

# ECsafeSEAFOOD

## Priority environmental contaminants in seafood: safety assessment, impact and public perception

Grant agreement no: 311820

### Deliverable D2.9

### Correlation between biological parameters and level of contamination

**Due date of deliverable:** M38

**Actual submission date:** M47

**Start date of the project:** 02/2013      **Duration:** 48 months

**Organisation name of lead contractor:** ICRA, IMARES

**Revision:** V1

Project co-funded by the European Commission within the Seventh Framework Programme (2007-2013)	
Dissemination Level	
PU Public	x
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## Table of Contents

1. Summary .....	3
2. Introduction .....	3
2.1. Effect of size on the level of contaminants.....	4
2.1.1. Inorganic arsenic (iAs).....	4
2.1.2. Total Mercury (T-Hg) and methyl-mercury (MeHg).....	4
2.1.3. PBDEs .....	6
2.1.4. BFRs.....	6
2.1.5. PFAs.....	8
2.1.6. PHAs .....	8
2.1.7. UV Filters:.....	8
2.1.8. Musks .....	9
2.1.9. Pharmaceuticals:.....	11
2.1.10. Endocrine disruptors:.....	11
2.2. Effect of lipid content on the level of contamination.....	12
3. Conclusions .....	13

## 1. Summary

In this research, commercially available seafood samples were taken in two sampling rounds (2014 and 2015). Some types were sampled multiple times across their distribution range (e.g mussels), others were only regionally available. Within species, different sizes, if available, were sampled also. In these samples a wide variety of persistent contaminants such as Arsenic (As), mercury (Hg), brominated flame retardants (BFRs), perfluoralkylsubstances (PFAS) and Polycyclic aromatic hydrocarbons (PAHs) and pseudo-persistent contaminants such as UV-filters, musk, pharmaceuticals and endocrine disrupting compounds were analysed. The results of this whole sampling campaign are shown in deliverable D2.4.

In this report the main objective was to research if the observed levels of contaminants were related to biological parameters such as size and species. For this purpose, the approach followed was first to focus on contaminants levels above the method limits of quantification (>MQL), second to plot out the contaminants levels in species for which different sizes were available from the same location, and third, to represent the levels of different species (at least 3 with concentration >MQL) from the same location. This has been performed with wet weight concentrations measured in both commercial sampling carried out within the project, namely round I (R1; 2014) and round II (R2; 2015).

The results clearly show that size can have a large effect on contamination level of some contaminants, but not in all contaminants. Next to this, the effects of the two sampling periods were large too, and sometimes even more pronounced than the effect of size.

## 2. Introduction

In this project, the contaminant levels were analysed in a wide variety of seafood samples. The complete list of analysed species and contaminants are clearly described in Deliverable 2.4 Presence and levels of priority contaminants in seafood. In this report, the influence of species and size on the observed levels were to be investigated. Accumulation of contaminants can be species specific due to e.g. feeding behavior, but also within a species the size can be a contributing factor. Larger specimens tend to contain higher levels of bioaccumulating compounds.

To determine these effects, a proper set of samples as well as properly defined levels of contaminants are required. It became clear that large differences in levels were observed, depending on species, location and sampling round. However, in many cases the levels of contaminants in seafood species were too low to be quantified properly. The influence of the above mentioned parameters could therefore not be determined for all contaminants, nor could all samples seafood species be used in the comparison, as the levels were too low.

## 2.1. Effect of size on the level of contaminants

### 2.1.1. Inorganic arsenic (iAs)

Inorganic arsenic was only determined in mussels and crab. These species were not analysed at different, selected sizes and therefore iAs was not assessed.

### 2.1.2. Total Mercury (T-Hg) and methyl-mercury (MeHg)

Methyl Mercury (MeHg) and total mercury (T-Hg) were measured in pacific tuna, octopus and sole of different size classes from the Mediterranean and the results are shown in figure 1. Compounds were present in concentrations >MQL in both sizes. For T-Hg, a small increase in concentration was observed with increase in size for tuna and octopus in both sampling rounds. This tendency was not observed for sole.

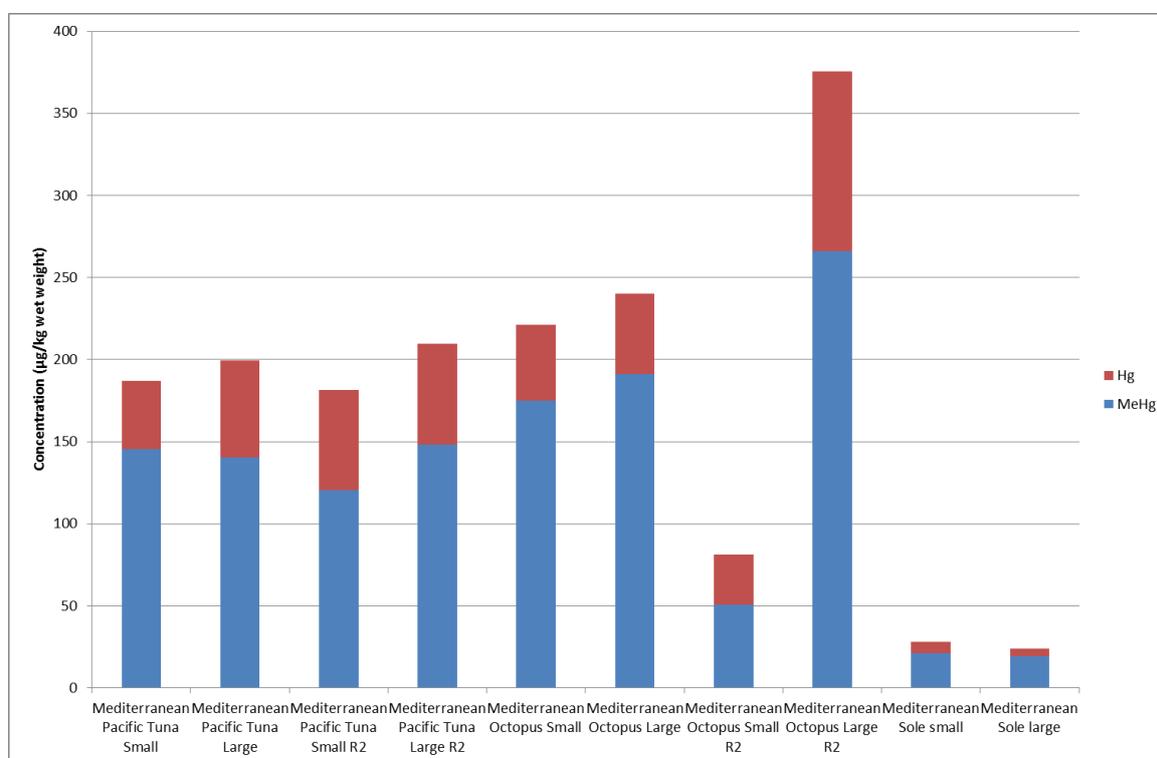


Figure 1. Levels of mercury in samples from species of different sizes from the Mediterranean in sampling round 1 and round 2 (R2)

