### RIJKSINSTITUUT VOOR VOLKSGEZONDHEID EN MILIEU man and environment NATIONAL INSTITUTE OF PUBLIC HEALTH AND THE ENVIRONMENT

RIVM report 639102 023

Scenario studies on maximum levels for dioxins, dibenzofurans and dioxin-like PCBs in

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July 2001



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This investigation has been performed as part of RIVM project 639102, "Dioxinen in voeding", by order and for the account of the Ministry of Health, Welfare and Sport.

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### **Abstract**

Dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls (together called 'dioxins') are persistent environmental contaminants with a comparable chemical structure and similar toxicological characteristics.

Since fish is one of the food items containing variable and sometimes high amounts of dioxins, the authorities asked RIVM, RIKILT and RIVO to investigate the possibility of setting maximum levels for dioxins in fish in order to protect fish consumers from high intake of these contaminants.

To this, scenario studies were performed estimating the effect of several hypothetical maximum levels for dioxins in fish on the intake of these compounds by fish consumers and the percentage of various fish species that can be expected not to comply with these maximum levels. The studies show that in general the intake reduction will be limited to about 2-8%, while the percentage of non-compliance varies between 0.1 and 10. The available datasets on which these estimations had to be based are very limited. Moreover, the regular consumption of fish has positive health effects too due to the amounts of  $\omega$ -3 unsaturated fatty acids.

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# Samenvatting

Dioxinen, dibenzofuranen en dioxine-achtige polychloorbiphenylen (samen aangeduid als 'dioxinen') zijn persistente milieucontaminanten met een vergelijkbare chemische structuur en overeenkomstige toxische eigenschappen. Om schadelijke gezondheidseffecten op de lange duur te voorkomen heeft de WHO in 1998 aanbevolen om de dagelijkse inname te beperken tot maximaal 1-4 pg TEQ (dioxine toxiciteitsequivalenten) per kg lichaamsgewicht. Het Scientific Committee on Food van de EC adviseerde in 2000 om de wekelijkse inname te beperken tot 7 pg TEQ per kg lichaamsgewicht.

De blootstelling van de algemene bevolking vindt voornamelijk plaats via de voeding. Momenteel bedraagt die blootstelling 1,3 pg TEQ per kg lichaamsgewicht per dag. De overheid streeft naar verlaging van die inname.

Vis als voedingsmiddel bevat sterk wisselende en soms hoge gehalten aan dioxinen, en draagt voor ca. 16% bij tot de dioxine-inname van de algemene bevolking; voor de regelmatige consumenten van vis echter bedraagt deze bijdrage ongeveer 34%. Teneinde de consumenten van vis te beschermen tegen een hoge dioxine-inname hebben de Ministeries van Volksgezondheid, Welzijn en Sport en Landbouw, Natuurbeheer en Visserij aan RIVM, RIKILT en RIVO gevraagd onderzoek te verrichten naar de mogelijkheid van normstelling voor dioxinen in vis.

Daartoe zijn scenario-studies uitgevoerd waarin een aantal hypothetische normen zijn onderzocht voor wat betreft de effecten op de dioxine-inname en het percentage van verschillende vissoorten dat als gevolg van die normen zouden moeten worden afgekeurd voor consumptie.

De studies tonen aan dat de effecten op de dioxine-inname beperkt blijven tot een reductie van 2–8%, terwijl het afkeuringspercentage varieert van 0,1 tot 10%.

Opgemerkt wordt dat de gegevens waar de berekeningen op zijn gebaseerd, uiterst beperkt van omvang zijn. Bovendien wordt aangetekend dat de consumptie van vis ook positieve gezondheidseffecten heeft als gevolg van het gehalte aan  $\omega$ -3 onverzadigde vetzuren.

## **Summary**

Dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls (together called 'dioxins') are persistent environmental contaminants with a comparable chemical structure, having similar toxicological characteristics. In 1998 the WHO recommended a health-based upper limit of intake of 1-4 pg TEQ (dioxin toxicity equivalents) per kg body weight per day, while the Scientific Committee on Food of the EC in 2000 advised to a temporarily tolerable weekly intake of 7 pg TEQ per kg body weight.

For the general population food is the major route of exposure. Currently the mean intake of the Dutch population amounts to 1.3 pg TEQ per kg body weight per day, the current policy aims at lowering this intake.

Fish is one of the food items containing variable and sometimes high amounts of dioxins, contributing approximately 16% to the total intake of the average Dutch individual, and approximately 34% to the total intake of the regular fish consumer. The Ministries of Health, Welfare and Sport, and Agriculture, Nature Management and Fisheries asked RIVM, RIKILT and RIVO to investigate the possibility of setting maximum levels for dioxins in fish in order to protect fish consumers from high intake of these contaminants.

To this purpose scenario studies were performed estimating the effect of several hypothetical maximum levels for dioxins in fish on the intake of these compounds by fish consumers and the percentage of various fish species that can be expected not to comply with these maximum levels.

The scenario studies show that in general the intake reduction will be limited to about 2-8%, while the percentage of non-compliance varies between 0.1 and 10.

It is noted that the available datasets on which these estimations had to be based are very limited. Moreover, the regular consumption of fish has positive health effects too due to the amounts of  $\omega$ -3 unsaturated fatty acids in fish.

## **Definitions**

'<u>Dioxins'</u> denotes the sum of the *polychlorinated dibenzo-p-dioxins* (PCDDs) and *polychlorinated dibenzofurans* (PCDFs).

'<u>Dioxins and related compounds'</u> denotes the sum of the PCDDs, the PCDFs, and the 'dioxin-like' polychlorinated biphenyls (PCBs).

'<u>Dioxin-like PCBs'</u> are *non-ortho-* and *mono-ortho* PCBs, for which a TEF has been established.

The 'consumer of fish' is defined as the individual consuming fish at least once per week (cf. paragraph 5).

### 1. Introduction

Fish is one of the major food items that contributes to the intake of dioxins and related compounds by consumers. The recent study by RIVM and RIKILT on levels of dioxins and related compounds, and dietary intake of these compounds has shown that for the average Dutch consumer, fish amounts to 16% of the total intake (Freijer et al., 2001). For regular fish consumers this value will be considerably higher.

