REVIEW OF DIOXIN EMISSIONS IN HONG KONG

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Comment on "An Assessment of Dioxin Emissions in Hong Kong" by Environmental Resources Management (ERM).

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1. INTRODUCTION

1.1 In December 1999 I was commissioned by Environmental Protection Department, Government of the Hong Kong Special Administrative Region (HKSAR) of the People's Republic of China to perform a study on "Review of Dioxin Emissions in Hong Kong" performed by Environmental Resources Management (ERM).

In my report I will discuss the ERM report and before that also some new data, which are not covered in the ERM report.

1.2 DIOXINS AND DIBENZOFURANS

Polychlorinated dibenzo-*para*-dioxins (PCDDs) and dibenzofurans (PCDFs) are chlorinated aromatic compounds with similar chemical properties. All are solids with high melting and boiling points, limited solubility in water and low vapour pressure. [1] The structures of the parent compounds are given in Fig. 1.



Fig. 1. Structural formulas of PCDDs, PCDFs and 2.3,7,8-

The generic term "dioxins" includes all PCDDs and PCDFs primarily those with 4,5,6,7 and 8 chlorine atoms. A large number of different combinations of both the number of chlorine atoms and position of substitution is possible, and as a result there are 75 different PCDDs and 135 PCDFs, or altogether 210 different compounds. [1] The number of isomers of PCDDs and PCDFs for a certain number of chlorine atoms is given in Table 2.1a in the ERM report.

The toxicity varies substantially among the different PCDDs and PCDFs. It is generally accepted that only 17 out of the 210 dioxin and dibenzofuran congeners are toxic. [1,2] The congener with the greatest toxic potency, and for which the greatest amount of toxicological information is available, is 2,3,7,8-tetra-CDD, see Fig. 1. In an attempt the TEF/TEQ approach has been adopted internationally [1,2]. Numerical Toxic Equivalent Factors (TEFs) have been developed, which enables the conversion from the concentrations of 2,3,7,8-tetraCDD based on their concentration to the toxicity.

In Table 2.2a in the ERM report the current Toxic Equivalent Factors (I-TEFs) for the 17 toxic dioxins and dibenzofurans are given. In June 1997 a group of experts assigned new TEF for Mammals/ Humans, Fish and Birds, including PCDDs, PCDFs and also the factors for the dioxinlike PCBs. The result for Mammals/Humans are collected and given see Table 2.2a in the ERM report. This scheme has been called the WHO-TEFs. [3] The TEF approach can be used to transform analytical results of total sum of all PCDDs and PCDFs into toxic equivalents, TEQs. [2,3] My personal advice is that if you will start an investigation now, you should analyze the PCDDs, PCDFs and also the dioxinlike PCBs and include the data in the WHO-TEF system. At the same time you have the value of the I-TEF and it is easy to compare this new value with the earlier results. This is true for the food data and later on it could be good to have the PCB-values for the CTWC.

The commonly used sub-units of a gram are: milligram, mg (1/1000) microgram, μg (1/1000 000) nanogram, ng (1/1000 000 000) picogram, pg (1/1000 000 000 000) femtogram, fg (1/1000 000 000 000)

1.3 SOURCES

It is now well established that PCDDs and PCDFs are ubiquitous all over the world, even in the Arctic and Antarctica. [4] They also undergo long-range aerial transport. [5]

Environmental contamination by PCDDs and PCDFs can be attributed to a series of primary sources like effluents, air, water and biota, while human exposure is due to secondary sources including food intake, (>98%), inhalation of air, drinking water and dermal contact. Other secondary sources are abiotic reservoirs like soil and sediments. The identified primary sources are mainly anthropogenic, but natural formation of PCDDs and PCDFs has also been described, see below. The primary sources can be divided into four categories.

During many *chemical reactions* it has been found that PCDDs and PCDFs are formed as un-wanted byproducts. As a result many pesticides and industrial-chemical products, including chlorophenols and chlorophenoxy herbicides (2,4,5-T) and PCBs have been strictly regulated or banned in recent years. Another chemical process generating PCDDs and PCDFs is the bleaching of pulp with chlorine gas. [6]

Combustion processes are considered to be another important primary source of PCDDs and PCDFs. Most thermal reactions which involve burning of chlorinated organic or inorganic compounds result in the formation of PCDDs and PCDFs. Of special importance is the incineration of various types of wastes like municipal (MSW), hospital and hazardous wastes (see below) and the production of iron and steel and other metals like copper, magnesium, nickel. *Photochemical reactions* under atmospheric conditions or aerial transport can result in the formation of PCDDs and PCDFs, as well as in the degradation of these compounds. These reactions are of interest, since most combustion and incineration sources produce emissions that undergo long-range aerial transport where they can be degraded by sun-light. [5]

Besides non biological formation processes, *some biological processes* can result in the formation of PCDDs and PCDFs. Our first example including mixing of chlorophenols, hydrogen peroxide and a series of peroxidases at room temperature resulted in the formation of PCDDs and PCDFs. [7] Later on, in addition to this evidence from in vitro experiments, these reactions can also occur under true *in vivo* or environmental conditions such as partly in sewage sludge and compost, see also below. [8,9]

1.4 INCINERATION

The formation of dioxins (PCDDs) and dibenzofurans (PCDFs) during incineration of waste has been discussed for more than 20 years now. In March 1977 I was working together with H.R. Buser in Switzerland and we analyzed fly ash samples from two incinerators in Switzerland, one burning municipal solid waste (MSW) and the other chemical waste. In both cases we were able to identify a long series of PCDDs and PCDFs, including 2,3,7,8-tetraCDD. Before we had the opportunity to publish our data it was reported by professor Otto Hutzinger in June 1977 at a meeting in Italy that Dutch incinerators generated a series of compounds which could be identified as PCDDs and PCDFs. Later in 1977 this investigation was published. [10] The Swiss results were published in 1978. [11, 12].