For MeHg and T-Hg in Plaice from the North Sea and the Channel a decrease in concentration was observed with increase in size for each species, except for Round 2 in the Channel (Figure 2). This could be caused by the fact that smaller plaice is resident in shallower waters, close to the coast, while larger plaice migrates to deeper water, further from the coast. Mercury pollution can be lower in open seas; the larger size class has experienced more “growth-dilution” than the smaller size-class.

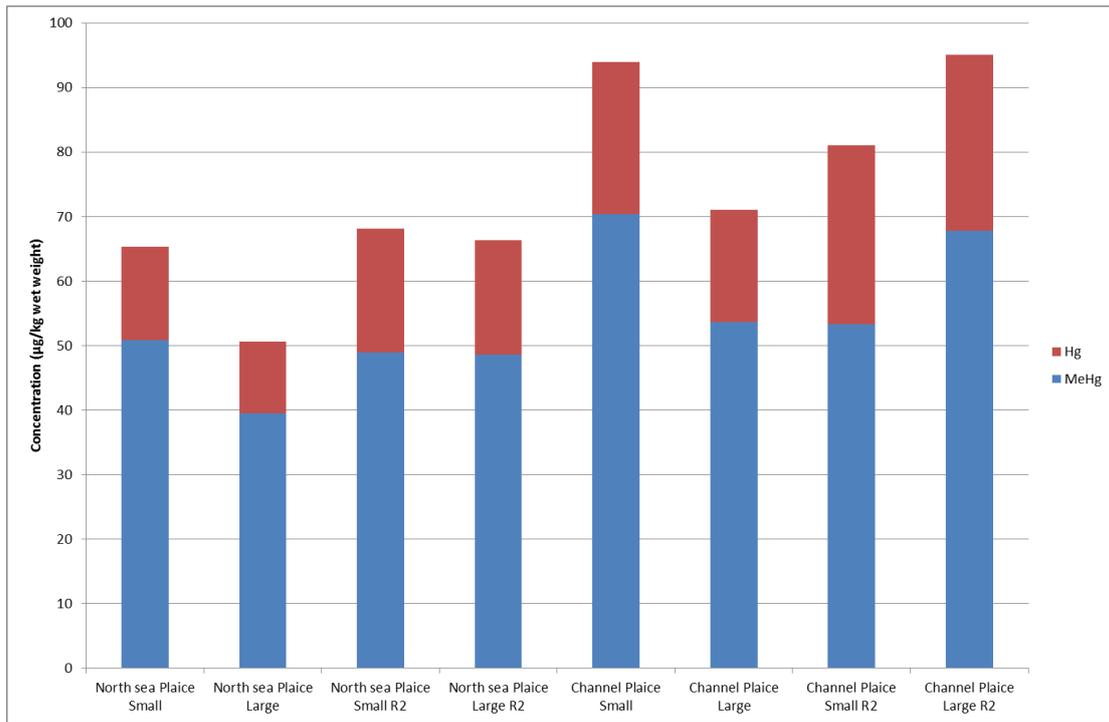


Figure 2. Levels of mercury in samples from species of different sizes from the North Sea and the Channel in sampling round 1 and round 2 (R2)

For MeHg and T-Hg in species from Portugal, South America and Africa an increase in concentration was observed with increase in size for each species (Figure 3).

