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Distribution of Flame-Retardant Compounds Across Different Trophic Levels in Coastal Lagoons

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Abstract: Plastics, textiles and electronic products emit flame retardants, which were deposited in coastal lagoons; sediment retention was favoured by a reduced water exchange. This investigation brings together concentrations of polybrominated diphenyl ethers (PBDEs), halogenated norbornenes and organophosphate flame retardants (OPFRs) in water, sediment, and organisms from lagoon food webs, estuarine, and marine food webs, and establishes a consistent sampling, mass-spectrometric, and stable-isotope approach for lagoon application. The lipid-normalised concentration and trophic magnification factors (TMFs) were used to assess trophic transfer. The concentrations of halogenated flame retardants in Bizerte Lagoon sediments were not detected to 51.8 ng/g dry weight, while the concentration of PBDEs in sea urchins were between 56.9 ng/g lipid weight. The total fish burden ranged from 137 ± 168 to 5755 ± 2679 ng/g lipid weight, with legacy OPFRs ranging from 55.9 ± 27.5 ng/L in seawater to 102 ± 69.8 ng/g dry weight in sediment in a tropical estuary. Total OPFRs decreased from phytoplankton (922 ng/g dry weight) to fish (58.2 ng/g dry weight), while TPHP (TMF 1.55) and EHDPHP (TMF 3.61) increased, and TCP, TEHP and iDDPHP decreased (TMF 0.505–0.519). This trophic transfer is thus specific to the chemical and depends upon hydrophobicity and biotransformation. Sediments were found to be long-term reservoirs; and reported hazard quotients for seafood consumption were well below 1. A multi-trophic, water and sediment monitoring system is recommended for lagoon management.

Keywords: Flame Retardants; Coastal Lagoons; Pbdes; Opfrs; Bioaccumulation; Trophic Magnification; Sediment Contamination.

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I. Introduction

Flaming materials, such as plastics, textiles, electronic goods, furniture, and building products have added flame retardants to slow combustion or delay ignition. Several formulations of brominated products were limited, leading to a growth of the use of alternatives containing phosphorus, estimated to have represented approximately 20% of the European consumption of flame retardants in 2006 (Van der Veen & de Boer, 2012). Organophosphate compounds are found in the environment as a result of their leaching from products and discharge from wastewater, and are ubiquitous in air, water, sediment and biota (Wei et al., 2015).

Coastal lagoons (CLs) are shallow, semi-enclosed marine areas with limited connections to the ocean. The high rate of retention is favoured by limited circulation, high sedimentation, organic rich sediments, and continuous urban, industrial and agricultural inputs. The highest contamination rates were found in urban areas, such as the shoreline in the city and enclosed water bodies, among European aquaculture sites (Aznar-Alemany et al., 2018). In Bizerte Lagoon, Tunisia, the levels of OPFRs in sediments (9.77–164 ng/g dry weight) were also higher than those of the halogenated flame retardants (3.30–28.5 ng/g dry weight) and were significantly correlated with TOC, while the latter were not (Mekni et al., 2020).

Legacy polybrominated diphenyl ethers (PBDEs) and organophosphate flame retardants (OPFRs) have been widely reported in marine, estuarine and lagoon environments in previous studies. Restricted exchange and high retention for organic C in semi-enclosed coastal systems have been shown in early studies to provide a permanent reservoir of hydrophobic halogenated compounds in the sediment, while polar organophosphates are highly dissolved and bioavailable in the water column. For example, estuarine and

lake food web studies showed that, while total summed OPFR burdens often show trophic dilution from primary producers to apex consumers, individual OPFs show compound-specific bioaccumulation that is dependent on the lipophilicity, protein binding in the organism, and transformation rates within the organism.

Transfer via food chains is compound-specific. A large number of phosphorus flame retardants were an order of magnitude higher in the Western Scheldt estuary than the brominated analogues, while the majority of the PBDEs were amplified (except BDE-209) and most of the phosphorus compounds were trophically diluted (Brandsma et al., 2015). Trophic magnification factors (TMFs), calculated by regressing log concentration on trophic level, are the preferred measure of food-web magnification and are supplemented by bioaccumulation factors (BACFs) and biota–sediment accumulation factors (BACFs) (Borgå et al., 2012; Conder et al., 2012). In a tropical South China Sea food web, the total OPFRs decreased with the trophic level, and increased tripentyl phosphate was observed in an unexpected manner (Ding et al., 2020). Biotransformation was reflected in the biomagnification of TMFs for coral reef fish but not in the Pearl River estuary, except for metabolite enrichment at high trophic levels for TPHP (Wang et al., 2022; Bekele et al., 2019).

