Dioxin production in the PEMEX PVC plant: possible environmental and health effects.

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Introduction

PVC is one of the most widely used plastics, with around 25 million tonnes being produced each year. Production of 429 000 tonnes was reported for Mexico in 1998 (Stringer & Johnston 2001). PEMEX produces the intermediates for PVC, vinyl chloride and ethylene dichloride at the Complejo Paharitos in Coatacoalcos, Vera Cruz state, Mexico.

In 2001, Greenpeace published a report on the environmental pollution around the plant (Stringer *et al.* 2001). Subsequently, a number of samples of hazardous wastes from the complex were obtained, specifically two samples of liquid waste and one sample of ash from the incinerator in which the liquid wastes are routinely burned. These were subjected to a comprehensive suite of analyses, including for chlorinated dioxins and furans, which are known waste products of both PVC manufacturing processes and incineration of chlorinated wastes.

The organic waste samples were characteristic of PVC production residues, including dioxin and furan profiles similar to those for wastes from other PVC sites around the world. The incinerator ash was also contaminated with dioxins resulting from the combustion of the chlorinated wastes.

Dioxins and furans are among the most toxic manmade chemicals ever produced. They cause a wide range of toxic effects in humans and animals, including cancer and damage to the reproductive, immune and hormone systems. The unborn and the developing young are especially sensitive to toxic effects of these pollutants. In addition to being extremely toxic, the dioxins and furans are very persistent in the environment. Moreover, they can build up in the food chain and are retained for many years in the human body. Dioxins released into the aquatic environment will accumulate in fish and other animals. Consequently, individuals who are exposed to elevated doses of dioxins either directly through working at the PEMEX plant or through regularly eating contaminated fish may be at risk of these health effects.

Sample collection and analysis

Samples were obtained from the PEMEX incinerators at the Paharitos Chemical complex at Coatzacoalcos, Vera Cruz State, Mexico, in April 2001. Two samples (AM01007 and AM01008) consisted of liquid wastes destined for the hazardous waste incinerator; the third (AM001009) was ash from the economiser or heat exchanger of the same incinerator.

All three samples were returned to the Greenpeace Research Laboratories, University of Exeter, for analysis, where they were screened to isolate and identify as many organic components as possible. Each sample also underwent quantitative analysis; the ash sample was analysed for a range of toxic metals and the two liquid wastes were analysed for ethylene dichloride (also known as 1,2-dichloroethane or EDC), one of the chemical intermediates in the PVC production process. Analytical methods for the analyses carried out at the Greenpeace Research Laboratories were unchanged from those used for the earlier environmental survey and reported in 2001 by Stringer and coworkers (Stringer *et al.* 2001).

In addition, subsamples were sent to an independent accredited laboratory for the determination of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs).

Results and discussion

Concentrations of the PVC intermediate 1,2-dichloroethane (EDC), a vital step in the production of PVC in the two liquid waste samples are given in Table 1 below. The findings that samples AM01007 and AM01008 contained 440g/l (44%) and 400g/l (40%) EDC respectively confirms that the wastes are from the PVC industry rather than any other of the processes operated at the Paharitos complex.

	AM01007	**AM01008**
Ethane, 1,2-dichloro-	440g/l	400g/l

Table 1: Quantitative results for 1,2-dichloroethane (EDC) in the two liquid waste samples.

Organic screen analysis of all three samples identified numerous other volatile and semivolatile organochlorines (Table 2). Many of these were also found during the earlier environmental survey (Stringer *et al.* 2001). Overall, these screening analyses establish a clear and direct link between the manufacture and disposal of PVC precursor chemicals and the environmental contamination around the Paharitos complex.

Concentrations of metals in the incinerator ash are shown in Table 3. The metal present in the highest concentrations was iron, almost certainly a result of the corrosion of the structure of the incinerator by hydrogen chloride, the acidic gas generated by the burning of organochlorines. This is supported by the presence of manganese, nickel and chromium, also components of steel.

