

TOXIC CHEMICAL CONTAMINANTS: REVIEW

STATE OF THE GULF OF MAINE REPORT

COMPANION DOCUMENT TO TOXIC CHEMICAL CONTAMINANTS THEME PAPER



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The Gulf of Maine Council on the Marine Environment was established in 1989 by the Governments of Nova Scotia, New Brunswick, Maine, New Hampshire and Massachusetts to foster cooperative actions within the Gulf watershed. Its mission is to maintain and enhance environmental quality in the Gulf of Maine to allow for sustainable resource use by existing and future generations.

1. Issue in Brief

The GULF of MAINE IS BEING SUBJECTED TO AN EVER-INCREASING NUMBER of chemical contaminants that are being introduced by human activities both locally and globally. Some of these contaminants make seafood unsafe for human consumption, are quite toxic to coastal organisms and are suspected of altering ecosystem composition and functioning (Figure 1).

Humans first began to noticeably alter the planet by clearing the forests, cultivating the soil, sluicing the salt marshes or otherwise modifying the landscape. This led to increased soil erosion, leaching of heavy metals to the sea and increased siltation of estuaries, coastal habitats and basins. This stage was quite recent in the Gulf of Maine region, starting with the Acadian and New England settlers in the 1600s. Second, the invention of the steam engine in the 1770s culminated in the industrial revolution fueled by fossil fuels, first coal followed by oil and gas (Steffen et al. 2007). This introduced not only the greenhouse gases carbon dioxide (CO_2), methane (CH_3) and nitrous oxide (N_2O) to the atmosphere, but also contributed toxic organic hydrocarbons, such as polycyclic aromatic hydrocarbons (PAHs), that precipitate, settle or partition to the both terrestrial and aquatic environments. Third, a chemical industrial revolution followed in the middle of the twentieth century with a plethora of synthetic chemicals being created and produced for use in almost every human activity, from manufacturing, agriculture, aquaculture, and forestry to medicine and cosmetics.

Polycyclic aromatic hydrocarbons, organochlorines, organometals and metals are all well-documented contaminants of concern in the Gulf of Maine. Various research groups over the past thirty years have documented this contamination most comprehensively with tissue analysis of marine mammals and mussels. There are many more categories of contaminants that are known to be of environmental concern such as organobrominated compounds, perfluorinated compounds, organophosphates, pyrethroids, antibiotics, other pharmaceuticals, and steroidal hormones; however, there is limited information on them in the Gulf of Maine. Some contaminants are known to be highly toxic or disruptive to the normal physiology of marine organisms which results in species depletion and changes to the ecosystem (Johnston and Roberts 2009). Not surprisingly, some species are evolving a resistance to certain toxicants in contaminated areas. The evolution of resistance is more rapid the simpler the organism, such as bacteria, which is partly a reflection of their shorter generation times (Grimes et al. 1984; Barkovskii et al. 2010). An increased resistance and/or reduced genetic diversity have been found in benthic invertebrates living in areas heavily contaminated with hydrocarbons and metals (Klerks and Weis 1987; Levinton et al. 2003; Ross et al. 2002; Street and Montagna 1996). However, aquatic communities exposed to contaminants are universally reduced in biodiversity (Johnston and Roberts 2009) and resilience to further perturbations (Hooper et al. 2005). An additional concern is that a number of the organochlorines (OCs) and methylmercury (MeHg) have the additional property of incrementally increasing in concentration at each level of the marine

LINKAGES

See also the following theme papers in the State of the Gulf of Maine Report:

- Microbial pathogens and toxins
- Eutrophication
- Climate change and its effects on ecosystems, habitats and biota
- Climate change and its effects on humans
- Emerging Issues
- Toxic chemical contaminants

food chain (biomagnification), with the OCs and MeHg residing in the lipids and proteins of the organisms, respectively. Alternately, some metals and polyaromatic hydrocarbons (PAHs) have been found to decrease up the trophic chain (Wang 2002, Campbell et al. 2005, Wan et al. 2007). Knowing the physicochemical properties of these contaminants and their toxicity has enabled the prediction of their potential to both bioaccumulate and alter the composition of the ecosystem (Mackay 1982; McCarty and MacKay 1993; Mackay et al. 1999), thus affecting the overall health and functioning of the ecosystem.

