GEOGRAPHICAL INVESTIGATION FOR PCDD/F AND PCB IN FISH COLLECTED FROM UK AND PROXIMATE MARINE WATERS

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Introduction

Marine environments are recognised sinks for a range of environmental contaminants, and uptake and bioaccumulation by various fish and shellfish species has been widely documented. Bivalve shellfish have a recognised potential for bio-accumulating contaminants and some species, such as mussels, are commonly used as early indicators of local pollution. Marine fish and shellfish are known to make a significant contribution to human exposure of a range of environmental contaminants.

In an effort to reduce or prevent inputs that could cause pollution, affect human health or adversely impact legitimate uses of the marine environment, the Marine Strategy Framework Directive[1] encourages collaboration and coordination between individual EU Member States with the aim of protecting and preserving marine ecosystems. In the context of the present study, one of the targets for good environmental status under the Directive is the limiting of contamination in fish and other seafood to achieve compliance with maximum contaminant levels established by European Commission Regulation, or other relevant standards.

Fish species available to consumers in the UK are sourced from a variety of locations but, keeping in mind the context of the current study, the main areas targeted for investigation were the North Sea and the Greater North Sea sub-region extending towards Norway, the Irish Sea and the Celtic Sea sub-regions extending off the North-Western coast of France and the European coastal North Atlantic regions, including Biscay and extending as far south as the Algarve.

The data from previous studies[2] provide a strong indication that oily fish species such as sardines, sprats, sea bass, mackerel and herring, are likely to show the highest levels of contaminants and would therefore represent the greatest likelihood of non-compliance. However, other species such as turbot, halibut, dogfish etc. were also included. These are moderately oily but are eaten in smaller quantities and are less abundant. Fish liver was not included in the investigation as a high rate of non-compliance would be expected (contamination levels are generally very high and the limit was set by the Commission to enable trade in fish liver to continue) and it was considered that this should not be included as an indicator for Good Environmental Status.

Materials and methods

192 samples from UK and proximate marine waters were obtained from Billingsgate Fish Market, London, or collected during routine research trawls (Table 1). Although the primary intention was to target UK waters, occasional samples from other European waters taken at Billingsgate were included to enable additional intra-species comparison and because the samples were in any case destined for the UK food chain. On receipt at the laboratory each sample was given a unique laboratory reference number and the sample details were logged into a database.

Sample preparation mirrored domestic consumption practices. Depending on the species, samples were dissected to collect edible muscle tissue and exclude skin, bones and organs. However for some species such as sprats, whole fishes were used. The selected tissue (or whole fish) was minced and homogenised by blending. The homogenate was freeze-dried and the resulting powders were re-homogenised.

Table 1: Overview of samples

Species	Sardines	Mackerel	Herring	Grey Mullet	Sprats
Sample number	n=16	n=41	n=19	n=26	n=25
General Location	England-South Coast, Northern Brittany, Algarve	Norwegian Coast, Scotland, England-South Coast, Irish Sea, Northern Brittany, Algarve	Scotland, England-South Coast, Irish Sea, Northern France	England-South Coast, East Coast, Welsh Coast, Northern France, Western Mediterranean	Scotland, England-South Coast, East Coast, Irish Sea, Welsh Coast
Species	Sea Bass	Turbot	Dogfish (various spp)	Other spp - Halibut, Haddock, Plaice, Lemon sole, Witch, Megrim, Monkfish	
Sample number	n=25	n=16	n=14	n=10	
General Location	England-South Coast, East Coast, Welsh Coast, Northern France	Scotland, England-South Coast, East Coast, Irish Sea,	England-South Coast, East Coast, Irish Sea,	England, East Coast, NE-Atlantic, Holland	

Results and discussion:
PCDD/Fs and PCBs were detected in all fish samples at concentrations in the range 0.01-2.6 WHO_{2005}-TEQ/kg for dioxins and 0.02-11.2 WHO_{2005}-TEQ/kg for dioxin-like PCBs. The sum of ICES-6 PCBs ranged from 0.1 to 145 µg/kg whole weight. As expected, certain species – notably sea bass, sprats, and sardines - showed a greater tendency to bio-accumulate these contaminants with average total WHO_{2005}-TEQ values of 2.5, 2.0 and 2.0 ng/kg respectively. These mean concentrations are lower than those for composite samples of the same species collected in the UK about a decade earlier with total WHO-TEQ values of 3.1, 3.6 and 4.9 ng/kg for wild sea-bass, sprats and sardines respectively (the original reported results have been recalculated using WHO_{2005} TEFs). The current regulation sets maximum limits for fish and fisheries products at 3.5 pg/g for dioxins and 6.5 pg/g for the sum of dioxins plus dioxin-like PCBs.[3] The corresponding limit for non dioxin-like PCBs is 75 ng/g for the sum of the ICES6 PCBs 28, 52, 101, 138, 153 and 180.

