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PCDD/FS AND PCBS IN BROWN CRAB (CANCER PAGURUS) FROM THE NORTHERN COAST OF FRANCE AND THE INFLUENCE OF THE COOKING PROCESS ON THE COMPLIANCE WITH EU LEGISLATION

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1. Introduction

While the consumption of seafood is linked to health benefits for the general population, the risks connected with the potential exposure to toxic contaminants have also been addressed (1, 2). For example, the edible brown crab has been shown to accumulate chemical pollutants such as metals (3) and organochlorinated contaminants (4). The consumption of seafood is considered as being one of the main routes of human exposure to PCDD/Fs and PCBs (5).

Since 2011, the maximum levels of PCDD/Fs and DL-PCBs for crustaceans should be applied solely to muscle meat from appendages, excluding the muscle from cephalothorax and the brown meat (6). Moreover, the maximum levels for PCDD/Fs and DL-PCBs in food were updated and values for NDL-PCBs were also set (7). Previous works showed that brown crabs from the Eastern English Channel had pollutant concentrations below regulatory limits (8), but other have also reported higher levels of contaminants in brown meat of decapod crustaceans than in muscle tissue (3). The effects of cooking on PCDD/F and PCB concentrations in appendage muscle due to possible contamination from other tissues, mainly from brown meat, have not yet been documented. Considering that according to EU regulation, PCDD/F and PCB limits are only established for muscle meat from appendages (5), it was essential to investigate if a transfer of these pollutants could occur between tissues during cooking. Hence, the present study reports the PCDD/F, DL-PCB and NDL-PCB concentrations and PCB profiles of raw and cooked muscle and brown meat of C. pagurus from the northern French coast. Besides providing data on the current contamination status of this specie in a region previously identified as polluted, the objective was to assess the influence of different cooking processes on the chemical status and conformity of brown crabs with the established maximum limits for PCDD/Fs, DL-PCBs and NDL-PCBs.

2. Materials and methods

In June 2015, 45 brown crabs were captured off the coast of Dieppe (Seine-Maritime department, France). Crabs were divided into five groups according to cooking procedures investigated: raw (control), boiled for 15 min, boiled for 30 min, steamed for 15 min, frozen for one week followed by boiling for 15 min (Fig. 1). Raw and cooked crabs were dissected and the edible tissues of each individual were separated into muscle meat from appendages (legs and claws), muscle from the cephalothorax, and brown meat. Afterwards, tissues were freeze-dried, homogenized with a grinder and stored pending analysis.

PCDD/Fs and PCBs congeners have been monitored using ISO17025 accredited methods based on mass spectrometry determination, as described elsewhere (9). Briefly, samples were extracted automatically in a pressurized solvent extraction device using a mixture of toluene and acetone as solvent. Next, the solvent was evaporated and the lipid content of the sample was determined gravimetrically. Purification and fractionation of PCDD/Fs and PCBs were carried-out in four sequential chromatographic steps involving multilayered sulfuric acid-activated silica gel, water deactivated Florisil® and carbon columns. The target contaminants were quantified by gas chromatography (7890A; Agilent Technologies, USA) coupled to a double electromagnetic sector high resolution mass spectrometer (JMS-700D and 800D; Jeol, Japan) set at a resolution of 10 000. Acquisition was performed in the single ion monitoring mode and for quantification as required by the isotopic dilution method principle. Toxic Equivalent Quotient values (TEQ) were calculated according to the 2005 World Health Organization Toxic Equivalency Factors (2005-WHO-TEF) and expressed on a fresh-weight basis for seafood.

3. Results and discussion:

Considering EU regulatory limits (5), the muscle meat from legs and claws of brown crabs harvested off the coast of Dieppe were considered safe for human consumption. The PCDD/F and PCB concentrations measured in brown meat were however well above the values measured in muscle tissues, reaching levels 20-fold higher than those measured in the appendages. These values were furthermore higher than the limits set for appendages' muscle, demonstrating that PCDD/F and PCB burdens in C. pagurus brown meat can represent a potential health risk for population consuming all edible tissues. Therefore, crustaceans' consumers should be advised to eat exclusively muscle from legs and claws of C. pagurus. Furthermore, future risk assessments should be conducted taking into account the whole crab, i.e., muscle from appendages and cephalothorax, and brown meat.
The most abundant congener found in both muscle from appendages and cephalothorax, and in brown meat was the 2.3.7.8-TCDF followed by PeCDFs. As it is known that 2,3,7,8-TCDF and 2,3,4,7,8-PeCDF may also derive from technical PCB solutions (10), the predominance of these compounds in crab samples could therefore be explained by the proximity of the Seine Bay, which was exposed for a long time to commercial PCB mixtures. Regarding PCBs, the most abundant congener was NDL-PCB 153. The DL-PCB and NDL-PCB profiles for the analyzed crab tissues showed higher proportions of DL-PCBs 156, 157, 167, 189 and NDL-PCB 180 in brown meat compared to muscle. The brown meat is mainly composed of hepatopancreas, which is the main site of cytochrome P450-dependent biotransformation of PCBs in crustaceans. The different PCBs profiles between brown meat and muscle may thus reflect the inability of the species to metabolize these congeners.

