The English Channel: contamination status of its transitional and coastal waters

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Abstract

The chemical contamination (organic compounds, metals, radionuclides, microplastics, nutrients) of English Channel waters has been reviewed, focussing on the sources, concentrations and impacts. River loads were only reliable for Pb, whereas atmospheric loads appeared robust for Cd, Pb, Hg, PCB-153 and γ-HCH. temporal trends in atmospheric inputs were decreasing. Contaminant concentrations in biota were relatively constant or decreasing, but not for Cd, Hg and HBCDD and deleterious impacts on fish and copepods were reported. However, data on ecotoxicological effects were generally sparse for legacy and emerging contaminants. Intercomparison of activity concentrations of artificial radionuclides in sediments and biota on both Channel coasts was hindered by differences in methodological approaches. Riverine phosphate loads decreased with time, while nitrate loads remained uniform. Increased biomass of algae, attributable to terrestrial inputs of nutrients, has affected benthic production and shellfisheries. A strategic approach to the identification of contaminant impacts on marine biota is recommended.

Keywords: English Channel, organohalogens, metals, radionuclides, microplastics, nutrients

Contents

1. Overview of the English Channel environment

1.1 Rationale

The first holistic assessment of the English Channel was given in the North Sea Task Force Sub-Region 9 Assessment (Reid et al., 1993) followed by an update by Tappin and Reid (2000), where particular emphasis was given to contaminants and their effects. In 2000 and 2010, Quality Status Reports (QSR) for the Oslo and Paris Commission (OSPAR) maritime area (essentially part of the NE Atlantic) were published (OSPAR, 2000; 2010a). In those reports, the English Channel was subsumed into the Greater North Sea, and its presence was overshadowed by the larger, and arguably, physico-chemically different North Sea partner. Nevertheless, the overview provided in the current paper has been guided by the previous QSRs, notably in respect of contaminants and their impacts i.e. the OSPAR Convention Thematic Strategies on hazardous and radioactive substances, and eutrophication, in relation to human uses and impacts. This approach also addresses issues of concern within the EU Marine Strategy Framework Directive (i.e. quality descriptors concerning concentrations of contaminants, contaminants in fish and other seafood, and human-induced eutrophication; Law et al., 2010) and within the EU Water Framework Directive. There are 8 groups of priority contaminants listed in the hazardous substances theme, of which 6 are discussed in this paper; these can loosely be described as ‘legacy’ contaminants. Tri-butyl tin is not included here as this is the subject of a separate paper in this Special Issue. There are also additional sections which serve to highlight emerging contaminants, including metals and micro – plastics, and their potential impacts on biota within the Channel.

1.2. Physical characteristics

The western limit of the English Channel is a line from 48°38′23″N 4°34′13″W to 50°04′N 5°43′W (essentially Ushant to the Scilly Isles) and the eastern limit (the Dover Strait) a line joining the Walde lighthouse (France) at 51^{o} 00' N 1°55'E and Leathercoat Point (England) at 51°10'N 1^{o} 55'E (IHO, 1953). It is widest in the west (200 km) and narrows to 30 km at the Dover Strait to give an overall area of 77000 km^{2} (Fig. 1). The Channel is also deepest in the west, the Hurd Deep is 174 m, and it shallows eastward (Fig. 1). Although the Channel region has many distinguishing geological features between the British and French catchments and coasts, from the viewpoint of its Quaternary history it can be considered as an integrated system (Gibbard and Lautridou, 2003).

There is considerable annual variability in salinity and temperature of the seawater, giving rise to the advance and retreat of the seasonal thermocline, as illustrated in Fig. 2a-e (Uncles and Stephens, 2007). Winds are predominantly from the south west and can produce long-period swell waves whose energy is dissipated before reaching the Straits of Dover, in the form of flooding which augments the spread of contaminants (Sibley and Cox, 2014). Over the long-term, coastal flooding will also be accentuated by predicted rises in sea level, which have been forecast from the records of sea level obtained from 16 stations around the English Channel coast (Haigh et al., 2011).

As a consequence of the persistent south west winds, residual currents generally flow from west to east (Fig. 2f), although locally current direction is complex, especially around headlands (Salomon and Breton, 1993; Bailly du Bois and Dumas, 2005). Tidal and density-driven currents are also responsible for seabed stresses that influence the transport and grain size distribution of mobile sediments, although within the near shore, wind-induced currents

become more important in this respect (Uncles and Stephens, 2007). As a result, sand and gravel predominate throughout the area, with rock outcrops nearer shore. In the central eastern English Channel gravel dominates due to high current scour (Fig. 2g). Locally, fine sands and muds predominate in estuarine bays and tidal flats, notably in the Baie de Seine, parts of the Brittany and Picardie-Artois coasts, and along the east Devon, Sussex and Kent coasts of England (Larsonneur et al., 1982). Sources of suspended particulate matter (SPM) to the Channel include rivers, coastal erosion, the Celtic Sea to the west, bed sediment resuspension and autochthonous biogenic production. River inputs of sediment are dominated by the River Seine and are of a similar order to the erosion of cliff sediments along the French coastline. Concentrations of SPM are highest in winter and are dominated by coarse silt (clay aggregates, clastic minerals), while spatially the coastal waters exhibit the highest concentrations of SPM. Mass balance considerations suggest most SPM is advected through the Dover Strait into the North Sea (Paphitis et al., 2010).

1.3 Catchments and anthropogenic pressures

The average population is ca 500 and ca 230 inhabitants km^{-2} along the English and French coasts respectively, and there is a general increasing density of population from west to east (Fig. 1). There is a wide range of economic activity, from heavy industry to agriculture, as exemplified in Fig. 1. Because of anthropogenic pressures, many transitional (= estuarine) and coastal waters are subject to contamination from point and diffuse sources, in particular the Brittany and Artois-Picardie coasts, as shown in Fig. 3(a). In addition, and as shown in Fig. 3(b), > 90 % of French transitional and coastal waters east of Sud Contentin fail to achieve EU Water Framework Directive (WFD) defined good chemical status (ETC/ICM, 2012). Counter-intuitively, while > 90 % of the south-eastern Channel coast of England has its transitional and coastal waters classified as good chemical status, > 90 % of

the waters are classified as less than WFD good ecological status, suggesting factors other than chemical contamination are affecting water quality (ETC/ICM, 2012). River inputs to the Channel occur from five River Basin Districts (RBD), the South West and the South East RBD in England and the Loire, Seine and Scheldt RBD in France (European Environment Agency, 2014). Many of these rivers are monitored for flow and chemical concentrations under the OSPAR Comprehensive Study on Riverine Inputs and Direct Discharges (RID) programme (OSPAR, 2013a). The rivers and their regional locations are given in Table 1 and shown in Fig. 3. The total long-term mean river flow into the Channel from both coasts is ca 1250 $m^3 s^{-1}$, of which 84 % is from the French coastline and 41 % from the Seine alone.

Offshore industry can also impinge on water quality. A recent economic and social analysis by OSPAR classified the following sectors as important for marine management: ports and shipping, commercial sea fisheries, tourism and recreation, aquaculture, oil and gas, renewable energy and aggregate extraction (OSPAR, 2013b). For example, there are 400-500 daily ship movements through the English Channel, and within the last 50 years there have been nearly 200 shipping incidents involving sinking, loss of cargo and contamination/pollution (Fig. 4; e.g. Law and Allchin, 1994; Law et al., 2003; Mamaca et al., 2009; Dauvin, 2012). Ships are also a source of anti-fouling agents to the water column, including metals and organo-metallic compounds, as well as aromatic hydrocarbons (OSPAR, 2009b, 2010a). A number of sites are used for the dumping of sediments dredged from ports and harbours, and less frequently, estuaries. These activities can redistribute historically buried contaminants, including metals, halogenated organics and aromatic hydrocarbons, among others (OSPAR, 2013c). There are also ca. 50 legacy munitions dump sites, mostly along the French Channel coast (Fig. 4), with the potential for the dispersal of hazardous substances (OSPAR, 2010b).

Following the introduction of the EU Marine Strategy Framework Directive (MSFD, Directive 2008/56/EC), participating countries were required to develop strategies to achieve Good Ecological Status (GES) of their marine waters by 2020 and in doing so allow the Cost of Degradation of marine resources to be evaluated (OSPAR, 2013b). The MSFD does not set out specific measures to be adopted to achieve GES, except for the establishment of Marine Protected Areas (MPAs) as required by the OSPAR Convention (OSPAR, 2013d). The MSFD is relevant to the Exclusive Economic Zone (up to 200 nautical miles) of the participating country and complements the EU WFD (covering transitional and coastal waters up to 1 nautical mile), and provides a framework for other European Directives and UK legislation. Thus, the MSFD should ensure improved integration of coastal zone management, protection of the marine and socio-economic activities. Most of the MPAs in the English Channel are already designated as Special Protection Areas and Special Areas of Conservation (OSPAR, 2013d). Implementation of the MSFD is work in progress.

2. Organic Contaminants

2.1. Halogenated compounds

2.1.1 Polychlorinated compounds

2.1.1.1 Polychlorinated biphenyls

The use of polychlorinated biphenyls (PCBs) in electrical and electronic equipment has been severely restricted under EU and other international legislation for a number of decades (OSPAR, 2010a). As a result, releases of PCBs to the aquatic environment appear to be decreasing. While there appear to be no estimates of measured atmospheric loads of PCBs to the Channel (OSPAR, 2009c, 2013e), the net (deposition minus re-emission; OSPAR, 2009c) modelled loads of PCB - 153 fell from 24 kg a^{-1} in 1990 to 4 kg a^{-1} in 2006 (Fig. 5). In contrast to the atmospheric inputs, trends in total PCB loads from Channel rivers are more difficult to discern. The OSPAR RID programme reported relatively large loads in the early 1990s, with the R. Seine dominating the input; however, information on the Seine has not been reported to the RID programme since 1996. The most recent data show PCB loads from the UK Channel coast to be up to ca. 40 kg for 2011, an order of magnitude larger the atmospheric loads of 2006 (Fig. 5).

Predicting the fate of PCBs in aquatic systems is conditional on their tendency to partition between the water and various suspended and deposited solid phases. Cailleaud et al. (2007a) determined the concentrations of PCBs 52, 87, 101, 118, 138, 153, 180 and 187 in solution, SPM and in the copepod *Eurytemora affinis* from the Seine Estuary. The highest mean dissolved concentration was for PCB - 101 (1.4 ng L^{-1}), whereas PCB-153 had elevated concentrations in SPM (mean, 39 ng g^{-1}) and *E. affinis* (mean, 203 ng g^{-1} dry wt). Partitioning of PCB - 153, as defined by the partition coefficient K_d, was of the order 5.6 x 10^4 L kg^{-1} for

SPM and 2.1 x 10^5 L kg^{-1} for *E. affinis*. Other PCBs showed similar trends, with consequences for the uptake of PCBs into the food web of the Seine Estuary.

Elevated values of the OSPAR priority PCBs (the ICES 7 congeners 28, 52, 101, 118, 138, 153, 180) have been reported for sediments, fish and shellfish in the English Channel, as summarised in Table 2. Concentrations appear to be generally decreasing, particularly along the French coast (Table 2; OSPAR, 2012b, 2013f); however, the temporal trends of concentrations of the sum of the 7 PCBs in samples of mussel tissue from the coastal region near the mouth of the Seine, at Villerville, showed that there was no systematic decline, at least up to 2005 (Fig. 6a). Compositional profiles of PCB contamination in particulate matter generally matched those in the associated marine biota. For example, tissue-specific bio-accumulation of PCB - 153 concentrations in spider crabs (*Maja brachydactyla*) increased with age and levels in the hepatopancreas were at least 10- to 50-fold higher than those in their gonads and muscles (Bodin et al., 2007a). Also, the accumulation of PCBs in several species of edible crabs (*M. brachydactyla*, *Cancer pagurus*, *Necora puber*) showed that the tissue-specific concentrations of the ICES-7 congeners were an order of magnitude higher in the Bay of Seine compared to those on the coast of Brittany (Bodin et al., 2007b).

PCB-118, one of the more toxic congeners, has occurred above the Ecological Assessment Criteria (EAC) for sediment off the coast of the SWRBD (OSPAR, 2012b); nevertheless, in 2009, the concentrations of 25 summed PCBs (including the ICES congeners) were below detection at 23 of the 24 stations sampled in the central and western Channel (CEFAS, 2012). Contamination by PCBs remains of serious concern because of the link between the body burden of the blubber of porpoises stranded on the coasts of the UK and their susceptibility to infectious diseases and death (OSPAR, 2010a).

2.1.1.2 Polychlorinated-p-dibenzo-dioxins and polychlorinated-p-dibenzo-furans

Polychlorinated-p-dibenzo-dioxin (PCDD) and polychlorinated-p-dibenzo-furan (PCDF) compounds are formed from a variety of natural and anthropogenic processes. They are persistent in the environment, bioaccumulate and are extremely toxic to organisms; for this reason they are OSPAR priority pollutants and their releases are subject to the UNEP Stockholm Convention on Persistent Organic Pollutants and EU legislation (OSPAR, 2009e). There are no data on riverine and atmospheric loads to the Channel, although within the OSPAR area estimates suggest emissions to the atmosphere decreased by 50 % between 1998 and 2007 (OSPAR, 2009c), implying reductions in loads to the marine environment. The concentrations of seven PCDD congeners, ten PCDF congeners and four coplanar PCBs were determined in the mussel *Mytilus edulis* and the sea star *Asterias rubens* (Danis et al., 2006). Values were relatively high in the sea stars, leading to the induction of the biomarker cytochrome P450 immuno-positive protein CYP1A IPP. In the Bay of Seine, concentrations of 17 PCDD/F congeners in edible crabs, marine mussels and sea bass were in the range 12 to 123 pg g^{-1} (dry wt) (Bodin et al., 2007c). Temporal trends (1981-2005) in the concentrations of 17 congeners of 2,3,7,8 - PCDD/F in tissues of *M. edulis* were reported by Munschy et al. (2008). The data showed that the most recent concentrations were generally lower than earlier in the time series, notably in mussels collected from the mouth of the Seine at Villerville (Fig. 6b). Nevertheless, it is noteworthy that the concentrations reported in Fig. 6b are two orders of magnitude above those observed at remote coastal locations in Europe, such as the coasts of Corsica where values are about 1.9 pg g^{-1} (dry wt). Nevertheless, the ecotoxicological effects of the higher concentrations of these compounds in these biota is not completely understood (OSPAR, 2009e).

2.1.2 Polybrominated compounds

2.1.2.1 Polybrominated diphenyl ethers

Polybrominated diphenyl ethers (PBDEs) are a class of 75 brominated compounds widely used as flame retardants in Europe. Two priority sub-classes, pentaBDE and octaBDE, were banned for use in 2004 because of their toxicity and persistence (OSPAR, 2009e,f), while decaBDE uses have been restricted since 2008 (Covaci et al., 2011). PBDE compounds are found throughout the marine environment but the main route of introduction (e.g. atmospheric, river) is unknown (OSPAR, 2009e). Aeolian particle dry deposition fluxes of tetra-brominated BDE-47, penta-brominated BDE-99 and DPTE (the non-PBDE flame retardant 2,3-dibromopropyl-2,4,6-tribromophenyl ether) to the English Channel were estimated to be 30 – 100 pg m^{-2} d^{-1} in 2008 (Xie et al., 2011).

Total concentrations of 9 PBDE congeners plus DPTE in the dissolved phase and SPM of Channel waters were 0.81 pg L^{-1} and 0.77 pg L^{-1}, respectively (Xie et al., 2011). In sediments along the English coast, measurements from 2009-2010 showed that PBDEs were dominated by the fully brominated (deca-) BDE-209, with BDE-47 contributing < 8 % of the overall concentrations (CEFAS, 2012). While concentrations were generally low, there is concern that BDE-209 may debrominate *in situ* to yield more mobile and toxic products (Law et al., 2014). Dab (*Limanda limanda*) liver samples collected in 2010 along the English coastline had Σ6 PBDE concentrations 71 - > 200 times higher than a proposed WFD Environmental Quality Standard (CEFAS, 2012). Temporal trends (2003 – 2010) in dab liver concentrations at these sites varied from no trend to significantly downward (CEFAS, 2012). Thirteen PBDE congeners were determined in contemporary and freeze-dried archived mussel samples collected along the French coast. PBDE concentrations increased

significantly over a decade from 1981, effectively doubling every 5 years, and have now levelled off and may be in decline due to tighter EU regulation. Highest concentrations were obtained in samples from the Baie de Seine, with BDE-47, BDE-99 and (penta-) BDE-100 occurring up to 10 ng g^{-1} (dry wt) (Johansson et al., 2006). Notably, the highest concentrations coincided with the resuspension and down-estuary transport of historically contaminated sediments by extreme river flows and extensive dredging of sediments related to the port development at Le Havre (Johansson et al., 2006). Analyses of PBDE congeners in worms, bivalves, sole and eels (*Nereis diversicolor, Scrobicularia plana, Solea solea, Anguilla anguilla*) from the Seine Estuary also showed elevated levels of BDE-47 (Bragigand et al., 2006; Munschy et al., 2011), although the daily uptake of PBDEs via human consumption of seafood from this area was below levels likely to cause adverse effects (Bragigand et al., 2006). Within the Bay of the Seine, concentrations of 13 PBDE congeners were significantly elevated for sea bass, as compared to edible crabs and marine mussels, implying that sea bass were unable to metabolise these compounds. Concentrations of PBDEs in *S. solea* did not correlate with fish fecundity indices in the Seine Estuary, although the fish did exhibit an enhanced degradation capacity for these compounds (Munschy et al., 2011). Banned PBDEs have been replaced by so-called 'novel' flame retardants (Covaci et al., 2011) and they are attracting their own interest because of their apparent ability to bioaccumulate. Munschy et al. (2011) provide the only reported studies of these compounds in the English Channel, finding that concentrations of BTBPE (1,2-bis(2,4,6-tribromophenoxy)ethane) and DBDPE (decabromodiphenylethane) in livers of *S. solea* collected from the Seine Estuary were low and not influenced by local anthropogenic activity.