Fish consumers eating fish from highly contaminated areas, for example eel caught in polluted rivers, can be exposed to very high levels of dioxins and related compounds. In such cases an advice to limit consumption of fish from such areas may be the most appropriate risk management option to decrease the intake of dioxins and related compounds. On the other hand, consumption of fish, in particular fatty fish, has shown to have beneficial health effects due to the presence of high levels of  $\omega$ -3 unsaturated fatty acids (Oomen et al., 2000). These beneficial effects may very well outweigh possible adverse effects of dioxins and related compounds in fish.

## 2. Terms of reference

In order to protect consumers from high dioxin intakes, the Ministry of Health, Welfare and Sports and the Ministry of Agriculture, Nature Management and Fisheries, have requested RIVM, RIVO and RIKILT to investigate the possibility to set maximum levels for dioxins and related compounds in fish and fish-derived products <sup>1</sup>). The advice should involve both dioxins (i.e., PCDDs and PCDFs) and the so-called dioxin-like PCBs, i.e., the non- and mono-ortho PCBs, which show similar toxicological properties <sup>2</sup>).

If possible the advice should take into consideration possible effects of food processing, like smoking, frying and cooking, on levels of dioxins and related compounds.

Furthermore, in view of the current costs and limited analytical capacity of the GC/MS reference method for dioxins in some countries, attention should also be given to the relationship between the so-called indicator PCBs (which are relatively easy to analyse) and the total amount of dioxins and related compounds. This aspect, however, was later decided to become the subject of a separate advisory report by RIVO.

Finally, the ALARA principle (As Low As Reasonably Achievable) should be applied wherever possible.

Currently eel is the only fish species for which maximum levels of dioxins are decreed (Warenwet, 2000, 2001).

<sup>&</sup>lt;sup>2</sup>) Up to now rules for maximum levels of dioxins in foodstuffs have been limited to dioxins only (*i.e.*, the sum of PCDDs and PCDFs). However, since the toxicity of the dioxin-like PCBs is very similar to the PCDDs and PCDFs (all include an interaction with the so-called Ah-receptor, see WHO, 2000) and the amount of dioxin-like PCBs in fish contribute for 50 % or more to the total amount of WHO-TEQ in fish (Scoop, 2000), while modern chemical-analytical facilities allow for a reliable determination of all three compound classes, it was decided to include the dioxin-like PCBs in the current advisory report.

## 3. Tolerable intake of dioxins and related compounds

Based on various effects in animals, like developmental, immunological and reproductive effects, the World Health Organisation (WHO) recommended in 1998 a tolerable daily intake (TDI) for dioxins and related compounds of 1 to 4 pg WHO-TEQ <sup>3</sup>) per kg body weight (bw) per day (WHO, 2000). It is important to understand that health effects are thought to be due to the long-term exposure of the most sensitive part of the population, to a large extent caused by the accumulation of these compounds in the body. Hence the TDI aims at lowering the intake of dioxins and related compounds in order to prevent tissue levels from reaching critical concentrations. Fluctuations in the daily exposure, not leading to a clear effect on tissue concentrations, are therefore thought to be of no relevance.

Recognising the above, and considering that dioxins and related compounds generally have very long half-lives in the human body, the Scientific Committee on Food (SCF) of the European Commission in its opinion of November 2000 (SCF, 2000) found it more appropriate to express the tolerable intake on a weekly rather than a daily basis. Consequently it derived a temporary TWI of 7 pg WHO-TEQ per kg bw per week (equivalent to 1 pg/kg bw/day). The SCF noted that at present a considerable proportion of the European population will exceed this t-TWI, and emphasised that a TWI is not a lower bound of toxicity, but an estimate of a safe level of intake which is derived in a conservative way using uncertainty factors applied to no-observed-adverse-effect levels or lowest-observed-adverse-effect levels.

Although the ultimate goal is the reduction of the exposure of the population to a level below the TWI of 7 pg WHO-TEQ/kg bw/week, this is not feasible on short notice. The current daily exposure level of the Dutch population to dioxins and dioxin-like PCBs is still at average 9.2 pg WHO-TEQ/kg bw/week (1.3 pg/kg bw/day), with a 90<sup>th</sup> percentile of 13.6 pg WHO-TEQ/kg bw/week (1.9 pg/kg bw/day) (Freijer et al., 2001).

In the current advisory report the value of 28 pg WHO-TEQ/kg bw/week (4 pg/kg bw/day, the upper limit of the TDI as established by the WHO) is used as the upper tolerable limit of exposure, resulting from an occasional consumption of fish containing the highest acceptable level. This value is used to compare the intake resulting from consumption of fish containing the highest 'tolerated' level of dioxins and related compounds. In addition, a weekly intake of 14 pg WHO-TEQ/kg bw/week (2 pg/kg bw/day) is used as reference point for consumption of fish on an average level of contamination.

<sup>&</sup>lt;sup>3</sup>) TEQ: Toxicity equivalents, see van den Berg et al., 1998.

# 4. Current consumption patterns of fish and fish-derived products in The Netherlands

Two food surveys were used to estimate the current consumption pattern of fish and fish products in The Netherlands. The first one, DNFCS (Dutch National Food Consumption Survey, in Dutch VCP: voedselconsumptiepeiling), is based on an accurate registration of food items consumed during two consecutive days by 6250 consumers. This survey allows a good estimation of the portions consumed from each food item, including various species of fish (see Table 1). The second one, the ANI-food frequency questionnaire (Dooren-Flipsen en van Klaveren, 1998, Dooren-Flipsen et al., 1999), involves the registration of the consumption frequency for specific food items by 1588 persons. In combination, these two surveys allow an estimation on the frequency of consumption of each type of fish or fish product, and on the portion size. Based on these estimates, three groups of consumers have been defined:

- (1) the average Dutch consumer (this is the mean consumer, including both fish consumers and non-consumers),
- (2) the fish consumer, eating fish at least once a week (constituting 20% of the total population), and
- (3) the frequent fish consumer, eating fish at least twice a week (constituting 6% of the total population).

Table 1 shows the fish consumption patterns for each of these consumer groups. It is important to note that frequent fish consumers (group 3) are included in the fish consumers (group 2), and that all fish consumers are included in the group of average consumers (group 1). The latter group also includes the group of consumers that do not eat fish products at all (40% of the total population). For the average consumer (group 1), fish contributes 16% of the total daily intake. In other words, the average consumer is exposed to 1.5 pg WHO-TEQ/kg bw/week (0.2 pg/kg bw/day) through the consumption of fish, and to 7.8 pg WHO-TEQ/kg bw/week (1.1 pg/kg bw/day) from sources other than fish (Freijer et al., 2001).