All of the results for fish taken from UK waters were within the limits for dioxins, total TEQ and non dioxin-like PCBs. Two samples, one each of sea bass and mackerel, exceeded the limits for total TEQ with levels of 12.5 and 7.5 ng WHO-TEQ /kg respectively. The sea bass sample also had exceeded the limit for non dioxin-like PCBs with a level of 145 µg/kg Both samples were landed in Boulogne and the results are consistent with the fish having come from the southern part of the Channel, which is known to be especially contaminated in the Seine Bay, where sardine fishing is banned due to high PCB levels.

Conclusions
This study has measured dioxins and PCBs in a number of commonly consumed fish species, taken from marine waters around the UK and from other proximate fishing areas from which retail fish in the UK are commonly sourced.

Table 2 Summary of PCDD/F, PCB WHO-TEQ and ICES-6 PCB concentrations ng/kg WHO-TEQ Whole weight (upper bound)

PCDD/F and PCB	Sardines (n=16)			Mackerel (n=41)			Herring (n=19)			Mullet (n=26)		
	MIN	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**
PCDD/F	0.13	0.40	1.20	0.04	0.43	1.62	0.34	0.64	1.55	0.02	0.14	0.51
non ortho-PCB	0.47	1.48	3.16	0.06	0.90	5.56	0.23	0.56	1.27	0.07	0.47	1.91
Ortho-PCB	0.03	0.09	0.33	0.01	0.07	0.37	0.02	0.05	0.12	0.01	0.06	0.22
Sum WHO-TEQ	**0.63**	**1.97**	**4.37**	**0.10**	**1.40**	**7.51**	**0.64**	**1.24**	**2.78**	**0.11**	**0.67**	**2.36**
Sum ICES-6 PCB, µg/kg	**5.41**	**16.6**	**54.9**	**0.86**	**10.6**	**63.6**	**3.76**	**8.49**	**17.8**	**0.89**	**12.2**	**43.8**
	Sprat (n=25)			Sea Bass (n=25)			Turbot (n=16)			Dogfish -various spp (n=14)		
	MIN	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**	**MIN**	**MEAN**	**MAX**
ng/kg												
PCDD/F	0.13	0.91	2.55	0.09	0.44	1.34	0.02	0.17	0.44	0.02	0.12	0.30
non ortho-PCB	0.09	1.02	2.25	0.23	1.92	10.38	0.05	0.47	1.37	0.01	0.14	0.46
ortho-PCB	0.01	0.07	0.15	0.02	0.14	0.84	0.01	0.03	0.10	0.01	0.07	0.21
Sum WHO-TEQ	**0.23**	**2.00**	**4.35**	**0.35**	**2.50**	**12.49**	**0.07**	**0.67**	**1.91**	**0.03**	**0.32**	**0.93**
Sum ICES-6 PCB, µg/kg	**1.35**	**11.1**	**28.3**	**2.76**	**22.2**	**145**	**0.52**	**4.98**	**17.2**	**0.11**	**9.82**	**34.0**

Although it remains to analyse the data more closely for geographical variations, which will include examining differences in ratios between the contaminants reported here and other contaminants measured in the investigation and reported elsewhere, the early indication is that UK marine waters currently meet the MSFD criterion for Good Environmental Status for Descriptor 9 (food safety). As expected, sea bass, sprats, mackerel, sardines and herring appear to show the highest levels of contamination. An initial impression of the geographical spread suggests that fish taken waters around the Southern UK/Northern French coasts and the Irish Sea tend to show higher levels of contamination, albeit still within compliance.

A small reduction in concentration levels has been seen for PCDD/Fs and PCBs since the last survey was conducted about a decade ago, and this data will be useful in allowing an updated risk assessment associated with the consumption of these species.

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References:

1. European Commission (2008) Directive 2008/56/EC of the European Parliament and of the Council of 17 June 2008 establishing a framework for community action in the field of marine environmental policy (Marine Strategy Framework Directive) Official Journal (OJ L 164/19)
2. Fernandes A Mortimer D.N., Rose, M., Knowles, T.G & Gem, M. (2009) The Occurrence of Dioxins (PCDDs, PCDFs) and PCBs in Wild, Farmed and Processed Fish, and Shellfish Food Additives and Contaminants B, 2(1), 15-20.
3. European Commission (2011) Commission Regulation (EU) No 1259/2011 of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs.