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Shellfish and Residual Chemical Contaminants: Hazards, Monitoring, and Health Risk Assessment Along French Coasts

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Abstract:

Shellfish farming is a common industry along European coasts. According to the 2005–2006 data from the French National Shellfish Farming Committee (CNC – *Comité National de la Conchyliculture* 2010; see Table 1 for a list of acronyms and abbreviations used in this chapter), Spain is the largest shellfish producer in Europe (270,000 t) and France ranks second, producing 200,000 t of shellfish annually. France is the leading European oyster producer, with an annual output of 130,000 t of *Crassostrea gigas,* and ranks fourth in the world after China, Japan, and Korea. The top three European mussel (*Mytilus edulis* and *Mytilus galloprovincialis*) producers are Spain (260,000 t), Denmark (80,000 t), and France (65,000 t). For other shellfish, the French annual output level is 15,000 t for king scallops (*Pecten maximus*) and a few thousand tons for *Ruditapes* clams (*Ruditapes decussatus* and *Ruditapes philippinarum*) and cockles (*Cerastoderma edule*). The economic impact of shellfish farming is considerable; despite fairly long production lead times and difficult operating conditions, shellfish farming generates annual sales of more than 650 million Euros in France, owing to its high added value.

1. Introduction

Shellfish farming is a common industry along European coasts. According to the 2005-2006 data from the French National Shellfish Farming Committee (CNC – *Comité National de la Conchyliculture* 2010;see Table 1 for a list acronyms and abbreviations used in this article), Spain is the largest shellfish producer in Europe (~270,000 t) and France ranks second, producing 200,000 t of shellfish annually. France is the leading European oyster producer, with an annual output of 130,000 t of *Crassostrea gigas,* and ranks fourth in the world after China, Japan and Korea. The top three European mussel (*Mytilus edulis* and *M. galloprovincialis*) producers are Spain (260,000 t), Denmark (80,000 t) and France (65,000 t). For other shellfish, the French annual output level is 15,000 t for king scallops (*Pecten maximus*) and a few thousand t for *Ruditapes* clams (*R. decussatus* and *R. philippinarum*), and cockles (*Cerastoderma edule*). The economic impact of shellfish farming is considerable; despite fairly long production lead times and difficult operating conditions, shellfish farming generates annual sales of more than 650 million Euros in France, owing to its high added value.

The main species of shellfish consumed in France are the Pacific oyster (*C. gigas*), mussels (*M. edulis* and *M. galloprovincialis*), king scallop (*P. maximus*), winkle (*Littorina littorea*), whelk (*Buccinum undatum*), cockle, *Ruditapes* clams and scallops (*Pecten spp*., *Chlamys spp.*).

Current European regulations focus on regulating microbiological agents, phycotoxins and some chemical contaminants. Since 2006, these regulations have been compiled under the name of the ‘Hygiene Package’. Because of increasing concern for the presence of contaminants in the marine environment, the French Food Safety Agency (AFSSA; now named the French Agency for Food, Environmental and Occupational Health & Safety, ANSES) issued a report in 2008 on the monitoring of chemicals in shellfish farming areas, and on health risks associated with shellfish consumption (AFSSA 2008b).

The purpose of this review is to address the residual chemical hazards that exist in shellfish that are routinely sampled from the natural marine environment and from the market place. We have included data on exposure levels and body burdens of many contaminants, and have related these data to human health risks. We have also addressed the concentration of contaminants found in the context of current regulatory and food safety standards. The data compiled here are designed to provide readers with a basis for assessing whether or not it is necessary to continue, or even extend environmental chemical contaminant monitoring to other chemicals that pose significant potential consumer health risks.

2. Regulation of Shellfish Food Safety in Europe

Food safety monitoring of shellfish farming areas falls under European regulatory jurisdiction and is defined in the “Hygiene Package”, which came into force on 1 January 2006. There are several specific sections of this regulation that apply to live bivalve molluscs. Two of these regulations (EC 2004a,b) are directed toward industry professionals (N°852/2004 and N°853/2004), and two others (EC 2004c; EC 2006b) apply to competent authorities having to do with official controls (N°854/2004 and N°882/2006). Directive (EEC) N°492/91 (EEC 1991), which had previously set the hygiene rules for the production and marketing of live bivalve molluscs, was repealed. A general presentation of these regulations is presented below, and deals only with the sections on residual chemical contaminants.

2.1. Provisions of the Hygiene Package

Regulation (EC) 852/2004 (EC 2004b) lays down general rules on food hygiene, and applies to primary production (farm and fishery products). It is complemented by Regulation (EC) 853/2004 (EC 2004a), which lays down additional specific hygiene rules for products of animal origin. Annex III, Section VII of Regulation (EC) 853/2004 specifies the requirements for live bivalve molluscs. Regulations (EC) 854/2004 and 882/2006 (EC 2004c ; EC 2006b) apply to official control bodies, and define a legal framework for setting the locations and boundaries of production, and relaying areas (depurating areas). The regulations also require food safety monitoring, by sampling, to screen for chemical and microbiological contaminants.

A clear distinction must be made between primary production of shellfish and the other operations that are required to bring shellfish to the market, because the regulatory obligations are different. Primary shellfish production concerns all operations carried out before shellfish reach an approved purification establishment: rearing, harvesting and transport of the produce. Annex I of Regulation (EC) 852/2004, and some provisions in Annex III, Section VII of Regulation (EC) 853/2004 apply to primary producers. Producers must be registered, but are under no obligation to set up Hazard Analysis Critical Control Point (HACCP) procedures. The activities of the purification and dispatch establishments (finishing, packing, etc.) are not regarded to constitute primary production. The provisions of Annex II of Regulation (EC) 852/2004 and of Annex III, Section VII of Regulation (EC) 853/2004 apply to these establishments. These establishments must be approved by the competent authority, and are under an obligation to introduce HACCP procedures.

The classification of production into Class A, B and C areas is based solely on measures having to do with microbiological contamination; these measures are defined by the Hygiene Package, and Regulations 853/2004 and 854/2004, in particular:

- Class A areas are those from which live bivalve molluscs may be harvested for direct human consumption;

- Class B areas are those from which live bivalve molluscs approach conformity, but before being marketed for human consumption require a short, but sufficient purifying treatment;

- Class C areas are those from which live bivalve molluscs can be harvested only after relaying (depurating) for a long period, with purification, or after intensive purification by an appropriate method.

At the EC level, the Hygiene Package regulates the monitoring of production areas during operations (854/2004, Annex II, Chapter II.b) for three types of hazards: microbiological, phytoplanktonic/phycotoxic and chemical. Thus, although under the Hygiene Package there is no obligation to test for chemical contaminants for the purposes of classifying the production areas; however, there is an obligation to chemically monitor these areas.

2.2. Provisions on Chemical Contaminants

To be regarded as edible bivalve molluscs must also comply with maximum levels of certain contaminants defined in Regulation (EC) 1881/2006 of 19 December 2006 (EC 2006c), which replaces Regulation (EC) 466/2001 (EC 2001), as amended by Regulation (EC) 629/2008 of 2 July 2008 (EC 2008a). These contaminant thresholds (Table 2) apply to the edible parts of bivalve molluscs, i.e., the whole flesh, except for the king scallop, for which the digestive gland is not taken into account (Article 1 of Regulation (EC) 1881/2006). Nonbivalve molluscs (gastropods), echinoderms and tunicates are not covered by the European regulations, but in France, in a recommendation issued on 31 October 2007 (AFSSA 2007b), AFSSA considers that the cadmium threshold set by decree on 21 May 1999 is appropriate: 2 mg kg^{-1} fresh mass for whelks (gastropod, *B. undulum*) (JORF 1999). For echinoderms and tunicates, given their particularly low levels of consumption, it is not regarded as necessary to set a regulatory threshold, but rather a guideline value of 2 mg kg^{-1} fresh mass (AFSSA 2007b).

3. Identifying Residual Chemical Hazards in the Marine Environment and in Shellfish

To identify the risks of chemical residues in the marine environment being transferred to bivalve molluscs, and thence to humans, it is necessary to target, among the many potentially toxic chemicals, those that have a likelihood of being released by human activities in the vicinity of shellfish farming areas. That does not mean that contamination of the environment and of the bivalve molluscs by the chemicals addressed in this paper has always been demonstrated. Hazard identification is usually conducted independently of the likelihood of an accident occurring. Consequently, hazard identification does not include chemicals that may be released into the environment from hitherto unidentified sources or following accidental spills, irresponsibility or acts of malice.

The main sources of contaminants are of human origin (Manta et al. 2002). They involve the following: terrestrial and marine crop and livestock farming; human habitation (energy production, building and demolition, wastewater, incineration of household waste, heating, etc.); land transport (infrastructures, vehicles); energy production; industry (solid waste, liquid effluents and gas emissions, end-of-life of products, etc.); maritime transport and related activities (port activities, dredging, etc.), as well as some leisure activities (golf courses, water sports, sailing, etc.). Pharmaceutical residues have been found in environmental waters and in the marine environment, so they also could qualify as pollutants (Walraven and Laane 2009; Fatta-Kassinos et al. 2011).

Crop and livestock farming activities results in the release of organic matter and nutrients (nitrates, phosphates, potassium) into the environment; these can contribute to the eutrophication of the marine environment and cause major changes to aquatic community dynamics. Many chemicals are, or have been used in farming: plant protection products, biocides, veterinary drugs (including antibiotics), any of which may contaminate the marine environment at some time (Schaffner et al. 2009). Human habitations can also be major sources of organic matter release into aquatic environments, particularly in coastal areas, via wastewater release (Heinzow et al. 2007; Schaffner et al. 2009). Incinerators and domestic heating equipment release persistent organic pollutants (POPs), such as dioxins, PCBs, PAHs, etc. (Lewtas 2007; Van Caneghem et al. 2010). Industrial activities also release a very wide range of toxic chemicals. Transport and energy production release such substances as PAHs, trace elements, radionuclides and many atmospheric pollutants (England et al. 2001). Through their toxic potential, these substances can cause direct adverse effects on the marine environment, on farmed molluscs and indirect effects on human consumers.

3.1. Inorganic Contaminants

Metals (trace elements) are naturally present in many rocks and minerals. Due to natural weathering of the earth's crust, they are found in all environmental compartments, including seawater. Some trace elements that are absorbed by living organisms accumulate in the food chain, and therefore present a risk to humans, who are the final consumers at the top of the food chain (Hamilton 2004 ; Hillwalker et al. 2006). Shellfish filter large amounts of water to extract their food, and are excellent bioaccumulators (Claisse 1989). Any contaminants in the water, from natural sources or pollution, are easily concentrated in shellfish flesh, particularly metals, such as the following: mercury, cadmium, lead, copper and zinc. Metals are mainly fixed in particular organs, such as the digestive gland (Soto et al. 1996), which plays a part in assimilation, excretion and detoxification (Johnson et al. 1996). These organs are generally the parts of the organisms that are eaten by humans (except for king scallops whose flesh is consumed only in France).

In Tables 3 and 4, we summarize the main metal contaminants found in the environment, their human-activity sources and we categorize their toxicity and risk levels. Levels of contamination in marketed shellfish are given by species for the three regulated metal contaminants (lead, cadmium and mercury); the results come from the CALIPSO (2005) and first Total Diet Study (EAT 2004) which were performed in France (Table 3). The levels reported in these tables can be compared with the maximum permitted levels set for fishery products. For example, cadmium levels are above the maximum permitted limits in some scallop species (1.14 mg kg^{-1} fresh wt), while the other bivalve molluscs show lower levels – no more than 0.040 mg kg^{-1} fresh wt. For lead and mercury, none of the species sampled were above the maximum permitted levels (lead < 0.26 mg kg^{-1} fresh wt, and mercury < 0.003 mg kg^{-1} fresh wt). The observed values in French shellfish farming areas (Fig. 1A, B, C and E) are very close to those observed in marketed shellfish just before consumption.

Table 3 also shows that molluscs have high concentrations of arsenic, the highest levels being found in whelks (15.8 mg kg^{-1} fresh wt). However, contamination levels in shellfish are lower than those in crustaceans, fish and other seafood; the highest levels were found in octopus (42 mg kg^{-1}fresh wt: Leblanc et al. 2006; Sirot et al. 2009). In 1988, the mean arsenic levels in bivalve molluscs (mussels and oysters) along the French coast ranged from 10 to 30 mg kg^{-1} (Michel 1993); arsenic residues were the most frequently encountered, irrespective of geographical area and species. It is difficult to link the highest levels with possible pollution sources. For example, organisms in the major estuaries (Seine, Loire and Gironde rivers) are less contaminated than those in adjacent coastal areas. It seems that the levels of arsenic in the environment derive less from bioaccumulation than from whether the metal is in organic or inorganic form (Michel 1993). In laboratory experiments, the oyster *C. virginica* bioaccumulates little inorganic arsenic and only a fraction of the organic arsenic present in the phytoplankton (Sanders et al. 1989). The arsenic fixed on inert particles of seston is poorly bioconcentrated in the oyster *C. gigas* (Ettajani et al. 1996), but the small amount that passes through the oyster causes intense erosion of the mitochondrial cristae, leading eventually to cellular respiratory failure. In the peppery furrow shell (or sand gaper) *Scrobicularia plana*, bioconcentrated arsenic levels match the levels of sediment contamination (Langston 1983). In the winkle, arsenic levels vary from 9 to 70 mg kg^{-1} dry wt, their exact level depending on the degree of contamination of their food sources (*Fucus spp.*), and the environment (Bryan et al. 1976, 1983). Among other unregulated metals, zinc and magnesium levels are higher in oysters than in mussels (Table 4).

Polonium (Po^{210}) is one of the radionuclides that may have a health impact (exposure threshold 2 millisieverts (mSv) yr^{-1}; Table 5). Exposure by ingestion is significant, and annual intake can reach hundreds of µSv per year in adults (Pradel et al. 2001).

3.2. Organic Contaminants

Bivalve molluscs are exposed to a multitude of persistent or non-persistent organic contaminants belonging to very different chemical families. Tables 6 and 7 give a summary description of the main data available in the literature on pollutants identified in water, sediments and in bivalve molluscs (Leblanc et al. 2006; OSPAR 2008) and include information on toxicity and risk category.

In regard to regulated organic contaminants (Table 6), PCBs and dioxins (PCDD/Fs) are found at levels far below the regulatory thresholds (8 pg g^{-1} of dl-PCBs + dioxins) in oysters (< 0.6 pg g^{-1}), mussels (< 0.6 pg g^{-1}) and king scallops (< 0.4 pg g^{-1}). The benzo[*a*]pyrene sanitary threshold is neither exceeded in marketed mussels (Table 6), nor in those that are farm-sourced (Fig. 1D and E). Some data on contamination of shellfish flesh are also available for unregulated organic contaminants (Table 7). Of about one hundred existing organostannic compounds, mono-, di- and tri-butyltin (MBT, DBT and TBT) and mono-, di- and tri-phenyl tin (MPT, DPT and TPT) are the most frequently found in fishery products. Octyltins are not detected in fishery products. Based on available data, results of two recent studies were that exposure to organotins through seafood does not seem to present a risk for the adult consumer (AFSSA 2006 ; Guérin et al. 2007). There are other relevant contaminating organic compounds, but very few data are available for them:

- synthetic musks, nitro-musks and polycyclic musks from the perfume industry;

- octylphenol ethoxylates (OPEs) and nonylphenol ethoxylates (NPEs), from industrial cleaning, maintenance of public places and processing of leather and textiles;

- hydrocarbons, particularly toluene, ethyl benzene, xylene (BTEX) and phenols, from the offshore oil industry via sludge and drill cuttings, process water and accidental spills or illegal discharges;

- substances on the list of 33 priority substances in Annex X of Directive (EC) N°105/2008 (EC 2008b), especially alachlorine, chloroalkanes, chlorfenvinphos, chlorpyrifos, di(2-ethylhexyl)phthalate (DEHP), diuron, endosulfan, hexachlorobutadiene, isoproturon, pentachlorobenzene, pentachlorophenol, trichlorobenzene and trifluralin;

- emerging contaminants including pharmaceuticals, hormones and endocrine disrupting compounds also present in aqueous environment (Richardson and Ternes 2005).

3.3. Accumulation of Contaminants in Molluscs and Factors of Variation

Shellfish are filter feeders that concentrate contaminants, and also have the ability to detoxify themselves. The balance between these two processes is not fixed but depends on many factors.

Contamination may be direct (from water), or via food ingestion. Food contamination in filter-feeding molluscs occurs via seston (suspended particulate matter, inert or living). As with inert particles, phytoplankton becomes contaminated by adsorbing chemical compounds onto their cell surfaces; sometimes, these chemicals are absorbed by diffusion into the cells. Food contamination (phytoplankton) generally leads to longer half-lives than does direct water contamination. The longer the duration of contact, the higher the level of contamination and the longer decontamination takes. The ration of organic to inorganic contaminants influences their distribution in organisms and their elimination rate.

3.3.1. Bioconcentration Factors (BCF)

The concentration factor (CF) or bioconcentration factor (BCF) is a concept that was introduced by Polikarpov (1960). It is based on a relatively simple concept that a relationship exists between the concentration of a substance in an organism and the concentration of the same substance in the surrounding water. However, CFs are not easy to estimate; to do so, the two concentrations must remain constant. It is difficult to experimentally maintain constant concentrations in water for long periods of time, and *in situ* water concentrations fluctuate widely. No method for standardizing the estimation of CFs has been proposed. Numerous studies have been carried out to address this problem (Chong and Wang 2001 ; James et al. 2006 ; Miramand et al. 1980 ; Murray et al. 1991 ; Pruell et al. 1986). CF data for various organic pollutants have been recorded by different agencies (e.g., the International Atomic Energy Agency (IAEA) and the Groupe Radioécologique Nord-Cotentin) and have been published (Amiard-Triquet and Amiard 1980). CF values vary widely among different animal types and the resultant bioaccumulation values are influenced by many abiotic and biotic factors.

The best estimations of CFs are those that are determined in experiments that are performed *in situ* over long periods of time. Since the Water Framework Directive (WFD), Directive (EC) N°60/2000 (EC 2000) has come into force, water authorities are obliged to assess concentrations of pollutants in total seawater, dissolved concentrations and amounts in particulates. However, hydrophobic pollutants are essentially adsorbed onto particulates and their concentration is dependent on the concentration of these particulates in water. Such particulate concentrations fluctuate widely in space and time, so direct measurements in water were abandoned more than 20 years ago, under the French National Monitoring Network (RNO – *Réseau national d'observation*) and the OSPAR convention. The French Research Institute for Exploitation of the Sea (IFREMER – *Institut français de recherche pour l'exploitation de la mer)* considers that, at least for non-hydrophilic substances, the most effective monitoring target for contaminants are media that concentrate these substances: sediments and/or biota and particularly mussels and oysters, the two usual sentinel species. However, to meet the requirements of the WFD, the levels measured in these media must be converted into water concentrations. The tissue concentration in the molluscs is equal to the concentration in the water multiplied by the BCF. It is therefore possible to calculate the water concentrations, if the CF is known. James et al. (2006) provided BCFs for most substances that the EU considers to be priority ones (Table 8).

3.3.2. Seasonal Fluctuations in Contaminant Concentrations

Concentrations of chemical contaminants in bivalve molluscs fluctuate according to the time of year. This was noticed from the start of the RNO monitoring program in the early 1980s (Claisse 1992). The pattern for inorganic compounds is 'biological dilution', when bivalves reach sexual maturity; this occurs when the amount of contaminants remain the same, but the organism's body mass increases, and thus metal concentrations fall. This has been observed for cadmium, copper, lead and zinc in mussels (Amiard et al. 1986) and oysters (Amiard and Berthet 1996). The highest concentrations are recorded in winter and spring and the lowest in summer and autumn, with ratios of up to 1:4 depending on the contaminant and the species (Devier et al. 2005). The reverse pattern is found with lipophilic organic compounds, such as DDT in the oyster *C. virginica*; concentrations increase at sexual maturity, when oysters produce lipid-rich gametes (Butler 1973). Oysters also eliminate these pollutants through spawning (release of eggs into the water). With *C. virginica*, the risk to humans is therefore greatest at the moment of sexual maturity.

Because contaminants are only monitored annually, and because of the kinetic behavior of contaminants in molluscs, tracing individual contamination events over short periods of time is not possible. Therefore, the established programs are effective for monitoring chronic contamination, but not for short duration events; such events may thus go unnoticed between any two samplings of the sentinel species. However, alarms may be sounded from accidental discharges as a result of triggering increased mortality at sensitive developmental stages.

3.3.3. Detoxification Mechanisms

Detoxification of Trace Elements

Invertebrates exposed to toxic trace elements respond with two types of detoxification mechanisms (Amiard 1991). The first response is to render the metal insoluble by immobilizing it in the form of a salt. This occurs with silver sulphide in oysters, for example (Martoja et al. 1988). The second response is to induce metallothioneins (MTs), which are capable of detoxifying various trace elements (Amiard et al. 2006). MTs form complexes with the trace elements and render them harmless. Metallothioneins are stored in lysosomes and their concentration is proportional to that of toxic trace elements in the environment, as shown by an experiment with transplanted mussels in the western Mediterranean Sea (Mourgaud et al. 2002). Detoxification mechanisms in invertebrates vary widely from one species to another. In various oyster species, mobile cells called amoebocytes accumulate complexed metal from the blood. In *Ostrea edulis*, some amoebocytes accumulate copper, others zinc, or copper and zinc simultaneously. Other oyster species, such as *O. angasi* and *C. gigas*, only have one amoebocyte type, which accumulates copper and zinc equally well (George et al. 1984). Some species of molluscs (e.g., oysters and mussels) are capable of regulating the internal concentration (homeostasis) (within certain concentration limits) of certain essential trace elements, such as copper and zinc (Amiard et al. 1987).

The particular physical-chemical form of inorganic contaminants that are stored have consequences for the subsequent transfer of trace elements within trophic networks. The two above-mentioned detoxification processes (insolubilization and metallothionein induction) are very efficient, and species that use them can live in heavily contaminated environments. Such species may accumulate high levels of contaminants in some of their tissues. When these species are consumed, the metal-metallothionein complexes are ingested and digested, releasing the metals into the consumer's body in a manner that favors the assimilation of the metals. Therefore, the levels transferred to and absorbed by the consumer may be high. In contrast, when detoxification occurs by insolubilization, the resultant granules are poorly digested by the consumer or predator; hence, bioavailability is low.

Detoxification of Organic Pollutants

Some invertebrates are able to biotransform organic pollutants in special organs (e.g., the digestive gland) that render pollutants hydrosoluble, and therefore more easily eliminated (Narbonne and Michel 1997). This metabolic process occurs in two biotransformation stages: (1) phase I, oxidation and/or (2) phase II, conjugation. Phase I is controlled by P450 cytochromes or by flavin monooxygenases. In phase II, conjugation frequently takes place with glutathione and is catalyzed by glutathione S-transferase (GST). Occasionally, biotransformation activates a metabolite to a form that is more toxic than the parent molecule. A third detoxification pathway is possible, and involves the glycoprotein Pg170 (phase III). In phase III, organic pollutants are expelled from the cell. This protective elimination mechanism is efficient in molluscs (Bard 2000), and is known as multixenobiotic resistance (Pain and Parant 2003).

The Effect of Shellfish Purification on Chemical Contaminants

In the course of shellfish production, shellfish are purified to reduce the risk of microbiological contamination. The question is whether this microbiological purification helps reduce the amounts of any chemical contaminants also present in the shellfish.

Microbiological purification consists of immersing live shellfish in tanks continuously fed clean seawater for a period that is sufficient to eliminate microbiological contaminants and render the shellfish suitable for human consumption. The regulatory definition of "clean seawater" is found in point *h* of Article 2 of Regulation (EC) 852/2004 (EC 2004b). This very vague definition sets goals, without clearly defining the criteria to be fulfilled. The French Directorate for Food (DGAL), therefore, commissioned AFSSA to establish seawater quality criteria suitable for handling fishery products. AFSSA delivered its opinion on 26 July 2007 (AFSSA 2007a). Microbiological purification is required only for shellfish from Class B and C production areas, and the produce from these areas can be harvested, but cannot be directly marketed. The time required for purification varies between two and several days, depending on the system used. In France, the duration for purification is 48 hours for Class B shellfish (industry recommendation). The duration of purification may be reduced for some fragile shellfish species (e.g., wedge shells and *Ruditapes* clams); the regulations do not impose a minimum duration.

When kept in large quantities of clean seawater, contaminated marine organisms purify themselves, eliminating the chemical contaminants that they have accumulated in their soft tissues. The measure used to track elimination rate is biological half-life, i.e., the time required for half the amount of a substance to disappear from the organism or organ.

The kinetics of decontamination depend not only on the difference in initial concentration, but also on the following factors (Casas and Bacher 2006) :

- chemical-specific factors- (type(s) of the contaminant(s), level(s) of contamination, variations in contamination over time, contamination pathways (i.e., water, food or inert particles));

- physiological factors of the organism (growth rate, mass variation over time, type of sexual state maturity, physiological status, differences between species, etc.);

- environmental factors (temperature, and food quantity and quality).

From the foregoing, it is obvious that the elimination kinetics, the mechanisms of elimination, and quantities of toxicants eliminated will be species-dependent. Mussels are capable of eliminating cellular organelles (lysosomes) that were involved in detoxifying various contaminants, whereas oysters retain their lysosomes for life (George et al. 1978). In some species, certain cumulative toxins continue to be accumulated throughout an animals' lifetime.

In Table 9 we provide examples of the chemical half-lives of several contaminants in bivalve molluscs. Although this table is far from exhaustive, it indicates the wide variations in half-life elimination times for various contaminants and species.

The above information disclosed on elimination half lives of various chemicals indicates that the 48 hour immersion time, used to purify microbes from Class B shellfish, is far from sufficient to also remove chemical contaminants (organic and metal). In fact, considerably more research results are needed to achieve reliable estimates of the half-lives in shellfish species of the main contaminants found in the marine environment. These data would be extremely useful in estimating the dissipation times, and therefore the seriousness of accidental chemical pollution or spills. Of course, the key question after such events occur is how soon, and under what conditions marketing of exposed shellfish can be resumed. Despite the usefulness of such information, the current regulations do not require that the disposition of chemical contaminates be considered.

4. Chemical Monitoring in the Environment and in Shellfish

4.1. Environmental Chemical Monitoring Programs

Shellfish are at risk of from pollutants primarily because of their environmental exposure. To protect shellfish from chemical contamination, systems have been set established to periodically monitor waters of coastal areas for selected contaminants (Apeti et al. 2010 ; Cantillo 1998 ; Claisse 1989 ; Franco et al. 2002 ; O'Connor 1998). The goal of the OSPAR convention for the protection of the northeast Atlantic marine environment is to reduce pollution. The OSPAR Hazardous Substances Committee listed the substances to be monitored in order of priority, taking into account those that are already prioritized by other regulations, e.g., under the WFD. Under the Mediterranean Action Plan (MAP) of the United Nations Environment Programme (UNEP), the Barcelona Convention for the Protection of the Mediterranean Sea Against Pollution (MED POL) has implemented phase III of the MED POL monitoring program.

European Directive (EC) N°105/2008 (EC 2008b), which amends Directive (EC) N°60/2000 (EC 2000), and lays down the environmental quality standards for water, provides for updating the list of priority substances. The updates give the maximum allowable concentration of each substance (set up to avoid serious and irreversible consequences of acute short-term exposure for an ecosystem), as well as the allowable mean annual concentration (to avoid long-term irreversible consequences).

In France, the monitoring of water contamination along the French coast has been performed by the RNO, renamed ROCCH (*Réseau d'Observation de la Contamination CHimique du milieu marin*), in 2008. ROCCH was established by the French Ministry of the Environment in 1974, and is coordinated by IFREMER. Its purpose is to assess levels and trends in chemical contamination along the coast. Until 2007, the RNO monitored only sediments and bivalve molluscs, in which contaminants are concentrated, to meet French obligations under the OSPAR and Barcelona conventions. In addition to sediments and bivalves, ROCCH also monitors the biological effects of contamination by organic forms of tin (which cause imposex; Huet et al 2003).

4.1.1. Monitoring Contaminants: the RNO Program and its Successor (ROCCH)

Because of the difficulty in obtaining valid samples suitable for water trace analysis, and the low spatial and temporal representativeness of such samples, RNO monitoring has focused on the matrices that absorb contaminants, i.e., biota and sediments. Therefore, bivalve molluscs (mussels and oysters) are used as quantitative contamination indicators (Claisse 1999).

The concepts of indicator- and sentinel-species are widely used in many countries, e.g., Mussel Watch in the USA (Cantillo 1998 ; Claisse 1989 ; Goldberg et al. 1983 ; O'Connor 1998 ; Sukasem and Tabucanon 1993 ; Tripp et al. 1992).

In France, testing for chemical contaminants was performed annually in November for all substances, and biannually (February and November) for trace elements (Table 10). The interpretation of the analytical results requires consideration of the differences among species in bioaccumulation; for example, the concentration ratios between oysters and mussels are approximately 50 for silver, 2.5 for cadmium, 10 for copper and 15 for zinc (Claisse et al. 2006).

The RNO results have also sometimes been used for monitoring food safety, together with results from official regulatory controls.

The main achievements of the RNO from 1979 to 2007 included the following:

- establishment of national baseline levels for nine trace elements, 14 organochlorine chemicals and 37 PAHs (Table 10);

- identification of reference or control sites for monitoring if

    - natural contaminants are present at representative levels, or
    - synthetic chemicals exist at levels that do not reflect significant inputs, and
    - hotspots exist (particularly contaminated areas; e.g., the Gironde is a hotspot for cadmium, and the Seine for PCBs);

- determination of temporal trends for 33 contaminants;

- assembling a bank of stabilized mollusc samples beginning in 1981;

- organization and management of national and international collaborations through European conventions and international programs previously cited at the beginning of paragraph 4.1; and

- implementation of data quality management, which is a driver for achieving the "state of the art" in marine environmental chemical analyses.

Although the RNO was designed for environmental monitoring purposes, it has also performed annual monitoring for food safety purposes to classify the shellfish-farming areas, and has conducted discrete site-specific studies.

In 2008, IFREMER established ROCCH (formerly RNO) for the French Ministry of the Environment, although ROCCH is partly financed by water authorities. The main purpose of ROCCH is to address the chemical monitoring needs of the WFD, and the OSPAR and Barcelona international conventions. ROCCH, contrary to RNO, performs chemical monitoring of WFD substances directly in the water, but, to the detriment of monitoring shellfish. In particular, the February surveys of shellfish have been discontinued. However as an annual peak in shellfish contamination was regularly observed, this change may be prejudicial for food safety monitoring, so, since 2008, DGAL has financed a February monitoring survey. The number of sampling points has been increased by 60% for this February survey to improve coverage of the shellfish farming areas. Similarly, the number of taxa monitored has been increased to also address farmed species. Analytical results of the monitoring are published no more than three months after the sampling, compared to ten months post-monitoring under the RNO system.

Up to the present, food safety monitoring has only applied to three trace elements. However, starting in 2011, DGAL and IFREMER will initiate monitoring for dioxins, dl-PCBs and benzo[a]pyrene, to comply with Regulation (EC) 1881/2006 (EC 2006c), and to follow the recommendations published in AFSSA's opinion of 21 March 2008 (AFSSA 2008a).

The monitoring work undertaken by RNO and ROCCH are described in Table 10, in the context of the various conventions and directives.

4.1.2. Examples of Contaminant Testing

In this section, a coastal lagoon (Arcachon Bay, *Bassin d'Arcachon* in French) and an estuary (Bay of Seine) have been taken as examples:

Bassin d'Arcachon

The mean concentrations of lead, cadmium, mercury and other contaminants detected in Bassin d'Arcachon are shown in Table 11. The mean concentrations recorded by the RNO in oysters from Bassin d'Arcachon are 0.18 ± 0.04 mg kg^{-1} fresh wt for lead, 0.23 ± 0.09 mg kg^{-1} for cadmium and 0.03 ± 0.01 mg kg^{-1} for mercury. These figures are well below the regulatory limits (Table 2). High concentrations of copper are found in oysters (24.51 ± 9.69 mg kg^{-1} fresh wt flesh). The concentrations have risen over the past 20 years, probably because copper has replaced the TBTs in anti-fouling paints (Claisse and Alzieu 1993).

In regard to the TBTs, mussels transplanted to oyster farms have revealed concentrations of approximately 30 µg kg^{-1} dry wt, and showed increases in July and August (Devier et al. 2005). No trace of TBTs has been detected in the water. However, in mussels transplanted to harbor areas, concentrations of 800 to 2400 µg kg^{-1} dry wt have been recorded, with peaks occurring between April and September. Devier et al. (2005) attribute this increase to spring and summer nautical activities. TBT concentrations measured in the surface waters of Arcachon harbor range between 2 and 7 ng L^{-1} (samples taken from May to August); the corresponding BCF values range from 2.8×10^5 to over 1.3×10^6. These are the highest BCF values recorded in the literature for mussels (*Mytilus sp.*). TBT levels of 400 µg Sn kg^{-1} dry wt, measured in sediments, are responsible for the high contamination levels found in mussels, and result from sediment resuspension (Devier et al. 2005). The observed speed of TBT bioaccumulation is high, and is consistent with data in the literature (stabilization after 25 days). Devier et al. (2005) concluded that Arcachon harbor is severely contaminated by organotins, because of their persistence in sediments from use as an anti-fouling treatment for boats; the organotins continue as significant contaminants several years after their use has been banned. The concentrations recorded in mussels transplanted to the harbor highlight the role this hotspot plays in local contamination, and the hazard it represents for the entire Arcachon Bay. These data confirm the work of Auby and Maurer (2004), who revealed TBT levels (between 1997 and 2003) in Arcachon Bay waters near the harbor service station that ranged from 5.7 to 21.9 ng L^{-1}. The toxic effects on plankton and molluscs associated with these TBT concentrations in seawater have been recorded by Alzieu et al. (1991) and Michel and Averty (1999). They reported that even for a TBT concentration in seawater of less than 1 ng L^{-1}, the females of some gastropods may develop male sexual characteristics (imposex). At concentrations exceeding 1 ng L^{-1}, diatom growth and zooplankton reproduction are restricted; above 2 ng L^{-1}, oyster shells show calcification anomalies, and above 20 ng L^{-1}, reproductive anomalies are observed in bivalves.

High levels of PAHs were measured in mussels transplanted in Arcachon harbor, with peaks occurring in May-June and August (the annual means at this site range from 1.45 $X10^6$ to

1.62 $X10^6$ pg g^{-1} dry wt, depending on the specific PAH, with a maximum of 2.7 $X10^6$ pg g^{-1} dry wt) (Devier et al. 2005).

Regarding indicator PCBs (sum of PCB 28, 52, 101, 118, 138, 153 and 180), the levels measured in mussels are low (annual means of 5.4 and $7X10^3$ pg g^{-1} wet wt). Concentrations in oysters are similar, with 5.2 ± $3.6X10^3$ pg g^{-1} wet wt.

Twenty-one pesticidal and biocidal active substances have been detected in the waters of the Arcachon Bay during the summertime from 1999 to 2003, at concentrations ranging from a few ng L^{-1} to several hundred ng L^{-1}. Most of these substances are herbicides, including some that are now banned (Auby and Maurer 2004). According to Auby and Maurer (2004), the presence of these substances may impact the development of the small phytoplankton on which oyster larvae feed, but probably do not affect oyster larval development.

The studies of Auby and Maurer (2004), and Devier et al. (2005) thus emphasize the need to monitor TBT and PAH contamination levels in shellfish farming areas of Arcachon Bay. Doing so will ensure that TBT and PAH pollution does not migrate from the harbor to the oyster and mussel farming areas.

The need to monitor TBT and PAH contamination levels in shellfish farming areas, as observed at Arcachon Bay, can be extended for the entire French coast, since organostannic and PAH compounds are present in similarly semi-enclosed waters elsewhere along the coast. The highest concentrations of TBTs and their degradation products are recorded in harbor areas, e.g., Brest (1.5 mg kg^{-1} of TBT) and Lorient (0.44 mg kg^{-1}) on the Atlantic coast, and Gulf of Fos (1.1 mg kg^{-1}), Toulon (4.1 mg kg^{-1}) and Gulf of Saint-Tropez (1.55 mg kg^{-1}) (Averty et al. 2005) on the Mediterranean coast. Relatively high levels of TBT and PAH are also found in other coastal areas such as the Seine estuary, the Basque coast and Thau Lagoon.

Bay of Seine

A study of metal contamination of the main marketed species in Bay of Seine was conducted in 2000. The aim was to assess levels of contamination by lead, mercury, cadmium, chromium and silver in five commercial species of interest: whelk, king scallop, plaice/sole, cod and rock salmon. The study (Chiffoleau et al. 2002) shows that whelks were heavily contaminated with cadmium — above the French regulatory limit, in very large specimens (over 70 mm). Based on this finding, a local decree was issued in July 2002, classifying whelks of over 70 mm as ‘Class D’ (French classification grade designating that harvest is prohibited) and whelks of less than 70 mm as "provisional Class A", throughout Bay of Seine and the coasts of Seine Maritime district. In 2002 and 2003, whelk sampling was intensified, particularly for small specimens, to determine the size, on average, above which the 2 mg Cd kg^{-1} wet wt threshold (French decree of 21 May 1999) would be exceeded.

The mean concentrations of cadmium, mercury, lead and benzo[*a*]pyrene in mussels are given in Table 12. The concentrations of these four contaminants are below the regulatory limits (Table 2).

4.1.3. Active Environmental Biomonitoring: a Promising Procedure for the Future

Researchers have been conducting active biomonitoring using various shellfish species for several years. For example, the study of Devier et al. (2005) used transplantation experiments. Active biomonitoring has a number of advantages over conventional monitoring (Andral et al. 2004). The transplanted shellfish have a known history, their exposure time is controlled, the siting of the station is chosen independently of bathymetry, and each specimen's position in the water column is controlled. Measurements are optimized, because samples are more homogeneous owing to the selection of specimens for the experiment (parental origin, size, age, healthy site of origin, etc.). There are some constraints, such as complicated logistics, data interpretation that depends on the trophic and physico-chemical variability of the destination site; additional biometric parameters must therefore be measured. The abundant literature in this field (Berthet et al. 2008; De Kock and Kramer 1994; Mourgaud et al. 2002) provides transplantation protocols that include the time required to establish equilibrium with the new environment, the initial stress and the trophic factors of the destination site.

Transplantation is a promising procedure for the future because of numerous benefits already cited, nevertheless, one aspect thus far neglected is the possibility of theft by ill-intentioned people.

4.2. Chemical Monitoring for Marketed Shellfish

Those who produce and/or market bivalve molluscs are subject to self-inspection and mandatory product traceability to provide information on quality, including analysis of chemical contaminants and shellfish mortality. For marketed shellfish, the public health authorities responsible for official controls must follow the provisions of the Annex II of Regulation (EC) 854/2004 (EC 2004c). The French Directorate General for Food (DGAL-*direction générale de l'alimentation*) is in charge of these controls and has drawn up annual monitoring programs since 1998 to assess the contamination levels of marketed shellfish.

4.2.1. Self-inspection

Self-inspection is a key tool for shellfish operators to optimize their effectiveness in meeting the requirements of the Hygiene Package. In addition, self inspection during production, transportation, purification, maturing and finishing also ensures the food safety of shellfish when they reach the consumer. Self-inspection is carried out for microbiological and chemical contaminants, both in the water and in the shellfish. Sampling is performed by third-party professionals who send their samples to a laboratory of their choice.

4.2.2. Monitoring and Management of Shellfish Mortality

Operators must report each event of mortality that exceeds 20% of individuals, within a 15 day period to the responsible authority. IFREMER then conducts a survey to determine the cause of the mortality, and whether it has an environmental, microbiological (often involving *Vibrio*, viruses, fungi or parasites) or a zootechnical origin. For animal health reasons, IFREMER produces periodic reports on national and regional oyster mortality, through the Mollusc Pathology Network (REPAMO — *Réseau de PAthologie des MOllusques*), and other organizations. Mortality occurs in patches within an area and generally affects only one species. It is thought to be multifactorial (Oyster Summer Mortality programme (i.e., MOREST *Mortalité ESTivale d'huîtres* and REPAMO), and involve oyster physiology, environmental factors (it does not occur below a temperature of 19°C) and/or aggravating factors (viruses, bacteria) (Samain and McCombie 2008). According to Gagnaire et al. (2006), pesticides may be among the triggering factors.

The epidemiological aspect of these die-offs and the zootechnical and environmental context provide guidelines for diagnosis. For example, if several species are affected simultaneously, an environmental or toxic origin will be strongly suspected. Blooms of *Gymnodinium spp.*, transfer stress and anoxia are known to cause die-offs. However, it is difficult to precisely identify causes, because operators sometimes take their samples at intervals of two weeks or more, (e.g., where concessions are accessible only during low spring tides). These mortality events also require dealing with decomposing shellfish, which can affect the microbiological quality of the water in a confined environment. Summer mortality of Pacific oysters (*C. gigas*) on the French coast is regularly reported, but has not endangered this species, which was considered to be invasive up until three years ago. Recurrent seasonal mortality has also been reported in *Ruditapes* clams and cockles, but not at the same time of year (in spring for *Ruditapes* clams, after stormy episodes for cockles). In 2008 and 2009, there was high mortality among Pacific oysters in France. Laboratory experiments have shown that certain pollutants can affect the genetic, immunity and trophic characteristics of oysters; in 2009, the combined presence of the OsHV-1 virus and the bacterium *Vibrio splendidus* seems to have played a major part in the mortality incident (Sauvage et al. 2009)

No oyster pathogen is known to also be pathogenic for humans. In some cases of abnormal mortality in marine species (e.g., several species suddenly, simultaneously and massively affected), a more thorough toxicological investigation may be undertaken to test for pesticides or biocides.

4.2.3. Monitoring Program for Chemical Contaminants in Marketed Shellfish

Two offices of the DGAL are involved in monitoring chemicals in shellfish: (1) the Office for the Quality and Safety of Food Products from Fresh and Marine Waters (BQSPMED – *Bureau de la Qualité Sanitaire des Produits de la Mer et d'Eau Douce*), responsible for monitoring chemical contaminants in bivalve molluscs and (2) the Office of Food and Biotechnology Regulations (BRAB – *Bureau de la Réglementation Alimentaire et des Biotechnologies*), responsible for the EU dioxin monitoring program. The International Health and Safety Coordination Mission ((MCSI – *Mission de Coordination Sanitaire Internationale*, part of DGAL) is also involved by sampling imports. The screened chemical contaminants are trace elements (lead, cadmium, mercury), indicator PCBs (7 congeners: 28, 52, 101, 118, 138, 153 and 180) and PAHs (15 since 2006). In earlier monitoring and control programs (1998-2002), pesticides and antibiotics (EC 2000) were tested. The number of bivalve mollusc samples to be tested each year, under the chemical contaminants monitoring program, is 400 altogether (all species and all chemical contaminants); this number includes farmed shellfish (oyster, mussel, cockle and *Ruditapes* clams) and wild populations of pectinids fished in French waters.