Aquatic toxicological studies during the recent past use the stable nitrogen isotope (δ^15 "N") profiling along with trophic magnification factors (TMFs) to quantify this food-web transfer objectively. Previous surveys conducted in other temperate and tropical systems like the Western Scheldt estuary, Taihu Lake and South China Sea showed that most OPFRs show a decrease in concentration over tropho level (TMF<1), but some constituents like triphenyl phosphate (TPHP) and 2-ethylhexyl diphenyl phosphate (EHDPHP) consistently biomagnify (TMF>1.5-3.6). In addition, parallel research into alternative compounds and naturally occurring halogenated natural products (e.g., marine invertebrates containing methoxylated PBDEs) demonstrated that biological exposure does not necessarily mean anthropogenic industrial exposure, and therefore there is a need for multi-compartment, integrated monitoring frameworks.

This systematic analysis of flame-retardant occurrence within coastal lagoon compartments indicates that there is significant contamination variability depending on compartment and compound type. The hydrophobicity of polybrominated diphenyl ethers (PBDEs) causes these compounds to bind to sediment, thereby creating an environmental sink, while the polar OPFRs are soluble in and available in the water column. This food web relationship exhibits a dual character; while the sum of all OPFR contaminants is diluted from the base trophic level to the apex predator through the food web, some specific OPFR compounds, such as TPHP and EHDPHP, biomagnify with trophic magnification factors greater than one. This is dictated by compound hydrophobicity, lipid availability, and biotransformation by organisms. Therefore, water-only sampling significantly understates the actual levels of contamination within lagoons. In order to manage contamination in aquatic ecosystems and seafood safety, management regimes must implement multiple-compartment testing methods.

Integrated datasets covering water, sediment, primary producers, invertebrates and fish within one lagoon remain scarce. The research questions are:

RQ 1: Which PBDE and OPFR compounds dominate water and sediment.

RQ 2: How concentrations differ among species and trophic levels.

RQ 3: Which compounds bioaccumulate or magnify.

RQ 4: Which implications arise for ecosystems and seafood consumers.

This paper contributes by integrating concentrations of PBDEs, halogenated norbornenes, and OPFRs across water, sediments, and multi-trophic food webs, while defining a reproducible sampling, mass-spectrometric, and stable-isotope protocol for coastal lagoons. It uniquely demonstrates that while summed OPFR burdens undergo trophic dilution, individual compounds like TPHP and EHDPHP biomagnify, highlighting the need for comprehensive, multi-compartment monitoring.

The paper is divided into four sections. Section I (Introduction) summarizes the use of flame retardants, susceptibility of lagoons, and research questions. Section II methods, Section III (Results) includes water, sediment, organism profiles and TMFs. Lastly, there are Section IV Discussion - which examines distribution in the compartments and ecological risks, and Section V for the conclusion and monitoring recommendations.

II. Materials and Methods

A data-synthesis design was used along with a standardised lagoon protocol. o new field measurements were made, and concentrations were compiled from Bizerte Lagoon (Tunisia), Nandu River Estuary (Hainan Island, China), the South China Sea, Taihu Lake and Bohai Sea bays. The lagoon reference is Bizerte Lagoon, a very industrialised Mediterranean lagoon. The three categories of sites are defined by the protocol: Site A (industrial or port influence), Site B (urban drainage influence) and Site C (reference). The height of the site, depth, salinity and temperature are measured at each sampling location.

The study adopts a data-synthesis design coupled with a standardized protocol across industrial, urban, and reference lagoon sites. The data were compiled from various aquatic ecosystems without generating new field measurements. Surface water, sediments, and four trophic categories of biota were systematically monitored across wet and dry seasons. Target analytes encompassed PBDEs, halogenated norbornenes, and legacy or emerging OPFRs. Sample preparation involved solid-phase extraction, ultrasonic or pressurized liquid extraction, and lipid removal, followed by quantification via GC-MS/MS and LC-MS/MS under strict quality control. Finally, stable nitrogen isotopes determined trophic levels to calculate BAF, BSAF, and TMF.

