# The Dioxin Problem in The Netherlands: Regulatory Approach and its Impact on Emissions and Human Exposure

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### Abstract

Because of the relatively high exposure to dioxins of the Dutch population expensive measures have been taken to reduce emissions. The success of this policy is reflected in a 90% reduction in emissions to air and a significant decrease in dietary exposure and levels in human milk. It is expected that exposure to dioxins will drop below a level of concern for human health. The importance of an international approach is emphasized.

### 1. Introduction

The description 'dioxins' stands for two groups of closely related compounds: polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs). Dioxins are very persistent organic compounds. Of 210 possible congeners the most persistent are the seventeen 2,3,7,8chlorinated ones and these are extremely toxic. The most toxic one, 2,3,7,8-TCDD, is used as a reference for the toxicity. Toxicity equivalency factors have been assigned to the others. This international accepted system to express the toxicity of mixtures of dioxins has been of great help in risk assessments and standard setting [NATO/CCMS 1988, Van Zorge et al. 1989].

Dioxins are ubiquitous environmental pollutants of anthropogenic origin. They are microcontaminants in many processes: combustion, incineration and industrial. Because of their lipophilicity and persistence, dioxins accumulate in the fatty tissues of exposed organisms, including humans. They build up in the food chain and therefore present a hazard for predatory animals. Human exposure is mainly (> 95%) by food and is at a level of concern. The human health hazard of 2,3,7,8-TCDD has been recognized for four decades now. Several coutries have set a Tolerable Daily Intake (TDI) for dioxins and many countries have developed policies to control emissions of dioxins.

# 2. Historical Background

The first Dutch confrontation with dioxin (2,3,7,8-TCDD) was on March 6 in 1963 when an explosion in a 2,4,5-trichlorophenol production site in Amsterdam contaminated the plant. Of the 106 workers involved in the cleaning operation 44 developed chloracne, a persisting and disfiguring skin disease. In 1957 dioxin had been identified as a very active chloracnogenic contaminant in the chlorophenol production by he German physician Schulz. Since it appeared to be impossible to clean the plant it was closed down and ten years later it was carefully pulled

down. The debris was put up in concrete and dumped into the Atlantic Ocean.

In 1980 the chemical contamination of the 'Volgermeer polder' was discovered. This polder had been used for long as a municipal waste dump site for Amsterdam, but it appeared that during the fifties and sixties illegally thousands of tons of chemical waste had been dumped there, also from the plant mentioned above. Elevated levels of dioxin were found in soil, sediments and fish. The 60 hectares dump site was closed, partly covered with soil in order to prevent distribution of the pollution and fenced off.

A reseach group of the University of Amsterdam was the first to report that PCDDs and PCDFs are emitted by municipal solid waste incinerators (MSWIs) [Olie et al. 1977]. This was the start of more government funded investigations in this field. But it was the detection in 1989 of dioxins in the milk of cows grazing near the most important MSWI in the country, at levels that raised concern for the public health, that has initiated an extensive national research programme on dioxins.

By that time it was realized that what originally had started as a single substance problem (2,3,7,8-TCDD) was in fact a multi substances problem (PCDDs and PCDFs). In recent years it has been proposed to extend the substances involved with several PCBs, which because of their structural similarity have a comparable mechanism of action as the dioxins. This means that these dioxin-like PCBs should be included in risk evaluations and standard setting [Ahlborg et al. 1994, Health Council 1996, Van Leeuwen 1997].

### 3. the Lickebaert Affair

The detection of high levels of dioxins in cow's milk near MSWIs usually is referred to as the Lickebaert affair, after the name of the agricultural area near Rotterdam where the identification of this problem started. It not only gave the impulse for the national dioxin research programme but also for the emission reduction policy.

### 3.1 Emission reduction measures

In August 1989 a guideline on incineration of municipal solid waste and related processes, like incineration of hazardous waste, hospital waste and sludges, was published. In this guideline an emission standard for dioxins of 0.1 ng TEQ/m<sup>3</sup> was set for new incinerators. Existing incinerators had to meet this standard the first of December 1993. In February 1993 the guideline became part of the Law on Environmental Management and because of problems with the accommodation of the existing incinerators the date of enforcement was delayed to the first of January 1995. In practice it took until the start of 1997 before all the MSWIs complied with the standards. For other dioxin emitting processes no standards have been set until now. Licence providing authorities however have been instructed to ask for maximum acceptable emission reduction. If necessary for certain sources, emission maxima will be presented in the National Emission Guideline.

