Organic UV filters in marine environments: an update of analytical methodologies, occurrence and distribution

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Abstract

Ultraviolet filters (UV Filters) are compounds that are widely employed in personal care products such as sunscreens to protect the skin from sun damage, but they are also added to other products, such as food packaging, plastics, paints, textiles, detergents, etc. The continuous use of these products causes the release of a substantial amount of these products into the marine environment through direct input or wastewater discharge, and thus they are becoming an important class of contaminants of emerging concern. A correlation between their occurrence and different negative effects on marine biota has been reported.

Taking into account all the possible impacts on the environment, knowledge of their presence and distribution in the different compartments of the ecosystems, ranging from waters and sediments to aquatic organisms, which potentially suffer from bioaccumulation and biomagnification processes, is essential. High concentrations of ultraviolet filters have been found in samples collected from across the entire planet, even in polar regions, revealing their global distribution.

Therefore, interest in the sensitive determination of ultraviolet filters in several marine matrices has increased. In this article, an overall review of the more recently reported analytical chemistry methods for identifying and quantifying these compounds in marine environmental samples is presented. We compare and discuss the potential advantages and disadvantages of every step involved in the analytical procedure, including the pre-treatment, treatment and extraction processes that are required to avoid matrix effects. Moreover, we describe the worldwide occurrence and distribution of those most important UV filters.

Keywords: organic ultraviolet filters, personal care products, sunscreens, analytical methodologies, marine environment occurrence

List of abbreviations:

4-MBC, 4-methylbenzylidene camphor

Ac, acetone

ACN, acetonitrile

APCI, atmospheric pressure chemical ionisation

APGC, atmospheric pressure gas chromatography

ASE, accelerated solvent extractor

BAME, bar adsorptive microextraction

BP-3, benzophenone-3

BP-4, benzophenone-4

BSTFA, N,O-bis(trimethylsilyl) trifluoroacetamide

CHL, chloroform

d.w., dry weight

DCM, dichloromethane

DLLME, dispersive liquid-liquid microextraction

DTS, drometrizole trisiloxane

EA, ethyl acetate

EHS, 2-ethylhexyl salicylate

ESI, electrospray ionisation

FPSE, fabric phase sorptive extraction

GC, gas chromatography

Hex, hexane

HCl, hydrochloric acid

HMS, homosalate

HRMS, high resolution mass spectrometry

HS, headspace

IL-MSA-DLLME, on-line *in-syringe* magnetic stirring assisted ionic liquid dispersive liquid-liquid microextraction

IL-SDME, ionic liquid-based single-drop microextraction

IL, ionic liquid

IMC, isoamyl p-methoxycinnamate

iSAME, in-situ suspended aggregate microextraction

l.w., lipid weight

LC, liquid chromatography

LD, liquid desorption

LLE, liquid-liquid extraction

LOD, limit of detection

LOQ, limit of quantification

MAE, microwave-assisted extraction

MBP, methylene bis- benzotriazolyl tetramethylbutylphenol

MeOH, methanol

MNPs-based dSPE, magnetic nanoparticles dispersive solid-phase extraction;

MS, mass spectrometry

MSA-DLLME, magnetic stirring assisted dispersive liquid-liquid microextraction

USAEME, ultrasound assisted emulsification microextraction;

MSPD, matrix solid-phase dispersion

OC, octocrylene

OD-PABA, ethylhexyl dimethyl para-aminobenzoic acid

OMC, ethylhexyl methoxycinnamate

IPA, isopropanol

PCPs, personal care products

PDSA, terephthalylidene dicamphor sulfonic acid

PHWE, pressurised hot water extraction

PLE, pressurised liquid extraction

QTOF, quadrupole time-flight

QuEChERS, Quick, Easy, Cheap, Effective, Rugged and Safe technique

SBDLME, stir-bar dispersive liquid microextraction

SBSDME, stir-bar sorptive-dispersive microextraction

SBSE, stir-bar sorptive extraction

SPE, solid-phase extraction

SPLE, selective pressurised liquid extraction

SPME, solid-phase microextraction

TD, thermal desorption

TPs, transformation products

UHPLC: ultra-high performance liquid chromatography

USE, ultrasonic extraction

USSPME, ultrasonic extraction followed by SPME

VE, vortex extraction

w.w., wet weight

Contents

1. Introduction

Short-term exposure to ultraviolet (UV) radiation exerts positive effects on human health, *i.e.*, it facilitates D vitamin synthesis. However, excess exposure to solar radiation can cause numerous harmful effects, such as sunburns and skin cancers [1]. Additionally, the human tendency to spend more time in the sun is attributed to growth of the solar protection industry [2]. Since the late nineteenth century, several formulas containing organic and inorganic UV filters have been developed to protect the skin from UV damage [3].

Currently, the European legislation set the maximum allowed concentration for each UV filter in cosmetic products (Regulation no. 1223/2009 of the European Commission) [4]. The European Union (EU) allows the use of 27 UV filters in concentrations ranging from 2% to 15% [5], of which only two are inorganic (titanium dioxide and zinc oxide). Organic UV filters are the most popular and widely used filters in sunscreens and personal care products (PCPs) [6,7]. They frequently contain single or multiple aromatic structures attached to hydrophobic groups to improve their properties, with a limited absorption band spectrum. Therefore, different combinations are generated to obtain the desired protection against both regions of solar radiation: UVA (320-400 nm) and UVB (280-320 nm) [8]. Although the main use of these compounds is associated with PCPs, these compounds are also present in other industrial goods, such as food packaging, plastics, paints, textiles, products for vehicle maintenance to prevent polymer and pigment photodegradation [9], detergents and disinfection products [10]. The increasing production and use of organic UV filters has generated a new kind of environmental pollutants [11]. For example, ethylhexyl methoxycinnamate (OMC), a common UV filter, was included in 2015 in the Watch List under the Environmental Quality Standards Directive [12], although it was removed in 2018 due to the few data reported in sediments [13].

Organic UV filters are classified based on their structure according to the chemical family and main physicochemical properties, as shown as in Table 1. Most exhibit some of the typical characteristics of priority organic pollutants [14]. The common feature is the presence of an aromatic moiety with a side chain displaying different grades of saturation [15], high lipophilicity and stability against biotic degradation [16]. These physicochemical properties determine the fate of organic UV filters in the environment, and it is a relevant issue when choosing an appropriate analytical method for determining the levels of these filters in the different matrices. K_{ow} provides information about their distribution. Compounds with K_{ow} values <1 are considered hydrophilic, while compounds with values >4 are hydrophobic. Analytes with K_{ow} >8 are not considered readily bioavailable and compounds with values >10 are considered not bioavailable at all [17]. Solubility (S) also provides information about the likely distribution of compounds between the different environmental compartments, particularly the soil/sediment and water. The majority of organic UV filters are slightly soluble, as shown in Table 1.

Organic UV filters follow two major pathways to enter the environment: direct input from human activities through wash off from skin and clothing during recreational activities, and indirect entry through industrial discharges, wastewater effluents, runoffs and domestic uses

[9,18,19]. When the compounds are released into sewage and reach the wastewater treatment plants, some might be transported to the sludge due to their high lipophilicity and poor biodegradability [20]. This sludge may be destined for landfills or used in agriculture, which potentially pollutes underground water [9]. A fraction of treated wastewater containing organic UV filters will be discharged into natural water supplies [21] are potentially retained in sediments [22] or bioaccumulated in biota [3,9,23].

Moreover, some organic UV filters undergo photodegradation upon exposure to UV radiation or biodegradation in environmental matrices; accordingly, they are often not detected because they are degraded into transformation products (TPs) [15,24]. Nevertheless, scarce information is available on UV degradation in marine environments [25].

Different negative effects of organic UV filters on marine ecosystem have been described. For example, these filters significantly increase the viral abundance in marine bacterioplankton through prophage induction, and they also modify the carbon, nitrogen and phosphorous biogeochemical cycle in seawater [26]. Furthermore, hard corals exposed to organic UV filters suffer rapid and complete coral bleaching, even at extremely low concentrations. Additionally, the toxic effects of benzophenone-3 (BP-3), OMC and 4-methylbenzylidene camphor (4-MBC) on marine organisms at three different trophic levels were reported and, show similar toxicity to copper, mercury, cadmium lead and zinc [27].

The presence of organic UV filters must be analysed to determine their impact on the environment and their possible deleterious on human health [28]. Due to their low concentrations in environmental samples and the appearance of matrix effects, different methods have been developed to analyse several families of organic UV filters in a variety of matrices, including marine samples such as seawater [29], sediments [30] and biota [10].

The aims of this review are describe and compare, the available information about the analytical procedures for the extraction and determination of the concentrations of organic UV filters in different marine matrices (seawater, sediments and biota) in the last ten years (2008-2018). Although this review encompasses global studies, only the compounds allowed in the European Union for which determination methods are described in the literature have been included. Similarly, the present work provides a broad overview of the occurrence and distribution of these compounds in different marine compartments, which highlights their extensive use.

2. Analytical procedures for detecting organic UV filters in the marine environment

The concentration of organic UV filters measured in the marine environment are very low and these filters are encapsulated in complex matrices. Thus, suitable preparation techniques must be applied to the samples to isolate and preconcentrate the analytes prior to their determination. Tables 2, 3 and 4 summarize the procedures for the extraction and determination of organic UV filters in marine samples.

2.1. Sample extraction

The employed extraction methods depend on the properties of the matrix and the analytes. The extraction techniques used for different marine environmental compartments are summarised in the next sections.

2.1.1. Sampling and pre-treatment

The determination of this kind of contaminants presents several issues. Sample contamination is a common risk due to the occurrence of organic UV filters in different products. For this reason several authors suggest that the analyst avoid the use of PCPs that contain organic UV filters, before [31–33] and during the sampling [10,33] and in the laboratory [34–36]. Due to the lipophilic behaviour of these compounds, they are easily transferred to the glass (adsorption problems), for that, organic modifiers are added to the bottle that contain the sample [37,38].

Analysis of environmental samples frequently requires a pre-treatment, depending on the subsequent extraction technique and the type of matrix.

For liquid samples, the collected samples are stored in an amber glass bottles [10,39–41], previously washed with an organic solvent [10,42,43] or in a certified clean amber bottle [31]. Acidification [10,40,41,44] and filtration [10,27,39,40,42,45] are also common pre-treatments. When the water is filtered, the fraction resulting should also be analysed because a fraction of the target analytes is likely adsorbed into the particles and their concentration might be underestimated [46]. Furthermore, unfiltered seawater has also been used in some studies [44,47]. If the samples are not analysed immediately, they must be kept refrigerated [29,40,42,45] in the dark [29,43] and in amber glass bottles [42,48,49] to prevent photodegradation [29]. For seawater, a continuous sampling process has also been reported. This procedure consists on a semipermeable membrane devices (SPMDs), where the sample passed through a membrane and the analytes are retained [50]. This device is attached to the boat in order to sample during a cruise route and it not requires pre-treatment.

In the case of sediments, the samples are often taken with a stainless steel grab [51,52]. The possible pre-treatments include homogenization [35,51,53], air dried [51,53] or dried at high temperature [36,54,55] and sieved using different size fractions [51,53,54]. Other pre-treatments are frozen [35,51,52], freeze-dried [35,52,56] and ground into powder [52]. This kind of samples are stored in aluminium boxes [51] or amber glass containers [36,54] in the dark [51,55] and dry environment [51], at the freezer [36,53,54], or in a dessicator in the dark at the freezer [57].

For biota samples, it is important maintain it fresh during the transportation [58,59], and then measure their length and weight [59,60]. Different pre-treatments are carried out depending on the species, such as skin removed, bone removed, peeled, dissection, deshelled, etc. [58,60–64]. The homogenization could be done in wet and then freeze-dried [58,59] or homogenized after the lyophilisation [60,62,64–68]. Normally the samples are grounded into powder [58,64,67,68] and stored in aluminium foils [60,65] or amber container [65] at the freezer [61,62]. Other pre-treatments are homogenization in wet and dry at high temperatures [10], or homogenized and extracted in wet [61].

Regarding the quality assurance, different criteria related with repeatability, sensitivity and extraction efficiency must be taken into account. Inter-day or intra-day repeatability, expressed as relative standard derivation (RSD) performed for 3 or more replicates has been reported with acceptable ranges from 0.2% [69] to below 20% [43,46,51,59,70–73]. Other quality criteria are also employed, such as blanks below the limits of detection (LODs) [32] or less than 1% [52]. In relation with the sensitivity, the LODs are determined as three times the standard deviation of blank peaks areas [32,58,70]. The extraction efficiency, reported as recoveries are carried out with spiked samples with a known concentration [51] or with surrogate standards [51,53,55,59].

2.1.2. Seawater

Several extraction techniques have been employed for measuring organic UV filters in seawater, although solid-phase extraction (SPE) is the most frequently used, as summarized in Table 2. SPE is simple and easy to perform; nevertheless, its main disadvantage is related to the occasional consumption of a high sample volume, sometimes up to 1 L [74]. C_{18} SPE cartridges are commonly employed because they retain a large amount of organic analytes. Recoveries ranging from 80% to 113% were obtained during the extraction of a mixture of BP-3, 4-MBC, octocrylene (OC), OMC, ethylhexyl dimethyl para-aminobenzoic acid (OD-PABA), 2-ethylhexyl salicylate (EHS) and homosalate (HMS) [45].

However, SPE is not adequate for the extraction of polar analytes [50,75]. New polymeric reverse phase sorbents that extract diverse analytes with distinct properties are used for this process [50]. Due to the relatively polar characteristics of the majority of organic UV filters, methanol (MeOH) is the solvent that is most frequently used as the eluent [44]. Other more polar solvents have also been used; for example, a mixture of dichloromethane (DCM) and ethyl acetate (EA) has been used and achieved recoveries ranging from 79% to 110% [40].

On-line SPE is an automated version of the conventional SPE procedure that presents advantages such as better reproducibility and reduced sample preparation, minimizing sample contamination. The study by Montesdeoca-Esponda et al. [39] is the only one to apply this technique in the extraction of methylene bis-benzotriazolyl tetramethylbutylphenol (MBP), obtaining similar recoveries (61-66%) to traditional SPE (60-61%). However, the LODs achieved using on-line SPE were lower (1.1 $ng \cdot L^{-1}$) than conventional SPE (6.2 $ng \cdot L^{-1}$) due to a higher preconcentration factor.

Another variant of SPE, the magnetic nanoparticles dispersive solid-phase extraction (MNPs-based dSPE), has also been employed to extract some UV compounds [48]. This technique consists of a SPE mediated by nanoparticles that are released into the sample. The advantages are its application in a wide pH range, it is a matrix independent method, it requires less time for the extraction (5 min) and it reduces the solvent volumes required, 3 mL [48], compared to traditional SPE, 30 mL [76]. The MNPs-based dSPE technique using hexane was successfully applied to quantify BP-3, isoamyl p-methoxycinnamate (IMC), 4-MBC, OC, OMC, OD-PABA, EHS and HMS, obtaining good recoveries (70-128%) [48].

Microextraction techniques have also been used to extract organic UV filters. Specifically, solid-phase microextraction (SPME) by headspace (HS) was used. The main advantages of this technique are the reduction of the matrix effect and the reuse of the fibres. It does not require the use of organic solvents for thermal desorption (TD), integrating the extraction and preconcentration in one step. It is also faster than conventional SPE [70]. Good recoveries (85-102%) were obtained for the extraction of a group of nine organic UV filters using the *in-vial* SPME technique [16]. This technique consists of the addition of acetic anhydride (acetylation) to the sample prior to extraction for the derivatization of the polar compounds. The main advantage of this methodology is the low cost of the derivatization process. Acetylation and extraction are performed in the same vial, reducing the analyte loss and sample contamination, and less time is required than conventional derivatization, reducing the overall analysis time. The obtained LODs using *in-vial* SPME [16] are lower (0.060-0.84 $ng \cdot L^{-1}$) than conventional SPME (0.068-12 $ng \cdot L^{-1}$) [70] (Table 2).

Stir-bar sorptive extraction (SBSE) combined with liquid desorption (LD) [49], which is compatible with liquid chromatography (LC), was employed to extract BP-3, OC, OMC, OD-PABA, EHS and HMS, with recoveries ranging from 64 to 85% [43]. Compared to conventional SPME, this technique has a better sensitivity for BP-3, OC and EHS because of its higher surface contact area [77]. Nevertheless, it requires a long analysis time, as 5 hours are required in some cases [43,77], which is the main disadvantage compared to SPME and *in-vial* SPME that require 10 min [16,70].

Different devices have been employed to solve the disadvantages of SBSE. Stir-bar dispersive liquid microextraction (SBDLME) uses a stir bar coated with a magnetic ionic liquid. It requires less time (10 min) and presents better recoveries (91-117%) [78] than SBSE (time, 3-5 hours [49,77]; recoveries, 18.4-100% [37,43,49]). Stir-bar sorptive-dispersive microextraction (SBSDME) uses a stir bar that is coated with a hydrophobic magnetic nanosorbent; compared to SBSE, the extraction time is shorter (25-30 min) [71,79] and it presents better recoveries (83-120%) [71,79,80]. Nonetheless, the extraction time is longer than SBDLME [78]. Bar adsorptive microextraction (BAME) uses a bar coated with an appropriate powdered sorbent subjected to an ultrasonic treatment [81]. Compared to traditional SBSE, it provides the possibility of selecting the most suitable sorbent for the target analytes. Better recoveries were obtained for the extraction of BP-3 (76.6-98.4%) [82] than SBSE (27.6%) [38].

