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Spatial analysis of polybrominated diphenylethers (PBDEs) and polybrominated biphenyls (PBBs) in fish collected from UK and proximate marine waters

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Abstract: Some commonly consumed marine fish species are considered to display a higher risk of bio-accumulating organic environmental contaminants such as PBDEs. As part of a study to investigate the spatial distribution of these contaminants, data on polybrominated diphenlyethers (PBDEs) and polybrominated biphenyls (PBBs) were collected and analysed by introducing a web-based resource which enables efficient spatial, species and concentration level representations. Furthermore, hierarchical cluster analyses permits correlations within the data to be predicted. The data provide current information on levels of PBDE and PBB occurrence, allowing identification of locations that show higher contaminant levels.

135 fish samples of various species were analysed from UK marine waters, but encompassing the waters around Norway in the North and to the Algarve in the South. PBDEs were observed in all samples with the majority of measured congeners being detected. The concentrations ranged from 0.087 µg/kg to 8.907 µg/kg whole weight (ww) for the sum of all

measured PBDE congeners. PBBs occurred less frequently showing a corresponding range of < 0.02 µg/kg to 0.97 µg/kg ww for the sum of seven PBB congeners. Concentrations vary depending on species and locations where landed, e.g. PBBs occurred more frequently and at higher levels in grey mullet from French waters. The high frequency of PBDE occurrence makes it prudent to continue the monitoring of these commonly consumed marine fish species. The web-based resource provides a flexible and efficient tool for assessors and policy-makers to monitor and evaluate levels within caught fish species improving evidenced-based decision processes.

Keywords: polybrominated diphenlyethers, polybrominated biphenyls, fish, spatial analysis, visualization, hierarchical clustering

1. Introduction:

The bioaccumulation of environmental contaminants by various marine fish and shellfish species has been widely documented (Bruggeman et al., 1984; Magalhaes et al., 2003; El-Moselhy et al., 2014; Hashizume et al., 2014). Thus, the consumption of edible species of marine fish and shellfish has the potential to make a significant contribution to the human exposure of these 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 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 along with compliance with maximum contaminant levels established by European Commission regulation, or other relevant standards. In addressing this aim, complex and large datasets require analyses which encompasses spatial, species and concentration levels presenting a challenge to assessors to be able to clearly identify trends or correlations.

PBDEs are classified as legacy brominated flame retardants (BFRs) that were manufactured for incorporation into a number of commonly used commercial materials such as plastics, rubbers, textiles and electronic components. These open-ended applications allow the PBDEs to diffuse out of materials into the environment, and this can occur during manufacture, use and disposal of the product.

As an important analogue of PBDE, polybrominated biphenyls (PBBs) had been primarily produced as BFRs during the early 1970s until the Michigan incident in 1973 (Luross et al., 2002). Due to their persistence in the environment, PBBs still show (environmental, food?) occurrence in the reports published recently (Luross et al., 2002; Bramwell et al., 2017; Chang et al., 2017). Both PBDEs and PBBs. could bioaccumulate through the food chain, and cause endocrine system disruption, neurobehavioral effects and reproductive toxicity (McDonald, 2002). Additionally, they may be particularly harmful during a critical window of brain development during pregnancy and early childhood (Fernandes A, 2012; Chiaramonte and Zota, 2016; Schrenk and Cartus, 2017). The detection and analysis of the concentration level and geographical dispersion of these contaminants are of great significance, which will provide a useful reference for the development of related policy. Even though their occurrence in food has been investigated (FSA, 2006; Fernandes A, 2009), a more comprehensive investigation is still needed.

As an example, PBDE/PBB concentration data in various fish species caught across various locations are analysed (Fernandes et al., 2004; Fernandes et al., 2008). This dataset exemplifies the problem of variation in concentration levels across congeners (orders of magnitude), species and geographical locations and we present a proposed methodology to efficiently explore spatial distribution in species and congeners to demonstrate an efficient and intuitive representation method.

