was considered to be as a consequence of egg consumption, although this was calculated using battery reared chickens as the egg source. Free-range chickens, or those exposed to soil, may have higher average PCDD/F residues in their eggs than battery-reared chickens. The extent to which eggs become contaminated with PCDD/Fs is, however, dependent on more than whether the chickens are raised in cages or outdoors. The less densely packed (i.e. the freer the range) the chickens are, the greater their access to soil organisms. It is these soil organisms (annelids, insects etc.) which are the predominant source of PCDD/Fs in soil. The density of soil organisms is inversely correlated with chicken density, so the higher the density of chickens, the less exposed they become to these contaminants (Fürst, Fürst et al. 1993; Schuler, Schmid et al. 1997). Regular consumption of contaminated eggs can result not only in an increased daily intake of PCDD/Fs but also in a significantly elevated body burden for a single exposed person (Schuler, Schmid et al. 1997). The residues of PCDD/Fs found in eggs from Swiss chickens reared in wire cages is generally in the region 0.5-2.0 pg I-TEQ g^{-1}. This is a similar range of values to those found in the remainder of western Europe (Schuler, Schmid et al. 1997).

3.4 Fruit, Vegetables And Other Crops

Pesticides are widely used in agriculture and horticulture to secure attained production and quality targets and for the control of weeds. Inevitably some residues remain on food crops when these methods are used (Dejonckheere, Steurbaut et al. 1996). An important factor in the level of these residues is the period allowed between application and harvest. The longer the period allowed, generally the lower the residue detected. But rainfall levels also need to be taken into account. During dry periods a longer time may need to be allowed between application and harvest for residues to diminish (Frank, Braun et al. 1990a). Other compounds (e.g. PCDD/Fs) known to be ubiquitous in the atmosphere may be found on fruit and vegetables as a consequence of aerial deposition either from the vapour phase or through the deposition of particulates.

The contaminant burden of a crop is usually a combination of uptake via the root system; direct foliar uptake and translocations within the plant and surface-deposited material associated with fine particulate matter (Jones 1991). Several studies have indicated, however, that the waxy outer surface of plants will accumulate lipophilic organic pollutants from the atmosphere (Nakamura, Matsueda et al. 1994). Application of pesticides to growing crops is not, however, the only manner in which food items may become contaminated. In areas where public health pesticide use is prevalent for control of diseases such as malaria, the spraying of residential properties may result in concurrent contamination of food items. A study undertaken in India found that wheat stored in gunny sacks could accumulate high concentrations of DDT and HCH, to levels above the Codex Alimentarius MRL for these substances, as a result of standard residential property fumigation practices (Singh, Battu et al. 1991).

India also has a serious problem with pesticide residues on food items and in particular on cereals, pulses, fruit and vegetables. A study conducted there, however, found that household preparation of fruit and vegetables resulted in a significant lowering of the pesticide residues (60-95%) found in them. Initial washing of the items was responsible for the removal of the majority of the contaminants (Ramesh and Balasubramanian 1999).

Table 3.15: Maximum residue limits (MRLs) for various pesticides on vegetables and fruit published by the Joint FAO/WHO Food Standards Programme Codex Alimentarius Commission (1998).

Chemical	Vegetables	Fruit
Aldrin & Dieldrin	20-100	50
Chlordane	20	20
DDT	100-200	-
Endrin	50	-
Heptachlor	20-500	10
Hexachlorobenzene	No MRLs established or revoked	No MRLs established or revoked
Lindane	50-2000	500-3000

Footnote: Where a range of values are given these refer to and lowest and highest values given for different vegetable species (e.g. the lowest value may refer to cabbage and the highest to potatoes). Values are in $\mu g\ kg^{-1}$.

In Canada fruit and vegetables are routinely analysed for pesticides and maximum residue limits are established for many of them. Generally common pesticides presently in use are found with greater overall frequency and over a greater range of concentrations than are those which have been banned from use. Banned pesticides are found very infrequently and usually at very low concentration. This latter group, in Canada, includes many of the organochlorines such as DDT and its metabolites, aldrin, dieldrin, endrin, heptachlor and lindane. Produce imported into Canada contains residues of these compounds at slightly higher frequencies than produce grown domestically. Crops that mature on, or below, the soil surfaced were found to contain pesticide residues at a higher frequency than other foods suggesting that some uptake from soil was occurring (Neidert, Trotman et al. 1994).

Aldrin and Dieldrin

A study of different types of tea in Spain found 78.3 $\mu g\ kg^{-1}$ of aldrin in camomile tea. The authors suggested that this was indicative of recent application of aldrin to the plants, although no details were given of where these were grown (Fernández, Sierra et al. 1993).

DDT

In a study of Egyptian vegetables and fruit samples of carrot and eggplant contained trace amounts of p-p'-DDE and p-p'-DDT (carrot p-p'-DDE 30 $\mu g\ kg^{-1}$; p-p'-DDT 40 $\mu g\ kg^{-1}$; eggplant p-p'-DDE 10-20 $\mu g\ kg^{-1}$) but no trace of these compounds was found in any other vegetables analysed. In general residues of DDT and DDE have diminished considerably in vegetables and fruit grown in Egypt since DDT was banned from use in 1987 (Dogheim, Alla et al. 1996).