Obviously group 1 cannot be used to estimate the exposure to dioxins and related compounds from fish, because this would severely underestimate the intake of fish consumers. The data on fish consumption by the third group bear a substantial number of uncertainties due to the limited size of this group. It was thus decided to select the second group ('fish consumers') for further calculations. For this consumer fish contributes approximately 34% of the total daily intake of dioxins and related compounds.

Only fish species of which relatively large quantities are consumed by the Dutch consumer were used for the calculations. Table 2 shows the contribution of the various fish species to the exposure to dioxins and related compounds through fish consumption. These data are based on the average amounts consumed (cf. Table 1) and the levels of dioxins and related compounds in the different fish species as described in paragraph 6 and Table 3. The selection used for further estimates thus excludes species like sole, trout or turbot, of which on average only very small amounts are consumed.

## 5. Levels of dioxins and related compounds in fish

For a reliable prediction of the effects of different maximum levels on intake, substantial data on the levels of dioxins and related compounds in the various fish species are needed. Unfortunately, only very few studies are available on levels of both dioxins (PCDDs and PCDFs) and dioxin-like PCBs in fish. Recently RIVO performed a study (Leonards et al., 2000), using 1 to 9 pooled (ca. 25 fishes per pool) samples per type of fish. In general the levels of PCDDs, PCDFs and non-ortho PCBs are in a similar range as reported by other studies (SCOOP 2000), possibly with the exception of herring caught in the Baltic sea, and salmon. Most of the data reported in the SCOOP report originate from studies in the UK.

Next to this study 30 samples of fish have been analysed in the period 1998/99 by RIKILT (Freijer et al., 2001). In this study dioxins and non-ortho PCBs have been measured. Mono-ortho PCBs, normally contributing a relatively small amount to the total TEQ, were not reported. The RIKILT data were corrected for this by using the ratio of the TEQ of mono-ortho PCBs to the total TEQ, derived from the RIVO-study. The RIKILT and RIVO data were then pooled and used in further calculations (Table 3).

Additional information was obtained from a British dataset (Parsley et al., 1998). Although the fish measured in this study was sampled in 1995/96, it appears to be a useful additional dataset. The British study includes information on mono-ortho PCBs. Data from this study are also included in Table 3.

## 6. Effects of cooking, frying and smoking

The actual human intake of dioxins by consumption of fish is influenced by the commonly used cooking practices. Removal of the toxic substances may occur by volatilisation, extraction in the cooking oil or by discarding the fat drippings and removal of the (contaminant-rich) skin. Several studies reported reductions of PCBs and PCDDs/PCDFs, depending on the applied cooking process (Zabik et al., 1992, 1996; Zabik and Zabik, 1995; Salama et al., 1998; Schecter et al., 1998; Wilson et al., 1998).

Zabik et al. (1996) reported a reduction of 40% of the sum of PCBs through smoking of lake trout, whereas baking, charbroiling and (salt) boiling showed reductions of approximately 15%. In another study (Zabik et al., 1992) the total amount of PCBs in boiled blue crab was reduced by 25-36% compared with the raw material. Salama et al. (1998) found reductions of PCBs in Atlantic bleufish filets with smoking and microwave baking of 65 and 60%, respectively. Skin-off charbroiling, skin-on charbroiling, pan-frying and convection oven baking showed reductions of 46, 37, 27 and 39%, respectively. The sum of the congeners of PCDDs and dioxin-like PCBs in catfish were reduced by 43 and 32% wet weight, respectively (Schecter et al., 1998). The amount of PCDFs, however, was increased by 67%, caused by an increase of octachlorodibenzofuran, for which no explanation was given.

Reductions are presumably a result of the reduction of the amount of fat in the sample: Zabik and Zabik (1995) showed PCDD reductions through cooking of skin-on fillets of about 40 to 80% for different species, while cooking of skin-off fillets showed smaller reductions. Finally, Wilson et al. (1998) reviewed the available data from different studies and reported average PCB mass reductions of 28, 68, 28, 48 and 30% for baking, boiling, broiling, frying and smoking, respectively. Microwaving was applied in one study, reporting 26% reduction of PCBs. The same reductions may apply for PCDDs and PCDFs.

Although the average reductions due to processing appear to be significant, the data from the different studies were inconsistent and showed a considerable range. Besides, data were limited to a few species and do not cover all regularly consumed species.

It is extremely complicated to take the effect of cooking into account when assessing the human intake of dioxins and related compounds, as cooking methods vary (temperature, cooking time etc.), different methods are applied for different species and tissues, and some consumers might use the cooking liquid as a basis for sauce. In addition, cooking methods differ geographically and, in practice, this step is not controlled as are laboratory experiments. It will therefore be difficult to produce a quantitative estimate of the reduction of dioxins and related compounds for different species and cooking methods. In general, it can be estimated that cooking of fish can reduce levels of PCDDs, PCDFs and PCBs by some 15–20 %. But due to the lack of consistency in the available data sets it is at present not possible to quantify more precisely the effect of food processing on the levels of dioxins and related compounds in fish. The scenario studies as described in paragraph 8 thus ignore food processing.

#### 7. Scenario studies

## 7.1. Principles

Three principles have been applied in the derivation of possible maximum levels of dioxins and related compounds in fish. Firstly a particular species of fish should not contain that much dioxins and related compounds that the consumption of one portion of that particular fish results in a weekly intake exceeding the upper limit of exposure, 28 pg WHO-TEQ/kg bw (see paragraph 7.2). Secondly, because a fish consumer might eat different fish species in different quantities, it should be avoided that the upper bound intake of 28 pg WHO-TEQ/kg bw (see paragraph 7.3) is exceeded when different fish species with the maximum tolerated level of contamination are consumed. In addition, the average intake of the hypothetical fish consumer should be lower than 14 pg WHO-TEQ/kg body weight per week, in order to reduce body burdens in the longer term. The third principle is the ALARA-approach (As low As Reasonable Achievable). This principle is only applied for those scenarios meeting principles 1 and 2 (see paragraph 7.3).

