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# Industrial Contaminants and Pesticides in Food Products

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### 1. Introduction

In our modern world a large number of man-made chemicals are being used. As a consequence their widespread presence in the environment is becoming increasingly well documented (Vethaak et al, 2002; Peters et al., 2008). They are found in a vast range of consumer products and include plasticizers, emulsifiers, flame retardants, perfluorinated compounds, artificial musks and organotin compounds. While they have undoubtedly improved the quality of our lives, a consequence of their intensive use is a widespread presence in the environment. Human exposure to these compounds may be through contact with consumer products containing such chemicals as additives, but also through food products. Since many of these compounds have a lipophilic nature there is a potential for bio-accumulation through the food chain especially in products with a high fat content. This is reflected in the presence of persistent organic compounds such as organochlorine pesticides and polychlorinated biphenyls that can be found in food products although there use has been seized many years ago. Many of these compounds have also been found in human blood indicating that humans are exposed to these chemicals (CDC, 2001, 2003; Guenther et al., 2002). This exposure may be through different routes. One is the use of these chemicals as additives in consumer products such as carpets, curtains, toys and electronic equipment. The exposure of these chemicals in house dust indicates the potential for human exposure. Another route for human exposure is, of course, through food products. Since many of these compounds have a lipophilic nature, they can be bio-accumulated through the food chain especially in products with a high fat content. This study focused on the presence and concentrations of a number of typical man-made chemicals in food products that many of use daily. The chemicals considered in this study are: brominated flame retardants (BFR's), phthalates, artificial musks, alkylphenols (AP's), organochlorine pesticides (OCP's), polychlorobiphenyls (PCB's), organotin compounds (OT's) and perfluorinated compounds (PFC's).

### 2. Methods and materials

## 2.1 Sampling and sample pre-treatment

All samples, mostly fresh food products were purchased in regular shops in nine European countries including the Netherlands, the United Kingdom, Germany, Finland, Sweden,

Spain, Poland, Italy, Estonia and Greece. Samples were sent to the laboratory where laboratory samples were prepared and stored at -18°C until analysis. In general, solid food samples were cut into small pieces and homogenised with a blender. If not the entire sample was used or homogenised, proportional sub-sampling was applied and the collected sub-samples were homogenised. Milk was acidified with formic acid and the solid part containing the proteins and fat was separated from the liquid phase. Both parts were stored for analysis. Orange juice was centrifuged and vacuum filtrated and the solid and liquid parts were stored for analysis. A selected number of chemical parameters were determined in each sample, based on expectations and reports in the literature.

# 2.2 Chemical parameters

The chemical parameters determined in this study are listed in table 1, including the abbreviations that are used throughout the text and in the result tables. Note that not all parameters are determined in all samples.

## 2.3 Analytical procedures

For the determination of the OCP's, PCB's, BFR's, phthalates and artificial musks, a weight sub-sample of the homogenised laboratory sample was mixed with anhydrous sodium sulphate in a mortar and spiked with internal standards. The internal standards used were <sup>13</sup>C-labelled standards for PCB's and BFR's, <sup>2</sup>D-labelled standards for OCP's and phthalates, and a surrogate standard for the artificial musks. The samples were Soxhlet extracted for 16 hours using a mixture of 10% diethyl ether in hexane. For milk and orange juice a proportional amount of the liquid phase was pre-extracted with hexane and this hexane extract was used in the Soxhlet extraction of the solid part of these samples. Olive oil was directly diluted in hexane. One procedural blank, consisting of 40 g anhydrous sodium sulphate, was included in every batch of 10 samples. All extracts were concentrated to a volume of 50 ml and split into two equal parts of 25 ml. For the determination of the OCP's, PCB's and BFR's, one part of the extract was washed several times with sulphuric acid of increasing concentration to remove the major part of the lipids. The remaining extract was concentrated and purified over a glass chromatographic column packed with florisil and capped with anhydrous sodium sulphate to isolate the fraction containing the OCP's, PCB's, PBDE's and HBCD. The eluent was concentrated to a small volume and a syringe standard (1,2,3,4-tetrachloronaphthalene) was added. This final extract was analysed on an Agilent 6890 series gas chromatograph coupled to an Agilent 5973 mass spectrometer (GC/MS) and equipped with a HP-5-MS, 30 m  $\times$  0.25 mm (i.d.), film thickness 0.25  $\mu$ m, fused silica capillary column. The mass spectrometer was operated in the selected ion monitoring mode and typically two or three characteristic ion masses were monitored for each analyte. The samples were analyzed for the following OCP's;  $\alpha$ -,  $\beta$ - and  $\gamma$ -hexachlorohexane (HCH), hexachlorobenzene (HCB),  $\alpha$ - and  $\beta$ -chlordane, o,p'-, p,p'-DDE, o,p'-, p,p'-DDD and o,p'-, p,p'-DDT: The following PCB congeners: 18, 28/31, 22, 41/64, 44, 49, 52, 54, 56/60, 70, 74, 87, 90/101, 99, 104, 105, 110, 114, 118, 123, 138, 141, 149, 151, 153/168, 156, 157, 158, 167, 170, 177, 180, 183, 187, 188, 189, 194, 199 and 203: The following PBDE congeners: 17, 28, 32, 35, 37, 47, 49/71, 66, 75, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 166, 181, 183, 184, 190, 191, 196, 197, 206, 207 and 209 and HBCD.

For the determination of the phthalates and artificial musks the second part of the extract was purified using a dimethylformamide-hexane partitioning to remove lipids. In this

Organochlorine pesticides (OCPs):	Phthalates:							
α-hexachlorohexane (α-HCH)	di-methyl phthalate (DMP)							
β-hexachlorohexane (β-HCH)	di-ethyl phthalate (DEP)							
γ-hexachlorohexane (γ-HCH) = lindane	di-isobutyl phthalate (DiBP)							
hexachlorobenzene (HCB)	di-butyl phthalate (DBP)							
α-chlordane	benzylbutyl phthalate (BBP)							
β-chlordane	di-(2-ethylhexyl) phthalate (DEHP)							
o,p'-DDE	di-isononyl phthalate (DiNP)							
p,p'-DDE	di-isodecyl phthalate (DiDP)							
o,p'-DDD								
p,p'-DDD								
o,p'-DDT								
p,p'-DDT								

Polychlorinated biphe	enyls (PCBs):	Polybrominated diphen	ylethers (PBDEs):
PCB-18	PCB-118	BDE-17	BDE-138
PCB-28/31	PCB-123	BDE-28	BDE-153
PCB-22	PCB-138/158	BDE-32	BDE-154
PCB-41	PCB-141	BDE-35	BDE-156
PCB-44	PCB-149	BDE-37	BDE-166
PCB-49	PCB-151	BDE-47	BDE-181
PCB-52	PCB-153/168	BDE-49/71	BDE-183
PCB-54	PCB-156	BDE-66	BDE-184
PCB-56/60	PCB-157	BDE-75	BDE-190
PCB-64	PCB-167	BDE-77	BDE-191
PCB-70	PCB-170	BDE-85	BDE-196
PCB-74	PCB-177	BDE-99	BDE-197
PCB-87	PCB-180	BDE-100	BDE-206
PCB-90	PCB-183	BDE-119	BDE-207
PCB-99	PCB-187	BDE-126	BDE-209
PCB-101	PCB-188		
PCB-104	PCB-189	Other brominated flame	retardents:
PCB-105	PCB-194		
PCB-110	PCB-199	tetrabromobisphenol-A (	TBBPA)
PCB-114	PCB-203	hexabromocyclododecan	e (HBCD)