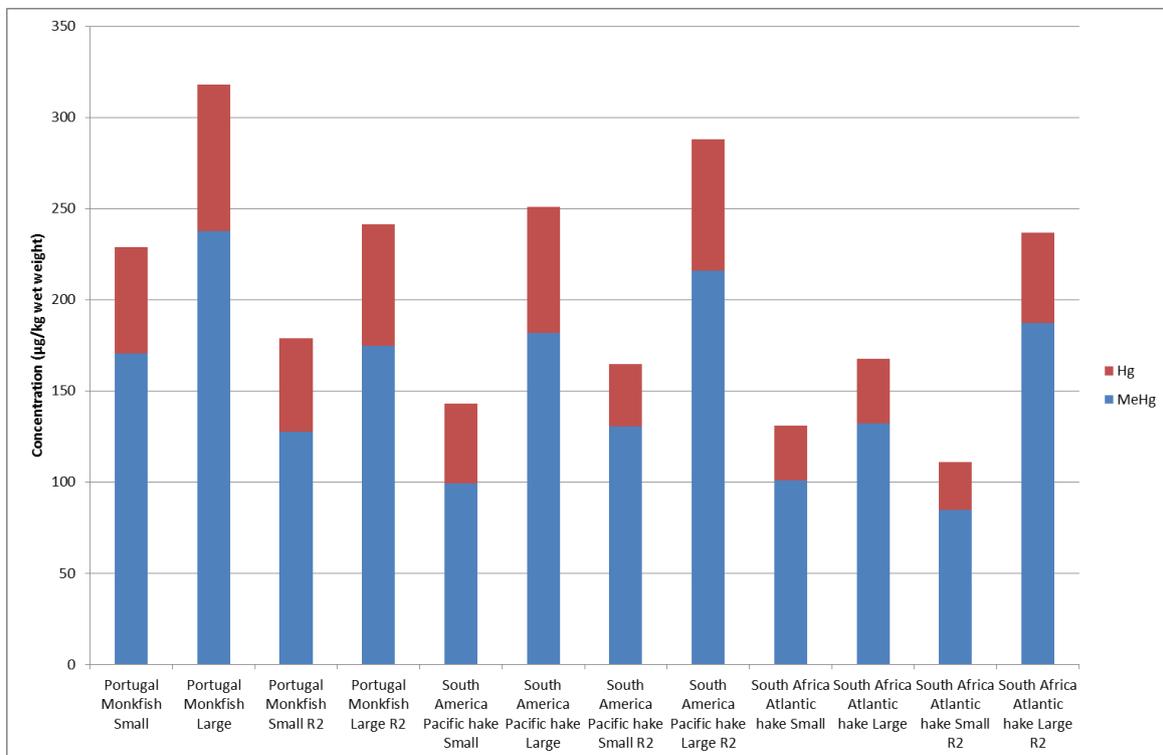


Figure 3. Levels of mercury in samples from species of different sizes from Portugal, South America and South Africa in sampling round 1 and round 2 (R2)

Overall in round 1, when it comes to the relation between size and levels, results for plaice in the North Sea and the Channel are different from those found at other locations, probably caused by the different habitat choices during their life cycle.

### 2.1.3. PBDEs

For PBDEs, analysis was performed on species of different sizes from the same location only in round 2.

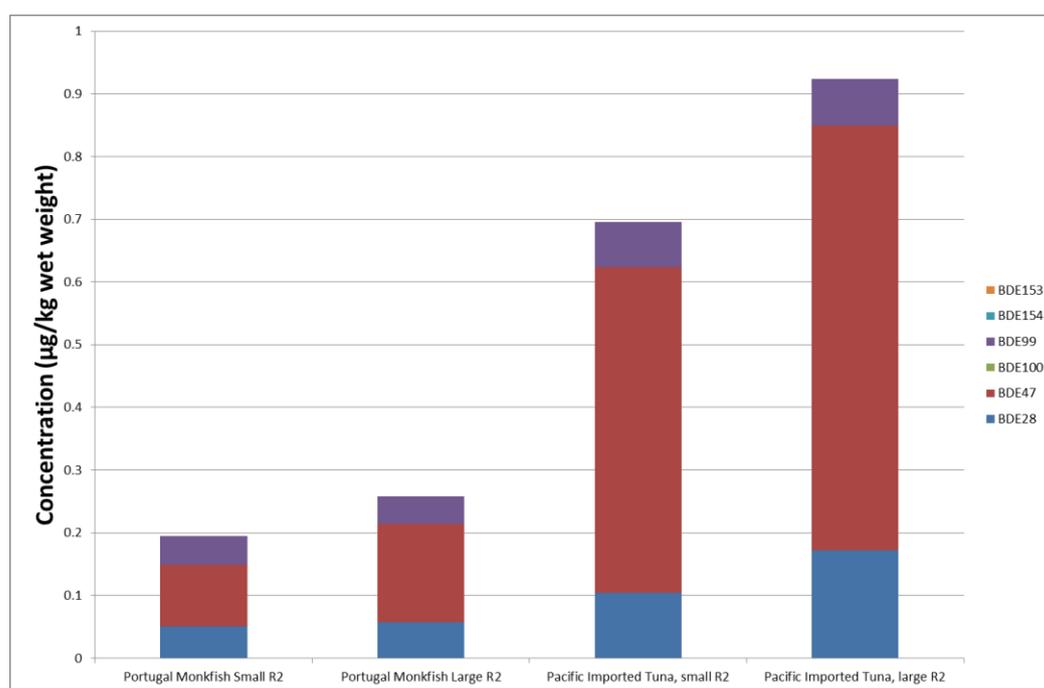


Figure 4. Levels of PBDEs in species of different sizes from the same location acquired in sampling round 2

BDEs 28, 47 and 99 were detected >MQL for Monkfish and Tuna of different size classes (Figure 4). The profile in both species was similar with BDE47 being present in the highest concentration followed by 28 and 99. Concentrations also increased with species size for all congeners, though the effects were not large.

### 2.1.4. BFRs

For new BFRs, such as HBB and 2,4,6- tribromophenol no analysis was performed on species of different sizes from the same location in round 1. In round 2 no effect of size was observed for HBB (Figure 5), the levels of 2,4,6- tribromophenol were too low to quantify.