In Australia monitoring studies have indicated that the prolonged use of DDT in vegetable, fruit and cotton growing areas during the 1970s and 1980s has left considerable residues in soil which has contaminated crop produce. P-p'-DDE was the predominant compound in most foodstuffs analysed suggesting that these residues were derived mainly from past use of DDT. Considerable proportions of p-p'-DDT and o,p'-DDT in coffee (Kannan, Tanabe et al. 1994). (74 and 15% respectively) and tea (40 and 20%) suggests a recent application of DDT on plantation crops. Vegetables such as tomato, pumpkin and cabbage also contained a higher proportion of p-p'-DDT than would be expected from historical rather than current use which suggests that DDT continues to be used illegally in Australia despite its ban in 1981 (Kannan, Tanabe et al. 1994). DDT was present at significant concentrations in human adipose tissue from Papua New Guinea (PNG) implicating the extensive use of DDT in malaria eradication programmes. This has also been reported from other South Pacific islands (Kannan, Tanabe et al. 1994).

Lindane and Other HCHs

Egypt banned the use of HCH pesticides in 1987 and residues of these compounds are now low or undetectable in vegetables grown in that country (Dogheim, Alla et al. 1996).

A high percentage incidence of α-HCH was found in samples of black tea, camomile tea and linden tea purchased and analysed in Spain, although none of the samples contained residues exceeding the EEC allowed limits (10 $\mu g\ kg^{-1}$ for total HCHs). Generally camomile and linden teas were more contaminated than were black teas. The maximum residues found were 353 $\mu g\ kg^{-1}$ of lindane and 28 $\mu g\ kg^{-1}$ of

α-HCH both in linden tea (Fernández, Sierra et al. 1993).

In Thailand HCH residues were generally low in foodstuffs when compared with other countries, reflecting the low usage of these compounds there and no foods analysed exhibited residues in excess of maximum residue limits (Tanabe, Kannan et al. 1991).

PCDD/Fs and PCBs

PCB and PCDD/Fs foodstuff monitoring programmes have in the past concentrated primarily on fatty foods such as dairy products, fish and meat. Relatively few studies have investigated the levels of these contaminants in fruit and vegetables. Those that are contained in the literature suggest that levels are usually below 10 μg kg^{-1} (Foxall, Lovett et al. 1995). Accumulation of PCDD/Fs in plants occurs primarily through atmospheric deposition. Soil-related pathways are negligible, probably due to the relative immobility of these compounds in soil and their strongly hydrophobic nature. Exceptions to this generalisation do, however, occur. Courgettes (zucchini) produce a root exudate which is capable of mobilising PCDD/Fs within the soil. Lipophilic compounds secreted by the plant are then able to translocate the PCDD/Fs throughout the tissues (Hülster and Marschner 1995).

PCDD/Fs concentrations on fruit and vegetables may have a seasonal element. A study in the Netherlands that investigated the use of kale plants as bioindicators for PCDD/Fs found that PCDD/Fs emissions rose during the winter months and that deposition on the kale plants increased as a result (Köhler and Peichl 1996).

An intensive study of these compounds in fruit and vegetables was, however, undertaken in the United Kingdom beginning in 1991. This survey was initiated primarily as a result of public concern over the operations of a chemical waste incinerator and once elevated levels of these compounds had been detected in soil and air further investigations were undertaken to discover whether elevated levels of PCDD/Fs and PCBs might occur in locally grown fruit and vegetables. The findings indicated that very slightly elevated residues were found in these crops when compared with similar crops grown in a rural area, but that increases in the average daily intake of PCBs and PCDD/Fs by people consuming these items would be limited to between 3 and 8% (Foxall, Lovett et al. 1995).

In Thailand PCB residues were found in all food samples analysed. Lowest residues were present in rice grains (0.22 μg kg^{-1} wet weight) and the highest in corn oil (51 μg kg^{-1}). Fruit and vegetables generally, however, contained the lowest concentrations when compared with other foodstuffs examined such as meat and fish (Tanabe, Kannan et al. 1991). There are no statutory thresholds for PCBs in food in Vietnam, but although dietary intake of these compounds by the population was roughly comparable with PCB intakes in industrialised countries, the source of this intake differs being predominantly as a result of consumption of cereals (34%) and vegetables (35%) rather than meat and fatty foods as is the case elsewhere (Kannan, Tanabe et al. 1992a).

4. PCB AND DIOXIN CONTAMINATION INCIDENTS

Over the years there have been numerous contamination incidents which have either caused serious public health impacts or have caused restriction of foodstuffs to protect human health. Some of the worst have been caused by PCBs, dioxins or a combination of the two.