Perfluorinated chemicals (PFCs):	Artificial musks:
perfluoro-octanoic acid (PFOA)	galaxolide (HHCB)
perfluoro-octane sulphonate (PFOS)	tonalide (AHTN)
perfluoro-octane sulfonamide (PFOSA)	musk xylene (MX)
perfluoro-nonanoic acid (PFNA)	musk ketone (MK)
perfluoro-decanoic acid (PFDA)	
perfluoro-undecanoic acid (PFUnA)	
perfluoro-dodecanoic acid (PFDoA)	Organotin compounds:
perfluoro-tetradecanoic acid (PFTrA)	
	mono-butyltin (MBT)
Alkylphenols (AP):	di-butyltin (DBT)
	tri-butyltin (TBT)
nonylphenol isomers (NP)	mono-octyltin (MOT)
octylphenol isomers (OP)	di-octyltin (DOT)

Table 1. Chemical parameters determined in this study including abbreviations used in the text and result tables

partitioning the hexane extract is extracted with dimethylformamide (DMF) to isolate the phthalates and artificial musks. After removal of the hexane layer, water is added to DMF fraction and the analytes are re-extracted into fresh hexane. This extract was concentrated and purified over a glass chromatographic column packed with florisil and capped with anhydrous sodium sulphate to isolate the fraction containing the phthalates and artificial musks. The eluent was concentrated to a small volume, the syringe standard was added and this final extract was analysed with the identical GC/MS system as described above. The mass spectrometer was operated in the selected ion monitoring mode and typically two or three characteristic ion masses were monitored for each analyte. The samples were analyzed for the following phthalates; di-methyl- (DMP), di-ethyl- (DEP), di-isobutyl- (DIBP), di-butyl- (DBP), benzylbutyl- (BBP), di-(2-ethylhexyl)- (DEHP), di-isononyl- (DINP) and di-isodecyl phthalate (DIDP): The following artificial musks; musk ketone (MK), musk xylene (MX), tonalide (AHTN) and galaxolide (HHCB).

AP's and the brominated flame retardant tetrabromobisphenol-A (TBBPA) were isolated using a steam distillation procedure described by Guenther et al., 2002. In the round bottom flask of the steam distillation apparatus typically 10 g of the laboratory sample was spiked with an internal standard and mixed with 250 ml Milli-Q water to which was added 1 ml of concentrated hydrochloric acid and 20 g of sodium chloride. During the overnight distillation process the organic phenols are isolated in the organic solvent, in this case hexane, in the backflow cooling system of the apparatus. The hexane extract is isolated, dried with anhydrous sodium sulphate and concentrated. Following drying the extract was reduced in volume nearly to dryness under a stream of nitrogen and re-dissolved in 1 ml of methanol. During concentration of the hexane extract care has to be taken to avoid losses of the AP's and rinsing of the glass surfaces with methanol is necessary. The methanol extract is analysed with liquid chromatography coupled with mass spectrometry (LC/MS) in the selected ion monitoring mode.

For the determination of perfluorinated compounds, specifically perfluoro-octanoate (PFOA), perfluoro-octane sulphonate (PFOS) and perfluoro-octane sulphonamide (PFOSA) sub-samples of 5 gram were collected in 50 ml poly-propylene tubes and extracted using acetonitrile. The samples were centrifuged and the clear liquid is decanted and purified over a glass chromatographic column packed with florisil, silica, LC-NH<sub>2</sub> and activated carbon. The residue in the poly-propylene tube is extracted two times more and each extract is decanted over the same chromatographic column, thus combining the purified extracts. 0.5 ml octanol is added as a keeper and the extracts are concentrated to a small volume.

### 3. Results

# 3.1 Validation of analytical methods

The analytical methods used were validated previous to the execution of this study. The parameters determined were linearity, repeatability, recovery from the rainwater matrix and method detection limits (MDL). For the determination of the organochlorine pesticides, polychlorinated biphenyls, brominated flame retardants, phthalates, artificial musks and organotin compounds internal standards were added to each sub-sample prior to analysis. For the perfluorinated compounds two relevant samples are spiked with the compounds of interest and analysed. The recovery of the internal standard and spikes were in the range of 70 to 140% with the exception of artificial musks where recoveries in the range of 56 to 87% were found, and the organotin compounds where recoveries were in the range of 67 to 99%.

With the exception of PCBs and organotin compounds, the results are not corrected for the recovery of the internal standards or spikes since the spikes used were not compound specific and their recovery is only used to evaluate the performance of the method. With each series of ten samples a blank sample was included. For the blank analysis the complete analytical procedure was followed, including all chemicals and solvents, but no sample was added. Blank results were only found for the phthalates DEHP, DIBP and DBP, The results were corrected for these blank values and the detection limits were raised to 10 ng/g for DIBP and DBP, and to 20 ng/g for DEHP.

# 3.2 Organochlorine pesticides

DDT, pesticides (OCPs) Organochlorine include compounds like lindane, hexachlorobenzene and chlordane, among others. DDT is a well-known agricultural insecticide that has been used extensively on a global basis for over 40 years. Although their manufacture and application are now largely prohibited or restricted in industrialized western countries because of their toxicity and persistence, they can still be found in environmental and biological matrices due to their persistence. Pesticide exposure has been associated with arthritis, diabetes, neurobehavioral changes and DNA damage Cox et al., 2007; Lee et al. 2007; Jurewicz et al., 2008; Rusiecki et al., 2008). The structures of p,p'-DDT and it's breakdown product p,p'-DDE, are shown below.

As a part of its monitoring program, the Food and Drug Administration (FDA) determines the levels of pesticide residues in a wide variety of foods typically consumed by Americans. Over the past ten years, these surveys have detected DDE and other OCPs in a variety of foods including meat, fish and shell fish products, eggs, root vegetables, legumes (beans, peas, and peanuts), some fruits, and leafy greens. In 1999 DDE or DDT were detected in 22% of the 1,040 food items analysed in the FDA Total Diet Study (FDA, 1999). The results for the 2003 Total Diet Study indicate DDT, but mainly DDE in 18% of the various food items in concentrations ranging from 0.1 to 11 ng/g (FDA, 2003). Those for chlordane and lindane range from 0.1 to 3.8 and from 0.1 to 8.4 ng/g product. In general, the concentrations as well as the frequency of detection of OCPs were lower in the 2003 study. The results of the OCPs in this study are presented in table 2 at the end of this section, and graphically in figure 1. OCPs are found in 17 of the 25 samples. The predominant OCPs that are detected are p,p'-DDE and HCB both found in 15 of the 25 samples. In addition o,p'-DDE and cis-chlordane were found in one and two samples respectively. The maximum concentration found for p,p'-DDE was 5.6 ng/g in a sample of pickled herring. The median concentrations for p,p'-DDE and HCB were 0.43 and 0.14 ng/g. Compared to the FDA's Total Diet Study, p,p'-DDE and HCB are found more frequently but in lower concentrations. This compares with a recent study of Schecter et al. who reported p,p'-DDE in 23 out of 31 samples in concentrations ranging from 0.06 to 9.0 ng/g with a median value of 0.51 ng/g (Schecter et al., 2010).

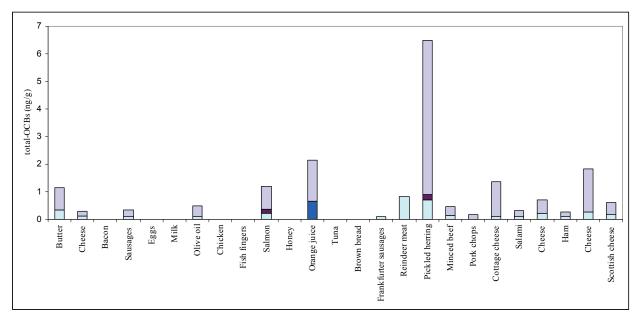


Fig. 1. Graphic presentation of the total-OCP concentration in 25 food items

#### 3.3 Polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) are marketed as cooling or insulating fluids for transformers, as softeners in the varnish and adhesive industries, and as hydraulic fluids. These compounds are not combustible, are heat resistant and make good solvents. On the other hand there is a severe toxic effect of PCBs, which damage the organs responsible for metabolism and also the nervous system (Guo et al., 1995; Ribas-Fito et al., 2001). Because of their persistence, PCBs are widely spread in the environment and, due to their good liposolubility, are easily deposited and concentrated in human, animal and plant tissue. The structures of two of the seven indicator PCBs, PCB-28 and PCB-180 are shown below.