There are many thousands of synthetic chemicals that have purposely or inadvertently been introduced to the environment by various human activities. Municipal wastewater chemicals are not well understood and individual standards to protect human health and aquatic life have not been established. The scientific community has always been "playing catch-up" to determine the consequences of specific contaminants, as in the approximately 20-year lag between widespread application and restriction of the use of DDT and other organochlorines. There are many contaminants, both known and unknown, present at any time and place that standard toxicity tests cannot possibly be used to attribute effects (Emlen and Springman 2009; Hartung 2009). The possibility of establishing a cause and effect in the environment, therefore, is often not practical, because all the contaminants, their effects and/or their interactions are not known. The presence and potential interactions of individual contaminants is cause for concern. The environmental and human health risk of a number of individual contaminant categories has resulted in national and international restrictions on their production and use. As a result the use of some of these contaminants has been discontinued; however, many of these same chemicals will remain in the environment for quite some time. A huge challenge lies in the future to not only establish the individual toxicity of an ever-increasing number of environmental contaminants but to evaluate the interactive effects of a wide variety of chemical contaminants on the community or ecosystem level (Rohr et al. 2006).

This document provides a review of the literature related to toxic chemical contaminants in the Gulf of Maine and organizes it according to the Driving Forces-Pressures-State-Impacts-Responses framework (DPSIR, Figure 1). It is intended to be a detailed background document for the Toxic Chemical Contaminants theme paper for the State of the Gulf of Maine Report.



Figure 1: Driving forces, pressures, state, impacts and responses (DPSIR) to toxic contaminants in the Gulf of Maine. In general, the DPSIR framework provides an overview of the relation between different aspects of the environment, including humans and their activities. According to this reporting framework, social and economic developments and natural conditions (driving forces) exert pressures on the environment and, as a consequence, the state of the environment changes. This leads to impacts on human health, ecosystems, and materials, which may lead to societal or government responses that feed back on all the other elements.

Definitions of Terms

Bioaccumulation – the uptake and accumulation of a contaminant or element by an organism from food or seawater at a greater rate than that by which the substance is lost.

Bioconcentrate - the uptake of a contaminant directly from seawater.

- Biodilution the decrease in the concentration of an element or contaminant with each subsequent trophic level (step in the food chain).
- Biomagnification the increase in the concentration of a contaminant or element with each subsequent trophic level (step in the food chain). This means that consumers near the end of the food chain have higher levels of the element or contaminant than producers or consumers lower on the food chain.
- Contaminant any element or compound introduced into the environment by human activity.
- Endocrine disrupter an endocrine disrupter is an external chemical that has the ability to enter and deceive the endocrine system of an organism by altering its normal hormone synthesis.
- Metabolize a process by which organisms use and break down various substances they have ingested or absorbed.

ppb – parts per billion, expressed as ng/g dry or wet weight or μ g/L.

ppm – parts per million, expressed as μ g/g dry or wet weight or mg/L.

Pollutant – any contaminant that is known to be toxic to organisms.

Toxicant - a toxic substance made by humans or created by human activity.

Toxic – causes damage to organisms.

Trophic levels – the succession of steps from producers to ultimate consumer in a food chain.

2. Driving Forces and Pressures

The ever-increasing human population, with its demands for more food and improved life style, has resulted in a multitude of chemical contaminants, both natural and synthetic, being introduced into the Gulf of Maine. Some of these are released locally within the Gulf of Maine and its watershed, whereas many are semi-volatile and brought in atmospherically from distant industrial, urban and agricultural centres. The chemical composition of coastal ecosystems is continuously changing both seasonally due to environmental conditions and geographically due to differences in habitat characteristics. The future fate of an introduced chemical is determined by its persistence in the environment, solubility in seawater, the strength of its adsorption to particulate surfaces, including life forms, and its absorption into living organisms. Compounds with a low solubility in seawater (hydrophobic) enter into a complex flux, or exchange, between depositional, resuspension, lateral removal and degradation processes (Figure 2, Table 1).