The surface water (0.5 m depth, 1–2 L, three replications) is collected in pre-cleaned, amber glass bottles, filtered and divided into dissolved and particle fractions. Surface sediment (0–5 cm) is collected with a stainless-steel grab, frozen at −20 °C, freeze-dried and sieved; organic carbon, grain size and moisture are measured. Biota include phytoplankton and algae, zooplankton and bivalves, shrimp, crab and small fish, and predatory fish, all of which are in four trophic categories. A minimum of three replicates per species/site is recorded length, weight, feeding behaviour and lipid content. Samples are taken twice a year, in wet and dry seasons.

Target compounds comprise BDE-28, -47, -99, -100, -153, -154 and -209; legacy OPFRs (TEP, TCEP, TCPP, TDCPP, TPHP, TNBP, TBOEP, TCP, EHDPHP, TEHP); emerging OPFRs (RDP, BDP, iDDPHP, TDtBPP, V6); and emerging halogenated compounds (DBDPE, HBB, PBEB, norbornenes). Solid phase extraction of water. Tissue and sediment (5 g) are spiked with isotope labelled surrogates then extracted with hexane–dichloromethane and cleaned on silica or Florisil columns, with lipids removed from tissue extracts. For PBDEs, quantification is based on GC-MS/MS and for OPFRs, quantification is based on LC-MS/MS. Quality control includes solvent, method and field blanks, matrix spikes, duplicates and calibration standards. Reported surrogate recoveries ranged from 49.6% to 138%, spike recoveries ranged from 65% to 127%, and method detection limits ranged from 0.002 to 0.873 ng/g/lipid weight for estuarine OPFR methods. Non-detects are NOT set to zero, but are treated using censored-data methods.