2.1.2.2 Hexabromocyclododecanes

Hexabromocyclododecanes (HBCDDs), used as flame retardants in polystyrene foam, are persistent, bioaccumulate and toxic, and are likely to be subject to legislation at some point (OSPAR, 2009f). They are the second most used flame retardants in Europe. The HBCDD family is made up of three stereoisomers, α-, β- and γ-HBCDD, of which α-HBCDD predominates in the tissues of marine mussels, accounting for typically 99 % of the total. Concentrations in the tissues of mussels sampled between 2008 and 2010 were in the range 0.06 to 0.41 ng g^{-1} (wet wt) (Munschy et al., 2013). Analyses of archived freeze-dried *M. edulis* samples from a site in the Seine Estuary showed a trend of increasing concentrations, with a doubling time of about 8 years, as shown in Fig. 6c (Munschy et al., 2013). This may reflect increasing use and / or release of these compounds to the environment.

2.1.3 Polyfluorinated compounds

Polyfluorinated compounds (PFCs) have been used in domestic and industrial products for 50 years. Seawater is the now the main environmental reservoir, where PFCs appear to be persistent, bioavailable and potentially toxic (Pistocchi and Loos, 2009). Of particular concern are the PFCs perfluorooctanoic acid (PFOA) and perfluorooctane sulphonate (PFOS). Estimates of PFOA loads from the Seine to coastal waters, at 0.2 t a^{-1}, are relatively modest compared to other large European rivers (Pistocchi and Loos, 2009). PFOS is more bioavailable than PFOA, and in *M. edulis* collected from the Seine Estuary in 2010 the concentration of PFOS was in the range 0.01 to 0.7 ng g^{-1} (wet wt) (Munschy et al., 2013). Freeze-dried samples of *M. edulis* tissue archived since 1981 showed an increase in concentrations in mussel tissues up to the mid-1990s followed by a linear decrease to relatively low concentrations by 2011 (Fig. 6d; Munschy et al., 2013). However, PFOS

concentrations remain high by European standards and information on the ecotoxicological implications of the PFOS loadings in marine mussels is sparse (OSPAR, 2009e). Since 2007 most uses of PFOS have been banned in the EU (Pistocchi and Loos, 2009) and from 2009 use has been restricted by the Stockholm Convention on Persistent Organic Pollutants (OSPAR, 2011b), and so environmental concentrations may be expected to decline further.

2.1.4 Pesticides

Gamma-hexachlorohexane (γ-HCH, or Lindane) is a priority pesticide whose use was banned by EU legislation in 2008 (OSPAR, 2009e). It is one of the six priority pesticides due for withdrawal from use by 2020, the others being endosulfan, methoxychlor, dicofol, pentachlorophenol and trifluralin (OSPAR, 2010a). While there are no measurements of γ-HCH undertaken in atmospheric dust and rainfall from the region (OSPAR, 2009c, 2013e), modelled net deposition shows a marked decrease from 1500 kg a^{-1} to 110 kg a^{-1} between 1990 and 2006 (Fig. 7). In contrast, trends in the RID data cannot be identified because of inconsistent reporting for the R. Seine (no data for 1990-1996, 2004-2006, 2010) and the large range in the low-high estimates, particularly for the French rivers (OSPAR, 2013a). It is notable that the recent estimates of the upper river loads are up to 5-fold higher than the atmospheric loads (Fig. 7). There are few recent measurements of γ-HCH in water, sediments and biota in the Channel. Most data are for sediments and biota along the French coast; here, concentrations were generally low in both matrices except for biota along the Brittany coast, where concentrations above the EAC occurred (Table 2; OSPAR, 2012b). Temporal trends in concentrations appear to be declining (Table 2), which may reflect time-dependent trends in loads to the area (with the caveat of the uncertainty in the river load estimates). Measurable concentrations of the legacy organochlorine pesticides hexachlorobenzene and pp´DDE (a metabolite of DDT) have recently been reported in edible crabs from the coasts of Brittany

and Normandy (Bodin et al., 2007b). While concentrations were highest in samples collected from the Baie de Seine, in general, concentrations were similar to those reported for other European waters.

2.2 Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are toxic, hydrophobic compounds that exist in samples as mixtures of complex organic compounds, containing 2 to 6 benzene rings. The main sources to the environment are from incomplete combustion of organic material, including from power generation, and from spills of crude oil. They also arise from a myriad of other industrial processes. OSPAR has included PAHs as voluntary determinands under the OSPAR atmospheric and river load monitoring programmes. However, very little load data appears to have been reported and none were available for the English Channel. Spills of crude oil that occur in the English Channel (OSPAR, 2010a) will contribute to water column concentrations of PAHs; however, there is a paucity of studies reporting such data. Addition of an Arabian light crude oil (similar to that spilled by the Amoco Cadiz in 1978) into Channel water showed a rapid dissolution of PAHs from the oil in to the seawater, increasing dissolved PAH concentrations by two orders of magnitude compared to the controls (Table 3). Exposure of the juvenile sea bass, *Dicentrarchus labrax*, a common fish in European transitional and coastal waters, to the oil contaminated seawater showed increased concentration of total PAH in the muscle tissue (Table 3) and longer term reductions in fish condition indices (Kerambrun et al., 2012a).

Along the industrialised harbours and estuaries of the both France and the UK, concentrations of PAHs in sediments, in particular, and biota are elevated above background values (Table 2; OSPAR, 2012b). For example, sediments located off the R. Tamar (SWRBD)

have on 46 % of occasions between 2005 – 2010 exceeded the Effects Range Low Sedimentary Quality Guideline for either low molecular mass PAHs (2-3 rings) or high molecular mass PAHs (5-6 rings) (CEFAS, 2012). In the Seine Estuary, Cailleaud et al. (2007a) determined the concentrations of various PAHs in solution, in SPM and in *E. affinis*. Phenanthrene, pyrene and benzo[b+k]fluoranthrene were the most predominant compounds in the water column and the maximum concentrations of PAHs in SPM and *E. affinis* occurred during winter. The highest mean dissolved concentration was for pyrene (2.8 ng L^{-1}) and for SPM and *E. affinis* the highest mean concentrations were 579 and 1124 ng g^{-1} (dry wt) for benzo[b+k]fluoranthrene, respectively. Consequently, using the summed PAH values for dissolved (11.1 ng L^{-1}), SPM (3361 ng g^{-1}) and *E. affinis* (879 ng g^{-1} dry wt) gives elevated K_ds of 3 x 10^5 L $kg^{-1}$and 5.2 x 10^5 L kg^{-1}, respectively. Experiments on the behavioural responses of *E. affinis* to sub-lethal concentrations of a 3-component mixture of PAHs (40 ng L^{-1}) in estuarine waters showed that swimming speed and activity were affected (Michalec et al., 2013). Clearly, PAHs are highly particle-reactive and significant uptake into the copepod suggests potential transmission through the marine food chain (Cailleaud et al., 2007a). The unknown factor is whether the uptake of PAHs by *E. affinis* was active or passive and is a topic for further investigation.

2.3 Alkylphenol-polyethoxylates

Alkylphenol-polyethoxlates are non-ionic surfactants having a wide range of industrial uses, both onshore and offshore. The most common compounds are nonylphenol-polyethoxylates (NPnEOs; where n=number of ethoxy units). They easily degrade, under both aerobic and anaerobic conditions, to yield short chain ethoxylates (NPE), nonylphenol (NP) and octylphenol (OP). NP, OP and short-chain NPE are lipophilic and hence bioaccumulate, exhibit endocrine disruption properties and are toxic to aquatic organisms

(Blackburn et al., 1999; OSPAR, 2009g). Indeed, experiments using NP at a concentration of 2 µg L^{-1} showed impacts on the swimming speed and activity of *E. affinis* (Michalec et al., 2013).

There are no reported load data for these compounds to the English Channel. Concentrations of dissolved NP and NP mono- + di-ethoxylate in estuaries along the UK Channel coast were less than 200 ng L^{-1} and 600 ng L^{-1}, respectively, and biological effects were thought unlikely (Blackburn and Waldock, 1995; Blackburn et al., 1999). There appears to have been little subsequent research in UK waters since this time. In the oligohaline region of the Seine Estuary the combined concentrations of NP and NPE compounds were in the range 399 to 2214 ng L^{-1} in solution and 405 to 9636 ng g^{-1} in SPM (Cailleaud et al., 2007b). In solution, nonylphenol-ethoxyacetic acid had the highest mean concentration (647 ng L^{-1}), while in SPM, nonylphenol had the highest mean value (1672 ng g^{-1} dry wt). Lowest total concentrations occurred during the spring and autumn blooms, perhaps due to biodegradation, while concentrations in the SPM were highest in winter. NP di-ethoxylate was the most bioaccumulated compound in *E. affinis*, with concentrations in the range 2890 to 6013 ng g^{-1} (dry wt), although NP was also significant. The K_d for *E. affinis* was ca. 5.3 x 10^3 L kg^{-1}; while this value is 2-3 orders of magnitude lower that the K_d*s* for PCBs and PAHs, this degree of partitioning may be sufficient to contribute to the endocrine disruption of fish observed in the Seine system (Cailleaud et al., 2007b).

2.4 Phthalates

The phthalates of most concern are diethylhexylphthalate (DEHP), dibutylphthalate (DBP) and butylbenzylphthalate (BBP) (OSPAR, 2009e). They are used as plasticisers in a wide range of products, from which they can be easily leached. There are no input data for

the Channel, but the emission of DEHP from adjacent river basins is estimated to be > 5 kg km^{-2} a^{-1} for the Seine and Scheldt, 0.5 – 5 kg km^{-2} a^{-1} for the SERBD and < 0.5 kg km^{-2} a^{-1} for the Loire Channel coastline (ETC/ICM, 2012). In Europe, > 5 kg km^{-2} a^{-1} is the highest emission category, which implies the potential for greatest impact is in the eastern English Channel; nevertheless, there are few reported studies of phthalates in estuarine and coastal waters of this region. Dissolved concentrations of six phthalates in the Seine Estuary, including DEHP, BBP, dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl-phthalate (DnBP), di (2-ethylhexyl) phthalate (DEHP) and di-n-octyl-phthalate (DnOP) were reported by Dargnat et al. (2009). Concentrations were in the range 160 to 314 ng L^{-1} for DEHP, 71 to 181 ng L^{-1} for DEP, and 67 to 319 ng L^{-1} for DnBP, whereas BBP and DnOP had very low concentrations or were not detected. The ecotoxicological implications of these concentrations has not been reported. The distribution of the phthalate concentrations appeared to correlate with industrial discharges from wastewater treatment plants. Experiments have been conducted to investigate the transfer mechanisms involving phthalates in the Seine river-estuary (Teil et al., 2013). Locations where elevated concentrations of DEHP occurred were identified in both rural and urban locations and plumes of DEHP were tracked to allow estimates of its dispersion and dilution from the source input under different hydrological conditions.

3. Metal contaminants

3.1 Cadmium, lead and mercury

The emphasis in this section is on the priority contaminant metals Cd, Pb and Hg, where the concentrations for total river loads are based on dissolved and particulate metals and for total atmospheric loads on wet and dry deposition (OSPAR, 2009c, 2013a). Relatively large river loads were obtained between 1990 and 1995, which diminished in the mid-2000s but have increased over the past few years (Fig. 8). These observations apply with more certainty to Pb rather than Cd and Hg because of the variability in their loads. Uncertainties in the estimation of metal loads is exemplified by reported inputs from the Seine for 1995; for Cd, the load calculated by Chiffoleau et al. (1999) is an order of magnitude larger than the RID load estimate (OSPAR, 2006b) even though the former is for the dissolved metal only, while for Pb the estimates are more consistent, with the total load a factor of 2-3 larger than the dissolved load (Fig. 8). Input of metals from the atmosphere to the surface of the English Channel have been obtained by numerical modelling and have decreased since the early- to mid-1990s, largely due to industrial control measures; indeed, Channel waters may now be a net source of Hg to the atmosphere. Comparison of the total river and atmospheric loads indicates that the inputs are now comparable for Pb, but for Cd and Hg such conclusions cannot be drawn because of the wide range in river load estimates. Unquantified, but possibly significant, groundwater inputs to the coastal sea in the Pays de Caux region, to the north of the mouth of the Seine Estuary, compound the uncertainty for Hg (Fig. 8; Laurier et al., 2007). Integrated studies between French and UK scientists in the 1990s (FLUXMANCHE, FLUXMANCHE II) quantified the transport of metals in the eastern Channel and their export to the southern North Sea (Fig. 8). For Cd the total loads were dominated by the dissolved phase in both regions of the Channel (ca. 86 %), while

dissolved Pb was more important in the central Channel (64 %) than in the Dover Straits (14 %). The large difference in the estimated particulate Pb loads for the central Channel and Dover Straits waters (77 and 1000 t a^{-1}, respectively; Statham et al., 1999) implied a source of particulate Pb in the eastern Channel significantly augmenting Pb export to the North Sea.

Along Channel coastlines, estuarine and near shore sediments contain the highest concentrations of metals, either because of naturally mineralised catchments (SW England), dumping of dredged spoil and domestic wastes or discharge from industry. Thus, locally high concentrations occur at sites on both sides of the Channel (e.g. Turner, 2000; Varma et al., 2011; Kadlecová et al., 2012). On a larger scale, the Seine Estuary and the Baie de Seine are two of the most metal contaminated sea areas in Europe because of intensive industrial activity in the hinterland, coupled to a large population in the RBD (Fig. 1). Both Cd and Pb have been reported at markedly higher concentrations in both sediments and biota, relative to other European estuaries (Dauvin, 2008). In the case of Hg, the French Mussel Watch programme has found that the highest concentrations were in shellfish from the eastern part of the Baie de Seine (Laurier et al., 2007). In contrast, sediments, limpets, fish and crabs collected from the industrialised coastline of the nearby Cherbourg peninsula showed that there was little evidence of significant contamination for Hg, Cd and Pb and that the concentrations were in the same ranges as those observed in similarly industrialised locations (Connan and Tack, 2010).

With respect to the ecotoxicological impacts of metals on marine biota, it is the sedimentary pool which appears to be the most important. Broadly, bioaccumulation within benthic organisms follows some measure of bioavailability of the metal in the sediment, however this is assessed. For example, Amiard et al. (2007) examined patterns of labile and total metal concentrations in surficial sediments and metal bioaccumulation in marine

organisms, including the deposit feeding *N. diversicolor* collected from the mouth of the Seine, from Boulogne harbour and from Restronguet Creek. Cadmium was the most labile metal of those studied, and concentrations of Cd in *N. diversicolor* increased in proportion with the fraction of sediment Cd that was extractable at pH 6. The toxicity effects of reported metal body burdens have been observed at the level of the cell and individual, but deleterious changes to benthic organisms at the population and community level have been more difficult to quantify (Dauvin, 2008). Recently, the benthic dwelling European flounder *Platichthys flesus* has been chosen as a test organism for biological effects monitoring in transitional and near shore waters (Henry et al., 2012). *P. flesus* typically spawns during winter at the mouths of estuaries and, after the larval stage, the juveniles spend approximately two years living in estuaries. The juvenile fish, which tend to be more sensitive to metal contamination than the adults, have been used to assess habitat quality of the Authie, Canche, Seine and Somme estuaries (Henry et al., 2012; Kerambrun et al., 2013) and their coastal waters (Henry et al., 2004). The concentrations of metals in the sediments and their bioaccumulation in the livers and gills of juvenile flounders have also been compared between the contaminated Seine and the more pristine Canche estuaries. The results for Cd, Hg and Pb show that the flounder from the Seine displayed the highest concentrations for each metal and also in both the livers and gills (Fig. 9). Significant statistical relationships were obtained for metal concentrations in the sediment and (a) the fish liver for Cd, Hg and Pb and (b) the gills for Pb. The fish condition indices for the samples from the Seine were poorer than those from the Canche, implying negative effects on juvenile survival and adult recruitment (Kerambrun et al., 2013). Assessment of habitat quality using juvenile fish, while promising, is acknowledged to be at an early stage of development.

Temporal trends in Cd, Hg and Pb concentrations sediments and biota along the French and UK coasts offer a mixed picture, as shown in Table 2, while data from the French Mussel watch programme, summarised in Fig. 10, indicates that there has been some diminution in concentrations of Hg in mussels between the early-1990s and the mid-2000s, particularly in the Pays de Caux region to the north of the Baie de Seine (Laurier et al., 2007).