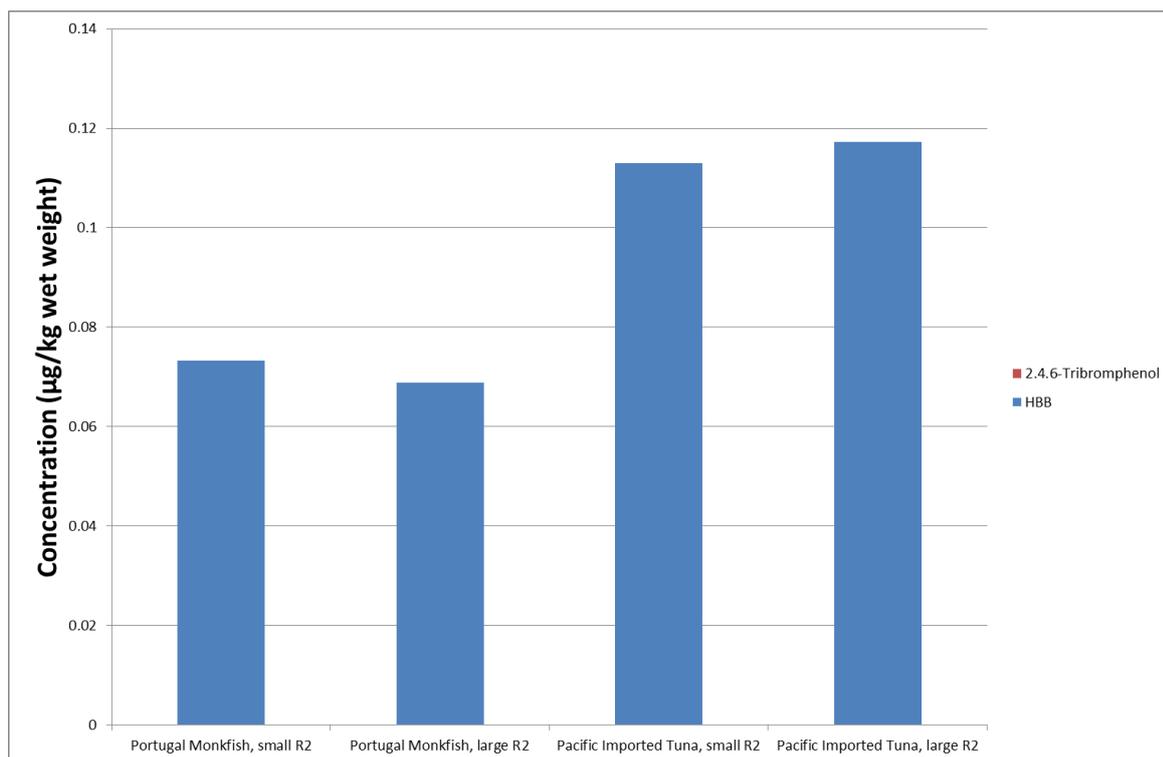


Figure 5. Levels of HBB and tribromophenol in samples from the same species with different sizes from the same location in sampling round 2

Concerning MBDE congeners, a small effect was observed in round 1, with increased levels for larger tuna. On the other hand, in round 2 the levels were very different between large and smaller tuna. While small tuna only presented contamination levels of 6-MBDE-47; larger tuna was mainly contaminated with 2-MBDE-68 (Figure 6).

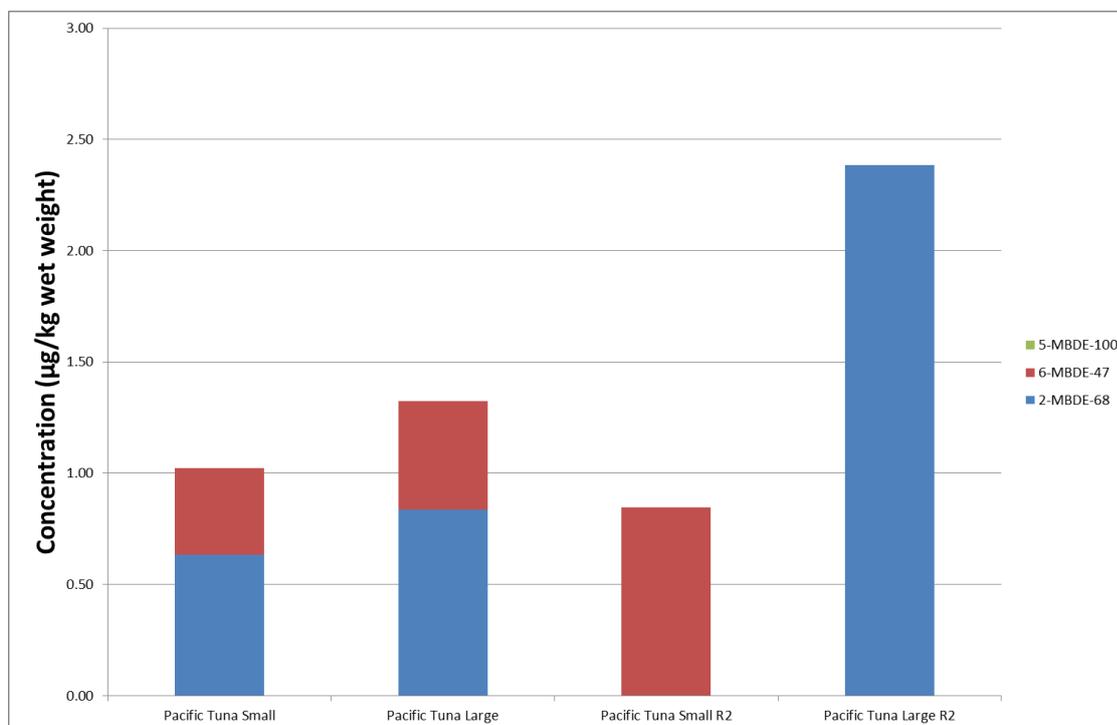


Figure 6. Levels of MBDEs in species of different sizes from the same location acquired in sampling round 1 and round 2 (R2)

### 2.1.5. PFAs

Several PFASs were detected >MQL in species with different sizes from the same location in round 1 and round 2 (Figure 7). In general levels were low, which makes the comparison between different sizes difficult. Although in specific samples (e.g. Tuna and Octopus) large differences were observed between small and large specimen, there is no clear relation between pollution level and size. The level of PFAS in the large tuna from round 2 was relatively high.