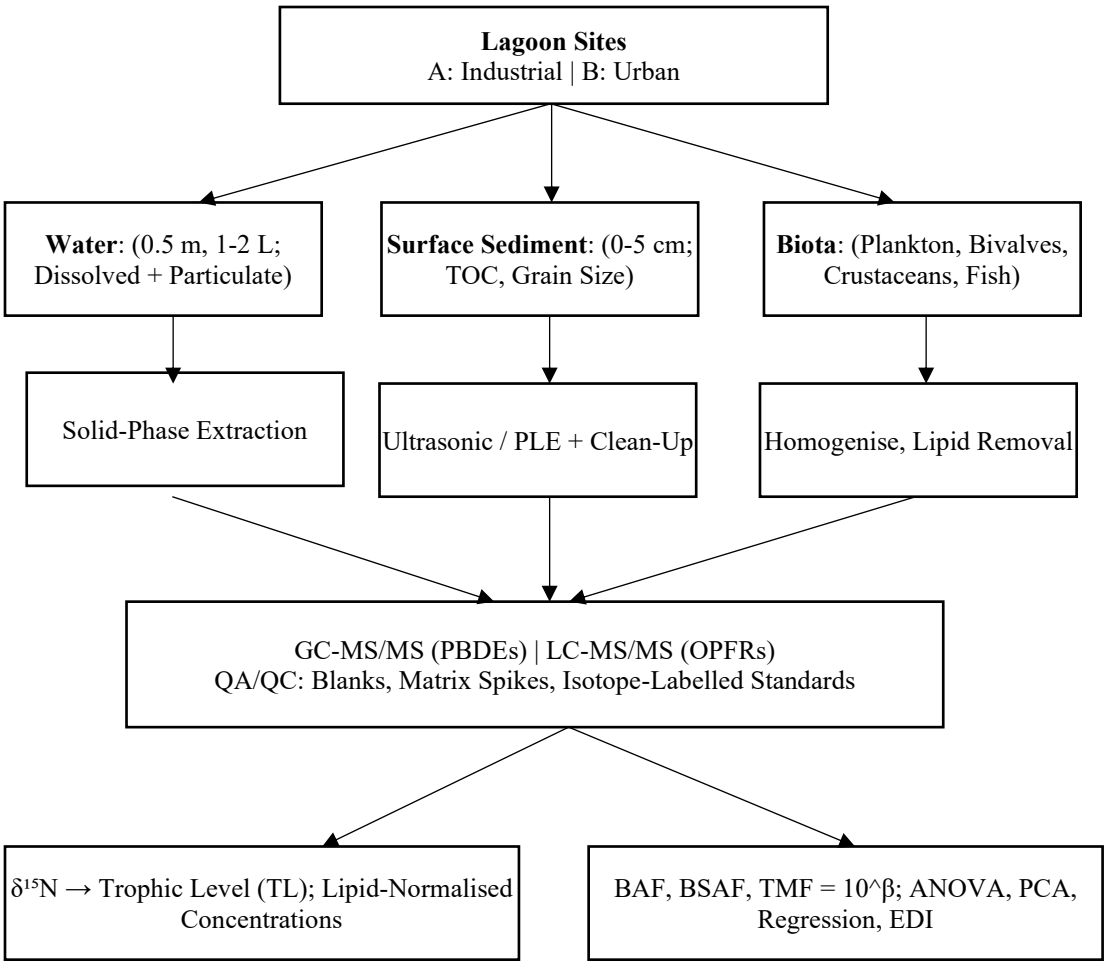

Figure 1: Methodological Workflow

Figure 1 links three lagoon sites (industrial, urban and reference) to parallel sampling of water, surface sediment and biota. All matrices are treated with solid phase extraction, ultrasonic or pressurised liquid extraction or tissue homogenisation with lipid extraction. The number of extracts is quantified for PBDEs and OPFRs using blank and spike and isotope labelled standard control in GC-MS/MS and LC-MS/MS, respectively. Nitrogen isotope ratios define trophic level, supporting calculation of bioaccumulation factors, biota–sediment accumulation factors and trophic magnification factors, followed by multivariate and regression statistics and mathematical formulations are defined in equation (1) – (3).

Trophic level (TL) is estimated from stable nitrogen isotopes using a trophic enrichment of 3.4‰:

$$\mathrm{TL} = \frac{\delta^{15}\mathrm{N}_{\mathrm{consumer}} - \delta^{15}\mathrm{N}_{\mathrm{baseline}}}{3.4} + \mathrm{TL}_{\mathrm{baseline}} \qquad (1)$$

Bioaccumulation (BAF) and biota–sediment accumulation (BSAF) factors relate lipid-normalised biota concentrations to water and organic-carbon-normalised sediment concentrations, respectively:

$$\mathrm{BAF} = \frac{C_{\mathrm{biota}}}{C_{\mathrm{water}}}; \quad \mathrm{BSAF} = \frac{C_{\mathrm{biota}}}{C_{\mathrm{sediment}}} \qquad (2)$$

The TMF follows from the regression slope β of log concentration against TL:

$$\log_{10} C = \alpha + \beta(\mathrm{TL}); \quad \mathrm{TMF} = 10^{\beta} \qquad (3)$$

TMF > 1 indicates magnification and TMF < 1 indicates dilution, judged by slope significance and confidence intervals. Human exposure is estimated as EDI $= \frac{C \times \mathrm{IR}}{\mathrm{BW}}$, with C the edible-tissue concentration, IR the ingestion rate, and BW body weight. Statistics include descriptive measures, ANOVA among sites and species, principal component analysis for source patterns, Spearman correlation with organic carbon, and Monte Carlo simulation of TMF uncertainty.

III. Results

The concentration of halogenated FRs ranged from not detected to 51.8 ng/g dry weight in sediments from Bizerte Lagoon, while no concentrations of methoxylated PBDEs were detected in sediment. In the Nandu River Estuary, the mean concentration of legacy OPFRs in seawater was 55.9 ± 27.5 ng/L and the mean concentration of emerging OPFRs in sediment was 350 ± 283 ng/g dry weight, both of which were dominated by TDtBPP. Different hydrophobicity with EHDPHP and TEHP being the most prevalent compounds in seawater and TEP and TDCPP in sediment.

Table 1: Reported Concentrations in Environmental and Biological Matrices

Ecosystem	Matrix	Compound Group	Concentration
Bizerte Lagoon	Sediment	Halogenated FRs	ND–51.8 ng/g dw
Bizerte Lagoon	Sea Urchin	PBDEs	3.67–56.9 ng/g lw
Bizerte Lagoon	Sea Urchin	Halogenated Norbornenes	4.52–116 ng/g lw
Bizerte Lagoon	Sea Urchin	MeO-PBDEs	ND–364 ng/g lw
Nandu Estuary	Seawater	Legacy OPFRs	55.9 ± 27.5 ng/L
Nandu Estuary	Seawater	Emerging OPFRs	309 ± 109 ng/L
Nandu Estuary	Sediment	Legacy OPFRs	102 ± 69.8 ng/g dw
Nandu Estuary	Sediment	Emerging OPFRs	350 ± 283 ng/g dw

ND, not detected; dw, dry weight; lw, lipid weight.