# 7.2. Upper-bound limits for maximum levels in different fish species

Based on the consideration that the weekly intake of dioxins and related compounds should not exceed 28 pg WHO-TEQ/kg bw, and using the average portion size for each fish species as given in Table 1, upper limits for dioxins and related compounds in various fish species were estimated.

Assuming an average body weight of 65 kg and the current exposure from food items other than fish of 7.8 pg WHO-TEQ/kg bw/week (cf. paragraph 4), the <u>maximally</u> acceptable contribution from one weekly portion of fish is:  $65 \times (28-7.8) = 1313 \text{ pg WHO-TEQ}$ . Thus, one portion of herring (110 grams) is allowed a maximum upper level of 1313/110 = 12 pg WHO-TEQ/gram fish. Similarly, upper-bound levels can be calculated for the other fish species, resulting in 22 for mackerel, 11 for salmon, 24 for tuna, 9 for cod and coalfish, 8 for plaice, 24 for shrimps and 9 for mussels. In the case of eel the maximum level is 38 (which is slightly lower than the limit of 44 corresponding with the current maximum level of 8 pg TEQ/gram for the sum of PCDDs and PCDFs only, cf. Warenwet 2000, 2001)).

The above presented levels are the upper-bound limits and should ensure that the upper-bound level of the TWI (28 pg WHO-TEQ/kg bw/week) is not exceeded when eating once a week a portion of fish containing dioxins and related compounds <u>at the maximum</u> level.

## 7.3. Scenario studies

For a number of hypothetical maximum levels the effects on both the *maximum* intake of dioxins and related compounds (based on consumption of only fish with dioxins and related compounds at the particular maximum level) and the *average* intake were calculated, using the data shown in Tables 1 and 3.

The results of these various scenario studies are summarised in Tables 4a and 4b. Table 4a presents the scenario studies based on the Dutch dataset only (Leonards et al., 2000;

Freijer et al., 2001), Table 4b presents similar scenario studies based on the combined data of The Netherlands and the UK (Parsley et al., 1998).

For these scenario studies various maximum levels in different fish species were hypothesised. For fatty fish (eel excepted) levels ranging from 5 to 10 pg WHO-TEQ per gram were applied, and for lean fish levels of 2 and 3 pg WHO-TEQ per gram. For mussels and shrimps two maximum levels (5 and 8) were used. Regarding eel, maximum levels of 30 and 35 pg WHO-TEQ per gram eel were applied, based on (1) the ALARA principle, and (2) the very small proportion of highly contaminated eel in the total consumption of eel.

For each fish species a statistical distribution of the concentrations of dioxins and related compounds was made using a log-normal transformation of the mean and the standard deviation (cf. Table 3). The result of such a transformation for herring is illustrated in Figure 1. To model the daily variation in fish consumption, (theoretical) samples were taken from these distribution curves using Monte Carlo probability sampling techniques. Samples from this distribution curve exceeding the proposed maximum level, were assumed to be removed from the food market and not reaching the consumer. Consequently these samples were not included in the average intake in Tables 4a and 4b.

From these distribution curves also the level of non-compliance, i.e., the percentage of fish exceeding the hypothetical maximum level, can be determined. The resulting levels of non-compliance are also shown in Tables 4a and 4b. However, because the information on dioxin- (PCDDs and PCDFs) and PCB-levels in fish is limited, the estimation of the non-compliance percentage can only be made with a rather large margin of uncertainty.

These Monte Carlo simulations were done in first phase for the Dutch data only (Leonards et al., 2000; Freijer et al., 2001), of which the results are shown in Table 4a. However, since we are aware of the limitations due to the small numbers of dioxin data, in a second phase a British dataset was included (Parsley et al., 1998), shown in Table 4b.

### 8. Conclusions

#### 8.1. General

As stated in paragraph 7.2 herring should not contain more than 12 pg WHO-TEQ per gram fish and eel should not contain more than 38 pg WHO-TEQ/gram. Similarly, the upper-bound level for salmon is 11, for mackerel 22, for tuna 24, for cod and coalfish 9, for plaice 8, for shrimps 24, and for mussels 9 pg WHO-TEQ/gram fish. Based on the ALARA-principle and the observed levels of dioxins and related compounds (Table 3), maximum levels for salmon, mackerel, cod, coalfish, plaice, shrimps and mussels can be set at a lower level.

All scenarios result in maximum intake levels clearly below 4 pg WHO-TEQ/kg bw/day, and in average intake levels of approximately 1.6–1.7 WHO-TEQ/kg bw/day. The fractions of non-compliance are in general rather low. Only a scenario based on one limit for all fish species of 6 pg WHO-TEQ per gram product (not shown in Tables 4a and 4b) resulted in relatively high levels of non-compliance for some fish species (i.e., 5% for herring, 23% for tuna and 53% for eel).

Of all fish species considered, only the levels of dioxins and related compounds in eel and salmon (the two species commercially farmed) can actually be influenced by a vigorous control of the levels of these contaminants in the feed. Levels of dioxins and related compounds in farmed eel in The Netherlands are already relatively low compared with freshwater eel (Leonards et al., 2000), in contrast to the levels in salmon reared in marine environments. The levels in reared salmon can thus be decreased actively by careful selection of the feed. Applying the ALARA principle, scenario 5 estimates the effect of a separate and rather low threshold limit for salmon of 5 pg WHO-TEQ/gram fish.

Obviously most sets of hypothesised maximum levels result in only very small fractions of non-compliance. Consequently their effect on the reduction of the average exposure is very small. The estimated reduction in intake of dioxins and related compounds is in general limited to less than 1.5%.

However, it must be emphasised that for most fish species the datasets are very limited, which results in large uncertainties in the estimations of the fraction of noncompliance. This is illustrated by the fact that for most species measured in the recent Dutch study no levels exceeding 10, 2 and 5 for fatty fish, lean fish, and mussel/shrimp, respectively, have been observed (Leonards et al., 2000; Freijer et al., 2001). Therefore, the fractions of non-compliance are theoretical figures, based on statistical processing of the available data sets. An exception to this situation is eel, where samples exceeding levels of 30 and even 40 pg WHO-TEQ/gram fish have been observed. Although it applies for most types of fish, especially data for tuna should be interpreted with great care, since only results from three samples of tuna were used in this assessment.