The local veterinary authorities (DDSV – *Direction Départementale des Services Vétérinaires*) communicates confirmed positive results to DGAL without delay to the local Maritime Affairs authorities (DDAM – *Direction Départementale des Affaires Maritimes*) and to IFREMER. An investigation is then carried out to identify the contamination source and any corrective measures that are required.

For cadmium, this process has brought several non-compliances to light. In Table 13, the first screening gives some non-compliance data for cadmium which were refined by a second analysis using a more sophisticated method, and by taking into account deviations from the standard. In view of the results from 2005, DGAL conducted a control study that had five samplings in March 2006. The goal was to check the level of contamination in problem areas and in the smaller neighbouring area of Pertuis d'Antioche (French Atlantic coast). These samplings also resulted in four non-compliant results for cadmium, and they were confirmed by AFSSA/LERQAP (two from the Pertuis Breton (French Atlantic coast) and two from

Arcachon Bay). Because of these results, imposition of possible management measures are under examination in collaboration between the French Directorate General for Health (DGS – *Direction Générale de la Santé*) and the Directorate for Marine Fisheries, and Aquaculture (DPMA – *Direction des Pêches Maritimes et de l'Aquaculture*).

With respect to the specific DGAL monitoring programs conducted in 2009, the presence of lead (Pb), cadmium (Cd) and mercury (Hg) concentrations in the white and dark meats of 108 batches of crustaceans (lobsters, spider crabs, common crabs, swimming crabs and king crabs) were found. These organisms, under investigation by the French National Reference Laboratory (NRL), were collected in France between April and December, 2009, and in marine gastropods (common winkles, common whelk, abalone and murex), echinoderms (purple sea urchin and black sea cucumber) and tunicates (ascidians) (Noël et al. 2011, *in press*). The results show mean concentrations for crustacean white meat of 0.041, 0.132 and 0.128 mg kg^{-1} for Pb, Cd and Hg, respectively. These values were always lower than the European legislation maximum level of 0.50 mg kg^{-1} Cd. The concentration in the dark meat of common crabs (mean concentration: 11.8 mg kg^{-1} and maximum of 14.3 mg kg^{-1}) is well above the observed levels for white meat. The results for gastropods, echinoderms and tunicates show that the highest levels of Hg and Cd were found in murex, 0.185 mg kg^{-1} and 0.853 mg kg^{-1}, respectively; whereas the highest level of Pb was detected in ascidians (0.505 mg kg^{-1}). Hg and Pb concentrations were systematically below the maximum regulatory levels (0.5 mg Hg kg^{-1} and 1.5 mg Pb kg^{-1} wet wt). For Cd, only two samples of murex (2.09 ± 0.42 mg kg^{-1} and 2.33 ± 0.46 mg kg^{-1}) exceeded the French maximum level of 2.0 mg kg^{-1} wet wt.

Other of the data on contaminants contained in marketed shellfish are presented in Tables 3 and 6.

4.2.4. European Data on Chemical Contamination of Shellfish

There is no specific EU reference laboratory (EU-RL) for monitoring chemical contaminants in shellfish. However, there are four EU-RLs that test for lead, cadmium, mercury, PAHs, dioxins and PCBs in animal tissues as indicated in Regulation (EC) N°776/2006 (EC 2006a).

Chemical contamination levels in shellfish are monitored in many national and international surveys. However, many of these are not published. It would be useful for many researchers and governmental personnel to bring these scientific data together in a single national or European database that could be accessed through the internet.

5. Impact on Humans

Health risks associated with chemical contaminants are difficult to assess, owing to the fact that many produce only long-term action (chronic risk), and such contaminants reach humans through so many different sources (food, water, air, occupational, etc.). To assess the health impact of contaminated shellfish consumed by humans, exposure has to be estimated from contamination levels and consumption data.

5.1. Consumption Data for the General Population (INCA 2 2009)

Data on food consumption for the general population (including consumers and non-consumers of shellfish) may be taken from the INCA 2 (2009) survey (*Enquête Individuelle et Nationale sur la Consommation Alimentaire*), conducted in 2005-2007 by the Food Consumption and Nutritional Epidemiology Unit (OCA-EN) at AFSSA. In this survey, all types of food intake was recorded by respondents over a period of one full week. To account for seasonal effects, the survey was carried out in four phases spread over a period of one year. Food consumption data were obtained from consumption diaries that respondents kept over the targeted seven-consecutive-day period; in their diaries respondents identified the foodstuffs and portions that were shown in a booklet of photographs (Suvimax 2002). The survey included 4000 adults and children that were representative of the French population. To ensure that the sample was nationally representative, it was stratified by region of residence and town size, and a quota method was used for age, sex, occupation, socio-occupational category and size of household. The adult sample included 2624 individuals aged 18 and over. A special method was used to exclude bias resulting from underestimation of food consumption by some respondents; those for whom the ratio between calories consumed and the basal metabolism, calculated using the Schofield method, was below a certain threshold, were excluded from the calculations (706 were excluded). The collection of "normal" adults thus included 1918 individuals. The sample of children included 1455 individuals aged 3 to 17. This sample was not adjusted, because there was no formula for identifying low-food-consuming subjects among children. In this survey, only the edible parts of foodstuffs were used to establish quantities consumed. The food groups counted as 'solid foods' included all food groups in the INCA 2 (2009) nomenclature except for milk, water, soft drinks, alcoholic drinks, hot drinks and soups.

At the most detailed level of the INCA 2 (2009) nomenclature, the reliability of the data for foods such as molluscs is not certain, because consumption was recorded for only one week. Amounts consumed were very low (Table 14). For comparison, Table 14 gives data on the percent consumption of meat and fish by INCA 2 (2009) survey respondents. Mean daily consumption of shellfish, in the general population, was estimated to be 4.5 g in adults; this value varied widely by region and season of the year. Using this consumption level, Shellfish consumption represented 0.16% of overall solid food intake. However, the INCA 2 survey (2009) was not well-suited to estimating shellfish consumption, because it included only a small number of shellfish consumers.

In conclusion, consumption of bivalve molluscs in France contributes little to the general population's overall food intake. Notwithstanding this conclusion, for the sake of regular shellfish consumers, continuing vigilance is necessary.

5.2. Consumption Data for High Consumers of Seafood (CALIPSO)

After the first INCA study (INCA1 1999), a specific work, called CALIPSO, was devoted to high consumers, i.e., adults who eat fish or seafood products at least twice a week (Leblanc et al. 2006). In Table 15 we present the data on mollusc consumption among high consumers of seafood products that were included in the CALIPSO survey (n=1011 adults, including 246 men aged 18 to 64, 641 women aged 18 to 64, and 124 persons aged 65 and over). The results are given as means across the four sites studied, without distinction for age or gender (Leblanc et al. 2006).

Consumed bivalve species included cockle, mussel, king scallop, queen scallop, other scallops, razor clam, *Ruditapes* clams, other clams, oysters, warty venus and wedge shell. Consumed gastropods included winkles, whelks, abalones and limpets. The only echinoderm eaten in France is the sea urchin and the only tunicate eaten is the sea squirt.

In CALIPSO, mean consumption of bivalve molluscs among adults is estimated at 153 g per week (8 kg yr^{-1}). The highest mean consumption is for king scallops (39 g $week^{-1}$), followed by oysters (34 g $week^{-1}$) and mussels (22 g $week^{-1}$).

Overall, these high consumers of seafood products eat, on average, twice the quantity of bivalve molluscs as do the shellfish consumers in the general population (INCA 2 2009); this is about the same level as the mean consumption of fish in the general population.

5.3. Exposure to Contaminants via Shellfish Consumption

Cadmium

In France, CALIPSO data show that the mean cadmium intake from shellfish is 1.26 µg $week^{-1}$ in adults (Leblanc et al. 2005, 2006), which is about half of the new tolerable weekly intake (TWI) value that was recently revised by the European Food Safety Authority (EFSA); this value was revised from 7 to 2.5 µg kg^{-1} bwt per week (EFSA 2009). A recent PTMI (provisional tolerable weekly intake) value was given by JECFA (25 µg kg^{-1} body wt mon^{-1}); this value corresponds closely to a PTWI value of 5.3 µg kg^{-1} bwt per week. Shellfish consumed by adult men, who are high seafood consumers, leads to a cadmium intake of more than twice that of the average total intake from food, in non-smoking adult men (EAT total diet survey by AFSSA). The cadmium intake varies from 8.2, 10 and 23 % of the PTWI, depending on the threshold that is selected (Table 3). The contribution of shellfish differs between regions. The shellfish that contribute most to cadmium intake by humans (CALIPSO survey) are king scallops (14% in Le Havre and 20% in Toulon), whelks (21%), scallops (19%) and oysters (11% in La Rochelle) (Leblanc et al. 2006).

Lead

In France, the mean intake of lead has fallen considerably in recent years. In 2005, the EAT survey indicated an average intake from food of 18 µg day^{-1} per adult, which amounts to 7% of the PTWI value set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1986 (Leblanc et al. 2005). The mean lead intake from shellfish (CALIPSO survey) is 0.26 µg kg bwt^{-1} day^{-1} in adults (from Leblanc et al. 2005, 2006). However, in June 2010, the JECFA concluded that the PTWI could no longer be considered health protective and withdrew it (JECFA 2010b). EFSA came to the same conclusion in its opinion of March 2010 (EFSA 2010). Consumption of seafood (fresh fish, crustaceans and molluscs) accounts for 3% to 11% of lead intake from total food. Shellfish contribute 0.7 µg day^{-1} of that intake. According to the CALIPSO survey, the main shellfish concerned are king scallops in Le Havre (22%), mussels in La Rochelle (16%) and sea urchins in Toulon (14%) (Leblanc et al. 2006).

Mercury

In seafood products, mercury is mainly present as methylmercury (see section 5.4). For methylmercury (MeHg), EFSA and AFSSA have both acknowledged that some population groups are particularly at risk: pregnant and breast-feeding women, very young children, fishing communities in heavily contaminated areas (EFSA 2004a; AFSSA 2004). Both agencies recommend that special information be aimed at these groups to encourage them to eat a wider range of fish species. In France, exposure study results show that values are two times lower than the PTWI (of 4 µg) for inorganic Hg kg^{-1} body weight. In adult males, who are high seafood consumers, the CALIPSO data suggest that shellfish result in an average intake of 0.47 µg day^{-1} of MeHg per adult, which approaches 1.2 % of the PTWI

(Leblanc et al. 2006). In general, fish contribute 86%, and molluscs and crustaceans 13% of MeHg exposure (EAT 2004 ; Leblanc et al. 2005 ; Sirot et al. 2008).

Arsenic

In 2003, the mean total arsenic intake in Europe was estimated at 125 µg day^{-1} in adults (SCOOP 2004); seafood accounted for over 50% of this exposure. The mean arsenic intake from shellfish from CALIPSO data is 84 µg kg bw^{-1} week^{-1} in adults (Leblanc et al. 2005, 2006). The seafoods that contribute most to the French population's inorganic arsenic exposure are king scallops (8.6% of intake from seafood) and oysters (7.0%) (Sirot et al. 2009). In the general population, shellfish contribute 0.2% of the PTWI for total arsenic (EAT 2004 ; Leblanc et al. 2005). However, it was noted in the 72nd JECFA committee meeting that the PTWI of 15 µg kg bwt^{-1} (equivalent to 2.1 µg kg bwt^{-1} per day) approaches the benchmark dose lower limit (BMDL$_{05}$), and therefore the PTWI is no longer appropriate. The committee withdrew the previous PTWI (JECFA 2010a). EFSA concluded that the overall range of BMDL$_{01}$ values of 0.3 to 8 µg kg^{-1} bwt per day should be used, instead of a single reference point, in characterizing the risk of inorganic arsenic (EFSA 2009).

Organostannic Compounds

In France, the average exposure of high seafood consumers to nine organostannic compounds is far below the tolerable daily intake. This intake is 8 to 19% of the TDI of 0.1 µg Sn kg^{-1} bwt set by EFSA for the combined total from the following Sn compounds: tributyltin (TBT), dibutyltin (DBT), triphenyltin (TPT) and di-octyltin (DOT) (AFSSA 2006; EFSA 2004b; Guerin et al. 2007).

Dioxins

The mean dioxin intake from shellfish for the sum of PCDD/Fs and dl-PCBs is 18.7 µg.kg^{-1} bwt.week^{-1} in adults (from CALIPSO data; Leblanc et al. 2006). However, it is important to note that these values are overestimated, because cooking seafood reduces the PCDD content (Hori et al. 2005). The shellfish contribution to the tolerable intake is low (5.73% for all species).

PCBs

Only 28% of high seafood consumers show indicator PCB (sum of PCB 28, 52, 101, 118, 153 and 180) levels below the TDI of 0.02 µg kg^{-1} bwt day^{-1}, the average being 0.40 ± 0.55 µg kg^{-1} bwt day^{-1}. Shellfish only contribute 9.5% to the TDI, 45% of which comes from the king scallop (Leblanc et al. 2006).

Body Burdens for these Trace Elements

The CALIPSO survey provides data on the body burdens (saturation) of high seafood consumers (adults) in lead, cadmium, mercury and arsenic. However, from these data, it is not possible to determine how much shellfish contributes to the body burden. Concentrations of these chemical contaminants, measured in blood and urine, are compared with a 'basal value'. This basal value is defined as the value found at the 95th percentile (P95) of the general French population that is not occupationally exposed. This value should not be interpreted as a maximum allowable quantity, but it makes it possible to identify a possible body overload. In conclusion, high seafood consumers do not display a significantly higher body burden than does the P95 of the general population for lead, cadmium or mercury. For lead, 6% of high seafood consumers exceed the basal value of 90 µg L^{-1} for men, and 70 µg L^{-1} for women. There were no observed blood concentration of lead >200 µg L^{-1}, the concentration above which a person is put under medical observation. For cadmium, fewer than 5% of individuals retained cadmium levels in urine higher than the basal value of 2 mg kg^{-1} creatinine. For mercury, only 3% of the values exceeded the basal value of 10 µg L^{-1} in blood. No signs of a health risk impairment were identified for any of these three contaminants. However, 22% of individuals displayed inorganic arsenic levels that exceeded the basal concentration in urine of 10 mg kg^{-1} creatinine, which is the P95 value for the general population (INRS 2010; Pillière and Conso 2007).

We conclude from the foregoing that, for high seafood consumers, the contribution of shellfish to inorganic contaminants was 1 to 10 % of TWI or PTWI for Cd, MeHg, and Sn (up to 19% for Sn), and the arsenic body burden was higher for 22% of individuals studied. These percentages will differ if the established effective regulatory threshold is different (Table 3).