Table 1 summarizes data reported from a Mediterranean lagoon and a tropical estuary and its biota. Sediments and sea urchins were collected in Bizerte Lagoon for the analysis of halogenated compounds while seawater and sediments were collected in estuarine waters for the analysis of OPFRs. Legacy OPFRs were surpassed by emerging OPFRs (led by TDtBPP) in seawater and sediment. Concentration bases vary between rows (dry weight, lipid weight and volume), and the values represent the occurrence within each concentration base, and should not be ranked across ecosystems or compound groups.

The profiles of chemical compounds were specific to the species. TBOEP was the most abundant among estuarine fish, for Epinephelus fasciatus (4721 ± 2317 ng/g lipid weight), while TPHP was most abundant in Siganus canaliculatus (475 ± 192 ng/g lipid weight). Emerging RDP was found at 287 ± 139 ng/g lipid weight in Stolephorus chinensis and 247 ± 65.8 ng/g lipid weight in Leiognathus brevirostris. However, naturally occurring MeO-PBDEs in Bizerte Lagoon were higher than anthropogenic PBDEs in sea urchins, which means that sea urchin pollutants cannot be entirely blamed on anthropogenic pollutants.

Figure 2a shows total OPFR concentrations falling from phytoplankton (922 ng/g dry weight) to fish (58.2 ng/g dry weight) across six organism groups of a tropical marine food web, indicating trophic dilution of summed burdens. Figure 2b shows large interspecific differences among estuarine fish, from 137 ± 168 ng/g lipid weight in Acanthopagrus latus to 5755 ± 2679 ng/g lipid weight in Epinephelus fasciatus. Concentration alone does not demonstrate magnification, because lipid content, habitat and metabolism also influence burdens.

