What about Environmental Quality Standards for PBDEs in European river biota?

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Introduction

Flame retardants (FRs) are widely used to increase the fire resistant of some materials. One of the most widely used FRs were polybromodiphenyl ethers (PBDEs). These compounds have a high lipophilicity and therefore they can be bioaccumulated and biomagnified along the food chain. Moreover, several toxic properties have been reported for PBDEs: they can act as endocrine disruptors and affect neurological, thyroid and liver activity. As a result in 2009 the Stockholm Convention decided to add the commercial mixtures with four, five, six and seven bromines to the Annex A, to end their production and use [1].

Moreover, the European Directive regarding priority substances in the field of water policy established in 2013 Environmental Quality Standards (EQS) in biota for some pollutants, such as PBDEs [2]. This standard was set at 0,0085 ng/g wet weight (ww), and refers to the sum of the concentrations of congener numbers 28, 47, 99, 100, 153 and 154.

There are several studies in the literature reporting PBDE levels in river fish samples. For instance, PBDE levels were recently determined in different fish samples collected in river basins from Spain, Italy, Greece, Slovenia, Croatia, Bosnia and Herzegovina and Serbia [3,4]. Despite its ban, and due to its persistent nature, PBDEs continue to be detected in all analysed samples showing their ubiquity in river environments.

The objective of this study was to collect published data (last 5 years) in European river basins, as well as in other places around the world, and to assess whether the established EQS are being met.

Data collection

Given the ban on the production and use of PBDEs in Europe since 2004, it is to be expected that environmental levels of PBDEs will decline over the years. Therefore, in order to evaluate the current situation, the literature search has been restricted to the last 5 years. On the other hand, and to assess the situation in Europe, but also in other areas of the world, we have collected data from different continents.

It should be pointed out that PBDE data are generally expressed as concentrations in lipid weight basis. However, EQS in biota are expressed in wet weight basis. That because, all collected data must be transformed from lipid weight (lw) to wet weight (ww) basis. However, not always the necessary information to perform this transformation was available. Taking into account that fish lipid content varies according to species, age, season and location, two different approaches were made defining two scenarios: the best scenario, in which fish lipid content was established at 0.5% ww; and the worst scenario, in which fish lipid content was set at 20% ww [5].

Results and discussion

In spite of the numerous studies carried out for the determination of PBDE levels in river fish, studies corresponding to the last 5 years (2012-2017) are not so abundant. Table 1 shows a summary of these works. As can be seen, we have PBDE data in different European countries (Belgium, Bosnia and Herzegovina, Croatia, Germany, Greece, Italy, Serbia, Slovenia, Spain and UK), as well as in North America (Canada) and Asia (China) [6-14].

Table 1. Summary of PBDE levels (expressed in ng/g ww) in fish samples collected around the world.

Country	Year	n	Range	% > EQS	% > EQS x 10	% > EQS x 100	% > EQS x 1000	% > EQS x 10000	Ref.
Germany	nr	61	< LOD – 46.3	87	98	94	51	-	[6]
Spain*	2010	48	< LOD – 2.60	73	69	5.7	-	-	[3]
			< LOD – 104	85	95	83	41	2	
Belgium	2000-09	26	0.77- 93	100	100	96	62	4	[7]
UK	2007-12	42	2.30 – 24.5	100	100	100	*nr*	*nr*	[8]
Italy, Greece, Slovenia, Croatia, Bosnia and Herzegovina and Serbia*	2014-15	27	0.03 – 3.26	100	85	15	-	-	[4]
			1.18 - 130	100	100	100	70	11	
Canada*	2008-12	50	0.47 – 120	100	100	*nr*	*nr*	*nr*	[9]
			18.8 - 4806	100	100	100	100	*nr*	
Canada	2006-13	470	1 - 390	100	100	100	*nr*	*nr*	[10]
China*	2010	34	0.18 – 2.8	100	100	*nr*	*nr*	*nr*	[11]
			7 - 112	100	100	100	*nr*	*nr*	
China*	2014	120	0.03 – 3.45	100	*nr*	*nr*	*nr*	*nr*	[12]
			1.38 - 138	100	100	100	*nr*	*nr*	

* Studies in which the transformation from lw to ww was not possible. Two scenarios were established: the top row corresponds to the best scenario (fish lipid content of 0.5% ww), and the bottom row corresponds to the worst scenario (fish lipid content of 20% ww).

nr Not reported

Despite the ban of PBDEs in Europe since 2004, these pollutants are still present in virtually all European river fish. In addition, between 73 and 100% of fish presented levels of contamination higher than EQS value of 0.0085 ng/g ww, established in the 2013/39/EU Directive. On the other hand, between 83 and 100% of fish exceeding the EQS value, their contamination was above 100 times the EQS value, and between 41 and 70% above 1000 times the EQS value. Similar situation was observed in other locations of the world, such as North America and China.

Restrictions in the use of PBDEs are beginning to yield declines in concentration in environmental samples. But in the case of POPs, their ban does not solve the environmental problem in the short or medium term. For instance, a study in the Lake Ontario (Canada) showed that since 1997, penta-BDE levels have declined at an annual rate of 4.5% [13]. Despite this decline, levels in fish were still above the guideline in the most recent years of monitoring.

The European directive established that, *with the aim of achieving good surface water chemical status, the EQS for substances such as PBDEs should be met by the end of 2027*. If we take into account an annual decrease of 5%, after 10 years, samples actually with levels higher than 100 times the EQS (between 83 and 100%), will present concentrations about 0.40 ng/g ww, being still 50 times above the value established by the European directive. Thus, it will take several decades to reach levels of pollution recommended by the legislation.

Based on these results, it would be important as proposed by the European directive "*taking measures additional to those already taken, including at international level, to reduce or eliminate discharges, emissions and losses of those substances so as to achieve the objectives*". In this sense, and taking into account that there is already a ban on the production and use of PBDEs at international scale through the Stockholm Convention [1], it is necessary to carry out new strategies. For instance, it is important to take into account what happens with waste from materials that still contain PBDEs. A considerable fraction of PBDEs containing waste is recycled, with little information about the fate during recycling. It is necessary to develop recommendations for waste management to ensure the minimum environmental impact and maximum transfer to appropriate final sinks.

Finally, due to the ongoing restrictions on PBDEs, there has been a shift in the use of alternative FRs. Attention must also be paid to the occurrence and time trends of these alternative FRs in river sediments and fish samples. Studies for these alternative FRs are strongly recommended.

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