Google Maps is a web mapping service which provides a powerful platform for geographical data visualization (Google Maps 2016). To have a better understanding of the PBDE dispersions around UK, and also to provide a convenient method to visualize geographical information for both, professionals and laypersons, this paper introduces a novel web-based resource developed from google maps to efficiently represent and explore the complex inter-relationships between the fish species, contaminants, catch locations and statistical clustering within the data. Each sample was presented with a circle or radiation pattern marked at its GPS position on an interactive webpage with different colours used to distinguish the species or contaminants, and radius (for circles) or offset distance (for radiation pattern) to represent the contaminant concentration. The webpage also allows users to select individual interested species and contaminants. Concentration levels across congeners may vary by orders of magnitude, therefore a scaling factor is included to allow easy calibration of the presented data. Thus, this paper aims to present a geographical data visualization method to present complex concentration data sets across multiple species, congeners and locations and provides results of PBDE/PBB data obtained from a study to investigate occurrence in commonly consumed marine fish species in UK and proximate marine waters as an example application.

2. Methods and Materials

2.1 Sample preparation and analysis

135 samples were collected mostly from waters around the UK, but extending to other North Atlantic regions around Norway, the Irish Sea, and the European coastal North Atlantic regions, including the North Western coast of France, Biscay and the Algarve. The main species

targeted were sardines, sprats, sea bass, mackerel, herring, grey mullet, but other species were also included.

Whole fish were dissected to collect edible muscle tissue and to exclude skin, bones and organs. However, for some species such as sprats, whole fish were used. This reflects consumer practice for fish consumption. The selected tissue was minced, homogenised by blending and freeze-dried. Dry samples were stored at -18 °C and re-homogenised before analysis.

The method used for the preparation, extraction and analysis of samples has been reported previously (Fernandes et al., 2004; Fernandes et al., 2008). In brief, samples were fortified with ^{13}C-labelled analogues of target compounds and exhaustively extracted using mixed organic solvents. PBDEs and ortho substituted PBBs were separated from other contaminants by fractionation on activated carbon. The fraction was purified using adsorption chromatography on alumina. Analytical measurement was carried out using high resolution gas chromatography-high resolution mass spectrometry (HRGC-HRMS). The analysis is ISO 17025 accredited (UKAS) and includes an in-house reference material and method blanks which were evaluated prior to reporting of sample data and used to determine the limits of detection.

PBDE congeners analysed (IUPAC numbers 17, **28**, **47**, **49**, 66, 71, 77, 85, **99**, **100**, 119, 126, **138**, **153**, **154**, **183** and **209**) include those specified in European Commission recommendation 2014/118/EU which are given in bold font. PBB congeners included: IUPAC numbers 49, 52, 80, 101, 153 and 209.

2.2 Software and data analysis

2.2.1 Interactive webpage for data visualization

To efficiently visualize the geographical dispersion of the contaminants, an interactive webpage (www.pbde.droppages.com) was designed based on Google Maps which utilised sample GPS data and the sample concentrations. As shown in Fig.1, the webpage primarily consists of the following control fields; species selection, contaminant selection, factor selection (for scaling), colour legend of selected species/contaminant and a help field. This permits a convenient facility to explore required permutations of the available underlying data.

Where a single fish species and contaminant are selected, each sample is presented with a circle located at its reported catch location based on the Global Positioning System (GPS) coordinates and where the radius of each circle reflects the level of the measured concentration (ng/kg) within the sample. The concentration in each sample is represented by a variable radius defined as in equation (1),

$$\text{radius} = \text{Factor} \times \text{C} \qquad (1).$$

Where *C* is the contaminant concentration, *Factor* is the scaling factor which is set by default automatically but which may be modified by the user dependent upon the contaminant concentration which vary by orders of magnitude between the congeners and also allows recalibration due to the map size selected.

Where multiple species or contaminants are selected, differentiation between the species/contaminants is achieved by the use of colour codes and line type (including solid, dashed and dotted) shown in the legend and which is applied to the relevant circle representations. Additional data retrieval within the same species may be achieved by placing the cursor over a particular sample, in which case an information window will be presented to display the cumulative distribution function of the sample set and which provides the ranking of current sample (red star) and, if required, specific details for that sample will be displayed if the cursor is moved over the red star.