During the period 1978-1982 a series of papers, reports, and reviews were published confirming the original findings regarding the contamination of fly ash. During this period less data have been published on the levels of PCDDs and PCDFs in other incineration by-products, e.g., particulates and flue gas condensate, and totally in flue gas, which are the true emissions. [13] During the second part of the 1980s and 1990s safe sampling methods have been developed, and today there is an overwhelming database on the contamination of PCDDs and PCDFs in the flue gas from various incinerators.

The emission from an incinerator are the flue gas, the fly ash and the slag. Up to now most studies are on the flue gas and some also on the fly ash and the slag. The levels of PCDDs and PCDFs in the fly ash and in the slag were quite high in the 1970s and 1980s, but with the technology used today it has dramatically decreased and in many samples they are difficult to measure. [14] The concentrations decrease with the temperature in the oven due to the volatility of PCDDs and PCDFs. However, in Sweden and in many other countries the fly ash and slag will be sent to a secure landfill, where the leachate will be collected and analyzed.

Studies in Japan and in Italy have recently shown that a secondary treatment of the fly ash and slag will result in detoxification and reduction of the amount of incinerator residues. The PCDDs and PCDFs were found to be reduced by more than 99% in this secondary melting process, [15, 16].

Incineration can be performed by many different technologies, partly depending on the nature of the waste namely:

Municipal solid waste (MSW) Open burning of houshold waste Hospital waste Chemical waste Spontaneous landfill fires House heating

1.4.1. Municipal Solid Waste

In March 1986, a working group of experts convened by the World Health Organization Regional Office for Europe reviewed the available data on emissions of PCDDs and PCDFs from municipal solid waste (MSW) incinerators. [13] It was found that the origin of these compounds was not completely understood, but they appear to result from a serious of complex thermal reactions occurring during periods of poor combustion or during the cooling period. Because of their high thermal stability, the PCDDs and PCDFs can be destroyed only after adequate residence times at temperatures above 800°C. [13, 17]

In 1986 Sweden introduced a guideline of 0.1 ng I-TEQ/m³ for MSW incinerators. [18] In the beginning this value was considered to be a too low value for a normal MSW incinerator. However today this is an operating guideline in most EU countries. The guideline now proposed by the US EPA corresponds to 0.2 ng I-TEQ m³. Japan has now accepted the 0.1 ng I-TEQ/m³ for their numerous MSW incinerators. Incinerators fulfilling this strict guideline can now be found in Sweden, Germany, Austria, The Netherlands, Japan, Hong Kong and Spain.

The method to achieve this dramatic reduction is based on improved burning technology sometimes including circulating fluidized beds, followed by flue gas cleaning technology as dry scrubbing, or wet scrubbing, use of special absorbents and also catalysts, which can destroy the PCDDs and PCDFs formed. In the countries, where these good incinerators operate, I understand that they are accepted by the authorities and also by the public. It has also been found that the inlet air to a normal MSW incinerator also contains some PCDDs and PCDFs.

1.4.2. Open burning of household wastes

Very few studies have been performed on emissions of PCDDs and PCDFs from the open buring of household waste in barrels. However, very recently Lemieux et al [19] reported on such a study. The condition used in these studies include the following features:

- 1) poor gas-phase mixing
- 2) low combustion temperatures
- 3) oxygen-starved conditions
- 4) high particulate-matter loading
- 5) particulate-matter bound copper
- 6) presence of HCl and/or Cl_2
- 7) significant gas-phase residence time in the lower temperature range (250-700°C)

In this study they used two different composition of the waste a) a non-recycler b) and avid recycler. The composition of their waste varied greatly, especially paper and bottles. They also found a higher value for PVC in the waste from the avid recycler.

The concentrations of the PCDDs and PCDFs emissions were higher for the avid recycler (two experiments) than for the non-recycler (two experiments). The chlorobenzenes were also found to follow this trend. In the four studies they found 5.4 and 1.2 μ g/kg for the avid recycler and 0.75 and 0.90 μ g/kg for the non-recycler. [19]

The study also include a comparison of these burn barrel incineration with emissions from various full scale incinerators. The burned barrel are found to be much higher I-TEQ values than for MSW that possess air pollution control equipment. This comparison is using an American generation rate and a MSW rate of 200 ton/day and the total emissions of PCDDs and PCDFs. They come to the conclusion that 2.5 household of avid recycler or 37 non-recycler are equivalent to the amount comparable to a well-operated RDF/MSW facility serving 37 000 non-recycling and 121 000 of avid recycling households. [19]

1.4.3. Hospital waste incinerators

Hospital waste incinerators have earlier been small units operating under bad burning conditions, without modern flue gas cleaning technology. They are found to emit more than > 0.1 ng I-TEQ/m³ (earlier up to several hundred ng I-TEQ/m³). They are being closed down in many countries. [1] In the Nordic countries the hospital wastes are sent to the MSW incinerators and follow the rules for these incinerators. It is my understanding that the same rules are followed in Germany and The Netherlands, but this has not been discussed in the literature.

1.4.4. Chemical Waste Incineration

Concerning the incineration of chemical waste, two different technologies have been used or are still in use.

A. Rotary Kiln

In this type of incinerator the feedstock is primarily *semisolid*, hazardous waste in drums, which is burned together with additional fuel. Example of this type is the SAKAB incinerator at Norrtorp in Sweden, which has been used for the destruction of PCB from capacitors. [20] In the beginning (1983) the guideline was 3 ng I-TEQ/m³, but some years later this was lowered to 0.1 ng I-TEQ/m³.