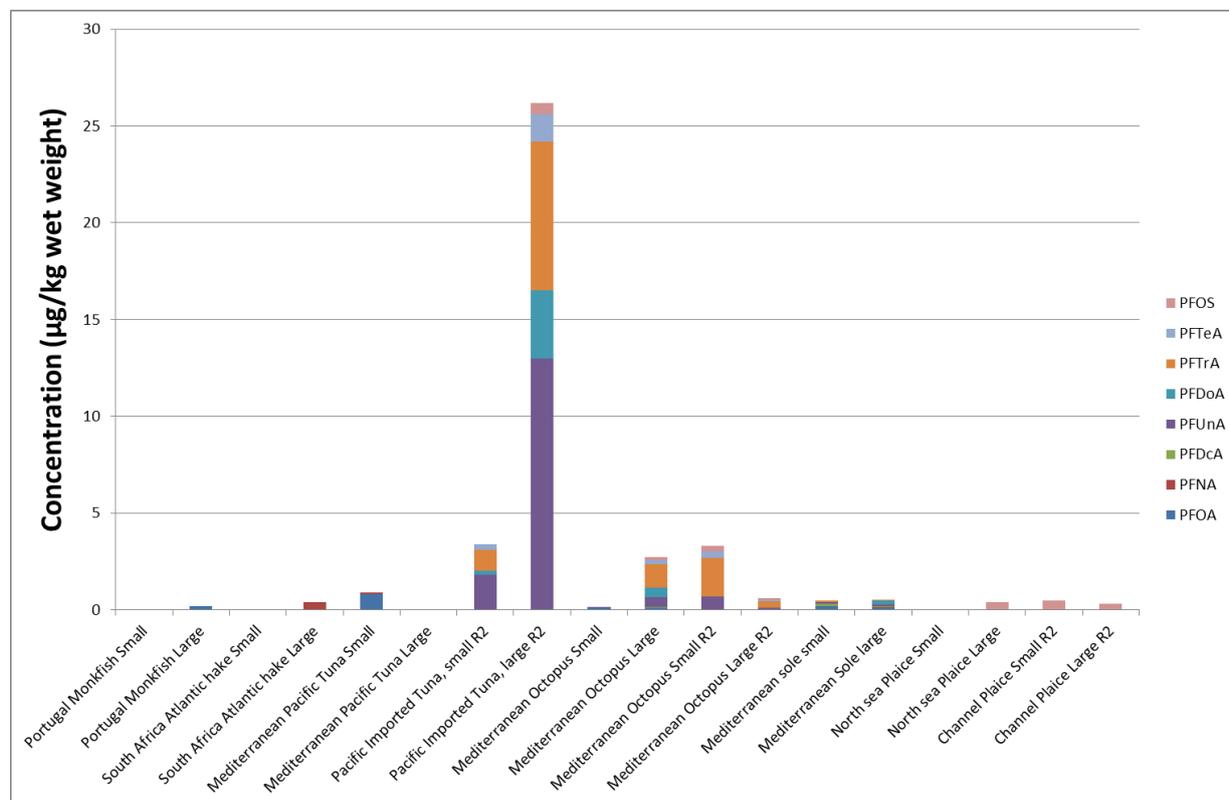


Figure 7. Levels of PFASs in samples from species of different sizes from the same location in sampling round 1 and round 2 (R2)

### 2.1.6. PHAs

PAHs were only determined in mussels and crab, as fish is capable of metabolisation of PAHs. Therefore, there is no data for species with different sizes from the same location.

### 2.1.7. UV Filters:

Monkfish, small and large, from Portugal and Tuna, small and large, from the Pacific Ocean were analysed in round 1 and 2, the results are shown in figure 8. An increasing trend in UV filters concentrations can be observed in monkfish with higher levels measured in large organisms. However, the specific compounds detected in round 1 and 2 were different. While in round 1 4-MBC, BP1, BP3, EHMC, and HS were present; in round 2 DHMB, OC and EHS were measured. Regarding Pacific tuna an opposite tendency was observed in round 1 and 2, besides the compounds measured were not the same.

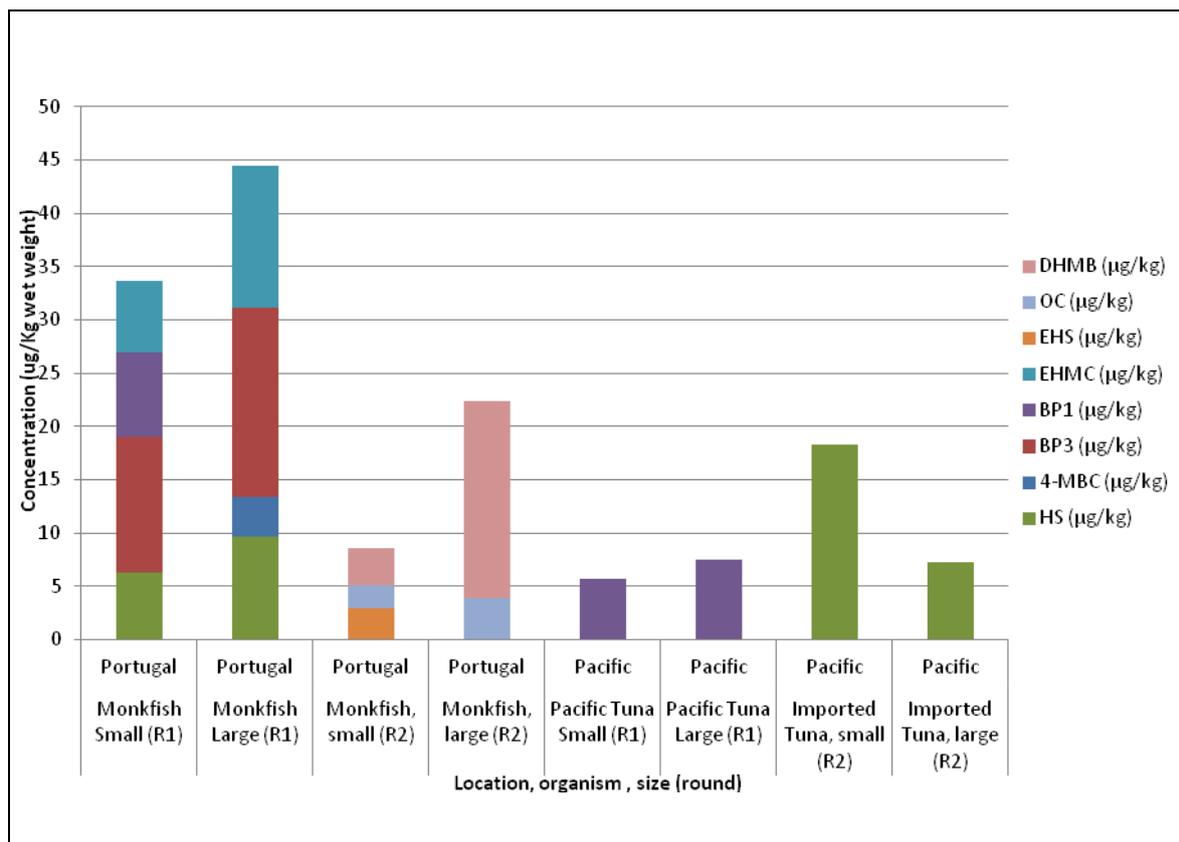


Figure 8. Levels of UV filters in samples from species of different sizes from Portugal and Pacific in sampling round 1 (R1) and round 2 (R2)

### 2.1.8. Musks

For this group of contaminant organisms with different sizes were analysed for monkfish, hake, plaice, octopus, sole and tuna from Portugal, South America, North Sea, English Channel, Mediterranean Sea and Pacific Ocean. The general trend is a decrease in the concentration when the size increases (Figure 9). This was observed in monkfish from Portugal, hake from South America, plaice from the English Channel, octopus from the Mediterranean Sea and tuna from the pacific. As previously reported from MeHg and T-Hg the decrease in concentration observed in plaice can be due to the fact that smaller plaice is resident in shallower waters, close to the coast, while larger plaice migrates to deeper water, further from the coast where the pollution is lower.