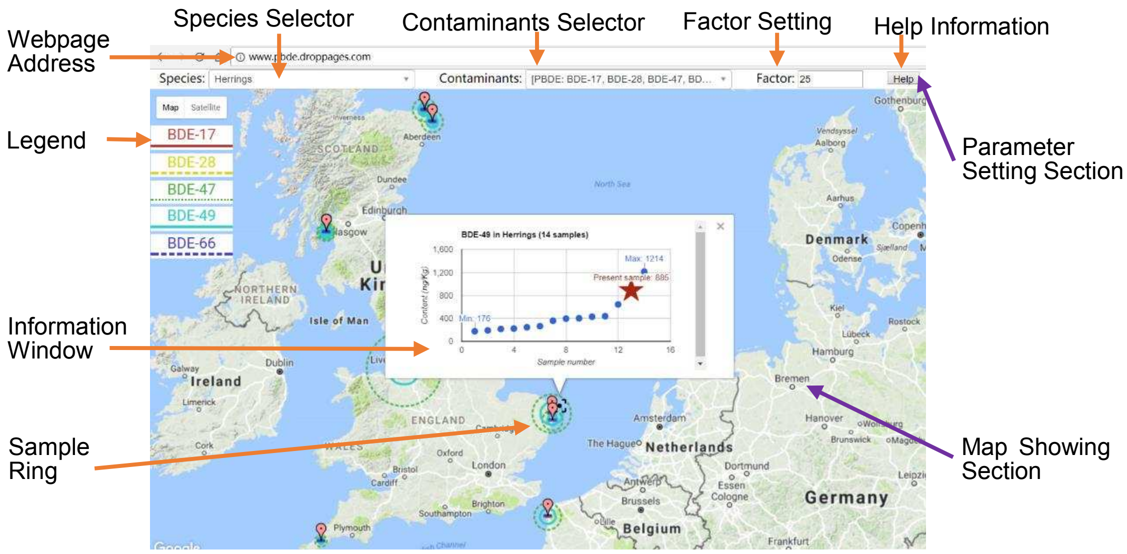

Fig.1 Interactive webpage for data visualization based on google maps GPS coordinates of where the sample was reported as caught. Circle radius indicates concentration level and inset indicates total sample set information. Additional fields show species selection, contaminant selection, factor selection (for scaling), colour legend of selected species/contaminant and a help field.

If multiple species and contaminants are selected simultaneously, each sample is presented with a radiation pattern as shown in Fig.2. Different colours are used to distinguish different species and all selected contaminants are arranged in sequence at the vertices of a k -polygon in a clockwise direction. The radiation pattern is centred at the sample catch location and is associated with the contaminant concentration as described in equations (2) and (3)

$$\theta_k = 360° / n \times k \qquad (2)$$

$$d_k = \text{Factor} \times C_k \qquad (3)$$

Where n is the total number of the selected contaminants, k is the sequence number of the k^{th} vertex which represents the k^{th} contaminant, θ_k is the heading angle of the k^{th} vertex measured clockwise from true north (0 degrees), d_k is the distance between the vertex and the sample location, which is proportional to the contaminant concentration C_k. Again, to provide user control, *Factor* is the scaling factor chosen dependent upon the map size and allows recalibration due to the levels of the contaminant concentration.

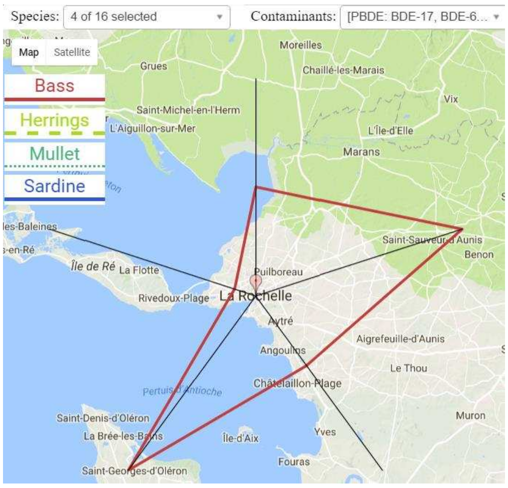

Fig.2 Illustration of the radiation pattern to display multiple species and contaminants simultaneously. The polygonal architecture is defined using equations (2) and (3).

2.2.2 Hierarchical Clustering analysis

Hierarchical Clustering (HC) is a typical algorithm to analyse the similarities (or dissimilarities) of objects in the variable space (Smoliński et al., 2002; Chen et al., 2005). To have a better understanding of the geographical distribution of the PBDE congeners, HC was employed to divide the fish samples into 3 clusters with the PBDE congener concentration as input variables to investigate if correlations exist between the species and spatial locations. This particular feature will enable assessors to investigate if certain locations or species are likely to have certain concentrations levels. The analysis was performed using R language with the Ward method as the amalgamation rule and Euclidean distance as metric.