Liquid-liquid extraction (LLE) is a traditional technique used to extract different contaminants from environmental samples [83]. The disadvantage of this technique is the use of large amounts of toxic solvents and the requirement of a post-extraction treatment, such as extract filtration [84] or solvent evaporation [85]. However, the sample volume used (200-500 mL) is comparable other techniques, such as SPE [42,76]. This process was used to extract four UV filter (4-MBC, OMC, OD-PABA and EHS) obtaining recoveries between 89 to 120%

Dispersive liquid-liquid microextraction (DLLME) relies on the relative solubility of analytes in two different immiscible liquids. In this method, a small volume of extractant solvent is dispersed by the action of a second solvent. DLLME uses a small volume of organic solvent compared to SPE (60 µL [86] and 6 mL [74], respectively); it has a low cost of implementation, is normally fast and decreases the matrix effect. The recoveries obtained for BP-3, IMC, 4-MBC, OC, OMC, OD-PABA, EHS and HMS mixture are equivalent to the recoveries obtained with other extraction processes, such as SPE [29,45], MNPs-based dSPE [48], LLE [87] and SPME [70].

In ionic liquid-based single-drop microextraction (IL-SDME), the extractant is an ionic liquid (IL) with organic salts as the acceptor phase [88]. In this technique, the use of organic solvents is minimized or eliminated, and therefore it is inexpensive, simple, fast, precise and sensitive compared with conventional LLE and SPE. Good recoveries (92-99%) have been obtained using a small sample volume (20 mL) of 1-hexyl-3-methylimidazolium hexafluorophosphate ([C_6MIM][PF_6]) as the IL for the extraction of six organic UV filters with an extraction time of 37 min [89]. The recoveries reported are comparable to SPE (Table 2).

On-line *in-syringe* ionic liquid magnetic stirring-assisted dispersive liquid-liquid microextraction (on-line *in-syringe* IL-MSA-DLLME) is a novel technique that is conducted inside an automated syringe containing a magnetic stir bar for the homogeneous mixing of the sample and dispersion of the extractant (*i.e.*, IL). The extract is aspirated and pushed into the detector [90]. This technique is environmentally friendly because it avoids the use of chlorinated solvents, minimizes waste generation, uses less sample volume (3.5 mL) and improves the analysis throughput [72]. Nevertheless, the recoveries obtained for BP-3, 4-MBC, OC and OD-PABA are lower (48-92%) [72] than the values reported using the IL-SDME technique (92-107%) [89].

During *in-syringe* magnetic stirring-assisted dispersive liquid-liquid microextraction (*in-syringe* MSA-DLLME), a derivatisation agent such as N,O-bis(trimethylsilyl) trifluoroacetamide (BSTFA) is added prior to the gas chromatography (GC) analysis. The advantage of this technique is that the extraction, derivatization and preconcentration performed at the same time, reducing the overall analysis time (6 min for each sample) [34]. It present better recoveries (88.4-111%) [34] than on-line i*n-syringe* IL-MSA-DLLME (46-92%) [72].

Other microextraction techniques have been also employed to measure organic UV filters. Ultrasound-assisted emulsification microextraction (USAEME) relies on the emulsification of the organic extractant in the sample by ultrasound and centrifugation. Comparable recoveries were achieved (75.5-108%) [69] compared to SPE (Table 2). In contrast to SPE, no significant matrix effect has been reported and hence the quantification is performed using conventional external calibration, presenting an advantage in the extraction of organic UV filters from complex matrices.

A new microextraction technique based on the use of a flexible and permeable substance coated with a sorbent chemically bonded to its surface, fabric-phase sorptive extraction (FPSE), was used for MBP, obtaining recoveries ranging from 32.4 to 51.4% [73]. The advantages of this technique are the low consumption of organic solvents, high surface area for the sorbent-analyte interaction, stability in a wide range of pH values and fast back extraction with a small solvent volume. When comparing the sample volume, FPSE usually uses less sample volume and organic solvent than SBSE. Furthermore, FPSE presents better recoveries than SBSE (18.4-19.9%) [37]. Nevertheless, SPE and on-line SPE result in higher recoveries, 60-61% and 61-66%, respectively [39].

In-situ suspended aggregate microextraction (iSAME) relies on the formation of a supramolecular aggregate phase, which is formed in the sample with a cationic surfactant. Afterwards, it is filtered to collect the aggregate and the elution performed by adding an organic solvent to dissolve the aggregate. The principal advantage of this technique is simple the requirement for fewer for its development; it is also simple, fast and precise [85].

Water samples have also been measured with an indirect analysis, using SPMDs that are based on a thin, lay flat tube composed of semipermeable polyethylene membranes. The devices are mounted and then exposed to the medium subject to analysis. This method is used to integrate *in situ* concentrations of more lipophilic compounds. The extraction step is performed using dialysis, and the solvents are generally cyclopentane or hexane. This technique is passive sample uptake and has been applied to extract BP-3, E-OMC, Z-OMC and 4-MBC during a raft expedition crossing the Pacific, expressing the concentration in pg·SPMD^{-1} [50].

Some of the new techniques have been applied only once to extract organic UV filters from seawater (Table 2), but resulted in satisfactory recoveries. The techniques that use IL or specific sorbents present the disadvantage that most reagents are not commercially available, thus they must be synthesize.

2.1.3. Sediments

Less information is available about techniques for extracting organic UV filters from sediments than from water (Table 3).

Traditional techniques, such as the Soxhlet technique have been used to extract organic UV filters from sediments [52,91]. Recoveries ranging from 70 to 90% have been achieved in the case of 4-MBC, OC and OMC, but its main disadvantages are the use of large quantities of organic solvents and the time required for the extraction (hours).

Microwave-assisted extraction (MAE) [51,54] and ultrasonic extraction (USE) [35,53] have been also used for the extraction of organic UV filters. In MAE, the extraction is faster and consumes less solvent [92], due to the use of microwaves to heat the solvent. In the USE technique, the clean-up step is performed at the same time as the extraction, and the method is efficient and selective; nonetheless, some matrix effects have been observed. For example, the extraction time for BP-3 was shorter (5 min) and better recoveries (98.3-115%) were obtained [36] than the process using MAE (time, 30 min; recovery, 80%) [54].

A similar extraction technique that uses a small volume of organic solvent (1-5 mL) is vortex extraction (VE) [32,36,55]. In this case, a lower volume of an appropriate organic solvent is added to the sample, and the extraction is performed with vortex agitation followed by centrifugation. After extraction, an additional step is usually required (SPE [32] and DLLME [55]) for clean-up or preconcentration. Recoveries of 58-76% were achieved for the extraction of BP-3, 4-MBC, OC, OMC and OD-PABA using 2 mL of Ac and n-hexane [32], a lower volume than employed in the USE technique for the same compounds (8 mL) [35].

Pressurised liquid extraction (PLE) is the most frequently used extraction technique for measuring organic UV filters (Table 3). The clean-up step is frequently performed at the same time as extraction (in-cell clean-up) by adding sorbents such as alumina, copper or primary secondary amine in the extraction cell [61,93]. Additionally, these compounds facilitate the removal of matrix interferences. Depending on the mixture of the target analytes, different solvents are employed, and DCM is the typical solvent [93–95]. The PLE technique is also performed using an accelerated solvent extractor (ASE) system that automatically extracts the target analytes from the sediment samples [57,93–95]. The use of the PLE-ASE technique is increasing due to the reduced time (10-45 min) [30,57,93–96] compared to traditional techniques. It also increases the efficiency of extraction by operating at high temperature and pressure. In this way, only 15 minutes are needed to extract BP-3, obtaining recoveries raging from 70-100% [93], whereas MAE requires 30 min [54]. In addition, the LODs are very appropriate with the detection of trace contaminants (Table 3).

Selective pressurised liquid extraction (SPLE) is a variation of PLE that consists of incorporating matrix compound retainers into the extraction cell [97] using the ASE system. Compared to PLE-ASE, the addition of the hydromatrix increases the solvent flow through the ASE cell. Good recoveries were achieved (81-102%) when this technique was applied to the extraction of BP-3, OC, OMC and OD-PABA using an intermediate polarity mixture of hexane and DCM [61].

Pintado-Herrera et al. [38] applied a PLE modification known as pressurised hot water extraction (PHWE) that uses water at high temperatures (100-374.1ºC), which modifies its properties to resemble another solvent, to extract compounds with low and medium polarity and semi-volatile organic compounds from solid samples. The main advantage is the minimal use of organic solvents [98]. Organic solvents such as DCM, EA and hexane are employed in In SPLE and PLE-ASE (Table 3), while a mixture of water and MeOH (10%) are used in PHWE obtaining recoveries of 13.5 and 22.4% for the extraction of BP-3 and OC, respectively [38].

A technique that is typically applied to liquid samples, SPME, was used for first the time to extract organic UV filters from beach sand. The process consists of placing the sample in a vial containing Milli-Q water and sealing the vial. Then, the vial is submerged in a water bath with magnetic stirring. After equilibration, the SPME technique is performed using HS and the desorption is conducted using TD. The main advantage of this technique is that it avoids the use of organic solvents (environmentally friendly), requires a shorter extraction time and provides high sample throughput. Recoveries ranging from 70 to 124% were achieved during the extraction of BP-3, IMC, 4-MBC, OC, OMC, OD-PABA, EHS and HMS [99], consistent with other extraction techniques, such as Soxhlet [52] and MAE [51].

Another technique used for liquid samples, SBSDME, was also applied to beach sand. The application of this technique starts with the placement of the sample in a vial containing stir bar and water, which acts as the dispersion medium. The sample is stirred to extract the analytes and then the stir bar is subjected to LD. This technique reduces the sample manipulation and the use of organic solvent (150 µL) compared with the MAE (30 mL) [51] and USE (1 mL) [36] techniques. The LODs and recoveries (between 91-111%) [100] were consistent with the values obtained using other techniques, such as Soxhlet [52] and SPME [99].

Vila et al. [36] employed a combination of two techniques for extraction: ultrasonic extraction followed by SPME (USSPME). First, the USE technique was applied to the sample, and the extract (diluted with water) was placed in a vial containing a stir bar. Then, the SPME technique was performed using HS and the desorption was performed using TD. Because this technique reduced the interference from the matrix, external calibration was possible. During SPME, the extraction and preconcentration occur in a single step. In addition, the same authors compared the effectiveness of three different methods (USE, VE and USSPME) in extracting BP-3, IMC, 4-MBC, OC, OMC, OD-PABA, EHS and HMS. Higher recoveries were found using the VE technique, obtaining at the same time the lowest limits of quantification, proving to be most sensitive technique in determining the levels of organic UV filters (Table 3).

2.1.4. Biota

The most common extraction technique for marine biological samples is PLE using an ASE system (Table 4). This technique has been applied to determine organic UV filters in different biota samples, such as dolphin liver [65], clam tissues [10], fish fillets [10,59] and prawn tissues [58].

However, other techniques that have also been used to extract these compounds from sediment samples have been employed for the extraction of biota samples, shown in Tables 3 and 4. SPLE-ASE was employed in the wet extraction of BP-3, OC, OMC, and OD-PABA from the soft tissues of fish, achieving recoveries ranging from 51-85% [61]. This study is the only report to present extraction from wet samples; moreover, the compounds were extracted from a combination of all soft tissues (muscle, stomach, intestines and liver).

The USE technique was used to analyse 4-MBC, OC, OMC and OD-PABA in dolphin liver, with recoveries ranging from 60 to 115% [60]. Furthermore, the USE and PLE-ASE techniques both present similar recoveries (98.8-115.6% for USE [62] and 94.6-113.1% for PLE-ASE [58]) for OC and OMC extraction, from fish fillet.

A single study has reported the extraction and determination of organic UV filters in corals [32]. The VE technique was used to extract BP-3, 4-MBC, OC, OMC and OD-PABA from coral tissues, obtaining recoveries ranging from 61 to 86%.

MAE technique was also applied to extract OC, OMC and OD-PABA from mussels (soft tissues). Better recoveries were obtained (89-116%) [64] using a mixture of acetone (Ac) and heptane, and the values were comparable to the recoveries reported using the PLE-ASE technique (80-110%) for the same biota matrix and compounds with an EA and DCM mixture [58].

The Quick, Easy, Cheap, Effective, Rugged and Safe technique (QuEChERS) was employed to extract OC, OMC and OD-PABA from mussel samples [67], obtaining good recoveries (90-126%) using acetonitrile (ACN) and water as extractants. The main advantage of this technique is that it is flexible and selective. It is also simple, effective and uses a small amount of solvent compared to the PLE-ASE [10,58] and USE [60,62,66] techniques. However, it presents a low enrichment factor, which can be solved by the use of other extraction technique, such as DLLME.

Matrix solid-phase dispersion (MSPD) extraction was also used for the extraction of organic UV filters in biota matrices. In this technique, the sample is placed in a mortar with the bonded phase and blended. Then, the mixture is placed in an appropriate cartridge, depending on the analytes to be determined, and a solvent is added for extraction [101]. The extraction and clean-up steps are integrated in a single step; thus, it is fast, simple and uses a small volume of solvents (5-7 mL) [63,102] compared to the USE (60 mL) [62], PLE-ASE (25 mL) [58] and QuEChERS (10 mL) [103,104] techniques for fish fillet extraction. Moreover, the study by Tsai et al. [102] is the only report to present recoveries for each species studied (striped bass, cod and salmon), providing additional accuracy in the study.

After comparing the four techniques employed for the extraction of OC and OMC from mussels (QuEChERS, PLE-ASE, MSPD and MAE), QuEChERS presents better recoveries for both compounds (Table 4). MAE and MSPD obtained comparable results, whereas PLE-ASE presents lower recoveries.

2.2. Detection and determination

Chromatographic techniques are usually employed to determine the levels of organic UV filters because they are selective to both parental compounds and TPs, as many organic UV filters undergo different transformations processes. For polar and less volatile compounds, LC is preferred, while GC is chosen for volatile compounds (and their TPs) [105].

LC and ultra-high performance liquid chromatography (UHPLC) coupled with different detectors are used to measure the concentration of organic UV filters in cosmetics [106] and environmental samples, such as seawater, sediments and biota (Tables 2, 3 and 4). Diode-array detectors (DAD) or UV detectors are employed because they provide a fast analysis with good resolution [40,85,89,96]. However, mass spectrometry (MS) detection [31] or tandem MS/MS produce lower LODs [22,61,87]. The coupling with a quadrupole time-flight (QTOF) mass spectrometry detector provides accurate mass detection of the parent ions $[MH]^+$, and identifies and confirms their metabolites [27,61]

Due to the different characteristics of these analytes, they are able to be ionized by electrospray ionisation (ESI) and atmospheric pressure chemical ionisation (APCI) in positive or negative mode. The ESI mode is reported to display better sensitivity for identifying and quantifying compounds with medium to high polarity, while APCI presents better ionisation for compounds with low polarity [49,62]. ESI mode is the most frequently used mode because it provides an efficient ionization in a wide range of *m/z*. Nevertheless, it presents matrix effects [107], while APCI approach is less affected [107] due to the ionisation in the gas phase [49]. Nguyen et al. [49] reported a comparison between ESI and APCI approach for UV filters determination in seawater, in which APCI provided better sensitivity and reproducibility for BP-3, OC, OD-PABA, OMC, HMS and EHS.

GC is the most frequently used technique to determine the levels of organic UV filters in marine biota and sediments (Tables 3 and 4). Although GC coupled with MS or MS/MS detectors yields low LODs for the analysis of trace contaminants in environmental matrices [54], this approach presents an issue due to the low volatility and thermal stability of some organic UV filters, containing phenolic hydroxyl groups. Therefore, an additional derivatization step is needed [53] such as salicylates and benzophenones [16,34]. This step increases the volatility, reduces the polarity, prevents co-elution in complex matrices, and improves the reproducibility and sensitivity of the detection of polar compounds [16,53,108,109]. Different strategies, such as silylation, alkylation and esterification acylation, are the most frequently used derivatisation methods [86]. Agents such as N-(tert-butyldimethylsilyl)-N-methyltrifluoroacetamide, BSTFA and N-methyl-N-(trimethylsilyl)trifluoroacetamide are employed to improve the signal intensity and peak shape of some polar compounds [10,86,93]. Another type of derivatization, acetylation, is also used because it requires a shorter reaction time than silylation [16]. Some extraction techniques incorporate the derivatization step in the extraction process, resulting in an overall shorter analysis time [16,34]. Nonetheless, derivatization of the different matrix components can affect its precision and accuracy [107].

When the mixture of organic UV filters subject to analysis has a great range of different properties, LC coupled with a MS/MS detector is the main option [29,71]. Moreover, this technique is the most appropriated method for the simultaneous determination of the parental compounds and their TPs, which generally have a higher polarity than their parental compounds [107].

On the other hand, the matrix effect represents a potential problem for the quantitative determination of organic UV filters in environmental samples [110] using both GC and LC [107] because the coextracted matrix components may affect the analyte ionisation during MS detection [111]. However, the matrix effect in GC is not so critical [107] and it presents less matrix effect of coeluted lipids for thermally stable organic UV filters [110]. Even through LC presents higher matrix effect; this technique is the most frequently used in the organic UV filters determination in seawater (Table 2). Therefore, a highly sensitive and selective detection method to prevent a matrix effect is needed. However, in some cases, the matrix effect is unavoidable, and must be corrected [112] to prevent signal suppression or enhancement [113].

3. Occurrence in the marine environment

The global use of organic UV filters, 10000 tons annually [105], requires their frequent and recurrent detection in the marine environment. The next section presents the occurrence for those organic UV filters allowed in the EU for which extraction methodologies in seawater, sediments and biota samples collected around the world has been described in the literature. Detailed data are presented in the Supplementary Material (SM).

3.1. Seawater

A wide variety of organic UV filters has been detected in seawater samples collected around the world. Figure 1 illustrates the global occurrence and concentrations of the different organic UV filters. Detailed information is shown in the SM (Table S1).

Most of the samples are normally collected during the summer season, highlighting the importance of the direct input of these compounds, although high concentrations have also been detected in other seasons in areas near wastewater effluents [29,74]. Seasonal variations were observed in Hong Kong [24], Japan [84] and Korea [41], where lower concentrations were observed in winter (Table S1). In some cases, the UV filter concentration increased up to 4.4 times during the holiday season (June-August) compared to the pre-holiday period [41].