Additional dry scrubbing was introduced to achieve this new guideline.

B. Thermal oxidiser

Thermal oxidisers are used for *liquid solvents and gases*. Even chlorinated solvents with a chlorine content exceeding 50% Cl can be completely destroyed at temperatures above 1400°C. As a consequence, the destruction has shown to be very efficient and the garanteed emission values for dioxins are quite low, far below 0.1 ng I-TEQ/m³. These thermal oxidisers are totally accepted by the authorities in Germany and USA.

1.4.5. Landfill fires

There have been many reports on the formation of PCDDs and PCDFs from the MSW incineration, but only two reports on the formation of PCDDs and PCDFs from spontaneous landfill fires. [21, 22] According to an inquiry among Finnish waste landfil personel and the personel in fire departments, there are approximately 380 fires per year in Finnish waste landfills. In Sweden it has been estimated that it will be 220 fires/year. In pilot tests in Sweden it was estimated that the TEQ concentrations in these tests were in the range of 66-518 ng I-TEQ/m³. In the real study from Finland they reported a value of 0.05--0.43 ng I-TEQ/m³. The congener pattern are the same as in MSW fires and the authors claim that the acceptable daily intake by the personnel of PCDDs and PCDFs are exceeded. For that reason the authors claim that protective breathing equipment must be used by those working to distinguish a waste landfill fire.

1.4.6. House heating

In several studies it has been found that normal house heating can be found to generate PCDDs and PCDFs.

A. Wood

In a study performed in Switzerland by Schatowitz et al [23] they found that combustion of different types of wood could generate PCDDs and PCDFs, see Table 1. The concentrations ranged from 0.019 up to 14.42 ng I-TEQ/m³. In this study where they also used the combustible part of household waste they found the concentrations of the PCDDs and PCDFs to be 114.4 ng I-TEQ/m³.

Fuel	Furnace	ng I-TEQ/m ³ this study	ng I-TEQ/m ³ Literature
Beech wood sticks	Fire-place	0.064	0.072
Beech wood sticks	Stick wood boiler	0.019-0.034	0.064
Wood chips	Automatic chip furnaces	0.066-0.214	0.006-0.12
Chipboards uncoated	Automatic chip furnaces	0.024-0.076	0.001-0.021
Waste wood	Automatic chip furnaces	2.70-14.42	0.10-4.18
Combustible part of Household waste	Household stove closed	114.4	

 Table 1. PCDDs and PCDFs emissions from wood combustion: comparative figures

Based on this study Schatowitz found that the annual emissions of PCDDs and PCDFs from wood combustion in Switzerland should be in the range of 6.94-43.79 g I-TEQ. [23]

A study by Schramm et al [24] from Germany showed that the emissions from a wood heating facility. They found that the concentrations of I-TEQ was in the range of $0.35-5.7 \text{ ng/m}^3$, which in all cases are much higher than the allowed concentrations in MSW heating facility (below 0.1 ng I-TEQ/m^3). The highest readings were found when the wood being burned was treated wood and treated boards. The homologue pattern in these wood incinerators was dominated by the tetraCDDs and tetraCDFs, see Figure 2. [24]



Figure 2: The homologue pattern of PCDD/F of a wood incinerator.

B. Lignite

Thuss et al. [25] have studied the emissions of PCDDs and PCDFs from lignite combustion. They used a normal coal with 300 ppm w/w and also a salt-coal lignite with 2000 ppm w/w. The values calculated from the normal lignite (0.015 I-TEQ/m³) was lower than the lignite with added salt (0.109 ng/m³). The additional extra PCDDs and PCDFs values are due to the salt being added [25].

C. Coal

Thanner and Moche [26] have studied the emissions of PCDDs and PCDFs from coal combustion in a small residentail plants in Austria. In these studies they found a value of 13.8-87.2 ng I-TEQ/m³ (0% O_2) and this should correspond to an annual emission of 6-38 g I-TEQ in Austria. They also reported that the major component here was the sum of TCDFs (68%) followed by PeCDFs (22%) see Figure 3.



D. Miscellaneous

Dumler-Gradl et al [27] have analyzed chimney soot from house heating systems in Germany. In the study they include soot from:

- a) wood burning ovens
- b) wood burning tiled stoves
- c) wood burning heating systems
- d) wood/coal burning ovens
- e) wood/coal burning tiled stoves
- f) wood, wood/coal and waste burning ovens.

The mean values ranged between 549 to 6587 ng I-TEQ/kg, the highest value was found for category f above and the lowest for category e. [27]

1.5. NATURAL FORMATION OF DIOXINS

Natural formation of dioxins can occur by thermal and biochemical reactions. Burning of any organic material containing organochlorine compounds or inorganic chlorides like sodium chloride, can generate dioxins. [28] So already when the caveman was burning dried see-weeds, he generated dioxins.

The same is the case with forest fires, but the background concentrations associated with forest fires are relatively low.

Sewage sludge is not considered to be a *primary source* of dioxins. It is considered to reflect aerial deposition, industrial input and other anthropogenic sources. The dioxin profile of sewage sludge is totally dominated by octa CDD to a larger degree than any other identified dioxin source. [29] An explanation for this situation could be the natural formation of octa CDD from pentachlorophenol, a relation that we have identified in laboratory experiments with sewage sludge and compost. [7-9] A group in Germany have found that chlorinated phenols, including pentachlorophenol, can be formed during composting of garden waste. [30]

The same dominance of octa CDD has been found in sediment samples from pristine areas in southern Mississippi, USA as well as in sediment cores from the same area and also in lakes in Germany and in the Baltic Sea. [31] These historical samples represent a time of 50 - 100 years (or more) before the industrial use of chlorine. It is an interesting observation that the dioxin pattern found in the sediment or sediment cores is the same as the pattern found in very old Kaolin samples from Germany or in Bentonite samples (Ball Clay) from southern United States. All these samples are more than a million years old. [32] The most plausible explanation for these observations seems to be a natural formation of octa CDD.