3.2 Emerging metal contaminants

3.2.1 Thallium

Thallium, in contrast to the other Group 1 elements Na and K, is highly toxic, and is classed as a dangerous substance by the EU and as a Priority Pollutant by the USEPA. The principle sources of Tl to the environment are metal mining and smelting, cement production and coal combustion. There are few data on Tl in the marine environment. Concentrations of Tl in river, estuarine and waste waters of SW England were generally in a range from < 20 ng L^{-1} up to 450 ng L^{-1} for rivers in mineralised catchments subject to historic mining of metals. The highest value of 1400 ng L^{-1} was obtained for waters collected directly from an abandoned mine (Law and Turner, 2011). Tl can penetrate cell membranes of marine organisms, as Tl^{+}, through NaCl-KCl co-transporter sites and, possibly via transport through K^{+} ion channels. In estuarine and coastal seawaters spiked with dissolved Tl^{+}, the high biomass macro-alga *Ulva lactuca* (section 7) bioaccumulated Tl linearly with increasing dissolved concentrations (Turner and Furniss, 2012). Above a dissolved concentration of 10 µg L^{-1}, there were marked decreases in the efficiency (up to 70 %) of its photosystem II (Turner and Furniss, 2012). Across a range of estuaries of the SWRBD in the partitioning of Tl in to benthic organisms and algae was in the range $10^{2} – 10^{4}$ L kg^{-1}; while this is low relative to some other metals, the limited understanding of its biogeochemical behaviour and

potential toxicity would suggest it prudent to extend the study of this metal in the marine environment (Turner et al., 2013).

3.2.2 Platinum group elements

There is a potential global remobilisation of the platinum group elements (PGE) Pt, Pd and Rh, principally through their use in vehicle catalytic converters; these account for 50 – 60 % of the worldwide use of Pt and Pd, and ca. 100 % for Rh. From this diffuse source, and also from mining, these metals will enter estuaries and coastal seas either in particulate form or in solution (Ravindra et al., 2004). The few studies undertaken on the three metals showed that their estuarine reactivity was contrasting. Experiments using Plym Estuary (SWRBD) water and sediments showed that Pd had the greatest mobility from catalytic convertor particles, and had the greatest sediment lability and lowest particle reactivity (Turner et al., 2006). Thus this metal, of the group, showed the greatest potential for long range transport and bioaccumulation. In solution Pd was mainly associated with small (< 0.1 μm) hydrophobic colloids (aiding bioaccumulation?) which salt out at high salinities. Rhodium also exhibited this behaviour but to a lesser extent, and was also more particle reactive with K_ds in the range 10^3 - 10^4 (Cobelo-Garcia et al., 2008). Rhodium also showed the greatest sorption to, and uptake in to, the marine unicellular alga *Chlorella stigmatophora* (Shams et al., 2014). Kinetic constraints on the partitioning of Pt suggested that sorption of the dissolved metal to particles would be favoured in longer residence time coastal waters (Cobelo-Garcia et al., 2008). Studies using water, sediment and the polychaete *Arenicola marina* from the Erme Estuary (SWRBD) showed these benthic organisms potentially have an important role in the cycling and fate of both dissolved and catalytic convertor particle PGE in estuarine environments (French and Turner, 2008). There have been few toxicity studies of PGE undertaken on marine organisms. The cyto-toxic drug cispaltin, which

contains Pt, can enter estuaries and coastal waters in effluents from sewage treatment and hospitals. The impact of cisplatin to the photo-efficiency of energy conversion by *U. lactuca* was examined and found to have no effects at the concentrations (150 nM) used (Easton et al., 2011).

3.2.3 Antifouling paint particles

Antifouling paints are applied to boats and semi-submerged permanent structures, such as drilling rigs, pipelines, buoys, navigation lights, pontoons and piers. Those containing TBT are only used for vessels over 14 m in length; nevertheless, TBT is the main cause of poor chemical status in the transitional waters of the SWRBD (ETC/ICM, 2012). In non-TBT paints, the principal bioactive metals are Cu and Zn, together or separately, and to a lesser extent Cd, Cr, Ni and Pb (Turner, 2010). The general composition of an anti-fouling paint is shown in Figure 11. Although the metal component is only a small fraction of the total, the concentrations are very high. The many marinas and boat/ship repair yards along the English Channel coast are sites where metal contaminants may enter local waters as a result of the removal of antifouling paints from the hulls of vessels. Fine dusts, paint flakes and larger particles, produced by various forms of abrasion, are then washed into the near shore and largely deposited into the sediments (Turner, 2010; Takahashi et al., 2012). In Plym Estuary (SWRBD) sediments collected close to a boat repair yard, almost 1 % of the < 63 µm fraction, by mass, was paint chips (Turner, 2010). Once in the sediments benthic dwellers are exposed to the contaminants, and Turner (2010) describes the bioaccumulation of Cu and Zn in *M. edulis* and *A. marina* from deposited paint particles. A population of wild mute swans, *Cygnus olor*, are resident in the Tamar Estuary, where they feed on eelgrass and hull-bound algae on beached or poorly-maintained boats. Turner and Hambling (2012) simulated metal uptake by the swans, using a proxy test, from eelgrass and sediments spiked with antifouling

paint particles. Results showed that both Cu and Zn had elevated bio-accessibilities, up to 10%, in the gizzard, and that gut absorption was much higher for Cu than Zn, implying that Cu poses a threat to mute swans at sites contaminated with antifouling paint particles.

4. Mixtures of xenobiotic substances

Assessments of bioaccumulation and toxicity are usually undertaken on a single compound or single class of compounds, although in reality contaminants generally occur as mixtures (Hutchinson et al., 2013). A plethora of toxic substances, including PCBs, PAHs and metals, are known to be sorbed to estuarine SPM and sediments. In the Seine Estuary, Vincent-Hubert et al. (2012) showed that SPM was mutagenic, with SPM collected in the vicinity of a discharge from an industry manufacturing chemical dyes having the highest mutagenicity, followed by SPM influenced by discharges from petrochemical industries, a petroleum refinery and pulp/paper mills. In - situ experiments were conducted using juvenile *D. labrax* and turbot (*Scophthalmus maximus*) caged for 38 days at three sites in Boulogne harbour (Kerambrun et al., 2012b). At the site with the most contaminated sediments (e.g. Cd = 1.56 µg g^{-1}; Hg = 0.35 µg g^{-1}; ΣPAH = 4.12 µg g^{-1}; ΣPCB = 0.01 µg g^{-1}, all dry wt) all the fish died within 14 days, whereas at the least contaminated site (e.g. Cd < 0.01 µg g^{-1}; Hg = 0.01 µg g^{-1}; ΣPAH < 0.01 µg g^{-1}; ΣPCB = 0.01µg g^{-1}, all dry wt) most of the fish recovered when transplanted to a clean site. However, the surviving fish experienced significant decreases in growth and condition indices which appeared to be a result of metal accumulation in the gills. These studies were complemented by laboratory experiments on turbot using sediments from the same sites in Boulogne harbour and sediments from the mouth of the Seine (Kerambrun et al., 2012c), with similar results regarding physiological biomarkers, fish conditions and potential impacts on recruitment to the adult stages. In terms of biological effects-directed analysis (Hutchinson et al., 2013), an important operational outcome from these studies was the conclusion that physiological biomarkers of juvenile fish (*D. labrax*) could be used to assess stress caused by exposure to complex mixtures of contaminants. Several papers in the current SI demonstrate, further, the value of this

philosophy. Comparisons of the genetic make-up of persistent populations of *P. flesus* living in Seine Estuary waters containing complex mixtures of xenobiotic compounds, with those from less contaminated locations, have identified specific genes that enable *P. flesus* to better manage chemical stress. The consequences of improved resistance to chemical stress in heavily contaminated Seine Estuary waters were reduced condition factors and fecundity for *P. flesus* (Marchand et al., 2013). Other workers have investigated the use of molecular data, in this case proteomic markers (i.e. proteins whose abundance changes in response to external pressures and hence reveal potentially useful data on the function of an organism under adverse environmental conditions) to investigate stresses on biota in Channel waters. Galland et al. (2013) measured the proteomic composition of the livers of wild populations of *P. flesus* from the Seine, Canche and Tamar estuaries. The livers of samples from the Seine and Tamar estuaries, classed by the authors as highly polluted water bodies, were the most similar in relation to protein changes observed. In the Seine samples, protein changes were linked to oogenesis deregulation, implying the potential for effects at the population level. The use of proteomics possibly heralds the development of a new molecular biomarker; nevertheless, the development of robust correlations between exposures to contaminants and marker protein changes in wild populations of fish remains a challenge (Galland et al., 2013).

5. Radionuclides

5.1 Sources of anthropogenic and natural radionuclides

The inputs of radionuclides originate from four main sources: (i) Approved discharges of radionuclides from the French nuclear reprocessing plant at La Hague on the Cherbourg Peninsula (Fig. 1). This plant has been in operation since 1967 and produces artificial radionuclides, including ^{60}Co, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu and ^{241}Am. The maximum discharges of ^{137}Cs occurred in 1971, of ^{238}Pu, $^{239+240}$Pu and ^{241}Am in about 1974 and of ^{60}Co from 1983 to 1985 (Cundy et al., 2002). (ii) Approved discharges of radionuclides from French nuclear power stations at Flamanville, on the Cherbourg Peninsula, at Paluel and Penly east of Le Havre and at Nogent-sur-Seine upstream of Paris (Fig. 1). (iii) The dumping, between 1950 and 1963, of some 28,500 containers containing low-level radioactive waste, equivalent to ca. 60 TBq, into the Hurd Deep by the UK and Belgium (Hughes et al., 2011; Spiegel Online, 2013). The UK authorities have carried out a radiological surveillance programme in the waters of the Hurd Deep over several decades and evidence for releases from the containers has not been reported (CEFAS, 2013). (iv) Radionuclides, such as ^{3}H (tritium), ^{7}Be and ^{14}C, are produced cosmogenically and are delivered to the earth surface in wet and dry fallout. Also, ^{238}U, and its decay products, originate from weathering granitic deposits in river catchments. There is also a significant anthropogenic production of both ^{3}H and^{14}C.

5.2 Tritium discharges and distributions

Tritium is a natural radionuclide with a half-life of 12.3 years and emits β-particles with a mean energy of 5.7 keV. However, about 90 % of the liquid and gaseous forms of ^{3}H activity in the environment originate from nuclear re-processing plants, nuclear power stations, defence establishments and nuclear research facilities (Table 4). The total discharges

of ^{3}H, mainly as tritiated water, from La Hague and Sellafield have increased over the past two decades and there is little evidence of any systematic decrease, unlike the marked decreases in the discharge of α -emitters (Fig. 12a; OSPAR, 2013g). Consequent upon the quantities of ^{3}H being discharged, effort has been devoted to the modelling of its transport and fate within the English Channel, as shown in Fig. 12b (Bailly du Bois et al., 2005).

In 2012, maximum dissolved ^{3}H activity concentrations in freshwater streams in the vicinity of the La Hague facility were 529 ± 37 Bq L^{-1} and near the nuclear power station at Nogent-sur-Seine the mean value was 32.7 ± 3.8 Bq L^{-1} (IRSN, 2012). However, in seawater around La Hague the dissolved ^{3}H activity concentrations were typically < 15 Bq L^{-1}, as compared with background values of 0.1-0.2 Bq L^{-1} (Fiévet et al., 2013). In the western English Channel and around the Channel Islands, dissolved ^{3}H activity concentrations were typically < 2 Bq L^{-1} (CEFAS, 2013).The mean activity concentrations of ^{3}H in marine algae near Cherbourg were 1.3 ± 0.2 Bq kg^{-1} (dry wt) (IRSN, 2012) and were below detection in seaweeds and edible crabs in the Channel islands (CEFAS, 2013). The biogeochemical behaviour of ^{3}H in the aquatic environment is poorly understood, notably in respect of the extent to which natural organic matter controls its geochemistry and bioavailability (Turner et al., 2009). This may be important because evidence is emerging that its low energy β -emissions can cause damage to the DNA of *M. edulis* (Jha et al., 2005; Jaeschke et al., 2011). Thus, there remains growing international concern about future trends in the discharge of anthropogenic ^{3}H and its potential biological impact (Dallas et al., 2012).

5.3 Distribution of artificial radionuclides

Interest in the discharge, distribution and fate of artificial radionuclides (for example ^{99}Tc, ^{125}Sb, ^{137}Cs and the transuranics^{238}Pu, $^{239+240}Pu$, ^{241}Am) in the English Channel has

increased since the mid-1990s, mainly because of the potential for their trans-boundary movement. Long range aquatic transport has been identified using measurements showing the migration of ^{125}Sb from the French nuclear reprocessing plant at La Hague into the North Sea (Guéguéniat et al., 1995; 1997). In 1993, dissolved ^{99}Tc activity concentrations were in the range 0.19 to 0.93 mBq L^{-1}, with the highest values being to the east of La Hague (McCubbin et al., 2002). The distribution of particle-reactive artificial radionuclides, such as ^{238}Pu, $^{239+240}$Pu, ^{60}Co and ^{137}Cs, has been studied in the sediments of the English Channel where the maximum concentrations of ^{238}Pu, $^{239+240}$Pu and ^{60}Co were found immobilised on seabed sediments in the central Channel (Boust, 1999). Evidence of the eastward transport of particle-reactive radionuclides along the French coast has been found from sediment cores taken on salt marshes near Flamanville and in the Baie de Somme where the radionuclide signatures are specific to La Hague discharges. The activity concentrations were relatively low and, because of sediment mixing, evaluation of the discharge history could not be made (Cundy et al., 2002). Similarly, activity concentrations of ^{60}Co and ^{137}Cs, in the < 50 µm grain size fraction of beach sediments from the Baie de Seine, decreased eastwards (Dubrulle et al., 2007).

Although ^{14}C is regarded as a natural radionuclide, the radioactive releases of ^{14}C by the La Hague reprocessing plant are included here because the annual liquid and gaseous discharges for 2012 were significant at 7 TBq and 16 TBq, respectively (IRSN, 2012). Studies of the uptake of ^{14}C into marine biota, including the macro-alga *Fucus serratus*, *M. edulis*, the Pacific oyster *Crassostrea gigas* and the European lobster *Homarus gammarus*, showed that activity concentrations were confined to a narrow range of 10 - 45 Bq kg^{-1} (wet wt). Similarly, concentration factors were also in a narrow range of 1 - 5 x 10^3, if the

measurements of dissolved inorganic carbon (6.9 - 14 mBq L^{-1}) were used in their estimation (Fiévet et al., 2006).

Activity concentrations of ^{137}Cs in the English Channel are due to discharges from the installations identified in Table 4. The most important is La Hague, releasing 0.75 TBq in 2011 (IRSN, 2011). For 2012, the mean activity concentrations of dissolved ^{137}Cs in the English Channel, and the Channel Islands were from < 2 to 3 mBq L^{-1}(CEFAS, 2013) and the effective half-life in the Channel (taking into account physical and chemical processes as well as nuclear decay) was 1.2 ± 0.5 years. Dissolved^{137}Cs from La Hague was largely transported in the north-east direction, together with a contribution of about 1% of ^{137}Cs from Sellafield, on the north west coast of the UK, entering the western Channel via the Celtic Sea (Povinec et al., 2003). However, the hydrography around the Cherbourg Peninsula indicates that seawater is also swept in a south west direction mainly towards the Channel Islands of Jersey and, to a lesser extent, Guernsey and Alderney (Cundy et al., 2002). Long-term monitoring of ^{137}Cs in sediments in the Channel Islands has shown that the activity concentrations have decreased, particularly in muddy sediments from Jersey, over approximately two decades from a maximum of 8.3 Bq kg^{-1} in 1998 to 1.5 Bq kg^{-1} (dry wt) in 2012 (Fig. 13a). In comparison, the activity concentrations of ^{137}Cs in littoral sediments (no textural information was given) from Cherbourg to Le Havre were 0.36 to 0.97 Bq kg^{-1} (dry wt) (IRSN, 2012), substantially lower than the values in Fig. 13a. Activity concentrations of ^{137}Cs in *Fucus* spp. and edible crabs from the Channel Islands were mainly below detection limits typically < 0.06 Bq kg^{-1} (wet wt) (CEFAS, 2013). Algal samples from the French coast were in the range 0.24 to 0.40 Bq kg^{-1} (dry wt), with maximum values of 052 ± 0.12 Bq kg^{-1} (dry wt) near to La Hague and activity concentrations in crustaceans were < 0.19 Bq kg^{-1} (wet wt) (IRSN, 2011, 2012).

In 2012, 19.7 GBq of Pu was discharged from La Hague (IRSN, 2012). The activity concentrations of $^{239+240}$Pu in sediments from the west coast of the Cherbourg Peninsula were 0.364 ± 0.019 mBq kg^{-1} (dry wt) although no information was given concerning sediment texture (IRSN, 2012). In contrast, activity concentrations of $^{239+240}$Pu for sediment collection sites in Jersey (mud) and Guernsey (muddy sand) were 1.2 and 0.072 Bq kg^{-1} (dry wt), respectively, in 2012 (Fig. 13b). No measurements were reported for Alderney, presumably because only sand was present (CEFAS, 2013). Thus, the contemporary activity concentrations of $^{239+240}$Pu in sediments from the vicinity of La Hague were lower than the values in Jersey sediments. The activity concentrations of $^{239+240}$Pu in algae and crustaceans from the west coast of the Cherbourg Peninsula were 93 ± 5 mBq kg^{-1}(dry wt) and 13.3 ± 3.4 mBq kg^{-1} (wet wt), respectively (IRSN, 2012). These values were significantly higher than those for *Fucus* spp. and edible crabs collected from the Channel Islands, and reported in Figs. 13(c) and (d) respectively, presumably due to their proximity to the La Hague.

Direct comparison of activity concentrations in various sediment and biological samples from the UK and French coasts is made more complicated by an apparent lack of systematic sampling and analysis between the two countries, at least for data in the publications in the public domain. For example, comparison of activity concentrations of ^{137}Cs and $^{239+240}$Pu reported above suggest that lower values occur in French sediments taken in the vicinity of La Hague compared to those in Jersey. Potentially this difference could have arisen because sediments in the vicinity of La Hague are more coarse grained and therefore less likely to adsorb particle-reactive radionuclides. As activity concentrations of radionuclides in sediments are dependent, to a significant degree, on sediment texture, it is essential that this parameter is taken account of. Furthermore, the activity concentrations for samples of algae are reported as wet weight in the UK (CEFAS, 2013) and dry weight in

France (IRSN, 2012), thus making direct comparison difficult. These factors combine to illustrate the urgent need for a Channel-wide harmonisation in radionuclide methodologies and the reporting of data.