Finally, the samples were analysed for dioxins (PCDDs) and furans (PCDFs). There are 135 PCDDs and 175 PCDFs, which vary in the number and location of the chlorine atoms in the molecule. The individual compounds are called congeners and the analysis focuses on the seventeen congeners which have chlorine atoms in positions 2,3,7 and 8, and which are especially toxic.

Because the toxicity of the individual PCDD/F congeners varies, the concentration of each congener is multiplied by a toxicity equivalence factor (TEF) and the results added to give a total toxicity equivalence value (TEQ) for the sample. Table 4 presents the concentrations of each of the seventeen 2,3,7,8-substituted congeners and their estimated toxicity (TEQs) according to the most commonly used schemes, the ITEFs and WHO mammalian TEFs.

It is well established that the oxychlorination process used to manufacture EDC produces PCDD/Fs (Evers 1989, ICI 1994, Stringer *et al.* 1995). Some are also present in the vinyl chloride wastes. The extremely high content of EDC in the liquid wastes described above, indicate that they primarily consist of EDC production wastes. Purification of the reaction mix results first in the most volatile impurities being removed from the mix- the light ends. Subsequently, pure EDC is distilled off, leaving behind the least volatile impurities, variously known as “still bottoms” or “heavy ends”. Since the PCDD/Fs are less volatile than EDC, they tend to be present at the highest concentrations in the heavy ends. For example, ICI (1994) reported 0.027 ng/kg ITEQ in light ends, but 3100- 7560 ng/kg ITEQ in heavy ends. Stringer *et al.* (1995a) reported a maximum of 6 370 000 ng/kg in one sample of EDC heavy ends collected from a facility in the USA in 1994.

Sample numbers		
AM01007	AM01008	AM01009
Sample descriptions		
Liquid organic waste	Liquid organic waste	Incineration ash
Number of compounds isolated		
63	86	37
Compounds identified to better than 90%		
1,3-butadiene, 2-chloro-	1,3-butadiene, 2-chloro-	14-.beta.-h-pregna
1-propene, 1,1,2-trichloro-	1,3-BUTADIENE, HEXACHLORO-	1-docosene
1-propene, 1-chloro-2-methyl-	1-propene, 1,1,2-trichloro-	1-dodecene
1-propene, 3-chloro-2-(chloromethyl)-	1-propene, 3-chloro-2-(chloromethyl)-	1-hexadecanol
2-butene, 1,3-dichloro-, cis-	2-butene, 1,3-dichloro-, cis-	1-octadecene
2-butene, 1,3-dichloro-, trans-	2-butene, 1,3-dichloro-, trans-	1-tridecene
2-BUTENE, 1,4-DICHLORO-, TRANS-	2-BUTENE, 1,4-DICHLORO-, CIS-	BENZENE, 1,2,3,4-TETRACHLORO- *
benzene	2-butene, 1,4-dichloro-, trans-	BENZENE, 1,2,3,5-TETRACHLORO- *
BENZENE, 1,2-DICHLORO-	benzene	BENZENE, 1,2,4,5-TETRACHLORO- *
benzene, chloro-	BENZENE, 1,2-DICHLORO-	BENZENE, 1,2,4-TRICHLORO- *
CHLOROFORM	benzene, chloro-	benzene, 1,2-dimethyl-
ethane, 1,1,1,2-tetrachloro-	CHLOROFORM	BENZENE, 1,3-DICHLORO- *
ETHANE, 1,1,2,2-TETRACHLORO-	ethane, 1,1,1,2-tetrachloro-	benzene, 1,3-dimethyl-
ethane, 1,1,2-trichloro-	ETHANE, 1,1,2,2-TETRACHLORO-	BENZENE, 1,4-DICHLORO- *
ETHANE, 1,1-DICHLORO-	ethane, 1,1,2-trichloro-	benzene, chloro- *
ethane, 1,1'-oxybis[2-chloro- (chlorex)	ETHANE, 1,1-DICHLORO-	BENZENE, HEXACHLORO-
ETHANE, 1,2-DICHLORO- (EDC)	ethane, 1,1'-oxybis[2-chloro- (chlorex)	BENZENE, PENTACHLORO- *
ethane, 1-bromo-2-chloro-	ETHANE, 1,2-DICHLORO- (EDC)	cyclohexane, 1,3-dimethyl-
ethane, pentachloro-	ethane, 1-bromo-2-chloro-	cyclohexane, 1,4-dimethyl-
ETHENE, 1,1-DICHLORO-	ethane, pentachloro-	cyclohexane, ethyl-
ETHENE, 1,2-DICHLORO-, CIS-	ETHENE, 1,1-DICHLORO-	eicosane
ETHENE, 1,2-DICHLORO-, TRANS-	ETHENE, 1,2-DICHLORO-, CIS-	ETHANE, 1,1,2,2-TETRACHLORO-
ETHENE, CHLORO- (VINYL CHLORIDE)	ETHENE, 1,2-DICHLORO-, TRANS-	ethylbenzene
ETHENE, TETRACHLORO-	ETHENE, CHLORO- (VINYL CHLORIDE)	heptadecane
ETHENE, TRICHLORO-	ETHENE, TRICHLORO-	octachlorostyrene
METHANE, TETRACHLORO-	ETHENE, TETRACHLORO-	octane
	METHANE, TETRACHLORO-	pentadecane
		phenanthrene
		styrene
		tetradecane