The limitation of the small Dutch dataset was partly neutralised by introducing a British data set in the assessment. The results are listed in Table 4b. There is a remarkable change in the percentage non-compliance for herring. Setting a maximum level of 12 WHO-TEQ pg/gram herring will, according to this calculation, result in a percentage non-compliance of approximately 12%. This also results in a decrease in average intake of 1.79 to 1.65 WHO-TEQ/kg bw. Likewise the percentage of non-compliance of salmon in this calculation was slightly higher compared to the figures listed in Table 4a. As can be seen in Table 4b, dioxin limits for salmon of 5 or 6 WHO-TEQ pg/gram fish will result in a percentage non-compliance of 2.5% and 0.7%, respectively.

Finally it must be noted that limited data indicate a rather high content of dioxins and related compounds in products like fish liver. Since these products are only consumed by a

restricted group of very specific consumers, and the database is very limited indeed, it is not possible to quantify the health risks in consuming these products. However, from knowledge on the high PCB levels in fish livers, it is estimated that a serious health risk could be related to preference consumption of fish livers.

#### 8.2. Areas of concern

Problems may arise with respect to the import of fish from highly polluted waters, like cod and herring from the Baltic Sea, and possibly for tuna from the Mediterranean (but it should be noted that until now only one sample of tuna from that region has been analysed). Herring caught in or nearby Scottish estuaries seems to contain more contaminants than average (which appears to be the cause of the differences in herring data between the Dutch and British datasets). Also a proportion of eel caught in the IJsselmeer and the large rivers in The Netherlands may fail to meet the maximum levels as applied in this report.

## 8.3. Beneficial effects of fish consumption

The current advice regarding maximum levels for dioxins and related compounds in fish may be followed by specific maximum levels issued by the Dutch authorities. If so, such maximum levels may be accompanied by an advice with respect to the frequency of fish consumption. This must be balanced against the health-promoting effects of fish consumption which are the result of the high content of  $\omega$ -3 unsaturated fatty acids in fish. These unsaturated fatty acids are known to have a beneficial effect on the development of coronary heart diseases and atherosclerosis. Hence it has been recommended to consume fish at least once a week (Hartstichting).

At present, there is no formal advice from the Dutch Government or from the European Commission, although there is consensus about such an advice amongst health professionals (Kromhout, personal communication, 2001). This is based on scientific recommendations to reduce the intake of saturated fats by replacing them by mono- or polyunsaturated fatty acids from marine sources or vegetables (Wood et al., 1998; Oomen et al., 2000).

It is conceivable that a conflicting situation might occur due to the wish to reduce the intake of dioxins and related compounds by reducing fish consumption versus the positive effects of fish consumption as mentioned above.

#### **8.4.** Final considerations

In general, setting maximum levels for residues or contaminants in foodstuffs serves to protect the general population or well-defined sub-populations from exposures with potentially adverse effects on human health when consuming these foodstuffs.

In the present study it is demonstrated that setting maximum levels for dioxins and related compounds in fish according to the different scenarios above has only a marginal influence on the actual average exposure of the fish consumer to these compounds. In addition, the enforcement of those maximum levels is complicated. Only in farmed fish the

amount of dioxins and related compounds can be influenced actively, namely by decreasing their amount in the feed <sup>4</sup>).

In order to actively reduce the presence of dioxins and related compounds in feedingstuffs, maximum levels should be accompanied by measures stimulating a pro-active approach, such as the setting of action thresholds and target values for foodstuffs, in combination with measures to reduce emissions. In accordance with the opinion of the EC Scientific Committee on Food, target values indicate the contamination levels to be achieved in order to ultimately bring human exposure for the majority of the population down to the TWI value. Action thresholds are set as a tool to assist competent authorities to identify those cases where it is appropriate to initiate more detailed investigations into the identification and reduction or elimination of the sources if significant levels of dioxins and related compounds are found in foodstuffs. The approach will result in a gradual reduction of the levels of dioxins and related compounds in foodstuffs, whereby the target levels will ultimately be achieved after a certain period of time.

# Acknowledgement

We are indebted to the advises and critical remarks given by the members of the Working Group 'Dioxinen in voeding'.

<sup>&</sup>lt;sup>4</sup>) Recently the European Commission has proposed maximum levels for dioxins in animal foods and feeds, see the appendix.

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Fish consumption (in grams per week) by the average consumer, the fish Table 1. consumer (once or more per week), and the frequent fish consumer (twice or more per week)

			(	Consumption patte	rn
Category	Species	Portion size *)	Average consumer	Fish consumer (once or more	Frequent consumer
				per week)	(twice or more
					per week)
Fatty fish	herring	110	13.3	21.0	45.5
	mackerel	60	2.3	7.0	17.5
	salmon	125	8.7	12.6	27.3
	eel	35	0.9	2.1	4.9
	tuna	55	2.8	5.6	11.2
Lean fish	cod **)	152	21.5	69.3	106.0
	coalfish **)	152	10.0	46.9	70.3
	plaice	166	0.6	14.7	25.9
Shellfish	mussels	140	2.9	4.2	9.8
Crustaceans	shrimps	55	4.0	3.5	8.4
Total	-	-	67	187	327

Average size of the fish portion consumed per meal in grams (Dooren-Flipsen and van \*) Klaveren 1998, Dooren-Flipsen et al., 1999; Freijer et al., 2001)

Table 2. Contribution from different fish species to the intake of dioxins and related compounds from fish consumption \*)

Category	Species	Contribution to intake of dioxins and related compounds from fish consumption (%)
Fatty fish	herring	32
	salmon	21
	tuna	9
	eel	5
	mackerel	3
Lean fish	coalfish	11
	cod	3
	plaice	<1
Shellfish	mussel	8
Crustaceans	shrimp	2
	Others **)	5-6

Based on Leonards et al., 2000 and Freijer et al., 2001

<sup>\*\*)</sup> Cod and coalfish include products like "kibbeling", "lekkerbekjes" and fishsticks on a 50/50 basis

<sup>\*)</sup> \*\*) Including fish salad and other fish products

Table 3. Minimum and maximum levels of dioxins (PCDDs and PCDFs) and dioxin-like PCBs in different types of fish, as measured in The Netherlands \*) and the UK \*\*).