Fourteen of the twenty-five compounds allowed in the EU were detected in various countries; almost all the UV filter families (except triazines and benzylmalonate derivatives) have been detected. Based on the collected data, the most studies have been conducted in Spain, China and the USA. BP-3 is the most recurrent organic UV filter detected, probably because it is one of the most commonly used organic UV filters [86] and it is allowed in all countries [116]. It also has a slower photodegradation rate than other organic UV filters [25]. BP-3 appears in concentrations ranging from ng·L^{-1} to mg·L^{-1} (Table S1).

The highest BP-3 concentration (1.395 mg·L^{-1}) was found in USA at the Trunk Bay of St. John Island (USA Virgin Islands). Samples were collected from coral reef areas at approximately noon, when more than 180 swimmers were present in the bay at the time of sampling (the bay receives up to 2000 visitors per day [31]). In the same study, in Hawaii a high value was detected (19200 ng·L^{-1}), specifically in Maunalua Bay (Oahu, Hawaii, USA); sampling was performed in June in a public beach with over 500 swimmers per day in the peak of tourism season [31]. The second highest measured concentration was recorded in Spain (Galicia) (692000 ng·L^{-1}) in water samples collected during the summer season in different bathing areas [70].

Lower but significant concentrations of BP-3 were also detected in China (5429 ng·L^{-1}) in samples collected from a popular beach in Hong Kong during the summer season, as well as from the Victoria Harbour channel near to a wastewater effluent, which received 70% of discharges from the Hong Kong population. Those results represent the indirect and direct pathway of organic UV filters [29]. In Spain, a concentration of 3316.7 ng·L^{-1} was measured in a beach of Spain (Gran Canaria island), which is located close to various resorts and has an artificial barrier; therefore it is considered a semi-closed beach. Additionally, because of its good weather, the summer season lasts for most of the year in Gran Canaria island; thus, temporary fluctuations are less pronounced [40].

In South Carolina (USA), The Netherlands, Japan and Taiwan, BP-3 has also been frequently detected with average concentrations of 10-2013, 10-1540, 9-1258 and 18.8-1233 ng·L^{-1}, respectively. These values correspond to samples collected during the summer season at beaches where different recreational activities have been developed [33,44,45,87]. Other European countries, such as Norway, Slovenia and Portugal, present lower concentrations, 13-439.9, 96-380 and <300 ng·L^{-1}, respectively, although the samples were collected from beaches during the summer [54,74,82].

The second most frequently detected UV filter in seawater is OMC (Fig. 1). The highest concentrations were found in China (4043 ng·L^{-1}) [29], Spain (1200 ng·L^{-1}) [69] and Japan (1080 ng·L^{-1}) [84]. These high values also correspond to samples collected from beaches in the summer season (Table S1).

Another recurrent compound is OC, for which the maximum concentration (171000 ng·L^{-1}) was reported in bathing areas in Spain during the summer [69]. In Norway, a high concentration (7301 ng·L^{-1}) was also reported in samples collected from a crowded beach during the summer season, where the majority of users were children [74]. This compound was also detected in other areas, such as The Netherlands [33], China [29], USA [29,44,47], Spain [40,100], the Arctic [29], Italy [43] and Japan [29,45].

The predominant presence of these compounds corresponds with those organic UV filters (BP-3, OC, EHS, OMC and IMC) that are commonly added to PCP formulations [70,117] and frequently used during the summer season [77], when beaches tend to be crowded [78].

Two different studies were conducted in the same beach in Spain (Valencia) in summer using different extraction techniques (SBDLME and SBSDME) and comparable concentrations were obtained; the compound with the highest concentration was EHS in both cases [78,79]. Similarly, two studies were performed at Alicante (Spain) in the summer of 2009. The study by Tarazona et al. [86] reported a higher concentration of BP-3 (3300 ng·L^{-1}) than the study by Román et al. [48] (879 ng·L^{-1}); this discrepancy is potentially attributed to the dependence of the UV filter concentrations on the users (children or adults), the water tides, water renovation and other factors [78]. In Gran Canaria island (Spain), MBP was detected at two beaches, where

García-Guerra et al. [73] reported higher concentrations than those reported by Montesdeoca-Esponda et al. [39], probably due to visitor habits as tourists travel to these areas almost year-round [40].

The UV filter OD-PABA was measured in concentrations ranging from 0.03 to 1187 ng·L^{-1} in the Arctic [29], China [24,29,32], Japan [29,45], Spain [42,46,48,78,79,100] and USA [29,44,47]. However, this compound was not detected in Korea, probably because it is no longer used [41]. In addition, OD-PABA was the compound with the lowest concentration detected (5.8 ng·L^{-1}) in Gran Canaria island (Spain), likely; because this compound has been progressively excluded [40] for its potential photoallergic effect on humans [118]. However, it is a permitted organic UV filter in the EU [4].

Other less common organic UV filters have been also quantified in different places, such as the Arctic, China, Japan, Spain, USA, Korea, The Netherlands, Norway, Antarctic and Taiwan (Fig. 1). In the Antarctic Ocean waters, the reported concentrations are comparable to the levels detected in other parts of the world [10] (Table S1). The occurrence of organic UV filters in oceanic waters far away from coastal areas suggests their transport via ocean currents or the atmosphere [119].

3.2. Sediments

The sediment matrix constitutes a compartment that traps lipophilic compounds [51]. Thus, the sediments subject to wastewater discharges (submarine outfall) are a localised reservoir for these compounds, where the highest values are present at sites located close to the wastewater release and their concentrations decrease at greater distances [57,61]. Due to the limited light penetration, the photosensitive compounds are stably retained [51].

Limited information is available regarding the occurrence of organic UV filters in marine sediments compared with freshwater sediments [57]. Figure 2 presents the global occurrence and concentrations of organic UV filters in marine sediments; presenting more detailed information in the SM, Table S2.

Compounds with log K_{ow} between 4 and 7 show the potential to accumulate in sediments and the biota [120], similar to reports from different countries (Fig. 3A and 3B). Most of the organic UV filters present in this kind of matrix are considered hydrophobic (Table 1).

Much of the occurrence data has been reported in studies conducted in Spain and China (Table S2). The UV filter most frequently detected is OC, probably due to its highly lipophilic behaviour (log K_{ow}= 6.88) and its tendency to be absorbed in sediments or organic matter. The maximum concentration, expressed in dry weigh (d.w.), of this compound was reported in Spain (Gran Canaria island) (670 ng·g^{-1} d.w.), a crowded beach visited all year [86,99]. The second highest concentration of OC (551 ng·g^{-1} d.w.) was measured in two fishing harbours in China that are used for recreational activities and were built in semi-closed coastal regions that decrease water exchange [52].

Another recurrent compound is OMC (Fig. 2), for which the highest concentration was determined in China (456 ng·g^{-1} d.w.) in a fishing harbour [52]. With a log K_{ow}= 5.8, the adsorption of OMC by sediments is expected to contribute to its persistence against dilution effects [84].

BP-3 is also a recurrent compound that is present in sediments in different countries around the world [22,32,35,38,53–55,57,61,93–95,99,100]. Nevertheless, it is present at relatively low concentrations (0.05-47 $ng \cdot g^{-1}$ d.w.) due to its lower log K_{ow} (4.79), and thus it is less likely that other compounds to be found in sediments [93].

Although EHS is not one of the most frequently recurring compounds, it was detected at the second highest concentration in samples collected from beaches in Spain and Portugal during summer [55,99].

The highest concentration of OD-was detected in China (150 $ng \cdot g^{-1}$ d.w.) at Victoria Harbour that receives 70% of the wastewater from the total population in Hong Kong and Sai Kung (popular recreational area) [57]. In Spain, OD-PABA has been measured in concentrations 10.2 $ng \cdot g^{-1}$ d.w. as a consequence of recreational activities [55,100]. In other countries, such as Japan [57], Lebanon [51], the Northwest Pacific Ocean [30] and Norway [61], 0.8-13.9 $ng \cdot g^{-1}$ d.w. levels have been reported and are related to wastewater discharges from different origins (domestic, industrial and agricultural).

Two studies were conducted in different seasons (winter and summer) to analyse beach sand from a beach in Spain (Valencia), and similar concentrations of eight studied organic UV filters were observed in both seasons [55,100]. However, in two studies performed at Cadiz Bay (Spain), in different seasons, different concentrations were reported for BP-3 and OC. The concentrations reported for sediments collected in summer [38] were higher than sediments influenced by urban wastewater discharges in winter [94].

Sediments from the Oslo Fjord in Norway were also investigated, and the compounds BP-3, OC, OMC and OD-PABA were detected at concentrations of <5, <7-82.1, 8.5-16.4 and <4 $ng \cdot g^{-1}$ d.w., respectively. If we compare these results with the compounds reported in seawater samples collected from the same place, OD-PABA was not present in this matrix [61,74], potentially due to the higher degradation rate following exposure to natural radiation [121] and its hydrophobic behaviour (log K_{ow}=6.15) [17].

Two studies were performed along the Pearl River Estuary (China) and reported similar concentrations for 4-MBC, OC and OMC [35,95], because of the input from wastewaters. In addition, in the river outlet, another study reported lower concentrations [52]. Moreover, in Hong Kong (China), seasonal variations were reported between the wet and dry seasons [32]. During the wet season higher concentrations were detected for 4-MBC, OC, OMC, and OD-PABA due to the extensive increase of recreational activities during this season [32].

3.3. Biota

Little information is available about the occurrence of organic UV filters in marine biota compared with seawater and sediments. Due to the lipophilic characteristics of some compounds (with log K_{ow} between 4-8) and their relative stability against biotic degradation, they tend to accumulate in the food chain [15] and are transferred to humans through alimentation [59]. Different species have been collected from different part of the world and in different seasons with the aim of determining their occurrence (Table S3). Their concentrations have been reported in d.w., lipid weight (l.w.) and wet weight (w.w.).

The majority of studies examining the marine biota has been conducted in China, as shown in Figure 3B. Most studies have been conducted on fish fillets from a wide variety of species [102,104] (Table 4), because they are part of the human diet [122]. Other fish tissues that are not components of the human diet have also been examined, such as fish belly and other inedible fish tissues [62].

The most frequently detected compounds (BP-3, 4-MBC, OC, OMC, and OD-PABA) are widely used not only in PCPs but also in food additives [67], and they have been detected at different concentrations (Table S3). BP-3 was detected at higher concentrations (82.2 ng·g^{-1} d.w.) in fish fillets [103], while OC is the most frequently detected compound. It presents a greater log K_{ow} (6.88), which contributes to its bioaccumulation.

The study by Molins-Delgado et al. [59] reported the distribution and concentration of organic UV filters (BP-3, 4-MBC, OC, OMC and OD-PABA) in different parts of the fish (*Mugil liza*), including the gills. In addition, the same authors and Emnet et al. [10] reported greater accumulation of BP-3 in fish liver (7.55-74.4 and 41.0 ng·g^{-1} d.w., respectively) than in muscle tissues (<3.20-15.4 and <6.6-14.1 ng·g^{-1} d.w., respectively).

Cunha et al. [104] and Picot Groz et al. [67] studied the presence of OC, OMC and OD-PABA in mussels from Portugal. However, the first authors [104] did not detect any compound. A potential explanation for these results is the collection of samples from September-December, while Picot Groz et al. [67] collected samples in summer. Thus, recreational activities exert an effect on these organisms. These results are consistent with the findings reported in other places showing the seasonal variation in seawater [24,41,84]. In addition, OMC was detected after the summer period, suggesting that other sources in addition to recreational activities also contribute to its accumulation [64].

Also, OC and OMC were detected in *Mytilus galloprovincialis* and *Mytilus edulis* mussels, with the highest concentration observed for OC (7112 ng·g^{-1} d.w.) [64]. The samples were collected from June to November at a closed beach in France with a population size of greater than 50000 inhabitants, revealing the effects of both recreational activities and the geomorphological structure on the concentration of organic UV filters in this kind of organisms.

In addition, a bioaccumulation study in mussels (*Mytilus galloprovincialis*) was conducted under laboratory conditions by Vidal-Liñán et al. [115]. The mussels were exposed to artificial seawater that contained BP-3, benzophenone (BP-4), 4-MBC, OC and OD-PABA for 30 days. After exposure, rapid uptake of 4-MBC, BP-4 and OC was registered, while BP-3 and OD-PABA presented lower accumulation. Moreover, the mussels are able to biotransform OD-PABA after exposure until undetectable levels are observed. The concentrations observed after exposure are comparable to the results reported for wild mussels (*Mytilus galloprovincialis* and *Mytilus edulis*) [64], confirming the bioaccumulation of some organic UV filters in mussels.

In Brazil, two studies analysed organic UV filters in dolphins (Table S3). Gago-Ferrero et al. [65] were the first to report the levels of these compounds in dolphins (*Pontoporia blainvillei*) liver. The concentration of OC was up to 782 ng·g^{-1} l.w. The second study performed by Alonso et al. [60] was related to the transfer of organic UV filters between the dolphin mother to the foetus or calf in two different dolphin species, Franciscana (*Pontoporia blainvillei*) and Guiana (*Sotalia guianensis*). Of all the samples analysed, the most recurrent compound was OC, and the highest concentration of OC (11130 ng·g^{-1} l.w.) was measured in muscle of foetal Franciscana dolphins (Table S3). This study also reported the presence of other organic UV filters (4-MBC, OMC, and OD-PABA) in dolphins. These filters likely accumulate due to feeding, as these animals are homeotherms and their rates of feeding are higher than fish [122]. Therefore, biomagnification is also suggested [60].

Marketed mussels, octopus, crab and fish fillets have also been analysed with the aim of evaluating the presence of organic UV filters in seafood in the EU [103]. BP-3, IMC, 4-MBC, OC, OMC, EHS and HMS were detected at different concentrations. BP-3 appears to be the compound present at the highest concentration in fish fillets (82.2 ng·g^{-1} d.w.). In addition, the same organic UV filters were also detected in canned fish fillets (seabream) [103]. Other biota species, such as shrimp [61], sea urchin [10], clams[10,58,104], conch, squids, *Squilla* and sea snails [62], were also analysed and different organic UV filters were also detected (Fig. 3A and 3B).

Finally, in other kind of organism like corals, the occurrence of organic UV filters (BP-3, 4-MBC, OC, OMC and OD-PABA) was reported for five different species of corals (*Platygyra acuta*, *Porites sp.*, *Pavona decussata*, *Acropora valida*, and *Favites abdita*) sampled along the Pearl River Estuary (Hong Kong, China) [32]. In samples collected during the wet and dry seasons, BP-3 was the only compound reported in both seasons. Additionally, higher concentrations were reported for BP-3 in the wet season revealing seasonal variations, this is potentially attributed to the seawater patterns also observed in the same place [32].

4. Conclusions

Organic UV filters are present in several marine matrices and are compounds of increasing concern because their toxicity and adverse effects on different marine organisms have been already reported. The most frequently used separation and detection technique for their measurement in environmental samples is gas chromatography coupled to mass spectrometry. Liquid chromatography is also selected for non-volatile compounds.

Organic UV filters exhibit a wide range of different characteristics that must be considered before their extraction from the matrix. A wide variety of extraction techniques have been performed to extract these compounds from seawater, sediment and biota, due to the wide range of polarities, solubilities, and other properties. Each technique offers distinct advantages and disadvantages in its analytical approach. In this way the new on-line techniques offer a high sample analysis frequency, allowing to analyse 2-5 samples per hour. Moreover, they present considerable improvements respect to traditional techniques such as LLE and SPE, like the reduction of solvent, sample volumes and sample handling. These characteristics are consistent with the principles of the green chemistry for determining the levels of trace contaminants.

Solid matrices (sediments and biota) represent a special challenge due to the low concentrations and matrix effects. Usually, this kind of matrices requires pre-treatments, post clean-up process and preconcentration steps. In this sense, there is not a common pre-treatment stablished, resulting difficult to compare the values obtained in some cases. Moreover, in biota samples the concentration is not always presented in the same units of concentration (*i.e.* l.w., w.w., d.w.).

The developed techniques are usually focused in some compounds, setting aside other compounds. In seawater only for the 64% of the compounds allowed in the EU, an appropriated methodology have been performed. For sediments and biota the percentages fall to 48% and 40%, respectively. Further methodology capable of extract a large number of compounds and their TPs are needed.

Most of the studies conducted to determine the occurrence of organic UV filters in the environment have been performed in seawater, where seasonal variations have been observed. Highest concentrations are reported in summer, principally due to recreational activity. The occurrence of organic UV filters in seawater also depends on the presence of marine outfalls of sewage. Studies in seawater have been reported the presence of only fourteen of the twenty-five compounds allowed in the EU since there is no methodology developed for the rest of organic UV filters. BP-3 was the most recurrent compound. From the reviewed studies in seawater, BP-3 was present in the 85% of the cases, OC and OMC in the 58%, 4-MBC in the 55%, and HMS, EHS and OD-PABA from 29% to 35%.

In the case of sediments, OC and OMC was reported in the 80% of the analysed samples, BP-3 in the 73%, 4-MBC and OD-PABA in the 47% and EHS and HMS in the 33%. In biota samples, OC was found in the 92%, of the samples, BP-3 and OC in the 64%, OD-PABA in the 50% and 4-MBC in the 42%.

To analyse the frequency of the results obtained in biota, it is necessary to take into account the different kind of organisms. Different frequencies were found for organic UV filters in diverse tissues from dolphins. The same pattern was observed in fish tissues. Different canned organisms presented 4-MBC, HMS, BP-3, OMC, EHS and OC. 4-MBC and BP-3 have also been found in organisms that comes from aquaculture, while BP-3 and HMS were reported in analysed marketed organisms taken in different places. In addition, wild organisms presented OC as the most frequent compound. These results provide a broad perspective of the possible bioaccumulation process. In corals taken from different places from China, BP-3 was reported in the 100% of the analysed samples.

Furthermore, the presence of organic UV filters in biota and seawater in polar regions (Arctic and Antarctic) confirms their global occurrence and relation to wastewater release, which highlights the risk that these contaminants poses to vulnerable ecosystems.