2,4,4'-trichlorobiphenyl (PCB 28) 2,2',3,4,4',5,5'-heptachlorobiphenyl (PCB 180)

In the Total Diet Study of 2003 the levels of the sum of i-PCBs (e.g. PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-153 and PCB-180) ranged from 6 to 70 ng/g product. The Estonian Environment Research Centre determined levels of the sum of i-PCBs in Estonian butter and found a relative narrow concentration range of 5.2 to 8.8 ng/g lipid, translating to a range of 4.2 to 7.0 ng/g on a wet weight basis (EU, 2004). A study of the European Food Safety Association (EFSA) of different food types sampled across Europe between 1997 and 2003 reports median concentrations for the sum of i-PCBs ranging from 1.4 to 12 ng/g. Only fish oil showed a higher median concentration of 52 ng/g for the sum of i-PCBs (Gallani et al., 2004). The results for the PCBs in this study are presented in table 2 at the end of this

section. A graphic summary is given in figure 2. PCBs are found in every sample with PCB-18, -28 and -52, with the highest frequency. The highest PCB concentrations were found in the pickled herring and the salmon with concentrations for the sum of i-PCBs of 13 and 3.3 ng/g. The highest concentration of an individual PCB was for PCB-153, 5.5 ng/g in the salmon. The results found are comparable with those reported by EFSA for different food types sampled across Europe.

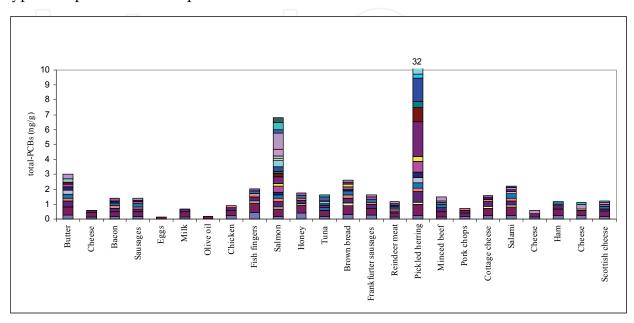


Fig. 2. Graphic presentation of the total-PCB concentration in 24 food items

# 3.4 Brominated flame retardants

Brominated flame retardants (BFRs) are widely used in electronic household equipment, plastics, textile and polyurethane foam in furniture and cars for safety reasons. Of the brominated products, about one-third contain tetrabromobisphenol-A (TBBPA) and derivatives, another third contains various bromines, including hexabromocyclododecane (HBCD) and the last third contains polybrominated diphenylethers (PBDEs). All three types of BFRs are determined in this study. The PBDEs are commercial mixtures with different degrees of bromination and used as additives to fireproof polymers. HBCD is a cycloaliphatic brominated chemical introduced as a replacement for the PBDEs and with the same applications. TBBPA is mainly used as a reactive (chemically bound) flame retardant in epoxy polymers such as printed circuit boards in electronic equipment. The structure of BDE-209 (better known as deca-BDE), HBCD and TBBPA is shown below. In a study by the Dutch National Institute for Public Health and the Environment the levels of various PBDEs, HBCD and TBBPA in 84 food products were determined (de Winter-Sorkina et al., 2003, 2006). With the exception of fish products PBDEs were absent or present in low concentrations (<0.1 ng/g) in food products. For the fish products the concentrations of the congeners BDE-28, -47, -99, -100, -153 and -154 ranged from 0.1 to 14 ng/g. BDE-209 was found in none of the 84 products while TBBPA was found in 7 products in concentrations ranging from 0.1 to 3.4 ng/g. Surprisingly, HBCD was found in 28 of the 84 samples in concentrations ranging from 0.1 to 8.9 ng/g product. TBBPA and HBCD were predominantly found in fish products, especially in eel.

A recent report on the results of a round robin exercise for BFRs in environmental, human and food samples gives some results for herring and salmon. The highest concentrations are found for BDE-47, up to 9.3 ng/g in herring and 0.89 ng/g in salmon. Those for the BDE-28, -99, -100, -153, -154 and -183 are in the range of 0.1 to 1.3 ng/g. BDE-209 and HBCD were not detected in these samples. The results for PBDEs in typical market basket studies show some differences, while studies in Spain (Bocio et al., 2003) and Japan (Ohta et al., 2002, 2008) show a predominance of the tetra- and penta-BDE with maximum concentrations up to 0.34 ng/g, an American study reports PBDE levels up to 3.1 ng/g product with surprisingly a predominance of BDE-209 which comprised as much as 50% of the total PBDE content in some of the samples (Schecter et al., 2004). The results for the BFRs in this study are reported in table 3 at the end of this section, and graphically in figure 3. PBDEs were found in 19 of the 24 samples. BDE-209, TBBPA and HBCD were found in none of the samples. BDE-47, -32 and -99 seem to be the predominant and the highest concentration for an individual PBDE was 0.65 ng/g found for BDE-47 in the sample of Scottish Cheese. Surprisingly, and different from other studies, only a limited number of BFRs were found in the fish products, in salmon no BFRs were found at all. The total PBDE concentrations ranged from 0.15 to 1.2 ng/g with the highest concentration in the sample of minced beef. The concentrations found in this study are therefore comparable with those found in the Spanish and Japanese study and much lower than those found in the American food study.

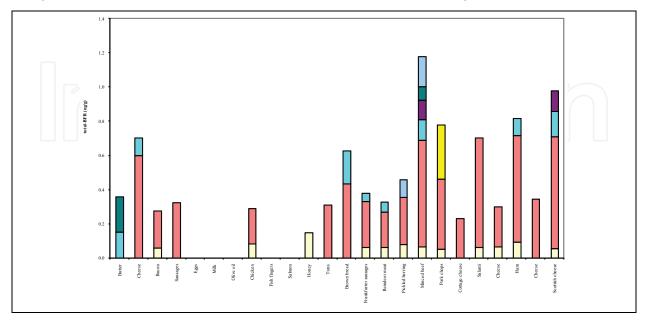


Fig. 3. Graphic presentation of the total-BFRs concentration in 24 food samples

#### 3.5 Perfluorinated compounds

Perfluorinated compounds (PFCs) are synthetic compounds characterised by an alkyl chain in which the hydrogen atoms are completely replaced by fluorine atoms. PFCs are heat stable, very resistant to degradation and environmental breakdown and have an amphiphilic nature (they repel water as well as oil). Because of these properties PFCs are used a myriad of applications, such as non-stick pans, stain and water repelling coatings for clothing, furniture and paper with typical brand names as Teflon, Gortex, Stainmaster and Scotchguard (3M-company, 1999). PFCs accumulate in the environment and they have been detected far from manufacturing plants in birds, marine plants and mammals from the Arctic to the Pacific and Indian Oceans and in land creatures in Europe and the USA (Kannan et al., 2002; Martin et al., 2003, 2004). In addition PFCs have been found in human blood (Kannan et al., 2004; Peters, 2005). The structures of the two most common PFCs, PFOA and PFOS are given below.

perfluoro-octanoic acid (PFOA)

perfluoro-octane sulfonate (PFOS)

Many studies focus on biota such as fish and birds and only limited information about levels of PFCs in food seem to be available. In 2001 the Centre Analytical Laboratory performed a study for the 3M-company as part of a Multi-City Study. PFCs were found in a limited number of samples. PFOS was found in five samples, four whole milk samples and a ground beef sample in concentrations up to 0.85 ng/g. PFOA was found in seven samples, two ground beef samples, two bread samples, two apple samples and one green been sample in concentrations up to 2.35 ng/g (Centre Analytical Laboratory, 2001). The results for PFCs are given in table 3 at the end of this section. PFOS and PFOSA are found in only one of the five samples that were analysed. The concentrations found in the sample of pickled herring, are 1.3 ng/g for PFOS and 0.2 ng/g for PFOSA. PFOS is widely detected in the environment, animals and humans and therefore expected. Surprisingly, Schecter et al. did not find PFOS, but do find PFOA in 50% of the analysed samples in concentrations ranging from 0.02 to 1.8 ng/g (Schecter et al., 2010).