Table 2: Organic contaminants found in the two waste samples and one ash sample. Pollutants given in capital letters were identified in discharges from the Paharitos complex or in the local environment (Stringer et al. *2001). Compounds were identified by GC/MS screening except for those marked with an asterisk* which were identified using selected ion monitoring.*

METAL	(mg/kg dry weight)
Cadmium	<1
Chromium	1444
Cobalt	37
Copper	518
Iron	534574
Lead	147
Manganese	3312
Mercury	0.78
Nickel	1550
Zinc	945

Table 3: Metals concentrations in sample AM01009, ash from the PEMEX incinerator.

Heavy ends would typically be expected to exhibit a tarry consistency, whereas samples AM01007 and AM01008 were colourless and highly fluid and are therefore most likely to represent either light ends or a mixture of light and heavy ends. The concentration of PCDD/Fs in the two liquid waste samples (around 400 ng/l ITEQ) are between those in light ends and those in heavy ends, though it remains possible that other wastes generated within the facility contain far higher concentrations of the dioxins and furans.

Examination of "congener profiles", showing the relative concentrations of different PCDD/F congeners is an important tool in identifying the source of dioxin and furan contamination. Wastes from the PVC industry exhibit a distinctive pattern, dominated by octachloro-dibenzofuran (OCDF). In many cases, the OCDF represents over 90% of the total concentrations of 2,3,7,8-substituted congeners (see eg ICI 1994, Stringer *et al.* 1995). Both the EDC waste samples show this pattern, as illustrated in Figure 1, providing further indications that these wastes are derived from the PVC production process.

The ash sample shows a different pattern, due to reactions taking place in the incinerator. This profile is more similar to some obtained from locations where PVC wire and similar materials have been burned (Harnly *et al.* 1995, van Wijnen *et al.* 1992).

It was beyond the scope of this study to determine dioxin concentrations in the environment. However, the results from these three samples combined with those from our earlier study confirm that the pollutants produced in the PVC production process are being released into the environment of Coatzacoalcos. If this is the case for so many organochlorines, it is must also be so for the PCDD/Fs.