5.4. Health Risk Assessment Uncertainty from Contaminant Bioavailability and Speciation Effects

The regulatory limits for lead, cadmium and mercury that were established in Regulation (EC) 1881/2006 (EC 2006c) are based on the total concentrations of them that exists in foodstuffs. However, only the bioavailable fraction can be transferred from shellfish to humans, during digestion. This fraction is influenced by several factors, and is rarely 100% of the amount present.

For mercury, it is the methylated form that predominates as a seafood residue, and this organic form is also the most toxic (Nakagawa et al. 1997 ; Storelli et al. 1998). A study of oysters and mussels sampled in 1996 under the RNO sampling program, show MeHg/THg (total Hg) ratios ranging from 11% to 88%. No notable differences were observed between the two mercury species, but there was considerable geographical variability (Claisse et al. 2001). Bioamplification has been observed in organic forms of mercury, with an increase in concentration at each trophic step in the food chain. In the CALIPSO survey, MeHg/THg ratios in shellfish ranged from 50% to 100% (Leblanc et al. 2006).

The toxicity of arsenic depends on its chemical form and its bioavailability. Inorganic forms of arsenic are more toxic than are the organic forms (Michel 1993 ; Sharma and Sohn 2009). A high proportion of the organic arsenic in seafood is in weakly toxic forms such as arsenobetaine and trimethylarsine. These forms are rapidly excreted (ATSDR 2007; Liber et al. 2006). According to the WHO, there are some (but limited) data showing that 25% of total arsenic in foodstuffs is in inorganic form. The data from a French study (Noël et al. 2003) suggest that, in fishery products, 5 to 10% of arsenic is in inorganic form, whereas the CALIPSO study gives figures ranging from 0.1 to 3.5% in fish, and from 0.1 to 6.7% in shellfish (Sirot et al. 2009). However, the percentage of inorganic arsenic is quite variable in fish and shellfish, and data from the international literature indicate that the percentage of inorganic arsenic in marine/estuarine finfish does not exceed 7.3%. However, in shellfish, it can reach 25% in organisms from presumably uncontaminated areas, although there are few data available for freshwater organisms. However, percentages can be much higher in organisms from contaminated areas and in seaweed (Schoof and Yager 2007; Lorenzana et al. 2009).

For mercury and arsenic it is thus difficult to conclude whether or not they pose a public health risk, when only the total levels present are given, and without differentiating between the proportion that is inorganic and organic.

Efforts have been made in various studies to quantify the bioavailability, or rather bioaccessibility, of trace elements that are accumulated by bivalves (He et al. 2010; Metian et al. 2009). Amiard et al. (2008) simulated human digestion *in vitro* with the flesh of naturally contaminated oysters, whelks, mussels, scallop species and *Ruditapes* clams. The total concentrations in these samples exceeded regulatory limits for the following (Amiard et al. 2008):

- Cd, in whelks (*B. undatum*) purchased in France, and in the adductor muscles of noble scallops (*Chlamys nobilis*) from Hong Kong;
- Pb in oysters (*O. edulis*) from Restronguet Creek, UK, Zn in whelks, and
- Cu and Zn in all samples of oysters from contaminated sites.

However, these comparisons are based on Australian and Asian standards that Europe does not recognize. If the concentrations recorded were indeed bioaccessible concentrations, only the levels of Cd in scallop species and Zn in whelks would be acceptable.

Although levels of arsenic in the urine and more specifically inorganic arsenic are satisfactory biomarkers for occupational and drinking water exposures, the literature data show that consumption of seafood gives variable results. The amount of total or inorganic arsenic in the urine is, therefore, not a relevant or usable indicator of the intensity of exposure to the most toxic forms of arsenic ingested with food, and with seafood in particular. To assess the health risk of ingesting arsenic via seafood, the species of arsenic must be taken into account, because there are significant differences in toxicity among the different chemical species. For example, the mean LD_{50} (lethal dose, 50%) in rats, expressed in mg kg^{-1} bwt, is 14 for potassium arsenite, 20 for calcium arsenate, 700 to 1800 for MMA (monomethylarsonic acid), 700 to 2600 for DMA (dimethylarsinic acid) and over 10,000 for arsenobetaine. In drinking water, arsenic is mainly found in the inorganic form, as the oxide anions arsenite and arseniate. The main foodstuffs supplying inorganic arsenic are cereals, flour and raw rice (Schoof et al. 1999), but seafoods contain several organic arsenic compounds and are a major food source of arsenic (Francesconi and Edmonds 1998 ; Munoz et al. 2000). Arsenic in fish, most shellfish and many crustaceans is mainly in the form of arsenobetaine, whose very weak toxicity has been established (Kaise et al. 1985 ; Sabbioni et al. 1991). Arsenobetaine is quickly excreted in an unaltered form in the urine (70% in 3 days) (Cannon et al. 1983), and does not react with the reagents used in urinary tests. Hence, arsenobetaine is clearly differentiated during arsenic speciation in the urine, and several experimental studies have shown that its consumption does not significantly alter the parameters of urine analyses for inorganic arsenic (Buchet et al. 1996 ; Heinrich-Ramm et al. 2002 ; Hsueh et al. 2002).

Algae, bivalves, crustaceans and fish all contain derivatives of ribose and arsenic called arsenoribosides (arsenosugars), which are metabolized and excreted in the urine, particularly as DMA (V), and in the form of dimethyloxarsylethanol and trimethylarsine oxide (Francesconi et al. 2002 ; Ma & Le 1998 ; Wei et al. 2003). It has been observed that ingestion of arsenoribosides via food invalidates the use of urine testing for inorganic arsenic derivatives, and as an exposure marker for these derivatives. As a result, these tests cannot satisfactorily reflect intake of inorganic arsenic (for which there is a risk of excess cancers) in individuals consuming seafood (Borak and Hosgood 2007 ; Heirich-Ramm et al. 2002 ; Ma and Le 1998). Considering that arsenic from seafood is usually eliminated within 3 days (Crecelius 1977 ; Freeman et al. 1979), it is recommended that urine tests for inorganic arsenic should occur at least 3 or 4 days after any seafood consumption (Foa et al. 1984 ; Kales et al. 2006).

As previously mentioned, health risks associated with chemical contaminants are difficult to assess because the risks they pose are normally of a chronic nature, and their sources of human exposure are numerous. Therefore, it is not possible to attribute a high body burden specifically to shellfish consumption, even though seafood is a major contributor of some contaminants, especially arsenic and mercury.

6. Conclusions

The major conclusions we have reached from compiling and reviewing the literature cogent to the topic of this paper are as follows:

- Both organic and inorganic chemicals have been identified as residual chemical contaminants in shellfish. Some contaminants, particularly metals, dioxins, dl-PCBs and PAH that appear as residues in molluscs, pose a potential hazard to consumers, which has resulted in European regulatory limits being established for them.

- To protect shellfish from chemical contamination, shellfish production and commercialization are managed according to safe practices which are under the European "Hygiene Package" regulations. Product quality is maintained by controlling facilities, tracking major steps in shellfish production and ensuring that defective batches are kept from the market. Such regulation ensures greater transparency and product quality for consumers. However, limits to regulating shellfish production also exist, because, in France, it is difficult to trace all production steps of living shellfish from the earliest to latest stages, particularly for oysters, wherein the same oyster may be successively raised at facilities in different areas.

- Although monitoring results show few non-conformities, the samplings that are made cannot be considered as representing all shellfish production in France, because the numbers of samples taken are limited. In addition, when residue levels are exceeded, they normally occur in oysters and mussels, which are the most commonly eaten species. Hence, self-inspection by producers, enforcement of compliance with good practices and regular checks on production are indispensable additional measures to ensure food safety

- Last but not least, is that the chemical monitoring network that has been set up in France (the RNO program and its successor ROCCH) to screen for contaminants clearly shows that there is low chemical contamination of molluscs, and of seawater in which the molluscs live. Moreover, when shellfish contamination occurs, it poses a generally low risk to the general French population, because the proportion of the diet that shellfish constitutes is low. The exceptions are when contaminants reach those people who are either high-consumers of shellfish, or are a more susceptible population, such as pregnant and breast-feeding women and very young children. Appropriate research programs should first be developed to protect these more susceptible categories of the population.

To improve chemical safety of shellfish, some suggestions can be given and they concern:

- the relaying (depuration) program currently used to purify shellfish of microbiological contamination before commercialisation , which should be further researched to determine if and how chemical residues in shellfish could be similarly reduced before consumption. The alternative that has been used, to date, i.e., closing contaminated areas for long time periods results in significant economic losses
- the monitoring of farmed shellfish which should be extended to other chemicals that are suspected to present a consumer risk, including arsenic, which the CALIPSO study disclosed to have high consumer urine levels, and intensive monitoring for cadmium, which was abnormally detected in some shellfish in 2009. We would also suggest monitoring for TBT and PAHs contamination levels, to ensure that these chemicals do not migrate from the harbor to oyster farms, as was observed to occur

Finally, from the data assembled in this review, we conclude that there is a strong argument not to curtail existing monitoring programs in edible shellfish. The major reason for continuing monitoring activities is that great variability exists in the magnitude to which different contaminants in shellfish bioconcentrate. Both environmental and species parameters are known to affect the degree of bioconcentration and bioaccumulation of potentially harmful residues, and, moreover residue loads are affected by the season during which the shellfish are harvested. Therefore, under equal conditions of environmental contamination, some species do exceed the European regulatory limits, whereas others do not. These variabilities explain the necessity of why monitoring was extended to farmed shellfish species by the ROCCH, and why monitoring activities should continue.

7. Summary

In this review, we address the identification of residual chemical hazards in shellfish collected from the marine environment or in marketed shellfish. Data, assembled on the concentration of contaminants detected, was compared with the appropriate regulatory and food safety standards. Moreover, data on human exposure and body burden levels were evaluated in the context of potential health risks.

Shellfish farming is a common industry along European coasts. The primary types of shellfish consumed in France are oysters, mussels, king scallops, winkles, whelks, cockles, clams and other scallops. Shellfish filter large volumes of water to extract their food, and are excellent bioaccumulators. Metals and other pollutants that exist in the marine environment partition into particular organs, according to their individual chemical characteristics. In shellfish, accumulation often occurs in the digestive gland, which plays a role in assimilation, excretion and detoxification of contaminants. The concentrations of chemical contaminants in bivalve molluscs are known to fluctuate with the seasons.

European regulations limit the amount and type of contaminants that can appear in foodstuffs. Current European standards regulate the levels of microbiological agents, phycotoxins and some chemical contaminants in food. Since 2006, these regulations have been compiled into the ‘Hygiene Package'. Bivalve molluscs must comply with maximum levels of certain contaminants as follows: lead (1.5 mg kg^{-1}), cadmium (1 mg kg^{-1}), mercury (0.5 mg kg^{-1}), dioxins (4 pg g^{-1} and dioxins + dl-PCBs 8 pg g^{-1}) and benzo[*a*]pyrene (10 µg kg^{-1}).

In this review, we identify the levels of major contaminants that exist in shellfish (collected from the marine environment and/or in marketed shellfish). The following contaminants are among those that are profiled: Cd, Pb, Hg, As, Ni, Cr, V, Mn, Cu, Zn, Co, Se, Mg, Mo, radionuclides, benzo[*a*]pyrene, PCBs, dioxins and furans, PAHs, TBT, HCB, dieldrin, DDT, lindane, triazines, PBDE, and chlorinated paraffins.

In France, the results of contaminant monitoring have indicated that Cd, but not lead (< 0.26 mg kg^{-1}) or mercury (< 0.003 mg kg^{-1}), have had some non-compliances. Detections for PCBs and dioxins in shellfish were far below the regulatory thresholds in oysters (< 0.6 pg g^{-1}), mussels (< 0.6 pg g^{-1}) and king scallops (< 0.4 pg g^{-1}). The benzo[*a*]pyrene concentration in marketed mussels and those coming from farmed shellfish do not exceed the regulatory threshold. Some monitoring data are available on shellfish flesh contamination for unregulated organic contaminants.

Of about a hundred existing organostannic compounds, residues of the mono-, di- and tributyl tin (MBT, DBT and TBT) and mono-, di- and tri-phenyl tin compounds (MPT, DPT and TPT) are the most frequently detected in fishery products. Octyltins are not found in fishery products. Some bivalve molluscs show arsenic levels up to 15.8 mg kg^{-1}. It seems that the levels of arsenic in the environment derive less from bioaccumulation, than from whether the arsenic is in an organic or inorganic form. In regard to the other metals, levels of zinc and magnesium are higher in oysters than in mussels.

To protect shellfish from chemical contamination, programs have been established to monitor water masses along coastal areas. The French monitoring network (ROCCH) focuses on environmental matrices that accumulate contaminants. These include both biota and sediment. Example contaminants were studied in a French coastal lagoon (Arcachon Bay) and in an estuary (Bay of Seine), and these were used to illustrate the usefulness of the monitoring programs. Twenty-one pesticidal and biocidal active substances were detected in the waters of Arcachon Bay during the summers from 1999 to 2003, at concentrations ranging from a few ng L^{-1} to several hundred ng L^{-1}. Most of the detected substances were herbicides, including some that are now banned. Organotin compounds have been detected in similarly semi-enclosed waters elsewhere (bays, estuaries, harbors). However, the mean concentrations of cadmium, mercury, lead and benzo[*a*]pyrene, in transplanted mussels, were below the regulatory limits.

In 2007, the mean daily consumption of shellfish in the general French population was estimated to be 4.5 g in adults; however, a wide variation occurs by region and season (INCA2 study). Tabulated as a proportion of the diet, shellfish consumption represents only 0.16% of overall solid food intake. However, the INCA 2 survey was not well-suited to estimating shellfish consumption, because of the small number of shellfish consumers sampled. In contrast, the mean consumption rate of bivalve molluscs among adult high consumers of fish and seafood products, i.e., adults who eat fish or seafood at least twice a week, was estimated to be 153 g a week (8 kg yr^{-1}). The highest mean consumption is for king scallops (39 g wk^{-1}), followed by oysters (34 g wk^{-1}) and mussels (22 g wk^{-1}).Thus, for high seafood consumers, the contribution of shellfish to inorganic contaminant levels is 1 to 10% of TWI, or PTWI for Cd, MeHg, and Sn (up to 19% for Sn), and the arsenic body burden is higher for 22% of individuals studied.