				Lev	els (pg WHO-7	ΓEQ/g fish)	
Category	Species			dioxins (PCDDs+PCDFs)	dioxin-like PCBs	comb	ined
			n	range	range	mean ± SD	range
Fatty	herring	NL	7	0.5 - 2.1	0.8- 3.9	3.0 ± 1.5	1.3- 5.7
		UK	10	0.3 - 3.8	0.5-10.4	8.6 ± 5.2	0.8-13.8
	mackerel	NL	6	0.2 - 1.3	1.1- 3.5	2.3 ± 1.2	1.3- 4.7
		UK	13	0.1 - 1.7	0.3- 6.0	3.1 ± 2.4	0.5- 7.5
	salmon	NL	4	1.1 - 1.4	2.1- 2.9	$3.9 \pm 0.4$	3.4- 4.3
		UK	12	0.6 - 1.0	1.3- 3.0	3.1 ± 0.6	2.2- 3.9
	tuna	NL	3	0.0 - 0.7	0.0- 9.0	4.7 ± 4.9	0.0- 9.8
	eel ***)	NL	9	0.7 - 3.9	3.1-32.8	13.9 ± 10.3	3.9-36.7
Lean	cod	NL	5	0.0 - 0.3	0.1- 0.5	0.4 ± 0.3	0.2- 0.8
		UK	30	0.0 - 0.1	0.0- 0.3	0.1 ± 0.1	0.0- 0.4
	coalfish	NL	4	0.1 - 0.1	0.1- 0.8	$0.5 \pm 0.3$	0.2- 1.0
	plaice	NL	4	0.2 - 0.3	0.2- 0.3	0.5 ± 0.1	0.4- 0.5
		UK	13	0.1 - 0.5	0.2- 0.9	$0.8 \pm 0.3$	0.2- 1.3
	haddock	NL	4	0.0 - 0.1	0.1- 0.2	0.2 ± 0.1	0.1- 0.3
		UK	26	0.0 - 0.1	0.0- 0.1	0.1 ± 0.0	0.0- 0.1
Shellfish	mussel	NL	4	0.5 - 1.5	0.7- 2.0	2.2 ± 1.0	1.2- 3.5
Crustaceans	shrimps	NL	6	0.1 - 1.0	0.1- 0.9	1.1 ± 0.6	0.2- 2.0

<sup>\*)</sup> Leonards et al., 2000; Freijer et al., 2001

<sup>\*\*)</sup> Parsley et al., 1998

<sup>\*\*\*)</sup> This includes both farmed eel and eel caught in the IJsselmeer, showing total dioxin and dioxin-like PCB levels of 3.9-10.7 and 8.7-36.7 pg TEQ/g, respectively

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Table 4a. Daily exposure of the fish consumer and non-compliance of fish species at different maximum levels, using Dutch data \*)

Due to a limited number of data points the intake reductions and the non-compliance fractions should be interpreted with caution.

		Hypothe	Hypothetical maximum level	imum leve	_	Intake	ke ¹)			Ē	raction of	non-con	) pliance	Fraction of non-compliance (percentage)			
Scenario		<u>'</u>	(pg TEQ/gram)	am)		(pg TEQ/kg	kg bw/day)		Fa	Fatty fish				Lean fish			
		Fatty fish		Lean	Mussel/	Max	Average	Herring	Mackerel	Salmon	Tuna	Eel	Cod	Coalfish	Plaice	Mussel	Shrimp
	ee	salmon	other	fish	Shrimp	2)	3)				<del>(</del>						
( <sub>s</sub> 0	1	1	-	1	1	-	1.67	0	0	0	0	0	0	0	0	0	0
_	30	10	10	3	8	3.29	1.65	0.3	0.1	0	9.6	1.2	0	0.1	0	0	0
2	30	10	10	3	5	3.24	1.64	0.3	0.1	0	9.6	1.2	0	0.1	0	1.5	0.1
က	30	10	10	2	80	3.00	1.64	0.3	0.1	0	9.6	1.2	0.1	0.5	0	0	0
4	30	10	10	2	5	2.95	1.64	0.3	0.1	0	9.6	1.2	0.1	0.5	0	1.5	0.1
2	30	5	10	2	5	2.81	1.64	0.3	0.1	1.3	9.6	1.2	0.1	0.5	0	1.5	0.1
9	35	5	10	2	5	2.82	1.64	0.3	0.1	1.3	9.6	0.8	0.1	0.5	0	1.5	0.1
7	35	9	10	2	5	2.84	1.64	0.3	0.1	0	9.6	0.8	0.1	0.5	0	1.5	0.1

-eonards et al., 2000; Freijer et al., 2001

Background exposure from non-fish food items is 1.11 pg WHO-TEQ/kg bw/day

Maximum intake calculated assuming that all fish consumed contains dioxins and related compounds at the upper-bound maximum level Estimated actual intake based on calculated distribution following removal of fish above the proposed maximum levels 

Tuna classified as fatty fish. Note that the small dataset for this species and the large range resulted in a large relative standard deviation which may

be the cause of a distorted non-compliance No limits applied

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Table 4b. Daily exposure of the fish consumer and non-compliance of fish species at different maximum levels, using Dutch\*) and British\*\*) data

Due to a limited number of data points the intake reductions and the non-compliance fractions should be interpreted with caution.

	Hypothe	Hypothetical maximum level	mum leve	_	Intake	ke ¹)			ш	raction of	non-com	pliance	Fraction of non-compliance (percentage)			
	<u>u</u> ;	(pg TEQ/gram)	am)		(pg TEQ/	(pg TEQ/kg bw/day)		Fa	Fatty fish				Lean fish			
	Fatty fish		Lean	Mussel/	Max	Average	Herring	Mackerel	Salmon	Tuna	Eel	Cod	Coalfish	Plaice	Mussel	Shrimp
ee	salmon	other	fish	Shrimp	2)	3)				4)						
1	-	-	ı	-	,	1.79	0	0	0	0	0	0	0	0	0	0
30	10	10	3	8	3.29	1.65	15.2	1.1	0	9.6	1.2	0	0.1	0	0	0
30	10	10	3	2	3.24	1.65	15.2	1.1	0	9.6	1.2	0	0.1	0	1.5	0.1
30	10	10	2	8	3.00	1.65	15.2	1.1	0	9.6	1.2	0.1	0.5	0	0	0
30	10	10	2	2	2.95	1.65	15.2	1.1	0	9.6	1.2	0.1	0.5	0	1.5	0.1
30	2	10	2	2	2.81	1.65	15.2	1.1	2.5	9.6	1.2	0.1	0.5	0	1.5	0.1
35	5	10	2	5	2.82	1.65	15.2	1.1	2.5	9.6	0.8	0.1	0.5	0	1.5	0.1
35	9	10	2	5	2.84	1.65	15.2	1.1	2.0	9.6	8.0	0.1	0.5	0	1.5	0.1