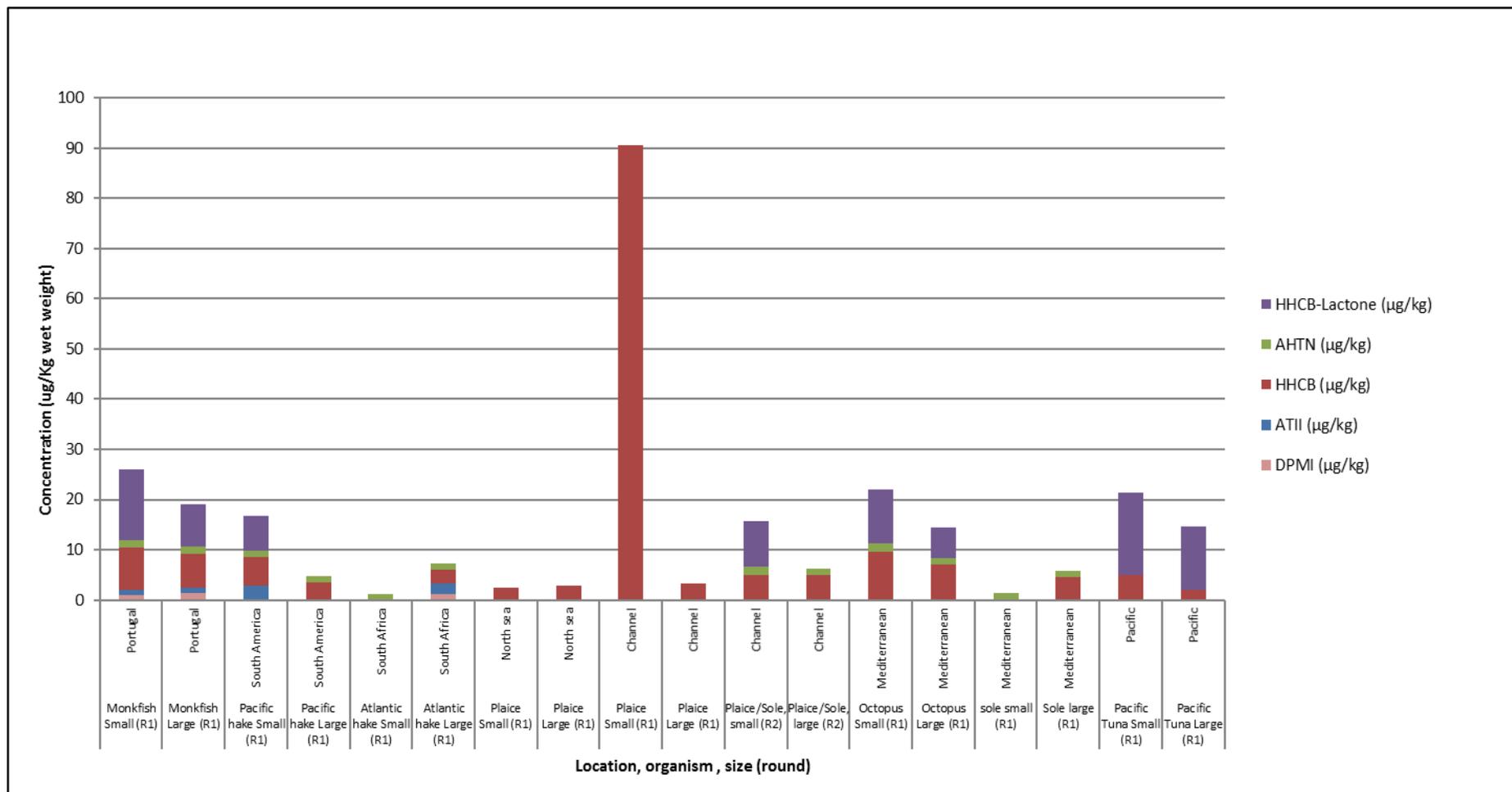


Figure 9. Levels of musk in samples from species of different sizes from Portugal, South America, North Sea, English Channel, Mediterranean and Pacific in sampling round 1 (R1) and round 2 (R2).

### 2.1.9. Pharmaceuticals:

This group of contaminants was quantified in very few occasions and only in round 1. A decrease in concentration was observed for tuna from the Pacific when the size increased for both sotalol and diazepam (Figure 10).