In general, the organic UV filters determination is focused in the compounds allowed in the country where the studied is carried out. Nevertheless, in some cases, several prohibited compounds have been found. This could be explained due to the use of PCPs brought by foreigner tourists, which demonstrates the widespread use of organic UV filters.

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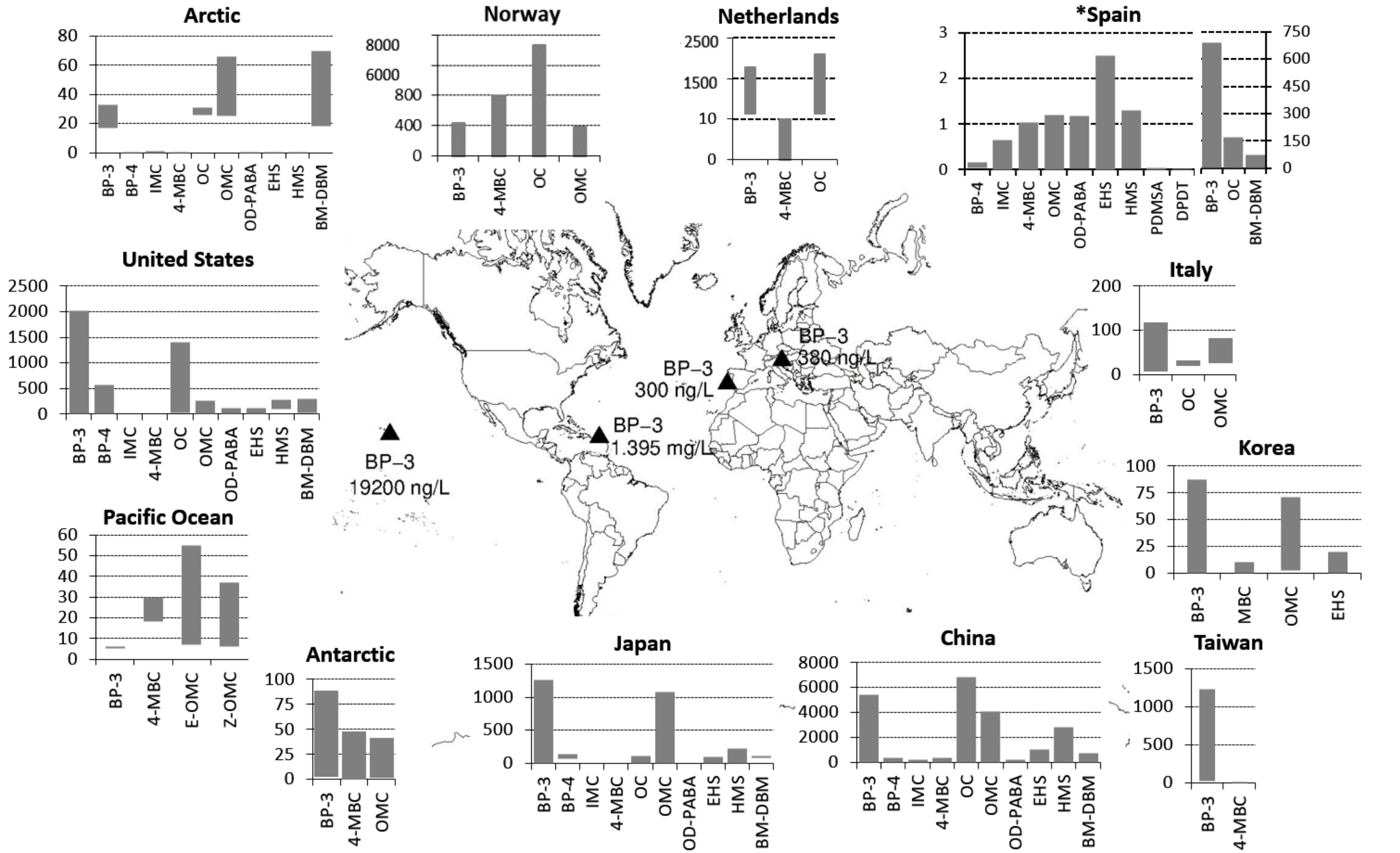
Figure captions:

Fig. 1. Global occurrence of UV filters in seawater. Y-axis are in ng·L^{-1}. Triangles represent a single value reported, * Values presented in µg·L^{-1}. Concentration expressed in pg·SPMD^{-1} for the Pacific Ocean.

Fig. 2. Global occurrence of UV filters in sediments.

Fig. 3A. European occurrence of UV filters in different marine biota. Y-axis are in ng·g^{-1} d.w. ^{x} Concentration in ng·g^{-1} w.w. □Concentration in ng·g^{-1} l.w. related to dolphins.

Fig. 3B. Not European occurrence of UV filters in different marine biota. Y-axis are in ng·g^{-1} d.w. ^{x} Concentration in ng·g^{-1} w.w. □Concentration in ng·g^{-1} l.w. related to dolphins.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20
40
60
80
BP-3
BP-4
IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
BM-DBM
Norway
0
400
800
6000
8000
BP-3
4-MBC
OC
OMC
Netherlands
0
10
1500
2500
BP-3
4-MBC
OC
*Spain
0
1
2
3
0
150
300
450
600
750
BP-4
IMC
4-MBC
OMC
OD-PABA
EHS
HMS
PDMSA
DPDT
BP-3
OC
BM-DBM
United States
0
500
1000
1500
2000
2500
BP-3
BP-4
IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
BM-DBM
Italy
0
100
200
BP-3
OC
OMC
BP–3
380 ng/L
BP–3
300 ng/L
BP–3
1.395 mg/L
BP–3
19200 ng/L
Korea
0
25
50
75
100
BP-3
MBC
OMC
EHS
Pacific Ocean
0
10
20
30
40
50
60
BP-3
4-MBC
E-OMC
Z-OMC
Antarctic
0
25
50
75
100
BP-3
4-MBC
OMC
Japan
0
500
1000
1500
BP-3
BP-4
IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
BM-DBM
China
0
2000
4000
6000
8000
BP-3
BP-4
IMC
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OC
OMC
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HMS
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Taiwan
0
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1000
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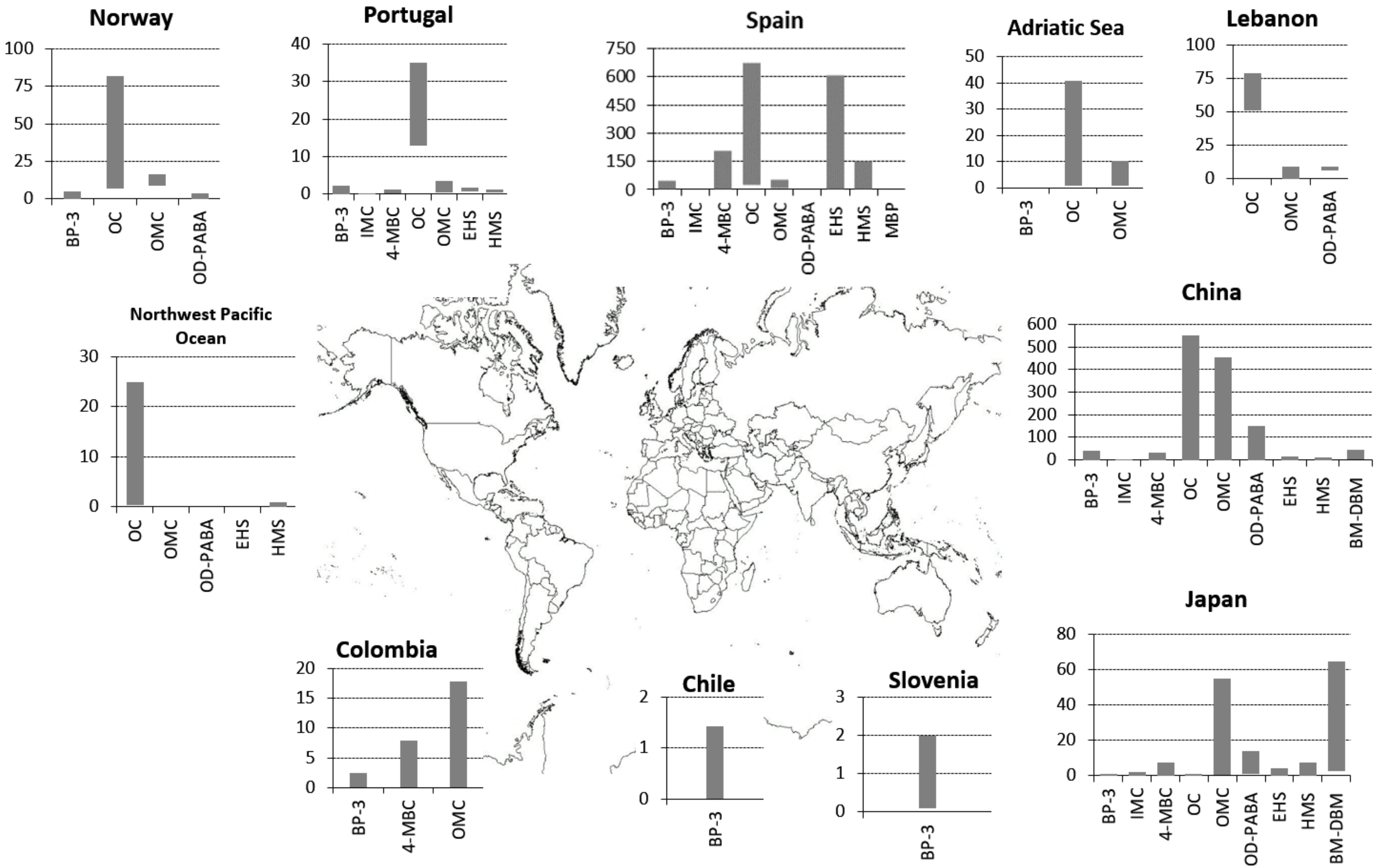
BP-3
4-MBC



Norway
100
75
50
25
0
BP-3
OC
OMC
OD-PABA
Portugal
40
30
20
10
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BP-3
IMC
4-MBC
OC
OMC
EHS
HMS
Spain
750
600
450
300
150
0
BP-3
IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
MBP
Adriatic Sea
50
40
30
20
10
0
BP-3
OC
OMC
Lebanon
100
75
50
25
0
OC
OMC
OD-PABA
Northwest Pacific Ocean
30
20
10
0
OC
OMC
OD-PABA
EHS
HMS
China
600
500
400
300
200
100
0
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IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
BM-DBM
Japan
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60
40
20
0
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IMC
4-MBC
OC
OMC
OD-PABA
EHS
HMS
BM-DBM
Colombia
20
15
10
5
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4-MBC
OMC
Chile
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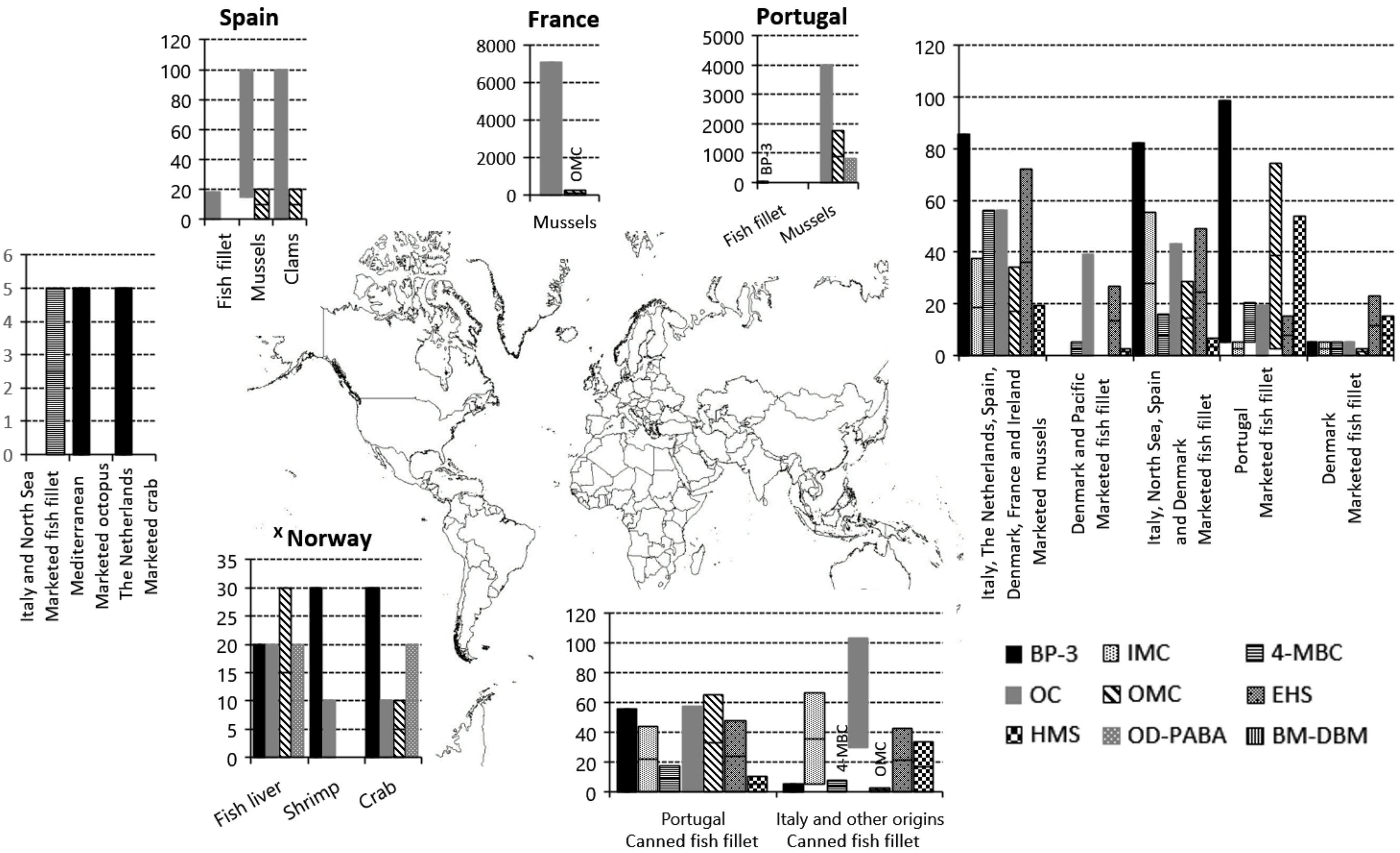
Slovenia
3
2
1
0
BP-3


Spain
120
100
80
60
40
20
0
Fish fillet
Mussels
Clams
France
8000
6000
4000
2000
0
OMC
Mussels
Portugal
5000
4000
3000
2000
1000
0
BP-3
Fish fillet
Mussels
6
5
4
3
2
1
0
Italy and North Sea
Marketed fish fillet
Mediterranean
Marketed octopus
The Netherlands
Marketed crab
x Norway
35
30
25
20
15
10
5
0
Fish liver
Shrimp
Crab
120
100
80
60
40
20
0
4-MBC
OMC
Portugal
Canned fish fillet
Italy and other origins
Canned fish fillet
120
100
80
60
40
20
0
Italy, The Netherlands, Spain,
Denmark, France and Ireland
Marketed mussels
Denmark and Pacific
Marketed fish fillet
Italy, North Sea, Spain
and Denmark
Marketed fish fillet
Portugal
Marketed fish fillet
Denmark
Marketed fish fillet
BP-3
IMC
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OC
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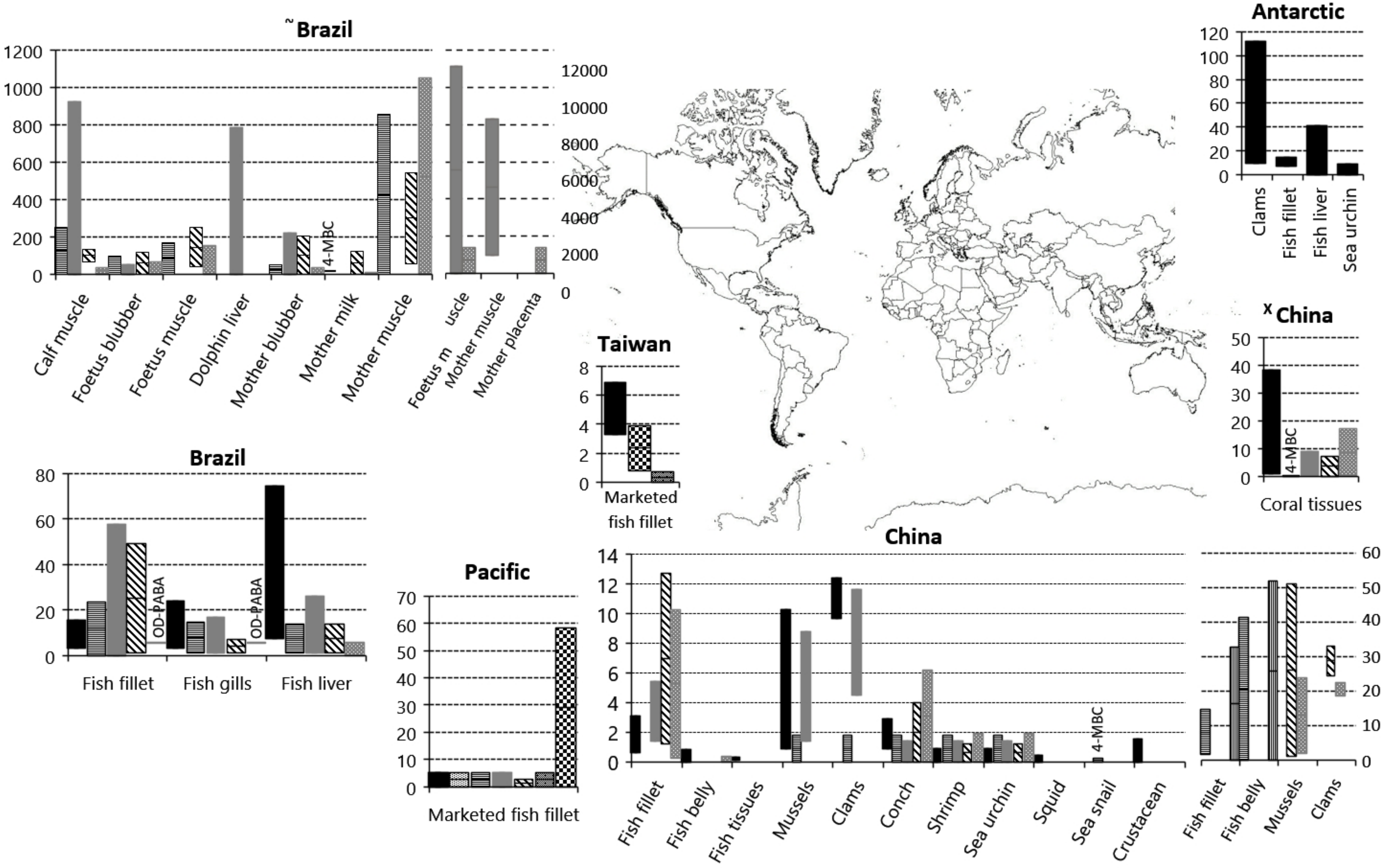

~ Brazil
Calf muscle
Foetus blubber
Foetus muscle
Dolphin liver
Mother blubber
Mother milk
Mother muscle
Foetus muscle
Mother muscle
Mother placenta
4-MBC
Brazil
Fish fillet
Fish gills
Fish liver
OD-PABA
OD-PABA
Pacific
Marketed fish fillet
Taiwan
Marketed fish fillet
China
Fish fillet
Fish belly
Fish tissues
Mussels
Clams
Conch
Shrimp
Sea urchin
Squid
Sea snail
Crustacean
4-MBC
Fish fillet
Fish belly
Mussels
Clams
Antarctic
Clams
Fish fillet
Fish liver
Sea urchin
x China
4-MBC
Coral tissues

Table 1. Main characteristic of organic ultraviolet filters authorized by the EU (regulation number 12223/2009).