1.6. NATIONAL SOURCE INVENTORIES

Due to the interest of the public and the mass media, several countries have performed national source inventories for dioxins in order to eliminate the major sources and to minimize the potential risk for the public and the environment. The first source inventory for air emissions was performed in 1990 by the Swedish EPA. [33] The dominating source at that time was MSW incineration followed by iron, steel, and non-ferrous metal works, pulp mills and exhaust gases from cars running on leaded gasoline containing halogenated scavengers. A substantial decrease in these emissions was postulated for the 1990s primarily due to the introduction of new technology for the MSW incineration (below 0.1 ng I-TEQ/m³) but also the introduction of new technology for pulp bleaching and unleaded gasoline without the halogenated scavengers. For the MSW incineration the air into the oven has been introduced to obtain a good turbulence by using both primary air and secondary air. The cooling phase for the hot gases have a rapid cooling in the temperature zone of 500°C to 200°C. The electrostatic precipitator (ESP) if they are still in use is connected to an additon of lime and charcoal followed by a baghouse [34,35,36]. The major dioxin source for PCDDs and PCDFs in Sweden in the mid 90s should be the metal production and the metal treatment. [33] At the moment most interest is focused in the steel industry on the sintering process [37].

A much more detailed inventory has been performed for the Netherlands. In 1992 they found the major source in this country to be MSW incineration, but in the year 2000 they consider the earlier use

of pentachlorophenol (banned already in the 1980s) to be the major source. The material treated with pentachlorophenol will still be in use in year 2000 and be the major source for the environmental contamination. [38]

More recently HMIP published an estimated inventory of emissions of PCDDs and PCDFs to the atmosphere in UK. [39] The dominant source was the incineration of municipal solid waste (MSW) contributing an average of 70% of the emission from industrial sources, see also Table 3.3a in the ERM report.

The new data with wood burning, house heating and landfill fires are not included in the HMIP and ERM report.

1.7. ENVIRONMENTAL CONCENTRATIONS

Abiotic samples

Background concentrations of PCDDs and PCDFs have been reported in a series of abiotic reservoirs like soil and sediments but also in air and snow. Air, soil and sediments will be discussed below.

Air

Although inhalation is only a minor route of human exposure to PCDDs and PCDFs (below 2%), air measurements have been performed in many countries like Germany, The Netherlands, Sweden, and USA. A series of investigations in Germany has given the following typical concentrations of annual PCDD and PCDF values for air and aerial deposition in ambient air and soil, as given in Table 4. [40]

	Air	Soil	Deposition
	pg/m ³	pg/g	pg/m^2 , d
Rural areas	< 0.07	<10	5-22
Urban areas	0.071-0.350	10-30	10-100
Industrial areas	-	30-300	-
Close to major	0.351-1.600	100-80 000	123-1293
source			

Table 4. Values of I-TEQ in samples of air, aerial deposition and soil in Germany

Soil

Soil samples have been frequently analyzed primarily in Germany, USA and UK. Based on these studies, the following typical concentrations (in TEQ) for soil samples in Germany counted on a dry weight basis are given in Table 4. [40]

In another study soil samples from plowland, grassland and forests, all representing rural background areas in Germany, were analyzed. They found a very interesting difference in the concentrations, the highest values were found for forest soil. A direct consequence is that it is of major importance to control the nature of a sampling site. [41]

Deposition

Very few studies on deposition of PCDDs and PCDFs can be found in the literature. Values for Germany are given in Table 4. [36] A recent study from the southern part of the State of Mississippi, USA shows a much lower value, $0.4 - 3 \text{ pg TEQ/m}^2$, d. [42] No deposition measurements have been published from Hong Kong.

Sediment

Analysis of sediments can give valuable information concerning the spatial variation of contamination, and sediment cores can be very useful to investigate historical trends. As PCDDs and PCDFs are fat soluble or bound to organic particulate matter, the best way to obtain comparability between different sediments is to relate the concentrations to the organic matter by means of loss of ignition (LOI). However, up to now most data are reported on a dry weight basis. The concentrations vary greatly due to local point sources.

2. THE SITUATION IN HONG KONG

2.1. Significant sources of dioxin in Hong Kong

This section has been covered in section 3 in the ERM report. The Consultants list the type of dioxin sources primarily covered by the UK report. The unit used in Table 3.3a are g I-TEQ per annum as in Table 3.3d.

It is very difficult for me to decide the amount of I-TEQ coming from other sources like burning of household wastes around Hong Kong or from landfill fires in Hong Kong or from burning of household wood and coal in this area. However, compared to some other amounts in the Table 3.3d this contribution might be quite important. The higher values measured in the winter period could be an indication that long-range transport and additional demand for electricity on the winter period. Another minor possibility could be that burning of household wastes and landfill fires also could be of some importance. A sampling period during the winter period could possibly indicate the importance of the long-range sampling by higher reading up-wind of Hong-Kong.

I agree in the Consultants recommendation of a pilot sampling program to verify this inventory, but I would also like to see samples taken outside central Hong Kong to see the importance of a long-range aerial transport.