Leonards et al., 2000; Freijer et al., 2001

Parsley et al., 1998

Background exposure from non-fish food items is 1.11 pg WHO-TEQ/kg bw/day £ \$ - 2 £ 4

Maximum intake calculated assuming that all fish consumed contains dioxins and related compounds at the upper-bound maximum level Estimated actual intake based on calculated distribution following removal of fish above the proposed maximum levels

Tuna classified as fatty fish. Note that the small dataset for this species and the large range resulted in a large relative standard deviation which may be the cause of a distorted non-compliance

No limits applied

In RIVM-report 639102 023 "Scenario studies on maximum levels for dioxins, dibenzofurans and dioxin-like PCBs in fish", the lower left panel of figure 1 (page 24) has disappeared in the printing process. The complete figure is presented below.

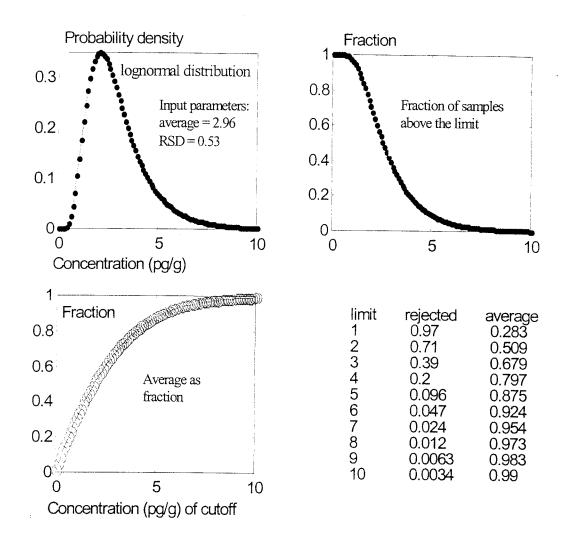


Figure 1 of RIVM-report 639102 023

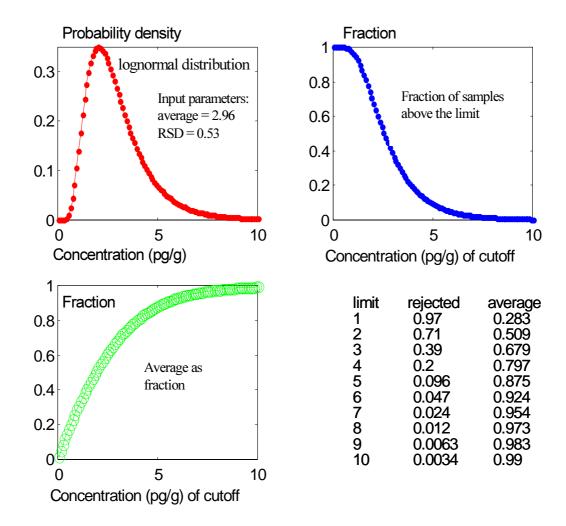


Figure 1

Illustration of the calculated distribution of levels of dioxins in Dutch herring (upper left panel). The fraction of fish above a certain limit value is equal to the surface of the corresponding tail of this distribution. For each limit value such a tail can be calculated. The result is shown in the upper right panel. Assuming that the critical level is fully effective as cut-off limit, the average concentration of the remaining part, relative to the average of all samples, is shown in the lower left panel. Finally, the lower right panel shows some tabulated values for the estimated fraction of rejected samples as the average of the remaining fraction divided by the average of all samples as function of the limit values.

A significant reduction of the average can only be achieved by rejection of a substantial part of the distribution.

Note that the distribution is calculated on the basis of the result of only 4 measurements, and will therefore have a large uncertainty.

### **APPENDIX**

# Effect of the recently proposed EC-maximum levels for dioxins (PCDDs and PCDFs) on intake and non-compliance

#### Introduction

Very recently the EC issued a draft proposal to set new maximum levels for dioxins (PCDDs and PCDFs only, so excluding dioxin-like PCBs) in different foodstuffs (SANCO/0384/01, 20 March 2001). For fish and fish products a limit of 4 pg WHO-PCDD/PCDF-TEQ per gram fresh product is proposed as of 01-01-2002, to be decreased to 3 pg TEQ/gram as of 01-01-2006.

Table 1(app) shows the relative contribution of the PCDDs and PCDFs to the total level of dioxins and related compounds (PCDDs, PCDFs and dioxin-like PCBs), expressed in WHO-TEQ, for the data presented in Table 3. In addition, the fraction of samples exceeding the EC-proposed limits of 4 and 3 pg TEQ per gram fish are given, following fitting of the data according to a log-normal distribution.

#### Results

The data in Table 1(app) clearly demonstrate that PCDDs and PCDFs in general contribute for about 20-50% to the total level of dioxins and dioxin-like compounds. In the case of lean fish these levels (PCDDs and PCDFs) are well below 1 pg TEQ/gram, and the EC-proposed limits would not constitute any non-compliance problem. The same holds for mussels and shrimps, which in the Dutch study (Leonards et al., 2000; Freijer et al., 2001) reached levels up to 1 and 1.5 pg TEQ/gram, respectively. Dioxin levels in fatty fish (tuna, salmon and mackerel) were below 2 pg TEQ/gram, and as such well below the newly proposed limits. Herring may pose a problem, in particular when the limit would be reduced to 3 pg TEQ/gram. However, again this is due to the herring from the UK-study, sampled in Scottish harbours, and possibly different from the herring consumed in The Netherlands.

The major problem appears to be eel. Based on fitting of the data on PCDDs and PCDFs of the Dutch study (Leonards et al., 2000; Freijer et al., 2001), 9% of the samples would exceed a limit of 4 pg TEQ/g, and 20% of the samples would exceed a limit of 3 pg TEQ/g.