Families	**Name (INCI nomenclature)**[a]	**Abbreviation**	**CAS Nº**	**Log K$_{ow}$**[b]	**Solubility (g·L^{-1})**[g]	**pKa**[h]
Benzophenones	Benzophenone-3	BP-3	131-57-7	3.79[c]	0.21	7.56[f]
	Benzophenone-4	BP-4	4065-45-6	0.37[d]	0.65	-0.70[f]
p-aminobenzoic acid and derivatives	Ethoxylated ethyl 4-aminobenzoate	PEG-25 PABA	116242-27-4	-0.66[e]	-	
	Ethylhexyl dimethyl PABA	OD-PABA	21245-02-3	6.15[f]	2.1x10^{-3}	2.39[f]
Salicylates	Homosalate	HMS	118-56-9	6.16[d]	0.02	8.09[f]
	2-ethylhexyl salicylate	EHS	118-60-5	5.97[f]	0.028	8.13[f]
Cinnamates	Ethylhexyl methoxycinnamate	OMC	5466-77-3	5.8[d]	0.15	-
	Isoamyl p-methoxycinnamate	IMC	71617-10-2	4.33[d]	0.06	-
Camphor derivatives	Camphor benzalkonium methosulfate	CBM	52793-97-2	0.28[e]	0.007	-
	Terephthalylidene dicamphor sulfonic acid	PDSA	92761-26-7 / 90457-82-2	3.83[e]	0.014	-1.05[h]
	Benzylidene camphor sulfonic acid	BCSA	56039-58-8	2.22[e]	0.038	-0.7[h]
	Polyacrylamidomethyl benzylidene camphor	PBC	113783-61-2	-	-	-
	4-methylbenzylidene camphor	4-MBC	36861-47-9 / 38102-62-4	4.95[d]	5.1x10^{-3}	-
Triazines	Ethylhexyl triazone	OT	88122-99-0	17.05[e]	-	3.17[f]
	Diethylhexyl butamido triazone	DBT	154702-15-5	14.03[e]	4.6x10^{-7}	3.04[f]
	Bis-ehylhexyloxyloxyphenol methoxyphenyl triazine	EMT	187393-00-6	8.03[e]	4.9x10^{-8}	6.37[h]
	Tris-biphenyl triazine	-	31274-51-8	10.38[e]	5.5x10^{-10}	1.2[h]
Benzotriazoles	Drometrizole trisiloxane	DTS	155633-54-8	10.82[e]	1.3x10^{-5}	9.72[h]
	Methylene bis-benzotriazolyl tetramethylbutylphenol	MBP	103597-45-1	12.46[e]	3x10^{-8}	7.56[h]
Benzimidazole derivatives	Phenylbenzimidazole sulfonic acid	PMDSA	27503-81-7	-0.16[e]	0.26	-0.87[f]
	Disodium phenyl dibenzimidazole tetrasulfonate	DPDT	180898-37-7	-6.79[e]	0.5	-0.27[f]
Dybenzoyl methane derivatives	Butyl methoxydibenzoylmethane	BM-DBM	70356-09-1	4.51[d]	0.037	9.74[f]
	Diethylamino hydroxybenzoyl hexyl benzoate	DHHB	302776-68-7	6.54[e]	9.5x10^{-4}	7.29[h]
Crylenes	Octocrylene	OC	6197-30-4	6.88[d]	2x10^{-4}	-
Benzylmalonate derivatives	Polysilicone-15	BMP	207574-74-1	-	-	-

[a] INCI International Nomenclature for Cosmetic Ingredients; [b] Octanol-water partition coefficient (K$_{ow}$); [c] Experimental values from Syracuse Research Corporation database; [d] Estimated values from Syracuse Research Corporation database; [e] Calculated by use of Estimation Program Interface (EPI) suite v4.11 (2012); [f] Software calculated value, from SciFinder Scholar Database 2006: http://www.cas.org/products/sfacad/; [g] From Díaz-Cruz et al., [15]. Indicates in water at 25ºC; [h] Values obtained from Chemicalize website

Table 2. Analytical methodologies for UV filters determination in seawater. Grouped by extraction techniques

Extraction technique	**Kind of sorbent**	**Compounds**	**Instrumental method**	**Eluent**	**Recoveries (%)**	**LOD (ng·L^{-1})**	**LOQ (ng·L^{-1})**	**Reference**
SPE	STRATA-X	BP-3 4-MBC OC OMC	GC-TOF-MS	EA and DCM mixture	60	1-5	-	[74]
	OASIS HLB	BP-3 BP-4 IMC 4-MBC OC OD-PABA BM-DBM PMDSA DPDT	HPLC-ESI-MS/MS	MeOH	79 91 66 69 66 71 69 88 81	7 10 46 20 18 12 25 8 25	-	[42]
	OASIS HLB	BP-3 E-OMC Z-OMC 4-MBC	GC-MS	MeOH	-	1-5	-	[50]
	OASIS HLB	BP-3 BP-4 IMC 4-MBC OC OMC OD-PABA PMDSA	HPLC-ESI-MS/MS	MeOH	128 105 77 84 88 103 76 85	4 0.9 5 2.7 30 3 1.6 0.8	13 3 16 9 99 10 5 3	[76]
	OASIS HLB	BP-3 BP-4 OC OMC OD-PABA BM-DBM	HPLC-ESI-MS/MS	MeOH and Ac mixture	71-111	0.5 1 25 25 1 12.5	-	[44]

	Discovery DSC-18LT and Discovery DSC-PH	BP-3 4-MBC OC OMC OD-PABA EHS HMS	GC–MS	DCM	80-113	0.1-3.0	-	[45]
	OASIS HLB	BP-3 BP-4 4-MBC OMC	HPLC-ESI-QTOF-MS	MeOH	-	-	10 - 9 10	[27]
	Bond Elut C18	BP-3 BP-4 IMC 4- MBC OC OMC OD-PABA EHS HMS BMDBM	HPLC–ESI-MS/MS	MeOH and EA mixture	93 104 77 83 76 83 73 63 65 74	0.04 0.03 1.04 0.28 1.38 0.41 0.03 0.10 0.11 0.13	-	[29]
	STRATA X	BP-3 4-MBC OC OMC OD-PABA HMS BM-DBM DHHB	UPLC-DAD	EA and DCM mixture	94-104 91-98 80-100 79-92 84-93 78-110 86-90 88-91	1.4 0.9 2.8 1.6 1.2 2.4 2.0 1.3	4.8 3.1 9.3 5.2 3.9 8.0 6.7 4.2	[40]
SPE	OASIS HLB	BP-3	GC-MS	EA	95	0.5	2.0	[54]
	OASIS HLB	BP-3 BP-4 OC OMC	HPLC-ESI-MS	MeOH and Ac mixture	100	0.5 12.5 25 25	-	[47]

		OD-PABA BM-DBM				12.5 1		
	OASIS HLB	BP-3 4-MBC OMC	GC-MS	DCM and MeOH mixture	124.4 118.7 94.5	-	2.6 3.2 1.9	[10]
	C18E	BP-3	GC-MS	Ac and DCM	>95	100	5000	[31]
	C18E		HPLC-MS	MeOH	>85	100	5000	
	Bond Elut C18	BP-3 4-MBC OC OMC OD-PABA	UPLC-ESI-MS/MS	MeOH and EA mixture	93 83 76 83 73	0.04 0.28 1.38 0.41 0.03	-	[32]
	SPE-DEX 4790	BP-3 4-MBC OMC OD-PABA EHS	GC-MS	Methylene chloride, EA and mixture of methylene chloride and EA	50-130	1.0 0.5 1.0 1.0 2.0	-	[41]
	LC-18 SPE	BP-3 4-MBC OC OMC OD-PABA	UPLC-ESI-MS/MS	MeOH and Mili-Q water	94.3-105.2 85.3-110.3 87.7-104.6 91.6-114.4 101.3-111.2	2.12 2.59 3.03 3.25 4.91	6.41 7.84 9.19 9.85 14.88	[24]
	ENVI-Chrom-P 500	BP-3 4-MBC OC	HPLC-ESI-MS/MS	MeOH	80 73 63	6.3 5.1 3.2	10 10 20	[33]
On-line SPE	OASIS HLB	MBP	UPLC-MS/MS	MeOH	61-66	1.1	3.8	[39]
MNPs-based dSPE	$CoFe_2O_4$-oleic acid	BP-3 IMC 4-MBC OC OMC OD-PABA	GC-MS	Hex	125 80 80 88 73 101	0.2 6.0 5.8 1.8 2.5 3.1	0.8 20.0 19.3 5.9 8.3 10.2	[48]

		EHS			86	0.2	0.5	
		HMS			81	0.4	1.5	
SPME	Polyacrylate fibre	BP-3	GC-MS/MS	-	73-115	1.5	-	[70]
		IMC			94-108	0.068		
		4-MBC			73-108	1.5		
		OC			104-128	0.16		
		OMC			75-117	0.22		
		OD-PABA			82-106	0.25		
		EHS			84-112	0.69		
		HMS			89-117	0.34		
		BM-DBM			-	12		
		DHHB			89-99	6.0		
		DTS			94-121	3.0		
In-vial SPME	Divinylbenzene-carboxen-polydimethylsiloxane fibre	BP-3	GC-MS/MS	-	86.9-89.7	0.30	1.0	[16]
		BP-4			86.9-89.7	0.30	1.0	
		IMC			94.9-96.5	0.069	0.23	
		4-MBC			90.6-95.3	0.84	2.8	
		OC			98.5-102	0.18	0.60	
		OMC			86.8-98.0	0.060	0.20	
		OD-PABA			95.9-101	0.096	20	
		EHS			93.1-95.9	0.066	0.22	
		HMS			85.6-90.8	0.15	0.49	
SBSE	Polydimethylsiloxane coated stir bar (Twister)	BP-3	HPLC-APCI-MS/MS	MeOH	71-100	80	25	[49]
		OC				200	101	
		OMC				70	25	
		OD-PABA				10	25	
		EHS				2650	3900	
		HMS				1700	3900	
	Polydimethylsiloxane coated stir bar (Twister)	BP-3	HPLC-APCI-MS/MS	MeOH	64	0.9	3.0	[43]
		OC			76	3.3	11.1	
		OMC			84	2.8	9.2	
		OD-PABA			82	0.6	2.4	
		EHS			83	114	382	
		HMS			85	94	313	

	Polydimethylsiloxane coated stir bar	MBP	UHPLC-MS/MS	ACN	18.4-19.9	22.9	76.3	[37]
	Polydimethylsiloxane coated stir bar	PB-3 OC	GC-MS	EA	27.6 59.6	2 0.6	-	[38]
	Polydimethylsiloxane coated stir bar	BP-3 4-MBC OC OMC OD-PABA EHS HMS BM-DBM	GC-APGC–TOF-MS	EA	-	0.17 0.01 0.02 0.46 0.6 0.28 0.44 12.4	-	[77]
SBDLME	$[P^+_{6,6,6,14}][Ni(hfacac)_3^-]$	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	GC-MS	-	91-95 109-113 97-102 95-103 95-91 110-112 114-117 102-104	10.4 13.1 15.2 21.2 15.3 26.7 9.9 11.3	34.3 43.1 50.2 69.9 50.5 88.8 32.5 37.3	[78]
SBSDME	$CoFe_2O_4$-oleic acid	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	HPLC-UV	Ethanol	84-116 79-116 96-120 98-103 97-107 100-107 83-95 87-97	30600 2400 3200 2700 2400 3000 3000 3200	100000 8000 10700 9100 8000 9900 9700 10000	[71]
	$CoFe_2O_4$-oleic acid	BP-3 IMC 4-MBC OC OMC OD-PABA	GC-MS	-	103 104 112 89 88 111	148 28 23 27 28 30	493 95 78 91 95 99	[79]

		EHS HMS			109 112	23 13	77 43	
	CoFe$_2$O$_4$-SiO$_2$-nylon	BP-4 DPDT PMDSA PDSA	HPLC-UV	HCl	95-103 91-97 104-115 97-105	1600 1900 2800 2900	5400 6300 9200 9600	[80]
BAME	Modified pyrrolidone	BP-3	HPLC-DAD	MeOH and ACN mixture	76.6-98.4	300-400	1000- 1300	[82]
LLE	-	4-MBC OMC OD-PABA EHS	GC-MS	Hex	89 90 86 120	0.15 0.082 0.096 0.099	0.5 0.27 0.32 0.33	[84]
	-	BP-3 4-MBC	HPLC-MS/MS	n-octanol	86.2-109.3 95.0-109.7	10.3 10.9	34.4 36.4	[87]
DLLME	-	BP-3	GC-MS	Ac dispersive solvent CHL extractant	82-126	33	110	[86]
	-	BP3 4-MBC	GC–MS	Ac dispersive solvent CHL extractant	-	-	-	[11]
	-	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	GC-MS	Ac dispersive solvent CHL extractant	111-114 97-107 82-88 91-104 87-99 90-95 112-117 88-97	30 23 10 27 14 29 26 14	99 78 33 91 47 98 85 46	[46]
IL-SDME	-	BP-3 IMC 4-MBC OC OMC OD-PABA	HPLC-DAD	[C$_6$MIM][PF$_6$]	99 92 96 92 107 92	110 160 60 3000 190 70	370 530 200 10000 640 230	[89]

On-line *In-syringe* IL-MSA-DLLME	-	BP-3 4-MBC OC OD-PABA EHS HMS	HPLC-UV	ACN dispersive solvent $[C_6MIM][PF_6]$ extractant	92 52 48 49 46 51	180 80 2500 890 11820 1024	620 250 8340 2980 39390 34150	[72]
In-syringe MSA-DLLME	-	BP-3 4-MBC OC OMC EHS HMS	GC-MS	Ac dispersive solvent Trichloroethylene extractant	95.6-104.4 88.4-111 95.1-110.7 82.5-108.3 98.4-104.3 95.4-110.8	79 380 291 191 31 95	23 160 130 86 19 28	[34]
USAEME	-	IMC 4-MBC OC OMC OD-PABA EHS HMS	GC-MS//GC-MS/MS	CHL	98.4-105 92.4-96.7 97.7-102 90.8-102 75.5-84.2 98.0-108 91.5-94.2	5.8//1 0.22//0.29 25//0.5 2.1//0.66 5.4//0.08 3//0.29 15//1.3	19//3.3 0.73//9.7 83//1.7 7//2.2 18//0.27 10//0.97 50//4.3	[69]
FPSE	Sol-gel poly(dimethyldiphenylsiloxane)	MBP	UHPLC-ESI-MS/MS	MeOH	32.4-51.4	2.72	9.08	[73]
iSAME	Cetyltrimethylammonium bromide and sulfosalicylic acid	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	HPLC-UV	2-propanol	102 88 93 93 89 95 80 84	1500 300 300 800 300 300 1700 1700	4800 1000 900 2600 1100 1100 5700 5800	[85]
SPMDs	Semipermeable-membrane	BP-3 E-OMC Z-OMC 4-MBC	GC-MS	Cyclohexane	-	(pg/SPMD) 150-510	-	[50]

Table 3. Analytical methodologies for UV filters determination in sediments. Grouped by extraction technique.

