Occurrence of PPCPs in the marine environment: a review

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Abstract

Little research has been conducted on the occurrence of pharmaceuticals and personal care products (PPCPs) in the marine environment despite being increasingly impacted by these contaminants. This article reviews data on the occurrence of PPCPs in seawater, sediment and organisms in the marine environment. Data pertaining to 196 pharmaceuticals and 37 personal care products reported from more than 50 marine sites are analyzed taking into account sampling strategies and analytical methods. A particular attention is given to the most frequently detected substances and at highest concentrations. A snapshot of the most impacted marine sites is provided by comparing the highest concentrations reported for quantified substances. The present review reveals that i) PPCPs are widespread in seawater; particularly at sites impacted by anthropogenic activities and that ii) the most frequently investigated and detected molecules in seawater and sediments were antibiotics, such as erythromycin. Moreover, this review points out other PPCPs of concern such as ultraviolet filters and underlines the scarcity of data on those substances despite recent evidence on their occurrence in marine organisms. Thus, the review discusses the exposure of marine organisms in regard to these insufficient data.
Keywords: pharmaceuticals, personal care products, marine environment, seawater, sediment, marine organisms

Introduction

The expression ‘pharmaceuticals and personal care products’ (PPCPs) encompasses many different substances, such as drugs used in human and veterinary medicine, fragrances, sun-screen agents and cosmetics ingredients (Daughton and Ternes, 1999; Richardson et al., 2005). More than 3000 PPCPs are currently marketed and new molecules enter the market yearly. Pharmaceuticals (PPs), after their therapeutic use, and some personal care products (PCPs), after regular use, are released in raw sewage waters and then often inefficiently treated in sewage treatment plants (STPs) and finally discharged in rivers, streams or lakes. These compounds are ubiquitously detected in treated wastewater, which is the main source of surface water contamination (Verlicchi et al., 2012). The PPCPs present in solid waste discharge, sewage sludge and in manure applied as fertilizer may leach from fields and, depending on their mobility in the soils, end up in ground waters (Daughton and Ternes, 1999; Diaz-Cruz et al., 2003). Some PCPs are also directly released into water during swimming and bathing activities in lakes or rivers (Balmer et al., 2005). PPCPs used throughout the world are thus widely detected in freshwater environments. Recent reviews have reported the occurrence of PPCPs in surface water (Brausch and Rand, 2011; Pal et al., 2010) and in groundwater (Lapworth et al., 2012).

Marine ecosystems are also affected by PPCP contamination (Klosterhaus et al., 2013) but little data has been published on PPCPs in seawater. However, coastal zones are subject to growing pressure due to the increase in human activities. PPCPs are released into seawater through submarine or marine STP outfalls (Fenet et al., 2014) or in runoff via rivers and streams (la Farre et al., 2008). Other sources of PPCPs in the marine environment are fish farming, for antibiotics (Zou et al., 2011) and antiparasitic drugs (Rico and Van den Brink, 2014), and recreational activities, especially regarding sun screen agents, which are washed from the skin into the water (Bachelot et al., 2012; Langford and Thomas, 2008). Fewer studies have been conducted in the marine environment because PPCP levels are expected to be low due to dilution and diffusion, and because of the complex hydrodynamics of the marine environment in coastal zones, for instance. Vidal-Dorsch et al. (2012) observed a significant dilution of STP effluent discharge in seawater at submarine outfalls and highlighted the difficulty to determine the environmental relevance of the low level PPCPs observed in seawater. However, several authors reported the occurrence of PPCPs in marine organisms (Klosterhaus et al., 2013; Nakata et al.,

2012). Hence, it is essential to evaluate the contamination range while identifying vulnerable marine sites so as enhance assessment of the exposure of marine organisms.
This paper reviewed current knowledge on PPCP contamination of the marine environment. Data pertaining to 196 pharmaceuticals and 37 personal care products reported from more than 50 marine sites were analyzed taking into account sampling strategies and analytical methods. A snapshot of the marine sites the most impacted is provided by comparing the highest concentrations reported for quantified substances to reveal areas of concern. The exposure of marine organisms was discussed through the scarce selected data on their PPCPs occurrence.

1- Framework used to record data on the occurrence of the investigated compounds

Although studies on the occurrence of PPCPs in freshwater have increased markedly over the past 20 years, studies in marine environment are more recent. Therefore, 71 articles providing data on the occurrence of PPCPs in seawater, sediment and marine organisms published since 2002 (Table A – supplementary material) were analyzed in the present work. The concerned areas were mainly placed in Asia (18 articles), North America (18 articles) and in southern Europe (16 articles), often located on the coasts, in areas influenced by STP effluents, bays, harbors and estuaries. Lagoons, gulfs and oceans were studied in less than five articles for each of them. Together, these studies target a total of 233 compounds (196 PPs and 37 PCPs), parent molecules and some of their metabolites. Data reported in the reviewed articles were analyzed by differentiating PPs and PCPs. PPs were classified according to their therapeutic classes. In the reviewed articles, the sampling strategies, analytical methods and studied molecules differed, which could appear to be a limitation for data comparison; these points are discussed below.

Study design

The sampling strategies deployed were highly dependent on the objectives of the reported studies. When the aim was to evaluate the spatial distribution of PPCPs within a defined area receiving a point source pollution (STP, rivers, etc.), samples were collected at regular distances from the source (Rubinfeld and Luthy, 2008), in different directions (Fang et al., 2012) or at different depths (Liang et al., 2013). Most often, concentrations were generally found to decrease with increasing distance from the pollution source such as an STP outfall (Amine et al., 2012; Minh et al., 2009), a bathing area (Langford and Thomas, 2008) or a more polluted area such as a river or an estuary (Andresen et al., 2007; Weigel et al., 2002), emphasizing the role of dilution in the marine environment. However, the observed concentration was also largely dependent on the coastal hydrodynamics as shown by Bayen et al. (2013). Indeed, these authors demonstrated that the long residence times created by the hydrodynamic resulted in highest concentrations of pollutants than those in the proximity to the STP reject. Depth was involved differently in the dilution from point sources of pollution. When contaminated fresh water comes from the bottom, as it is the case with submarine outfalls, Fenet et al. (2014) showed that a thermocline observed in summer limits the PPCP diffusion from the bottom to the surface. In estuaries, diffusion from fresh water carrying the PPCPs to the salt water was limited with depth, as observed by Liang et al. (2013), which reported that the total concentration of antibiotics of the Pearl River estuary was significantly higher in the surface water than in the bottom water layer during both dry and wet seasons. In marine water, tidal stage could have an influence on dilution and diffusion of a contaminant. However, Fair et al. (2009) have studied the influence of the tidal stage on triclosan concentrations in the Charleston Harbor, and found no significant difference between the low tidal stage and the high tidal stage. The synthesis that can be made from these results is that, even more than in the case of fresh water, the site conformation and hydrodynamics will determine the spatial distribution of concentrations found for PPCPs. The parameters which may influence the temporal variation of concentrations in seawater and sediments are seasons (dry or wet season) and recreational activities (tourism) as indicated by Liang et al. (2013) and (Knee et al., 2010).
Beyond the sampling strategy, the method used to sample the different compartments could have an impact on the results. In most of the reviewed articles (43), water was collected by active sampling with pumps or grab sampling. Passive integrative samplers for polar molecules (POCIS) were used by Alvarez et al. (2014), Martinez Bueno et al. (2009) and Munaron et al. (2012). Active sampling is easier to implement than passive sampling and enables direct quantification of PPCPs in water.