#### 3.6 Phthalates

Phthalates are one of the most ubiquitous classes of chemical contaminants in our everyday environment as a consequence of their high volume uses in open applications. They are used as plasticizers to increase the flexibility of high molecular weight polymers (mainly in PVC), as heat-transfer fluids and as carriers, and can be found in ink, paint, adhesives, pesticides, vinyl flooring (Vethaak et al., 2002), but also in cosmetics and personal care products. Consequently, the potential for human exposure is very high. Di-(2-ethylhexyl) phthalate (DEHP) and di-ethyl phthalate (DEP) are two of the most common used plasticizers. DEHP is nowadays gradually replaced by iso-alkyl phthalate mixtures like di-isononyl phthalate (DINP). The chemical structure of DEP and DEHP is shown below.

There is not much information about concentrations of phthalates in food. Most attention has been focused on phthalates in plastic wrapping materials for food products. An older study dating from 1994 deals with the determination of DEHP in milk, cream, butter and cheese (Sharman et al., 1994). DEHP was found in all these products in concentrations ranging from 330 to 980 ng/g. More recent information is available from the UK Food Standard Agency and is concerned with the presence of phthalates in infant formulae (Joint Food Safety and Standards Group, 1998). Seven phthalates including DEHP were determined in 39 samples of infant formulae. In 12 of the 39 samples none of the phthalates were found. In the remaining samples di-butyl phthalate (DBP), benzylbutyl phthalate (BBP) and DEHP were found. DEHP was the most abundant individual phthalate in concentrations ranging from 50 to 440 ng/g product. DBP was found in concentrations up to 90 ng/g and BBP up to 15 ng/g product. Concentrations of other phthalates were less than 10 ng/g. The results for phthalates in this study are presented in table 3 at the end of this section, and graphically in figure 4. 16 of the 19 samples analysed for phthalates did contain one or more of these compounds. In eggs, milk and orange juice no phthalates were detected. DIDP was the only phthalate that was not found in any of the samples. As expected DEHP is the predominant phthalate found in 16 of the 19 samples with concentrations ranging from 20 to 24,000 ng/g. It should be mentioned that the latter concentration is an exception and was found in the sample of olive oil. The neck of the olive oil bottle contains a polymer spout that may be responsible for the high DEHP concentration in the olive oil. Other phthalates that are frequently found (>50% of the samples) are DBP and BBP, be it in lower concentrations than DEHP.

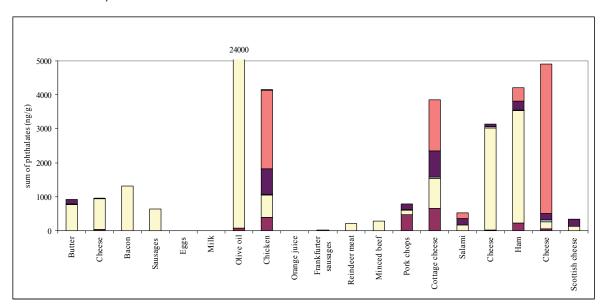


Fig. 4. Graphic presentation of the sum of the eight phthalates in 19 food items

#### 3.7 Alkylphenols

Alkylphenols, but primarily alkylphenol ethoxylates are used as additives in plastics and as surface-active ingredients in industrial detergents and emulsifiers. The ethoxylates are produced by a condensation reaction of alkylphenols with ethylene oxide. Alkylphenols commonly used are nonylphenol (NP) and to a lesser extent octylphenol (OP), in both cases pre-dominantly the para-substituted isomers (>90%). Alkylphenols are the common products of bio- or chemical degradation of the ethoxylates. The chemical structure of n-NP is shown below.

As with the phthalates only little information seems to be available about levels of alkylphenols in food. Guenther et al. determined NPs in 60 different commercially available foodstuffs and concluded that NPs are ubiquitous in food (Guenther et al., 2002). The concentrations of NPs (sum of the isomers) varied between 0.1 and 19.4 ng/g product and were found in all samples. Despite the lipophilic properties of NPs, high concentrations of NPs were not only found in fatty foods but also in non-fatty food products. In another study OP and NP were determined in composite foods (Fernandes et al., 2003). OP was found in only one sample in a concentration of 8.7 ng/g while NP was found in concentrations up to 25 ng/g. In a previous TNO study alkylphenols were determined in wrapped fresh meat and cheese products (Geenen, 2003). Since the alkylphenols were determined in slices of the product collected directly below the foil or wrapper, the results are not representative for the entire product. OP was detected in none of the samples while NPs were found in five of the eight sub-samples in concentrations ranging from 9 to 590 ng/g. For one sample the whole food item was analysed resulting in a much lower concentrations in the order of 1 ng/g for NP. The results for alkylphenols in this study are given in table 3 at the end of this section. NP was found in 2 of the 19 samples, the samples of butter and bacon in concentrations around 5 ng/g. OP was found in none of the samples. Although the concentrations are in the range of what Guenther found, the results are different because the frequency of detection in this study is far lower. Perhaps this is a result of the way sub-samples are collected since higher concentration may be found in top-layers beneath the packaging foil.

#### 3.8 Musk compounds

In nature, musk is a compound produced by a gland in male deer which has been used in perfumes, but the increasing demand resulted in the production of artificial musk compounds. The most well-known are nitro musks like MX and MK that are nowadays replaced by polycyclic musks like AHTN and HHCB. Musks are used as additives for perfumes, in detergents and soaps, in body lotions and deodorizers. The structure of MK and HHCB is presented below.

As far as we know there are no reports or studies concerning the presence of artificial musks in food or food products. However, since 1981 it is known that artificial musks can be found in fish (Yamagishi et al., 1981, 1983; Rimkus & Wolf, 1995; Fromme et al., 2001; Gatermann et al., 2002), and as a results artificial musks may be present in fish products. The results for musks in this study are given in table 3 at the end of this section. The nitro-musks MK and MX are not found in the four fish products analysed for artificial musks. The polycyclic musks AHTN and HHCB are found in two of the samples, the samples tuna and pickled herring in a maximum concentration of 0.56 ng/g for the latter sample. As in other environmental matrices and human blood the HHCB concentrations are about twice that of the AHTN concentrations. That the concentrations are lower than those reported for fish in the literature is probably because most literature studies report results for fish in waterways connected to sewer effluents and not for typical marine fish species.