3. Results & Discussion:

3.1 General comparison between different species

PBDEs were observed in all samples with most measured congeners being detected. The concentrations ranged from 87 ng/kg to 8907 ng/kg whole weight for the sum of all measured PBDE congeners (64 ng/kg to 8635 ng/kg for the ten EU listed PBDEs). A summary of the data are presented in Table 1. There are only minor differences between the average values for both the sum of the 23 congeners (PBBs included) and the sum of the 10 congeners specified in the

EC recommendations, which confirms a more informed choice of congeners for the EU list.

PBBs occurred less frequently showing a corresponding range of < 0.02 μg/kg to 0.97 μg/kg ww for the sum of seven PBB congeners.

Fig.3 shows the dependence of mean value for the sum of the PBDEs on the fish species, and the concentration distribution of the top five congeners, including BDE-47, BDE-49, BDE-100, BDE-99 and BDE-154, in different species. In terms of the sum of the PBDEs in each sample, Herring and Bass ranked at the top level with a mean value around 2100 ng/kg, Sprat, Mackerel and Mullet at the second with mean values around 1200 ng/kg, Sardine and Turbot at the lowest level with mean values around 400 ng/kg. The change in certain congener concentrations in the samples approximately agreed with the trend of the PBDEs sum, but in some cases the congeners could show quite different patterns. For example, BDE-99 in Sea Bass were as low as Sardine and Turbot, which suggests that the difference of the congener level in fish is also closely correlated with the congener types.

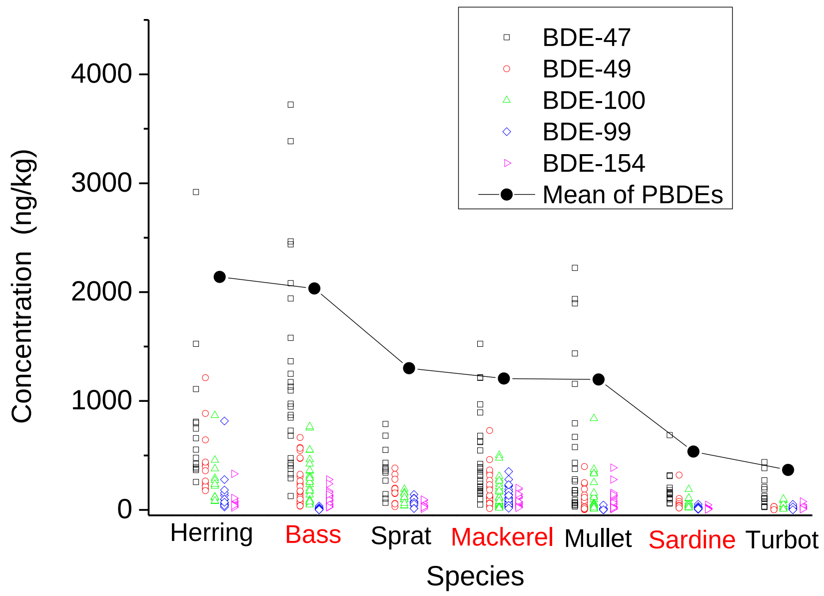

Fig.3 Dependence of mean value for the sum of PBDEs on fish species, and the concentration distribution of the top five congeners

Table 1 Summary of the detected PBDEs in the samples

PBDE Concentrations, ng/kg whole weight	Sardines (n=15)				Mackerel (n=27)				Herring (n=14)				Grey Mullet (n=25)			
	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX
Sum measured PBDEs	159	415	535	2206	169	894	1206	3636	808	1634	2140	8906	147	577	1198	5422
*Sum PBDEs (EU list)	133	388	497	2118	141	806	1096	3381	757	1553	2039	8635	77	555	1094	5360
	Sprat (n=12)				Sea Bass (n=25)				Turbot (n=13)				Others (n=4)			
	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX	MIN	MEDIAN	MEAN	MAX
Sum measured PBDEs	383	975	1301	4620	297	1806	2033	5747	87	300	367	859	100	141	169	293
*Sum PBDEs (EU list)	354	911	1236	4560	270	1736	1972	5641	64	268	332	792	78	119	146	268

*- Sum BDE-28, 47, 49, 99, 100, 138, 153, 154, 183 and 209 (EU recommendation 2014/118/EU)

Therefore, it could be expected that the geographical distribution of the congeners would be affected by both the fish species and the congener types. To avoid the mixing of the two variables, it is necessary to analyse the data in single species successively. However, it would be unpersuasive to draw a distribution pattern if the sample numbers were too small, so, only the species with more than 15 samples were analysed in Section 3.2, including Sea Bass, Herring, Mackerel, Mullet and Sardine. Clearly, if more data become available, confidence in the statistical calculations will increase. Therefore, as more data are accumulated they may be used to augment or update the current data provided the sampling conditions remain the same e.g. catch location.