However, to be representative of the studied area, numerous samples should be collected and more if a temporal variation was researched. Passive sampling allows an estimation of the target analyte concentration during an exposure time, enabling the semi-quantification of some PPs depending on the sampling rate (Rs), predefined in the laboratory. However, harmonizing measurements during laboratory calibration is difficult (Munaron et al., 2012), which explains the low use of this technique at the moment.
Sediment depth sampled in the reviewed articles varied from 2-3 cm (Long et al., 2013; Maruya et al., 2012) to 40 cm (Miller et al., 2008) requiring different equipments. Surface sediment was sampled using a stainless steel Van Veen type grab sampler whereas sediment cores were sampled with gravity corers. The sample pre-treatment (freeze drying, homogenization, sieving) may influence the analysis efficiency. Belkessam et al. (2005) revealed that the increasing sample homogeneity improved analysis reproducibility and extraction recovery of PAH compounds. This influence observed for persistent contaminants could be studied for PPCPs, which should possibly have a different behavior. The storage may have an influence on the stability of the compounds. Negreira et al. (2014) showed that environmental samples such as wastewater had to be frozen immediately after collection and had to be analyzed as soon as possible in order to minimize the degradation of cytostatic drugs. The influence of freeze on PPCPs in sediments remains to determine. Most of the articles have studied the spatial variation of PPCP contamination in surface sediments whereas some authors have investigated the geochronology of the contamination by sampling sediment cores (Cantwell et al., 2010; Miller et al., 2008). The contaminant distribution in sediments varies in function to the sediment composition, which depends on sedimentation rates, and is variable in line with the characteristics of the studied sites (topography and hydraulic conditions). Beretta et al. (2014) reported the influence of the location in an area of low energy with high sediment rates on the composition of muddy sediments, composed predominantly of clay and silt, prone to become a sink of organic contaminants such as PPCPs. Moreover, transformation processes in sediment such as aerobic and anaerobic degradation are dependent on the location and depth in the sediment (Hektoen et al., 1995; Robinson and Hellou, 2009). This latter point may affect the rate of degradation and detection of PPCP transformation products (Cantwell et al., 2010).
Concerning marine organisms, sampling was performed mainly to assess the status of contamination of studied sites subjected to anthropogenic pressure. Bivalves (mussels, oysters, scallops and clams) were the most frequently studied organisms, by direct sampling (Bachelot et al., 2012; Dodder et al., 2014; Klosterhaus et al., 2013; Li et al., 2012; Martinez Bueno et al., 2014; Na et al., 2013; Nakata et al., 2007; Picot Groz et al., 2014) or via caging at sites of interest (Hoenicke et al., 2007; Kookana et al., 2013; McEneff et al., 2014; Wille et al., 2011). Bivalves, such as mussels have often been used as bioindicators for aquatic pollution monitoring (Quinete et al., 2009), and they are representative of the sampling area because of their sedentary behavior (De Donno et al., 2008), whereas fishes are less representative of a site. Fishes are highly mobile and may have a migratory behavior (Ferreira et al., 2009), thus complicating data interpretation. However marine mammals (dolphins, sea lions, otters, seals) and fishes (sharks, salmons, flatfishs, eelpouts, rays) were also studied in four papers (Kannan et al., 2005; Maruya et al., 2012; Nakata et al., 2007; Rüdel et al., 2006). In fish and marine mammals, liver was often sampled because biotransformation enzymes are mainly distributed in this organ (Matsuo et al., 2008; Teramitsu et al., 2000), while in mussels, all the soft tissues were mainly sampled, although xenobiotics are accumulated in the digestive gland, responsible for metabolic activity (Fasulo et al., 2012).

As a conclusion of this section, although several parameters influence the results of the analysis as well as in freshwater studies, the issue of the sampling site choice is a critical aspect of the study design and appears to depend mostly on the knowledge of hydrodynamic characteristics of the studied site.

Analytical procedures
Beyond the sampling strategies, one of the difficulties in reviewing research in this area is to compare data derived from various analytical procedures. The different analytical steps, such as the extraction process, quantification methods and the addition of an internal standard have consequences on the

analytical performances, such as the limit of quantification or repeatability. Multiple residues analyses were often performed in the reviewed articles. As regards the extraction steps for the aqueous samples, in the last 30 years solid phase extraction (SPE) has replaced the traditional liquid-liquid extraction (LLE) and has become the technique most commonly used for sample preparation. At present, hydrophilic-lipophilic-balanced-reversed-phase (HLB) sorbent is best used for the extraction of organic pollutants from water samples, due to its ability to retain analytes in a wide range of polarities and stability throughout the pH range. One of the great advantages of these adsorbents is the possibility of multi-analyte method work at neutral pH (Gomez et al., 2006), which greatly simplifies the treatment of the sample. However, for very polar and water-soluble analytes, such as ranitidine, recoveries may be low if the samples are not adjusted to the appropriate pH (Pichon, 2000).
For the analyses, liquid chromatography-mass spectrometry (LC-MS or LC-MS/MS) and gas chromatography-spectrometry (GC-MS or GC-MS/MS) were used to quantify concentrations in samples in 32 and 37 articles, respectively. However, matrix effects are widely described in LC-MS analyses of complex matrices (Petrović et al., 2005), and are most frequent than in GC-MS analyses (Caban et al., 2012). The use of an isotope-labeled standard in isotope dilution-based methods helps correct this issue (Hernandez et al., 2005). More broadly, the use of an internal standard (IS) can correct errors related to extraction processes, injection and ionization such as matrix effects. The use of an IS was reported in more than 80% of the papers reviewed, and a third of these 80% used isotopic dilution. Isotope dilution can significantly reduce systematic errors from several sources, including sample stability prior to analysis, analyte loss during extraction procedures and matrix effects (Boden and Reiner, 2004; Vanderford and Snyder, 2006). However, limited availability of isotope labeled standards is problematic for broad screening analysis, for some PPCPs and particularly for metabolites. For example, Martinez Bueno et al. (2014) analyzed metabolites of venlafaxine, N-desmethylvenlafaxine, in mussels by LC-MS/MS with venlafaxine-d6 as a surrogate because no isotope labeled standard is marketed for this venlafaxine metabolite. In the present review, the reported occurrence values in articles were selected when the use of an internal standard was reported.
Furthermore, the limits of quantification (LOQ) and limits of detection (LOD) for a molecule varied in the reviewed articles with the overall analytical procedures. For example, for sulfamethoxazole, LODs ranged from 0.084 (Yang et al., 2011) to 300 ng/L (Bayen et al., 2013). Thus, the authors are aware that when a detection frequency was calculated for PPCPs detected at the different sites reviewed in the present work, there was bias due to the difference in LODs at the different studies.
As presented above, there are a wide variety of analytical procedures applied to the environmental samples studied in the reviewed articles. A standardization of the analytical procedures, mainly for multi-residue analyses, could allow an accurate comparison of the results.

Data analysis considerations

As the authors are aware of the uncertainties and the limits of the comparison of the results in this review and as previously mentioned, regarding the available articles, for occurrence studies (detection frequencies and concentrations), data were selected only when an internal standard (surrogate or isotopic) was used. Furthermore, as recommended by the European Commission's 'Guidance documents on pesticide residue analytical methods' ("SANCO/825/00," 2010), the data were reported in this occurrence review, when the reported recovery ranged from 70% to 120% and the precision, expressed as the relative standard deviation (RSD), was lowest than 20 %. Furthermore, passive sampling data (POCIS) were not included in the occurrence study as we consider that data are semi-quantitative. Based on the selected data, the detection frequencies of studied PPCPs were calculated in aqueous phase by including all sampling sites of each study selected in the present review. The detection frequency for each PPCP expressed in % was calculated with the formula $(d/i) \times 100$, where d is the number of sampling sites where the specific compound was detected and i is the number of sampling sites where the compound was investigated. The frequency was calculated when a compound had been detected in at least ten sampling sites, and at least two distinct geographical areas. For some articles, information on detected but not quantified compounds was missing. In these cases, the reported frequency in the present review work should be considered as underestimated. However, the detection frequencies gave a good overview of the occurrence of PPCPs in the marine environment. For the presentation of the reviewed data, the maximum concentrations were chosen to

reflect the occurrence of the studied compounds as these values were the most commonly cited values in the reviewed articles. Moreover, maximum concentrations far from the limit of detection were likely more independent of the variability in the sensitivity of the analytical techniques than the concentrations close to the limit of detection, such as minimal concentrations. The median values would have been the preferred values as they were less impacted by the extreme values chosen, but these data were not available for all the reviewed studies. Thus, the results were expressed as follows: number of sampling sites where the molecule was investigated (s), detection frequency for all sites (f) %, and number of studies (n).