#### 3.9 Organotin compounds

The main OTCs to be found in food are likely to be tri-substituted compounds, tributyltin (TBT) and triphenyltin (TPT), which have been used extensively as biocides in wood preservatives, in antifouling paints for boats and as pesticides. Mono- and di-substituted OTC's (dibutyltin, mono-n-octyltin and di-n-octyltin) are used as stabilizers in PVC plastics, and di-alkyltins have been approved as PVC stabilizers for food contact materials. OTC's tend to accumulate in fish and other aquatic organisms and tri-alkyltins are bio-degraded to di- and mono-alkyltin compounds and therefore these may be found also in addition to the tri-substituted OTCs. The structures of TBT and TPT are presented below.

triphenyltin (TPT)

Based on an EU SCOOP report the European Food Safety Authority estimated that the median concentrations of TBT, DBT and TPT in fish and fishery products are 7.0, 2.5 and 4.0 ng/g product (EC, 2003; EFSA, 2004). The EU SCOOP report contains very few data on DOT, which were always below the limit of detection. The results for organotin in this study are given in table 3 at the end of this section. Organotin were found in three of the four samples that were analysed. The highest concentration of 9.0 ng/g was found for mono-butyl tin (MBT), a degradation product of TBT in the sample of tuna. Di-butyl tin (DBT) and TBT were also found in this sample. The pickled herring and the fish fingers contained butyl-tin as well as octyl-tin compounds.

# 3.10 Result tables of the concentrations of determined parameters in 25 samples of food

Tables 2 and 3 contain the full results of the study. Note that not all parameters are measured in all samples. In those cases the positions in the table are left blank. If concentrations of the parameters in a sample were below the detection limit, this is indicated with a "<" sign. The limits of detection for each parameter are printed directly after the compound name of the parameter. All results are expressed in ng/g product.

compound	limit of detection	cheese 1	cheese 2	cheese 3	scottisch cheese	cottage cheese	butter	eggs	milk	bacon	sausages	chicken breast	frankfurthers	reindeer beef
	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g
Organochlorin	-													
а-НСН	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
β-НСН	0.1	<	<	<	<	<	</td <td>&lt;</td> <td>\ \&lt;_</td> <td>&lt;</td> <td>&lt;</td> <td>&lt;</td> <td>&lt;</td> <td>&lt;</td>	<	\ \<_	<	<	<	<	<
ү-НСН НСВ	0.1	0.12	0.22	0.26	0.18	0.10	0.34	<	<	< <	010	_<	0.10	0.83
α-chlordane	0.1	< 0.12	< _	0.26	0.16	< 0.10	< 0.54	< /	<	<	0.10	<	< 0.10	< 0.83
β-chlordane	0.1	3	\ \ \ \ \			<	<	<	<	~	\ \	~	<	<
o,p'-DDE	0.1	) <	<	<	<	<	<	<	<	<	<	<	<	<
p,p'-DDE	0.1	0.18	0.48	1.6	0.43	1.3	0.79	<	<	<	0.25	<	<	<
o,p'-DDD	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
p,p'-DDD	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
o,p'-DDT	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
p,p'-DDT	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
Polychlorinate	d biphenyls													
PCB 18	0.1	0.13	0.15	0.22	0.16	0.24	0.25	<	0.12	0.19	0.16	0.20	0.26	0.14
PCB 28/31	0.1	0.31	0.22	0.37	0.36	0.49	0.56	0.16	0.36	0.32	0.35	0.40	0.44	0.26
PCB 22	0.1	<	<	0.11	<	0.14	<	<	<	<	<	<	<	<
PCB 41/64	0.1	<	<	<	<	<	0.27	<	<	<	<	<	<	<
PCB 44	0.1	<	<	<	0.18	<	0.27	<	<	0.18	0.24	<	0.18	0.14
PCB 49	0.1	<	<	<	<	0.14	0.18	<	<	0.19	0.13	0.13	0.16	0.13
PCB 52	0.1	0.16	<	<	0.15	0.29	0.40	<	0.21	0.21	0.16	0.17	0.27	0.15
PCB 54	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 56/60 PCB 70	0.1 0.1	< <	<	<	< 0.12	< 0.16	< 0.20	<	<	< 0.21	< 0.12	<	< 0.17	< 0.11
PCB 74	0.1	<	<	<	< 0.12	0.16	0.20	<	<	0.21	0.12	<	< 0.17	0.11 0.12
PCB 87	0.1	<	<	<	<	< 0.14	< 0.22	<	<	0.11 <	<	<	<	< 0.12
PCB 90/101	0.1	<	<	<	<	<	<	<	<	<	0.11	<	<	<
PCB 99	0.1	<	<	<	<	<	0.14	<	<	<	<	<	<	<
PCB 104	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 105	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 110	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 114	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 118	0.1	<	<	<	<	<	0.23	<	<	<	<	<	<	<
PCB 123	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 138	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 141	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 149	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 151	0.1	<	<	<	<	<	<	<	\ \<_	<b>*</b>	<	<	_	<
PCB 153/168	0.1	<	0.22	0.31	0.16	<	0.29	<	<	<	0.12	<	0.14	0.11
PCB 156	0.1	<	<		<	<	<	<	<	<	<	<	<	<
PCB 157	0.1	4	<	0.12	< 0.10	<	<	<	<	</td <td>/ &lt; /</td> <td>&lt;</td> <td>&lt;</td> <td>&lt;</td>	/ < /	<	<	<
PCB 158	0.1	<	<	0.12	0.10	<	<	<	<	<	<	<	_ <	<
PCB 167	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 170 PCB 177	0.2 0.2	< <	<	<	<	<	<	<	<	< <	<	<	<	<
PCB 177 PCB 180	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 180 PCB 183	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 187	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 188	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 189	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 194	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 199	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
PCB 203	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<

Table 2. Results for organochlorine pesticides and polychlorobipenyls expressed in ng/g product.