3.2 Analysis of the PBDEs in single fish species

3.2.1 Sea Bass

Within the sample set, 25 Sea Bass samples were collected with the sum of the 23 detected congeners in individual samples varying between 297 to 5747 ng/kg with an average value of 2033 ng/kg as shown in Table 1. The concentration distribution of all the 23 congeners in Sea Bass is shown in Fig.4. The PBDE pattern was dominated by BDE-47, followed by BDE-100 > BDE-49 > BDE-154 > BDE-28, and these top 5 congeners represented 94.71% of the summed PBDEs. Typically, within these datasets, levels below the limit of detection (LOD) occur and so in utilising tools of this form, decisions regarding the representation of these values should be agreed.

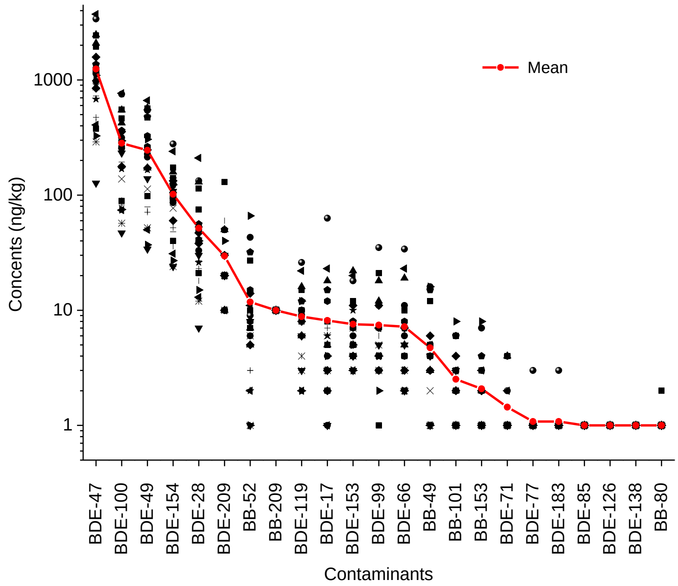

Fig.4 Concentration distribution of the detected congeners in Sea Bass.

For spatial or geographical analysis, BDE-47 was selected as a typical example. BDE-47 in Sea Bass, was measured at between 127 to 3722 ng/kg and exhibited significant geographical variation. As shown in Fig.5 (a), 4 of the top 6 concentration values occur where the waters of the English Channel and the North Sea meet, and the other two were located off the coasts of Wales and La Rochelle in France. Ten other congeners, including BDE-17, BDE-28, BDE-49, BDE-66, BDE-77, BDE-99, BDE-100, BDE-119, BDE-153 and BDE-154, exhibited a similar geographical distribution trend with BDE-47. Such distribution indicates that Sea Bass from the south-east marine region of the UK show higher PBDEs contamination when compared with other regions.

