SURVEY OF TOXAPHENE CONCENTRATIONS IN FISH FROM EUROPEAN WATERS

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Introduction

Toxaphene, a suspected carcinogen¹ is a broad spectrum chlorinated pesticide consisting of a complex mixture of chlorinated bornanes (CHBs)² which has been produced in comparable amounts to PCBs³. The objective of this study was to provide information on the toxicological risks posed by toxaphene to the consumer of fish from European waters. The levels of 3 toxaphene congeners in various fish species from different geographical locations were determined. These data were then used to provide information on the exposure of toxaphene to the consumer of fish.

Methods and Materials

221 samples were collected, covering 23 different species, from European waters (Fig.1). Where possible, samples were collected in accordance with the JAMP Guidelines⁴. Each laboratory used their own analysis method, 3 used GC-ECD and 1 GC/NCI-MS. CPSIL 8 and 19 columns were used in all cases. Tissue extraction techniques used were Soxhlet and blender solvent extraction. Clean-up methods included GPC, HPLC, sulphuric acid and alumina. All laboratories used silica gel for the separation of the different groups of chlorinated compounds. The comparability of these methods was assessed through a series of intercomparison exercises. In addition, herring oil was analysed as a reference material with each batch of samples to assess the within and between laboratory variation. Based on the results of these studies CHBs 26, 50 and 62 were selected for analyses. In order to maximise the information obtained from the baseline survey 3 sampling strategies were incorporated into the survey with a view to measuring the:

- 1. variation in toxaphene concentration in tissues of similar individuals of the same species.
- 2. geographical distribution of toxaphene in marine fish species expressed on a lipid basis.
- 3. human exposure to toxaphene from fish consumption.

Results and discussion

Variation study on individual fish: Table 1 shows the summarised results of the variation study. The ranges and median for Barents Sea golden redfish are higher that the other three groups. An ANOVA shows no significant difference (p>0.05) in the concentration of the Σ 3CHBs in the lipids of cod liver, herring and mackerel, but the concentration in the golden redfish is significantly higher than the other three species. The Σ 3CHBs measured in the variation study suggested higher

levels, on a lipid basis, in fish from the Barents Sea than from the other three regions. The relative contribution of individual CHB congeners to the Σ 3CHB congeners varied considerably within 3 of the groups but not in the golden red fish from the Barents Sea. It is unclear if this is due to analytical uncertainties or is a true reflection of the variances within the populations.

| | Mean | CV | Range | Interquarile |
|---|-------|------|--------------|--------------|
| Mackerel (n=24) Atlantic west of Ireland – Region D | | | | |
| Σ3CHBs | 19.4 | 52.6 | 5.7 -40.2 | 11.1 - 24.4 |
| Length | 255 | 14.3 | 210 - 310 | 219-290 |
| Lipids | 9.5 | 63.2 | 2.2-22.5 | 4.6 - 15.3 |
| Golden redfish (n=24) - Barents Sea – Region B | | | | |
| Σ3CHBs | 76.2 | 40.3 | 37.0 - 181.0 | 58.0 - 85.2 |
| Length | 288.5 | 9.6 | 230 - 350 | 280 - 310 |
| Lipids | 2.8 | 42.7 | 1.3 - 6.2 | 1.7 –3.6 |
| Cod liver (n=25) – North Sea – Region E | | | | |
| Σ3CHBs | 20.2 | 44.2 | 12.7 - 45.48 | 16.1 - 28.5 |
| Length | 420.8 | 6.2 | 380 - 490 | 400 - 430 |
| Lipids | 50.5 | 21.2 | 28.3 - 70.7 | 42.6 - 54.2 |
| Herring (n=26) – Baltic Sea – Region F | | | | |
| Σ3CHBs | 22.3 | 76.9 | 8.0 - 88.0 | 11.5 - 23.7 |
| Length | 234.6 | 5.4 | 220 - 260 | 222 - 240 |
| Lipids | 5.7 | 57.7 | 1.0 - 16.0 | 4.0 - 6.7 |

Table 1: The mean, coefficient of variation (CV), range and interquartile range (of the sum of CHB26+50+62) in μ gkg⁻¹ lipid weight, length (mm) and % total lipid.