The PVC story discussed in the HMIP review is partly a study from my own group from a fire in January 1987 where a warehouse outside Umea in Holmsund was burned down resulting in a measured amount of only 3 mg I-TEQ. The higher values (13 kg of total PCDDs and PCDFs) are personal calculations by Dr. Meharg to Dr. Dyke (from 1994, unpublished). It has been found that any burning of chlorinated compounds can generate PCDDs and PCDFs. Another important source is the

production of chlorine in the chloralkali factory. I would like to know if such plants have been found in Hong Kong.

2.2. Contamination of air and soil

Table 4.2a is a summary of the concentrations of PCDDs and PCDFs at two different locations in Hong Kong, The Central/Western area close to Victoria Harbour and also Tsuen Wan, north of CWTC. The highest concentrations of PCDDs and PCDFs are found for Februrary 1999 for Tsuen Wan and Central/Western. The Tsuen Wan location is situated 3.5 km north of the CWTC. During this time of the year the winds are blowing from the north (see Table in the Tenth Monitoring Report). It seems rather impossible that the emissions from CWTC could account for this. During the summer season when the wind was blowing from the south, the values at the Tsuen Wan station is approximately ten times lower. The general concentrations found in Hong Kong are the same as what can be expected from other locations around the world, see also Table 4 in this report.

The concentrations of PCDDs and PCDFs in the CWTC stack has been analyzed now up to 95 measurements see Table 4.3a. For 93 of these 95 analyses these concentrations are within the 0.1 ng I-TEQ/m³, while 2 of the 95 measurements exceeded that condition by a factor of 2-4. I would like to know the CO values for these two measurements and to compare these with the other 93 readings. In my opinion the CO readings could be a very good indicator, see below.

In Table 4.3b is the management of the ambient air at the roof of the Ching Yung House. The value I have got here in my calculation is 0.15 pg I-TEQ/m³ but since this station is placed north of the CWTC it is not clear to me if this can be a clear indicator of the activity at CWTC in the wintertime. The blank sample of unknown origin analyzed at the same time gave a value of 0.01 pg I-TEQ/m³. All the congeners in this sample are below the detection limit.

In Table 4.3b they include the non-detects at half of the detection level. In the analysis given in Table 4.3b only 2,3,7,8-TeCDF and two heptasubstituted congeners (HpCDD+HpCDF) plus the two octasubstituted ones are included, all the remaining are based on half of the non-detects. The really detects are 0.018 pg I-TEQ/m³ compared to the total amount of 0.15 pg I-TEQ/m³.

The calculations performed in 1991 by Enviropace Limited indicate that the ambient air concentrations will increase by a factor of 1-4% by the CWTC. The data found in Table 4.3b now indicate that these emissions could be 0.1-0.4% of the existing ambient air concentrations.

It is now generally accepted that food can account for more than 98% of the total uptake from the environment [43]. Among the other exposure route inhalation can contribute to up to 2% of the total uptake of PCDDs and PCDFs. From Table 4.2a the ambient air concentrations are given in Hong Kong. It is also accepted that the total intake of PCDDs and PCDFs to 2 pg I-TEQ/day or 0.03 pg I-TEQ/kg bw, day. The total dietary intake of PCDDs and PCDFs has been estimated to 105 pg

I-TEQ/day, which is similar to data from other countries. I agree in the calculation that the CWTC could contribute up to 0.1-0.4% of the existing ambient air concentrations. The available data has not adversively impacted *the ambient* air in the vicinity of the plant and that it will not have any *adversively related effect on the health of the exposed population*, see also Section 2.3b below.

Concerning the PCBs I would like to use the terminology "the dioxinlike PCBs". They are non- and mono-chlorinated compounds and they have earlier been called the "co planar" or "planar" PCBs. In the text the WHO has also used the dioxinlike PCBs". [3].

In the study the Consultant (ERM p. 27) have used a 1:2 basis for the conversion of the dioxinlike I-TEQ to the total I-TEQ (1.5 pg I-TEQ versus 3.0 pg I-TEQ/kg bw day). This value is based on the consumption of contaminated food in many countries, where the 1:2 ratio has been found. However, the leaking of pure PCBs directly into the environmental and the organisms can be due to many reasons.

On the other side contribution of the dioxinlike PCBs to the emissions from MSW incinerator has not been studied in detail. I know of one example [44] and in this case it is less than 20% of the total emissions coming from the dioxinlike PCBs. I suggest that the dioxinlike PCBs should be studied in Hong Kong both for the food-intake and perhaps also from emissions from CWTC.

The analytical procedures at Dow Chemical was concentrated to the occurrence of PCDDs and PCDFs. They did not study the dioxinlike PCBs. [45]

The dispersion modelling work predicted that each site treating 6,000 t/d would contribute a maximum of 0.001 pg I-TEQ/m³ to ambient air levels. The incremental environmental burden associated with these emissions is sufficiently low, such that even if two sites for incineration facilities were proximate to each other, the cumulative health impact of the emissions would not constitute a significant additional exposure. The dispersion modelling is dependent on the height of the smoke stack. The Consultants also recommend that the concentrations of PCDDs, PCDFs and PCBs should be studied in vegetation, soil, dust and food. I agree with this. However in the case of soil samples it is of importance to collect these samples in open fields. I also agree that the monitoring locations should include rural areas as well as industrial sites.