4.1. PCB contamination: Yusho & Yu-Cheng

On three separate occasions, PCB oils have got into the human food supply. Two of these incidents were caused by PCBs leaking into cooking oil from heat exchange equipment. The first, the "Yusho" incident, occurred in the Kyushu region of Japan in 1968, and affected around 1700 people. The second, the "Yu-Cheng" incident in Taichung Province, central Taiwan in 1979, affected some 2000 people (Chen and Hsu 1987; Kashimoto and Miyata 1987; Chen, Guo et al. 1992; Rogan 1995). “Yusho” and “Yu-Cheng” both mean “oil disease”.

The ingested dose of PCBs in Yusho was estimated to average 2 g with a minimum dose; in Yu-Cheng the dose was estimated as 0.8-1.8g (Ikeda 1994). The PCBs involved had degraded to some extent. Consequently the oil responsible for the Yusho incident also contained dioxins (PCDDs), dibenzofurans (PCDFs), polychlorinated quaterphenyls (PCQ) and sulphur derivatives of the PCBs (Ikeda 1994; Masuda et al. 1998); that responsible for the Yu-Cheng incident also contained PCDFs, PCDDs, PCQs, sulphur derivatives of PCBs, polychlorinated terphenyls (PCTs) and polychlorinated naphthalenes (PCNs) (Ryan and Masuda 1994; Guo, Lambert et al. 1995).

Different components of Yusho oil were extracted and tested on monkeys and rats. Of these, the furans produced Yusho-like symptoms, but the PCBs and PCQs did not (Ikeda 1994). A similar picture emerged from calculations of the dioxin-type toxicity (TEQs) of different components of the Yu-Cheng oil. PCDDs represented a TEQ of 4.472 ng g^{-1} (12% of the total TEQ); coplanar PCBs contributed 13.154 ng g^{-1} (35%) and the PCDFs contributed 19.568 ng g^{-1} (53%). Moreover, workers exposed to uncontaminated PCBs exhibited less toxic responses than Yusho/Yu-Cheng patients (Ikeda 1994).

The initial symptoms of Yusho and Yu-Cheng included dermal abnormalities (acne, patches of dark pigment, skin swelling and thickening); eye problems (including discharges and pigmentation change); respiratory problems (chronic bronchitis); neurological disorders (headache and visual impairment) and other effects such as fatigue and anorexia. Yu-Cheng patients suffered significantly increased mortality rates over the next 13 years with the greatest increase in chronic liver disease and cirrhosis (rate ratio 5.3, 95% CI 1.9-14.7). Death from circulatory system disease, including heart disease, and diseases of the respiratory system and digestive system were also significantly elevated, though the observed increase in cancer deaths was not significant (Yu, Guo et al. 1994).

Children *in utero* at the time of the poisoning were severely affected. Thirteen women were reportedly pregnant during the Yusho incident. Eleven children born subsequently had excessive pigmentation, including one that was stillborn. Among these prenatally exposed children there were also nine cases of conjunctivitis, eight of hyperbilirubinemia, four of pigmented gums and four of low birth weight for gestational age (Rogan 1995).

Yu-Cheng children have been followed and monitored closely, providing an unrivalled understanding of the effects of acute organochlorine exposure on human development. This cohort includes children born from 1978 (who were not exposed *in utero*, but only via breast milk) until 1985 (Guo, Lin et al. 1994; Guo, Lambert et al. 1995). Symptoms at birth were similar to those of the Yusho and of 39 babies *in utero* at the time of the incident, eight died in the first few years of life. In 1985, Yucheng children ranging in age from 6 months to 7 years old were compared with matched controls. Significant differences were found in height, weight, pigmentation, occurrence of acne, formation of the nails, and frequency of respiratory infection. (Guo, Lambert et al. 1995).

Six years later, the Yu-cheng children remained shorter and had a lower total lean mass (Guo, Lin et al. 1994; Guo, Lambert et al. 1995). Abnormalities of the nails and immune deficit persisted; respiratory infections and chronic otitis media (infection of the ear) were both significantly elevated (Chao and Hsu 1994; Guo, Lambert et al. 1995). Overall neurological and behavioural development of the children was affected; amongst other measures, cognitive development was reduced as measured by the Stanford Binet test and the Weschler Intelligence Scale for Children, Revised (Chen, Guo et al. 1992; Guo, Lambert et al. 1995). There was also preliminary evidence of visual recognition memory deficits (Ko, Yao et al. 1994). Neurological effects were present in children born up to six years after the incident and they persisted up to seven years of age (Chen, Guo et al. 1992; Guo, Lambert et al. 1995). An eighth year study reported that Yu-Cheng children's performance in the Weschler test and in Raven's Standard Progressive Matrices had improved to the extent that, although they still tended to score lower than the control children, the differences were no longer significant (Lai, Chen et al. 1994). Boys appeared to be more severely affected than girls, possibly because of prenatal endocrine disruption (Guo, Lai et al. 1995).

Penile length in older boys (11-14 years) was significantly reduced. The authors caution that this represents only a preliminary study of sexual development and confirmatory research will be needed. They also hypothesise that reduction in the length of the penis could be caused by endocrine disruption, though all parameters measured were found to be normal (Guo, Lai et al. 1993; Guo, Lambert et al. 1995).