The conclusion that dioxins and furans are being released into the environment by the PEMEX operations has serious implications. The PCDD/Fs are arguably the most hazardous industrial pollutants in existence. As well as being highly toxic, they are extremely persistent, which means that they can remain in the environment for thousands of years (Hashimoto *et al.* 1990). They are very poorly soluble in water, but dissolve instead in fat. Consequently, they are taken up from the environment by animals such as fish and can build up in the food chain and in humans.

	AM01007, liquid waste			AM01008, liquid waste			AM01009, ash		
	concentra-tion	ITEQ	WHO TEQ	concentra-tion	ITEQ	WHO TEQ	concentra-tion	ITEQ	WHO TEQ
Dioxins	(ng/l)	(ng/l)	(ng/l)	(ng/l)	(ng/l)	(ng/ml)	(ng/kg)	(ng/g)	(ng/g)
2378-TCDD	n/d	n/d	n/d	n/d	n/d	n/d	2.5	2.5	2.5
12378-PeCDD	n/d	n/d	n/d	n/d	n/d	n/d	78	39	78
123478-HxCDD	20	2	2	30	3	3	220	22	22
123678-HxCDD	40	4	4	30	3	3	450	45	45
123789-HxCDD	40	4	4	40	4	4	330	33	33
1234678-HpCDD	32	0.32	0.32	40	.4	0.4	4500	45	45
OCDD	91	0.091	0.0091	110	0.11	0.011	12000	12	1.2
Furans									
2378-TCDF	n/d	n/d	n/d	n/d	n/d	n/d	18	1.8	1.8
12378-PeCDF	n/d	n/d	n/d	n/d	n/d	n/d	330	16	16
23478-PeCDF	n/d	n/d	n/d	n/d	n/d	n/d	210	110	110
123478-HxCDF	540	54	54	500	50	50	3100	310	310
123678-HxCDF	640	64	64	570	57	57	930	93	93
123789-HxCDF	710	71	71	610	61	61	430	43	43
234678-HxCDF	50	5	5	90	9	9	790	79	79
1234678-HpCDF	2400	24	5	2900	29	29	11000	110	110
1234789-HpCDF	290	2.9	2.9	270	2.7	2.7	6800	68	68
OCDF	170000	170	17	175000	175	17.5	19000	19	1.9
TOTAL	**174700**	**401**	**248**	**180000**	**394**	**236**	**61000**	**1040**	**1050**
	(ng/l)	(ng/l)	(ng/l)	(ng/l)	(ng/l)	(ng/l)	(ng/kg)	(ng/g)	(ng/g)

Table 4: Concentrations of 2,3,7,8-substituted dibenzo-p-dioxins and dibenzofurans in waste samples from the Complejo Paharitos. Data are given in terms of concentrations of each congeners and toxic equivalents (TEQs) according to the International TEF and WHO mammalian TEF schemes.