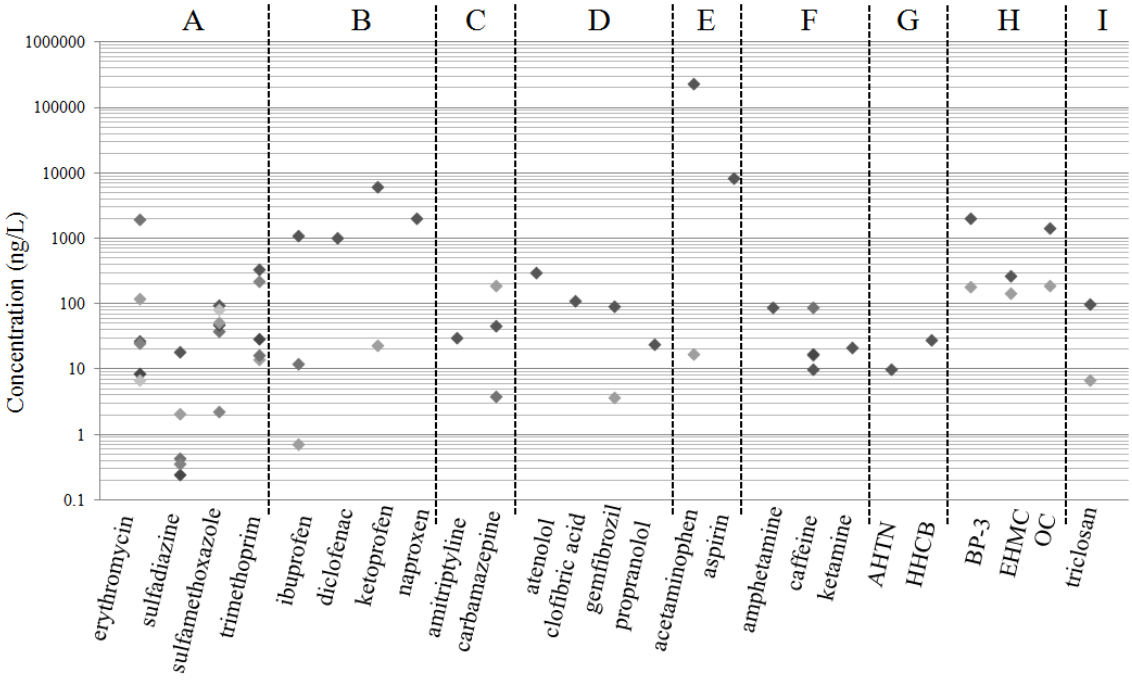
2- Occurrence in seawater

PPCPs were investigated worldwide in seawater in North America (Canada and USA), in northern Europe (United Kingdom (UK)), Ireland, Belgium, Germany, Netherlands, Denmark, Sweden and Norway), in southern Europe (France, Spain, Majorca, Portugal, Italy and Greece) and in Asia (China, Taiwan and Singapore) (Table A – supplementary material). For PPs, different trends between continents and countries could have been expected due to differences in prescription habits, authorized drugs and patient consumption. However the data were too scarce to study any difference between these continents and countries.

On the 100 investigated compounds (parents and metabolites) in water meeting the entered criteria, the half was detected. The most frequently investigated and detected compounds were antibiotics with erythromycin (s: 220; f: 77%; n:7), sulfamethoxazole (s: 251, f: 71%; n:9) and trimethoprim (s:168, f:80%; n:6), antiepileptics with carbamazepine (s: 85, f: 23%, n:5), caffeine (s:76, f: 86%, n:5), anti-inflammatories with ibuprofen (s:79, f:38%, n:5) and analgesics with acetaminophen (s:65, f:81%; n:3).

Maximum **PP** concentrations reported in seawater were in a large range of concentrations from the LOQ (few ng/L) to 230,000 ng/L (Fig.1 A-F). **Antibiotic** concentrations ranged from 0.24 to 1900 ng/L (Fig.1 A). The most frequently detected compounds were erythromycin, sulfamethoxazole and trimethoprim. The highest concentration was quantified for erythromycin, i.e. 1900 ng/L at the site immediately adjacent to the effluent discharge of a STP located in the typhoon shelter situated in Victoria Harbor (Hong Kong, China), where the water exchange to the outside sea is particularly low (Minh et al., 2009). **Non-steroidal anti-inflammatory drugs (NSAIDs)** (Fig. 1 B) were detected in seawater at maximal concentrations ranging from 0.7 ng/L to 6100 ng/L (Jiang et al., 2014; Togola and Budzinski, 2008; Weigel et al., 2004). Although the highest concentration was found for ketoprofen (s:60, f:68%,n:2), ibuprofen was the most frequently investigated and detected NSAID. For **central nervous system drugs** (Fig.1 C), carbamazepine was the most frequently detected compound in water (Jiang et al., 2014; Togola and Budzinski, 2008; Wille et al., 2010). Amitriptyline was less often screened and presented lower concentrations (30 ng/L) (Togola and Budzinski, 2008). Among **cardiovascular system drugs** (Fig.1 D), atenolol was detected at the highest level (293 ng/L) although its detection frequency was very low (s:64, f:3%, n:2). Gemfibrozil was the most frequently detected compound (s:68, f:40%, n:3). **Other less investigated classes** such as analgesics, analeptics and illicit drugs were reported in Fig.1 (E-F). The most frequently investigated and detected compound was caffeine. The highest levels were reported for acetaminophen (230,000 ng/L) and aspirin (8300 ng/L) in sampling sites close to the pollution source, at less than 500 m from an STP outfall (Marseille, France) without secondary treatment, where the effluent was rejected in a semi enclosed area, poorly submitted to dilution (Togola and Budzinski, 2008). Among **PCPs** (Fig.1 G-I), little data was available. Musks and disinfectants are rejected in seawater with STP discharges and were detected at sites influenced by anthropogenic pressures, such as estuaries or harbors. Musks AHTN (Tonalide®), HHCB (Galaxolide®) were detected at levels up to 28 ng/L for HHCB (Sumner et al., 2010). Triclosan was detected at concentrations up to 99.3 ng/L in Victoria Harbor waters (China) (Wu et al., 2007). Concerning UV filters, most of studies were conducted along the coasts, as recreational activities are the main source of contamination. Highest concentrations were reported for benzophenone 3 (BP-3) and octocrylene (OC) with 2013 ng/L and 1409 ng/L respectively in the most highly frequented beaches sampled by Bratkovics and Sapozhnikova (2011). Tovar-Sanchez et al. (2013) studied the intra-day variation in concentrations of sunscreen agents and found the maximum levels in seawater when the sunlight radiation was maximum, when the skin applications are highest.

This broad range of concentrations was noted for most of the investigated PPCPs. This could be due to the sampling sites (proximity of the sources of contamination with direct or indirect input) and sampling strategies (depth in the water column, etc.), the hydrodynamics of the investigated sites with various dilution and diffusion processes and probably to variable attenuation processes. Little data is available on these two latter points regarding PPCPs in the marine environment.