2.3 HEALTH EFFECTS BY PCDDs and PCDFs

2.3a Exposure to PCDDs and PCDFs at incinerators

The highest concentration of PCDDs and PCDFs is certainly found inside the incinerator among the people working there. The levels of PCDDs in blood from 11 workers at Swedish hazardous waste incinerator where the emission guideline was 3 ng I-TEQ/m³ fell within the range of the normal background. [20] The same result was reported in a study from Germany for 31 workers at three hazardous waste incinerators. Another German study also analysed 37 blood samples of employees at a

municipal solid-waste incinerator. They reported that no increased concentrations could be attributed to the professional activities by the donors. [46, 47] Similarly, Päpke *et al.* found that I-TEQ values in 10 blood samples from workers employed at municipal solid-waste incinerators in Germany were the same as for other people. [48]

In order to determine occupational exposure of employees in three German hazardous waste incinerators, 25 workplace air measurements were analysed. The highest concentration measured was 3.79 pg I-TEQ/m³, corresponding to 7.6% of the German occupational technical exposure limit (TRK) of 50 pg I-TEQ/m³. All the sampling took place during a normal working day at the plants. [49] However, in the studies performed no increase in the "workers" blood or air could be identified.

2.3b Cancer

It is very often claimed that "dioxins are recognized human carcinogens". IARC (International Agency for Research on Cancer) is a WHO agency located in Lyon, France with special responsibility to evaluate the carcinogenic risks to humans. In February 1997 I participated in an 8 day long evaluation of the carcinogenic risks of PCDDs and PCDFs, and I will now describe the process. [1] The invited experts are divided into four groups: exposure, epidemiology, animal carcinogenicity and mechanistic data. I participated in the exposure assessment group.

The exposure assessment group identified several groups of heavily exposed workers, primarily from factories producing 2,4,5-trichlorophenol. The blood or adipose tissue from these workers have been analyzed and strongly elevated concentrations of 2,3,7,8-tetra CDD have been reported up to 10 000 times over the concentrations found in the general population (up to 70 000 pg/g fat). We also included data from the analyses of the population exposed in connection with the accident in northern Italy (Séveso) in 1976.

The epidemiology group had access to a series of studies. The most instructive and useful studies were those where the exposure had been certified by chemical analyses and discussed in the exposure assessment group. After a careful examination of all these studies, the epidemiology group came to a conclusion that based on these studies it was only **limited evidence** that 2,3,7,8-tetra CDD was a human carcinogen.

According to the old IARC rules this situation should end up in a II A/II B classification for the overall evaluation. However, IARC recently modified their evaluation criteria to include Exceptionally relevant mechanism of carcinogenicity with this category.

Group 1 - The agent (mixture) is carcinogenic to humans. The exposure circumstance entails exposures that are carcinogenic to humans. This category is used when there is sufficient evidence of carcinogenicity in humans. Exceptionally, an agent (mixture) may be placed in this category when evidence in humans is less than sufficient but there is sufficient evidence of carcinogenicity in experimental animals and strong evidence in exposed humans that the agent (mixture) acts through a

relevant mechanism of carcinogenicity.

This situation was foreseen by IARC, and they had also invited a large group of biologists to the working group "Other relevant data". They concluded that the mechanism of action is via the Ahreceptor, and most of the discussions before the final voting was whether this was a "relevant mechanism of carcinogenicity". The group of experts including the chairman of the group (prof. Stahlmann) and the Ahreceptor specialist (prof. Poellinger) was hesitant to say yes or no. However, in the final voting there was a **14** - **10 result in favour of a group 1 carcinogen**.

Other PCDDs and PCDFs

Two PCDFs (2,3,4,7,8-penta CDF and 1,2,3,4,7,8-hexa CDFs) were discussed. After a 12 - 11 voting they were included in the group III carcinogen "Not classifiable". Due to lack of data all other PCDDs and PCDFs were in the same group III.

Carcinogenicity and TEQ

Only 2,3,7,8-tetra CDD is a group I carcinogen, all the other are not classifyable. Emissions from incinerators contain a multitude of PCDDs and PCDFs, making up a TEQ-value. Since 2,3,7,8-tetra CDD contributes to less than 4% to the TEQ-value, the TEQ-value for incinerator emissions has very little correlation to carcinogenicity.

Overall Conclusion

Only 2,3,7,8-tetra CDD has been classified as a Group 1 carcinogen. Even in the case where a massive exposure has been documented, resulting in body burdens at 1000 to 10 000 times the background, the epidemiology **data gave limited evidence for carcinogenic effects in humans**. In this connection it is of interest to refer to the following paragraph in the evaluation. [1]

"In view of the results mentioned above, it should be noted that the present background levels of 2,3,7,8-TCDD in human populations (2-3 ng/kg) are 100 to 1000 times lower than those observed in this rat carcinogenicity study. Evaluation of the relationship between the magnitude of the exposure in experimental systems and the magnitude of the response (i.e. dose-response relation-ships) do not permit conclusions to be drawn on the human health risks from background exposures to 2,3,7,8-TCDD."

Summing up in my opinion it is completely irrelevant to state that any non-occupationally exposed person should be at a greater risk to develop cancer.

Other effects

The Consultants (Appendix B) discuss some other effects which could be caused by exposure to PCDDs and PCDFs. In general it is less clear if these effects are really caused by the exposure to PCDDs and PCDFs because other confounding factors are present as well [50, 51].

2.4. DIOXIN CONTROL MECHANISMS

The Consultants describe many parameters by which the emissions of PCDDs and PCDFs could be below 0.1 ng I-TEQ/m³. The feedstock and the feed control could be quite important and the ram feeding system seems to be the best. The role of PVC is another important issue. However, it has recently been found that under normal contamination levels of the waste the influence by added PVC is the same as in the case where you add sodium chloride. These emissions are also found to give very low emissions. For this purpose a waste feed was used and PVC and/or sodium chloride was added [28]. This proves that for generating a chlorine source it is independent if you start with PVC, after organochlorine compounds or sodium chloride. They all give minor effects on the formation of PCDDs and PCDFs, see also ERM page C5.

It is suggested that CWTC will co-incinerate clinical waste for 8-12 hours per day and then go back to chemical waste. This seems to be all right for me.