5.4 Naturally-occurring radionuclides

The naturally occurring radionuclides, ^{210}Pb and ^{210}Po, which are ^{238}U decay products, are of particular importance because both are α-emitters with energies in excess of 5 MeV. As a result of their particle reactivity, these radionuclides are taken up by suspension or bottom feeding marine organisms and the α-radiation can cause internal genetic damage. Activity concentrations of ^{210}Po in *M. edulis*, and *C. gigas*, were similar and in the range 90 to 600 Bq kg^{-1} (dry wt), whereas activity concentrations in *F. serratus* were lower and between 4 and 16 Bq kg^{-1} (dry wt). Tissue-specific analyses in *C. gigas* showed that the digestive glands, gills and gonads had the highest activity concentrations. Estimates of the contribution to the internal dose from ^{210}Po received by humans consuming 5 kg a^{-1} each of mussels and oysters was in the range 50 to 130 μSv a^{-1} (Connan et al., 2007). Doses of this magnitude may make a minor contribution to the recommended maximum "additional" dose to the general public, 1 mSv a^{-1}, which is in addition to the natural radioactivity received by humans across the globe of 2.4 mSv a^{-1} (UNSCEAR, 2000).

As a consequence of their particle-reactivity, the solid phase speciation of ^{210}Po and ^{210}Pb in sediments is important with respect to their bioavailability and potential diagenetic behaviour, particularly for ^{210}Po which is bound to organic matter. Sediment samples from the vicinity of Cherbourg had pore water concentrations of ^{210}Po and ^{210}Pb between 1 and 20 mBq L^{-1} and 2.4 and 3.8 mBq L^{-1}, respectively. These activity concentrations were higher

than those in seawater implying that the sediments act as a source of both radionuclides to the overlying seawater (Connan et al., 2009).

In conclusion, there is a paucity of data on the mutagenic effects of chronic exposure to ionising radiation on the genetic integrity of marine organisms. Future research could involve studies on the biological responses of marine organisms to low doses in order to establish improved criteria for the protection of the marine environment.

6. Micro-plastic contaminants

Plastics production in Europe has increased nearly 200 – fold since the 1950s to 280 million tonnes in 2011 (Wright et al., 2013). As a consequence, plastic particles are being found in ever increasing amounts in seawater (Thompson et al., 2004). Micro-plastics, typically < 1 mm in diameter, are either manufactured directly or are derived from large plastic particles which degrade into smaller fragments as a consequence of photolytic and mechanical breakdown, and biodegradation. Nevertheless, it is sewage effluent that appears to contribute the most to micro- plastic loads found on shores and strandlines, as concluded from the similarities in plastics composition between the two end – members (67 – 78 % polyester, 17 – 22 % acrylic). The use of washing machines appears to promote micro-plastic fibre generation (Browne et al., 2011). Micro - plastics from past (now banned) dumping of sewage at sea have also been found at dump sites, including one in the western English Channel; these can have > 250 % more micro-plastics buried in the sediments compared to the sediments of reference sites (Browne et al., 2011).

Within the inter-tidal sediments of the Tamar Estuary 65 % of all plastic particles found were < 1 mm in diameter and the polymer types were polyolefins, expanded polystyrene, polyethylene and polypropylene (Browne et al., 2010). Analyses of floating plastic debris collected at the mouth of the Tamar Estuary, using a net with a 300 µm mesh, showed that the plastics comprised 40 % polyethylene, 25 % polystyrene and 19 % polypropylene. The particles were typically < 5 mm diameter and comprised 82 % of the total captured. Detailed variations in the size ranges observed were dependent on tidal state, thus indicating the need for clearly defined sampling strategies (Sadri and Thompson, 2014).

The implications arising from the presence of micro-plastics in marine sediments and waters is the potential for their ingestion by wild and farmed marine animals, such as mussels, barnacles, amphipods and lugworms. Lusher et al. (2013) reported that of the 504 pelagic and demersal fish sampled from the English Channel, 35.6 % of the gastrointestinal tracts examined contained micro – plastics, at an average count of 1.9 pieces per fish. Of the 351 items analysed for composition, rayon (57.8%) and polyamides (35.6%) were most common and there were no differences between the two fish types. More than 60 % of beach – washed northern fulmars (*Fulmarus glacialis*) in the Channel had more than 0.1 g of plastic in their stomachs, significantly higher than the Ecological Quality Objective of 10 % (OSPAR, 2010a). As a cause of concern is the first reported 'natural' transfer of micro - plastics between trophic levels in laboratory experiments involving *M. edulis* and the crab *Carcinus maenas* (Farrell and Nelson, 2013).

An additional complication with micro-plastics in estuaries and coastal waters of the English Channel is their propensity to scavenge persistent organic pollutants from the dissolved phase. Bakir et al. (2012, 2014) have investigated the particle-water interactions of ^{14}C-labelled phenanthrene (Phe) and DDT using micro-plastic particles of polyvinyl chloride (PVC) and polyethylene (PE) at five different salinities. The values of K_d for each plastic-contaminant combination PVC-Phe, PE-DDT, PVC-Phe and PE-DDT were largely independent of salinity and were in the ranges $2 – 2.5 \times 10^3$, $2 - 7.9 \times 10^5$, $5 – 12.5 \times 10^4$ and $2 - 7.9 \times 10^5$ L kg^{-1}, respectively. These studies illustrate that there is not only the potential for physical harm to marine organisms from the uptake of plastics but also the chemical impact from adsorbed contaminants; these concerns are the subject of continuing research.

7. Nutrients and algae

In the context of the MSFD, human-induced eutrophication is one of eleven quality descriptors for evaluating ecosystem function. It is defined as a 'process driven by enrichment of water by nutrients, especially compounds of nitrogen and/or phosphorus, leading to: (a) increased growth, primary production and biomass of algae, (b) changes in the balance of organisms, and (c) water quality degradation. The consequences of eutrophication are undesirable if they appreciably degrade ecosystem health and/or the sustainable provision of goods and services' (Ferreira et al., 2011). Excess phytoplankton growth leading to undesirable disturbance has been termed harmful algal bloom, or HAB, and Ferreira et al. (2011) have described three types: (a) toxic algae harmful to shellfish, even at low abundance, (b) potentially toxic algae, and (c) high biomass blooms, including both suspended phytoplankton and bed - anchored macro- algae. The source of the nutrients is primarily land-based and hence excess algal growth is generally observed in transitional and near shore coastal waters that are not light limited.

The mean riverine concentrations of nitrate-N and phosphate-P for the RBDs draining into the Channel are given in Table 5. The Seine, Loire, Scheldt and South East RBDs have high concentrations of nitrate-N (3.6 – 5.6 mg N L^{-1}), reflecting the high applications of nitrogen fertiliser and intensive animal husbandry in these regions (ETC/ICM, 2012; Dupas et al., 2013). Phosphate-P is also high along the UK coast, especially the SERBD, and in the Scheldt basin. Riverine loads of total N, nitrate-N, total P and phosphate-P into the Channel for 1990 – 2011, shown in Fig. 14, reveal that there has been a marked decline in P loads over this period, consistent with EU mitigation controls on point sources, while N loads have remained relatively unchanged, reflecting the difficulties in controlling diffuse sources, coupled to N contamination of long residence time ground waters (Billen et al., 2009;

Howden and Burt, 2009). It is noteworthy from Fig. 14 that atmospheric inputs of N were also largely unchanged (data for 1995 – 2005 only), and at ca. 60 kt a^{-1}, or a third of the riverine load, were a significant input. It is likely that both riverine and atmospheric nutrient inputs have contributed to the 'problem' eutrophication status of some Channel waters, as shown in Fig. 15 (OSPAR, 2010a).

Along the eastern Channel coast of France, nutrient loads, particularly nitrogen, from the rivers Seine, Somme, Authie and Canche have contributed to the enhanced blooms of diatoms and flagellates since the early 1980s, with concentrations of chlorophyll a reported to be 30 – 40 µg L^{-1}, and sometimes 60 - 70 µg L^{-1} (Cugier et al., 2005a; Boulart et al., 2006; Lefebvre et al., 2011). Toxic and potentially toxic suspended algae, including the diatoms *Pseudo-nitzschia* and the dinoflagellates *Dinophysis*, *Prorocentrum* and *Gymnodinium-Gyrodinium* have been reported in these waters, as has the high biomass alga *Phaeocystis globosa* (Lefebvre et al., 2011; Hernández-Fariñas et al., 2013). Deleterious impacts on the functioning of shellfisheries and intertidal benthic nutrient cycling and community production from toxic algae and *Phaeocystis*, respectively, have been reported (Spilmont et al., 2009; Dauvin, 2012). In UK estuarine and coastal waters, toxic (*Alexandrium*, *Gyrodinium*) and high biomass (*Chaetoceros*, *Emiliania huxleyi*, *Noctiluca*, *Phaeocystis*) suspended plankton have also been observed (Environment Agency, 2000). Blooms have been reported in the western Channel, including toxic (*Pseudo-nitzschia*, *Dinophysis*, *Prorocentrum*) and high biomass (*Chaetoceros*, *Noctiluca*, *Karenia*) algae (Johns, 2006); these blooms were not obviously linked to land run-off or aeolian inputs and were probably due to favorable hydrographic and climatic conditions (Johns, 2006; Vanhoutte-Brunier et al., 2008). High biomass macro-algal growth (*Ulva*, *Enteromorpha*) is a frequent occurrence along the French coast west of Brittany and along sections of the UK coast, and has been linked to high

nutrient inputs, including those from sewage (Ménesguen et al., 2006; Dauvin, 2012; Environment Agency, 2012).

The few extended time series of phytoplankton community composition for open Channel waters have revealed that potentially important compositional changes have and are occurring, as exemplified by temporal changes in the Phytoplankton Colour Index (PCI) shown in Fig. 16. These changes have been explained by large scale climatic factors such as the North Atlantic Oscillation (NAO) and its influence on sea surface temperature (Southward et al., 2005; Johns, 2006; Widdicombe et al., 2010; Smyth et al., 2010). Since 1992, i.e. after the beginning of increased riverine nutrient loads to coastal waters, the French REPHY and SRN phytoplankton and nutrient monitoring programmes have generated time – series data for the coastal strip for the Somme northwards to Dunkirk (Lefebvre et al., 2011; Hernández-Fariñas et al., 2013). Even in these nutrient replete waters, initial conclusions were that the NAO and/or the Atlantic Multidecadal Oscillation can influence phytoplankton community structure, not only via water temperature and mixing, but also by altering the hydrographic functioning of catchments (Hernández-Fariñas et al., 2013). This inter-dependency is clearly complex and poses a challenging management problem (Ferreira et al., 2011). The use of advanced coupled hydrodynamic – biogeochemical models may be one tool to aid water quality regulators. For example, changing patterns of potentially toxic dinoflagellate abundance in the Baie de Seine plume in relation to changes in river flows, land use patterns and nutrient discharge controls have been examined using (off line) coupled catchment and estuarine-coastal water models (Cugier et al., 2005a,b).

8. Summary and Conclusions

This review relies heavily upon the data accumulated by the OSPAR Commission in the pursuit of its goal to implement the statutory guidelines, framed under the OSPAR Convention, to protect the marine environment of the NE Atlantic. Without such long-term commitment to routine environmental monitoring by the 15 countries involved there would be no framework for the effective management of the seas around Europe, including the English Channel. However, effective monitoring has to be complemented by research into fundamental marine environmental processes and the review indicates that, over the past decade, the bulk of the effort in process-oriented research with respect to contaminants has been led, largely, by French scientists. Their studies are no doubt driven, to some extent, by the complex problems posed in managing a large, impacted estuarine system such as the Seine. The OSPAR and independent research datasets comprising the review are summarised in Appendix 1.

Presently, the estimated river loads are most reliable for Pb, nitrate and phosphate, but much less so for Cd, Hg, PCBs and γ-HCH, despite the best intentions of the OSPAR RID programme. Major uncertainties lie in the datasets for the R. Seine, which dominates fluvial input to the Channel and to the eastern Channel in particular. Actual measurements of atmospheric inputs of hazardous contaminants to Channel waters are few, and load data largely rely on modelling. The data appear robust for Cd, Pb, Hg, PCB(-153) and γ-HCH. For the other contaminants discussed in this article, there is simply no information on riverine and atmospheric inputs. Load data from other sources, for example submarine groundwater discharges or shipping, also appear to be lacking for all constituents. Temporal trends in reliable data show persistent reduction in loads, probably as a result of control measures on contaminants. However, river nitrate loads have remained stubbornly uniform with time,

reflecting difficulties in controlling land-based diffuse sources. Similarly, atmospheric loads of nitrate and ammonium have also remained uniform over time, despite European legislation designed to limit atmospheric emissions of nitrogen. Strategies to promote recovery of the estuarine and coastal waters of the English Channel, and thereby enhance its ecological status, are likely to be thwarted because of a lack of coherent spatial and temporal data on contaminants and an incomplete understanding of their marine environmental pathways.

The various biological impacts of contaminants on the ecological functioning of the Channel are difficult to discern. For the hazardous contaminants assessed herein widespread eco-toxicological impacts have not been observed, although OSPAR monitoring data indicate toxicologically important body burdens in some organisms. Moreover, recent research-based studies have shown impaired functioning of fish and copepods following exposure to water and sediments from the Seine Estuary. In addition, a number of contributions in this volume describe novel field- and laboratory-based protocols which characterise biological responses to contaminants in Channel environments using, pertinent (validated) model organisms and advanced molecular analysis (omics). Despite recent advances, such approaches, combining mode of action / adverse outcome pathways, designed to examine the effects of multiple stressors, have yet to gain traction in studies on contaminant impacts in English Channel waters. It is clear that the biological effects of mixtures of toxic substances must be a focus for future research, despite the difficulties in distinguishing natural and anthropogenic causality in the field and complications in simulating environmental conditions in the laboratory. Clearly, identification of the toxic moieties in contaminant mixtures, and their synergistic or antagonistic impacts on organisms, requires a high level of excellence in marine analytical chemistry and biology that will be difficult to achieve without adequate resourcing.

The process-oriented projects alluded to above will require holistic, inter-disciplinary, international research that combines radical experimental design, advanced instrumentation, innovative methodologies and intellectual prowess. Arguably, the last time such exacting scientific criteria were met was during the EU FLUXMANCHE programmes from 1990 and 1994. New, advanced scientific insights would contribute significantly to policy development within both the Water Framework Directive and Marine Strategy Framework Directive, the major future drivers of good water quality in Europe's coastal seas. In the longer term the datasets could be of great value to the visionary EU Copernicus Marine Environment Monitoring Service. It is evident greater effort should be devoted to bringing together the leading research groups, from both sides of the English Channel, to conduct these major inter-disciplinary research programmes.

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Table 1. River systems in OSPAR Region II subject to RID reporting (from Uncles and Stephens, 2007; OSPAR, 2013a).

OSPAR region	Country region	OSPAR zone code	River name	Long-term mean freshwater flow ($m^3 s^{-1}$)
II (13)	Pas de Calais	II-AP-PC-Aa	Aa	32
	Somme	II-AP-SO-Canche	Canche	53
		II-AP-SO-Somme	Somme	37
	Normandie	II-SN-NO-Bethune	Béthune et Bresle	24
		II-SN-NO-Saane	Saane	34
	Seine	II-SN-SE-Seine	Seine	519
		II-SN-SE- Andelle	Andelle	8
		II-SN-SE-Eure	Eure	26
		II-SN-SE-H7	-	18.5
		II-SN-SE-Risle	Risle	19
	Nord Cotentin	II-SN-NC-Dives	Dives	15
		II-SN-NC-Douve	Douve	7.2
		II-SN-NC-Orne	Orne	29
		II-SN-NC-Seulles	Seulles	4
		II-SN-NC-Touques	Touques	12
		II-SN-NC-Vire	Vire	26
	Sud Cotentin	II-SN-SC-I6	-	13.6
		II-SN-SC-Selune	Sélune et Sée	23
		II-SN-SC-Sienne	Sienne	15.4
	Nord Bretagne	II-LB-NB-Aulne	Aulne	77
		II-LB-NB-Couesnon	Rance et Couesnon	25
		II-LB-NB-J1J2	-	42.3
	SERBD[a]	E13	Medway	11.7
		E13	(Great) Stour	3.3
		E13	Rother	2.5
		E14	Adur	2.5
		E14	Ouse	4.2
		E14	Cuckmere	1.3
		E14	Arun	8.5
		E15	Itchen	1.9
		E15	Test	11.2
		E15	Blackwater	0.9
	SWRBD[b]	E16	Frome	8.9
		E16	Stour	-
		E16	Avon	3.3
		E17	Axe	5.0
		E17	Dart	11.1
		E17	Exe	23.3
		E17	Gara	-
		E17	Otter	3.1
		E17	Teign	9.3
		E18	Cober	-
		E18	Erme	1.9
		E18	Fal	10.0
		E18	Fowey	5.1
		E18	Gara	-
		E18	Par	-
		E18	Porthleven	-
		E18	St Austel	-
		E18	Plym	22.5 (Tamar) 44.7 (Plymouth Sound)
		E18	Lynher	
		E18	Tavy	
		E18	Tamar	

[a]SERBD, South East River basin District; [b]SWRBD, South West River Basin District

Table 2. Summary of data from the OSPAR Coordinated Environmental Monitoring Programme (CEMP) for metal and organic contaminants in sediments and biota (from OSPAR, 2009e).