Extraction technique	**Compounds**	**Instrumental method**	**Eluent**	**Recoveries (%)**	**LOD (ng·g^{-1} d.w.)**	**LOQ (ng·g^{-1} d.w.)**	**Reference**
Soxhelt	4-MBC	GC-MS	DCM and Hex mixture	93	-	-	[56]
	4-MBC OC OMC	GC-MS	DCM	70-90	0.01-0.17	-	[52]
MAE	OC OMC OD-PABA	GC-MS/MS	Ac and heptane mixture	97-115 99-113 98-104	2.0 1.5 1.5	6.0 5.0 5.0	[51]
	MBP	UHPLC-ESI-MS/MS	ACN	50.1-55.7	0.0533	0.176	[114]
	BP-3	GC-MS	MeOH and Ac mixture	80	0.1	0.2	[54]
USE	BP-3 EHS HMS	GC-MS/MS	EA and MeOH mixture	98.9-101.3 99.4-102 97.4-101.3	0.28 0.11 0.12	0.90 0.36 0.40	[53]
	BP-3 4-MBC OC OMC OD-PABA	HPLC-ESI-MS/MS	MeOH	55.1 68.9 117.6 101.4 86.9	-	0.3 0.1 0.03 0.5 0.03	[35]
	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	GC-MS/MS	EA	98.3-115 100-107 96.3-107 98.3-117 107-110 88.2-104 100-101 84.2-103	-	2.5 0.40 4.6 0.43 1.8 0.10 0.32 2.0	[36]

VE	BP-3	GC-MS	Ac	106	0.041	0.140	[55]
	IMC			86	0.041	0.140	
	4-MBC			92	0.029	0.096	
	OC			95	0.035	0.117	
	OMC			82	0.018	0.061	
	OD-PABA			84	0.046	0.150	
	EHS			80	0.038	0.130	
	HMS			9	0.053	0.180	
	BP-3	UPLC-ESI-MS/MS	Ac and n-hexane mixture	81	0.43	-	[32]
	4-MBC			58	0.09		
	OC			76	0.09		
	OMC			76	7.55		
	OD-PABA			63	0.16		
	BP-3	GC-MS/MS	EA	86-121	-	3.2	[36]
	IMC			96.4-106		0.42	
	4-MBC			99.7-99.2		4.9	
	OC			103-112		0.33	
	OMC			100-112		1.6	
	OD-PABA			89.7-106		0.30	
	EHS			92.2-103		0.16	
	HMS			91-95.3		2.3	
PLE-ASE	BP-3	UPLC-ESI-MS/MS	MeOH and EA mixture	83	0.71	-	[57]
	IMC			82	2.10		
	4-MBC			91	7.33		
	OC			89	0.58		
	OMC			100	0.51		
	OD-PABA			94	0.61		
	EHS			84	4.26		
	HMS			75	7.55		
	BM-DBM			78	3.94		
	BP-3 OC OMC	GC-MS	DCM	70-100	0.003- 0.54	-	[93]
	BP-3	GC-MS/MS	DCM	61-91	0.009	0.029	[94]

	4-MBC			53-91	0.221	0.737	
	OC			92-120	0.024	0.080	
	OMC			86-134	0.039	0.129	
	OD-PABA			85-138	0.408	1.361	
	EHS			68-94	0.065	0.216	
	HMS			70-130	0.022	0.073	
	BP-3	GC-MS/MS	DCM	61-91	0.009	0.029	[95]
	4-MBC			53-91	0.221	0.737	
	OC			92-120	0.024	0.080	
	OMC			86-134	0.039	0.129	
	EHS			68-94	0.065	0.216	
	HMS			70-130	0.022	0.073	
	4-MBC	UPLC-UV	EA and Hex mixture	74.4-102.4	-	0.00036	[96]
	OD-PABA			66.4-77.0		0.00040	
	BP-3	UHPLC-ESI-MS/MS	DCM	92-106	0.03	0.1	[30]
	IMC			98-108	0.02	0.07	
	4-MBC			97-100	0.12	0.38	
	OC			103-108	0.084	0.28	
	OMC			100-101	0.016	0.06	
	OD-PABA			90-99	0.001	0.004	
	EHS			88-105	0.02	0.07	
	HMS			83-94	0.007	0.024	
	OT			<5	-	-	
	DBT			<5	-	-	
SPLE-ASE	BP-3	UPLC-MS/MS	MeOH	125	0.4	1.3	[22]
	4-MBC			89	1.1	3.6	
	OC			85	9.9	33	
	OMC			90	4.1	14	
	OD-PABA			120	0.7	2.5	
	BP-3	UPLC-HRMS and GC-HRMS	Hex and DCM mixture	72	5	10	[61]
	OC			102	7	5	
	OMC			98	-	5	
	OD-PABA			81	4	4	

PHWE	BP-3	GC-MS	Water 10% MeOH	13.5	0.07	-	[38]
	OC			22.4	0.3		
SPME	BP-3	GC-MS/MS	-	77.6-107	0.052	0.17	[99]
	IMC			83.8-104	0.010	0.033	
	4-MBC			89.6-106	0.014	0.046	
	OC			89.0-119	0.059	0.18	
	OMC			70.8-111	0.087	0.15	
	OD-PABA			70.1-124	0.001	0.003	
	EHS			93.3-111	0.031	0.053	
	HMS			93.8-120	0.023	0.039	
SBSDME	BP-3	GC-MS	Ethanol	99-111	0.55	1.79	[100]
	IMC			100-110	0.02	0.07	
	4-MBC			105-107	0.15	0.49	
	OC			94-98	0.01	0.04	
	OMC			103-107	0.02	0.05	
	OD-PABA			98-108	0.04	0.12	
	EHS			95-103	0.03	0.09	
	HMS			91-103	0.04	0.14	
USSPME	BP-3	GC-MS/MS	MeOH	81.3-98.1	-	0.30	[36]
	IMC			85.5-86.8		0.080	
	4-MBC			91.5-96.3		0.50	
	OC			96.7-116		0.50	
	OMC			80.6-89.8		0.30	
	OD-PABA			88.4-91.8		0.010	
	EHS			87.9-94.4		0.10	
	HMS			88.3-95.6		0.10	

Table 4. Analytical methodologies for UV filters determination in marine biota. Grouped by extraction technique.

Extraction technique	Compounds	Type sample	Instrumental method	Eluent	Recoveries (%)	LOD (ng·g^{-1} d.w.)	LOQ (ng·g^{-1} d.w.)	Reference
PLE-ASE	OC	Dolphin liver expressed in ng·g^{-1} l.w.	UPLC-ESI-MS/MS	DCM and Hex mixture	-	23	75	[65]
	BP-3 4-MBC OMC	Clams tissues	GC-MS	Water and IPA mixture	53.0 - -	-	6.6 8.0 4.8	[10]
	BP-3 4-MBC OMC				67.4 - -	-	6.6 8.0 4.8	
	BP-3 4-MBC OC OMC OD-PABA	Fish fillet	ULPC-ESI-MS/MS	EA and DCM mixture	94.8-113.0 88.6-96.4 94.6-113.1 94.9-99.8 89.2-110.0	0.9 1.8 1.4 1.2 1.9	2.9 6.0 4.5 4.0 6.2	[58]
	BP-3 4-MBC OC OMC OD-PABA	Mussels tissues			95.5-102.9 90.9-95.0 83.7-98.0 80.4-93.7 88.8-109.8			
	BP-3 4-MBC OC OMC OD-PABA	Fish tissues			92.2-114.8 94.8-113.8 95.7-106.8 92.7-112.3 94.4-111.4			
	BP-3 4-MBC OC OMC	Fish fillet	HPLC-ESI-MS/MS	EA and DCM mixture	107 95 75 66	0.93 0.39 0.39 0.33	3.20 1.30 1.30 1.10	[59]

	OD-PABA				42	1.77	5.90	
SPLE-ASE	BP-3 OC OMC OD-PABA	Fish tissues expressed in ng/g w.w.	UPLC-HRMS and GC-HRMS	Hex and DCM mixture	75 75 85 51	-	20 20 30 20	[61]
USE	BP-3 4-MBC OC OMC OD-PABA BM-DBM	Fish fillet	UHPLC-APCI-MS/MS	MeOH	88.3-102.0 86.0-102.4 97.8-115.6 98.3-109.5 85.5-102.3 41.1-82.8	-	0.08 0.2 0.1 10 0.005 1	[62]
	BP-3 4-MBC OC OMC OD-PABA BM-DBM	Fish belly			93.6 80.8 87.9 81.1 64.2 58.4			
	4-MBC OC OMC OD-PABA	Dolphin liver expressed in ng/g l.w.	HPLC-ESI-MS/MS	Hex and DCM mixture	60-115	1.50-25	1.90-75	[60]
	BP-3 4-MBC OC OMC OD-PABA BM-DBM	Fish fillet	UHPLC-MS/MS	DCM and EA	70-120	-	0.003-1.0	[66]
VE	BP-3 4-MBC OC OMC OD-PABA	Coral tissues and skeleton	UPLC-ESI-MS/MS	Ac and n-hexane mixture	86 83 65 64 61	0.50 0.11 0.12 7.06 0.22	-	[32]

MAE	OC	Mussels tissues	GC-MS/MS	Ac and heptane mixture	89-101	2	5	[64]
	OMC				89-99	2	5	
	OD-PABA				103-116	2	5	
	OC	Mussels tissues	GC- MS	Ac and heptane mixture	89-101	2	5	[68]
	OMC				89-99	2	5	
QuEChERs	OC	Mussels tissues	GC-MS/MS	ACN	99-126	5	5	[67]
	OMC				93-106	1	5	
	OD-PABA				90-93	2.5	10	
	BP-3	Fish fillet	GC-MS	Deionized water and ACN	72-83	3	20	[104]
	IMC				89-95	6	20	
	4-MBC				79-86	2	5	
	OC				75-76	23	100	
	OMC				93-115	3	20	
	OD-PABA				69-79	2	5	
	EHS				83-91	2	5	
	HMS				76-82	6	20	
	DHHB				59-62	-	-	
	BP-3	Fish fillet	GC-MS/MS	Deionized water and ACN	72-77	0.5	2	[103]
	IMC				68-77	1	5	
	4-MBC				57-88	2	5	
	OC				77-79	3	10	
	OMC				90-107	0.5	1	
	OD-PABA				61	2	5	
	EHS				70-82	2	5	
	HMS				92-108	2	5	
	DHHB				82	7	20	
MSPD	BP-3	Fish fillet	GC-MS	ACN	97-99	9	28	[63]
	IMC				97-104	3	10	
	4-MBC				97-101	4	12	
	OC				99-106	1	4	
	OMC				94-98	2	6	
	OD-PABA				86-96	4	12	
	EHS				70-76	6	18	
	HMS				84-93	9	28	

	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	Mussels tissues			89-96 94-107 90-101 96-112 97-111 70-101 80-97 80-85			
	BP-3 EHS HMS	Striped bass fillet	GC-MS/MS	ACN	90 84 98	0.03 0.02 0.02	0.1 0.05 0.05	[102]
	BP-3 EHS HMS	Cod fillet			75 88 76			
	BP-3 EHS HMS	Salmon fillet			96 77 78			
	BP-3 BP-4 4-MBC OC OD-PABA	Mussels tissues	HPLC-ESI-MS/MS	ACN	90-110	-	0.2-3	[115]

Supplementary Material.

Table S1. UV filters occurrence in seawater. Chronological order.

Kind of matrices	Location	Compounds	Concentration (ng·L^{-1})	Reference
Seawater (coastal areas and fjord)	Norway (Bærum)	BP-3 4-MBC OC OMC	n.d- 439.9 n.d- 798.7 n.d- 7301.0 n.d- 389.9	[74]
Seawater (0, 10 and 100 m off a WWTP discharge)	Norway (Oslo Fjord)	BP-3 4-MBC OC OMC	n.d., 15.5-22.5, 13.7-35.5 n.d., n.d.- 17.2, 2.6- 5.3 n.d., n.d.- 31.2, n.d.- 24.8 32.6- 164.1, n.d.- 189.3, n.d.- 178.9	
Seawater (beach site)	Spain (Galicia)	BP-3 BP-4 IMC 4-MBC OC OD-PABA BM-DBM PMDSA DPDT	<LOD 38-138 <LOD <LOD <LOD <LOD <LOD <LOD-42 <LOD	[42]
Seawater (ocean water)	Pacific Ocean (crossed from Peru to Polynesia)	BP-3 4-MBC E-OMC Z-OMC	(pg/SPMD) <LOD- 34310 <LOD 11464- 27058 3432- 8484	[50]
Seawater (surface microlayer)	Pacific Ocean (near to Polynesia)	PB-3 4-MBC E-OMC Z-OMC	5- 6 18- 30 7- 55 6- 37	
Seawater (Ria water)	Spain (A Coruña)	BP-3 BP-4 IMC 4-MBC OC OMC OD-PABA PMDSA	n.d.***	[76]
Seawater (beach sites)	Spain (Valencia and Alicante)	BP-3 IMC 4-MBC OC OMC OD-PABA	n.d.***	[89]
Seawater (beach sites)	Spain (Murcia and Alicante)	BP-3	1340- 3300	[86]
Seawater (beach site)	Spain (Alicante)	BP-3 IMC 4-MBC OC OMC OD-PABA	254- 879 245- 645 358- 758 <LOQ- 440 409- 774 682- 1187	[48]

		EHS HMS	792- 1222 625- 1030	
Seawater (coastal areas)	Italy (Liguria)	BP-3 OMC	<LOQ- 118 <LOQ- 83	[49]
Seawater (coastal areas)	United States (South Carolina)	BP-3 BP-4 OC OMC OD-PABA BM-DBM	10-2013 <1 <25-1409 30-264 <1-111 62-303	[44]
Seawater (beach site)	Italy (Genoa)	BP-3 OC	8-13 19-32	[43]
Seawater (beach sites)	Spain (Gran Canaria Island)	MBP	<LOD-5.2†	[39]
Seawater (surface water)	Spain (Cádiz)	BP-3 OC	70 100	[38]
Seawater (beach sites)	Spain (Majorca Island, Palmira beach)	BP-3 4-MBC	143.6 62.5	[11]
	Spain (Majorca Island, Santa Ponça beach)	BP-3 4-MBC	76.2- 314.8 47.5- 65	
	Spain (Majorca Island, Ses Salines Cape)	BP-3 4-MBC	36.3 26.6	
Seawater	Portugal (Costa de Caparica)	BP-3	<LOD	[82]
Seawater (beach sites)	Japan (Okinawa Island)	BP-3 OC OMC OD-PABA EHS HMS	n.d.- 1258 n.d.- 79 n.d.- 143 n.d.- 0.8 n.d.- 10 n.d.- 214	[45]
Seawater (river and reef sites)	Japan (Okinawa Island)	BP-3 OC OMC OD-PABA EHS HMS	n.d.- 9.0 n.d.- 8.1 n.d.- 3.9 n.d. n.d.- 1.8 n.d.- 3.2	
Seawater (beaches sites)	Spain (Galicia, Coira beach)	BP-3 BP-4 4-MBC OMC	68.6 164.4 84.6 52.5	[27]
	Spain (Galicia, Toralla beach)	BP-3 BP-4 4-MBC OMC	21.7 58.8 - 35.7	

Surface samples	Slovenia (Novigrad)	BP-3	96	[54]
	Slovenia (Ankaran)		340	
	Slovenia (Portorož)		380	
Surface samples	Spain (Cádiz)	BP-3	60	[77]
		4-MBC	46	
		OC	49	
		OMC	36	
		EHS	<LOD	
		HMS	9	
Seawater (surface water)	China (Hong Kong)	BP-3	39- 5429	[29]
		BP-4	54- 389	
		IMC	63- 173	
		4- MBC	173- 379	
		OC	103- 6812	
		OMC	89- 4043	
		OD-PABA	95- 182	
		EHS	61- 1030	
		HMS	66- 2812	
		BM-DBM	24- 721	
	Japan (Tokyo Bay)	BP-3	24- 86	
		BP-4	71- 136	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	87- 108	
		OMC	46- 95	
		OD-PABA	<LOD	
		EHS	71- 95	
		HMS	65- 110	
		BM-DBM	78- 104	
	United States (New York)	BP-3	23- 178	
		BP-4	89- 574	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	117- 128	
		OMC	89- 150	
		OD-PABA	<LOD	
		EHS	<LOD	
		HMS	91- 114	
		BM-DBM	70- 87	
	United States (Los Angeles)	BP-3	227- 601	
		BP-4	<LOD	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	145- 377	
		OMC	91- 138	
		OD-PABA	<LOD	
		EHS	53- 120	
		HMS	142- 270	
		BM-DBM	67- 109	

	China (Shantou)	BP-3	55- 188	
		BP-4	<LOD	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	75- 107	
		OMC	52- 78	
		OD-PABA	<LOD	
		EHS	<LOD	
		HMS	<LOD	
		BM-DBM	53- 100	
	China (Chaozhou)	BP-3	37- 49	
		BP-4	<LOD- 49	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	36- 102	
		OMC	<LOD- 79	
		OD-PABA	<LOD	
		EHS	121- 128	
		HMS	<LOD	
		BM-DBM	<LOD	
	Arctic (Arctic Ocean and Chukchi Sea between 65 and 75ºN)	BP-3	17- 33	
		BP-4	<LOD	
		IMC	<LOD	
		4- MBC	<LOD	
		OC	26- 31	
		OMC	25- 66	
		OD-PABA	<LOD	
		EHS	<LOD	
		HMS	<LOD	
		BM-DBM	18- 70	
Seawater (beach sites)	Spain (Majorca Island, Palmira beach)	BP-3	308	[46]
		IMC	280	
		4-MBC	192	
		OC	260	
		OMC	260	
		OD-PABA	246	
		EHS	880	
		HMS	310	
Seawater (beach sites)	Spain (Valencia, Malvarrosa beach)	BP-3	200	
		IMC	251	
		4-MBC	220	
		OC	317	
		OMC	250	
		OD-PABA	390	
		EHS	750	
		HMS	280	
Seawater (beach sites)	Spain (Valencia, Pineda beach)	BP-3	<LOQ	
		IMC	118	
		4-MBC	<LOD	
		OC	<LOQ	
		OMC	91	
		OD-PABA	163	
		EHS	440	
		HMS	157	