PCBs, PCDDs and PCDFs were still detectable in the blood and sebum of Yusho and Yucheng victims collected in 1994 -1996, between 17 and 26 years after exposure (Iida, Hirakawa et al. 1999). Further insights into human response to these chemicals will clearly continue to be derived from ongoing research programmes.

4.2. PCB Contamination: the Belgian scandal

Both the Yusho and Yu-Cheng incidents occurred over 20 years ago, when PCBs were still in widespread use. In Europe they have not been manufactured for many years and legislation is in place to ensure that all remaining stocks be destroyed (EC 1996). However, this did not prevent PCBs getting into the European food chain in an incident with had severe repercussions for the farming industry.
The incident in question began in Belgium in January 1999. The amount of PCBs involved was small; a team involving personnel from the Belgian Federal Ministry of Agriculture report the amount to have been around 50kg of PCBs containing a total of 1g of PCDD/Fs (Bernard, Hermans et al. 1999).

At the beginning of the incident, it was not realised that PCBs were the agent. When the customers of the de Brabander feed producer reported that their chickens were becoming ill, and the firm contacted their insurance company. The symptoms resembled "chick oedema disease" previously caused by PCBs and dioxins (Bernard, Hermans et al. 1999) and the expert appointed by the insurance company recommended dioxin analyses. This happened in March 1999; confirmation of the presence of dioxin came in April. However, the Belgian government commissioned further analyses and so it was not until late May that they informed the European Commission and advised that all chicken and egg products be removed from sale (Ashraf 1999).

It transpired that the contamination originated in mid-January from one tank containing approximately 80 tonnes of recycled animal fat at the Verkest Company. From Verkest, the contaminated fats were supplied to a maximum of nine feed-producing companies (including de Brabander) who then supplied animal feed containing PCBs and dioxins to farm producers of chickens, eggs, pork and beef until the end of January. Most of the feed producers were in Belgium, but some were also in France and the Netherlands.

Although the monitoring data for this crisis are not yet fully available, some compilations of information were obtained from the Belgian Department of Health and from other sources during June 1999. Almost all of the data available at that time were for dioxins.

Although initially all attention was focussed on the dioxins, it soon became apparent that dioxin-contaminated PCBs were the source of the problem. The PCB congener profile in the feed closely resembled that of Aroclor 1260 (Bernard, Hermans et al. 1999). Since testing for PCBs was quicker and cheaper and could be carried out by a greater number of laboratories, this was used as a monitoring tool, based on an approximated ratio of PCB:PCDD/F of 22 000:1 (Bernard, Hermans et al. 1999). The EC Scientific Committee on Food (SCF 1999) reported that PCBs contributed 80% of the total PCB/dioxin ITEQ in some egg samples from the Belgian incident. This means that the total dioxin toxicity equivalents in Belgian samples could be 5 times higher than that of the dioxins alone, though this may not necessarily apply to other products (e.g. pork, milk).

The concentration of PCDD/Fs in one chicken feed sample was 752 pg ITEQ g^{-1} product; this is over 1500 times the EC limit for dioxins in feedstuffs (specifically citrus pulp) set in 1998 (EC 1998).

Some of the samples were collected from breeding stock which were not destined for human food. Nevertheless, it is worth comparing the concentrations with those published on chickens for human consumption. Belgium set a limit of 5 pg ITEQ g^{-1} fat for milk and milk with a higher concentration of dioxin than this should have the cream removed and destroyed (de Fre and Wevers 1998). This is a useful benchmark against which to judge concentrations in food-related products.

With the exception of one sample (1.23 pg ITEQ g^{-1} fat), all Belgian chicken samples were very highly contaminated. Apart from this one, all samples were between 10 and approximately 5000 times higher than the Belgian milk guideline (range 50.9 to 2360 pg ITEQ g^{-1} fat. They were also many times above the concentrations found in poultry meat

in Europe or the US during the 1990s (Fiedler et al. 1997; Fürst et al. 1990; Harrison et al. 1998; Malisch et al. 1998c).

Concentrations of dioxins in Belgian eggs ranged from 6.7 to 685 pg ITEQ g^{-1} fat. These exceeded all values found in the scientific literature for the 1990s (0.2-2.1 pg ITEQ g^{-1} fat) (Fiedler, Cooper et al. 1997; Harrison, Wearne et al. 1998; Malisch 1998b). All were higher than the Belgian guideline for dioxins in milk (5pg ITEQ g^{-1} fat). The most contaminated sample exceeded this limit by over 130 times.

None of the samples of pork analysed contained concentrations of dioxin in excess of the Belgian guideline for milk (5 pg ITEQ g^{-1} fat). It was later reported that some pork samples contained excessive concentrations of PCBs, though no data were provided (Bernard, Hermans et al. 1999).

Extensive restrictions on a worldwide scale were placed on Belgian food products once the contamination became known. On the 2nd June, Belgium placed restrictions on 500 pig holdings that were thought to have received contaminated feed and, on the 3rd June, it informed the Commission that some cattle had also received contaminated feed (EC 1999b). On the 4th June, the USA banned all imports of pork and poultry form the EU (Ashraf 1999). The EC also acted swiftly once it had been informed of the problem. On the 3rd June, the EC decided that products derived from poultry reared in Belgium between the 15th January and 1st June 1999 should not be sold unless they are they are known to be uncontaminated (EC 1999a). A similar Decision was passed on 4th June, referring to the products from cows and pigs (EC 1999b). The various Directives were repeatedly amended and/or superseded. At the time of writing, a maximum concentration of 200 ng (7 PCBs) g^{-1} fat was allowed in eggs, egg products, fresh poultry meat and derived products, fresh pig meat and derived products from Belgium. Exceptions can be made if analyses have established that the products are not contaminated or the animals were slaughtered after 20th September 1999 (EC 1999c; EC 2000).

Although it has been stated that "*it is unlikely that the isolated episode of contamination in Belgium will cause adverse effects on the general population*" (Bernard, Hermans et al. 1999), there is little room for complacency. Individuals known to have been exposed should be monitored for contaminant levels and health outcomes. The slowness of the response also exacerbated the problem. In all, there was a four month delay between the first symptoms being seen in chickens (end of January) and restrictions being placed on foodstuffs (end of May). The delay in restricting sales and exports of contaminated foodstuffs undoubtedly caused people to be exposed unnecessarily. Even if health impacts are not great, the economic impact on the Belgian food industry was enormous and ongoing, due to the need to continuously monitor all potentially affected products. All in all, this could hardly be a more illuminating example of the potential for PCB wastes to cause harm some 20 years after its removal from use. The case clearly illustrates that contamination can travel through the human foodchain rapidly on a global scale, even though the pollution originated from a local source.
The contamination of Belgian chickens illustrates the point that a very small amount of regulated POPs that have been banned still represent a global threat. It brings to light issue regarding safety of existing stockpiles of unused POPs and the potential for more contamination incidents to occur. For instance, there are huge stockpiles of unwanted pesticides in Africa and Asia. These unused, unusable and unwanted products are often

stored in damaged containers under highly inadequate conditions. The main cause of the problem in Africa is excessive inappropriate pesticide donations given as donor assistance. Industry and agencies appear to be favouring incineration of obsolete stocks and contaminated soils. This is however a grossly unacceptable solution given many of the toxic products of incomplete combustion including PCDD/Fs and PCBs. A more viable solution is that manufacturers and donors must take responsibility for the obsolete stocks by repackaging them and safely storing them in the manufacturers home country to be subsequently disposed of by zero-emission technology when available (Johnston and Stringer 1992).

4.3. Brazilian lime waste

Shortly before the Belgian food contamination scandal, hazardous waste found its way into the European food chain from another source. However, this incident was by no means as widely reported. An increase in concentrations of dioxins in milk and butter (from a baseline of about 0.6-0.7 pg ITEQ g^{-1} fat) was noted in Germany in September 1997 through routine monitoring. In March 1998, milk from a tanker that collected from some 70 farms was found to contain 7.4 pg ITEQ g^{-1} fat. This was in excess of the German standards (5.0 pg ITEQ g^{-1} fat) and so was unfit for sale. Subsequent investigations determined that the source of the contamination was Brazilian citrus pulp pellets (CPP) which could constitute up to 25% of cattle feed. The contaminated feed was taken off the market across Europe and the milk contamination abated (Malisch 1998a; Malisch, Bruns-Weller et al. 1998; Traag, Mengelers et al. 1999). Beef was also contaminated, though no data are available (MacKenzie 1999). In 12 EU member states, approximately 92,000 tonnes of pulp pellets were either landfilled or destroyed (Malisch, Berger et al. 1999).

Around 1.5 million tonnes of pulp pellets, with an estimated value of 200-300 million US$ are sold each year. Around 60% of this comes from Brazil (Malisch 1998a; Malisch, Berger et al. 1999). The citrus pulp pellets are based on citrus pulp, peel and seeds (by-products of orange juice production) and contain approximately 2% lime, which absorbs some of the moisture and neutralises its natural acidity (EC 1999e; Malisch, Berger et al. 1999).

Waste lime produced by Solvay in Brazil was identified as the most likely source of the contamination (MacKenzie 1999). In January 1999 representatives of the European Commission visited Brazil to investigate the source of the dioxin-contaminated lime. In the report on this trip (EC 1999e) one specific supplier, which, until 1996, had generated “lime milk” as a by-product of acetylene production, and had approximately 1 million tonnes still in storage, was identified as the source of the contamination. This waste was supplied to another company who “converted” it into hydrated lime and then sold it on to the citrus pulp producers.