	Concentrations		**Temporal trend in concentrations**		**Concentrations**		**Temporal trend in concentrations**	
	UK coast				**French coast (Seine only)**			
	Sediment	**Biota**	**Sediment**	**Biota**	**Sediment**	**Biota**	**Sediment**	**Biota**
PCB	Red	Red	None - Up	None - Down	Green - Red	Red	nd	None - Down
γ - HCH	nd	nd	nd	nd	Green	Green – Red	nd	None - Down
PAH	Green - Red	Red	None - Up	None	Green - Red	Red	nd	Down
Cd	Green	Amber	None – Up	None - Down	Red	Amber	nd	None
Pb	Red	Amber - Red	None-Up	None - Down	Red	Amber	nd	Down
Hg	Green - Red	Amber	None - Up	None	Red	Amber	nd	None
					French coast (no Seine)			
PCB					nd	Green – Red	nd	Down
γ - HCH					nd	Blue – Red	nd	None - Down
PAH					nd	Green	nd	None - Down
Cd					nd	Blue – Amber	nd	None - Up
Pb					nd	Blue – Amber	nd	None - Down
Hg					nd	Blue – Amber	nd	None - Up

Key:

Red : Status is unacceptable. Concentrations are at levels such that there is an unacceptable risk of chronic effects occurring in marine species, including the most sensitive species (PAHs and PCBs in biota; PAHs, PCBs and metals in sediment), or are greater than EU dietary limits for fish or shellfish but the extent of risks of pollution effects is uncertain (metals in biota).

Amber : Status is uncertain. Concentrations of metals in biota are lower than EU dietary limits for fish and shellfish and above background but the extent of risks of pollution effects is uncertain.

Green : Status is acceptable. Concentrations of contaminants (except metals in biota) are at levels where it can be assumed that little or no risks are posed to the environment and its living resource at the population or community level.

Blue : Status is acceptable. Concentrations are near background (metals, PAHs) or close to zero (PCBs) i.e. the ultimate aim of the OSPAR Strategy for Hazardous Substance has been achieved.

nd, no data

Table 3. Time-dependent dissolution of ΣPAH from Arabian crude into seawater and their subsequent uptake into the muscle of sea bass, *Dicentrarchus labrax* (from Kerambrun et al., 2012a).

Time, h	Dissolved ΣPAH, µg L^{-1}		Muscle ΣPAH, µg g^{-1} (dry wt)	
	Control	Oil	Control	Oil
0	0.43 ± 0.23	48.7 ± 15.7		
48	0.48 ± 0.68	30.4 ± 12.9	0.49 ± 0.13	5.72 ± 1.40
96	0.48 ± 0.93	10.9 ± 2.80	0.62 ± 0.16	2.29 ± 0.47

Table 4. Liquid discharges (TBq) of radionuclides from nuclear industries to the English Channel for 2011[b] and 2012[a] (France) (OSPAR, 2013g; IRSN, 2011; 2012) and 2012 (UK) (CEFAS, 2013). [a]The Sellafield data are reported for comparison. Values in brackets are for gaseous discharge.

Location	Activity	Discharge of ^{3}H, TBq	Other Radionuclides
France			
La Hague	Reprocessing	11600[a] (55)[a]	Total α=0.02 TBq Total β=2.85 TBq Plus smaller quantities of γ-emitting radionuclides
Flamanville	Power Station	63.5[b]	^{58}Co; ^{60}Co; ^{110m}Ag; ^{125}Sb; ^{134}Cs; ^{137}Cs
Paluel	Power Station	110[b]	^{58}Co; ^{60}Co; ^{110m}Ag; ^{125}Sb; ^{134}Cs; ^{137}Cs
Penly	Power Station	58.4[b]	^{58}Co; ^{60}Co; ^{110m}Ag; ^{125}Sb; ^{134}Cs; ^{137}Cs
Nogent-sur-Seine	Power Station	37.9[a]	^{58}Co; ^{60}Co; ^{110m}Ag; ^{125}Sb; ^{134}Cs; ^{137}Cs
UK			
Sellafield[a]	Reprocessing	1050 (102)	Total α=0.142 TBq Total β=9.49 TBq Plus quantities of γ-emitting radionuclides
Devonport	Defence Facility	0.21	^{14}C; plus β- and γ - emitting radionuclides
Dungeness A	Power Station	0.08	^{137}Cs
Dungeness B	Power Station	162	^{35}S; ^{41}Ar; ^{14}C
Winfrith	Research Establishment	26.9	Total α=9.9 x 10^{-5}TBq ^{137}Cs=9.27 x 10^{-5} TBq

Table 5. Concentrations (mg L^{-1}) of nitrate-N and phosphate-P in catchment rivers during 2010 (from ETC/ICM, 2012).

	Nitrate-N	**Phosphate-P**
SWRBD[a]	2.0 -3.6	0.05 – 0.10
SERBD[b]	3.6 – 5.6	0.20 - 0.40
Loire RBD	2.0 -3.6	0.02 – 0.05
Seine RBD	3.6 – 5.6	0.02 – 0.05
Scheldt RBD	3.6 – 5.6	0.05 – 0.10

[a]SWRBD, South West River basin District; [b]SERBD, South East River Basin District

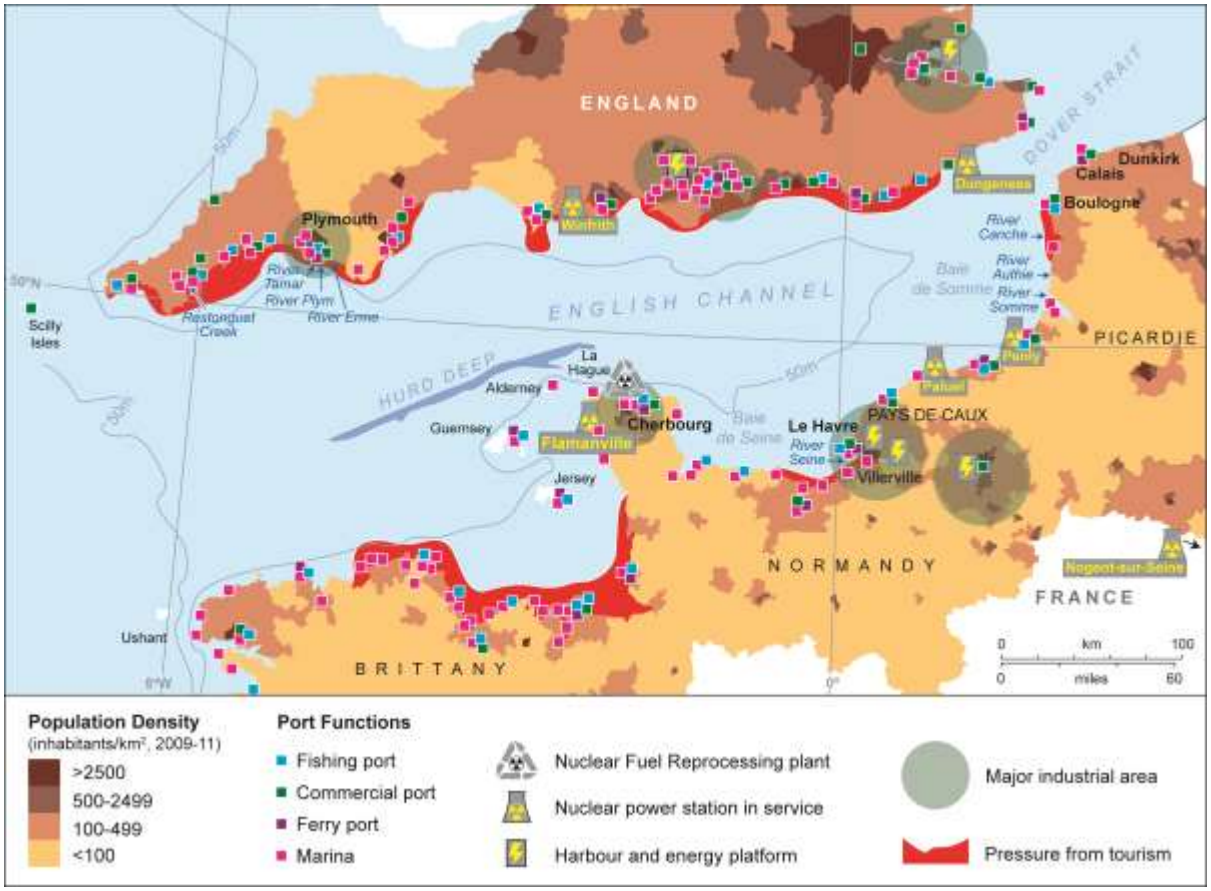

Figure 1. Geography and bathymetry of the English Channel. The population density and coastal industries are also shown, as are place names given in the text (adapted from Uncles and Stephens (2007) and University of Caen (2014)).

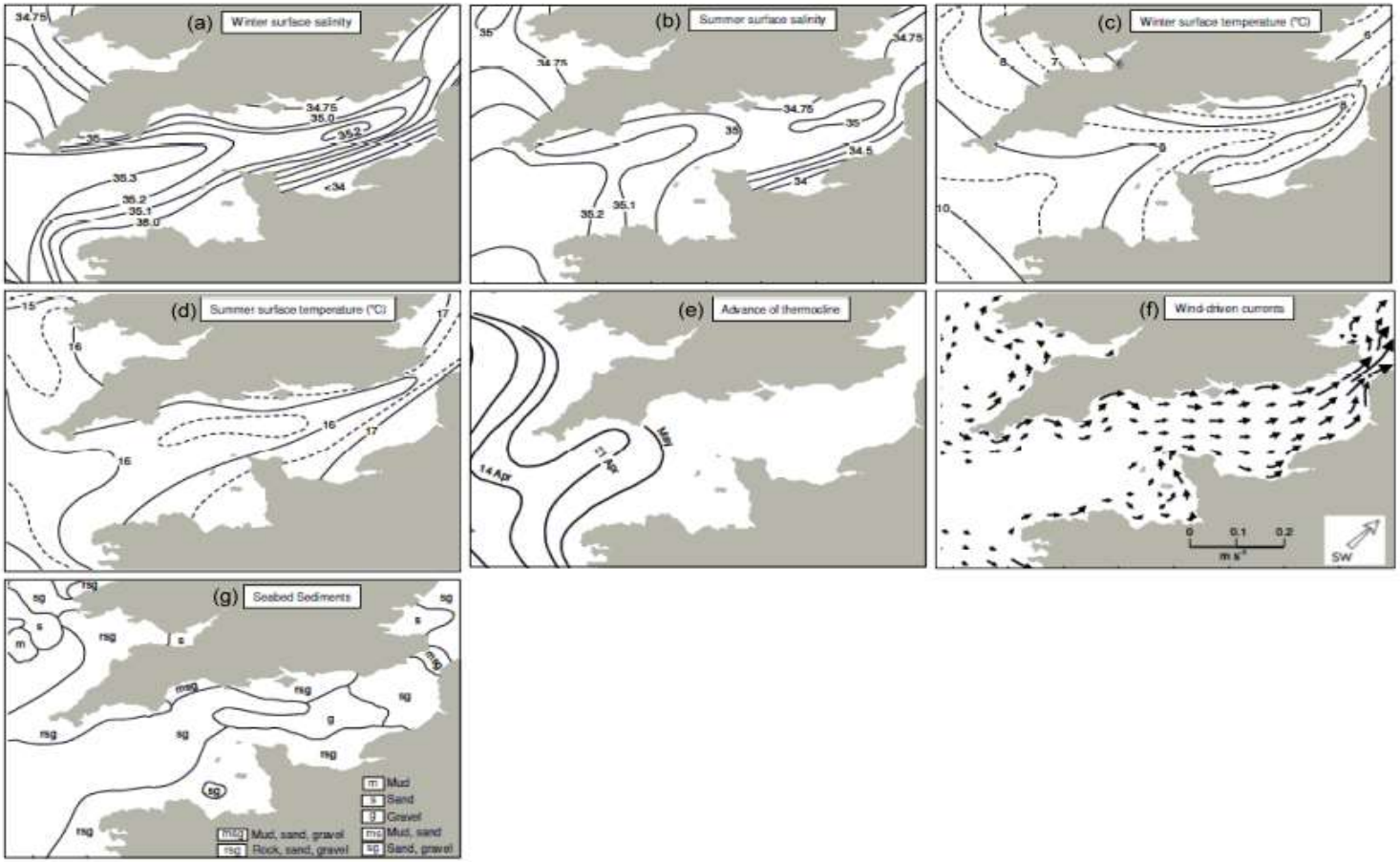

Figure 2. A summary of the physical oceanography and sedimentology of the English Channel. (a) winter surface salinity, (b) summer surface salinity, (c) winter surface temperature, (d) summer surface temperature, (e) temporal advance of the seasonal thermocline (f) residual currents for a southwest wind (arrows representing residual velocities of < 0.01 m s^{-1} have been omitted) and (g) distribution of surficial seabed sediment types (from Uncles and Stephens, 2007).

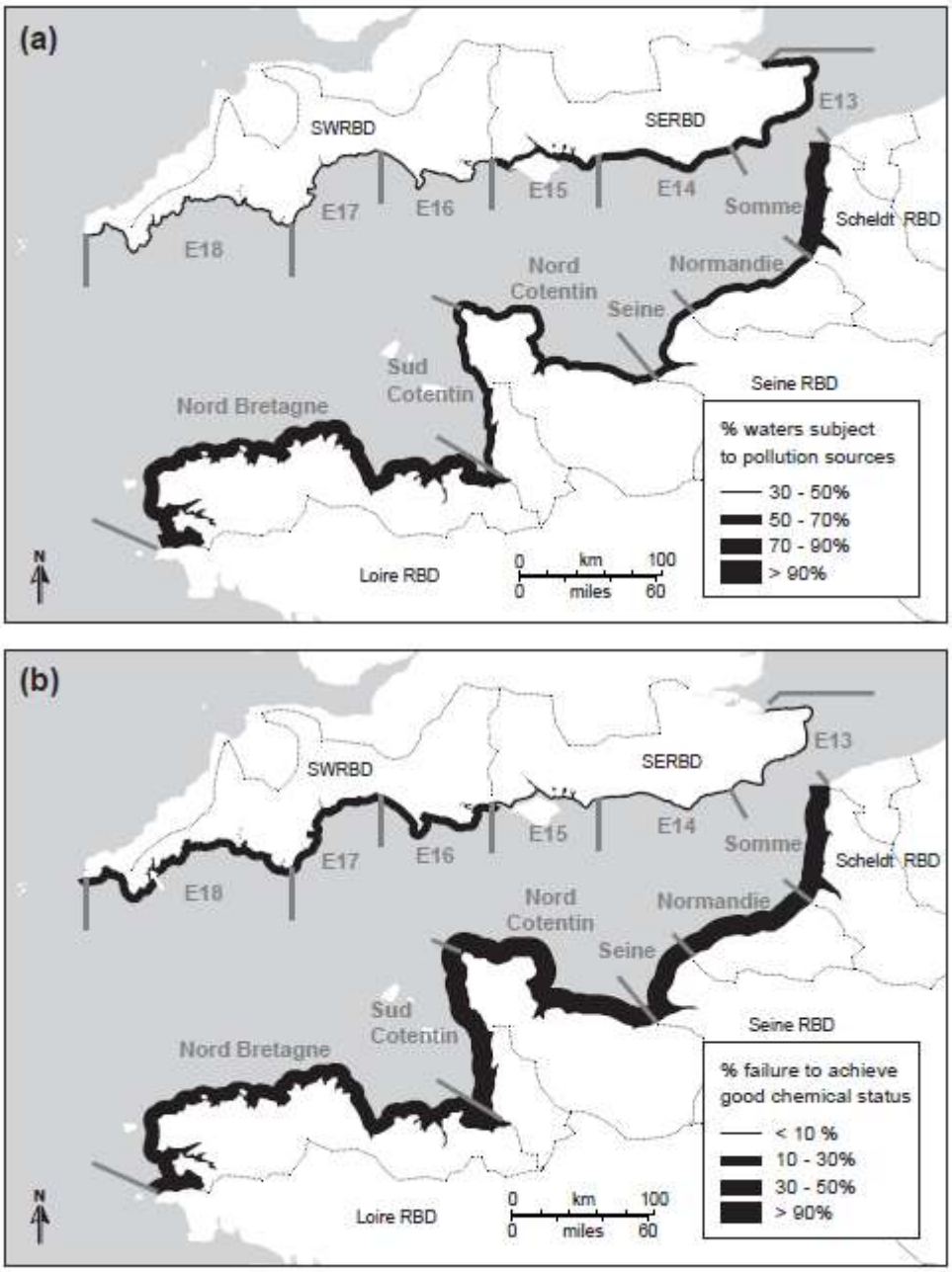

Figure 3. River basins draining into the English Channel and OSPAR RID reporting zones. Proportion (as a percentage) of transitional and coastal waters (a) subject to point and diffuse source pollution pressures and (b) failing to achieve good chemical status. (Adapted from ETC/ICM, 2012; OSPAR, 2013a; European Environment Agency, 2014). SWRBD, South West River Basin District; SERBD, South East River Basin District.