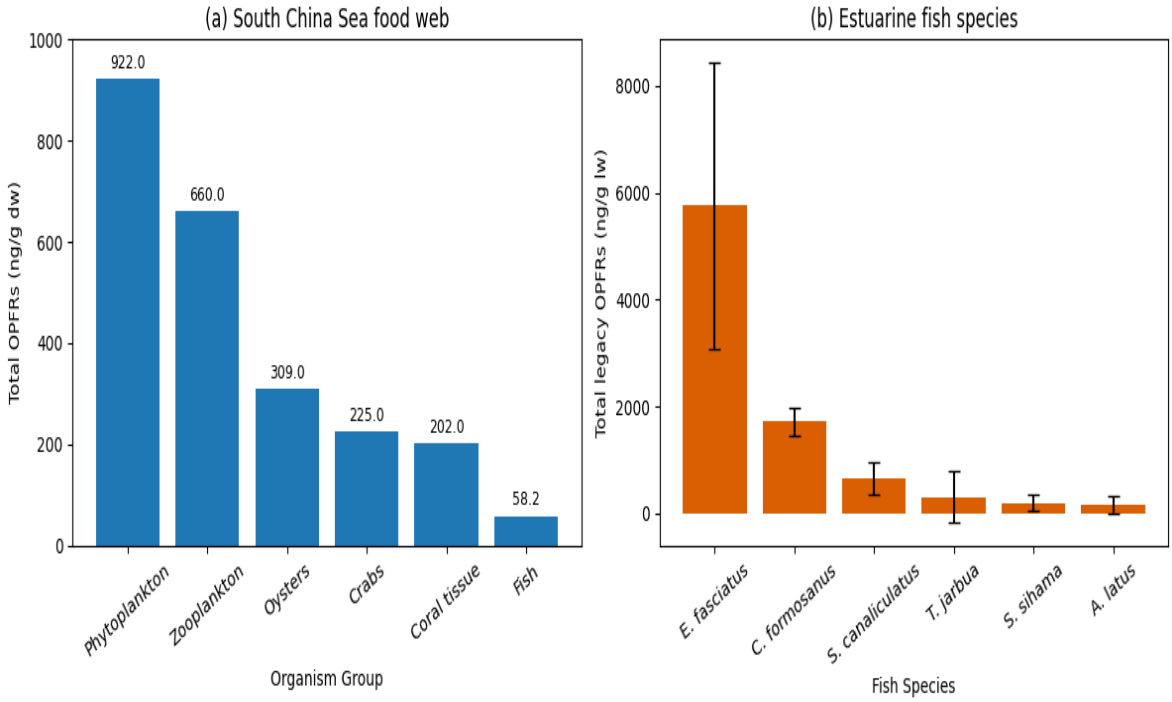

Figure 2: Total OPFR Concentrations Across Organism Groups And Fish Species (a) South China Sea Food Web (b) Estuarine Fish Species

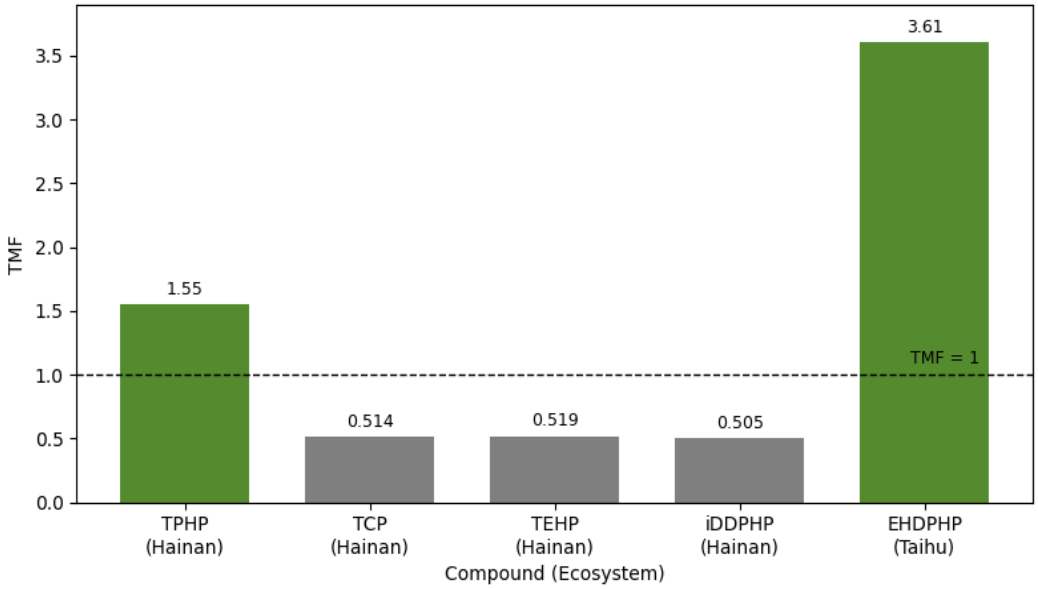

Figure 3: Reported Trophic Magnification Factors

Figure 3 shows a comparison of reported TMFs, the dashed line indicating no net trophic change. TPHP (1.55) and EHDPHP (3.61) were both greater than 1, which means they were magnified in a tropical estuary and in Taihu Lake, respectively. The values of TCP, TEHP and iDDPHP were set around 0.5, which shows the biodilution. These differences are found in the same food web and show that the trophic behaviour is not exclusively dependent on the food class, but also on the actual chemical properties and biotransformation of the organisms. Values come from various ecosystems, and should be compared with caution.

Table 2: Trophic Magnification Factors and Interpretation

Ecosystem	Compound	TMF	Interpretation
Nandu Estuary	TPHP	1.55	Magnification
Nandu Estuary	TCP	0.514	Dilution
Nandu Estuary	TEHP	0.519	Dilution tendency
Nandu Estuary	iDDPHP	0.505	Dilution
Taihu Lake	EHDPHP	3.61	Magnification
Western Scheldt (benthic)	TBOEP / TCIPP / TCEP	3.5 / 2.2 / 2.6	Tentative magnification
Western Scheldt	PBDEs except BDE-209	> 1	Magnification
Bohai Sea	Most OPFRs	< 1	Dilution

Table 2 lists TMFs and their interpretation for OPFRs and PBDEs. For the Western Scheldt, the following were reported: Magnification for TPHP, tentative benthic values for TBOEP, TCIPP and TCEP, and for all PBDEs except BDE-209. Most OPFRs and TCP, TEHP were characterised by dilution in the Bohai Sea. A TMF greater than 1 implies that there is a statistically significant positive regression slope between log concentration and trophozoits; the tentative values should be verified using a larger data set.