Figure 2: An illustration of the transport and processes involved in contaminant flux or exchange over the estuarine, coastal and shelf regions of the Gulf of Maine.

2. Driving Forces and Pressures

Table 1: Primary sources of the major chemical contaminant groups in the Gulf of Maine as determined by this review, including past usage. The number of + signs indicates the degree to which each source contributes to levels of contaminants in the Gulf of Maine, with + indicating a small contribution and +++ a major contribution. The – sign indicates that the source does not contribute to levels of that contaminant.

					SO	URCES		
		NATURAL	URBAN	INDUSTRIAL	Shipping/ Harbours	Agriculture: Animals	Agriculture & Silvaculture: Crops	AQUACULTURE
	PAHs	+	+++	+++	+	-	-	-
	PCBs	-	++	+++	+	-	-	-
	CHBs	-	-	-	-	-	+++	-
	PCDDs/PCDFs	+	++	+++	-	-	-	-
	∑DDT	-	+	-	-	-	+++	-
ts	CBs	-	+	+	-	-	++	-
	HCHs	-	+	-	-	-	+++	-
	CHLs	-	+	-	-	-	+++	-
	Mirex	-	+	-	-	-	+	-
nar	Aldrin/Dieldrin	-	+	-	-	-	+++	-
Ĩ	OBs	-	+++	++	-	-	-	-
onto	PFCs	-	+++	++	-	-	-	-
0	OPs flame retardants	-	+++	++	-	-	-	-
	OPs pesticides	-	+	-	-	-	+++	+++
	Pyrethroids/pyrethrins	+	+	-	-	-	+++	+++
	Antibiotics	+	+++	+	-	+++	-	+++
	Pharmaceuticals	-	+++	-	-	+++	-	-
	Steroidal hormones	+	+++	-	-	+++	-	-
	Butyltins	-	-	-	+++	-	-	-
	Mercury/MeHg	+	++	+++	++	-	-	+
	Trace metals	+	+	++	-	-	+	+

(PAHs are polyaromatic hydrocarbons; PCBs are polychlorinated biphenyls; CHBs are polychlorinated bornanes; PCDDs/PCDFs are dioxins and furans; ∑DDT is total DDT; CBs are chlorobenzenes; HCHs are hexachlorocyclohexanes; CHLs are chlordanes; OBs are organobromine compounds; PFCs are perfluorinated compounds; OPs are organophosphates).

2.1 NATURAL

A number of chemical elements and compounds, also known to be contaminants, are also introduced into the Gulf of Maine through natural processes. The weathering of continental crust involves the dissolution of the earth's elements, such as the metals (e.g., mercury), their transfer in the hydrosphere via groundwater to stream to river to estuarine to coastal environments and beyond. This process created the natural baseline in marine sediment layers that predated the European arrival that drastically altered both the terrestrial and aquatic environments. This has been documented for mercury from the sedimentary record in the bottom mud of Passamaquoddy Bay (Sunderland et al. 2010). Natural hydrocarbon seeps occur over fossil petroleum deposits and recent organic deposits. Natural petroleum seeps are not known for the Gulf of Maine, although hydrocarbon seeps are expected to occur frequently in the biologically productive estuarine habitats of the Gulf of Maine. Forest and field fires and volcanic activity result in the production and atmospheric transport of polycyclic aromatic hydrocarbons (PAHs), dioxins and furans, and also metals like mercury.

The hydrosphere is the total mass of water on the planet. The water cycle, also known as the hydrologic cycle, is the movement of water around, over, and through the Earth, including its transormation into the different states of liquid (water), air (water vapour) and solid (ice).