The dioxin emission of MSWIs has to be monitored at least twice a year. As the result of a round robin study an instruction for sampling, sample treatment and analysis was published in 1993, in which also detection limits for the different congeners were dictated [Ministry 1993].

Since 1989 six of the twelve existing MSWIs were closed, two of them were replaced by new ones and another three new incinerators have been build. Action was taken to reduce the amount of waste which has to be incinerated. Separated collection and composting of the organic fraction of municipal waste now is widely introduced in the country.

#### 3.2 Emergency measures

In order to prevent the consumption of milk with high dioxin levels in 1989 a standard of 6 pg TEQ/g fat was set for dairy products. This standard was derived from the Tolerable Daily

Intake for dioxins, the assessed exposure by non-dairy products and the average daily consumption of dairy products on a fat base. Milk of dairy farms near MSWIs was monitored and in areas where the standard was exceeded the milk was destroyed. Animals in areas exceeding the milk standards also were taken out of the consumption chain.

As an illustration of the effectivity of emission reduction measures Figure 1 shows the dioxin levels in milk from reference farms in the Lickebaert area situated near the MSWI AVR, which with a capacity of ca. 1 milion tons of waste per year is the largest MSWI in the country [Liem and Van Zorge 1995]. The retrofitting of this incinerator was completed in 1994 which was clearly reflected in the dioxin levels in milk. Moreover the levels in milk were in good agreement with the calculated values from a chain model that was developed to relate emissions to levels in cow's milk [Slob et al. 1993].



Fig. 1. Time course of dioxin levels in cow's milk from dairy farms in the Lickebaert area

# 4. The National Dioxin Research Programme (1989-1994)

Formally the programme was finished in 1994 but monitoring programmes are still going on in order to follow trends and to detect the effect of the measures which have been taken. The targets of this programme were:

- i detection of dioxin sources in the country, quantification of their emissions and the use of these data for an emission reduction strategy,
- ii analysis of human exposure to dioxins and related compounds by food consumption and the monitoring of milk of dairy farms in suspected areas,
- iii detection of environmental levels, background levels as well as levels in contaminated areas.

# 4.1 Dioxin emissions in The Netherlands

The study on sources and emissions made it clear that for the Netherlands MSWI's, with about 80% of the total emission, were by far the most important source of dioxins emission into the air. Table 1 presents the emissions of dioxin sources in 1989 and 1991 together with the estimates for the year 2000 [Bremmer et al. 1996]. It clearly shows that municipal solid waste incineration changes from a major to a minor source of dioxin emission. Today wood combustion for house heating, industrial processes and the use of pesticides have become the major sources. The latter one is dominated by the extensive former use of pentachlorophenol (PCP) as a wood

preserver. PCP contains relatively large concentrations of dioxins. The use of PCP was terminated at the start of 1991.

Estimated loads of fresh surface water are presented in Table 2. Direct discharges on surface water are small in comparison to deposition from the air and municipal runn off.

Emissions to soil are dominated by deposition from the air with the application of sewage sludge in agriculture as a second source (estimated loads in 1990 resp 325 and 7 g TEQ).

Year	1989	1991	2000
Municipal solid waste incineration	697	382	3
Incineration of chemical waste	43	16	0.1
Incineration of tip gas, biogas, sludge	0.4	0.3	0.5
Incineration of hospital waste	4	2.1	0
Incineration of waste oil	2	2	2
Incineration of wood	16	12	10
Cable burning	6	1.5	0
Fires, flares	9	9	9
Traffic	7	5	2
Metal industry	45	31	7
Application of pesticides	50	42	20
Chemical processes	5	3	3
Various processes	15	10	8
TOTAL	899	516	63

Table 1. Sources of dioxin emissions into the air in The Netherlands

### Table 2.Estimated load of fresh surface water (g TEQ/year)

1985	1990	1995	2000	
6	4	4	4	
37	30	14	14	
18	13	4	5	
61	47	22	23	
	6 37 18	6 4 37 30 18 13	6 4 4 37 30 14 18 13 4	6     4     4     4       37     30     14     14       18     13     4     5

# 4.2 Environmental levels and fluxes

Levels in surface soil (depth 0-5 cm) ranging from 2.2 to 16.4 ng TEQ/kg dry weight were obtained from 32 rural areas. Near MSWIs surface soil levels of 2.5-252 ng TEQ/kg dry weight were found. In several floodplains, where the soil can be contaminated by sludge from the river,

soil levels were 17-78 ng TEQ/kg dry weight [Hendriks et al. 1996]. High levels (60-98,000 ng TEQ/kg dry weight) in soil were detected on sites where wires and cars had been burned for metal reclamation [Van Wijnen et al. 1992].