Regarding the influence of cooking processes on the contamination levels, Table 1 shows the PCDD/F, DL-PCB and NDL-PCB concentrations in crab tissues using common culinary practices. The results are presented in terms of dry weight to allow direct comparison between muscle and brown meat since differences in loss of water content were observed depending on the cooking process. No evidence was observed for a decrease of lipid content in the cooked tissues. The present study showed that when crabs are cooked whole, the PCDD/F and PCB levels in appendages' muscle are not significantly higher than those determined in raw crabs. Therefore, consumers who do not deliberately eat the cephalothorax content or the brown meat of C. pagurus knowing the potential health risk associated with their ingestion, can still freeze, boil or steam the intact crabs without concern of possible transfer of PCDD/Fs and PCBs from brown meat to muscle from legs and claws.

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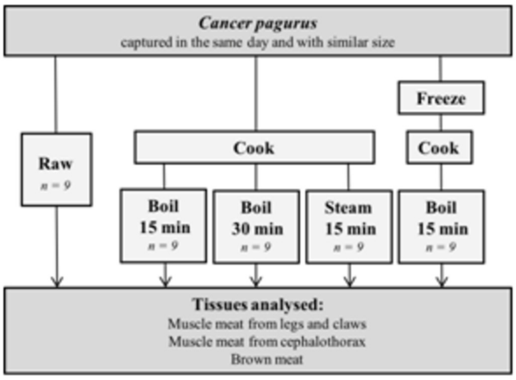

Fig. 1. Schematic representation of the experimental design.

Table 1. Mean values and standard deviations of water and lipid content (%), and concentrations of PCDD/Fs, DL-PCBs and NDL-PCBs in muscle from appendages and cephalothorax, and from brown meat of crab Cancer pagurus, raw and prepared using different cooking methods (n=9 crabs in each group). Concentrations are expressed on a dry weight basis.

		Raw	Boiled 15 min	Boiled 30 min	Steamed 15 min	Frozen and boiled 15 min
Water (%)	Muscle appendages	20.6 ± 4.5	25.7 ± 0.6	25.9 ± 1.8	26.6 ± 2.2	23.4 ± 3.0
	Muscle cephalothorax	20.4 ± 3.5	24.8 ± 2.4	24.9 ± 1.5	25.6 ± 1.5	23.1 ± 2.3
	Brown meat	33.0 ± 6.0	37.8 ± 1.0	31.6 ± 8.3	33.5 ± 2.1	31.7 ± 3.5
Lipids (%)	Muscle appendages	0.8 ± 0.2	1.3 ± 0.1	1.1 ± 0.2	1.2 ± 0.1	0.8 ± 0.1
	Muscle cephalothorax	0.9 ± 0.1	1.4 ± 0.1	1.4 ± 0.2	1.4 ± 0.2	1.3 ± 0.2
	Brown meat	12.0 ± 4.8	15.5 ± 2.3	11.8 ± 4.0	11.7 ± 1.5	13.3 ± 0.6
Σ_{17} PCDD/Fs ($pg.g^{-1}$)	Muscle appendages	25.8 ± 13.8	24.5 ± 5.1	16.2 ± 2.5	16.4 ± 1.4	15.5 ± 7.8
	Muscle cephalothorax	18.4 ± 4.3	22.7 ± 13.4	24.6 ± 13.8	21.0 ± 4.1	18.3 ± 10.2
	Brown meat	158.2 ± 71.5	116.0 ± 15.2	169.8 ± 71.4	152.5 ± 50.4	185.9 ± 44.0
Σ_{12} DL-PCBs ($ng.g^{-1}$)	Muscle appendages	5.0 ± 1.9	4.1 ± 1.1	4.2 ± 1.4	4.2 ± 0.7	3.7 ± 1.2
	Muscle cephalothorax	5.3 ± 1.2	4.5 ± 1.2	8.9 ± 5.7	8.1 ± 3.2	9.3 ± 5.3
	Brown meat	99.7 ± 22.9	70.0 ± 20.2	101.0 ± 60.2	106.1 ± 41.3	131.9 ± 17.8
Σ_6 NDL-PCBs ($ng.g^{-1}$)	Muscle appendages	16.7 ± 5.3	13.8 ± 4.6	14.7 ± 6.1	13.4 ± 1.3	13.0 ± 4.6
	Muscle cephalothorax	19.7 ± 5.0	18.2 ± 4.2	34.8 ± 20.8	31.5 ± 11.2	40.9 ± 24.7
	Brown meat	421.8 ± 79.2	314.6 ± 97.9	445.8 ± 264.6	444.5 ± 163.1	617.6 ± 87.0