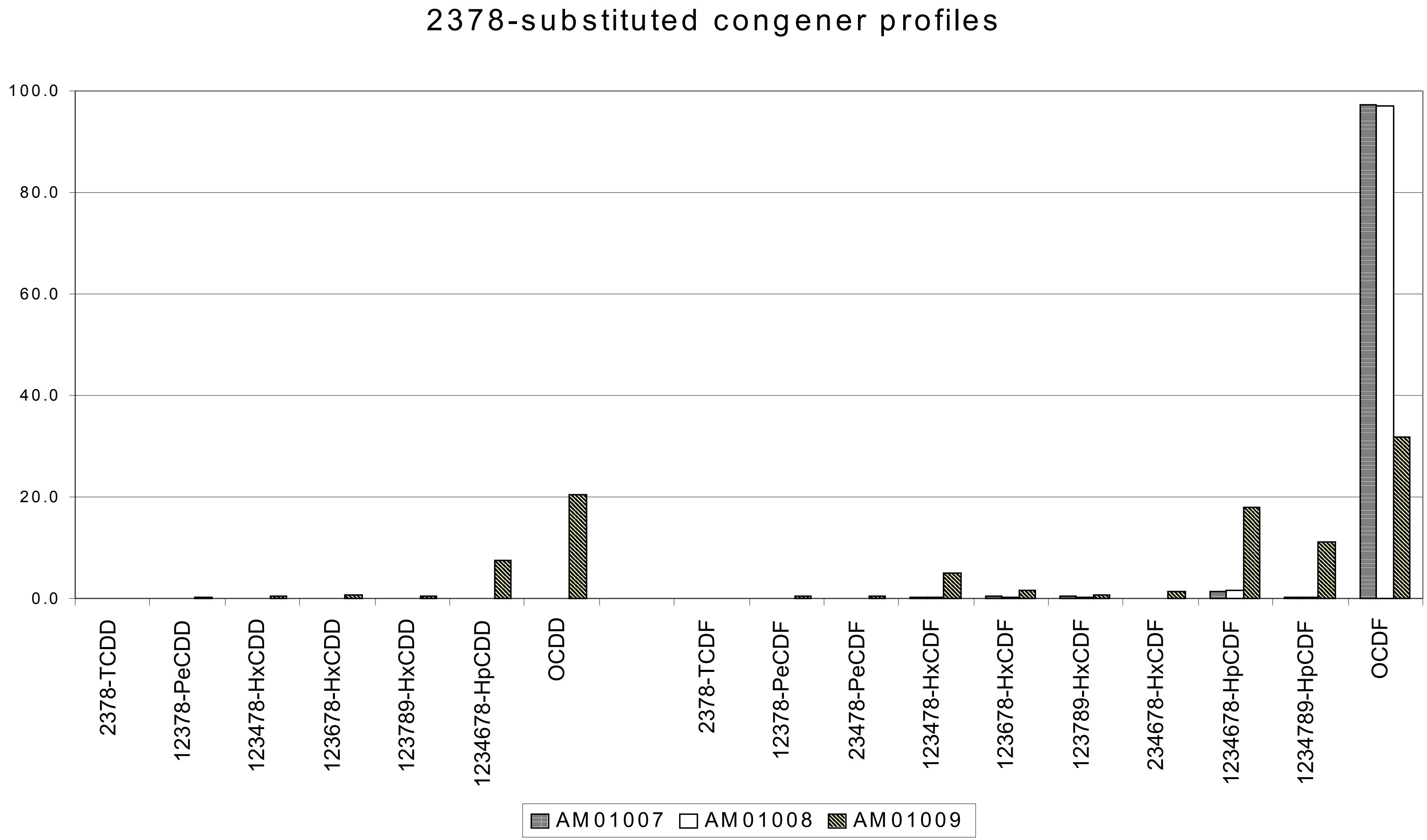

Figure 1: 2,3,7,8-Substituted congener profiles for liquid wastes and incinerator ash from PEMEX, Coatzacoalcos.

Humans typically receive 90% of their dioxin intake from food (DHHS 2001, EC 2001). Once in the body, they are only eliminated slowly. Half a person's body burden of dioxin would be eliminated over a period of about 7 to 10 years (USEPA 2000). However, since they are taking in more each day via their diets, an individual's dioxin body burden will tend to increase gradually through their lifetime. The only notable exception to this is that when women have children, they pass some of their body burden on to their children in the womb and subsequently during breastfeeding.

Studies of workers in others areas of the chemical industry show that workers who have to handle dioxin-contaminated wastes can become contaminated (see eg Svensson *et al.* 1993, Stringer *et al.* 1995b). However, other workers may also be vulnerable; in one study undertaken at a UK factory, an electrician exhibited comparatively high level of PCDD/Fs in his blood. The delicate nature of the work would have necessitated him to work without gloves, making him vulnerable to exposure through his skin.

Thus there are two groups of people in Coatzacoalcos who are at risk from the dioxins produced at the PEMEX plant; plant workers, and local people who eat locally caught fish or shellfish.