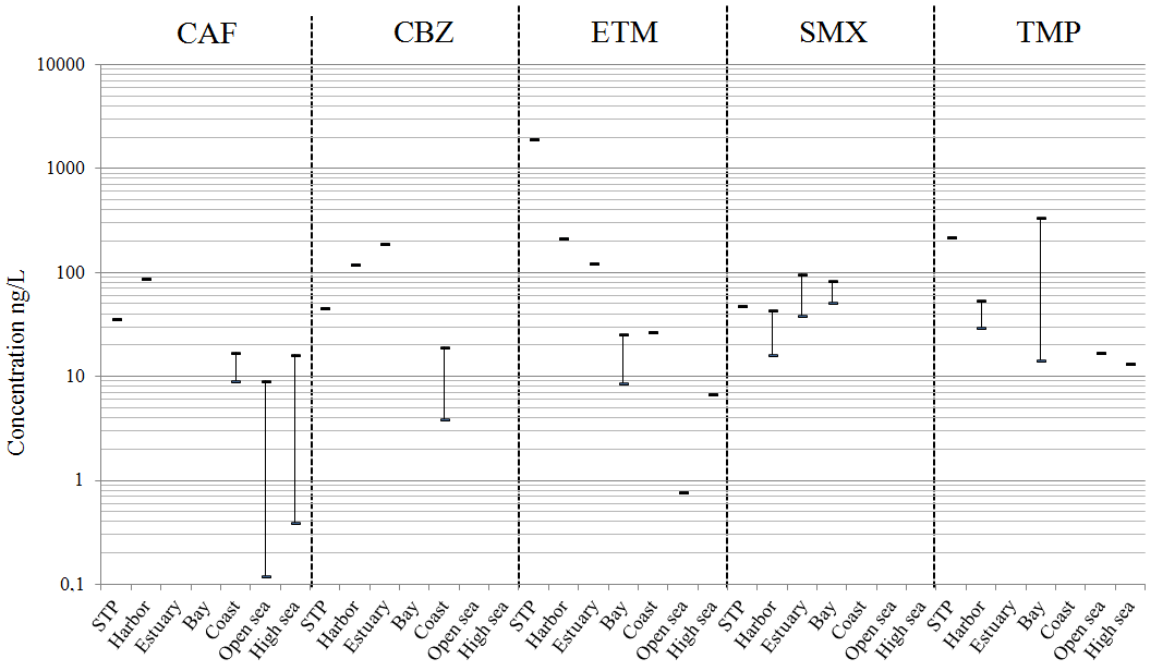


Benedé et al., 2014; Bratkovics and Sapozhnikova, 2011; Jiang et al., 2014; Knee et al., 2010; Liang et al., 2013; Minh et al., 2009; Na et al., 2013; Sumner et al., 2010; Thomas and Hilton, 2004; Togola and Budzinski, 2008; Weigel et al., 2002, 2004 ; Wille et al., 2010; Wu et al., 2007; Xie et al., 2008; Zhang et al., 2012, 2013a, 2013b

Figure 1: Maximum PPCP concentrations in seawater (ng/L). PPs: A-antibiotics/B-NSAIDs/C-central nervous system drugs/ D-cardiovascular system drugs /E-analgesics/ F-other drugs; PCPs: G-musks/H-UV filters/I-disinfectants

For the sampling sites, as data concerning the most frequently quantified compounds were available for numerous sites, the results are presented according to the nature of the sampling sites. As to Fig. 2, the sites were classified as sites heavily influenced by STP, harbors, estuaries, bays, coasts, open seas and high seas to study the influence of the distance to a source of pollution on PP concentrations. As one might expect, highest concentrations were detected in zones closely impacted by STP effluents, harbors and bays where urban activities were located and estuaries subject to surface water contamination. In open seas and high seas, concentrations were lower (up to 16.6 ng/L) due to the lower anthropogenic pressure and to higher dilution or advection mechanisms. For example, for erythromycin, the highest reported concentrations were reported at a site directly impacted by STP (Minh et al., 2009), then in a harbor and an estuary, with maximum levels ranging from 121 to 212 ng/L (Liang et al., 2013; Minh et al., 2009), whereas in bays, coasts, open seas and high seas, maximum concentrations ranged from 0.76 to 26.6 ng/L (Zhang et al., 2013a, 2013b, 2012). In marine water, as expected, the maximum concentrations of PPCPs reported in the literature were lower than those reported in freshwater. For example, carbamazepine was found at levels ranging from 9 to 2,000 ng/L in surface waters (Fent et al., 2006) whereas concentrations reported in marine waters ranged from 3.89 ng/L along the coasts (Jiang et al., 2014) to 185 ng/L in estuaries (Wille et al., 2010). Triclosan was detected with concentrations ranging from 0.1 to 1023 ng/L in freshwater (Bu et al., 2013; Gorga et al., 2013) and from 6.87 to 99.3 ng/L in marine water (Wu et al., 2007; Xie et al., 2008). Similarly, 2-ethyl-hexyl-4-trimethoxycinnamate (EHMC) was detected in seawater at levels of 144-264 ng/L (Benedé et al., 2014; Bratkovics and Sapozhnikova, 2011) whereas in freshwater, the

concentrations range was 7-3009 ng/L (Balmer et al., 2005; Rodil et al., 2009). In the same way, OC was detected in the reviewed studies with the lowest concentrations in seawater, ranging from 187 to 1409 ng/L (Benedé et al., 2014; Bratkovics and Sapozhnikova, 2011) and 5 to 4301 ng/L in freshwater (Kameda et al., 2011; Rodil et al., 2009).



Jiang et al., 2014; Knee et al., 2010; Liang et al., 2013; Minh et al., 2009; Na et al., 2013; Togola and Budzinski, 2008; Weigel et al., 2002, 2004 ; Wille et al., 2010; Zhang et al., 2012, 2013a, 2013b

Figure 2: Concentration ranges of caffeine (CAF), carbamazepine (CBZ), erythromycin (ETM), sulfamethoxazole (SMX), and trimethoprim (TMP) at different sampling sites in seawater: STP, harbor, estuary, bay, coast, open sea and high sea (ng/L)

3- Occurrence in sediment

Most of the studies have been conducted on the aqueous compartment insofar as most of PPCPs are molecules particularly water-soluble. However, some PCPs are more lipophilic compounds, such as OC or EHMC (log K_{ow} close to 6), and may be highly subject to sorption by sediment (Amine et al., 2012). A relationship between the degree of sorption and organic matter has been shown to some PPCPs. Sorption to sediment is a complex process influenced simultaneously by hydrophobic interactions (Van der Waals forces), ionic interactions (electrostatic interactions, complexation), hydrogen bonding etc., i.e. electrostatic forces are stronger than Van der Waals forces (von Oepen et al., 1991). More recently, studies investigated the sorption of PPs pointed out that the degree of ionization of the functional groups is a factor which influences the sorption on sediment. Numerous environmentally relevant PPs contain one or several dissociable functional groups (acidic/basic). Therefore, these compounds are in dissociation equilibrium depending on the seawater pH. Particularly for compounds with pK_a within the seawater 7.4-8.35 pH range (Byrne, 2002), a highly pH-dependent sorption behaviour could be expected (Franco et al., 2009; Franco and Trapp, 2008; Kah and Brown, 2007). Studies performed on the sorption process of antibiotics by Vasudevan et al. (2009) revealed that ciprofloxacin was able to mainly undergo cationic exchange and that its sorption was strongly dependant on the soil CEC at all tested pH values. Yamamoto et al. (2009) reported the existence and dominant role of cation exchange processes for atenolol, positively charged at neutral pH. However, studies on sorption processes of PPs were mainly conducted in freshwater-sediment systems. Few data are available on the sorption of PPs in marine sediments while the degree of salinity

is an additional factor that may control the PP sorption onto natural sediment. Wang et al. (2010), who studied the sorption of antibiotic tetracycline onto clays and marine sediment from seawater, found that sorption decreased with increased salinity.