The human health risks associated with consuming chemical contaminants in shellfish are difficult to assess for several reasons: effects may only surface after long-term exposure (chronic risk), exposures may be discontinuous, and contamination may derive from multiple sources (food, air, occupational exposure, etc.). Therefore, it is not possible to attribute a high body burden specifically to shellfish consumption, even if seafood is a major dietary contributor of some contaminant, e.g., arsenic and mercury.

The data assembled in this review provide the arguments for maintaining the chemical contaminant monitoring programs for shellfish. Moreover, the results presented herein suggest that monitoring programs should be extended to other chemicals that are suspected of presenting a risk to consumers, as illustrated by the high level reported for arsenic (in urine) of high consumers of seafood products from the CALIPSO study. In addition, the research conducted in shellfish farming areas of Arcachon Bay highlights the need to monitor TBT and PAH contamination levels, to ensure that these chemical pollutants do not migrate from the harbor to oyster farms.

Finally, we have concluded that shellfish contamination from seawater offers a rather low risk to the general French population, because shellfish do not constitute a major contributor to dietary exposure of chemical contaminants. Notwithstanding, consumer vigilance is necessary among regular shellfish consumers, and especially for those residing in fishing communities, for pregnant and breast-feeding women and for very young children.

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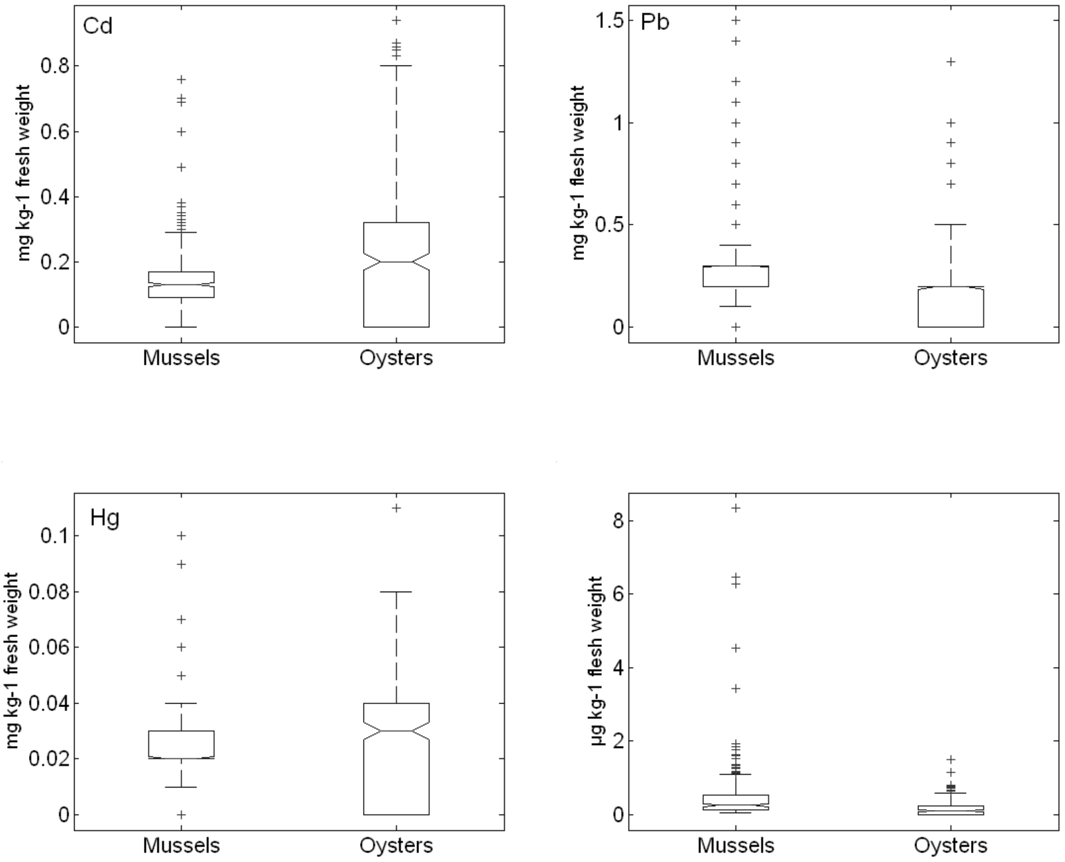
Figures

Fig. 1 Distribution of contamination in mussels and oysters in French shellfish farming areas from 2003 to 2007 (data from Claisse et al. 2006 for 2003 to 2005; unpublished results from the same authors for 2006 to 2007 period). (A) Cadmium (B) Lead (C) Mercury in mg kg-1 fresh wt, and (D) benzo[a]pyrene in µg kg-1 fresh wt (E) provides values used to construct graphs A to D



E	Mussels	Oysters
Cadmium	0.15 ± 0.09 (n = 374)	0.34 ± 0.18 (n = 239)
Lead	0.03 ± 0.01 (n = 374)	0.04 ± 0.02 (n = 239)
Mercury	0.03 ± 0.02 (n = 374)	0.03 ± 0.02 (n = 239)
Benzo[*a*]pyrene	0.56 ± 1.01 (n = 180)	0.27 ± 0.24 (n = 180)

Tables

Table 1 : List of Abbreviations and Acronyms Used in this Review

AFSSA: *Agence Française de Sécurité Sanitaire des Aliments* (French food safety agency) (website : www.anses.fr)
ANSES: *Agence Nationale de Sécurité Sanitaire de l'alimentation, de l'environnement et du travail* (French agency for food, environmental and occupational health & safety) (website : www.anses.fr)
ATSDR: Agency for Toxic Substances and Disease Registry
BCF: Bioconcentration factors
BWT: Body weight
BMDL$_{01}$: Benchmark Dose (Lower Confidence Limit 0.01)
BMDL$_{05}$: Benchmark Dose (Lower Confidence Limit 0.05)
BQSPMED: *Bureau de la Qualité Sanitaire des Produits de la Mer et d'Eau Douce* (Office for the quality and safety of food products from fresh and marine waters)
BRAB: *Bureau de la Réglementation Alimentaire et des Biotechnologies* (Office of Food and Biotechnology Regulations)
BTEX: Benzene, Toluene, Ethylbenzene, Xylene
CALIPSO: *Etude des Consommations ALimentaires de produits de la mer et Imprégnation aux éléments traces, PolluantS and Omega-3* (Fish and seafood consumption study and biomarker of exposure to trace elements, pollutants and omega-3)
CF: concentration factor
CNC: French national shellfish farming committee
DBT: dibutyltin
DDAM: *Direction Départementale des Affaires Maritimes* (Local maritime affair authorities)
DDT: dichlorodiphenyl trichloroethane
DDE: dichlorodiphenydichloroethylene
DDD: dichlorodiphenyldichloroethane
DDSV: *Direction Départementale des Services Vétérinaires* (Local veterinary authorities)
DEHP: di(2-ethylhexyl)phthalate
DGAL: *Direction Générale pour l'Alimentation* (French directorate for food)
DGS: *Direction Générale de la Santé* (French directorate general for health)
DMA: dimethylarsinic acid
DOT: di-octyltin
DPMA*: Direction des Pêches Maritimes et de l'Aquaculture* (Directorate for marine fisheries and aquaculture)
DPT: diphenyltin
EAT*: Etudes Alimentaires Totales* (Total Diet Study (TDS))
EC: European Community
EEC: European Economic Community
EFSA: European Food Safety Authority
EPA: Environmental Protection Agency
EU-RL: EU reference laboratory
FAO: Food and Agriculture Organization of the United Nations
GST: glutathione S-transferase
HACCP: Hazard Analysis Critical Control Point
IAEA: International Atomic Energy Agency
IARC: International Agency for Research on Cancer
IFREMER*: Institut Français de Recherche pour l'Exploitation de la MER* (French research institute for exploitation of the sea)
INCA: *Enquête Individuelle et Nationale sur la Consommation Alimentaire* (Consumption data for the general population)

INRS: *Institut National de Recherche et de Sécurité* (National institute of research and safety)

IRSN: *Institut de radioprotection et de sûreté nucléaire* (French Institute for Radiation Protection and Nuclear Safety)

JECFA: Joint FAO/WHO Expert Committee on Food Additives

JORF: *Journal Officiel de la république Française* (Official Journal of the French Republic

LD_{50}: Lethal Dose 50%

LERQAP: *Laboratoire d'Etudes et de Recherches sur la Qualité des Aliments et les Procédés Agroalimentaires* (Laboratory of studies and research on food quality and food processes)

MAP: Mediterranean Action Plan

MCSI : *Mission de Coordination Sanitaire Internationale* (International health and safety coordination mission)

MeHg: methyl mercury

MED POL: Barcelona convention for the protection of the Mediterranean sea against pollution

MBT: monobutyltin

MMA: monomethylarsonic acid

MOREST: *Mortalité ESTivale d'Huîtres* (Oyster summer mortality program)

MPT: monophenyl tin

MT: metallothioneins

NPE: nonylphenol ethoxylates

NRL: National Reference Laboratory

OCA-EN: *Observatoire des Consommations Alimentaires- Epidemiologique Nutritionnelle* (Food consumption and nutritional epidemiology unit)

OPE : octyphenol ethoxylate

OSPAR: Convention for the protection of the marine environment of the North-East Atlantic

P95 : 95th percentile

PAH: Polycyclic aromatic hydrocarbon

PCB: Polychlorinated biphenyls

PCBi: indicator PCBs (sum of selected PCBs)

dl-PCB: PCB dioxin like

PCDD/Fs: polychlorinated dibenzo-dioxins/furans

PTMI: Provisional Tolerable Monthly Intake

PTWI: Provisional Tolerable Weekly Intake

REPAMO: *Réseau de Pathologie des Mollusques* – Mollusc pathology network

RNO: *Réseau National d'Observation* – French national monitoring network

ROCCH: *Réseau d'Observation de la Contamination CHimique du milieu marin* (French national monitoring network)

SCOOP: Scientific COOPeration

TBT: tribultytin

TDI: Tolerable Daily Intake

THg: total mercury

TPT: triphenyl tin

TWI: Tolerable Weekly Intake

UNEP: United Nations Environment Programme

WFD: Water Framework Directive

WHO: World Health Organization

WT: Weight

Table 2 Regulatory thresholds for consumption of various contaminants in bivalve molluscs (EC 2006c, amended by EC 2008a)

Contaminant		Maximum level (fresh wt)
Metals	Lead	1.5 mg kg^{-1}
	Cadmium	1 mg kg^{-1}
	Mercury	0.5 mg kg^{-1}
Dioxins and PCBs	Dioxins Dioxins + dl-PCBs	4 pg g^{-1} 8 pg g^{-1}
PAHs	Benzo[a]pyrene	10 µg kg^{-1}

Table 3 Levels of contamination in environment and in shellfish flesh sampled from the marketplace for three inorganic contaminants (Cd: cadmium; Pb: lead; Hg: mercury) regulated according to EC (2006c) and amended by EC (2008a). Arsenic (As), though not regulated, is included in the table, because it is also closely monitored

	Cd	Pb	Hg	As
Anthropogenic source [c]	Industry(coloring;stabilizer; cadmium plating)	Industry (printing. metallurgy, etc.)	Rare in the natural environment; electrical industry, etc.	Rare in the natural environment; Metallurgy industries, etc.[c]
Mean levels in the environment				
Seawater (µg L^{-1}) [a]	0.01-0.1	0.5-5	0.005-0.05	1-2[a]
Sediments (µg g^{-1} dry wt) [a]	0.1-1	5-50	0.05-0.5	5-3000[a]
Contamination in shellfish (mg kg^{-1} fresh wt)				
Regulatory threshold	1 [d]	1.5 [d]	0.5 [d]	
Oyster (min-max) (n=6) *	0.07-0.22 [d]	0.04-0.08 [d]	0.003-0.02 [d]	0.003[d]
Mussel (min-max) (n=6) *	0.06-0.18 [d]	0.14-0.26 [d]	0.003-0.02 [d]	0.88-3.39 [d]
Cockle (mean) (n = 2) **	0.04[b]	0.04 [b]	0.02 [b]	1.78[b]
Scallop (n = 1) **	1.14 [b]	0.09 [b]	0.01 [b]	2.42 [b]
Winkle (mean) (n = 3) **	0.19 [b]	0.09 [b]	0.01 [b]	6.39 [b]
Whelk (mean) (n = 3) **	0.78 [b]	0.06 [b]	0.03 [b]	15.8 [b]
King scallop (mean) (n = 4) **	0.27 [b]	0.07 [b]	0.03 [b]	2.96 [b]
PTWI (µg kg^{-1} bwt.wk^{-1}) PTMI (µg kg^{-1} bwt.month^{-1}) with (Px) = PTWI or PTMI values	7[b] (P1) 2.5 [e] (P2) 25 [g] (P3)	25 [b] (P4) withdrew in 2010 [g]	1.6 (MeHg) and 5 (Hg total) [b] (P5) 4 (Hg inorganic) [f] (P6)	Intake not to exceed [b] :15 (As^{III} and As^{V}) and 350 (total As) Withdrawn in 2010 [f]
Contribution of shellfish in % of PTWI or PTMI (/Px)				
High consumers (CALIPSO) [b]	8.22% (/P1)–23% (/P2) –10%(/P3)	0.8% (/P4)	0.12%(MeHg) (/P5) – 1.16% (/P6)	1.2% (As^{III} and As^{V}) -2.5% (total As)
General population (EAT) [d]	0.25%(/P1)–0.72%(/P2)–0.313% (/P3)	0.1% (/P4)	1.8%	0.2% (total As)
Mean saturation as % of basal value***				
Blood (basal value) [b]	62% (1 µg L^{-1} blood)	42% (90 µg L^{-1} blood)	37% (10 µg L^{-1} blood)	nd
Urine (basal value) [b]	35% (2 µg g^{-1} creat)	23% (25 µg g^{-1} creat)	nd	280% (10 µg g^{-1} creat. for inorg As) [e]
Risk category [d]	T; Cat. 1 IARC (human carcinogen)	T + N	T	T + N; Cat. 1 IARC[c]
Toxicity[d]	Renal damage; bone lesions; delayed foetal growth; reduced fertility	Neurotoxicity (saturnism); haematological toxicity (anaemia); congenital anomalies	Neurological damage; kidney failure; digestive tract inflammation	Acute: digestive disorders; Chronic: cancers of skin, lung, bladder, kidney. Skin disorders [j]

PTWI: provisional tolerable weekly intake; PTMI: provisional tolerable monthly intake

a. Merian *et al*. 2004; b. CALIPSO (Leblanc et al. 2006) c. INRS (2010) Toxicology data sheets d. EAT 2004 e. EFSA 2009 f. JEFCA 2010a. g. JEFCA 2010b

* Each sample consists in 5 sub-samples at most, weighted by main place of purchase main place of supply used by consumers on the Secodip panel. Analyses involved an amount of about 0.6 g per composite sample and replicate analyses were performed on each sample.