Geographical distribution: To investigate the geographical distribution of toxaphene in marine fish species, expressed on a lipid basis, the survey area was divided into 6 regions A to F (Fig. 1). Region A was not included in the comparative statistical analysis as no data for CHB26 were available for this region. The box and whisker plot (Fig. 2) shows the concentration ranges of the Σ 3CHBs (Σ 2CHBs for region A) in biota from the different regions. No significant difference (p>0.05) in the concentrations of the Σ 3CHB congeners in the lipids of fish and shellfish from the Barents Sea (region B) and Norwegian Sea (region C) was detected. Both these areas, however, show significantly higher levels (p<0.05) than observed in the Baltic Sea (region F), North Sea (region E) and Irish coastal waters (region D). These data suggest that fish from the Barents and Norwegian Seas have elevated concentrations compared to the other 3 regions. In all regions the highest concentrations of Σ 3CHB are generally measured in lipid rich species.

Human Exposure: Human exposure to toxaphene occurs mainly through the consumption of contaminated fish⁵. In this study, fish with a low lipid content contained levels for the Σ 3CHBs of 0.04-0.52, 0.01-0.90 and 0.02- 0.07 µgkg⁻¹ for cod, plaice and haddock, respectively. In lipid rich fish the ranges were 0.76-14.4, 13.5-18.4 and 0.11-5.90 µgkg⁻¹ for herring, Greenland halibut and mackerel respectively. In farmed salmon and trout the range was 1.83-12.4 µgkg⁻¹ wet weight. In general the concentrations in the baseline survey data set are lower than previously reported⁵.



Figure 1: The locations where samples were collected, the division of the study area into geographical regions A to F. The \blacklozenge denotes the location where the samples for the variation study were collected.



Figure 2: Box and whisker plot of the concentration, on a lipid basis, of the Σ 3CHBs 26, 50 and 62 (Σ 2CHBs, 26 and 50 for region A) in various fish species.

Tolerable Daily Intakes (TDI) are generally based on total toxaphene. To convert the Σ 3CHBs to total toxaphene conversion factors, based on the simultaneous analyses Σ 3CHBs and total toxaphene for 55 samples, were estimated. Conversion factors for marine fish, eel and mussel were

estimated as 12.4, 41.6, and 24.0, respectively. Using these factors the daily intake of total toxaphene, through the consumption of fish, was estimated. Fish consumption for Germany is 12 kg/person/year, for Ireland, Norway and The Netherlands it is 18, 46 and 12 kg/person/year, respectively⁶. Fig.3 shows the estimated daily intake of toxaphene, assuming single species consumption. For an average fish consumer with a body weight of 60 kg the estimated average daily intake of toxaphene for Norway was 1.2 µg, and 0.4, 0.5, and 0.2 µg for consumers of Germany, Ireland, and The Netherlands respectively.



Figure 3: The estimated daily intake of total toxaphene (μ g) from the consumption of various fish species in Germany, Ireland, Norway and The Netherlands. Solid lines indicate average daily intakes in each country. TDI levels from Canada (12 μ g) and the U.S. for chronic toxicity (15 μ g) are also shown.

Only the toxaphene intake from the consumption of 46 kg per year of Greenland halibut would exceed the Canadian TDI of 12 μ g per day for a person of 60 kg. Based on the information obtained in this study it can be concluded that no adverse toxicological effects from the exposure to toxaphene, through the consumption of fish from European waters, are expected.

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References

- 1. Saleh, M.A. (1991). Rev. Environ. Contam. Toxicol. 118, 1.
- 2. Muir, D.C.G. and J.de Boer (1995). Trends Anal. Chem. 14, 56.
- 3. de Boer, J. and P.G. Wester. (1993). Chemosphere 27 1879-1890.

4. JAMP Guidelines for Monitoring Contaminants in Biota (ASMO 97/4/2). OSPAR Commission, London, UK.

- 5. de Geus, H-J. Besselink, H. Brower, A. Klungsoyr, J. Mc Hugh, B. Nixon, E. Rimkus, Wester,
- P. G. and de Boer, J. (1998) Environ. Health Persp. 107 (Suppl. 1), 115.
- 6. Eurostst.http://www.eubusiness.com/fooddrin/980720es.htm