Seawater (beach sites)	Spain (Gran Canaria Island, Maspalomas-Los Ingleses beach)	BP-3	<LOD- 27.1	[40]
		4-MBC	<LOD- 7.2	
		OC	<LOQ- 359.1	
		OMC	<LOD- 16.1	
		HMS	<LOD- 51.5	
		BM-DBM	<LOD- 188.4	
		DHHB	<LOD- <LOQ	
	Spain (Gran Canaria Island, Puerto Rico beach)	BP-3	32.7- 979.8	
		4-MBC	4.1- 219.5	
		OC	61.2- 973.1	
		OMC	<LOD- 756.2	
		HMS	9.2- 536.2	
		BM-DBM	35.6- 1163.2	
		DHHB	<LOQ- 228.7	
	Spain (Gran Canaria Island, Amadores beach)	BP-3	12.7- 2675.7	
		4-MBC	<LOD- 104.8	
		OC	30.7- 766.7	
		OMC	<LOD- 276.8	
		HMS	<LOD- 319.0	
		BM-DBM	<LOQ- 792.0	
		DHHB	<LOD- 163.5	
	Spain (Gran Canaria Island, Mogán beach)	BP-3	54.2- 3316.7	
		4-MBC	<LOQ- 346.3	
		OC	37.8- 1324.9	
		OMC	<LOQ- 260.2	
		HMS	10.8- 526.1	
		BM-DBM	19.8- 1770.3	
		DHHB	<LOQ- 144.4	
	Spain (Gran Canaria Island, Las Alcaravaneras beach)	BP-3	<LOD- 158.0	
		4-MBC	<LOD- 29.7	
		OC	<LOD- 183.2	
		OMC	<LOD- 65.4	
		HMS	<LOD- 84.8	
		BM-DBM	<LOD- 314.3	
		DHHB	<LOD- 34.7	
	Spain (Gran Canaria Island, Las Canteras beach)	BP-3	<LOD- 182.6	
		4-MBC	<LOD- 1043.4	
		OC	<LOD- 768.5	
		OMC	<LOD- 109.9	
		HMS	<LOD- 102.2	
		BM-DBM	<LOD- 737.1	
		DHHB	<LOD- 176.3	
Seawater (beach sites)	United states (South Carolina)	BP-3	37.6- 591	[47]
		OC	41.1- 711	
		OMC	10.7- 96.9	
		OD-PABA	n.d- 36.7	
		BM-DBM	31.9- 234	
Ocean seawater	Antarctic (Cape Armitage, Winter Quarters Bay, Scott Base, and Cape Evans during 2009-2010)	BP-3	12- 88.4	[10]
		4-MBC	n.d- 47.5	
		OMC	<LOQ- 41.7	
Ocean seawater	Antarctic (Cape	BP-3	<LOQ- 3.7	

	Armitage, Winter Quarters Bay, Scott Base, and Cape Evans during 2012-2013)	4-MBC OMC	<LOQ- 5.8 <LOQ- 4.3	
Thawed sea ice	Antarctic (Cape Armitage, Winter Quarters Bay, Scott Base, and Cape Evans during 2012-2013)	BP-3 4-MBC OMC	<LOQ- 4.2 <LOQ- 4.3 <LOQ- 4.8	
Seawater (beach sites)	Japan (Kumamoto, winter)	OMC EHS	11-20 2.0-3.8	[84]
	Japan (Kumamoto, summer)	OMC EHS	210-1080 4.3-23.1	
Seawater (surface water)	United States (St. John Island)	BP-3	75000 (ng/L)- 1.395 (mg/L)	[31]
Seawater (surface water)	Hawaii (Oahu Island)	BP-3	<LOQ- 19200	
Seawater (surface water)	Hawaii (Maui Island)	BP-3	<LOQ	
Seawater (beach sites)	Spain (Valencia, Patacona beach)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	603 174 169 406 691 212 914 369	[79]
Seawater	Spain	BP-3 OC BM-DBM	692000 30000 72000	[70]
Seawater (beach sites)	Spain (Gran Canaria Island)	MBP	41.12- 544.9	[73]
Seawater	Spain	IMC 4-MBC OC OMC EHS HMS	88 <LOD 1100- 171000 1200 420 720	[69]
Seawater	Spain	OC OMC EHS HMS	14- 79000 10 34- 2500 1300	[16]
Seawater (coral ambient)	China (Hong Kong, Ung Kong, wet	BP-3 4-MBC OC	25.5- 26.1 <LOD 13.1- 13.2	[32]

	season)	OMC	<LOD	
		OD-PABA	15.1- 15.2	
	China (Hong Kong, Wu Pai, wet season)	BP-3	13.9- 14.0	
		4-MBC	<LOD	
		OC	11.8- 11.9	
		OMC	<LOD	
		OD-PABA	13.2	
	China (Hong Kong, Sharp Island, wet season)	BP-3	23.2- 25.6	
		4-MBC	<LOD	
		OC	9.6- 9.8	
		OMC	<LOD	
		OD-PABA	22.6 -22.7	
	China (Hong Kong, Sung Kong, wet season)	BP-3	12.9- 13.5	
		4-MBC	<LOD	
		OC	8.7- 9.0	
		OMC	<LOD	
		OD-PABA	14.8- 14.9	
	China (Hong Kong, Ung Kong, dry season)	BP-3	28.9- 29.2	
		4-MBC	<LOD	
		OC	14.1- 14.2	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Wu Pai, dry season)	BP-3	13.7- 13.8	
		4-MBC	<LOD	
		OC	10.7- 10.8	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, dry season)	BP-3	31.5- 31.9	
		4-MBC	<LOD	
		OC	13.2	
		OMC	<LOD	
		OD-PABA	<LOD	
Seawater (beach sites)	Korea (Gwangalli)	BP-3	n.d.- 17.3	[41]
		4-MBC	n.d.- 4.70	
		OMC	15.1- 70.5	
		OD-PABA	n.d.	
		EHS	n.d.- 16.7	
	Korea (Songjeong)	BP-3	13.5- 87.8	
		4-MBC	n.d.- 3.60	
		OMC	2.11- 10.9	
		OD-PABA	n.d.	
		EHS	n.d.- 11.7	
	Korea (Haeundae)	BP-3	8.48- 72.7	
		4-MBC	n.d.- 10.6	
		OMC	3.56- 15.6	
		OD-PABA	n.d.	
		EHS	4.25- 19.6	
Seawater (beach sites)	China (Hong Kong, winter)	BP-3	13.08- 70.55	[24]
		4-MBC	24.38- 74.50	
		OC	13.50- 53.86	
		OMC	34.71- 167.72	
		OD-PABA	<LOQ- 41.68	

Seawater (beach sites)	China (Hong Kong, summer)	BP-3 4-MBC OC OMC OD-PABA	27.36- 82.35 26.04- 67.78 15.50- 63.63 99.05- 191.67 <LOQ- 46.14	
Seawater (coastal areas)	China (Hong Kong, winter)	BP-3 4-MBC OC OMC OD-PABA	19.86- 32.47 20.39- 41.89 11.54- 43.95 34.15- 148.81 <LOQ- 25.42	
Seawater (coastal areas)	China (Hong Kong, summer)	BP-3 4-MBC OC OMC OD-PABA	26.03- 41.17 27.26- 46.26 12.72- 46.44 81.64- 182.12 <LOQ- 28.82	
Seawater (beach sites)	Spain (Puzol beach, Valencia)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	148 73 105 745 349 187 553 257	[78]
	Spain (Patacona beach, Valencia)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS	405 103 144 149 436 201 731 497	
Seawater (surface water)	Taiwan (Kenting National Park)	BP-3 4-MBC	18.8- 1233 2.40- 7.93	[87]
Seawater (beach sites)	Netherlands (Lac Bay)	BP-3 4-MBC OC	<10- 1540 <10 <20- 1950	[33]

n.d. Not detected

*** All compounds were not detected

† Performed using on-line SPE and UPLC-MS/MS

Table S2. UV filters occurrence in sediments and sand. Chronological order.

Kind of matrices	Location	Compounds	Concentration (ng·g^{-1} d.w.)	Reference
Coastal sediments	Spain (Valencia)	BP-3 EHS HMS	n.d. 13.3 n.d.	[53]
Coastal sediments	Lebanon (sewage outfalls along the El-Mina coastline)	OC OMC OD-PABA	79.0 9.0 9.0	[51]
Coastal sediments	Lebanon (commercial harbour and fishing harbour on the El-Mina coastline)	OC OMC OD-PABA	51.0 9.0 6.0	
Coastal sediments	Chile (Concepción Bay, San Vicente Bay and Coronel Bay)	BP-3 4-MBC OC OMC OD-PABA	n.d.- 1.42 n.d. - n.d -	[22]
Coastal sediments	Colombia (West coastline)	BP-3 4-MBC OC OMC OD-PABA	n.d.- 2.52 n.d.- 7.90 - n.d.- 17.8 -	
Sediments from 0, 10 and 20 cm depths	Spain (Cadiz Bay)	BP-3 OC	47, 26 and 38 53, 20 and 41	[38]
Coastal sediments (beach sediments)	Spain (Gran Canaria Island)	MBP	<LOD	[114]
Sediments close to marine outfalls at different distances from the coast		MBP	<LOD- 0.33	
Top 10 cm sediment layer	Slovenia (Novigrad)	BP-3	2.0	[54]
	Slovenia (Ankaran)		<LOD	
	Slovenia (Portorož)		<LOD	
Fjord sediments	Norway (along a transect from close to a WWTP discharge and southward)	BP-3 OC OMC OD-PABA	<LOD <LOD- 82.1 8.5- 16.4 <LOD	[61]
Different depths marine sediments	China (Hong Kong in the Victoria Harbour and Sai Kung)	BP-3 IMC 4-MBC OC OMC	0.05- 39.8 <LOD <LOD 0.04- 15.6 0.6- 447	[57]

		OD-PABA	1.5- 150	
		EHS	<LOD	
		HMS	<LOD	
		BM-DBM	4.3- 42.9	
	Japan (Tokyo Bay)	BP-3	<LOD	
		IMC	<LOD	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	0.3- 54.5	
		OD-PABA	0.8- 13.9	
		EHS	<LOD	
		HMS	<LOD	
		BM-DBM	2.5- 64.5	
Surface sediments	China (Pearl River estuary)	4-MBC	0.36- 3.68	[52]
		OC	6.26- 27.8	
		OMC	14.5- 81.6	
	China (fishing harbour)	4-MBC	2.16- 31.3	
		OC	18.1- 551	
		OMC	36.4- 456	
Surface and deep sea regions sediments	Italy (along the Adriatic Sea, Northern Adriatic)	BP-3	<LOD- 0.23	[93]
		OC	4.0- 40.7	
		OMC	1.0- 10.4	
	Italy (along the Adriatic Sea, Central Adriatic)	BP-3	<LOD- 0.1	
		OC	0.8- 33.7	
		OMC	0.9- 6.9	
	Italy (along the Adriatic Sea, Southern Adriatic)	BP-3	<LOD- 0.18	
		OC	0.9- 19.0	
		OMC	1.3- 10.0	
Different surface sediments	Spain (Huelva estuary, Cadiz Bay and Almeria coast)	BP-3	0.45- 1.5	[94]
		OC	0.73- 25.1	
		OMC	<LOQ- 26.2	
		EHS	2.3- 6.8	
		HMS	<LOQ- 9.7	
Surface sediments	China (along the Pearl River Estuary)	BP-3	n.d.- 4.0	[95]
		4-MBC	n.d.- 25.4	
		OC	6.2- 105.2	
		OMC	n.d.- 30.1	
		EHS	n.d.- 13.7	
		HMS	n.d.- 10.7	
Surface sediments	China (along the Pearl River Estuary)	BP-3	0.16- 1.07	[35]
		OC	91.7	
		OMC	22.4	
		OD-PABA	<LOQ	

Surface sediments (coral ambient)	China (Hong Kong, Ung Kong, wet season)	BP-3	6.1	[32]
		4-MBC	<LOD	
		OC	2.0- 2.2	
		OMC	<LOD	
		OD-PABA	3.4	
	China (Hong Kong, Wu Pai, wet season)	BP-3	9.7- 9.9	
		4-MBC	<LOD	
		OC	2.5- 2.6	
		OMC	<LOD	
		OD-PABA	4.3- 4.5	
	China (Hong Kong, Sharp Island, wet season)	BP-3	6.5- 6.6	
		4-MBC	<LOD	
		OC	2.7	
		OMC	<LOD	
		OD-PABA	4.9	
	China (Hong Kong, Sung Kong, wet season)	BP-3	8.0- 9.0	
		4-MBC	<LOD	
		OC	3.0- 3.1	
		OMC	<LOD	
		OD-PABA	8.0	
	China (Hong Kong, Ung Kong, dry season)	BP-3	3.4- 4.9	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Wu Pai, dry season)	BP-3	16.9- 17.1	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, dry season)	BP-3	8.1	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
Superficial sediments	Northwest Pacific Ocean (Laizhou Bay)	OC	<LOD- 25	[30]
		OMC	<LOD- 0.22	
		OD-PABA	<LOD	
		EHS	<LOD- 1.28	
		HMS	<LOD	
	Northwest Pacific Ocean (Bohai Sea)	OC	<LOD- 0.36	
		OMC	<LOD- 0.24	
		OD-PABA	<LOD	
		EHS	<LOD	
		HMS	<LOD- 0.06	
	Northwest Pacific Ocean (Yellow Sea)	OC	<LOD- 4.25	
		OMC	<LOD- 0.08	
		OD-PABA	<LOD- 0.004	
		EHS	<LOD- 1.35	
		HMS	<LOD- 0.94	
Beach sand	Spain (Valencia, Malvarrosa beach)	BP-3	1.0	[55]
		IMC	1.3	
		4-MBC	0.9	
		OC	8	

		OMC	2.1	
		OD-PABA	<LOQ	
		EHS	5.3	
		HMS	1.8	
	Spain (Valencia, Pinedo beach)	BP-3	<LOQ	
		IMC	<LOQ	
		4-MBC	<LOQ	
		OC	1.7	
		OMC	0.9	
		OD-PABA	<LOQ	
		EHS	2.6	
		HMS	1.06	
	Spain (Valencia, Patacona beach)	BP-3	<LOQ	
		IMC	<LOQ	
		4-MBC	<LOQ	
		OC	5.2	
		OMC	<LOQ	
		OD-PABA	<LOQ	
		EHS	1.8	
		HMS	<LOQ	
	Spain (Gran Canaria Island, Los Ingleses beach)	BP-3	<LOQ	
		IMC	1.2	
		4-MBC	2.0	
		OC	25	
		OMC	10	
		OD-PABA	<LOQ	
		EHS	12	
		HMS	4.9	
Beach sand	Spain (Patacona beach, Valencia)	BP-3	<LOD	[100]
		IMC	<LOD	
		4-MBC	4.9	
		OC	2.4	
		OMC	1.3	
		OD-PABA	0.52	
		EHS	4.7	
		HMS	4.8	
	Spain (El Saler beach, Valencia)	BP-3	3.3	
		IMC	1.1	
		4-MBC	6.7	
		OC	4.6	
		OMC	7.0	
		OD-PABA	1.7	
		EHS	6.9	
		HMS	7.1	
	Spain (Javea beach, Alicante)	BP-3	9.2	
		IMC	6.9	
		4-MBC	16.2	
		OC	8.2	
		OMC	14.0	
		OD-PABA	10.2	
		EHS	7.5	
		HMS	10.1	

	Spain (Maspalomas beach, Gran Canaria Island)	BP-3	10.2	
		IMC	6.5	
		4-MBC	13.8	
		OC	11.0	
		OMC	5.5	
		OD-PABA	5.8	
		EHS	5.9	
		HMS	7.4	
Beach sand	Spain (Galicia)	BP-3	<LOD- 33	[99]
		IMC	<LOD- 0.090	
		4-MBC	2.2- 206	
		OC	31- 454	
		OMC	0.21- 2.7	
		EHS	0.93- 609	
		HMS	1.6- 149	
	Portugal	BP-3	<LOD- 2.2	
		IMC	<LOD- 0.14	
		4-MBC	0.1- 1.2	
		OC	13- 35	
		OMC	0.46- 3.5	
		EHS	0.67- 1.8	
		HMS	0.34- 1.1	
	Spain (Gran Canaria Island)	BP-3	7.5	
		IMC	2.6	
		4-MBC	87	
		OC	670	
		OMC	54	
		EHS	83	
		HMS	34	
	Spain (Mallorca Island)	BP-3	<LOD- 0.85	
		IMC	<LOD	
		4-MBC	0.066- 1.0	
		OC	2.9- 20	
		OMC	0.45- 1.4	
		EHS	2.7- 6.9	
		HMS	3.5- 6.8	

Table S3. UV filters occurrence in marine biota. Chronological order.

Kind of matrices	Location	Compounds	Concentration (ng·g^{-1} d.w.)	Reference
Mussel (*M. galloprovinciali*)	France (Atlantic coast)	OC OMC	n.d.- 23 5- 45	[64]
Mussel (*M. edulis*)	France (Mediterranean coast)	OC OMC	n.d.- 7112 3- 256	
Mussels (*Mylitus galloprovincialis*)	Spain (Galicia)	OC	15- 20	[63]
Mackerel (*Scomber scombrus*)			18	
Dolphin liver (*Pontoporia blainvillei*)	Brazil (Espirito Santo)	OC	n.d.- 712 (ng·g^{-1} l.w.)	[65]
	Brazil (Rio de Janeiro)	OC	n.d.	
	Brazil (São Paulo)	OC	n.d.- 524 (ng·g^{-1} l.w.)	
	Brazil (Paraná)	OC	n.d.- 129 (ng·g^{-1} l.w.)	
	Brazil (Santa Carina)	OC	n.d.- 401 (ng·g^{-1} l.w.)	
	Brazil (Rio Grande do Sul)	OC	n.d.- 782 (ng·g^{-1} l.w.)	
Fish (*Striped bass*, marketed fish)	Taiwan	BP-3 EHS HMS	5.7 2.9 n.d.	[102]
Fish (Cod, marketed fish)		BP-3 EHS HMS	3.3 0.8 n.d.	
Fish (Salmon, marketed fish)		BP-3 EHS HMS	6.9 3.9 0.7	
Mussels (*M. galloprovincialis*)	Portugal (south of Portugal)	OC OMC OD-PABA	3992 1765 833	[67]
Antarctic clams (*Laternula elliptica*)	Antarctic (Winter Quarters Bay)	BP-3	9.2- 112	[10]
Sea urchin (*Sterichinus neumayeri*)	Antarctic (Cape Armitage)	BP-3	8.6	
Fish (*Trematomus bernachii*)	Antarctic (Cape Evans)	BP-3	<6.6- 14.1 (265- 1450 ng·g^{-1} l.w.)	
Fish liver (*Trematomus bernachii*)		BP-3	41.0 (1690 ng·g^{-1} l.w.)	