The extent of contamination of fish around Coatzacoalcos and its impact on the local ecosystem – including the fishing community- has not been documented and is hard to predict. Factors such as the trophic level and migratory patterns of marine organisms can significantly influence the extent to which they will accumulate PCDD/Fs from the environment. Similarly, there are no data on the exposure of the workers within the plant.

What is certain, however, is the serious nature of the health effects associated with exposure to PCDD/Fs. These are understood as a result of decades of research. Almost all laboratory research has been carried out on the most toxic congener, 2,3,7,8-TCDD, but the other sixteen 2,3,7,8-substituted dioxin and furan congeners act through the same mechanism and are assumed to exert the same effects. Studies of people exposed via dietary or industrial routes may be complicated by exposure to other chemicals as well as the polychlorinated dibenzo-p-dioxins and dibenzofurans. There is also some evidence that the effects vary depending on whether exposure is as a results of a single large dose, as might occur in an industrial setting, or due to repeated small doses more typical of dietary exposure. As with many sorts of toxic substances, the unborn and the developing young are more sensitive to dioxins and furans than are adults. However, there is comparatively little research on the effects on children, or, indeed, women, since most human studies have been conducted on the adult males employed at chemical factories. Despite these complications, the whole body of research on humans and animals provides a clear picture of the risks posed by dioxin exposure.

Table 5 below summarises many of the toxic effects that have been seen in humans or animals. 2,3,7,8-TCDD is an extremely potent multi-site carcinogen in animals and is also classified as a human carcinogen by both the International Agency for Research on Cancer (IARC) and the US Department of Health and Human Services (DHHS 2001). Several studies of workers suffering dioxin exposure showed elevated rates of cancer (USEPA 2000, DHHS 2001) and calculations show that it is possible that between one in 10,000 and one in 1000 ordinary people in the USA might suffer dioxin-related cancers (USEPA 2000).

Dioxin also affects several of the most important systems of the body; the immune system, the reproductive system and the hormone systems. Understandably, there is less research on the impacts of dioxins on the human immune system than there is on animal systems and the exact impacts may be difficult to predict in part because of the complexity of the immune response. Nevertheless, there is evidence of increases in infections suffered by children exposed to higher than normal levels of dioxins and dioxin-related compounds both through a short-term high dose and through longer term lower doses. Exposed workers showed a range of alterations in immune parameters, although with no consistent pattern.

This evidence, together with the laboratory studies, shows there is a real risk of immune disruption in humans. Moreover, because the animal research shows effects at extremely low concentrations, immune disruption is one of the effects of greatest concern to regulatory authorities such as the European Commission (CEC 2001, USEPA 2000).

Carcinogenesis
Carcinogenic to humans and animals
Immune system effects
Suppression of cell-mediated and humoral immunity; increased susceptibility to infectious challenge; auto-immune response
Male reproductive toxicity
Reduced sperm count; testicular atrophy; abnormal testis structure; reduced size of genital organs; feminized hormonal responses; feminized behavioural responses
Female reproductive toxicity
Decreased fertility; inability to maintain pregnancy; ovarian dysfunction; endometriosis
Developmental impacts
Birth defects; foetal death; impaired neurological development and subsequent cognitive deficits; altered sexual development Modulation of hormones, receptors, and growth factors Steroid hormones and receptors (androgens, estrogens and glucocorticoids); thyroid hormones; insulin; melatonin; vitamin A; EGF and receptor; TGF-a and TGF-b; TNF-a, IL-1b, c-Ras, c-ErbA
Other effects
Organ toxicity (liver, spleen, thymus, skin); diabetes; weight loss; wasting syndrome; altered fat and glucose metabolism

Table 5: Toxicological effects of dioxin (adapted from DHHS 2001, USEPA 1994, USEPA 2000 and Birnbaum 1994).

Both the male and female reproductive systems can be affected by the dioxins and furans. This is in part as a result of ability to disrupt the action of hormones which is exhibited by dioxin-like compounds. In particular, the action of the female hormone oestrogen are blocked and levels of the male hormone testosterone can be altered (USEPA 2000).