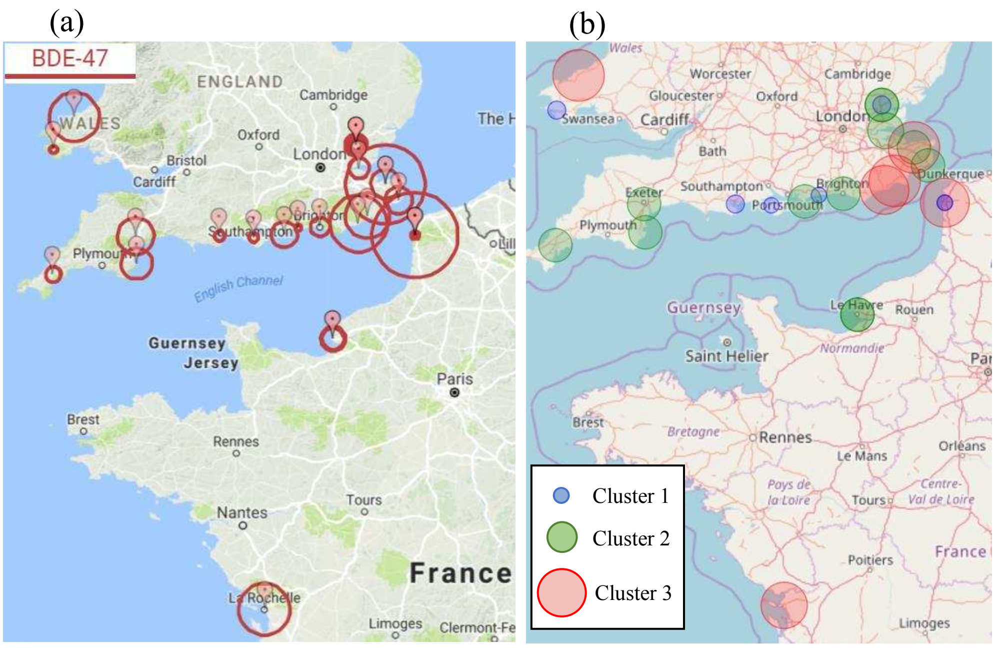

Fig.5 (a) Geographical distribution of BDE-47 in Sea Bass and (b) the Hierarchical Clustering results.

To confirm the distribution of the contaminants, HC analysis (Liang et al., 2011) was performed to divide all 25 samples into 3 clusters with their concentrations as input variables. The clustering results shown in Fig.5 (b) gave good agreement with the geographical distribution of the aforementioned congeners as BDE-47. The samples belonging to Cluster 3 located at the junction area of English Channel and the North Sea, Wales and La Rochelle, which contained higher concentration of contaminants, while Cluster 1 containing least contaminant levels were mainly located at the middle part of the south coast of the UK.

Furthermore, four of the congeners, including BDE-71, BDE-99, BB-49 and BB-52 showed no obvious regularity, and the other 8 congeners were at very low levels (most < LOD).

3.2.2 Herring

Of the 14 herring samples collected, the sum of the 23 detected congeners in single samples varied from 808 to 8906 ng/kg with an average value of 2140 ng/kg. To observe the different content levels of the congeners, the concentration distributions of all the 23 congeners were compared (shown in Fig.6) by arranging them in descending order of their mean value.

Similar with Sea Bass, BDE-47 was also at the highest level in herring, followed by BDE-49 > BDE-100 > BDE-209 > BDE-99, these top 5 congeners represented 88.27% of the summed PBDEs. Such difference of the congeners could also be intuitively observed by presenting their concentrations with rings in map at the same scale level (see Fig.7 (a)).

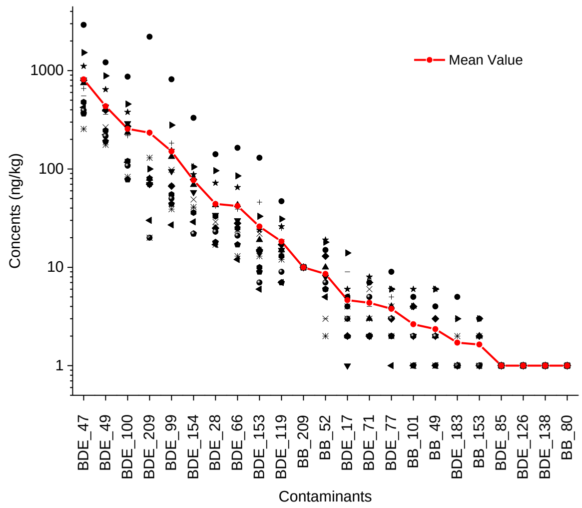

Fig.6 Concentration distribution of the detected congeners in 14 Herring samples.

Fig.7 (a) also reveals that herring samples from the central part of UK, i.e., Mersey, showed higher levels of most of the PBDE congeners. HC results as shown in Fig.7 (b) agreed well with this conclusion, only samples from Mersey were recognized as Cluster 3, which contained highest level of contaminants. All samples from the south-east part of UK belonged to Cluster 2 and samples from the north part showed no clear pattern. In Fig. 7(a) it is shown using the web page how mutiple contaminants within single species can be effectively displayed across the spatial region of interest.