Analysis of sediment samples from several rivers, lakes and canals indicated that approx 1-10 ng TEQ/kg dry weight can be considered as a present background level, but levels up to 420 ng TEQ/kg have been detected. Even higher levels were found in stagnant waters (up to 650 ng TEQ/kg) and harbours (up to 4,000 ng TEQ/kg). In estuaries and seas levels are 8-21 ng TEQ/kg dry matter. A considerable amount of the dioxins in the Dutch aquatic environment originates from transboundary sources [Liem et al. 1993].

Ambient air samples were collected at four locations across the country, indicating an average background concentration in air is approx 25 fg TEQ/m<sup>3</sup>. The transboundary flux of dioxins was estimated by combination of concentrations in air and weather conditions. From the environmental data the fluxes of dioxins in the Dutch environment were calculated. Figure 2 presents a schematized presentation of these fluxes [Liem et al 1993]. It illustrates the important contribution from abroad to the dioxin load in The Netherlands. This makes it clear that an international approach is needed for a decrease of the present background.



Fig. 2. Estimated flows of dioxins in the Dutch environment in g TEQ/year in 1990

# 4.4 Human exposure and body burden

Much effort has been put into investigations of dietary exposure. Two tracks were followed in these investigations:

- i analysis of duplicate diets for dioxins and related compounds,
- ii analysis of food catagories and calculation of exposure by use of the consumption data from the National Food Consumption Inquiry.

The latter approach has the advantage that information is obtained for different age categories and

for gender. The results of both studies were well in line with each other. The proportional contribution of food categories to the dietary intake for dioxins and dioxin-like PCBs is shown in table 3 [Liem et al. 1996]. In 1991 for dioxins the median and average daily intakes were resp. 55 and 67 pg TEQ. For dioxin-like PCBs these figures were resp. 71 and 90 pg TEQ.

Contribution to:	dioxins (% of TEQ)	PCBs (% of TEQ)
Beef	8.0	8.3
Pork	6.3	1.6
Mixed meat	9.2	5.1
Poultry and eggs	7.1	6.2
Dairy products	47.7	44.5
Fish	7.4	12.6
Industrial fats	10.4	14.6
Others	3.9	7.1

 Table 3.
 Proportional contribution of food categories to the dietary intake in The Netherlands

The results of duplicat diet studies are shown in table 4. There clearly has been a substantial decrease in dioxin levels in food from 1978 till 1994. Regression analysis indicated a log-linear fit of the data in TEQ for dioxins as well as for dioxin-like PCBs. Consequently the food intake in TEQs over these years shows a logarithmic decrease. In 1994 the average intake of PCDDs, PCDFs and dioxin-like PCBs for adults was 1.5 pg TEQ/kg b.w./day [Liem and Theelen 1997]. In view of the fact that important emission repressing measures have been taken after 1994, it is expected that this downward trend will continue and that an average daily intake of 1 pg TEQ/kg b.w. is well within reach as a result of all the measures which have been taken now.

Year	197	1978		1984/85		1994	
	mean	RSD (%)	mean	RSD (%)	mean	RSD (%)	
PCDDs and PCDFs	4.19	10	1.82	47	0.53	44	
non-orth PCBs	2.29	14	1.57	24	0.66	24	
mono-ortho PCBs	2.55	27	0.92	38	0.21	30	
di-ortho PCBs	2.01	21	0.14	23	0.05	24	
$\Sigma$ TEQ	11.0	14	4.45	20	1.45	29	

Table 4. Time course contribution to the diet from duplicate diet studies (in pg TEQ/day)

Dioxin body burdens will respond much slower to emission reduction measures. The long halflife in humans causes an accumulation that reaches steady state only after several decades. So past high exposures will work on for a long time. Nevertheless a reduction is observed in human milk, which is an indicator of body burden. In The Netherlands the mean levels in human milk decreased from 34.2 to 23.5 pg TEQ/g fat between 1988 and 1993 [Liem et al. 1995]. Based on

the decrease in dietary intake a further reduction of 30-40% is predicted for the additional study which is planned for 1998.