Overall, the male reproductive system seems to be more sensitive than the female system. There is little evidence that human fertility is reduced, although one study conducted after an industrial accident at Seveso in Italy showed an alteration in the ratio of boys and girls being born. While this observation has not been repeated elsewhere, there are many other studies where the children of exposed parents do show clear developmental effects. In one of the worst cases, in Taiwan, where rice oil became contaminated with PCBs and dibenzofurans, eight out of thirty-nine children whose mothers were pregnant at the time of the poisoning incident died during the first few years of life. The surviving children, as well as those who were born shortly before the incident and were therefore exposed via breastfeeding, showed a range of effects including impacts on the skin, liver and conjunctivitis. As they developed they were consistently smaller and lighter than unexposed children. They showed increased susceptibility to infection, and their neurological and behavioural development was impaired. The boys' neurological development was worse than that of the girls, possibly because of gender-specific endocrine disruption. Moreover the boys showed preliminary evidence of reduced sexual development, which is also highly dependent on correct hormonal control (Stringer & Johnston 2001, USEPA 2000).

Other effects which might occur as a result of exposure to dioxins and related compounds include altered liver chemistry and enzyme levels, diabetes and endometriosis (USEPA 2000).

Many people are already potentially exposed to doses of dioxins through their diets that could cause health effects (deVito *et al.* 1995, USEPA 2000). The most likely effects include biochemical alterations, diabetes, endometriosis and reduced developmental, immune and reproductive capability (deVito *et al.* 1995). Also, as mentioned earlier, there may also be an increased incidence of cancer in the general population (USEPA 2000).

The World Health Organisation has also acknowledged that subtle effects may be occurring in the industrialised countries and urged that measures be taken to reduce overall exposure to dioxins and related compounds via the diet to the lower end of their recommended tolerable daily intake (WHO 1998, van Leeuwen & Younes 1998). The EC has set maximum levels of dioxins and furans in food to remove highly contaminated food from the market. These come into force in July 2002 (EC 2001) and lower "action" and "target" concentrations will be set in the future to try to reduce dietary exposure further (CEC 2001).

It is impossible to estimate the extent of increased exposure to workers or the local population of Coatzacoalcos and what, if any effects may be being experienced. The necessary research would be extremely costly and would take years to complete. Moreover, since many of the effects may be subtle, it may be very difficult to prove beyond doubt the causes of any effects with current epidemiological techniques. Nevertheless it is evident that the PEMEX plant is generating wastes which are heavily contaminated with chlorinated dioxins and other chlorinated chemicals, and that it is acting as a source of hazardous chemicals into the surrounding environment.

Conclusions

The manufacturing of PVC precursor chemicals, predominantly 1,2-dichloroethane (EDC), and the subsequent incineration of the wastes generated, is contaminating the environment around the Paharitos Petrochemical Complex in Coatzacoalcos with toxic organochlorines. The evidence form this study indicates that among these are the polychlorinated dibenzo-p-dioxins and dibenzofurans, widely regarded as the most toxic man-made pollutants in existence.

The PCDD/Fs are toxic, persistent and bioaccumulative. They will remain in the environment effectively indefinitely and will be taken up into the local food web. This could lead to increased body burdens among consumers of seafood from this area. Plant workers may also be exposed during the course of their employment.

The conclusions from decades of research into the effects of dioxin and related chemicals in both humans and animals are that there is a real threat of serious health impacts including cancer, immune deficits, reproductive impacts and impaired neurological development. The only justifiable response to this threat is to reduce the exposure of the workers and population to dioxins and furans produced by the manufacturing of PVC precursors by PEMEX and the incineration of hazardous chlorinated wastes. The swiftest and surest way accomplish this would be to cease the production of all chlorinated products and replace them with less hazardous substitutes.

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