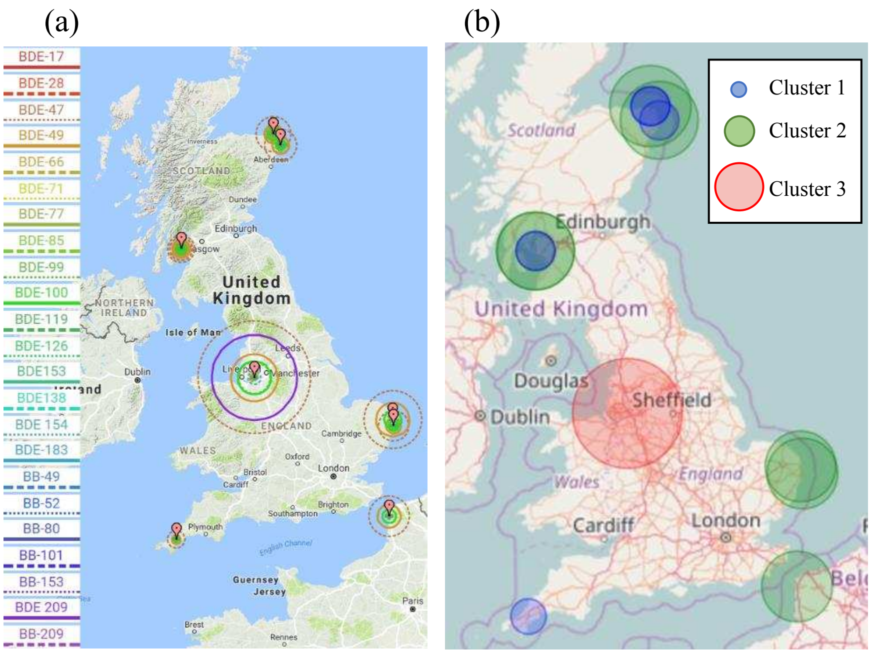

Fig.7 (a) Geographical distribution of detected congeners in Herring and (b) the Hierarchical Clustering results

3.2.3 Mackerel

27 Mackerel samples were collected in the location extending to Norway in the north and Algarve in the south. The sum of all detected congeners in individual samples ranged from 169 to 3636 ng/kg with an average value of 1206 ng/kg. As expected, BDE-47 was still the congener responsible for the highest level of contamination as it showed the largest average concentration. BDE-47 alone represented 38.57% of the summed contaminants, followed by BDE-49, BDE-100, BDE-99 and BDE-154, and the top 5 congeners represented 80.66% of the summed PBDEs.

Geographically, the north-east region of UK (mainly around Peterhead) and the English Channel area showed higher concentration of contaminants including BDE-17, BDE-28, BDE-47, BDE-49, BDE-66, BDE-99, BDE-100, BDE-119, BDE-153, BDE-154, BDE-209 and BB-52. The remaining congeners showed very low concentrations (< LOD) in Mackerel. HC results identified samples from Norway and Spain were recognized as Cluster 1, which contained the lowest level of contaminants.

3.2.4 Mullet

The 25 mullet samples were collected mainly from the south coast of UK and surrounding coastal area of France. The sum of all detected congeners in individual samples ranged from 147 to 5422 ng/kg with an average value of 1198 ng/kg. Similar with the previously described species, the top 5 congeners were BDE-47, BDE-100, BDE-209, BDE-154 and BDE-29 (See Fig.10), which represented 87.89% of the summed PBDEs. Eight of the congeners, including BDE-71, BDE-183, BDE-77, BDE-85, BDE-126, BDE-138, BB-80 and BB-209, showed concentration lower than LOD in most of the samples, which would not be considered for further analysis.

Geographically, it was found that the BDE compounds and the PBB compounds exhibited quite different distribution patterns. Samples from the coastal area of UK tended to show much higher levels of BDE compounds when compared with France, whereas the PBB compounds in samples from UK all showed concentrations lower than LOD, and all the samples with a concentration higher than the LOD came from the coast of France. This confirmed a trend observed in other studies (Fernandes et al., 2008), which may reflect a higher utilisation of PBBs in France relative to the UK.