Fish (Red snapper, farmed fish, fillet)	China (Pearl River estuary)	BP-3 4-MBC OD-PABA BM-DBM	0.59 14.7 0.239 33	[62]
Fish (Red snapper, farmed fish, fish belly)		BP-3 4-MBC OD-PABA BM-DBM	0.80 41.5 0.36 52	
Fishes (Pomfret, Flounder and Osteomugi, tissues)		BP-3 4-MBC OD-PABA BM-DBM	n.d.***	
Fish (Goby, tissues)		BP-3 4-MBC OD-PABA BM-DBM	0.276 n.d. n.d. n.d.	
Fish (Hairtail, tissues)		BP-3 4-MBC OD-PABA BM-DBM	0.106 n.d. n.d. n.d.	
Squid (Sleeve- fish, tissues)		BP-3 4-MBC OD-PABA BM-DBM	0.408 n.d. n.d. n.d.	
Crustacean (Squilla, deshelled)		BP-3 4-MBC OD-PABA BM-DBM	1.520 n.d. n.d. n.d.	
Sea snail (Whelk, whole body)		BP-3 4-MBC OD-PABA BM-DBM	n.d. 0.2 n.d. n.d.	
Fish liver (*Gadus morhua*)	Norway (Oslofjord)	BP-3 OC OMC OD-PABA	(ng·g^{-1} w.w.) <LOQ- 1037 <LOQ- 11875 <LOQ- 36.9 <LOQ- 21.3	[61]
Shrimp (*Pandalus borealis*)		BP-3 OC OMC OD-PABA	(ng·g^{-1} w.w.) <30- 68.9 <10- 23.1 <20 <20	
Crab (*Carcinus meanas*)	Norway (north of WWTP in Sjøstrand)	BP-3 OC OMC OD-PABA	(ng·g^{-1} w.w.) <30 <10 <10 <20	
Mussels (*Mytilus edulis* and *Mytilus galloprovincialis* meat and intervalvular fluid) and fish (*Platichthys fesus*)	Portugal (Tagus estuary) and Italy (Po estuary)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS	n.d***	[104]

		HMS DHHB		
Mussels (*Mytilus edulis* and *Mytilus galloprovincialis,* meat and intervalvular fluid)	Spain (Ebro delta)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d. n.d. n.d. <LOQ <LOQ n.d. n.d. n.d. n.d.	
Fish (*Liza aurata*)	Portugal (Tagus estuary)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	<LOQ n.d. n.d. n.d. n.d. n.d. n.d. n.d. n.d.	
Clams (*Chamelea gallina*, meat and intervalvular fluid)	Spain (Ebro delta)	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d. n.d. n.d. <LOQ <LOQ n.d. n.d. n.d. n.d.	
Dolphin (*Pontoporia blainvillei*, mother, blubber)	Brazil (Rio de Janeiro, Sao Paulo and Ceará state)	4-MBC OC OMC OD-PABA	(ng·g^{-1} l.w.) n.d.- 47.5 n.d.- 113 n.d.- 85.0 n.d.- 3.15	[60]
Dolphin (*Pontoporia blainvillei*, mother, muscle)		4-MBC OC OMC OD-PABA	(ng·g^{-1} l.w.) n.d.- 855 n.d. 54- 67.5 n.d.	
Dolphin (*Pontoporia blainvillei*, mother, milk)		4-MBC OC OMC OD-PABA	(ng·g^{-1} l.w.) 17.5- 20.0 n.d. n.d.- 120 n.d.- 8.5	
Dolphin (*Pontoporia blainvillei*, mother, placenta)		4-MBC OC OMC OD-PABA	(ng·g^{-1} l.w.) n.d. n.d. n.d. 1385	
Dolphin (*Pontoporia blainvillei*, calf,		4-MBC OC	(ng·g^{-1} l.w.) n.d. n.d.	

blubber)		OMC	67.0	
		OD-PABA	n.d.	
Dolphin (*Pontoporia blainvillei*, calf, muscle)			(ng·g^{-1} l.w.)	
		4-MBC	250	
		OC	925	
		OMC	133	
		OD-PABA	36.5	
Dolphin (*Pontoporia blainvillei*, fetus, blubber)			(ng·g^{-1} l.w.)	
		4-MBC	n.d.- 97.0	
		OC	n.d.- 50.0	
		OMC	n.d.- 117	
		OD-PABA	n.d.- 67.5	
Dolphin (*Pontoporia blainvillei*, fetus, muscle)			(ng·g^{-1} l.w.)	
		4-MBC	n.d.- 170	
		OC	n.d.- 11130	
		OMC	69.0- 250	
		OD-PABA	n.d.- 155	
Dolphin (*Sotalia guianensis*, mother, blubber)			(ng·g^{-1} l.w.)	
		4-MBC	n.d.- 48.0	
		OC	n.d.- 220	
		OMC	n.d.- 205	
		OD-PABA	n.d.- 34.0	
Dolphin (*Sotalia guianensis*, mother, muscle)			(ng·g^{-1} l.w.)	
		4-MBC	230- 570	
		OC	970- 8310	
		OMC	70- 545	
		OD-PABA	n.d.- 1050	
Dolphin (*Sotalia guianensis*, fetus, blubber)			(ng·g^{-1} l.w.)	
		4-MBC	n.d.- 34.0	
		OC	n.d.	
		OMC	n.d.	
		OD-PABA	n.d.	
Dolphin (*Sotalia guianensis*, fetus, muscle)			(ng·g^{-1} l.w.)	
		4-MBC	60.0- 80.0	
		OC	115- 240	
		OMC	40.0- 85.0	
		OD-PABA	17.0- 26.0	
Fish (*Lutjanus argentimaculatus*, *Lutjanus stellatus* and *Epinephelus lanceolatus*. Farmed fishes)	China (Hong Kong)	BP-3	<LOD- 3.1	[58]
		4-MBC	<LOD	
		OC	<LOD- 5.4	
		OMC	<LOD- 12.7	
		OD-PABA	<LOD- 10.3	
Mussels (*Perna viridis*)		BP-3	<LOD- 10.3	
		4-MBC	<LOD	
		OC	<LOD- 8.8	
		OMC	<LOD- 51.3	
		OD-PABA	<LOD- 24.1	
Clams (*Mactra antiquata* and *Corbicula sp.)*		BP-3	9.7- 12.4	
		4-MBC	<LOD	
		OC	<LOQ- 11.6	
		OMC	24.6- 33.1	
		OD-PABA	18.7- 22.6	

Conch (*Babylonia sp.*)		BP-3 4-MBC OC OMC OD-PABA	<LOD- <LOQ <LOD <LOD <LOQ <LOQ	
Shrimp (*Penaeus monodon*)		BP-3 4-MBC OC OMC OD-PABA	<LOD <LOD <LOD <LOD <LOD	
Sea urchin (*Anthocidaris crassispina*)		BP-3 4-MBC OC OMC OD-PABA	<LOD <LOD <LOD <LOD <LOD	
Golden pompano (*Trachinotus ovatus*)	China (Pearl River estuary)	BP-3 4-MBC OC	(ng·g^{-1} l.w.) 5.04 8.0 20.0	[66]
Bigeye herring (*Harengula ovalis*)		BP-3 4-MBC OC BM-BMD	(ng·g^{-1} l.w.) 5.82 13.4 17.4 2	
Gray's grenadier anchovy (*Coilia grayii*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 4.19 10.7 7.9	
Black pomfret (*Formio niger*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 1.62 5.4 11.5	
Bombay duck (*Harpadon nehereus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 1.77 8.7 10.4	
Yellow drum (*Nibea albiflora*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.96 13.9 13.7	
Bighead croaker (*Collichthys lucidus*)		BP-3 4-MBC OC BM-DBM	(ng·g^{-1} l.w.) 5.11 9.3 13.7 3	
Smallhead hairtail (*Eupleurogrammus muticus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.12 8.9 27.2	
Bigeye snapper (*Lutjanus lutjanus)*		BP-3 OC	(ng·g^{-1} l.w.) 2.14 20	

Shortnose ponyfish (*Leiognathus brevirostris*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 0.68 8.4 25.2	
Taileyed goby (*Parachaeturichthys polynema)*		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.26 16.9 5.3	
Silver sillago (*Sillago sihama*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.94 10.9 30.5	
Half-smooth golden pufferfish (*Lagocephalus spadiceus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.22 5.9 12.8	
Pike conger (*Muraenesox cinereus*)		BP-3 4-MBC	(ng·g^{-1} l.w.) 8.88 5.7	
Rice-paddy eel (*Pisodonophis boro*)		BP-3 4-MBC OC BM-DBM	(ng·g^{-1} l.w.) 3.97 4.7 20.2 3	
Macao tonguesole (*Cynoglossus sinicus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 5.26 12.0 31.8	
Bluespot mullet (*Moolgarda seheli)*		BP-3 OC	(ng·g^{-1} l.w.) 9.99 18.6	
Musket squid (*Loliolus beka*)		BP-3 4-MBC OC OMC	(ng·g^{-1} l.w.) 2.40 6.0 10.4 13	
Bigfin reef squid (*Sepioteuthis lessoniana*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 9.04 38.9 28.1	
Sword prawn (*Parapenaeopsis hardwickii*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 1.24 2.0 3.3	
Kuruma prawn (*Marsupenaeus japonicus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.93 3.3 5.0	
Japanese stone crab (*Charybdis japonica*)		BP-3 4-MBC OC BM-BDM	(ng·g^{-1} l.w.) 43.40 2.3 5.8 21	

Blue swimming crab (*Portunus pelagicus*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 0.94 3.5 2.4	
Mantis shrimp (*Oratosquilla oratoria*)		BP-3 4-MBC OC	(ng·g^{-1} l.w.) 2.30 12.2 16.6	
Coral tissues (*Favites abdita*)♦	China (Hong Kong, Ung Kong, wet season)	BP-3 4-MBC OC OMC OD-PABA	8.0- 14.3 <LOD 1.5- 2.1 <LOD <LOD	[32]
	China (Hong Kong, Wu Pai, wet season)	BP-3 4-MBC OC OMC OD-PABA	14.1- 21.8 <LOD 2.0- 4.3 <LOD <LOD	
	China (Hong Kong, Sharp Island, wet season)	BP-3 4-MBC OC OMC OD-PABA	14.1- 21.8 <LOD 3.1- 4.9 <LOD <LOD	
	China (Hong Kong, Sung Kong, wet season)	BP-3 4-MBC OC OMC OD-PABA	9.5- 11.2 <LOD 1.8- 2.6 <LOD <LOD	
Coral tissues (Porites sp)♦	China (Hong Kong, Ung Kong, wet season)	BP-3 4-MBC OC OMC OD-PABA	10.6- 22.7 <LOD 2.9- 6.3 <LOD 6.0- 17.1	
	China (Hong Kong, Wu Pai, wet season)	BP-3 4-MBC OC OMC OD-PABA	9.4- 15.7 <LOD <LOD <LOD <LOD	
	China (Hong Kong, Sharp Island, wet season)	BP-3 4-MBC OC OMC OD-PABA	22.1- 38.4 <LOD 6.2- 7.0 <LOD 8.4- 14.8	
	China (Hong Kong, Sung Kong, wet season)	BP-3 4-MBC OC OMC OD-PABA	11.3- 24.2 <LOD 6.5- 8.7 <LOD 4.4- 14.7	
	China (Hong Kong, Ung Kong, dry season)	BP-3 4-MBC OC OMC OD-PABA	5.6- 14.7 <LOD <LOD <LOD <LOD	

	China (Hong Kong, Wu Pai, dry season)	BP-3	10.3- 11.3	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, dry season)	BP-3	4.7- 14.0	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
Coral tissues (*Pavona decussat*)♦	China (Hong Kong, Ung Kong, wet season)	BP-3	2.3- 5.1	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Wu Pai, wet season)	BP-3	9.4- 15.7	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, wet season)	BP-3	13.9- 26.6	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sung Kong, wet season)	BP-3	3.9- 16.4	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Ung Kong, dry season)	BP-3	2.1- 7.8	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, dry season)	BP-3	1.0- 5.6	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
Coral tissues (*Acropora valida*)♦	China (Hong Kong, Ung Kong, dry season)	BP-3	9.9- 12.3	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
Coral tissues (*Platygyra acuta*)♦	China (Hong Kong, Ung Kong, dry season)	BP-3	1.0- 5.7	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	
		OD-PABA	<LOD	
	China (Hong Kong, Wu Pai, dry season)	BP-3	4.8- 6.1	
		4-MBC	<LOD	
		OC	<LOD	
		OMC	<LOD	

		OD-PABA	<LOD	
	China (Hong Kong, Sharp Island, dry season)	BP-3 4-MBC OC OMC OD-PABA	2.2- 6.0 <LOD <LOD <LOD <LOD	
Mussels (*Mytilus galloprovincialis*)	Spain (Galicia)	BP-3 BP-4 4-MBC OC OD-PABA	<LOQ- 80 6- 739 <LOQ- 801 <LOQ- 833 <LOQ- 46	[115]
Fish liver (*Mugil liza*)	Brazil (Ipiranga)	BP-3 4-MBC OC OMC OD-PABA	11.8- 74.4 7.16- 13.7 <LOQ- 25.9 <LOQ- 9.53 <LOQ	[59]
	Brazil (Itaipu)	BP-3 4-MBC OC OMC OD-PABA	7.55- 50.6 <LOQ- 11.7 <LOQ- 11.6 <LOQ- 14.0 n.d.- <LOQ	
Fish (*Mugil liza*, fillet)	Brazil (Ipiranga)	BP-3 4-MBC OC OMC OD-PABA	3.5- 15.4 <LOQ- 23.4 <LOQ- 57.8 <LOQ- 49.4 <LOQ	
	Brazil (Itaipu)	BP-3 4-MBC OC OMC OD-PABA	<LOQ- 4.84 n.d.- 16.4 n.d.- 22.3 <LOQ <LOQ	
Fish (*Mugil liza*, gills)	Brazil (Ipiranga)	BP-3 4-MBC OC OMC OD-PABA	<LOQ- 6.62 <LOQ- 7.47 <LOQ- 9.96 <LOQ <LOQ	
	Brazil (Itaipu)	BP-3 4-MBC OC OMC OD-PABA	<LOQ- 24.0 4.42- 14.5 <LOQ- 16.8 <LOQ- 7.27 <LOQ	
Fish (Mackerel, canned)	Portugal	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d.- 5.0 5.0- 43.9 5.0- 17.5 n.d.- 18.5 n.d.- 2.5 n.d. n.d.- 48.1 n.d.- 5.1 n.d.	[103]
Fish (Tuna, canned)	Portugal	BP-3 IMC 4-MBC	n.d.- 27.6 n.d.- 5,5 5,0-5,0	

		OC OMC OD-PABA EHS HMS DHHB	n.d.- 57.6 n.d.- 65.4 n.d. n.d.- 13.8 n.d.- 10.4 n.d.	
Fish (Sardine, canned)	Portugal	BP-3 4-MBC OD-PABA DHHB	55.72 14.09 n.d. n.d.	
Fish (Salmon, aquaculture, fish fillet)	Denmark	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d.- 2,5 n.d.- 5 5,0-5,0 n.d.- 5 n.d.- 2.5 n.d. n.d.- 23 n.d.- 15.3 n.d.	
Fish (Seabream, aquaculture, fish fillet)	Italy and other origins	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d.- 5.0 5- 66.7 n.d.- 8 30- 103.3 n.d.- 2.5 n.d. n.d.- 42.9 n.d.- 33.4 n.d.	
Mussel (soft tissues)	Italy, Netherlands, Spain, Denmark, France and Ireland	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d.- 85.5 n.d.- 37.3 n.d.- 56.2 n.d.- 56.0 n.d.- 34.2 n.d. n.d.- 72.1 n.d.- 19.1 n.d.	
Octopus	Mediterranean	4-MBC OD-PABA DHHB	5,0- 5,0 n.d. n.d.	
Crab	Netherlands	4-MBC OD-PABA DHHB	5,0- 5,0 n.d. n.d	
Fish (Cod, fish fillet)	Denmark, Pacific	4-MBC OC OD-PABA EHS HMS DHHB	5,0- 5,0 n.d.- 39.1 n.d. n.d.- 26.7 n.d.- 2.5 n.d.	
Fish (Mackerel, fish fillet)	Italy, North Sea, Spain and Denmark	BP-3 IMC 4-MBC OC OMC OD-PABA	n.d.- 82.2 n.d.- 55.5 n.d.- 15.7 n.d.- 43.2 n.d.- 28.7 n.d.	

		EHS HMS DHHB	n.d.- 49.1 n.d.- 6.4 n.d.	
Fish (Monkfish, fish fillet)	Portugal	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	5.0- 98.7 n.d.- 5.0 5.0- 20.4 n.d.- 19.3 2.5- 74.4 n.d. n.d.- 15.3 n.d.- 54 n.d.	
Fish (Plaice/Sole, fish fillet)	Italy and North Sea	4-MBC OD-PABA DHHB	5,0- 5,0 n.d. n.d.	
Fish (Tuna, fish fillet)	Pacific	BP-3 IMC 4-MBC OC OMC OD-PABA EHS HMS DHHB	n.d.- 2,5 n.d.- 5.0 n.d.- 5,0 n.d.- 5.0 n.d.- 2.5 n.d. n.d.- 5.0 n.d.- 58.5 n.d.	

♦ Expressed in ng·g^{-1} ww.

□ Concentration measured after 30 days of exposition