In the CWTC the kiln has the average temperature of $1050^{\circ}C \pm 100^{\circ}C$ and in the secondary combustion chamber (SCC) the temperature is at $1150^{\circ}C \pm 25^{\circ}C$. The residence time in the kiln is 6 seconds and in the SCC additionally 4 seconds. The excess oxygen is set at 6%. All these parameters are well inside the accepted areas: temperature 850°C, time 2 seconds and 6% excess oxygen. Before leaving the kiln the exit temperature is maintained at 380°C. The Reynolds Number for the SCC is more than 55 000.

The pollution control at CWTC has two spray dryer in which a slurry of lime and activated carbon is used and after this follows a fabric filter. These two spray dryer systems are working parallel and independent of each other.

This general configuration has been proven to be a standard practice and this has been found to result in I-TEQ values for beyond 0.1 ng TEQ/m³. The proposed arrangement with a limit below 0.1 ng I-TEQ/m³ can be used for the incineration of MSW, sewage sludge and animal carcass . It is also equipped with a failsafe system to meet process upset, plant malfunctions or power failure.

I totally agree with the Consultants that it is very unlikely that the released quantities of PCDDs and PCDFs could be detrimental to the human health. The emissions are close to the planned even during very abnormal operations.

Some concern has recently been associated with the content of PCDDs and PCDFs in the ash particles leaving the oven or combustion chamber which are boiler ash and fly ash. However, the data now provided on ash particles show that the situation at CWTC is quite good. From 1993 all the 77 concentrations are below the 1 ng I-TEQ/g which is the guideline for CWTC. In only one occasion in 1995 the value was in the range 0.3-0.4 ng I-TEQ/g and in another sample it was 0.16 ng I-TEQ/g. In all other analyses the value were below 0.1 ng I-TEQ/g.

2.5. STANDARDISATION OF DIOXIN MEASUREMENTS PROCEDURES FOR INCINERATOR WASTE GASES.

At present, the regulatory requirements for incinerator emissions vary widely among the countries of the European Union. The European Commission published a Council Directive on the incineration of hazardous waste which would require ' that the emission of PCDDs and PCDFs shall be minimized by the most progressive techniques' and which defines 0.1 ng/m^3 as a guide value which should not be exceeded by all average values measured over the sample period of 6 to 16h. [52]

Germany and the Netherlands have set daily average limit values of 0.1 ng I-TEQ/m³ of exhaust gas for PCDDs/PCDFs from municipal waste incinerator emissions; in Sweden, the corresponding value is 0.1-0.5 ng Eadon TEQ/m³. The United Kingdom has set a limit value of 1 ng I-TEQ/m³ with a goal to reduce PCDD/PCDF emissions to 0.1 ng I-TEQ/m³ for industrial and municipal waste incinerators. [1]

In Japan the emission standards up to 2002.11.30 is 80 ng I-TEQ/m³ for the existing incinerators. For new incinerators the limit is 0.1 ng I-TEQ/m³ for incinerators burning above 4 ton /hours. For smaller units they have the guideline of 1 ng I-TEQ/m³ (2-4 ton/hours) or 5 ng I-TEQ/m³ (below 2 ton/hours).

At the moment I am not aware of any discussion about the violation of these rules either from Europe or Japan.

It has been a very long discussion about the quality of the data from incinerators working with a 0.1 ng I-TEQ/m³ guideline. A working group under the European Committee for Standardisation (CEN) has now established a European Standard for the measurement of PCDDs and PCDFs [36]. Three different sampling procedures were selected: the filter/condensate method, the cooled probe method and the dilution method. They are all considered to be equivalent as a result of comparative measurements. The applicability of the European Standard has been evaluated and proven by validation measure, see Figure 4 and 5.

The result show that at a limit of 0.1 ng I-TEQ/m³ they can be measured with an internal variability of 0.01 to 0.1 ng I-TEQ/m³ and an external variability around 0.05 to 0.1 ng I-TEQ/m³ All these combined give a value of 0.3 ng I-TEQ/m³. All values are calculated with a 95% confidence level.

In Table 4.3a you have two figures which are above 0.1 ng I-TEQ/m. They are 0.214 ng I-TEQ/m³ for November 1998 and 0.4495 ng I-TEQ/m³ for February 1999. Using the data given above it can be concluded that *the November 1998 levels are inside the allowed dioxin limits (below 0.3 ng I-TEQ/m³)*. However, for the February 1999 value of 0.4495 is I-TEQ/m³ *this value is outside the allowed dioxin limit.* The February 1999 value is due to malfunction of the activated carbon dosing pumps as demonstrated by

the abnormality low consumption rate of the activated carbon. No explanation has been given for the November 1998 value, but as described above this value is inside the allowed dioxin limit.

The sampling method in Hong Kong is apparently taken from USA and this has not been fully tested in the European studies. Moreover the laboratory used is not included in the European studies but it has successfully participated in other intercalibration studies. [44]

2.6. Setting exposure limits and emergency control.

The Consultants discuss that fluctuation in emissions of PCDDs and PCDFs can result in unstably operations or malfunctions that can affect the public health. A value given by De Fré and Wavers [49] report a large variation between a sample taken after an exposure time of two weeks and a 6-hours sampling period. The data from DeFré and Wevers suggest that the standard 6 hours measurements *underestimates* the 'true' average emission by a factor of 30 to 50. Another possibility could be that the continously 2 weeks sampling *overestimates* the 6 hours sampling by the same figures. [53]

In the study from Belgium they are using a sampling principle based on stream and condensates that are drawn through a prefilter followed by an adsorber which will collect all the PCDDs and PCDFs in the flue gas. Thus the condensate contains the resin but also the water from the *wet scrubbing* unit. No test-burnes have been performed to show that the instrument is working properly in all sampling systems. [53, 54]

In the study in Belgium they are analyzing the content of PCDDs and PCDFs from an incinerator equipped with a wet scrubbing unit. In my group in Umea we have been analyzing several data from Avesta, a Swedish wet scrubbing unit. [55] In our study we analyzed samples taken at the same time and found that they showed an increase in the wet scrubbing system and we also found that this was due to a *new formation* of PCDDs and PCDFs, see also Figure 6 and 7.