Eel was further investigated recently by the Keuringsdienst van Waren (KvW; de Vries, 2001; Hoogenboom et al., 2001). The CALUX screening assay was used to select samples suspected to contain ≥8 pg TEQ (PCDDs and PCDFs) per gram (the maximum level for dioxins in eel as decreed by the Dutch authorities in October 2000; Warenwet, 2000, 2001). The criterium for this selection was an action limit of 30 pg TEQ (sum of PCDDs, PCDFs and dioxin-like PCBs) per gram as measured in the CALUX screening assay. In total 153 samples of eel were analysed, of which 22 samples were estimated to contain more than 30 pg TEQ per gram. Consequently these 22 samples were analysed by GC/MS for the exact amount of dioxins, in addition to 17 samples estimated to contain levels between 20 and 30 pg TEQ/g. Based on the GC/MS results, the EC-proposed limit of 4 pg TEQ/gram for dioxins results in about 11% non-compliance (non-fitted data), which is very similar to the approximately 9% non-compliance which results if a limit value of 30 pg TEQ/gram for the sum of dioxins and related compounds is applied according to the method outlined in paragraph 8 of the present advisory report. Likewise, the EC-proposed limit of 3 pg TEQ/gram for dioxins might result in at least 23% non-compliance. It must be noted, however, that in particular the latter figure is likely to underestimate the actual situation, since the selection was based on a suspicion of exceeding 8 pg TEQ/gram for dioxins, so that also

samples below the screening action limit have some probalility to exceed 3 pg TEQ/gram. In fact, all samples exceeding the action limit in the CALUX assay (30 pg TEQ/g) would actually have exceeded a dioxin limit (PCDDs and PCDFs) of 3 pg TEQ/gram, and 76% (13 samples) of the additional 17 samples, predicted to contain 20-30 pg TEQ/g, thus indicating that also part of the samples predicted to contain less than 20 pg TEQ/g might have exceeded this limit. In the case of a limit of 4 pg TEQ/gram these figures were respectively 59 (13 out of 22) and 24% 4 out of 17), indicating a lower chance of underestimation.

#### Effects on intake of dioxins by the consumer

Since the data presented in Table 1(app) suggest that a limit of 4 pg TEQ/gram for dioxins only would not result in high levels of non-compliance, the consequences for the actual exposure of the Dutch population seem to be very minor, and actually be less than the effects of the maximum levels used in the scenarios presented in Table 4b. This is primarily due to the data in UK herring, which partly exceed a limit of 10 pg TEQ/gram for dioxins and related compounds, but do not exceed the EC-proposed limit of 4 pg TEQ/gram for dioxins only. A further reduction to 3 pg TEQ/gram, however, could have more dramatic effects due to the higher fraction of herring and eel exceeding this level.

As demonstrated by the UK-data on herring, a maximum level of 4 pg TEQ/gram for dioxins only does not guarantee that the overall exposure to dioxins and dioxin-like PCBs will not exceed the upper limit of the TWI, i.e. 28 pg TEQ/kg bw/week.

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Dioxin levels and relative contribution of dioxins and furans (PCDDs and PCDFs) to the total level of dioxin-like compounds (expressed in TEQs) in different types of fish, as measured in The Netherlands \*) and the UK \*\*). Non-compliance data are calculated using a lognormal distribution Table 1(app)

				Dioxins (PCDDs/PCDFs)	Os/PCDFs)	Relative o	Relative contribution	Non-compliance	npliance
Category	Species			(pg TEQ/g)	(g/C	(%)	(%)	(%)	(9
			ㅁ	mean ± SD	range	mean ± SD	range	>4 pg TEQ/g	>3 pg TEQ/g
Fatty fish	herring	٦	7	$1.29 \pm 0.57$	0.5 - 2.1	$45 \pm 10$	32 - 54	0.2	1.4
		¥	10	$2.44 \pm 1.27$	0.3 - 3.8	32 ± 9	25 - 53	10.5	25.2
		a a	17	$1.96 \pm 1.17$				5.8	14.7
	mackerel	۲	9	$0.52 \pm 0.39$	0.2 - 1.3	19±8	15 - 28	0	0.2
		¥	13	$0.66 \pm 0.49$	0.1 - 1.7	23 ± 5	14 - 30	0.1	0.4
		a a	19	$0.62 \pm 0.45$				0.1	0.3
	salmon	٧	4	$1.30 \pm 0.11$	1.1 - 1.4	34 ± 4	29 - 38	0	0
		¥	12	$0.80 \pm 0.13$	0.6 - 1.0	26 ± 6	20 - 41	0	0
		a	16	$0.92 \pm 0.26$				0	0
	tuna	۲	က	$0.44 \pm 0.37$	0.0 - 0.7	18 ± 14	11 – 24	0	0.1
	eel ***)	۲	တ	$2.26 \pm 1.16$	0.7 - 3.9	18± 5	11 - 24	8.7	20.4
Lean fish	poo	۲	2	$0.11 \pm 0.11$	0.0 - 0.3	$32 \pm 11$	24 - 39	0	0
		¥	30	$0.04 \pm 0.03$	0.0 - 0.1	$42 \pm 18$	11 - 79	0	0
		a	35	$0.05 \pm 0.05$				0	0
	coalfish	۲	4	$0.11 \pm 0.02$	0.1 - 0.1	21 ± 16	10 - 33	0	0
	plaice	٧	4	$0.21 \pm 0.03$	0.2 - 0.3	52 (n=1)	52 (n=1)	0	0
		¥	13	$0.28 \pm 0.12$	0.1 - 0.5	37 ± 6	27 - 48	0	0
		a	17	$0.27 \pm 0.11$				0	0
	haddock	۲	4	$0.07 \pm 0.03$	0.0 - 0.1	48 (n=1)	48 (n=1)	0	0
		¥	56	$0.03 \pm 0.02$	0.0 - 0.1	$48 \pm 20$	10 - 81	0	0
		a a	30	$0.03 \pm 0.02$				0	0
Shellfish	mussel	٧	4	$0.98 \pm 0.40$	0.5 - 1.5	45± 4	42 - 48	0	0.1
Crustaceans	shrimps	¥	9	$0.60 \pm 0.30$	0.1 - 1.0	52± 4	49 - 58	0	0
1						: :		:	;

Data on fish in The Netherlands were taken from Leonards et al., 2000, and Freijer et al., 2001. Since in the latter case mono-ortho PCB levels were not determined, the relative contribution of dioxins was only based on Leonards et al. (2000)

<sup>\*\*)</sup> Data from Parsley et al., 1998
\*\*\*) Data from the KvW-report (de Vries, 2001) not included

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