A few studies have focused on PPCPs in marine sediment (15 papers between 2008 and 2014, Table A – supplementary material). PPCPs were investigated in North America in USA, in South America, in northern (UK) and southern Europe (France, Spain), in the Near East in Lebanon, and in Asia in China. These scarce publications showed that the marine sediment is also concerned by the presence of PPCPs. Twenty PPCPs (14 PPs and 6 PCP) were detected among thirty researched compounds, parents and metabolites (Table 1). Detection frequencies could not be estimated due to the lack of data. Among data selected as previously mentioned, the **PP** quantified at the highest levels was sulfameter (56.65 ng/g dry weight (dw)) in sediments located in the Dalian coastal environment (China) (Na et al., 2013). The reason could be that sulfameter had the highest sorption capacity among antibiotics investigated by Na and al. (2013).

Among **PCPs**, UV filters, musks and antimicrobials were detected and quantified. A general trend is difficult to highlight as it has been performed for aqueous compartment, because of the lack of data for marine sediments, however some studies highlighted, as expected, that the highest concentrations were mostly reported in zones impacted by anthropogenic activities. Cantwell et al. (2010) reported a high concentration of triclosan (400 ng/g dw) in the urbanized estuary of the Narragansett Bay (USA). It is acknowledged that STP effluents are the primary source of triclosan in estuarine sediments. Amine et al. (2012) studied levels and spatial trends of concentrations of three UV filters in coastal zones impacted by indirect inputs (rivers and STP), highest concentrations of UV filters were reported for OC (78 ng/g dw) in surface sediments receiving STP effluents along El Mina city coastline. Concerning musks, Beretta et al. (2014) detected AHTN and HHCB in sediments in the Todos os Santos Bay (Brazil), at highest levels, i.e. 52.5 ng/g dw for HHCB, at sites with the highest population density and the presence of muddy coastal sediments.

Despite the limited results available, it appeared that the concentrations found in marine sediments were lower than those observed in rivers sediments (Zeng et al., 2008). For example, sulfamethazine and triclosan were detected in river sediments in China at concentrations up to 248 ng/g dw (Yang et al., 2010) and 1329 ng/g dw (Zhao et al., 2010) respectively. In contrast, the maximum concentrations reported in this review were 3.24 ng/g dw for sulfamethazine and 400 ng/g dw for triclosan (Liang et al., 2013).

4- What about marine organisms?

Although the presence of PPCPs in organisms sampled in marine environment has been rarely reported in the literature, mostly because of the high polarity of some of them, these compounds (PCPs) were detected at levels up to 7112 ng/g dw (Bachelot et al., 2012). Thus, this aspect of PPCPs contamination in the marine environment has to be considered. Twenty articles were published between 2006 and 2014 and were reported in Table A – supplementary material. The occurrence of PPCPs in organisms has been documented in North America from San Francisco Estuary and Bay, along the California and Florida coasts to Charleston Harbor and South America along the Brazilian coasts, in Europe along the Portuguese, French, Ireland and Belgian coasts, in Asia and in Australia.

Ranges of concentrations detected in marine organisms are presented in Table 2A for PPs and 2B for PCPs. As regard to selected data, PPs were detected in bivalves. Antibiotics were the main represented pharmaceutical class with a little more than ten detected molecules. The highest concentrations were 490 ng/g dw for salicylic acid, 115 ng/g dw for acetaminophen in mussels *Mytilus edulis* of the Oostende marina in Belgium (Wille et al., 2011) and 19.9 ng/g ww for sulfamerazine in bivalves *Crassostrea gigas*, *Patinopecten yessoensis and Chlamys farreri* located at the Dalian coastal sites in China (Na et al., 2013). Martinez Bueno et al. (2014) detected the antidepressant venlafaxine and four of its metabolites in mussels *Mytilus galloprovincialis* sampled in the vicinity of a submarine outfall with industrial and urban discharges in Montpellier (France). Among PCPs, the highest concentrations were found in mussels *Mytilus galloprovincialis* and *Mytilus edulis* for OC and for the stabilizer UV-328 at levels up to 7112 ng/g dw at a the Mediterranean coastal site (Bachelot et al., 2012) and up to 14,000 ng/g lw in mussels *Perna viridis* and *Mytilus edulis* in Kohyongsong Bay in Korea (Nakata et al., 2012), respectively. Bachelot et al. (2012) reported highest levels of sunscreen agents in

recreational sites in summer period, in July and August. Picot Groz et al. (2014) observed similar seasonal trends of UV filters in mussels *Mytilus galloprovincialis* sampled along the Portuguese coasts, with highest concentrations in July. As noticed by Tovar-Sanchez et al. (2013), concentrations of UV filters in seawater vary throughout the day, depending on the arrival of bathers. This lead thus to an intra-day variation of organisms exposure which could be investigated through the measurement of concentrations in exposed organisms, helping to a better interpretation of pharmacokinetic studies concerning UV filters. However, for musks, a seasonal trend was not reported and their presence at sampling sites was due to the proximity of a STP (Picot Groz et al., 2014). Concerning marine mammals, Kannan et al. (2005) reported HHCB concentrations of 25 ng/g lw in striped dolphins and of 6.6 ng/g lw in pigmy sperm whales of the Florida coast.

Although PPs are found at relatively low levels in the environment, their possible risk for aquatic organisms is mostly related to chronic exposure. The real ecological risk of these compounds is thus difficult to apprehend. The studies show generally effects at concentrations highest than those reported in the environment. As an example, Huggett et al. (2002) observed a significant decrease in eggs production of the fish *Orizias latipes* after a 4-week exposition to propranolol at 500 ng/L which is higher than the maximum concentration observed in seawater of 52 ng/L (Wille et al., 2011). Recently, some studies reported results of toxicity at levels close to the environmental concentrations, which is subject of controversy. Di Poi et al. (2013) and Franzellitti et al. (2013) reported behavioral effects of antidepressant fluoxetine on the cuttlefish *Sepia officinalis* and transcriptional effects on the marine mussel *Mytilus galloprovincialis* respectively, both at concentrations of 1 ng/L or less. Sumpter and Margiotta-Casaluci (2014) have risen to question the relevance and the significance of these results in a commentary paper, and have highlighted the lack of pharmacological and pharmacokinetic data in aquatic organisms exposed to such molecules, helpful to confirm these results. Although pharmacokinetics have been studied concerning some marine species (Li and Randak, 2009), few studies have been published, mainly for invertebrates, for which it is difficult to know the fluid concentrations such as haemolymph in mussels (Franzellitti and Fabbri, 2014). Li and Randak (2009) showed that environmental conditions such as water temperature and salinity influence the pharmacokinetics in aquatic organisms. Metabolisation data are currently lacking to assess the loss of parent compounds by metabolite production. Ueno et al. (2001) studied the distribution of the antibiotic miloxacin in the fish *Anguilla japonica* and found the highest concentrations in kidney, in muscle and liver after oral administration. Smith et al. (2010) studied the hepatic metabolism of fluoxetine in freshwater fishes (rainbow trout, goldfish, killifish, zebrafish) and found that the metabolism was lower than in mammals, with differences noted regarding the metabolite nature and in proportions. Such studies have never been performed in marine organisms.