** Each sample of fresh product analysed consists in about 1000 g of product i.e., 5 sub-samples of 200 g. The origin and distribution of the 5 sub-samples was determined according to place of purchase selected from data on frequency of purchase in the consumer survey, which were weighted by frequency of consumption and quantities consumed.

***basal value: value found for the 95th percentile of the general French population not occupationally exposed (EAT 2004)

n: number of samples. nd: not determined; T: toxic; N: dangerous for the environment

Table 4 Levels of contamination in the environment and in shellfish sampled from the marketplace for unregulated inorganic contaminants

	Ni	Cr	V	Mn	Cu	Zn	Co	Se	Mg	Mo
Anthropogenic sources	Industry (production of stainless steel, catalysis, etc.)[b]	Industry (anti-corrosion, catalysis, pigments, etc.)[b]	Titanium industry, ports, petrochemicals [c]	Industry (catalysis, battery manufacture, etc.)[b]	Electrical industry, construction, etc.[b]	Industry (anti-corrosion coatings, alloys, etc.) [b]	Industry (alloys, pigments, fertilisers, etc.) [b]	Industry (electrical, metallurgy, etc.) [b]	Industry (chemical, alloys, etc.) [b]	Industry (alloys, catalysis, pigments, etc.) [b]
Mean levels in the environment										
Seawater (µg L^{-1})	0.6[a]	0.2[a]	1.9[a]	0.01[a]	0.005-0.05 [a]	0.5-5 [a]	0.002 [a]	0.09 [a]	$1.3\ 10^{6}$ [h]	nd [h]
Sediments (µg g^{-1} dry wt)	45[a]	60[a]	252[a]	1.2 [a]	5-50 [a]	50-500 [a]	0.045 [a]	$1.7\ 10^{-4}$ [a]	45 [h]	$8.\ 10^{-4}$ [a] (b)
Mean contamination of shellfish (mg kg^{-1} fresh wt)										
Oysters (min-max) (n=6) * Mussels (min-max) (n=6) *	0.03-0.17 [f] 0.20-0.53 [f]	0.02-0.15 [f] 0.07-0.25 [f]	6.3 [d] 7.3 [d]	3.18-7.07[f] 1.32-3.68 [f]	6.90-30.1 [f] 0.89-2.39 [f]	111-312 [f] 8.23-26.7 [f]	0.01-0.05 [f] 0.07-0.18 [f]	0.011 [f] 0.011 [f]	590-957[f] 160-673 [f]	0.02-0.20 [f] 0.05-0.51 [f]
Recommended nutritional intake per day (d^{-1})				2-3 mg d^{-1} [g]	0.8-2 mg d^{-1} [g]	6-19 mg d^{-1} [g]	0.6 µg d^{-1} [g]	20-80 µg d^{-1} [g]	80-420mg d^{-1} [g]	30-50 µg d^{-1} [g] (a)
Intake not to exceed	nd	nd	100 µg d^{-1} [g]	4.2-10 mg d^{-1} [g]	nd	15-40mg d^{-1} [g]	200 µg d^{-1} [g]	150 µg d^{-1} [g]	750 mg d^{-1} [g]	350 µg d^{-1} [g]
Intake from shellfish ingestion in adult men										
	0.76 µg d^{-1} [f]	0.23 µg d^{-1} [f]	nd	0.01 mg d^{-1} [f]	0.02 mg d^{-1} [f]	0.11 mg d^{-1} [f]	0.12 µg d^{-1} [f]	0.03 µg d^{-1} [f]	1.2 mg d^{-1} [f]	0.33 µg d^{-1} [f]
Risk category	Xn + T (monoxide) [b]	T + N; Cr(VI) Cat. 1 IARC (human carcinogen) [b]	Xn (divanadium pentaoxide); combustible (vanadium trioxide) [e]	Xn (Mn dioxide) [b]	Xn [b]	C (chloride); Xi (sulphate); T (chromate) + N [b]	T + N (Co sulphate); Xn (cobalt) [b]	T + N [b]	Xi (Mg chloride) [b]	Xi [b]
Toxicity	Digestive disorders; probable carcinogenic effect [b]	Cr(VI): Digestive disorders; kidney failure [b]	Severe systemic poisoning symptoms and death (divanadium pentaoxide); headaches, vomiting (vanadium trioxide) [e]	Chronic: nervous and respiratory disorders [b]	Chronic: hepatitis; neurological disorders [b]	Digestive disorders [b]	Irritative respiratory syndrome [b]	Digestive disorders; neurological signs [b]	Muscular tetany, digestive disorders [f]	Diarrhoea, anaemia, erythrocytic immaturity, uricemia [f]

a- Merian et al. 2004; b- INRS(2010) Toxicology data sheets; c- Saavedra et al. 2004; d- Roux et al. 2001; e- International chemical safety sheets 2010; f- EAT 2004; g - AFSSA 2008b; h- OSPAR 2008

* The shellfish contamination data were obtained from an individual composite sample of 5 sub-samples at most, weighted by main place of purchase used by consumers on the Secodip panel. Analyses involved an amount of about 0.6 g per composite sample and replicate analyses were performed on each sample.
nd: not determined; T: toxic; N: dangerous for environment; Xn: noxious; C: corrosive; Xi: irritant; (a) estimated adult requirement – no DRI value; (b) estimated concentration

Table 5 Radionuclides in the environment and in shellfish sampled from the marketplace

	Radionuclides ^{99}Tc. ^{129}I. ^{226}Ra. ^{210}Po. ^{238}U. ^{239}Pu. ^{240}Pu. ^{241}Am
Anthropogenic sources	Nuclear industry; fertilizer manufacture [a]; mining [c]
Mean levels in the environment	
Seawater (µg L^{-1})	^{137}Cs 0.002 – 0.500 Bq L^{-1} [a] ^{99}Tc 0.350 Bq L^{-1}[a] ^{210}Po 1 - 5 Bq m^{-3} [c]
Sediments (µg g^{-1} dry wt)	^{210}Po 9 - 125 Bq kg^{-1} [c]
Mean contamination of shellfish	
Mussels (min-max)	^{210}Po 150 - 600 Bq kg^{-1} dry wt [c]
Cockles (min-max)	^{210}Po 80 - 1200 Bq kg^{-1} dry wt [c]
Molluscs (mean)	^{210}Po 15 Bq kg^{-1} dry wt [b]
Intake not to exceed	Men 2 mSv yr^{-1} (probable maximum individual dose) [a]
Maximum estimated intake from shellfish ingestion, adult men	^{210}Po 160 µSv yr^{-1} [b]
Risk category	Radiological and chemical risk [a]
Toxicity	Irradiation, contamination, cancers

a- OSPAR 2007; b- Pradel et al. 2001; c- IRSN 2010

Table 6 Levels of contamination in the environment and shellfish flesh sampled from the marketplace for regulated organic contaminants under Regulation (EC) N°1881/2006 (EC 2006c)

Contaminants	PCBs		Dioxins and furanes	PAHs
Sources	Industrial products: transformer and condenser oils [b] Paint plastifiers and plastics, sealants [b]		Incineration, metallurgy processes [b] Use of active chlorine for bleaching paper pulp [b] Internal combustion engines, wildfire, wood burning [e]	Constituents of crude oil, incineration and incomplete burning of organic matter: wood, coal, heating oil [b]. Oil production. Offshore activities [b]. Coal tar coatings, exhaust gases, [b]. Wildfire, volcanic eruptions [b]
	Current reservoirs: soil, sediments, rubbish dumps/landfills, old infrastructures [b] Remobilisation of old sediments (dredging) [b] Rivers, atmosphere and ocean currents [b], professional or recreational nautical activities [c]			
Mean levels in the environment				
Seawater (ng L^{-1})	0.001 [b]		nd [e]	Benzo[a]pyrene 0.001 – 0.005[b] Fluoranthene 0.036 – 0.285 [b] Benzo[b+k]fluoranthene 0.001 – 0.017 [b] Pyrene 0.011 – 0.053 [b] Total PAHs < 0.0001 - 8500 [b]
Sediments (µg kg^{-1} dry wt)	Congeneric PCBs (28/52/101/138/153/180) < 0.010 – 0.116 [b]		0.020[e]	Benzo[*a*]pyrene 0.2 – 112 [b] Fluoranthene 0.72 – 160 [b] Benzo[*b*+*k*]fluoranthene 1.1 - 434 [b] Pyrene 0.6 – 128 [b] Total estuarine PAHs 200 - 6000 [b]
Mean contamination of shellfish pg g^{-1} fresh wt				
	ΣdI-PCBs	ΣPCBi	(ΣPCDD/F)	ΣPAH
Oysters	0.324[a]	2700 [a]	0.272 [a]	39.0- 337 [e]
Mussels	0.334 [a]	3950 [a]	0.228 [a]	
Scallops	0.193 [a]	4977 [a]	0.199 [a]	
Regulatory thresholds [d]	Σ(PCDD/F+dl-PCB) 8.0 pg g^{-1} [d]		ΣPCDD/F 4.0 pg g^{-1} [d]	Benzo[*a*]pyrene 10.0 pg g^{-1} [d]
TDI (ng kg^{-1} bwt d^{-1}.) PTMI (pg kg^{-1} bwt $month^{-1}$)	ΣPCB 20 (Aroclor eq.) [c] ΣPCBi 10 [g]		0.001 – 0.004 [b] Σ(PCDD/F+ ΣdI-PCBs) 70 [f]	
Daily intake from food [c]			Σ(PCDD/F+dl-PCB): 1.8 pg WHO-TEQ kg^{-1} bwt d^{-1}	Σ(6 PAH): 1.4 ng WHO-TEQ /kg bwt/d
Toxicity Ecotoxicity	Endocrine disruptor neurotoxic, immunotoxic [b]		Chloracne [c] Immunodepressor[c] Carcinogen (2,3,7,8-TCDD) [c]	Endocrine disruptor Benzo[a]pyrene: carcinogen [c]. Less bioaccumulative and biomagnifying than organochlorines. Slow metabolisation in mussels / fish [b]
Status	Main applications banned in France (1987). Total end to use in 2010 [b]		Two decrees in 2002 on waste incineration – limit value 0.1 ng TEQ m^{-3} [e]	Decrees in 1999 limiting PAH emissions to 0.1 mg Nm^{-3} for boilers and engines [e]

a. CALIPSO study (Leblanc et al. 2006); b. OSPAR 2008; c. AFSSA 2008b; d. Regulation (EC) 1881/2006 (EC 2006c); e. INERIS (2010)Toxicology data sheets; f. JECFA (2001); g. AFSSA (2005).
PCBi: sum of PCBs 28, 52, 101,138, 153, 180 (AFSSA 2005).

Table 7 Levels of contamination in shellfish sampled from the marketplace for some unregulated organic contaminants

	TBT Organostannic compounds	HCB	Dieldrin	DDT /DDE /DDD Total DDT	Lindane α–, β–, γ–HCH	Toxaphene	Triazines Atrazine, simazine	Dichlorvos	Brominated flame retardants, , Polybromodiphenyl ethers (PBDEs)	Chlorinated paraffins
Sources	Agricultural processes, antifouling agents, fungicide agents,[b] waste tips [b].Current reservoirs: soil, sediments, public waste tips and old infrastructures, remobilisation of old sediments (dredging) [b] Direct inputs from rivers, atmosphere and ocean currents [b], professional and recreational nautical activities [c]		Environmental stocks [b]		Agriculture, wood treatment, veterinary medicine, domestic use [d] Control of crop pests, cattle parasites and commensal insects [d]	Pesticide, complex mixtures of organochlorines [b]	Pesticide [b]	Used on salmon farms: parasiticide against sea louse, insecticide – acaricide [b]	Automobile and aeronautic padding, textile and polymer additives [b], water purification, photography	Plastifiers, additives for metal working fluids, flame retardants, leather industry [b]
Mean levels in the environment										
Seawater (ng L^{-1}) Sediments (µg kg^{-1} dry wt)	0.6[c] 0.001 [c]	≈ 1 [b] 0.040 – 0 .070 [b]	0.005 – 0.05[b] 0.0005 – 0.005[b]	0.005 – 0.05[b] 0.0005 – 0.005[b]	0.0005 – 0.005[b]		< 2 - 42[b]		DeBDE 1 - 1700[b]	10[b]
Ecotoxicological evaluation criteria*										
(mg/kg fresh wt)						10[b]				
Mean contamination of shellfish										
Mussel (µg kg^{-1} dry wt) Mussel (µg kg^{-1} fresh wt)	1.1[g]			DDE 5 – 50[b]	≈ 1 [b]	Nd	nd	nd	nd	nd
ADI (ng kg^{-1} bwt d^{-1}.) or other TRV	Σ(organoSn) 250[f]	160 [b]	aldrin + dieldrin 0.0001 mg kg^{-1} bwt d^{-1}		0.005 mg kg-1 bwt d^{-1}		Atrazine :500 ng/kg bwt/d Simazine :520 ng kg^{-1} bwt/d^{-1}	80 ng kg^{-1} bwt d^{-1} [e] DL_{50} rat 17 – 80 mg kg^{-1} bwt [d] DL_{50} mouse 61-135mg kg^{-1} bwt [d] DL_{50} rabbit 10–12. mg kg^{-1} bwt [d] DL_{50} dog 100 mg kg^{-1} bwt [d]	No TRV exists [a] LOAEL octaBDE: 8 mg kg^{-1} bwt d^{-1} LOAEL pentaBDE 72 mg kg^{-1} bwt d^{-1}	
Daily intake from shellfish ingestion, adult men										
(ng ind^{-1} d^{-1})	Ratio (ng ind^{-1} d^{-1})/(ADI x 60) = 0.0034 [f]		Nd	nd	nd				Fish and seafood: 85 [b] 150[a]	
Risk category	T. N[a]				R23/24/25. R36/38 [d]			R24/25 [d]		
Toxicity/Ecotoxicity	TBT. Endocrine disrupter [a] TPT. toxic for reproduction and development [a] DBT. TBT. TPT: immunotoxic [a]				Neurological disorders [d]			Acetylcholinesterase inhibitor Mutagenic Carcinogenic Reprotoxic [d]	Endocrine disrupter Neurotoxic, Potentially carcinogenic [b]	
Physico-chemical properties and phenomena determining fate of contaminants					Poorly hydrosoluble. Highly soluble in organic solvents [d]			Poorly hydrosoluble. Soluble in organic solvents [d]	Highly lipophilic. Poorly hydrosoluble Adsorbs strongly to sediments[b]	
Status	Total ban since 01/01/08 [c]		Banned [b]	Banned [b]		Not used in OSPAR area [b]	Banned in France[b] Limited uses [b]			End of use for short-chain paraffins scheduled [b]