The EC report did not name any of the companies involved, but they did report the maximum concentration of dioxins found in samples from their sites. The maximum concentration reported from lime from the “converter” was 15 270 ng ITEQ kg^{-1} dry matter and the maximum concentration reported from the company that supplied them with lime was 56 111 ng ITEQ kg^{-1} dry matter. A report by a German consulting firm (ERGO 1998) provides a detailed data set for samples analysed for ABECitrus, the Brazilian association of citrus producers. The sampling was also documented by the Brazilian Ministry of

Agriculture. The ERGO report contains full analysis results for two samples with identical dioxin concentrations to the two described above. Examination of the sample data included in the ERGO report confirms that the "converter" was a company called Carbotex and that their supplier was indeed Solvay.

Comparison of dioxin congener profiles is always an important factor in determining the sources of dioxin contamination. ERGO (1998) reported that samples with a congener profile similar to the citrus pulp were found at the Carbotex site, but did not investigate the subject in detail. However, another consultants' report (CEGEQ 1999), found close similarities between the dioxin profile in citrus pulp analysed in Germany and that in several of the samples from the Solvay and Carbotex sites. They were even able to offer experimental evidence to explain that if the lime from Solvay were heated, as would happen during conversion and manufacturing of CPP, it could change the dioxin profile in a way that would explain some of the apparent discrepancies between samples.

CEGEQ also reported high concentrations of other toxic organochlorines in lime samples originating from Solvay. These included tetra-, penta-and hexa-chlorinated benzenes, PCBs, hydroxy-PCBs and polychlorinated diphenyl ethers. Chlorophenols were not found. Concentrations of PCBs were generally high in samples which were heavily contaminated with PCDD/Fs, though there was no very close correlation. PCB concentrations ranged up to 52 200 $\mu g\ kg^{-1}$ in a sample from the Solvay site. This same sample also contained the highest reported concentrations of chlorinated benzenes; 190 $\mu g\ kg^{-1}$ (tetrachlorobenzenes), 3 000 $\mu g\ kg^{-1}$ (pentachlorobenzene) and 4 800 $\mu g\ kg^{-1}$ (hexachlorobenzene). The concentration of dioxins in this sample was 32 434 ng ITEQ kg^{-1}, among the highest reported during this incident. However, another sample from this report, also from the Solvay site, had the highest dioxin contamination of all: 56 390 ng ITEQ kg^{-1} (CEGEQ 1999).

In July 1998, the EC passed Directive 98/60/EC (EC 1998), which set a maximum limit of 500 pg ITEQ kg^{-1} in citrus pulp pellets for use in animal feed. This has been retained in a subsequent Directive and proposed Directive (EC 1999d; EC 1999g). Although the new EC regulations, and the safeguards instituted in Brazil (EC 1999f) will protect European food sources from any further contamination from this source, there was scope for considerable concern if this highly contaminated material were used for other purposes. For example, contaminated lime had been used for construction in Brazil (EC 1999e). Consequently, after protracted negotiations with the State Prosecutor and other interested parties, in December 1999, Solvay signed an agreement to clean up the site (Paolo 1999).

4.4. Fires, smelters and incinerators:

There have been numerous incidents where atmospheric emissions of dioxins from a range of combustion sources have impacted on food production nearby. The following are reported almost exclusively from the most highly developed countries, which have the facilities and funding to monitor food dioxin concentrations. They must represent only a small fraction of the incidents that have occurred and continue to occur all around the world.

4.5. PVC fires

A fire in a metal-processing plant in Lingen, Germany, on 4 November, 1996, involved the burning of around 10 tonnes of PVC. An atmospheric HCl level of 4ppm was recorded and the PVC combustion residues contained dioxins (6.26 µg ITEQ kg^{-1}) unidentified extractable organohalogens (21 mg kg^{-1} EOX), PAHs (93 mg kg^{-1}) and lead (23 mg kg^{-1}). A high wind blowing at the time kept the fumes close to the ground and increased the impact of the fumes from the fire on the immediate locality. Although soil samples collected after the fire did not show greatly increased dioxin concentrations, leafy vegetables and other foodstuffs collected up to with 2 km downwind of the fire were severely affected and their consumption had to be restricted (Thiesen, Hamm et al. 1997).

Kale collected up to 900m down-wind of the fire site during the 11 days after the fire contained PCDD/Fs in excess of German intervention values of 1 ng ITEQ kg^{-1} wet weight or 10 ng ITEQ kg^{-1} dry weight. Concentrations of PAHs and copper were also as much as three times over "normal levels". Hen's eggs and meat were contaminated to a maximum of 53 pg ITEQ g^{-1} egg fat and 26 pg ITEQ g^{-1}, an order of magnitude over background levels. No reduction in the concentration of dioxins was observed in free-range eggs laid four weeks after the incident. All free-range chickens, ducks and rabbits bred for human consumption in the worst affected area (30^{o} sector, 1.3 km downwind of the fire) were slaughtered (Thiesen, Hamm et al. 1997).

Cows fed on affected grass produced milk in excess of the highest German intervention value (5.0 pg ITEQ g^{-1} milk fat) making it unfit for sale. However, the cattle were switched to uncontaminated feed and the milk fell below this level less than three weeks after the fire (Thiesen, Hamm et al. 1997).