2.2 ANTHROPOGENIC

Human activities have accelerated the natural processes that introduce chemicals into the marine environment and have also introduced new, man-made contaminants. Disturbances by humans, such as the removal of flora/fauna and soil during construction projects or mining, can expose underlying rock to accelerated dissolution. Much of the northern and northeastern watersheds of the Gulf of Maine are granitic, with little buffering capacity, or sedimentary shales and schists that are highly acidic on exposure. This acidity results in the dissolution of metals that are then dispersed in the groundwater and can influence the estuarine and river mouth environments and potentially can alter the resident biological community. Clearcutting exposes the forest floor to soil erosion without the binding action of roots so that soils and nutrients are lost downstream. Clear cutting also increases the vulnerability of the dry slash to fire in the northern Gulf of Maine and this releases other contaminants to the atmosphere.

Humans are synthesizing and producing chemicals at an ever-increasing rate such that thousands of them have been intentionally or inadvertently released to the environment (Muir and Howard 2006). Manufactured or produced chemical contaminants are released into coastal waters during agricultural, industrial and domestic activities through surface runoff, effluent and sewage outfalls, surface spills, and atmospheric fallout (Figure 2). It is difficult to determine which route a specific contaminant follows because it is determined by both the local and distant source(s). Furthermore, no two contaminants behave the same in the atmosphere, hydrosphere or biosphere. High nutrient levels from agricultural practices that reach estuarine and coastal waters result in increased primary production and biomass, a process known as eutrophication (see Eutrophication theme paper). This, in turn, can result in increased uptake of hydrophobic pollutants that are consumed by the pelagic food chain (plants and animals that live in the water column, from plankton to fish and whales), or ultimately settle to the bottom, either as senescent (aging) cells or in fecal pellets, and enter the benthic or bottom-living food chain (Dachs et al. 2000; Hjort et al. 2008; Dueri et al. 2009). Conversely, extended eutrophication events could cause biodilution by reducing contaminant concentrations in phytoplankton at the base of the food web (Cox 1970). Also, urban sewage outfalls and aquaculture operations contribute high organic loads at local scales that combine increased microbial activity in the presence of antibiotics, pesticides and a variety of other contaminants.

2. Driving Forces and Pressures

Many contaminants enter the Gulf of Maine watershed primarily through atmospheric transport and deposition. The organochlorine and organobromine compounds and mercury are known to gradually drift towards the poles by atmospheric transport through a repeated process of evaporation and precipitation (Wania and MacKay 1993; Jurdao and Dachs 2008). Pesticides that have been banned in North America for 40 years but used in Central America, such as undegraded DDT, appear in present-day mussel monitoring programs in the Gulf of Maine (Jones et al. in prep). An array of chemicals, such as heavy metals, polycyclic aromatic hydrocarbons, alkanes and other hydrocarbons and one of the most toxic chemical groups known, the polychlorinated dibenzodioxins and furans (PCDD/DFs) are produced by combustion of fossil fuels for urban and industrial power, and by other human activities such as pulp and paper mills, chemical manufacturing, petroleum refining and metal smelting. This atmospheric input is inclusive of industrial stacks, domestic furnaces and transportation. The more troublesome chemicals, because of their toxicity and persistence, are those that have been created as pesticides and for high-pressure industrial use. The organohalides (e.g., PCBs, PFCs, PBDEs) are of particular concern because of their persistence, bioaccumulation and extreme toxicity in the environment.