TBT: tributyltin. TPT: triphenyltin, DBT: dibutyltin; HCB: hexachlorobenzene, T: toxic, N: dangerous for environment DDT, dichlorodiphenyltrichloroethane, DDE, dichlorodiphenyldichloroethylene, DDD, dichlorodiphenyldichloroethane, HCH, hexachlorocyclohexane, nd: no
determined; R23/24/25: toxic by inhalation, skin contact, ingestion; R36/38: eye and skin irritant R24/25: toxic by inhalation, skin contact, ingestion

a. CALIPSO (Leblanc et al. 2006) ; b. OSPAR 2008; c. AFSSA 2006; d. INRS (2010)Toxicology data sheets; e. INERIS 2010; f. EFSA 2004b; g. Guérin et al. 2007

Table 8 Bioconcentration factors for chemical contaminants in bivalve molluscs. Source: James et al. (2006)

Substance	BCF in molluscs
Anthracene	260 (*Macoma*)
Cadmium	994 (invertebrates)
C10-13 chloroalkanes	40900 (mussels)
Chlorfenvinphos	255 (*M. galloprovincialis*)
Diethylhexylphthalate	2500 (mussels)
Endosulfan	600 (*Mytilus*)
Fluoranthene	10000 (*Crassostrea*)
Hexachlorobenzene	7000 (bivalves)
Haxachlorobutadiene	2000 (*Mytilus*)
Hexachlorocyclohexanes (lindane)	161 (mussels) 240 (*Mytilus*)
Lead	2279 (molluscs)
Mercury*	$10^6 – 10^7$
Naphtalene	27 – 38 (mussels)
Nickel	270 (bivalves)
Nonylphenols	3000 (mussels)
Octylphenols	634 (calculated)
Pentachlorobenzene	2000 (bivalves)
Pentachlorophenol	390 (*Mytilus*)
Benzo[*a*]pyrene	12000 (*Mytilus*)
TBTs	11400 (*Crassostrea*)
Trifluraline	2360 (*Helisoma*)
Aldrin	43560 (calculated)
Dieldrin	7760 (calculated)
Endrin	5250 (calculated)
Isodrin	43650 (calculated)
Total DDT	45600 (molluscs)

* bioamplification taken into account

Table 9 Example half-lives for chemical contaminants that exist in bivalve molluscs

Species	Chemical contaminant	Biological half-life (days)	Reference
Mytilus edulis	TBT	21 – 36	Yang *et al*. 2006
	TBT	69	Page *et al*. 1995
	DBT	115	Page *et al*. 1995
	Fluoranthene	30	Pruell *et al*. 1986
	Benzo[a]anthracene	18	Pruell *et al*. 1986
	Chrysene	14	Pruell *et al*. 1986
	Benzo[*e*]pyrene	14	Pruell *et al*. 1986
	Benzo[*a*]pyrene	15	Pruell *et al*. 1986
	Indenol[*1.2.3-cd*]pyrene	16	Pruell *et al*. 1986
	PCB 28	16	Pruell *et al*. 1986
	PCB 101	28	Pruell *et al*. 1986
	PCB 128	37	Pruell *et al*. 1986
	PCB 153	46	Pruell *et al*. 1986
	Zn	76	Bryan 1976
Mytulis galloprovincialis	Hg	1000	Bryan 1976
Mya arenaria	TBT	71 – 94	Yang *et al*. 2006
Gafrarium tumidum	Ni	35 ± 7	Hédouin *et al*. 2007
Venerupis decussata	TBT	17 – 38	Gomez-Ariza *et al*. 1999
Crassostrea gigas	Cu	11.6 – 25.1	Han *et al*. 1993
	Zn	16.7 – 30.1	Han *et al*. 1993
	Cd	137	Geffard *et al*. 2002
	Cu	430	Geffard *et al*. 2002
	Hg	44	Bryan 1976
	Zn	335	Geffard *et al*. 2002
	Zn	255	Bryan 1976
Crassostrea virginica	Fluoranthene	26 – 32	Sericano *et al*. 1996
	Pyrene	10 – 12	Sericano *et al*. 1996
	Benzo[*a*]anthracene	13 – 15	Sericano *et al*. 1996
	Chrysene	12 – 16	Sericano *et al*. 1996
	Benzo[*e*]pyrene	12 – 16	Sericano *et al*. 1996
	Benzo[*a*]pyrene	9 – 10	Sericano *et al*. 1996
	Indenol[*1.2.3-cd*]pyrene	10 – 11	Sericano *et al*. 1996
	PCB 26	22	Sericano *et al*. 1996
	PCB 118	73 – 299	Sericano *et al*. 1996
	PCB 149	130 - > 365	Sericano *et al*. 1996
	PCB 153	51 – 102	Sericano *et al*. 1996
Ostrea edulis	Zn	890	Bryan 1976
Crassostrea belcheri	Cd	5 – 16	Lim *et al*. 1998
	Cu	5 – 9	Lim *et al*. 1998
	Pb	4 – 14	Lim *et al*. 1998
Crassostrea iredaleii	Cd	4	Lim *et al*. 1998
	Cu	6	Lim *et al*. 1998
	Pb	6	Lim *et al*. 1998
Isognomon isognomon	Ni	Infinite	Hédouin *et al*. 2007

Table 10 The RNO/ROCCH monitoring program for the various conventions and directives (Water Framework Directive (WFD), Oslo and Paris convention (OSPAR), Barcelona convention (MED POL), and for the French Directorate General for Food (DGAL), with regard to water, biota and sediment

RNO (1979-2007)

Sampling frequency			
Conventions / Directives	**Water**	**Biota**	**Sediment**
OSPAR & Barcelona		Annual, in November (at all 80 RNO sites)	Every 10 years (entire French coast)
DGAl		Annual, in February (at all 80 RNO sites)	
RNO contaminants (1979 – 2007)			
Metals	Cadmium (Cd), copper (Cu), mercury (Hg), silver (Ag), chrome (Cr), nickel (Ni), lead (Pb), vanadium (V), zinc (Zn)		
Organochlorines	DDT, DDD, DDE, lindane (γ-HCH), α-HCH, polychlorobiphenyls: indicator PCBs (28, 52, 101, 138, 153, 180) and dioxin-like PCBs (105, 118, 156)		
Polycyclic hydrocarbons (PAHs)	Naphtalene, mono-, di-, tri- and tetra-methyl naphthalenes, acenaphtylene, acenaphtene, fluorene, mono and di methyl fluorenes, phenantrene, anthracene, mono, di and tri methyl phenanthrenes/anthracenes, fluoranthene, pyrene, mono and di methyl pyrenes/fluoranthenes, benzo[a]anthracene, triphenylene, chrysene, mono and di methyl chrysene, benzofluoranthenes, mono methyl benzofluoranthenes, benzo[e]pyrene, benzo[*a*]pyrene, perylene, dibenzo[*a,h*]anthracene, benzo[*g,h,i*]perylene, indeno[*1,2,3-cd*]pyrene, sulphurated heterocycles: dibenzothiophene, mono, di and tri methyl dibenzothiophene, benzonaphtothiophenes, monomethyl benzonaphtothiophenes		

ROCCH (since 2008)

Sampling frequency			
Conventions / directives	**Water**	**Biota**	**Sediment**
WFD	Monthly for 12 months every 6 years (at all WFD sites)	Annual in November (at 25% of WFD sites)	Every 6 years (at 25% of WFD sites)
OSPAR & Barcelona		Annual in November (at 50% of WFD sites)	Every 6 years (at 50% of WFD sites)
DGAL		Annual in February, Cd, Hg, Pb (on 131 sites)	
ROCCH contaminants (WFD + OSPAR + DGAL)			
Metals	Cadmium (Cd), mercury (Hg), nickel (Ni), lead (Pb)		
Organic contaminants	Polychlorobiphenyls: indicator PCBs (28, 52, 101, 138, 153, 180) and dioxin-like PCBs (105, 118, 156) Alachlore, anthracene, atrazine, benzene, pentabromodiphenylether, octa-bromodiphenylether, deca-bromodiphenylether, C10-13 chloroalkanes, chlorfenvinphos, chlorpyrifos, 1,2 dichloroethane, dichloromethane, di (2-ethylhexyl)phtalate (DEHP), diuron, endosulfan (family), fluoranthene, hexachlorobenzene, hexachlorobutadiene, hexachlorocyclohexane (alpha, beta, delta), lindane, isoproturon, naphthalene, nonylphenols, 4-n-nonylphenol, para-nonylphenols, octylphenol, para-ter-octylphenol, pentachlorobenzene, pentachlorophenol, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*g,h,i*]perylene, benzo[*k*]fluoranthene, indeno[*1,2,3-cd*]pyrene, simazine, tributyltin, trichlorobenzene, 1,2,4-trichlorobenzene, trichloromethane (chloroform), trifluralin, aldrin, carbon tetrachloride, total DDT, para-para DDT, dieldrin, endrin, perchloroethylene (tetrachloroethylene), trichloroethylene, isodrin,		

Table 11 Concentrations of certain contaminants (fresh wt)* observed in oysters from Arcachon Bay (RNO survey 02/2000-11/2005), and in mussels (Devier *et al*. 2005)

Contaminant	Oysters (mean ± s.d.) (n)	Mussels (min-max of means depending on site) (n)
Inorganic (mg kg^{-1} fresh wt)		
Cadmium	0.23 ± 0.09 (54)	0.14– 0.18 (84)
Lead	0.18 ± 0.04 (54)	0.25 – 0.31 (84)
Mercury	0.03 ± 0.01 1 (54)	nd
Arsenic	Nd	2.5 – 2.9 (84)
Nickel	0.21 ± 0.04 (18)	0.20 – 0.25 (84)
Chrome	0.17 ± 0.08 (42)	0.23 – 0.34 (84)
Vanadium	0.33 ± 0.12 (18)	nd
Copper	24.51 ± 9.62 (54)	1.1 – 4.1 (84)
Zinc	372 ± 112 (54)	28 – 42 (84)
Selenium	Nd	1.6 – 2.2 (84)
Silver	0.79 ± 0.33 (18)	nd
Organic (pg g^{-1} fresh wt)		
Organostannics (amount in Sn)	Nd	7.2 – 394 10^3
PCBs (sum of 6 congeners)	5.2 10^3± 3.6 10^3 (21)	5.4 – 7.0 10^3
PAHs (amount EPA PAHs)	40 10^3± 11 10^3 (15)	13.3 – 262 10^3
DDT / DDE / DDD (sum of the 3)	2.3 10^3± 1.3 10^3 (24)	nd
Lindane (α-,γ-HCH) (sum of the 2)	0.23 10^3 ± 0.10 10^3 (24)	nd

* fresh weight obtained by multiplying dry weight value by 0.18; nd: not determined
n:number of samples. nd: not determined.

Table 12 Concentrations of certain contaminants (mg kg^{-1} fresh wt) observed in mussels from the Bay of Seine (RNO survey from 2003 to 2007)

Contaminant	Mean ± s.d.	Sample size
Cadmium	0.23 ± 0.09	48
Lead	0.49 ± 0.26	48
Mercury	0.04 ± 0.02	48
Benzo[*a*]pyrène	3.01 ± 4.10 10^{-3}	24

Table 13 Summary of cadmium non-compliances in reports from DGAL monitoring programs (2002-2005)

Scallops	Year of DGAL monitoring program	Fishing area	Cadmium test result (mg kg^{-1} fresh wt)	Cadmium confirmation result (mg kg^{-1} fresh wt) (AFSSA/LERQAP)
Chlamys varia	2005 Total no. of scallop samples = 14	Pertuis Breton	1.18	*1.26 ± 0.18
		Pertuis Breton	1.65	**1.64 ± 0.23**
		Pertuis Breton	1.12	1.05 ± 0.21
		Pertuis Breton	1.12	1.07 ± 0.21
		Pertuis Breton	1.40	1.07 ± 0.21
		Quiberon Bay	1.54	**1.62 ± 0.23**
		Quiberon Bay	1.5	**1.56 ± 0.22**
		Quiberon Bay	1.06	1.13 ± 0.16
Aequipecten opercularis	2004 Total no. of scallop samples = 3	Western Channel	1.13	1.33
Chlamys varia	2002 Total no. of scallop samples = 9	Not specified	1.5	1.7
			1.6	1.7

*Bold type: samples confirmed as non-compliant with Regulations (EC) 1881/2006 (EC 2006c)

Table 14 Daily human consumption (g d^{-1} of product consumed) according to the 2007 INCA 2 survey (INCA 2 2009)

	Adults (normal estimators) (N = 1918; aged 18 and older)				Children (N= 1455; aged 3-17)			
	% of Consumers	Mean	Standard deviation	Median	% of Consumers	Mean	Standard deviation	Median
Meat	92.0	49.7	37.5	42.4	91.5	38.1	28.8	32.9
Fish	79.3	26.5	24.7	21.2	78.7	18.3	17.6	14.3
Molluscs and crustaceans	33.5	4.5	9.3	0	17.9	1.4	5.1	0

Table 15 Detail of mollusc consumption by "high consumers" of seafood in the CALIPSO survey (Leblanc et al. 2006). Data given in g week^{-1} of fresh flesh

Molluscs	Mean (g week^{-1})	P5	P50	P95
Bivalves	119.7	7.5	79.8	350.3
Clam	0.2	0	0	0
Cockle	3.1	0	0	15.0
King scallop	39.3	0	25.0	156.3
Razor clam	0.4	0	0	0
Oyster	34.4	0	18.0	144.0
Mussel	22.5	0	17.5	70.0
Palourde clam	2.8	0	0	12.3
Other scallops	14.7	0	0	56.3
Warty venus	1.5	0	0	7.5
Wedge-shell, olive	0.3	0	0	0
Queen scallop	0.5	0	0	0
Gastropods	21.2	0	3.8	87.5
Winkle	4.2	0	0	25.0
Whelk	15.4	0	0	75.0
Abalone	0.6	0	0	0
Limpet	1.0	0	0	0
Echinoderms	11.6	0	0	52.5
Sea urchin	11.6	0	0	52.5
Tunicates	1.0	0	0	0
Sea-squirt	1.0	0	0	0
All	153.5	10.0	106.1	413.5