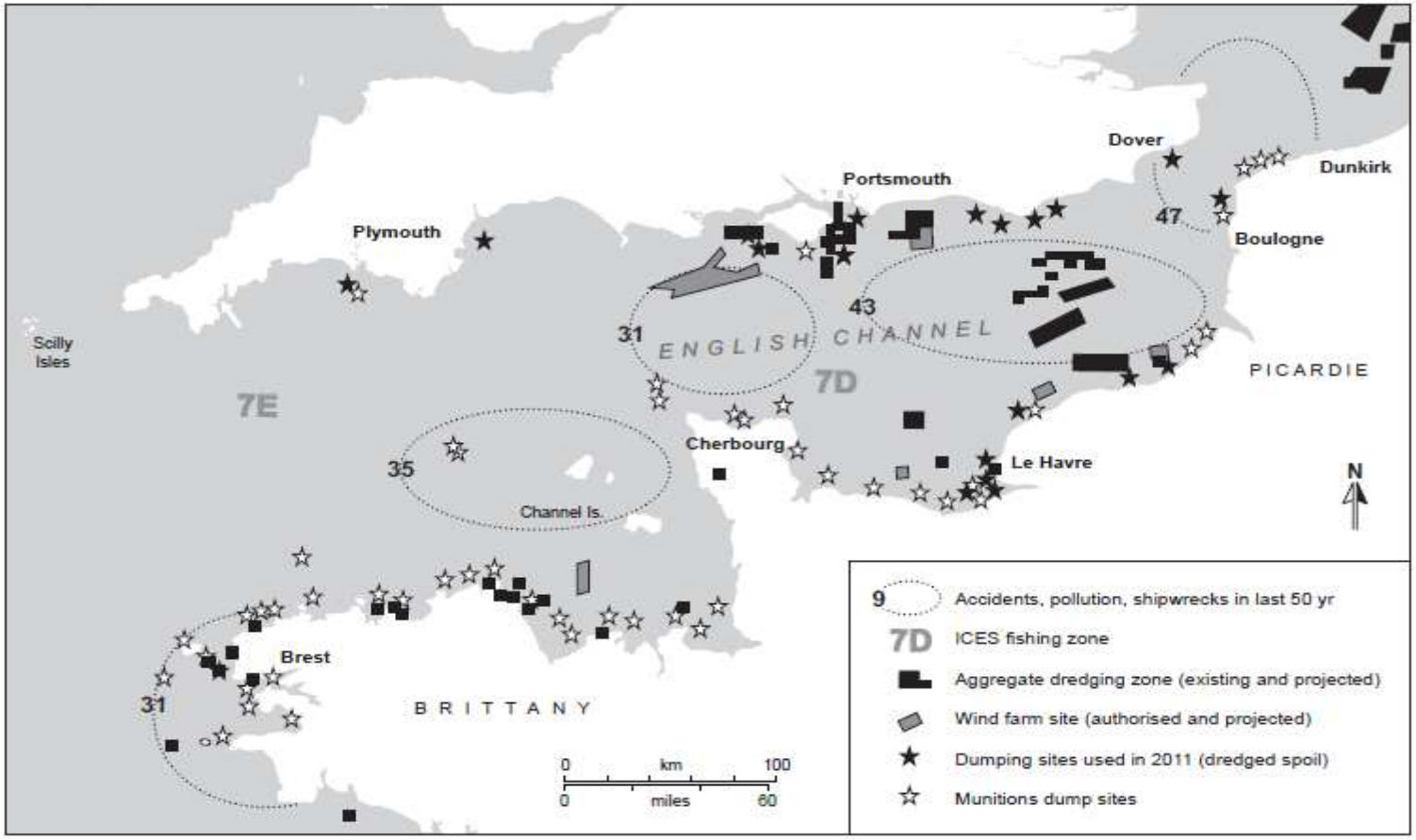

Figure 4. Offshore industry, dumping sites and accident/pollution frequency (adapted from OSPAR, 2009a, 2010a,b, 2013c; University of Caen, 2014).

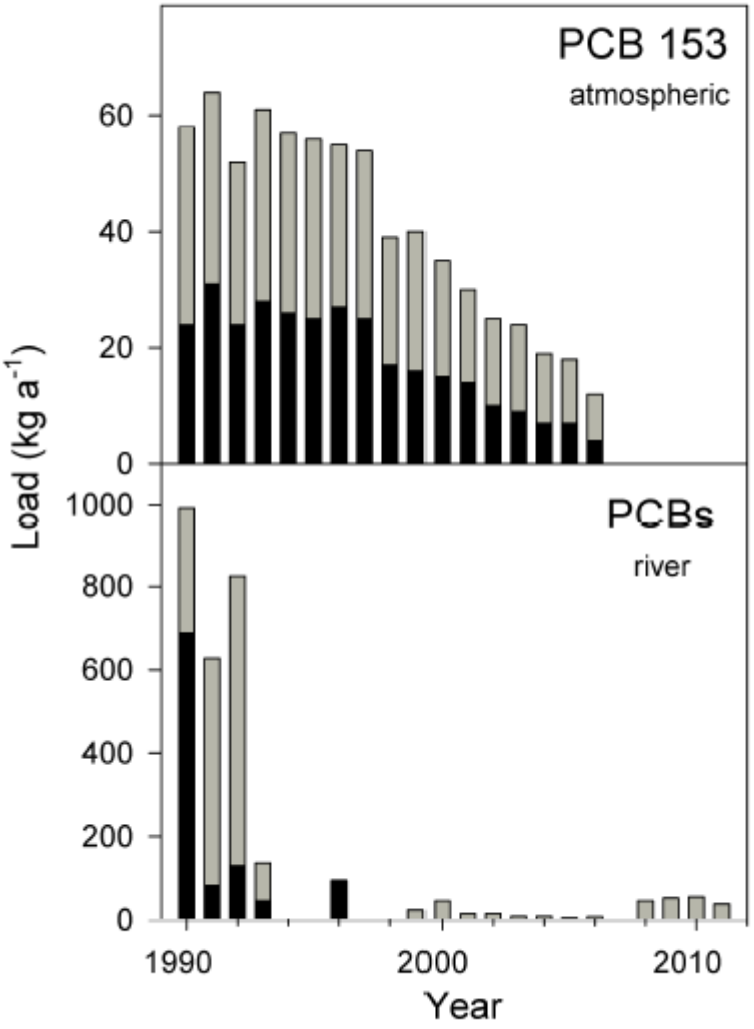

Figure 5. Total atmospheric and total riverine loads of PCBs to the English Channel (from OSPAR, 2001, 2002, 2004, 2005a, 2006a,b, 2007, 2008, 2009c,d, 2010c, 2011a, 2012a, 2013a). The atmospheric loads are modelled results. Atmospheric load: grey + black represents total deposition, black represents net deposition only. River load: grey represents upper estimate and black represents lower estimate where these can be differentiated.

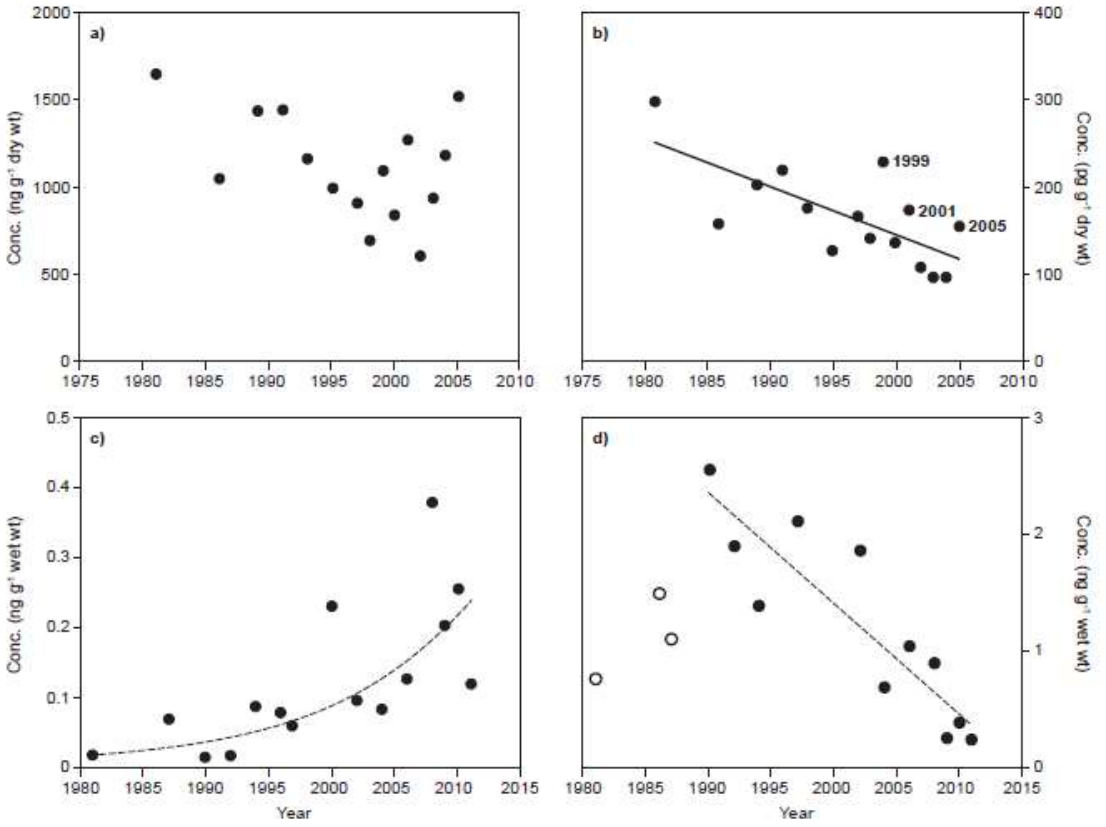

Figure 6. Temporal trends in the concentrations of (a) ΣPCBs (7 ICES congeners), (b) ΣPCDD/Fs (17 ICES congeners), (c) HBCDD (α-isomer only) and (d) PFOS in soft tissues of *Mytilus edulis* from the Seine Estuary (redrawn from Munschy et al., 2008, 2013).

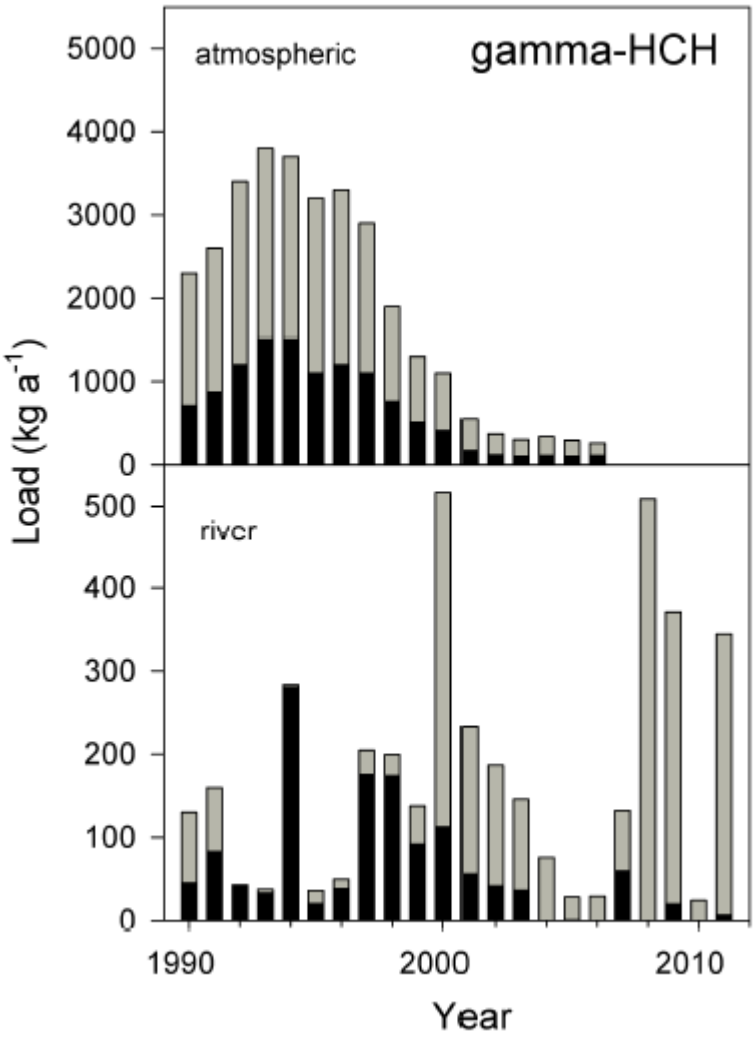

Figure 7. Total atmospheric and total riverine loads of gamma-HCH to the English Channel. See Fig. 5 for data sources. The atmospheric loads are modelled results. Atmospheric load: grey + black represents total deposition, black represents net deposition only. River load: grey represents upper estimate and black represents lower estimate where these can be differentiated.

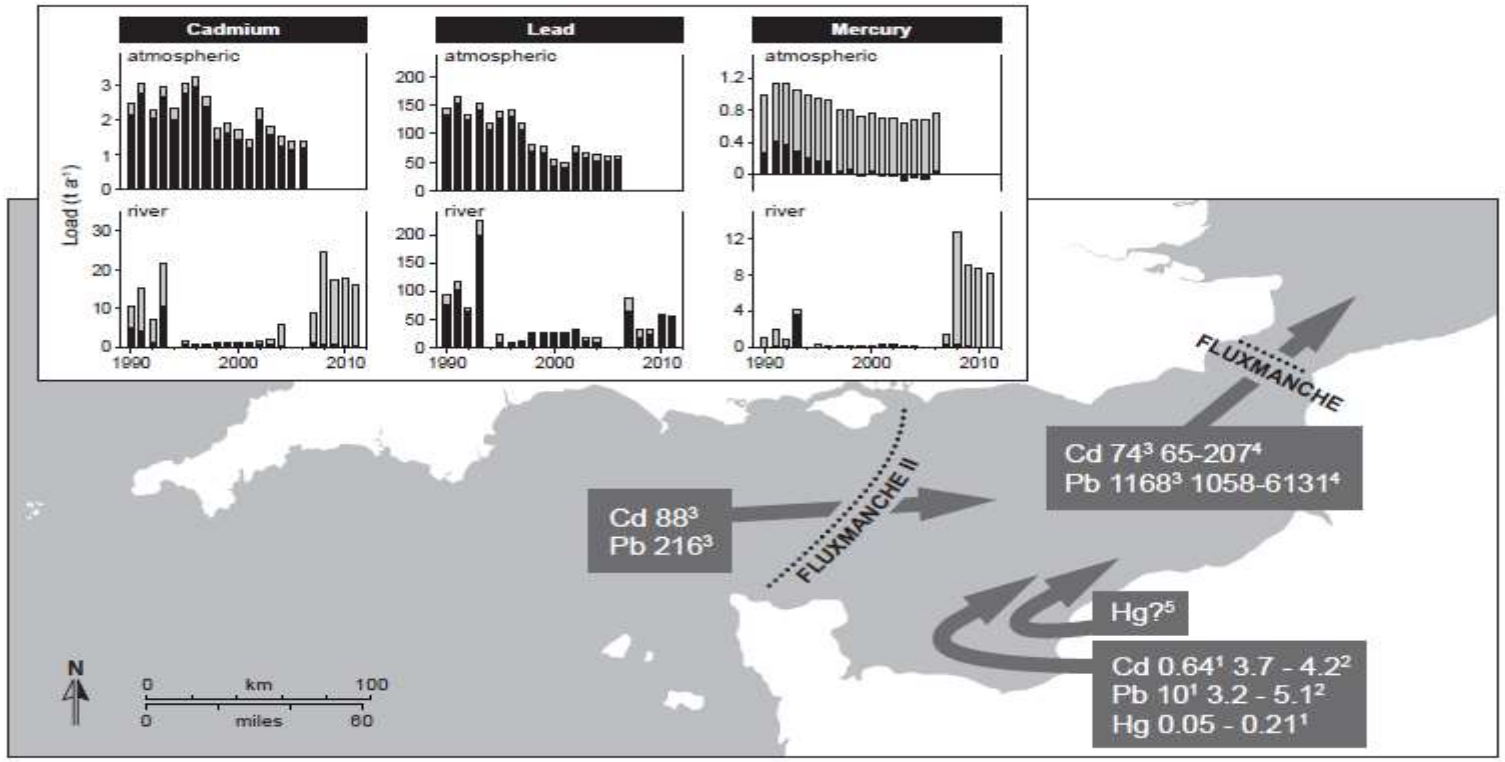

Figure 8. Total atmospheric and total river loads of lead, cadmium and mercury within the English Channel, total loads across the FLUXMANCHE transects (dashed lines) and the total[1] and dissolved[2] loads from the Seine river estuary. From: [1]OSPAR (2006b), [2]Chiffoleau et al. (1999), [3]Statham et al. (1999), [4]Tappin et al. (1997), [5]Laurier et al. (2007). See Fig. 5 for additional data sources, plus OSPAR (2005b). Inset, upper panel: Atmospheric load: grey + black represents total deposition, black represents net deposition only. Inset, lower panel: Total river load: grey represents upper estimate and black represents lower estimate where these can be differentiated.