Concerning some PCPs, more lipophilic than PPs, their risk may be predicted by their bioaccumulation or bioconcentration potential. The main property which influences bioaccumulation is the hydrophobicity of the substance, expressed as log K_{ow}. The log K_{ow} values of the PCPs discussed in the present review range from approximately 5.0 to 7.5. Beyond chemical properties, several biological parameters may influence the bioaccumulation. Borga et al. (2004) reported the influence of lipids, seasonality, reproduction, body size, age, sex, life cycle, biotransformation, habitat use, migration, and feeding ecology in bioaccumulation of organochlorine contaminants in arctic marine organisms. Although PCPs have different properties, the influence of such parameters should be taken into consideration. A few studies have been conducted on the distribution and bioaccumulation of PCPs in aquatic organisms, especially in marine organisms. Gatidou et al. (2010) and Kookana et al. (2013) studied the bioaccumulation of triclosan in marine mussels and reported a log BCF of 3.23 and 2.81 L/kg dw, at exposition levels in water of 300 and 100 ng/L, respectively. Gomez et al. (2012) studied the accumulation kinetic of OC and EHMC during 48h in *Mytilus galloprovincialis* exposed by food at 0h and 24h, and observed higher concentrations 24h after the second spiking than the initial concentration. Bioaccumulation can be observed in marine mammals which may particularly accumulate hydrophobic compounds because they are at the top of the food chain (Teramitsu et al., 2000). Indeed, Kannan et al. (2005) detected higher concentrations for musks AHTN and HHCB in marine mammals than in fishes sampled in the California seawater. Considering the available data, it appears that the highest levels were reported for UV filters, which are able to bioaccumulate in organisms. However, for the most lipophilic compounds such as sunscreen compounds, it remains to

obtain in the same time the concentrations in seawater and in the organisms, to calculate a BCF and assess the exposure of the organisms to these contaminants. The presence of PCPs in marine mammals confirms their bioconcentration potential and their possible risk.
Further investigations on temporal trends, regional and global monitoring of PPCPs is needed to understand their environmental profiles and potential risks to wildlife. As little or no information was being gathered on chemicals of emerging concerns (CECs) on a national scale in US coastal ecosystems, the Mussel Watch Program recommended, since 2009, the inclusion of CECs (in particular PPCPs), as pilot study and regular monitoring efforts are done to assess the risk posed by these contaminants to the marine wildlife. The questions of occurrence (frequency of detection, concentration) and its variability were raised and factors as land use, proximity of wastewater treatment plant outflow and stormwater runoff were discussed. To answer these questions, native mussels were collected and analyzed at 68 sampling sites along the California coasts to characterize the spatial extent and temporal trends in contaminant concentrations (Bricker et al., 2014; Dodder et al., 2014; Maruya et al., 2014a, 2014b). The results showed that 30 out of 88 PPCPs were detected and that frequencies of detection of lomefloxacine and sertraline exceed 60%. Dodder et al. (2014) revealed a widespread occurrence of PPCPs in mussels without discernible trend with land use. Moreover, neither discharge of WTP nor discharge of stormwater runoff dominated the input. Among CECs, lomefloxacine was proposed to be included in future coastal bivalve monitoring efforts because of its relatively high concentrations and detection frequencies. This study demonstrates that PPCPs have to be taken into account in pollution monitoring programs. Such pilot studies should be conducted in other countries to better understand the local specificities of this contamination.

CONCLUSION

Over the last few decades, the characterization of the PPCP contamination in the marine environment and the identification of sources that lead to contamination have been made possible by the widespread availability of sensitive analytical techniques designed to detect environmental traces. However, studies concerning the quantitative analysis of PPCPs in marine ecosystems are still somewhat limited. Reported concentrations were generally lower than those reported in freshwater environments making the assessment of ecological risks caused by this contamination challenging. Despite the scarcity of quantitative data, the contamination of the marine ecosystem is a new reality that leads to the exposure of the marine wildlife species, confirmed by the data of occurrence in marine organisms available in literature. It is essential to acquire more accurate data on the fate of PPCPs in the marine ecosystem and the effects of PPCPs on marine organisms. The main knowledge gaps and research needs highlighted by the present study address the following issues.

1) Through the analysis of reviewed papers, some factors influencing the distribution of the PPCPs concentrations in marine environment were emphasized. The site conformation and hydrodynamic conditions will largely determine the spatial distribution of marine PPCP concentrations. These particular aspects should be considered when designing an occurrence study at a marine site.

2) A need for harmonizing analytical procedures for multi-residue analyses in aquatic environments was highlighted. It would allow more effective data comparisons and make the characterization of the PPCP contamination more accurate. Moreover, there is a lack of specific quality criteria for organic contaminant multi-residue analyses. More specific guidelines should be completed to obtain higher quality occurrence data for PPCPs.

3) Research projects to assess the fate of PPCPs in the marine environment should be conducted. Sorption studies on marine sediments are rare in literature. For some PPCPs, degradation processes need to be better understood. With these data, hydrodynamic models could help predict the behavior of such substances on a local scale, by integrating degradation and sorption parameters.

4) The assessment of any potential risk of PPCP exposure to marine species should be completed with supplemental studies. The ecological significance of ecotoxicity data extrapolated to natural marine systems is difficult to understand. There is thus a need to conduct long-term ecotoxicological tests on sensitive species at environmental levels. The use of sensitive approaches and informative techniques, such as "omics" approaches, already used for other pollutants such as heavy metals or pesticides (Kwon et al., 2012; Milan et al., 2013) could help obtain this information.

5) In the future, marine pollution monitoring programs conducted at local scales should, to the extent feasible, include PPCPs to improve understanding of the local specificities of the PPCP contamination and thus better inform future management decisions concerning the quality of the marine environment.

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Type	Class	Compound	Concentrations ng/g dw	References
PPs	Antibiotics	chloramphenicol	2.31	Na et al., 2013
		doxycycline	1.54	Na et al., 2013
		erythromycin	2.29-14	Beretta et al., 2014 ; Liang et al., 2013
		roxithromycin	13.5	Liang et al., 2013
		sulfacetamide	1.39	Na et al., 2013
		sulfadiazine	1.68	Na et al., 2013
		sulfamerazine	3.67	Na et al., 2013
		sulfameter	56.65	Na et al., 2013
		sulfamethazine	1.76-3.24	Liang et al., 2013; Na et al., 2013
		sulfamethoxypyridazine	7.67	Na et al., 2013
		sulfathiazole	1.89	Na et al., 2013
		tetracyclin	1.74-7.13	Liang et al., 2013; Na et al., 2013
	Antiepileptics	carbamazepine	4.81	Beretta et al., 2014
	NSAIDs	diclofenac	1.06	Beretta et al., 2014
		ibuprofen	14.3	Beretta et al., 2014
	Others	caffeine	23.4	Beretta et al., 2014
		diazepam	0.71	Beretta et al., 2014
PCPs	Antimicrobials	triclosan	400	Cantwell et al., 2010
	Musks	AHTN	17.3-27.9	Beretta et al., 2014 ; Zeng et al., 2008
		HHCB	44.5-52.5	Beretta et al., 2014 ; Zeng et al., 2008
	UV filters	EHMC	9	Amine et al., 2012
		OC	78	Amine et al., 2012
		OD-PABA	9	Amine et al., 2012

Table 1: Occurrence of PPCPs in marine sediments (highest concentrations in ng/g dw)
OD-PABA:octyl-dimethyl-PABA

Class	Compound	Mollusks ng/g	References
Antalgics	acetaminophen	115	Wille et al., 2011
	salicylic acid	490	Wille et al., 2011
Antibiotics	chloramphenicol	3.23*	Na et al., 2013
	doxycycline	1.57*	Na et al., 2013
	ofloxacin	65	Wille et al., 2011
	oxytetracycline	9.45*	Na et al., 2013
	sulfacetamide	2.10*	Na et al., 2013
	sulfadiazine	5.21*	Na et al., 2013
	sulfamerazine	16.69*	Na et al., 2013
	sulfamethazine	3.93*	Na et al., 2013
	sulfamethiazole	2.05*	Na et al., 2013
	sulfamethoxypyridazine	6.11*	Na et al., 2013
	sulfathiazole	2.16*	Na et al., 2013
Antidepressants	NDV	3.0	Martinez Bueno et al., 2014
	NODDV	3.5	Martinez Bueno et al., 2014
	ODV	3.7	Martinez Bueno et al., 2014
	venlafaxine	2.7	Martinez Bueno et al., 2014
Antiepileptics	carbamazepine	11	Wille et al., 2011
Betablockers	propranolol	52	Wille et al., 2011