3. Status and Trends

3.1 POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds that are present in the environment as a result of incomplete combustion of organic material. They result both naturally from forest fires and from the burning of fossil fuels for energy. These compounds are of particular concern because of their widespread presence, toxicity of certain compounds and threat to human health (Bostrom et al. 2002; Santodonato 1997; Villeneuve et al. 2002). There is plenty of evidence for contamination of the sediments in the coastal bays of the Gulf of Maine (Johnson and Larsen 1985; Kennicutt et al. 1994; Windsor and Hites 1979). Coastal areas have ten times greater PAH concentrations than the deep basins of the Gulf of Maine due to their proximity to runoff from land. The PAH concentrations found in deep waters are mainly from atmospheric fallout with minor oceanic transport of resuspended sediments from contaminated coastal embayments (Windsor and Hites 1979; Larsen et al. 1986; Gustafson et al. 1998). The lower molecular weight PAHs are more volatile and widely distributed in the atmosphere (Brun et al. 2004). Mussels are known to rapidly take up PAHs in the vicinity of contaminated sediments and therefore bioaccumulate the higher molecular weight compounds (Pruell et al. 1986). The highest concentrations of PAHs in biota have been found in mussels off Massachusetts; these concentrations decreased northwards towards New Brunswick and Nova Scotia (Chase et al. 2001; Table 2). There was no overall Gulf-wide temporal trend observed between 1993 and 2008 in mussel levels of PAH, although there was an increasing trend for the combined New Hampshire sites. These results do not correspond to the steady decline observed in the atmospheric deposition of PAHs between 1985 and 2000 at the Kejimkujik weather station in southern Nova Scotia (Brun et al. 2004). This discrepancy between the recent decline in atmospheric input of PAHs in the Gulf of Maine region and the lack of a corresponding trend in the mussel tissues over the same period could be explained by the fact that the weathered higher molecular weight compounds dominate the coastal regions, as reflected in the sediments (Larsen et al 1983; Kennicutt et al 1994), whereas the atmospheric PAHs are the more volatile lighter weight compounds (Brun et al. 2004). A National Marine Fisheries Service survey of fishes conducted in 1980, which included Cape Cod Bay and Georges Bank, reported PAH concentrations 100 times lower than that found in coastal mussel tissues, which could be a reflection of proximity to source (Boehm and Hirtzer 1982). However, PAHs are known to biodilute in marine food chains from phytoplankton/seston to zooplankton, fish, and seabirds (Wan et al. 2007).

Table 2: Percentage of Gulf of Maine Gulfwatch sites within each state/province with low, medium and high organic contaminant concentrations in blue mussels (Jones et al. in prep). Contaminants were grouped into three categories—low, medium and high—using cluster analysis, so that "low" and "high" means low and high relative to the other levels measured.

		AVERAGE-ALL CHEMICALS	Pest	tDDT	tChl	Dield	PCBs	PAHs
Mass.	Low	1%	0%	0%	0%	7%	0%	0%
	Medium	32%	21%	36%	29%	21%	14%	71%
	High	67%	79%	64%	71%	71%	86%	29%
N.H.	Low	6%	8%	8%	8%	8%	0%	0%
	Medium	71%	75%	75%	58%	83%	75%	58%
	High	24%	17%	17%	33%	8%	25%	42%
Maine	Low	31%	22%	22%	39%	39%	28%	33%
	Medium	55%	67%	56%	50%	50%	61%	44%
	High	15%	11%	22%	11%	11%	11%	22%
N.B.	Low	21%	0%	13%	25%	25%	13%	50%
	Medium	69%	88%	75%	75%	50%	88%	38%
	High	10%	13%	13%	0%	25%	0%	13%
N.S.	Low	67%	91%	82%	64%	36%	82%	45%
	Medium	29%	9%	18%	36%	55%	18%	36%
	High	5%	0%	0%	0%	9%	0%	18%

(PEST is sum of all pesticides, tDDT is total DDT, tCHL is total chlordanes, Dield is dieldrin, PCBs are polychlorinated biphenyls and PAHs are polyaromatic hydrocarbons).

3.2 ORGANOCHLORINES (OCS)

An organochlorine is an organic compound containing at least one covalently bonded chlorine atom, which makes them more highly persistent in the environment. The OCs considered here are those synthesized for either their toxicity as pesticides or stability at high temperatures for industrial uses.

Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls (PCBs) are a man-made mixture of 140 to 150 chlorinated biphenyl compounds of a possible 209, known as congeners. PCBs were originally recognized for their application as heat and pressure resistant lubricating oils in electrical capacitors and transformers in the 1920s (Cairns et al 1986; Frame et al. 1996). It has been estimated that 1.2 million tonnes have been produced worldwide (Harrad et al. 1994). PCB levels have declined in the atmosphere (Rapaport and Eisenreich 1988; Simcik et al. 2000) and biota (Addison and Smith 1998; Braune et al. 2007) since the restriction of their use in the 1970s. However, it has been estimated that only 30% of the PCBs produced had dispersed into the environment by the mid-1980s and were still being released from disposed and stockpiled equipment (Tanabe 1988). PCBs remain the most abundant organochlorine in the marine sediments (Schneider et al. 2007) and biota (Skarphedinsdottir et al. 2010). The sediments of Casco Bay, Maine, have been surveyed for PCBs several times, most recently by Wade et al (2008) who compared their results of 2000/2001 to those of an earlier survey in 1991(Kennicutt et al. 1994). PCB levels at the inner bay region around Portland Harbor had declined slightly whereas in the outer bay the decline was twofold. This decline in the outer bay is probably due to resuspension and dispersion of particles towards the unprotected mouth of Casco Bay. Larsen et al (1985) had earlier reported that PCB concentrations in the deep basins of the Gulf of Maine declined with distance from Boston Harbor. Wilkinson Basin was more contaminated than either Georges Basin or Jordan Basin, being immediately downstream from Boston. The sediments in the estuaries along the east coast of the United States contain a large proportion of the PCBs present in the marine environment and these PCBs are readily available for desorbtion or solution in seawater after resuspension by storms (Schneider et al. 2007).

There was no temporal trend in PCB congeners in blue mussels in the Gulf of Maine from canned mussels analyzed from the 1940s and compared to the values obtained by the NOAA Mussel Watch collections in 1988 and 2007 (Apeti et al. 2010). A distinct spatial trend occurs in PCB concentrations with a tenfold decline from the southwest in Massachusetts to the northeast in Nova Scotia (Table 2, Figure 3). Vorkamp et al. (2010) reported that Boston Harbor had the highest blue mussel PCB concentration in a global bivalve survey in 2007. An assessment survey of demersal (near bottom-living) fishes between 1999 and 2001 found that PCBs were the most frequent organochlorine contaminant in the Northeast United States, being detected at 44% of the sites surveyed (Harvey et al. 2008). The eggs of four species of seabirds nesting in the Bay of Fundy were found to have similar low ppm wet weight PCB levels in the 1970s, despite their diverse feeding habits from fish-eating cormorants to benthic, mollusc-feeding eiders (Pearce et al. 1979).

Westgate et al. (1997) re-sampled the blubber of harbour porpoises from the Bay of Fundy and Jeffreys Ledge, Gulf of Maine, between 1989 and 1991 and found the levels had decreased by four times since an earlier study conducted between 1971-77 (Gaskin et al. 1983). PCBs are the prevalent OC in the blubber of harbour seals in the Gulf of Maine and levels had also declined about sevenfold between the 1970s and 1990s but thereafter remained stable suggesting an equilibrium has been reached (Gaskin et al. 1973; Lake et al. 1995; Shaw et al. 2005; Park

3. Status and Trends



Figure 3: Average polychlorinated biphenyl (PCB) concentrations (ng/g dry weight) from the Gulf of Maine mussel monitoring (Gulfwatch) program between 2000 and 2009 (ESIP 2012).

et al. 2009). The prevalence of PCBs in the biota in the Gulf of Maine, combined with their persistence and toxicity, make them a present day concern despite the restriction and ban on their use in the 1970s.