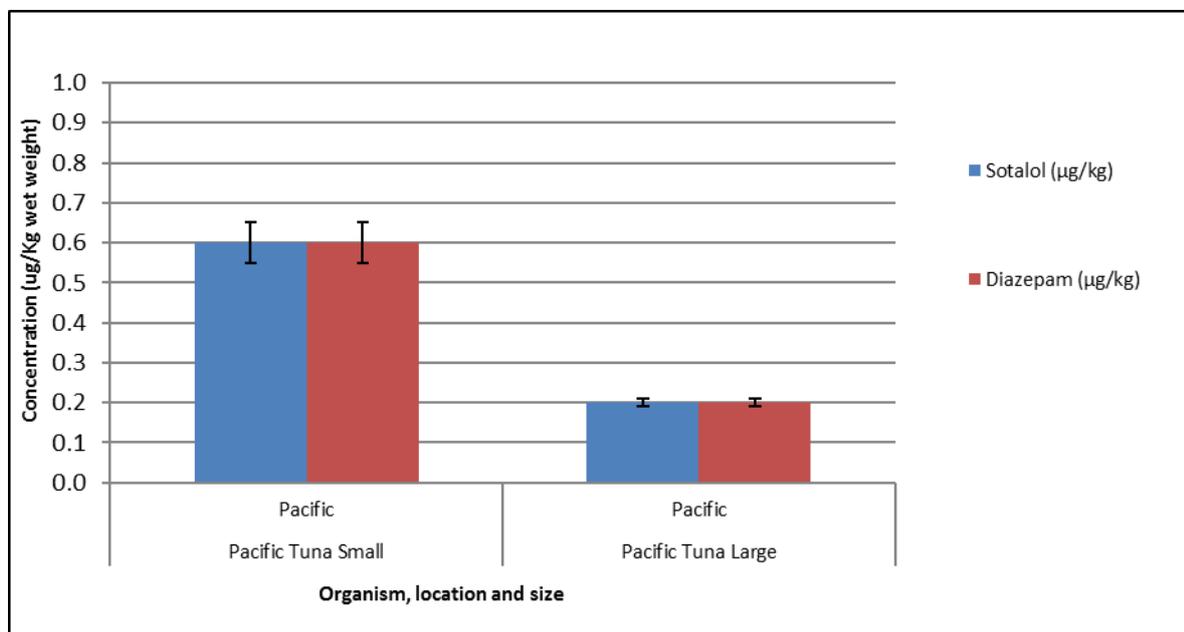


Figure 10. Levels of pharmaceuticals in samples from species of different sizes from the Pacific in sampling round 1

### 2.1.10. Endocrine disruptors:

Tuna, plaice, monkfish and sole from different sizes were analysed for EDCs (fig. 11). No changes in levels with size were detected in tuna from the Pacific, sole from the Mediterranean and plaice from the channel. However, in plaice from the North Sea an increase in the concentration of triclosan for larger organisms was observed, while in monkfish from Portugal and tuna from the Pacific a decreased in BPA level was measured in large specimens.

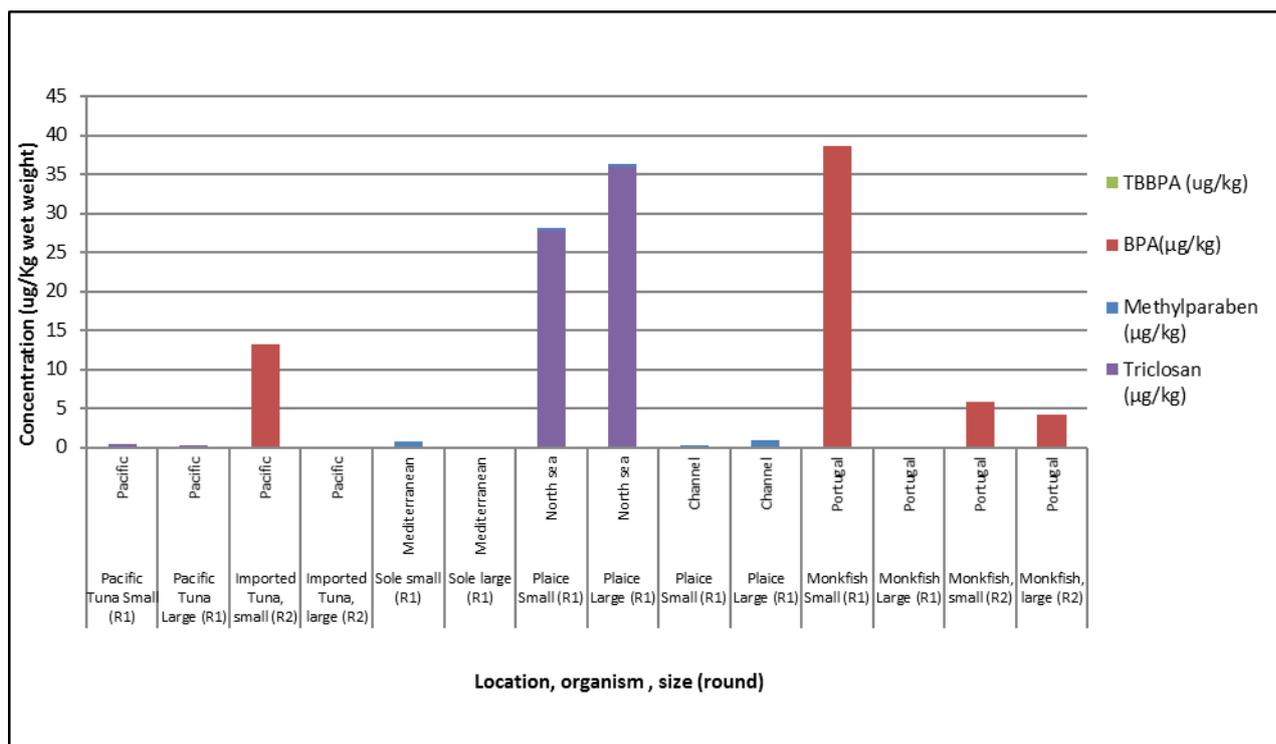


Figure 11. Levels of endocrine disruptors in samples from species of different sizes from the Pacific, Mediterranean, North Sea, English Channel and Portugal in sampling round 1 (R1) and round 2 (R2).

## 2.2. Effect of lipid content on the level of contamination

The effect of lipid levels on contaminant levels is relevant for those compounds that predominantly accumulate in fatty tissues, such as BFRs. For most of the compounds analysed, with a moderate hydrophilicity/lipophilicity the lipid content is not very important. For element like arsenic and (methyl) mercury lipid levels do not have any effect on accumulation. A confounding factor is that the accumulation of these lipophilic compounds is also influenced by diet and age.