p	limit of detection	eef	sd			ırs			pickled hering		ice	ead		
punodwoo	t of ¢	minced beef	pork chops	jĘ.		fish fingers	non		led h	<u>\$</u>	orange juice	brown bread	e oil	
com	limi	mine	pork	salami	ham	fish	salmon	tuna	pick	honey	oran	brov	olive oil	
	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	
Organochlorii	-													
а-НСН	0.1	<	<	<	<	<	<	<	<	<	<	<	<	
β-НСН	0.1	<	<	<	<	<	<	<	\ \ \	<	<	<	<b>~</b>	
ү-НСН	0.1	< 0.14	<	< 0.10	< 0.10	<	< 0.22	<	<	<	< 4	~	0.10	
HCB α-chlordane	0.1 0.1	0.14	<	0.10	0.10	< <	0.22 0.13	< <	0.7 0.2	< <	<	<	0.10	
β-chlordane	0.1	C\		7 🕻 🔪	7	<	0.13		<		<		<	
o,p'-DDE	0.1	Ų.	~	<	<			~	<	~	0.65	<		
p,p'-DDE	0.1	0.33	0.17	0.21	0.17	<	0.83	<	5.6	<	1.5	<	0.40	
o,p'-DDD	0.1	<	<	<	<	<	<	<	<	<	<	<	<	
p,p'-DDD	0.1	<	<	<	<	<	<	<	<	<	<	<	<	
o,p'-DDT	0.1	<	<	<	<	<	<	<	<	<	<	<	<	
p,p'-DDT	0.1	<	<	<	<	<	<	<	<	<	<	<	<	
Polychlorinate	ed biphenyls													
PCB 18	0.1	0.14	0.16	0.22	0.24	0.45	0.19	0.20	0.23	0.38		0.29	<	
PCB 28/31	0.1	0.36	0.23	0.59	0.43	0.63	0.49	0.38	0.76	0.55		0.61	0.19	
PCB 22	0.1	<	<	0.14	<	0.13	0.11	<	0.13	0.11		0.16	<	
PCB 41/64	0.1	<	<	0.14	<	<	<	<	0.38	<		<	<	
PCB 44	0.1	0.24	<	0.29	0.17	0.22	0.24	0.19	0.33	0.19		0.21	<	
PCB 49	0.1	<	0.12	0.19	0.11	0.21	0.23	<	0.22	0.15		0.19	<	
PCB 52	0.1	0.25	0.20	0.28	0.24	0.27	0.36	<	0.74	0.19		0.35	<	
PCB 54	0.1	<	<	<	<	<	<	<	<	<		<	<	
PCB 56/60	0.1	<	<	<	<	<	0.18	<	0.31	<		0.14	<	
PCB 70	0.1	0.13	<	<	<	0.14	0.38	<	0.71	0.16		0.17	<	
PCB 74	0.1	<	<	<	<	<	0.19	<	0.37	<		0.11	<	
PCB 87	0.1	<	<	<	<	<	0.12	<	0.41	<		<	<	
PCB 90/101	0.1	<	<	0.13	<	<	0.47	0.14	2.4	<		0.15	<	
PCB 99	0.1	<	<	<	<	<	0.24	<	0.95	<		<	<	
PCB 104	0.1	<	<	<	<	<	< 0.14	<	<	<		<	<	
PCB 105 PCB 110	0.1 0.1	< <	< <	< <	< <	< <	0.14 0.30	< 0.14	0.59 1.6	< <		< <	< <	
PCB 110 PCB 114	0.1	<	<	<	<	<	< 0.30	< 0.14	1.6	<		<	<	
PCB 114 PCB 118	0.1	0.11	<	<	<	<	0.40	0.11	1.8	<		<	<	
PCB 118 PCB 123	0.1	0.11	<	<	<	<	0.40 <	0.11 <	0.27	<		<	<	
PCB 138	0.1	<	<	0.10	<	<	0.24	0.20	1.5	<		<	<	
PCB 141	0.1	<	<	<	<	<	<	<	0.30	<		<	<	
PCB 149	0.1	<	<	<	<	_<	0.49	<	2.8	<		<	<	
PCB 151	0.1		<	<	<	<	0.17	<	0.92	<		<	<	
PCB 153/168	0.1	0.26	~	0.13	<	<	1.0	0.12	5.5	~		0.13	<	
PCB 156	0.1	<	<	<	<	<	<	<	0.22	<		<	<	
PCB 157	0.1	<	<	<	<	<	<	<	<	<		<	<	
PCB 158	0.1	<	<	7 < \	<	<	0.50	0.16	3.2	<		~	<	
PCB 167	0.1	<	_	<	_ <	<	<	<	0.19	<		_ <	<	
PCB 170	0.2	<	<	<	<	<	<	<	0.56	<		<	<	
PCB 177	0.2	<	<	<	<	<	<	<	0.56	<		<	<	
PCB 180	0.2	<	<	<	<	<	0.32	<	0.90	<		<	<	
PCB 183	0.2	<	<	<	<	<	<	<	0.43	<		<	<	
PCB 187	0.2	<	<	<	<	<	<	<	1.6	<		<	<	
PCB 188	0.2	<	<	<	<	<	<	<	<	<		<	<	
PCB 189	0.2	<	<	<	<	<	<	<	<	<		<	<	
PCB 194	0.2	<	<	<	<	<	<	<	<	<		<	<	
PCB 199	0.2	<	<	<	<	<	<	<	<	<		<	<	
PCB 203	0.2	<	<	<	<	<	<	<	<	<		<	<	

Table 2. (continued) Results for organochlorine pesticides and polychlorobipenyls expressed in ng/g product. Empty spaces, as PCBs in orange juice, indicate that the parameter was not determined in this sample.

punoduoo	limit of detection	cheese 1	cheese 2	cheese 3	scottisch cheese	cottage cheese	butter	eggs	milk	bacon	sausages	chicken breast	frankfurthers	reindeer beef
	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g
Brominated flam														
BDE-17	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-28	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-32 BDE-35	0.05 0.05	<	0.07	<	0.06	<	< <	< <	< <	0.06	<	0.08	0.06	0.06
BDE-37	0.05	~	<	<	<	~	<	<	<	~	<	<	~	
BDE-47	0.05	0.75	0.3	0.43	0.82	0.29	<	<	<	0.27	0.41	0.26	0.33	0.26
BDE-49-71	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-66	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-75	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-77	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-85	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-99	0.05	0.10	<	<	0.15	<	0.15	<	<	<	<	<	0.05	0.06
BDE-100	0.05	<	<	<	0.12	<	<	<	<	<	<	<	<	<
BDE-119	0.05	<	<	<	<	<	< 0.21	<	<	<	<	<	<	<
BDE-126 BDE-138	0.05 0.05	< <	<	<	< <	< <	0.21	<	<	< <	< <	< <	< <	<
BDE-138 BDE-153	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-153	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-156	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-166	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-181	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-183	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-184	0.05	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-190	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-191	0.1	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-196	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-197	0.2	<	<	<	<	<	<	<	<	<	<	<	<	<
BDE-206	1	< <	<	<	< <	<	<	<	< <	<	<	<	<	<
BDE-207 BDE-209	1 5	<	< <	< <	<	< <	< <	< <	<	< <	< <	< <	< <	<
Perfluorinated ch	-													
PFOA	0.2													
PFNA	0.2													
PFDA	0.2													
PFUnA	0.2													
PFDoA	0.2													
PFTrA	0.2													
PFOS	0.2													
PFOSA	0.2													
Phthalates	_													
DMP	1	<	<	<	< <	<	<	< <	< <	< <	<	4.9	<	<
DEP DIBP	1 10	< <	< <	4400	<	< 1500	5.6 <	< <	<	<	< <	24 2300	< <	<
DBP	10	<	76	190	200	780	132	<	<	<	<	760	<	<
BBP	10	21	32	50	9.9	25	17	<	<	2	<	11	<	<
DEHP	20	910	3000	210	130	890	770	<	<	1320	640	670	20	210
DINP	20	31	26	59	<	660	<	<	<	<	<	390	<	<
DIDP	20	<	<	<	<	<	<	<	<	<	<	<	<	<
Alkylphenols														
NP	2	<	<	<	_<	<	5.1	<	<	5.5	<	<	<	<
OP	2	<	<	<	<	<	<	<	<	<	<	<	<	<
Artificial musks														
AHTN	0.1													
HHCB	0.1													
MK MY	0.1													
MX Organotin compo	0.1													
MBT	0.2													
DBT	0.2													
TBT	0.2													
MOT	0.2													
MOI														

Table 3. Results for brominated flame retardants, perfluorinated chemicals, phthalates, alkylphenols, artificial musks and organotin compounds expressed in ng/g product. Empty spaces, as for the artificial musks, indicate that the parameter was not determined in this sample.