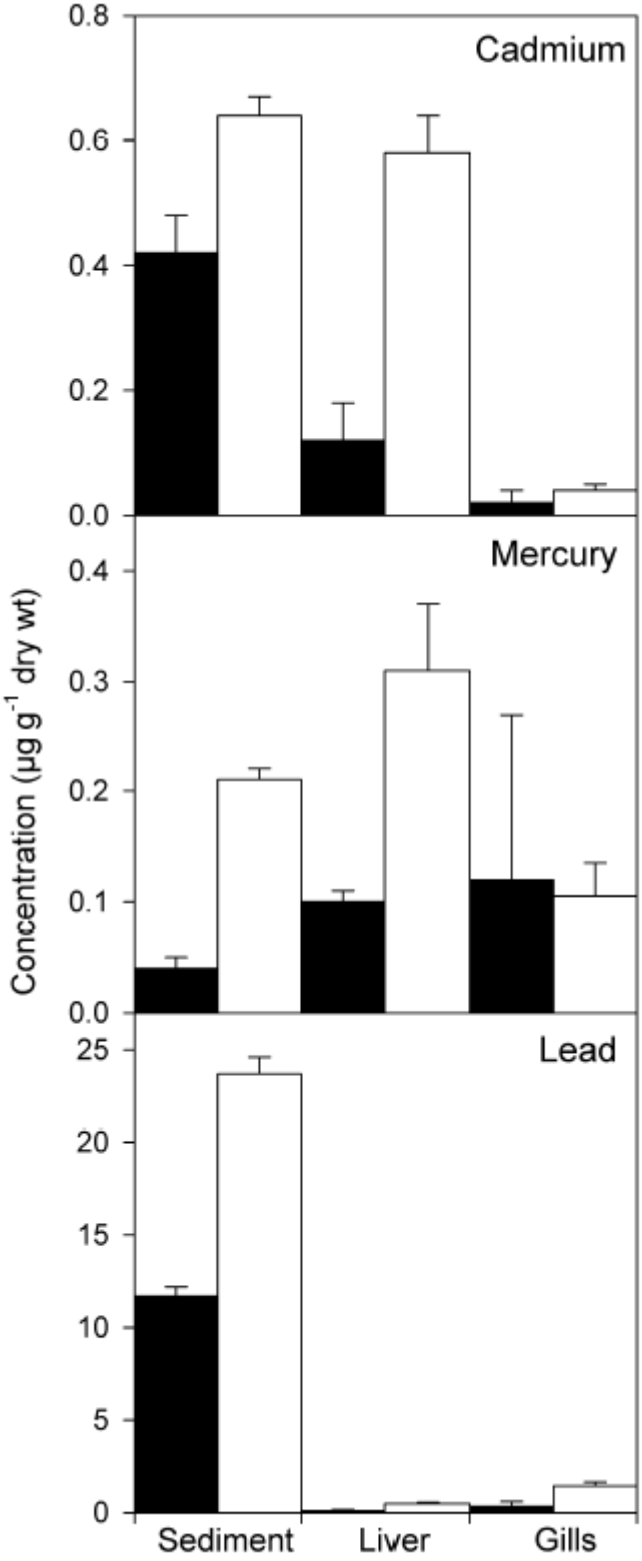

Figure 9. Mean concentrations (± 1σ) of Cd, Hg and Pb in the sediments, livers and gills of *Platichthys flesus* from the Canche Estuary (solid fill) and the Seine Estuary (no fill) (redrawn from Kerambrun et al., 2013).

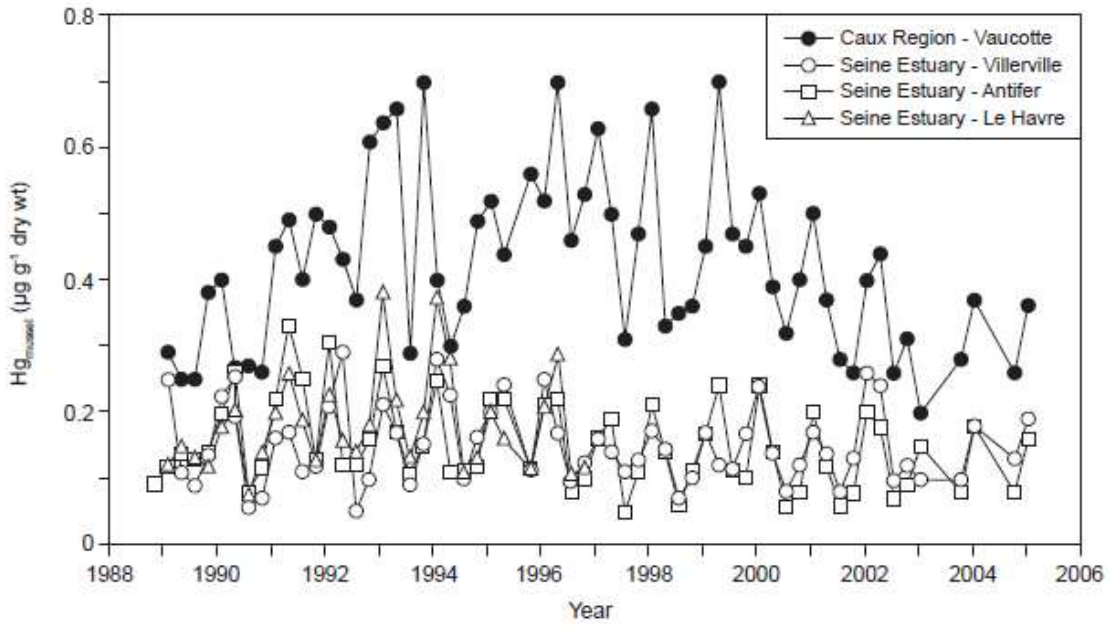

Figure 10. Temporal trend in the concentration of Hg in the soft tissues of *Mytilus edulis* from the Seine Estuary and Pays de Caux region (redrawn from Laurier et al., 2007).

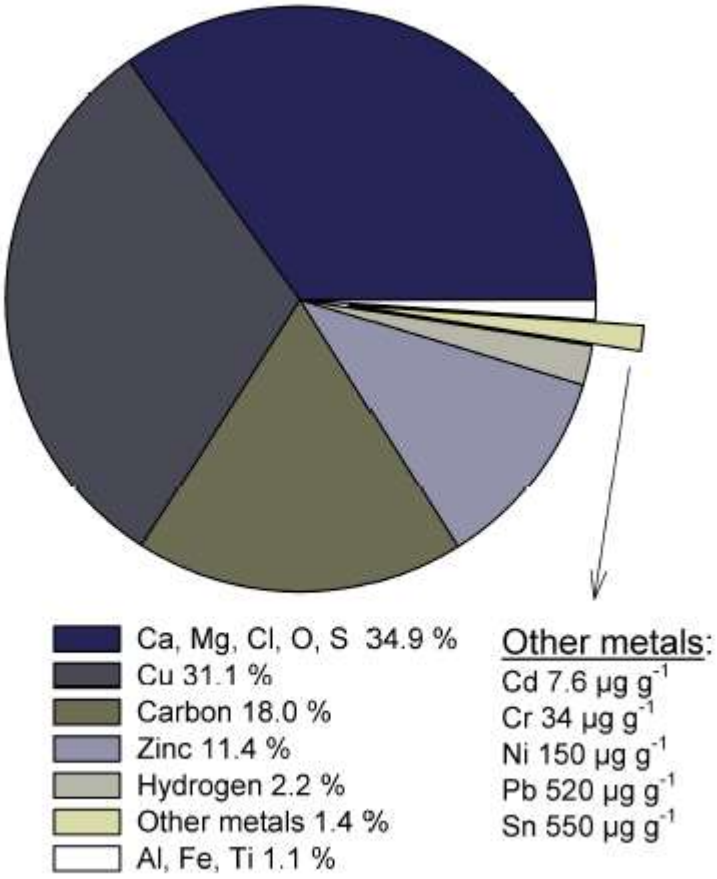

Figure 11. Elemental composition (dry wt) of an antifouling paint composite sample obtained from a leisure boat marina in Plymouth, SW England (redrawn from Turner, 2010).

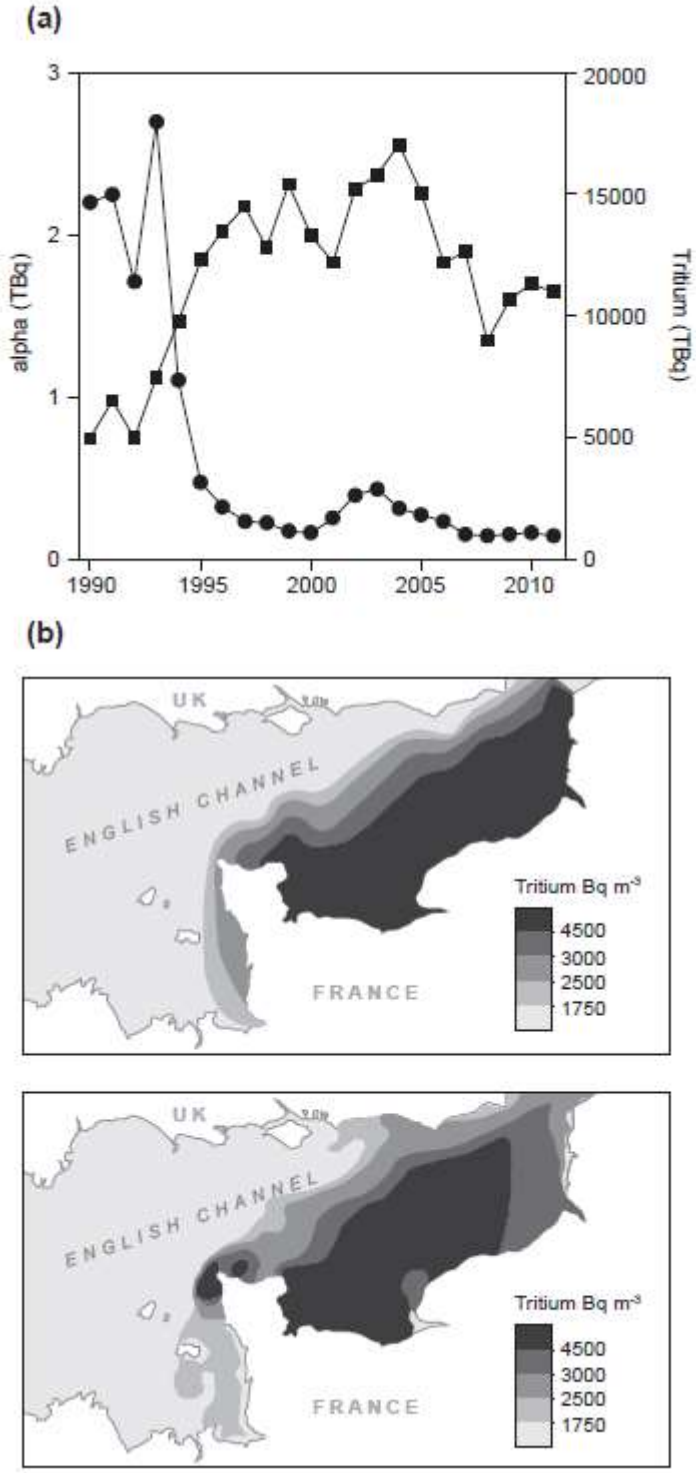

Figure 12. (a) Temporal trends in the discharge of tritium (■) and total alpha (●) from the nuclear reprocessing plants at La Hague and Sellafield (from OSPAR, 2013g) and (b) measured and simulated distributions of tritium in the English Channel (adapted from Bailly du Bois and Dumas, 2005).

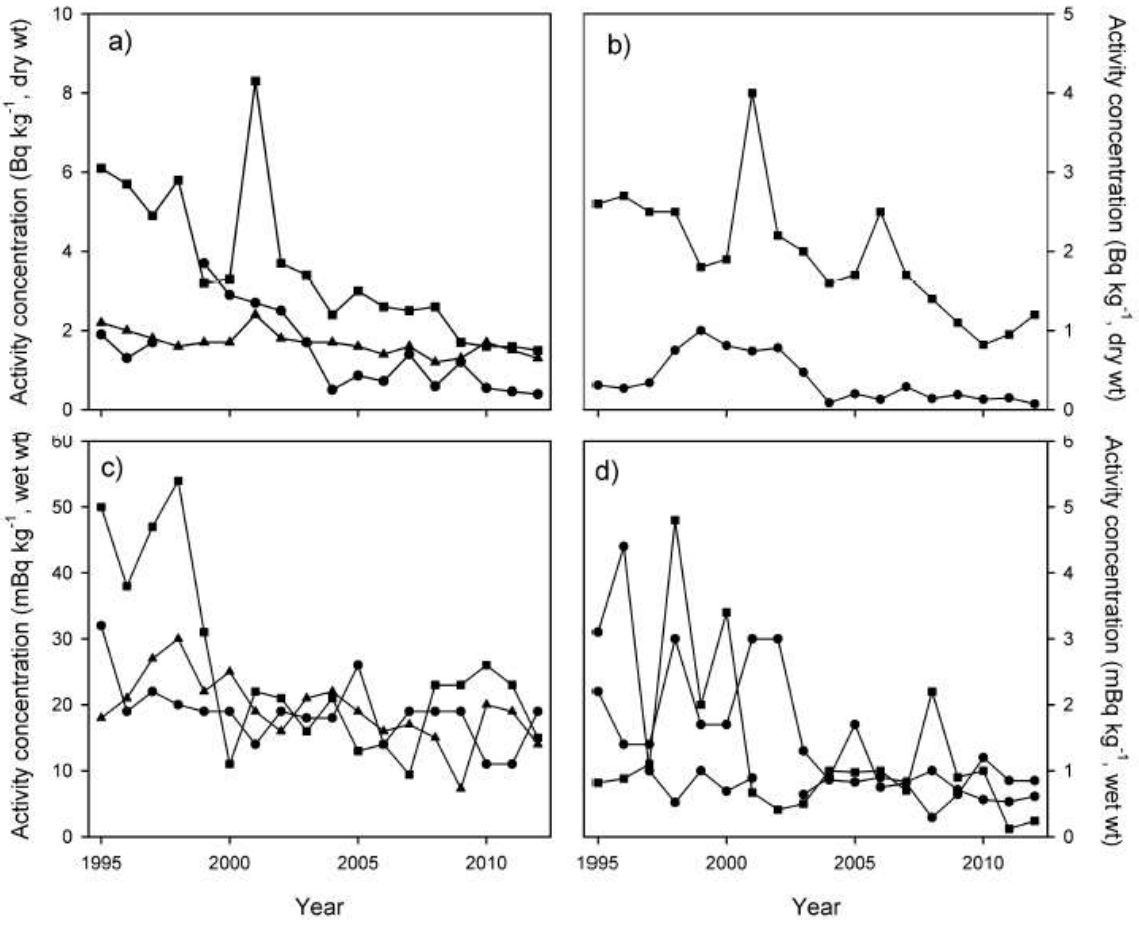

Figure 13. Activity concentrations of artificial radionuclides in sediments and marine biota from the Channel Islands for the period 1995 – 2012. (a) ^{137}Cs in sediments; note a value of 20 Bq kg^{-1} for 1998 has been omitted (b) ^{239,240}Pu in sediments (c) ^{239,240}Pu in *Fucus* spp. and (d) ^{239,240}Pu in edible crabs. ■ Jersey, ● Guernsey, ▲, Alderney. Data compiled in CEFAS (2013).

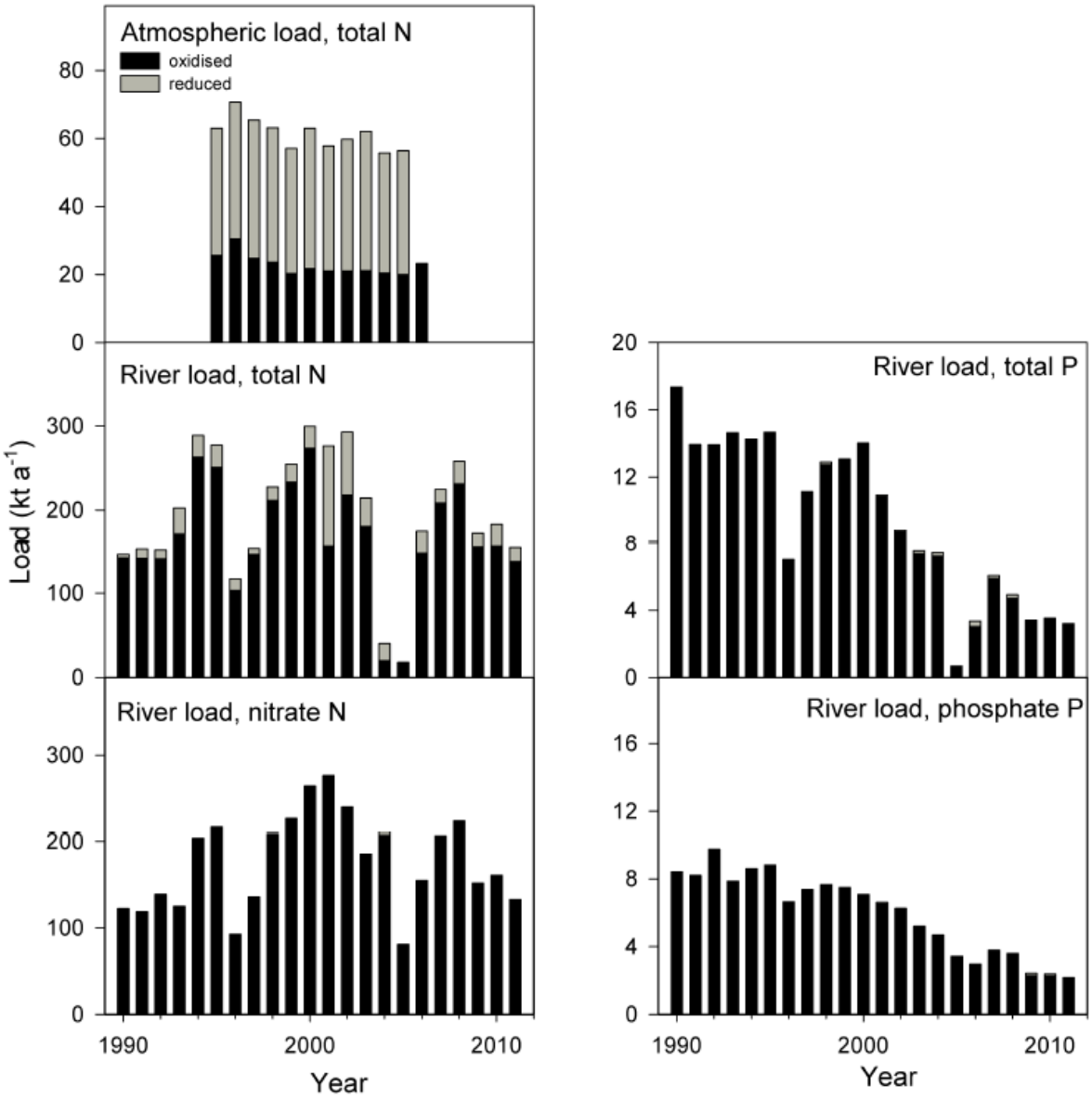

Figure 14. Atmospheric and riverine loads of total N, total P, nitrate-N and phosphate-P to the English Channel. The total fractions include organic forms of nitrogen and phosphorus. The UK did not undertake this analysis and so UK data are for nitrate-N and phosphate-P only. See Fig. 5 for additional data sources, plus OSPAR (2005b). River load: grey represents upper estimate and black represents lower estimate where these can be differentiated.