A previous incident in Lengerich, Germany in October 1992, where 450 tonnes of PVC-containing plastics burned under similar meteorological conditions, also caused high levels of dioxins in foodstuffs. In this case, kale sampled the day after the fire contained 33.6 ng ITEQ kg^{-1} dry weight (0.36 ng ITEQ kg^{-1} wet weight). This was in excess of the German intervention values mentioned above, but not as high as the sample collected on the day after the Lingen fire (94.8 ng ITEQ kg^{-1} wet weigh). Milk levels also exceeded the 5.0 pg ITEQ g^{-1} milk fat limit, with a maximum of 5.9 pg ITEQ g^{-1} milk fat being reached between three and four weeks after the fire. Hen's eggs were not badly affected in this case (Thiesen, Hamm et al. 1997).

4.6. Metals processing

The US EPA (EPA 1998) found little evidence of dioxin production in primary copper processing, but problems often arise in recycling of copper, especially scrap electrical cables. Copper and PVC are the main constituents of cable waste together making up 75% of the waste. Although cryogenic or mechanical methods of separating the components of the cables are available, the most widely used recycling method for electrical cables is burning off the PVC coating to recover the copper. This can be done either in an incinerator/smelter or in the open air. Even in the most affluent countries, open burning was conducted until very recently. In the US, open burning continued until at least the

1970s and contamination was still evident in the 1990s (Harnly, Stephens et al. 1995). A similar picture appears in Europe, where Dutch investigators found contamination from sites where small scale illegal burning of scrap cables and cars was carried out. Some of these practices had continued into the 1980s (van Wijnen, Liem et al. 1992).

It is well known that reclamation of copper from cables, in either a controlled or an uncontrolled fashion, results in the emission of toxic organochlorines. These include dioxins and furans (PCDD/Fs) (Marklund, Kjeller et al. 1986; Christmann, Kasiske et al. 1989), polychlorinated biphenyls (PCBs), chlorinated benzenes and chlorinated phenols. It is also recognised that PVC plastic is a major contributor to the production of these pollutants (Aittola, Paasivirta et al. 1993).

In Taiwan, soil samples from sites where imported electrical cables and credit cards were burned were found to contain high levels of PCBs and PCDD/Fs. PCDD/Fs were present at above 1 μg kg^{-1} (Huang, Miyata et al. 1992). Nearby aquaculture ponds and the fish raised in them were highly contaminated (Lu, Miyata et al. 1995b), principally by the coplanar PCBs (Lu, Miyata et al. 1995a). The aquaculture industry provides a significant contribution to the protein consumption of the people of South Taiwan. Consumption of 50 g of fish per day was estimated to provide a maximum intake of 10 800 pg TEQ kg^{-1} body weight day^{-1}, 2 700 to 10 800 times higher than the acceptable daily intake recommended by the World Health Organisation (1-4 pg TEQ kg^{-1} body weight day^{-1}) (Lu, Miyata et al. 1995b).

Austria's only copper reclamation plant is sited in an Alpine valley where most of the surrounding area is used for dairy farming. The waste smelted included electronic scrap. Samples containing high concentrations of dioxins have been collected from around this site since 1987. Contamination has been found in ambient air, spruce needles, soil, hay, fodder grass, cow's milk, human blood and breast milk, though the breast milk was not regarded as "conspicuously high". Subsequent to the recognition of the very high levels of contaminant from the plant, plastic was banned from the input copper scrap and an afterburner was installed. These and other measures reduced the atmospheric loading of dioxins from 26 g ITEQ $year^{-1}$ up until 1988 to less than 1 g $year^{-1}$ from 1989 onwards. The discontinuation of processing of plastic-containing materials was attributed with reducing the concentrations of PCDD/Fs in fodder grass by about 40%. Concentrations in cow's milk collected in February and April 1988 ranged from 14.0 - 69.5 ng ITEQ kg^{-1}. The average concentrations were 49 ng ITEQ kg^{-1} during the winter of 1987/8 and gradually reduced until they reached an average of 5 ng ITEQ kg^{-1} in the winter of 1992/3. In spring 1993, concentrations were in the range of 1.0 to 2.1 ng ITEQ kg^{-1}, which is within the normal range. However, concentrations of PCDD/Fs in the grass did rise suddenly for a brief period in May 1992, possibly indicating that occasional exceptional emissions from the smelter might still impact on local farms. Of the five blood samples collected from farmers and family members, one was exceptionally high (946 ng ITEQ kg^{-1} blood fat). The lowest concentration was 41 ng ITEQ kg^{-1} and the average 246 ng ITEQ kg^{-1} (Riss, Hagenmaier et al. 1990; Riss 1993).

In the Netherlands, smelting of copper cables contaminated the milk of cattle grazing within a few kilometres of the smelting site. The amount of dioxin in the milk was raised approximately three-fold, sometimes exceeding the Dutch maximum allowable level in milk (6 pg TEQ g^{-1} milk fat). The PCDD/F pattern associated with burning PVC was clear,

allowing the researchers to distinguish the farms affected by metal reclamation from those affected by other pollution sources (Liem, Hoogerbrugge et al. 1990; Liem, Hoogerbrugge et al. 1991).