Table 2A: Highest concentrations of PPs reported in marine organisms in the literature (ng/g, dry weight or wet weight*),
NDV: N-desmethylvenlafaxine, NODDV: N,O-didesmethylvenlafaxine, ODV: O-desmethylvenlafaxine

Class	Compound	Mollusks	Fishes	Mammals	References
Disinfectants	Triclosan	2578	-	-	Gatidou et al., 2010
Musks	AHTN	[2500]	1.7*	2.3*	Kannan et al., 2005; Nakata et al., 2012
	HHCB	12 ; [14,000]	6.6*	25*	Kannan et al., 2005; Nakata et al., 2012; Picot Groz et al., 2014
UV filters	EHMC	256-1765	-	-	Bachelot et al., 2012; Picot Groz et al., 2014
	OC	3992-7112	-	-	Bachelot et al., 2012; Picot Groz et al., 2014
	OD-PABA	833	-	-	Picot Groz et al., 2014
UV stabilizers	UV-320	[86]	-	-	Nakata et al., 2012
	UV-327	[300]	-	-	Nakata et al., 2012
	UV-328	[830]	-	-	Nakata et al., 2012

Table 2B: Highest concentrations of PCPs reported in marine organisms in the literature (ng/g, dry weight or wet weight* or lipid weight [ ])

Reference	Number of purchased compounds	Sampling	Type of water sampling	PP and/or PCP	Location	Site description
Alvarez et al., 2014	5	Water	POCIS	PP, PCP	USA	Eleven sampling stations along the California's San Francisco Bay and the Southern California Bight with urban and agricultural activities, stormwater discharges and STPs.
Amine et al., 2012	3	Sediment	-	PCP	Lebanon southern France	Sampling in coastal zones of three Lebanese rivers with urban and agricultural pressure, along the El-Mina coastline with a commercial harbor, a fishing harbor and four STPs. In France, sampling in coastal lagoons and in the vicinity of a submarine outfall.
Andresen et al., 2007	7	Water	Active	PCP	North Sea	Sampling sites in the German Bight, a region of special concern for fisheries, heavily influenced by shipping, fishing and recreational activities and by the river Elbe estuary.
Bachelot et al., 2012	3	Organism	-	PCP	France	Ten sites along the French coast, three in eastern France, seven in southern France, representing different anthropogenic pressures and geomorphologic configurations.
Bayen et al., 2013	4	Water	Active	PP, PCP	Singapore	Twenty four samples from eight sites around Singapore.
Benedé et al., 2014	8	Water	Active	PCP	Spain	Three sampling sites in the Palmira Beach (Majorca Island), the Malvarrosa Beach and the Pinedo Beach (Valencia).
Beretta et al., 2014	9	Sediment	-	P,PCP	Brazil	Seventeen sampling sites in the Todos os Santos Bay, receiving drainage from various watersheds and the main destination for the domestic and hospital sewage from the city of Salvador.
Bertin et al., 2011	1	Sediment	-	PP	Chile	Sampling at nine sites at the discharge locations of nine STPs distributed along the Pacific Ocean coastline of the Biobío region. STPs treat domestic and industrial waste waters with primary treatment mostly.
Bratkovics and Sapozhnikova, 2011	7	Water	Active	PCP	USA	Sampling at four sites along the Folly Beach, South Carolina, differing in access and in usage.
Cantwell et al., 2010	1	Sediment	-	PCP	USA	Samples collected in four urbanized estuaries situated along the Atlantic coast of the United States including Boston Harbor, Narragansett Bay, the Lower Hudson River and Chesapeake Bay.
Cavalheiro et al., 2013	9	Water	Active	PCP	France, Spain	Samples collected in the Gernika estuary (Bay of Biscay) and the Adour estuary (Bayonne).

Comeau et al., 2008	11	Water	Active	PP	Canada	Four samples in Halifax Estuary, three in Pictou Estuary, where both untreated and treated waste waters are released and two sampling points in the Cocagne Estuary, where there is no sewage release, but possibility of septic tank leaching.
del Rey et al., 2012	1	Water	Active	PP	USA	Fourteen samples along the Oregon coast, from Astoria in the North, to Brookings in the South, some with populated areas with pollution threats (STP, rivers) and some with sparsely populated areas with no identifiable pollution threats.
Dodder et al., 2014	86	Organism	-	PP, PCP	USA	Sixty-eight mussel stations along the coast of California. These stations were located in areas submitted to different pressures (receiving STP discharge, agriculture areas, urbanized areas and stormwater receiving areas).
Fair et al., 2009	1	Water, organism	Active	PCP	USA	Blood sampled from dolphins in Charleston and in Indian River lagoon. Eighteen samples were collected in estuarine surface waters of Charleston.
Fang et al., 2012	4	Water	Active	PP	Taiwan	Fourteen samples in Danshui Estuary where Bali STP discharges its effluents through an ocean outfall pipe. It receives untreated sewage from several industries and domestically treated and untreated wastewaters.
Fernandes et al., 2011	2	Sediment	-	PCP	Australia	Thirty sampling sites near the Barket Inlet mouth in South Australia, receiving effluents from the Bolivar STP.
Gago-Ferrero et al., 2013	1	Organism	-	PCP	Brazil	Fifty-six samples collected from dolphins found dead on beaches along the southern Brazilian coast (six sampling sites), impacted by domestic wastewaters.
Gatidou et al., 2010	1	Organism	-	PCP	Greece	Eighteen bivalve samples collected in the Thermaikos Gulf and the Lesvos Island.
Giokas et al., 2004	4	Water	Active	PCP	Greece	Samples collected along the coastal line of North Greece (Epirus), at a site of recreational and bathing activities.
Giokas et al., 2005	5	Water	Active	PCP	Greece	Sampling done in a closed gulf located in a popular touristic area.
Gulkowska et al., 2007	9	Water	Active	PP	China	Eight samples along Hong Kong coasts, one in Victoria Harbor, one farther away near an STP outfall, two as a reference location, and three in waters influenced by an STP outfall.
Hoenicke et al., 2007	8	Organism	-	PCP	USA	Mollusks deployed at 4 sites: Coyote Creek, Redwood Creek, Napa River, and Yerba Buena Island (San Francisco Estuary).
Jiang et al., 2014	31	Water	Active	PP, PCP	Taiwan	Fifty-three sampling stations in coastal south-western Taiwan, which

						includes heavily urbanized, industrial areas and activities such as animal husbandry and aquaculture.
Kannan et al., 2005	2	Organism	-	PCP	USA	Coastal waters of Alaska and California.
Klosterhaus et al., 2013	104	Water, organism, sediment	Active	PP, PCP	USA	Sampling in urbanized segments of San Francisco Bay influenced by municipal and industrial wastewaters.
Knee et al., 2010	1	Water	Active	PP	Hawaii	Samples at five sites along the north shore of the island of Kauai (Haena, Hanaley Bay, Hono Iki, Pier and Pavilion).
Kookana et al., 2013	2	Organism	-	PCP	Australia	Cages of mussels deployed at two sites along the Adelaide metropolitan coast. The sampling sites are adjacent to the Bolivar STP and to the Glenelg STP.
Langford and Thomas, 2008	4	Water	Active	PCP	Norway	Samples collected from sites with direct (bathing areas and marinas) and indirect sites receiving wastewater effluent) human influences.
Li et al., 2012	23	Organism	-	PP	China	Mollusk samples collected from nine coastal cities along the Bohai Sea. Most of sampling sites were large and busy seaports with a large population.
Liang et al., 2013	9	Water, sediment	Active	PP	China	Seven sampling sites located downstream of Pearl River Estuary, with four located in Shiziang channel, receiving runoff from the Guangzhou channel and the East River. Seven other sampling sites were distributed within the estuary.
Long et al., 2013	119	Sediment	-	PP, PCP	USA	Ten samples in Puget Sound and 30 samples in Bellingham Bay.
Magner et al., 2010	10	Water	Active	PP	Sweden	Eight samples collected in the central Bay of Stockholm, receiving effluents from Henriksdal STP. Four other samples were collected along the coast downstream of Himmerfjärdens STP.
Martinez Bueno et al., 2009	12	Water	POCIS	PP	Spain	Fish farm set up in the Mediterranean Sea in a coastal zone.
Martinez Bueno et al., 2014	5	Organism	-	PP	France	Thirty four mussel samples collected from different zones of the Mediterranean Sea in southeastern France (Marseille and Montpellier): a farming site, a lagoon influenced by human activities, in the vicinity of marine outfalls, and local markets.
Maruya et al., 2012	7	Sediment, organism	-	PP, PCP	USA	Four sites along the southern California coasts close to STPs, and a reference site.
McEneff et al., 2014	5	Water, organism	Active	PP	Ireland	Three sites: a control site and a two effluent exposure sites off the east and west coasts of Ireland, in an estuary and in a bay, respectively.
Miller et al., 2008	4	Sediments	-	PCP	USA	Sampling sites along the US East Coast near a wastewater outfall in the Chesapeake Bay watershed (Maryland) and in the Jamaica Bay (New