### 5. The Risk Assessment of Dioxins

The Netherlands was the first country to propose a health based exposure limit for dioxin. A TDI of 4 pg 2,3,7,8-TCDD/kg body weight was introduced [Van der Heijden et al. 1982] and almost unnoticed this afterwards was changed into the same value for TEQs. In 1991 the TDI of 10 pg 2,3,7,8-TCDD/kg b.w. which was proposed in 1990 by WHO [Ahlborg et al. 1992] was adopted and again this value was interpreted as TEQs. In spite of this raise in the TDI the policy remained to reduce dioxin levels as far as possible. The Health Council of the Netherlands as a result of a re-evaluation of dioxin toxicity data advised to reduce the health based exposure limit to 1 pg TEQ/kg b.w. [Health Council 1996]. Besides that it was advised to include dioxin-like PCB's in risk evaluations and standard setting. Herefore the use of the toxic equivalency factors, as proposed by WHO/EURO [Ahlborg et al. 1994], was recommended Based on the intake data for 1991-1992 only 1.5% of the population exceeded the existing TDI of 10 pg TEQ/kg b.w.. At that time the exceedence of a TDI of 1 pg TEQ/kg b.w. would have been 93%.

Because of the importance of international consensus on this subject the Dutch government decided in response to the advice of the Health Coucil to await the outcome of the WHO-coordinated expert consultation for the re-evaluation of the TDI, which is planned for the second quarter of 1998. Since there is a connection between the TDI and the standard for milk a change in the TDI would mean a re-evaluation (lowering) of this standard and the consequence of that would be an expensive control on levels in milk all over the country. In view of the goal of the government to lower the exposure to the level proposed by the Health Council, this would be a waste of effort and money. Recently the Dutch parliament has asked the government to present a plan in which the necessary steps to reach this goal and the time schedule for it are presented.

The levels of dioxins and related substances in human milk have raised much concern about the high exposure of breastfed babies. This exposure may be two orders of magnitude higher than for adults although only for a short period. However in a comparitive study between breastfed an formula fed children only marginal differences were detected [Koopman-Esseboom 1995, Huisman 1996] and overall the effect of breastfeeding was positive. In a risk assessment of breastfed infants by use of a kinetic model the liver concentration after four months of breastfeeding was calculated to be approximately two times higher than the liver concentration in adults, but well below concentrations for which adverse effects can be expected [Liem and Theelen 1997]. The Health Council concluded that limitation of breastfeeding should not be considered [Health Concil 1996].

### 6. European Community Measures

The WHO-coordinated human milk studies [WHO/EURO 1989, WHO/ECEH 1996] showed the highest concentrations of dioxins in the highly industrialized Western-European countries. Fig. 2 indicates that background levels in The Netherlands might be reduced by measures in neighbouring countries. For that reason the the influence of the policy of European Community should not be underestimated. The implementation of Community measures in national legislation is obligatory.

In the E.U. Directive on the combustion of hazardous waste, which has been implemented at the start of this year, an emission limit for dioxins of 0.1 ng  $TEQ/m^3$  is set. This means that facilities for destruction of hazardous waste or facilities combusting hazardous waste as a source

of energy, e.g. cement kilns, will have to meet this standard [Commission of the European Communities 1994]. The Directive on integrated pollution prevention control (IPPC) imposes the application of best available techniques (BAT) for indicated categories of industrial activities in order to reduce the impact on the environment [Commission of the European Communities 1996]. The existing EC-directives on municipal waste incineration date back to 1989 and do not dictate an emission limit for dioxins. Installations for municipal solid waste incineration however are included in the IPPC-list of activities for which BAT must be introduced.

In February 1993 the Council of Ministers of the European Union agreed upon the 5th European Community Environmental Programme in which a reduction target for emissions of dioxins of 90% was set for the year 2005 as compared to the reference year 1985.

### 7. Conclusion

In The Netherlands all the efforts and money spend on emission reduction of dioxins have caused a significant drop in human exposure. Probably a part of this drop has to be attributed to measures abroad. It is expected that the measures taken will effect a decrease of human exposure below a level that rises concern for their offspring. A condition for an ongoing downward trend is the maintenance of the measures which have been taken. Because of the transboundary flows of dioxins an international approach is necessary. Therefore the EU approach, as laid down in the 5th Environmental Programme, is promissing.

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