compound	limit of detection	minced beef	pork chops	salami	ham	fish fingers	salmon	tuna	pickled hering	honey	orange juice	brown bread	olive oil	
	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	
Brominated flame BDE-17	0.1	<	<	<	<	<	<	<	<	<		<	<	
BDE-28	0.1	<	<	<	<	<	<	<	<	<		<	<	
BDE-32	0.05	0.07	0.05	0.06	0.09	<	<	<	0.08	0.15		<	<	
BDE-35	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-37	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-47 BDE-49-71	0.05	0.78	0.52	0.81	0.78	<	< <	0.39	0.35	<		0.55	<	
BDE-49-71 BDE-66	0.05 0.05	< <	<	< <	< <	< <	<	< <	< <	< <		/\ <b>&lt;</b> \	< /	
BDE-75	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-77	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-85	0.05	<	0.32	<	<	<	<	<	<	<		<	<	
BDE-99	0.05	0.12	<	<	0.10	<	<	<	<	<		0.19	<	
BDE-100 BDE-119	0.05	0.11	<	<	<	<	<	<	<	<		<	<	
BDE-119 BDE-126	0.05 0.05	0.08	<	< <	<	< <	< <	< <	<	<		< <	< <	
BDE-126 BDE-138	0.05	< 0.08	<	<	<	<	<	<	<	<		<	<	
BDE-153	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-154	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-156	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-166 BDE-181	0.05 0.05	0.18	<	<	< <	< <	< <	< <	< 0.10	< <		<	< <	
BDE-181 BDE-183	0.05	0.18	<	<	<	<	<	<	0.10 <	<		<	<	
BDE-183	0.05	<	<	<	<	<	<	<	<	<		<	<	
BDE-190	0.1	<	<	<	<	<	<	<	<	<		<	<	
BDE-191	0.1	<	<	<	<	<	<	<	<	<		<	<	
BDE-196	0.2	<	<	<	<	<	<	<	<	<		<	<	
BDE-197 BDE-206	0.2	<	<	< <	<	<	<	<	<	<		<	<	
BDE-206 BDE-207	1 1	< <	<	<	< <	< <	<	< <	<	< <		<	< <	
BDE-209	5	<	<	<	<	<	<	<	<	<		<	<	
Perfluorinated ch	emicals													
PFOA	0.2					<	<	<	<			<		
PFNA	0.2					<	<	<	<			<		
PFDA PFUnA	0.2					<	<	<	<			<		
PFUnA PFDoA	0.2					< <	<	< <	<			<		
PFTrA	0.2					<	<	<	<			<		
PFOS	0.2					<	<	<	1.3			<		
PFOSA	0.2					<	<	<	0.2			<		
Phthalates														
DMP DEP	1 1	< <	< <	< <	1.1						< <		< <	
DIBP	10	<	<	165	400						<		<	
DBP	10	<	170	183	260						<		<	
BBP	1	<	13	12	17						<		340	
DEHP	20	290	140	160	3300						<		24000	
DINP DIDP	20	< <	470	< <	230						<		72	
Alkylphenols	20		7 //											
NP	2	<	<	<	<						<		<	
OP	2	<	<	<	<						<		<	
Artificial musks														
AHTN	0.1					<	<	0.18	0.29					
HHCB MK	0.1 0.1					< <	< <	0.27	0.56					
MK MX	0.1					<	<	<	<					
Organotin compo														
MBT	0.2					0.5	<	9	<					
DBT	0.2					<	<	1.1	0.6					
TBT	0.2					<	<	0.2	0.8					
MOT	0.2 0.2					0.3	<	<	0.8 1.2					
DOT	0.∠					<	<	<	1.2					

Table 3. (continued) Results for brominated flame retardants, perfluorinated chemicals, phthalates, alkylphenols, artificial musks and organotin compounds expressed in ng/g product. Empty spaces, as for the brominated flame retardants in orange juice, indicate that the parameter was not determined in this sample.

#### 4. Conclusions

In this study the concentrations of a number of typical man-made chemicals in food or food products were determined. The compound groups of interest were organochlorine pesticides, polychlorinated biphenyls, brominated flame retardants, alkylphenols, artificial musks, perfluorinated compounds and organitin compounds. The results show that many of these compounds are present food in the range of 0.1 to 10 ng/g with the exception of phthalates for which the typical concentrations are two orders of magnitude higher. Organochlorine pesticides were found in the 17 of the 25 samples. The main organochlorine pesticides found in food are p,p'-DDE, a metabolite of DDT, and HCB in concentrations up to 5.6 ng/g. Polychlorinated biphenyls were found in all samples with predominance for PCB-18, -28 and -52. The sum of the indicator-PCBs ranged from 0.16 to 13 ng/g and total PCBs up to 32 ng/g. The highest concentrations were found in fish. Brominated flame retardants were found in 19 of the 24 samples with predominance for the tetra- and penta-PBDEs, especially BDE-47, -32 and -99. Total PBDE concentrations ranged from 0.15 to 1.2 ng/g with the highest concentration found in meat and not in fish as in other studies. BDE-209, HBCD and TBBPA were not found in any of the samples. The prefluorinated compounds PFOS and PFOSA were found in one of the four samples analysed, a fish sample, in concentrations of 1.3 and 0.2 ng/g. The predominant phthalates in food were DEHP, DBP and BBP. Phthalates were found in 12 of the 19 samples. DEHP concentrations ranged from 20 to 24,000 ng/g, the latter for a sample of olive oil, with a median concentration of 640 ng/g. Median concentrations for DBP and BBP were 200 and 17 ng/g. Alkylphenols were detected in 2 of the 19 samples, in both cases nonylphenol in concentrations around 5 ng/g. Of the artificial musks the polycyclic musks HHCB and AHTN were found 2 of the 4 samples in concentrations up to 0.56 ng/g for HHCB. As in other matrices the AHTN concentrations are about half those of HHCB. Organotin compounds were found in three of the five samples. Apart from TBT and its metabolites DBT and MBT, two samples also contained octyltin compounds.

### 5. Acknowledgement

We kindly acknowledge the support of the WWF in the execution of this study.

# 6. References

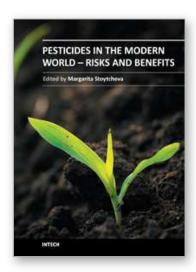
- 3M-company (1999). Fluorochemical Use, Distribution and Release Overview. EPA docket AR226-0550 (1999).
- Bocio, A.; Llobet, J.M.; Domingo, J.L.; Corbella, J.; Teixido, A.; Casa, C. (2003). Human exposure to polybrominated diphenyl ethers through th diet in Catalonia, Spain. *Journal of Agricultural and Food Chemistry*, Vol.51, (2003), pp. 3191-3195.
- CDC report (2001). National Report on Human Exposure to Environmental Chemicals. CDC, Atlanta, Georgia, (March 2001).
- CDC report (2003). National Report on Human Exposure to Environmental Chemicals. CDC, Atlanta, Georgia, (January 2003).
- Centre Analytical Laboratory (2001). Analysis of PFOS, PFOSA and PFOA from various food matrices using HPLC electrospray/mass spectrometry. Study number 023-057, (June 2001).

- Cox, S.; Niskar, A.S.; Narayan K.M.V.; Marcus, M. (2007). Prevalence of self-reported diabetes and exposure to organochlorine pesticides among Mexican Americans: Hipanic Health and Nutrition Examination Survey, 1982-1984. Environmental Health Perspective, Vol.115, (2007), pp. 1747-1752.
- De Winter-Sorkina, R.; Bakker, M.I.; van Donkersgoed, G.; van Klaveren, J.D. (2003). RIVM report 310305001, Dietary intake of brominated flame retardants by the Dutch population, (2003).
- De Winter-Sorkina, R.; Bakker, M.I.; Wolterink, G.; Zeijlmaker, M.J. (2006). RIVM report 320100002, Brominated flame retardants: occurrence, dietary intake and risk assessment, (2006).
- EC (2003). European Commission report. Report on Tasks for Scientific Cooperation (SCOOP), task 3.2.13. Assessment of the dietary exposure to organotin compounds of the population of the EU member states. European Commission, Directorate-General Health and Consumer Protection, Reports on tasks for scientific cooperation, (October 2003).
- EFSA (2004). European Food Safety Authority report. Opinion of the Scientific Panel on Contaminants in the Food Chain on a request from the Commission to assess the health risks to consumers associated with the exposure to organotins in foodstuffs. *The EFSA Journal*, Vol.102, (2004), pp. 1-119.
- EU (2004). EU-report: Dioxins & PCBs: Environmental levels and human exposure in candidate countries. Reference: ENV.C.2/SER/2002/0085, (June 2004).
- Fernandes, A.R.; Costley, C.T.; Rose, M. (2003). Determination of 4-octylphenol and 4-nonylphenol congeners in composite foods. *Food Additives and Contaminants*, Vol.20, (2003), pp. 846-852.
- FDA (1999). Food and Drug Administration Total Diet Study 1999. See internet site, http://vm.cfsan.fda.gov/~acrobat/pes-99rep.pdf.
- FDA (2003). Food and Drug Administration Total Diet Study 2003. See internet site, http://www.cfsan.fda.gov/~comm/tds-toc.html
- Fromme, H.; Otto, T.; Pilz, K. (2001). Polycyclic musk fragrances in fish samples from Berlin waterways, Germany. *Food Additives and Contaminants*, Vol.11, (2001), pp. 937-944.
- Gallani, B.; Boix, A.; Di Domenico, A.; Fanelli, R. Occurence of NDL-PCB in food and feed in Europe. *Organohalogen Compounds*, Vol.66, (2004), pp. 3561-3569.
- Gatermann, R.; Biselli, S.; Huhnerfuss, H.; Rimkus, G.G.; Hecker, M.; Karbe, L. (2002) Part 1: Species-dependent bioaccumulation of polycyclic and nitro musk fragrances in freshwater fish and mussels. *Archives of Environmental Contamination and Toxicology*. Vol.42, (2002), pp. 437-446.
- Geenen R. (2003). TNO report TR 03/725, The determination of additives in food products, (December, 2003).
- Guenther, K.; Heinke, V.; Thiele, B.; Kleist, E.; Prast, H.; Raecker, T. Endocrine disrupting nonylphenols are ubiquitous in food. *Environmental Science and Technology*, Vol.36, (2002), pp. 1676-1680.
- Guo, Y.L.; Lambert, G.H.; Hsu, C.C. (1995). Growth abnormalities in the population exposed in utero and early postnatally to polychlorinated biphenyls and dibenzofurans. *Environmental Health Perspective*, Vol.103, (1995), pp. 117-122.
- Joint Food Safety and Standards Group (1998). Joint Food Safety and Standards Group, Food surveillance information sheet, number 168, (December, 1998). See internet site,