						York).
Minh et al., 2009	16	Water	Active	PP	China	Twenty samples in Victoria Harbor, mainly near the sewage effluent discharge points.
Montesdeoca-Esponda et al., 2012	5	Sediment	-	PP	Spain	Sampling close to a submarine outfall located in the southern region of Gran Canaria Island, discharging treated waste waters from an STP
Moon et al., 2011	5	Organism	-	PCP	Korea	Fifty two samples of bubbler from dolphins entangled in fishing nets along the Yellow Sea and the South Sea.
Munaron et al., 2012	21	Water	POCIS	PP	France	Thirteen samples along the Mediterranean coast, from the Italian border to the East to the Spanish border to the West.
Na et al., 2013	20	Water, organism, sediment	Active	PP	China	Twenty sampling sites along the Dalian coasts in the Yellow Sea.
Nakata et al., 2007	5	Organism	-	PCP	Japan	Samples from the Ariake Sea, the Chiba coast, Shizuoka coast, Ise Bay, Mikawa bay, Taiji coast, Seto Inland Sea, Fukuoka coast and the Omura Bay.
Nakata et al., 2012	6	Organism	-	PCP	Cambodia, China, India, Indonesia, Japan, Korea, Malaysia, Philippines, USA, Vietnam	Coastal waters: harbors, fisheries, aquaculture, commercial, industrial, farming, residential areas and open coastal sites.
Nguyen et al., 2011	6	Water	Active	PCP	Italy	Ten samples, one blank at the Surla Beach in Genoa, nine others were collected in Santa Margherita, San Fruttoso and Camogli (Ligurian coast).
Picot Groz et al., 2014	9	Organism	-	PCP	Portugal	Four different beaches in the south of Portugal with different recreational and tourist population pressures.
Pintado-Herrera et al., 2014	2	Water, sediment	Active	PCP	Spain	Sampling sites in the province of Cadiz, in the Guadalete River estuary receiving urban, agricultural and industrial effluents discharges.
Rocha et al., 2013	2	Water	Active	PP	Portugal	Nine sampling sites in the Ria Formosa Lagoon along the coast covering urban centers and a protected natural park area.
Rubinfeld and Luthy, 2008	2	Water, sediment	Active	PCP	USA	Sediments collected from 14 sites located in the South San Francisco estuary intertidal zone. Water samples were collected from the outfall

						pipe of an local STP and the surrounding surface water.
Rüdel et al., 2006	8	Organism	-	PCP	North Sea, Baltic Sea	Samples from Environmental Specimen Bank (ESB) of mussels and fishes eelpout.
Siegener and Chen, 2002	1	Water	Active	PP	USA	Boston Harbor and Massachusetts Bay.
Singh et al., 2010	5	Water	Active	PP,PCP	USA	Eight samples in Kay Largo Harbor, largest island of Florida Keys, with several tank systems and 40 sampling points in Looe Key, a popular reef for scuba diving and snorkelling.
Sumner et al., 2010	6	Water, sediment, SPM	Active	PCP	UK	Eleven sampling sites in the Tamar Estuary, influenced by agricultural land runoff, discharges from STPs serving the city of Plymouth and commercial and industrial shipping activities in the area.
Thomas and Hilton, 2004	14	Water	Active	PP	UK	Twenty two samples in estuaries of the rivers Tyne, Tees, Mersey, and Thames and Belfast Lough.
Togola and Budzinski, 2008	17	Water	Active	PP	France	Samples in Cortiou rocky inlet (Marseille) in the Mediterranean Sea in the effluent plume of a wastewater treatment plant (no biological treatment).
Tovar-Sanchez et al., 2013	2	Water	Active	PCP	Majorca	Sampling in two areas corresponding to semi-enclosed and densely populated beaches and a third considered as a control, an open and scarcely used beach.
Vidal-Dorsch et al., 2012	20	Water	Active	PP, PCP	USA	Samples collected at five stations in the Southern California Bight, four located near municipal wastewater outfall effluent discharge areas and one from a reference area.
Weigel et al., 2002	2	Water	Active	PP	Denmark, Norway, UK, Netherlands	Fifteen samples in the North Sea.
Weigel et al., 2004	13	Water	Active	PP, PCP	Norway	Two samples in Breivika Harbor, one off the Tromso STP, two 300 m north of an STP, two in a marina, three on a sewage plume (upstream, downstream) and two reference points in the open North Atlantic Ocean. Treated STP and untreated effluents are discharged.
Wille et al., 2010	13	Water	Active	PP	Belgium	Eleven harbors (Ostend, Nieuwpoort and Zeebrugge), six offshore and two Schedt Estuary samples.
Wille et al., 2011	11	Organism	-	PP	Belgium	Cages of mussels from the Eastern Schedt placed in the marina of Nieuwpoort, Oostende, Zeebrugge, and in the open sea.
Wu et al., 2007	1	Water	Active	PCP	China	Sampling in Victoria Harbor and Tai Po Harbor (Hong Kong).

Xie et al., 2008	1	Water	Active	PCP	Germany, Denmark and Netherlands	Twenty four sampling sites in the German Bight.
Yang et al., 2011	9	Water, Sediment, SPM	Active	PP	China	One sample in the Yangtze River Estuary, two in coastal waters, and three around an STP (upstream, downstream and in the STP outfall).
Zeng et al., 2008	6	Sediment	-	PCP	China	Three sampling sites along the Macao coast, a know pollution deposition zone in the Pearl River Delta.
Zhang et al., 2012	13	Water	Active	PP	China	Twenty seven samples in Laizhou Bay (15 in inner bay and 12 in outer bay).
Zhang et al., 2013a	11	Water	Active	PP	China	Sixty two samples collected in the Bohai Sea semi-enclosed sea and in the Yellow Sea (open sea) where coasts have dense human populations, developed industries and agriculture, including animal husbandry and aquaculture.
Zhang et al., 2013b	11	Water	Active	PP	China	Fifty-six samples in Jiaozhou Bay and in Yantai Bay, close to urbanized areas with agriculture, fish farming, and industries.
Zheng et al., 2012	11	Water	Active	PP	China	Thirty-five samples in Beibu Gulf where aquaculture is developed.
Zou et al., 2011	21	Water	Active	PP	China	Twenty-eight samples in Bohai Bay, which receives wastewaters and municipal sewage from the city of Beijing

Table A: Main characteristics of sites where PPCP contamination was reported in the papers reviewed (PP: pharmaceutical, PCP: personal care product, SPM: suspended particulate matter, UK: United Kingdom, USA: United States of America)

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