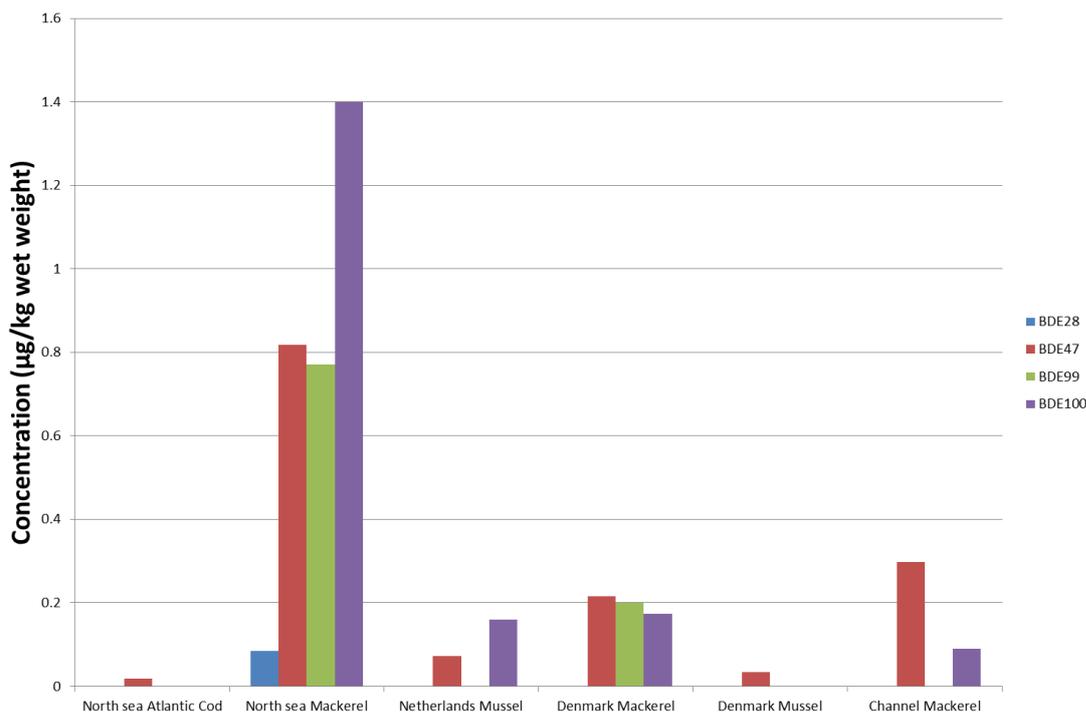


Figure 12. levels of PBDEs in species from the North Sea in round 1

Assessing the levels of BFRs, the levels in a fatty fish like Mackerel are as expected higher than in mussels and cod, both with very low lipid levels, from the same region. Figure 12 also clearly shows that the effect of location is very large. Lipid levels itself can therefore be indicators of levels of lipophilic contaminants levels, but the other parameters influencing levels (species, location, habitat and size) are equally important.

### 3. Conclusions

Overall, a small increase of the contaminant levels with size (table 1) has been observed. Still, this is dependent on the contaminant and species:

- T-Hg and MeHg: the contaminant levels were higher in larger species of Pacific tuna and Mediterranean octopus. In contrast, the larger specimen of Mediterranean sole and North Sea plaice contained lower levels. Nevertheless, Channel plaice showed higher contaminant levels with size in sampling round 2 and the opposite in round 1.
- PBDEs: only BDE 28, BDE 47 and BDE 99 were detected and for both Portuguese monkfish and Pacific Tuna, the contamination levels increased with size.
- BFRs: only HBB and MBDE congeners (2-MBDE-68 and 6-MBDE-47) were detected. While HBB contamination level increased with size for Portuguese monkfish; in Pacific tuna decreased with size. As for MBDE congeners, the contamination level increased with size in tuna, however for different compounds for the two sizes in sampling round 2.
- PFAs: the contaminant levels were higher in larger species of Pacific tuna and Mediterranean octopus.

- UV-filters: the contaminant levels were higher in larger species of Portuguese monkfish. On the other hand, in Pacific tuna, the contaminant levels were higher in larger species from round 1 and lower in round 2.
- Musks: the contaminant levels were higher in larger species of Atlantic hake and Mediterranean sole; while in larger species of Portuguese monkfish, Pacific hake, Channel plaice, Mediterranean octopus and pacific tuna the contaminant levels were lower.
- Pharmaceuticals (PhACs): the contaminant levels of sotalol and diazepam were lower in larger species of Pacific tuna.
- Endocrine disruptors (EDCs): the contaminant levels were higher in larger species of North Sea and Channel plaice; whereas larger species of Pacific tuna, Mediterranean sole and Portuguese monkfish presented lower contamination levels.

**Table 1.** The effect of size on the level of contaminants in samples from species from the same place in sampling round 1 (R1) and 2 (R2).

Species	Place	Sampling round	T-Hg	MeHg	PBDEs	BFRs	PFAAs	UV-filters	Musks	PhACs	EDCs
Tuna	Pacific	R1	↗	↗	n.a	↗	n.a	↗	↘	↘	n.a
Tuna	Pacific	R2	↗	↗	↗	n.a	n.a	↗	n.a	n.a	↘
Octopus	Mediterranean	R1	↗	↗	n.a	n.a	n.a	n.a	↘	n.a	n.a
Octopus	Mediterranean	R2	↗	↗	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Sole	Mediterranean	R1	↘	↘	n.a	n.a	n.a	n.a	↗	n.a	↘
Plaice	North Sea	R1	↘	↘	n.a	n.a	n.a	n.a	n.a	n.a	↗
Plaice	North Sea	R2	↘	↘	n.a	n.a	n.a	n.a	n.a	n.a	n.a
Plaice	Channel	R1	↘	↘	n.a	n.a	n.a	n.a	↘	n.a	↗
Plaice	Channel	R2	↗	↗	n.a	n.a	n.a	n.a	↘	n.a	n.a
Monkfish	Portugal	R1	↗	↗	↗	n.a	n.a	↗	↘	n.a	↘
Monkfish	Portugal	R2	↗	↗	n.a	↘	n.a	↗	n.a	n.a	↘
Hake	Pacific	R1	↗	↗	n.a	n.a	n.a	n.a	↘	n.a	n.a
Hake	Atlantic	R1	↗	↗	n.a	n.a	n.a	n.a	↗	n.a	n.a

↗ = increase of the contaminant level with size; ↘ = decreased of the contaminant level with size; n.a. = not analysed

This preliminary work shows that biological parameters, such as size and species, have influence on the contamination levels of several commercial available Seafood species studied within the ECsafeSEAFOOD project. The levels are clearly not only influenced by size or species, the sampling period can have large effects also.

This study was designed to get an overview of parameters that influence contaminant levels in a wide variety of commercial seafood species worldwide. Clearly worldwide, and species dependent, large differences in levels and types of contamination can be found. To assess the effect of biological parameters into detail a study should entail a few species only, with more sampling points focusses on biological parameters only.