The OPFR concentrations of surface water in three bays of the Bohai Sea were different, Laizhou Bay had the highest value of 255.8±36.44" ng/L" " followed by the Bohai Bay and the Liaodong Bay. The mean tissue burden of organisms in Laizhou Bay was also the highest at 70.56±61.36" ng/g" wet weight. Additionally, the seasonal changes in the concentration of OPFRs in the Pearl River estuary are different from the changes in sediment concentration which reflect a long-term equilibrium. The seasonal changes in the lagoon have not yet been measured and, in this regard, represent an important part of the new monitoring protocol standardized.

IV. Discussion

Hydrophobic PBDEs have a strong affinity to particulate matter and organic carbon, and can be stored in sediments for long periods of time in the environment, while polar OPFRs such as TCEP and TEP can remain in solution and be bioavailable in the water column. Chemical partitioning is compound specific: the significant correlation between OPFRs and organic carbon in Bizerte Lagoon and the uniform dominance of TDtBPP in both seawater and sediment. Thus, water-only monitoring is an underestimate of the contaminant loads that exist within coastal lagoons.

The observed decrease in summed OPFR concentrations from phytoplankton through fish is comparable with that seen in the Western Scheldt and in the Bohai Sea. However, the biomagnification of TPHP and EHDPHP is similar to the results obtained from the Pearl River estuary and Taihu Lake. This opposition can be explained by biotransformation into polar metabolites, chemical hydrophobicity, lipid distribution, growth dilution and dietary habits. Biomagnification can only be concluded with rigorous regression analysis across trophic levels, and does not provide direct evidence of biomagnification when concentrations are high in predators.

Potential sources for contamination include industrial effluents, domestic wastewater, urban run-off and plastic dust, port activities and electronic waste degradation. To determine the difference between these sources, spatial gradients, compound concentration ratios, and principal component are used. Possible biological effects include oxidative stress, endocrine disruption and developmental toxicity; these are not necessarily indicative of toxicity. Hazard quotients < 1 for South China residents and safe Bizerte sea-urchin consumption suggest low current exposure risk, but this is only applicable to the specific exposure scenario(s) that were formally evaluated.

V. Conclusion

All examined lagoon, estuarine and marine compartments contain flame retardants, but their presence is not even distributed throughout the tropho-levels. The presence of hydrophobic halogenated compounds in the sediments has been observed with concentrations up to 51.8 ng/g dry weight in Sediments of the Bizerte lagoon and concentrations up to 55.9 ± 27.5 ng/L for legacy compounds in a tropical estuary for polar OPFRs where these compounds are available in water column. Although the total OPFR burdens decreased from phytoplankton to fish, the total concentrations of TPHP (TMF 1.55), EHDPHP (TMF 3.61), and tentative benthic TMFs for TBOEP, TCIPP and TCEP were greater than one, indicating compound-specific magnification. Much more concern is given to TPHP, EHDPHP, sediment-associated PBDEs, and carnivorous/benthic-feeding fish (e.g. Epinephelus fasciatus) with 5755 ± 2679 ng/g lw of legacy OPFRs. Reported hazard quotients less than 1 do not suggest that the populations sampled are at high risk at present, but are not necessarily applicable to other lagoons. Monitoring priorities are the simultaneous measurement of PBDEs and emerging OPFRs in water, sediment, plankton, invertebrates and fish of the same lagoon, repeated by seasons, along with the measurement of trophic positions using δ15N and metabolites. Management should focus on controlling wastewater, urban runoff and port discharges, along with regular surveys of sediment and exposure to seafoods to ensure lagoon ecosystems and human health are maintained. Further study is recommended on the emerging flame retardants and their metabolites in aquatic compartments (water, sediment, multi-trophic biota) simultaneously monitored within the same lagoon, based on the paper provided. Furthermore, it should combine stable nitrogen isotope analysis with in-depth exposure assessment to aid in a more comprehensive assessment of food-web transfer and possible human health implications.

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