In all we now have three studies with the same effect: the Belgian study, the Avesta, Sweden study and a study from Coburn, Germany. In all these cases the sampling is from a *wet scrubbing* unit and the data indicate a pronounced difference between the samples, an underestimation or an overestimation.

However, in the study of CWTC discussed here in Hong Kong the air pollution control unit is a *dry scrubbing* unit, and consequently the values from a wet scrubbing unit is not appropriate here. As pointed out already in the paper by DeFré and Wevers this under- or overestimation of the amount of PCDDs and PCDFs is only connected with a wet scrubbing unit. The CWTC is a continously sampling dry scrubbing incinerator, which is not subjected to frequent shut-down and start-ups.

The Consultants have also shown that the emission of PCDDs and PCDFs from the CWTC in 1999 is 0.055 ng I-TEQ/m³. This average also includes the exceedance of 0.4495 ng I-TEQ/m³ in February 1999 which should be compared to a limit of 0.1 ng I-TEQ/m³. The months excluding February 1999

have an average concentration of PCDDs and PCDFs at 0.029 ng I-TEQ/m³. Calculated for the whole time since the CWTC started in May 1993 the value for all these emissions are in the range of 0.03 ng I-TEQ/m³ against the allowed concentrations of 0.1 ng I-TEQ/m³ which is only around 30% of the allowed limit.

The Consultants have used the emission figure of 0.1 ng I-TEQ/m³ and found that this contribution is 0.2% of the background concentration of 0.1 pg I-TEQ/m³. Using the figure discussed above, 0.03 ng I-TEQ/m³, this value should be 0.00006 pg I-TEQ/m³ (see Figure 6.3a) which corresponds to 0.06% of the background ambient air concentrations.

The introduction of a 2 ng I-TEQ/m³ as a trigger is in my opinion a good idea, see Figure 6.3b. The Consultants also discuss the "turn-over" time for the chemical analyses of PCDDs and PCDFs in a sample. They calculated this to be in the range of 4-8 weeks. If special precautions are given I am inclined to say that the "turn-over" time could be only 2-3 weeks. This will give the plant more time to revert to compliance with the stated emissions, say altogether two months instead of three months. So the value calculated here should be considered to be the higher value.

The Consultants have proposed Figure 6.3b. If the "turn-over" time in the chemical laboratory could be 2-3 weeks the result of the earlier sampling could be ready for the next sampling period. In general the CO monitoring, the temperature and the lime slurry with the activated carbon are the most important parameters to follow.

The WHO has now calculated a TDI of 1-4 pg I-TEQ/kg bw and day [3]. This was announced almost two years ago. In December 1999 the Scandinavian countries followed the WHO guideline. This value is primarily due to for the food intake. More than 98% of the daily intake is via the food. The TEF values for dioxins and PCBs are given in Table 2.2a.

It is recommended that the authorities in Hong Kong should monitor these concentrations using the values of PCDDs, PCDFs and also PCBs in meat and meat products, milk and dairy products, fish and also in vegetables. From such measurements a dietary intake can be made for the people living in Hong Kong. I should also like to see that the Government of Hong Kong will start to analyze the concentrations of PCDDs and PCDFs in mother's milk. If you follow the mothers with their first child the values found for PCDDs and PCDFs will be a very good indicator of the situation in the country. The next phase in the WHO series will probably be taken during the year 2000.

3. General conclusions.

The influence of CWTC with a limit of 0.1 ng I-TEQ/m^3 has now been studied for more than 6 years. The air quality recording is very good. The air-borne emission from this incinerator is less than 0.06% of the total.

From an analytical point of view the data set attached to the application could be a little bit more detailed. It could be interesting to know the outcome of the blank samples taken *before the high values reported* from February 1999. The value from the EU experience [48] indicate that this is an important problem. Some datapoints in that EU-study could not be included due to this reason in the final report.

In addition to the congeners now reported in the CWTC emission I would also like to see the totals included which are the totals of non-2,3,7,8-substinted PCDDs and PCDFs.

It is my opinion that the Government of Hong Kong should perform a dietary intake study to find out the major sources for PCDDs, PCDFS and PCB in the food intake. It could be valuable to make a comparison between this value and the direct inhalation of air and the concentrations of PCDDs and PCDFs in human milk.

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Figures



Fig. 4 PCDD/PCDF concentrations in the flue gases of a municipal waste incinerator equipped with an activated coke filter. Different sampling systems were employed on two measuring days. The samples were quantified by the same analytical laboratory based on the added analytical standards. A-B: Dilution method; C-D: Filter/condenser method; E-F: Cooled probe method; LIS: preliminary test measurement; H: additional laboratory (see text) employed by the plant operator.



Fig. 5 PCDD/PCDF concentrations in the flue gases of a municipal waste incinerator equipped with an activated coke filter. Results are shown from the second measuring day obtained by different sampling systems. The samples were quantified by the same analytical laboratory based on both the added analytical standards (2Ha) and the sampling standards (2Hs). A-B: Dilution method; C-D: Filter/condenser method; E-F: Cooled probe method; H: additional control laboratory (see text).





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