4.7. Municipal Waste Incineration

Local deposition of PCDD/Fs near to incinerators may cause serious contamination of vegetation and soil in the vicinity. In 1989/90, a study was conducted in the Netherlands to investigate the possible PCDD/F contamination of cow's milk from farms located near to municipal waste incinerators (MSWs), (Liem et al. 1991). The study found that increased levels of PCDD/Fs may occur in cow's milk in the vicinity of MSWs. Particularly high levels of PCDD/Fs (up to 13.5 pg TEQ/g fat) in milk from areas near Rotterdam ("Lickebaert area") and Zaandam. A potential health risk from such high levels could not be excluded and the Dutch government set an upper limit for dioxin levels in milk of 6 pg TEQ/g fat. Restrictions were put in place on the sale of milk from the affected areas. In April 1990, the MSW at Zaandam was closed. Subsequently, the levels of PCDD/Fs in the milk declined to levels below the 6 pg TEQ/g fat limit.

4.8. Hazardous waste incineration

The Coalite chemicals plant in Derbyshire, UK has manufactured organochlorine chemicals including chlorophenols since the 1960s (May 1973). In the early 1990s, severe dioxin pollution of the environment surrounding the plant was discovered (Berryman, Bennett et al. 1991; SAL 1991a; SAL 1991b; MAFF 1992c; MAFF 1992b; MAFF 1992d; MAFF 1992e). The first indications came when (in late 1990 and early 1991) the UK Ministry of Agriculture, Fisheries and Food (MAFF) collected samples of milk produced by several farms in the vicinity of Coalite. Milk from some of these farms were highly contaminated, with maximum concentrations of 1.8 ng ITEQ kg^{-1} and 1.9 ng ITEQ kg^{-1} whole milk at two farms producing milk for human consumption and 3.4 ng ITEQ kg^{-1} whole milk from a suckling herd at another farm (MAFF 1992c). These exceeded the UK's maximum tolerable concentration (MTC) for dioxins in milk, which was then set at 0.7 ng kg^{-1} ITEQ whole milk (MAFF 1992c; MAFF 1992a) and the three farms were prevented from selling their meat or dairy produce for over a year (ENDS 1993).

Her Majesty's Inspectorate of Pollution (HMIP) began trying to identify the source of the contamination on the farmland. The potential sources included a Seveso-type accident which had occurred on the site in 1968, a fire in a warehouse containing chlorophenols in 1986, waste treatment processes, ongoing organochlorine production processes and the on-site incinerator. It quickly became apparent that the incinerator in the chemicals plant was emitting significant amounts of dioxins (EA 1997; Sandalls, Berryman et al. 1997a). The incinerator was closed on the 30 November, 1991, and concentrations of dioxins in the milk of the local farms decreased thereafter.

In 1993, Coalite Products Ltd paid £200 000 in compensation (ENDS 1993) to the affected farmers in an out of court settlement (Leigh Day & Co Solicitors 1993). In 1996, Coalite Products Ltd was prosecuted for not having used the "best practicable means for preventing the escape of noxious or offensive gases" during incineration of their wastes. The company pleaded guilty and were fined £150 000 and ordered to pay the prosecution £300 000 costs as well as having to pay their own legal expenses (EA 1997). Statistical

analysis of the distribution of dioxins in the soil around the plant determined that the chances of the dioxins not having originated in or close to the Coalite plant was less than one in 10,000 (Sandalls, Berryman et al. 1997b).

The Coalite plant is still in operation and food contamination remains a concern; in 1996, there were two further milk contamination incidents. Increased concentrations of dioxins were found in milk from three farms close to Coalite (ENDS 1997b). The milk of the suckler herd, which had suffered the greatest contamination in 1991, again exceeded the MTC, which had been revised to 0.66 ng ITEQ kg^{-1} of whole milk (equivalent to 16.6 ng ITEQ kg^{-1} milk fat) for both dioxins and PCBs (MAFF 1998). The source of the contamination was not clear; it may have been partly a seasonal variation (MAFF 1998) and cattle ingesting mud during a wet spell was also suggested (ENDS 1997b). A farm near Rotherham, adjacent to the river Rother, was also affected, though the MTC was not breached. It was concluded that the most likely source was dioxins from Coalite deposited on the fields when the river flooded (ENDS 1997a; MAFF 1997a).

Routine monitoring in Germany found high levels of dioxins in a pheasant imported from close to the Bolsover site: the bird's fat contained 235 pg ITEQ g^{-1} (Malisch, Gleadle et al. 1999), compared with concentrations in chicken fat from the UK, Germany and the US which are in the region of 1pg ITEQ g^{-1} for samples collected during the 1990s (Fiedler, Cooper et al. 1997; Harrison, Wearne et al. 1998; Malisch 1998b). Although the congener profile cannot be assigned with certainty to a Coalite source, an association has not been ruled out (Malisch, Gleadle et al. 1999).

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