Figure 15. Eutrophication status of English Channel waters for 2001 – 2005, as classified by the OSPAR Common Procedure (redrawn from OSPAR, 2010a).

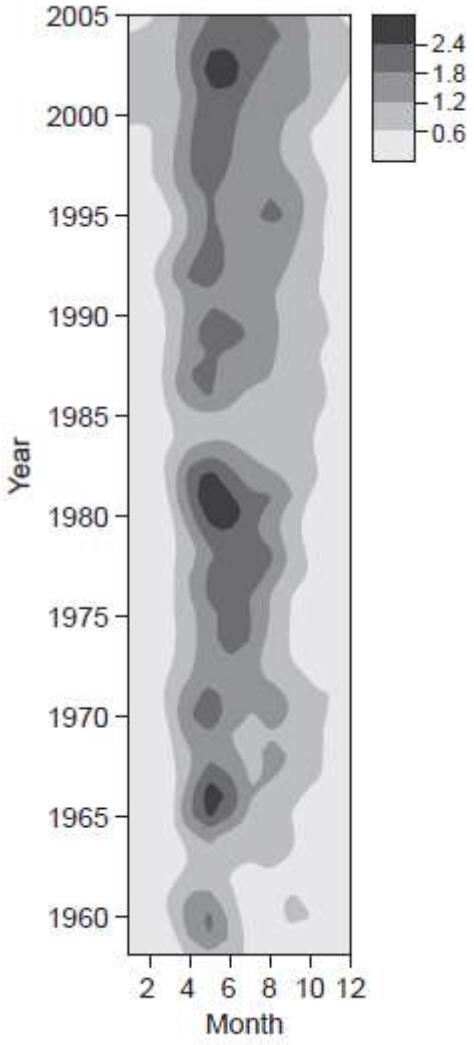

Figure 16. Temporal changes in the Phytoplankton Colour Index between 1960 and 2005. The scale is logarithmic, with black representing the highest primary production (adapted from Johns, 2006).

Appendix 1. Data sources for contaminants relevant to this review.

Data component	Data availability Year, (location, sample)	Data source
Polychlorinated biphenyls		
Aeolian load	1990-2006 (Channel, model results)	OSPAR, 2009c
River load UK	1990-2011	OSPAR, 2001, 2002, 2004, 2005a, 2006a, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
River load France	1990-1996 (Seine only)	OSPAR, 2001, 2006b
Water UK	no data[a]	
Water France	2002-2005(Seine)	Cailleaud et al., 2007a
Sediment UK	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2009	CEFAS, 2012
Sediment France	2002-2005 (Seine, SPM)	Caillard et al., 2007a
	1998-2012	OSPAR, 2009e, 2012b, 2013f
Biota UK	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2010 (*L. limanda*)	CEFAS, 2012
Biota France	2001 (Pays de Calais, *M. edulis, A. rubens*)	Danis et al., 2006
	1980-2005 (Seine, *M. edulis*)	Munschy et al., 2008
	2002-2005 (Seine, *E. affinis*)	Caillard et al., 2007a
	2003-2004 (Seine, *M. brachydactyla*)	Bodin et al., 2007a
	2003-2004 (Brittany, Normandy coasts, edible crabs, *M. edulis, D. labrax*)	Bodin et al., 2007b,c
	1998-2012	OSPAR, 2009e, 2012b, 2013f
Polychlorinated-p-dibenzo-dioxins / polychlorinated-p-dibenzo-furans		
Aeolian load	1998-2007(Channel, terrestrial emission to atmosphere)	OSPAR, 2009c
River load UK	no data	
River load France	no data	
Water UK	no data	
Water France	no data	
Sediment UK	no data	
Sediment France	no data	
Biota UK	no data	
Biota France	2001 (Pays de Calais, *M. edulis, A. rubens*)	Danis et al., 2006
	2003-2004 (Brittany, Normandy coasts, edible crabs, *M. edulis, D. labrax*)	Bodin et al., 2007c
	1981-2005 (Seine, *M. edulis*)	Munschy et al., 2008
Polybrominated diphenyl ethers		
Aeolian load	2008 (estimated particle dry deposition flux)	Xie et al., 2011
River load UK	no data	
River load France	no data	
Water	2008 (mid-Channel)	Xie et al., 2011
Sediment	2008 (mid-Channel, SPM)	Xie et al., 2011
	2009-2010 (UK coast)	CEFAS, 2012
Biota UK	2003-2010 (*L. limanda*)	CEFAS, 2012
Biota France	1981-2003 (Seine, *M. edulis, M.*	Johansson et al., 2006

	galloprovincialis)	
	2003-2004 (Brittany, Normandy coasts, edible crabs)	Bodin et al., 2007c
	Annual cycle (Seine, *S. plana, N. diversicolor, S. solea, A. anguilla*)	Bragigand et al., 2006
	2007-2009 (Seine, *S. solea*)	Munschy et al., 2011
Hexabromocyclododecanes		
Aeolian load	no data	
River load UK	no data	
River load France	no data	
Water UK	no data	
Water France	no data	
Sediment UK	no data	
Sediment France	no data	
Biota UK	no data	
Biota France	2008, 2010 (French coast, *M. edulis*, *M. galloprovincialis*, *C. gigas*)	Munschy et al., 2013
	1981-2011 (Seine, *M. edulis*)	Munschy et al., 2013
Polyfluorinated compounds		
Aeolian load	no data	
River load UK	no data	
River load France	2006-2007 (Seine only)	Pistocchi and Loos, 2009
Water UK	no data	
Water France	no data	
Sediment UK	no data	
Sediment France	no data	
Biota UK	no data	
Biota France	2010 (French coast, *M. edulis*, *M. galloprovincialis*, *C. gigas*)	Munschy et al., 2013
	1981-2011 (Seine, *M. edulis*)	Munschy et al., 2013
Pesticides		
Aeolian load	1990-2006 (Channel, model results)	OSPAR, 2009c
River load UK	1990-2011	OSPAR, 2001, 2002, 2004, 2005a, 2006a, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
River load France	1990-2011 (no Seine 1990-1996, 2004-2006, 2010)	OSPAR, 2006b, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
Water UK	no data	
Water France	no data	
Sediment UK	1998-2008	OSPAR, 2009e, 2012b
Sediment France	1998-2008	OSPAR, 2009e, 2012b
Biota UK	1998-2008	OSPAR, 2009e, 2012b
Biota France	1998-2008	OSPAR, 2009e, 2012b
	2003 (Brittany, Normandy coasts, edible crabs)	Bodin et al., 2007b
Polycyclic aromatic hydrocarbons		
Aeolian load	no data	
River load UK	no data	
River load France	no data	
Water UK	no data	
Water France	2002-2005 (Seine)	Cailleaud et al., 2007a
Sediment UK	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2000-2010	CEFAS, 2012

Sediment France	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2002-2005 (Seine, SPM)	Cailleaud et al., 2007a
Biota UK	1998-2012	OSPAR, 2009e, 2012b, 2013f
Biota France	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2002-2005 (Seine, *E. affinis*)	Cailleaud et al., 2007a
Alkylphenyl-polyethoxylates		
Aeolian load	no data	
River load UK	no data	
River load France	no data	
Water UK	1994-1995 (Channel coast)	Blackburn and Waldock, 1995; Blackburn et al., 1999
Water France	2002-2005 (Seine)	Cailleaud et al., 2007b
Sediment UK	no data	
Sediment France	2002-2005 (Seine, SPM)	Cailleaud et al., 2007b
Biota UK	no data	
Biota France	2002-2005 (Seine, *E. affinis*)	Cailleaud et al., 2007b
Phthalates		
Aeolian load	no data	
River load UK (emission data)	2008	ETC/ICM, 2012
River load France (emission data)	2008	ETC/ICM, 2012
Water UK	no data	
Water France	2005-2006 (Seine)	Dargnat et al., 2009
Sediment UK	no data	
Sediment France	no data	
Biota UK	no data	
Biota France	no data	
Cadmium, lead and mercury		
Aeolian load	1990-2006 (Channel, model results)	OSPAR, 2009c
River load UK	1990-2011	OSPAR, 2001, 2002, 2004, 2005a,b, 2006a, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
River load France	1990-2011	OSPAR, 2006a,b, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
	1994-1995 (Seine, Cd, Pb)	Chiffoleau et al., 1999
	2000-2001 (Pays de Caux, groundwater Hg)	Laurier et al., 2007
Water	1994-1995 (Cd, Pb, dissolved, SPM)	Statham et al., 1999
Sediment UK	No date (SWRBD)	Amiard et al., 2007
	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2010(SWRBD)	Varma et al., 2011
Sediment France	No date (Seine, Boulogne)	Amiard et al., 2007
	1998-2012	OSPAR, 2009d, 2012b, 2013f
	2002-2003 (Deûle River)	Kadlecová et al., 2012
	1990s on(Seine)	Dauvin, 2008
	2010(Canche, Seine)	Kerambrun et al., 2013
Biota UK	1998-2012	OSPAR, 2009e, 2012b, 2013f
	2010 (SWRBD, *F. ceranoides*)	Varma et al., 2011
	No date (SWRBD, *N. diversicolor*)	Amiard et al., 2007
Biota France	No date (Seine, Boulogne, *N. diversicolor*)	Amiard et al., 2007

	1990s onward (Seine, macrofauna)	Dauvin, 2008
	1998-2012	OSPAR, 2009e, 2012b, 2013f
	1989-2005 (Seine, Pays de Caux, *M. edulis*)	Laurier et al., 2007
	1 year (Cherbourg Peninsula, *Patella sp*., *L. bergylta*, *C. pagurus*)	Connan and Tack, 2010
	2008, 2010 (Authie, Canche, Seine, Somme, *P. flesus*)	Henry et al., 2012; Kerambrun et al., 2013
	1998 (Coastal waters, *P. flesus*)	Henry et al., 2004
Emerging Metal Contaminants		
Aeolian load	no data	
Load UK	no data	
Load France	no data	
Water UK	No date (Tl, SWRBD)	Law & Turner, 2011
Water France	no data	
Sediments UK	No date (PGE, sorption to sediments)	Turner et al., 2006; Cobelo-Garcia et al., 2008; French & Turner, 2008
	No date (Antifouling paint particles, SWRBD)	Turner, 2010; Takahashi et al., 2012
Sediments France	no data	
Biota UK	No date (Tl, uptake by benthic organisms and algae)	Turner et al., 2013
	No date (PGE, uptake by *A. marina, C. stigmatophora*)	French & Turner, 2008; Shams et al., 2014
	No date (Cu, Zn, *M. edulis, A. marina*)	Turner, 2010
Biota France	no data	
Mixtures of Xenobiotic Substances		
Aeolian load	no data	
Industrial load UK	no data	
Industrial load France	No date (Discharges from petrochemical industries, refineries, pulp/paper mills)	Vincent-Hubert et al., 2012
Water UK	no data	
Water France	no data	
Sediments UK	no data	
Sediments France	No date (Seine, genotoxicity of SPM due to industrial discharges)	Vincent-Hubert et al., 2012
	2010 (*S. maximus*, contaminated sediments)	Kerabrum et al., 2012c
Biota UK	2009 (Proteomic composition, *P. flesus*)	Galland et al., 2013
Biota France	2009 (Cage experiments, *D. labrax*, *S. maximus*)	Kerabrum et al., 2012b
	2009 (Proteomic composition, *P. flesus*)	Galland et al., 2013
Radionuclides		
Aeolian inputs UK	1995-2012 (Power stations, defence facility, research)	CEFAS, 2013 (CD-ROM version)
Aeolian inputs France	2010-2011(Power stations, reprocessing plant, defence facility, research)	
	2012 (Power stations, reprocessing plant, defence facility, research)	
Industrial load UK	1950-1963 (Waste disposal, Hurd Deep)	Spiegel Online, 2013
	2012 (Power stations, defence facility,	CEFAS, 2013

	research)	
Industrial load France	2010-2011 (Power stations, defence facility, research)	IRSN, 2011
	2011 (Power stations, reprocessing plant)	OSPAR, 2013g
	2012 (Power stations, reprocessing plant, defence facility, research)	IRSN, 2012
Water	1987-1992 (La Hague, Channel)	Bailly du Bois et al., 2005
	1987-1992 (La Hague, Channel)	Guéguéniat et al., 1995, 1997
	1993 (La Hague, Channel)	McCubbin et al., 2002
	1995-2012 (UK coast)	CEFAS, 2013 (CD ROM version)
	2000-2001(North Cherbourg Peninsula)	Fiévet et al., 2006
	1963-2003 (^{137}Cs)	Povinec et al., 2003
	No date(La Hague, Channel)	Fiévet et al., 2013
	No date (^{3}H)	Turner et al., 2009
Sediment UK	1990-2009 (Channel Islands)	Hughes et al., 2011
	1995-2012 (Channel Islands; UK coast)	CEFAS, 2013 (CD-ROM version)
Sediment France	1994 (Channel)	Boust, 1999
	2002-2003 (Le Havre, Somme, Seine)	Cundy et al., 2002; Dubrulle et al., 2007
	2006-2007 (Western Channel, Cherbourg Peninsula)	Connan et al., 2007, 2009
	2010-2011 (coastal, estuarine, river, some SPM)	IRSN, 2011
	2012 (coastal, estuarine, river, some SPM)	IRSN, 2012
Biota UK	1990-2009 (Channel Islands, algae, molluscs)	Hughes et al., 2011
	1995-2012 (Channel Islands, UK coast)	CEFAS, 2013 (CD ROM version)
	No date (DNA damage to biota from tritiated water and organic tritium)	Jha et al., 2005; Jaeschke et al., 2011; Dallas et al., 2013
Biota France	2000-2001(North Cherbourg Peninsula, *F. serratus*)	Fiévet et al., 2006
	2010-2011 (coastal, estuarine, biota)	IRSN, 2011
	2012 (coastal, estuarine, biota)	IRSN, 2012
Microplastic Contaminants		
Aeolian load	no data	
Industrial load	1950s - 2011 (Production in Europe)	Wright et al., 2012
Water UK	No date (Sorption of PAHs and DDT by microplastics)	Bakir et al., 2012; 2014
Water France	no data	
Sediments UK	No date (Tamar Estuary)	Browne et al., 2010
	No date (UK coast)	Browne et al., 2011
	2012 (Tamar Estuary)	Sadri & Thompson, 2014
Sediments France	no data	
Biota	2010 & 2011 (demersal fish, *F. glacialis*)	OSPAR, 2010a; Lusher et al., 2013
	No date (Trophic transfer)	Farrell & Nelson, 2013
Nutrients		
Aoelian load	1995-2006 (Channel, model results)	OSPAR, 2009c
River load UK	1990-2011	OSPAR, 2001, 2002, 2004, 2005a,b, 2006a, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a

	2010 (RBD concentrations)	ETC/ICM, 2012
River load France	1990-2011	OSPAR, 2006a,b, 2007, 2008, 2009d, 2010c, 2011a, 2012a, 2013a
	2010 (RBD concentrations)	ETC/ICM, 2012
Water UK	1920s onwards (Western Channel, nutrients, plankton and others)	Southward et al., 2005; Johns, 2006; Smyth et al., 2010; Widdicombe et al., 2010
Water France	1992-2011 (Seine, Boulogne, nutrients and phytoplankton)	Lefebvre et al., 2011; Hernández-Fariñas et al., 2013
Sediment UK	no data	
Sediment France	no data	
Biota UK	no data	
Biota France	no data	
On-line data resources		
Aoelian load	Organic contaminants, metals, nutrients	http://ebas.nilu.no/
River / industrial load	Organic contaminants, metals, nutrients, radionuclides	http://www.ospar.org
Water, sediment, biota	Organic contaminants, metals, nutrients, radionuclides	http://dome.ices.dk (OSPAR directed marine monitoring data) https://www.bodc.ac.uk (MERMAN, the UK database for mandatory marine monitoring) http://wwz.ifremer.fr (Quadrige-2 database of French marine measurements) https://www.bodc.ac.uk (EDMED, the European Directory of Marine Environmental Data) http://www.emodnet-chemistry.eu (EMODnet, the European Marine Observation and Data Network) http://www.eea.europa.eu (WFD directed monitoring data for coastal and transitional waters)

[a] no data. Data not found or data component not addressed in this article