- http://archive.food.gov.uk/maff/archive/food/infsheet/1998/no168/168phtha.h
- Jurewicz, J.; Hanke, W. (2008). Prenatal and childhood exposure to pesticides and neurobehavioral development: review of epidemiological studies. *International Journal of Occupational Medical and Environmental Health*, Vol.21, (2008), pp. 121-132.
- Kannan, K.; Corsolini, S.; Falandysz, J.; Oehme, G.; Focardi, S.; Giesy, J.P. (2002). Perfluorooctane sulfonate and related fluorochemicals in marine mammals, fish and birds from the Baltic and Mediterranean Sea. *Environmental Science and Technology*, Vol.36, (2002), pp. 3210-3216.
- Kannan, K.; Corsolini, S.; Falandysz, J.; Fillman, G.; Kumar, K.S.; Loganathan, B.G.; Mohd, M.A.; Olivero, J.; Van Wouwe, N.; Yang, J.H.; Aldous, K.M. (2004). Perfluorooctanesulfonate and related fluorochenmicals in in human blood from several countries. *Environmental Science and Technology*, Vol.38, (2004), pp. 4489-4495.
- Lee, D.H.; Steffes, M.; Jacobs, D.R. Jr. (2007). Positive associations of serum concentration of polychlorinated biphenyls and organochlorine pesticides with self-reported arthritis, especially rheumatoid type, in women. *Environmental Health Perspective*, Vol.115, (2007), pp. 883-888.
- Martin, J.W.; Mabury, S.A.; Solomon, K.R.; Muir, D.C.G. Bioconcentration and tissue distribution of perfluorinated acids in rainbow trout (Oncorhynchus mykiss). *Environmental Toxicology and Chemistry*, Vol.22, (2003), pp. 196-204.
- Martin, J.W.; Whittle, D.M.; Muir, D.C.G.; Mabury, S.A. Perfluoroalkyl contaminants in a foodweb from Lake Ontario. *Environmental Science and Technology*, Vol.38, (2004), pp. 5379-5385.
- Ohta, S.; Ishizuka, D.; Nishimura, H.; Teruyuki, N.; Aozasa, O.; Shimidzu, Y.; Ochiai, F.; Kida, T.; Nishi, M.; Miyata, H. (2002). Comparison of dioehyl ethers fish, vegetables, and meats and levels in human milk of nusing women in Japan. *Chemosphere*, Vol.46, (2002), pp. 689-696.
- Ohta, S.; Tokusawa, H.; Nakao, T.; Aozasa, O.; Myata, H.; Alaee, M. (2008). Global contamination of co-planar polybrominated/chlorinated biphenyls (Co-PXBs) in the market fishes from Japan. *Chemosphere*, Vol.73, (2008), pp. S31-S38.
- Peters, R.J.B. (2005). TNO report B&O-A R 2005/129, Man-made chemicals in maternal and cord blood, (March 2005).
- Peters, R.J.B.; Beeltje, H.; van Delft, R.J. (2008). Xeno-estrogenic compounds in precipitation. *Journal of Environmental Monitoring*, Vol.10, (2008), pp. 760-769.
- Rimkus, G.G.; Wolf, M. (1995). Nitro musk fragrances in biota from freshwater and marine environment. *Chemosphere*, Vol.30, (1995), pp. 641-651.
- Rusiecki, J.A.; Baccarelli, A.; Bollati, V.; Tarantini, L.; Moore, L.E.; Bonefeld-Jorgensen, E.C. (2008). Global DNA hypomethylation is associated with high serum-persistent organic pollutants in Greenlandic Inuit. *Environmental Health Perspective*, Vol.116, (2008), pp. 1547-1552.
- Ribas-Fito, N.; Sala, M.; Kogevinas, M.; Sunyer, J. (2001). Polychlorinated biphenyl (PCBs) and neurological development in children: a systematic review. *Journal of Epidemiological Community and Health*, Vol.55, (2001), pp. 537-546.

- Sharman, M.; Read, W.A.; Castle, L.; Gilbert, J. (1994). Levels of di-(2-ethylhexyl)phthalate and total phthalate esters in milk, cream, butter and cheese. *Food Additives and Contaminants*, Vol.11, (1994), pp. 375-385.
- Schecter, A.; Papke, O.; Tung, K.C.; Staskal, D.; Birnbaum, L. (2004). Polybrominated diphenyl ethers contamination of United States food. *Environmental Science and Technology*, Vol.38, (2004), pp. 5306-5311.
- Schecter, A.; Colacino, J.; Haffner, D.; Patel, K.; Opel, M.; Päpke, O.; Birnbaum, L. (2010). Perfluorinated compounds, polychlorinated biphenyls, and organochlorine pesticide contamination in composite food samples from Dallas, Texas USA. *Environmental Health Perspective*, Vol.118, (2010), pp. 796-802.
- Vethaak, A.D.; Rijs, G.B.J.; Schrap, S.M.; Ruiter, H.; Gerritsen, A.; Lahr, J. (2002). In: *Estrogens and xeno-estrogens in the aquatic environment of the Netherlands*. RIZA/RIKZ-report no. 2002.001, (February 2002).
- Yamagishi, T.; Miyazaki, T.; Horii, S.; Akiyama, K. (1983). Synthetic musk residues in biota and water from Tama River and Tokyo Bay (Japan). *Archives of Environmental Contaminants and Toxicology*, Vol.12, (1983), pp. 83-89.
- Yamagishi, T.; Miyazaki, T.; Horii, S.; Kaneko, S. (1981). Identification of musk xylene and musk ketone in freshwater fish collected from the Tama River, Tokyo. *Bulletin of Environmental Contaminants and Toxicology*, Vol.26, (1981), pp. 656-662.





### Pesticides in the Modern World - Risks and Benefits

Edited by Dr. Margarita Stoytcheva

ISBN 978-953-307-458-0
Hard cover, 560 pages
Publisher InTech
Published online 03, October, 2011
Published in print edition October, 2011

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