3.2.5 Sardine

The Sardine samples with a total number of 15 were collected mainly from the coastal areas around the English Channel. The sum of all detected congeners in individual samples ranged from 159 to 2206 ng/kg with an average value of 535 ng/kg. The top 5 congeners were BDE-47, BDE-209, BDE-49, BDE-100 and BDE-99, which represented 87.14% of the summed PBDEs. Thirteen of the congeners, including BDE-17, BB-52, BDE-71, BDE-77, BB-49, BDE-85, BDE-126, BDE-138, BDE-183, BB-80, BB-101, BB-153 and BB-209, showed concentrations lower than LOD in most of the samples, which would not be considered for further analysis.

In terms of the geographical distribution, samples from Poole in the UK showed higher concentration of PBDEs compared with other locations. However, the other regions showed no clear distribution patterns. It should be emphasised that this approach, although offering quantitative information does not address wider issues e.g. sampling levels and behavioural feeding patterns or ranges of species. Thus, if particular trends are observed a more detailed analyses should be conducted to establish causative factors and/or minimise sources of uncertainties.

3.2.6 Summary

As seen from the above analysis, BDE-47 was the congener of highest concentration in all species, and was always followed by congeners as, BDE-49, BDE-99, BDE-100, BDE-154 and BDE-209 (orders could be different), which agrees well with previous reports (Brown et al., 2006; Colombo et al., 2011). However, the BB compounds and several of the BDE compounds such as BDE-71, BDE-77, BDE-85, BDE-126, BDE-183 and BDE-138 always showed much lower concentrations than other congeners. For the PBDEs, this finding is consistent with the lower occurrence of these congeners in commercial mixtures (perhaps apart for BDE-183 which forms part of the deca-BDE product) Geographically, the distribution pattern of the PBDEs in different species varied significantly, even though the different congeners in the same species always followed a similar pattern.

It can generally be concluded that BDE-47, BDE-49 and BDE-100 were of much higher concentration than other congeners in most of the samples, and the coast around the English Channel shows higher PBDEs concentration levels when compared with other areas. Both these conclusions had been confirmed by the analysis in Section 3.1 and 3.2, and agreed with a previous report (Johansson et al., 2006), which reflects the influence of urban and industrial sources in these coastal areas.

3.4 Conclusions

This study has presented the use of an interactive webpage facility to rapidly evaluate complex environmental contaminant datasets which may comprise of multiple factors, e.g. congeners, species and sample locations. Qualitative assessment of correlations between the location and species with congeners is demonstrated which will enable more targeted detailed analyses when considering these complex multifactor datasets. As an example we have characterised PBDE and PBB occurrence in five commonly consumed fish species, (Sea Bass, Herring, Mackerel, Mullet and Sardine) taken from marine waters around the UK and from other North Atlantic fishing areas from which retail fish in the UK is commonly sourced. PBDEs were observed in all samples with the majority of measured congeners being detected. The concentrations ranged from 0.087 µg/kg to 8.907 µg/kg whole weight (ww) for the sum of all measured PBDE congeners. PBBs occurred less frequently showing a corresponding range of < 0.02 µg/kg to 0.97 µg/kg ww for the sum of seven PBB congeners. Generally, BDE-47 was the congener of highest concentration in all species, and was always followed by congeners as, BDE-49, BDE-99, BDE-100, BDE-154 and BDE-209 (orders could be different), the BB compounds and several of the BDE compounds such as BDE-71, BDE-77, BDE-85, BDE-126, BDE-183 and BDE-138 always showed much lower concentrations than other congeners. The web-based resource provides a flexible and efficient tool for assessors and policy-makers to monitor and evaluate levels within caught fish species and enables evidenced-based decisions to be justified. Spatial variation, species and congener correlations with clustering can be achieved offering a useful investigative resource to explore the relationships within large and complex datasets. Results show that the spatial distribution of the PBDEs in different species varied significantly, but the different congeners in the same species always followed a similar pattern. The coast area around the English Channel shows higher PBDEs concentration levels when compared with other areas. With the inclusion of additional data, confidence levels of suggested trends may be improved. Furthermore, this system could be readily enhanced to include consumption data and enable predictions of exposure estimates for comparison with UK or European tolerable daily intake (TDI) levels. The levels and frequency of PBDE occurrences make it prudent to continue the monitoring of these commonly consumed marine fish species from the point of view of public health, as well as the status of the marine environment. This approach may be more readily applied to land-based contaminant datasets and enable effective intervention monitoring.

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