Dioxins (polychlorinated dibenzo-p-dioxins; PCDDs) and Furans (polychlorinated dibenzofurans; PCDFs)

Some of the 210 congeners of PCDD/PCDFs are considered to be the most toxic man-made chemicals. They were never produced intentionally but are present everywhere both naturally as a combustion byproduct and as a result of industrial activities (Fletcher and McKay 1993). Agent Orange, a herbicide containing dioxin, was sprayed on forests in the Gulf of Maine watershed near Gagetown, New Brunswick in the 1960s to 70s (Wikipedia 2012). PCDDs and PCDFs have been measured in sediments of Casco Bay, Maine (Wade et al. 2008). The PCDD concentrations within Portland Harbor were slightly higher than at the mouth of the Casco Bay, indicating an urban source for PCDDs but not PCDFs. PCDD/ PCDF concentrations in aquaculture salmon raised at Maine and New Brunswick sites were low with only 20% of the fish found to have levels above the analytical detection limit (Shaw et al. 2006). Trace amounts of PCDDs and PCDFs were detected in the blubber of a few harbour seals from the coast of Massachusetts (Lake et al. 1995). Shaw et al. (2007) reported extremely low PCDD/PCDF concentrations in the Gulf of Maine female and pup harbour seals, which may reflect a species specific degradation capacity. Bottlenosed whale samples, from the mass mortality of 1987/88 off the Atlantic coast of the United States, were

found to have only six PCDD/PCDF congeners at levels slightly over the analytical detection level (Kuehl et al. 1991). The cause of this stranding is unknown but not thought to be due to the presence of PCDD/PCDFs. The discrepancy between PCDD/PCDF and PCB concentrations at the higher levels of the marine food web, given their chemical similarities, appears to be due to the ability of organisms to metabolize PCDD/PCDFs and thereby reduce their body burdens (Pruell et al. 2000; Shaw et al. 2007).

DDT and its metabolites (total DDT is the sum of all six isomers)

DDT, dichlorodiphenyltrichloroethane, was the first organochlorine compound discovered to have insecticidal properties as a neurotoxin in 1939 (Brooks 1974). It replaced pyrethrum by the 1950s and became the panacea for insect control in relation to agriculture, forestry and human health (malaria control). By the mid-1960s, it was discovered that DDT and its persistent degradation products p,p'-DDE and p,p'-DDD were resistant to further breakdown and implicated in the global decline of raptorial bird populations, particularly fish eaters, by interfering with eggshell formation (Ratcliffe 1958; Lundholm 1997).

Apeti et al. (2010) documents the first entry of DDT into marine organisms of the Gulf of Maine by analyzing mussels canned in the late 1940s. Both Gulfwatch and NOAA Mussel Watch monitoring programmes observed an elevated level of total DDT in blue mussels in the late 1980s to early 1990s with a decline in the 90s, followed by levelling out of concentrations at low levels in the first decade of this century (Apeti et al. 2010; Jones et al. in prep). The earlier DDT high values obtained between 1968 and 1970 from the United States national mussel program at Small Point, Maine (Butler 1973) suggest an exponential decline has occurred since the North American ban was imposed by 1972 (Apeti et al. 2010; Jones et al. in prep). Present day DDT concentrations in blue mussels decrease from the southwest to northeast regions of the Gulf of Maine, which suggests an important urban contribution (Table 2). This is surprising considering the large quantities of DDT sprayed aerially on the boreal forests of Maine and New Brunswick starting in 1958 and continuing into the 1960s (Dimond and Owen 1996). A similar decline of DDT concentrations in harbour porpoise has been documented in Gulf of Maine (Gaskin et al. 1971, 1982; Westgate et al. 1997). DDT also has declined significantly in harbour seals since the 1970s, however, the decline had slowed or reached an equilibrium by the 1990s (Gaskin et al. 1973; Shaw et al. 2005).

DDT levels in eggs of seabirds nesting in the Bay of Fundy during the 1970s were in the low ppm wet weight range, with fish-eating cormorants having greater concentrations than benthic-foraging eiders (Pearce et al. 1979).

In general DDT levels have declined in various biota by the turn of the century from the high values measured in the 1970s. Nevertheless, the levels of total DDT

in biota from the Gulf of Maine are relatively high decades after the pesticide's introduction in the 1950s because of the persistence of the equally toxic DDE breakdown product (Weisbrod et al. 2001).

Polychlorinated bornanes (CHBs)

Toxaphene, a product made up of polychlorinated bornanes, was widely used in the southern United States from 1947 as a pesticide on soybean and cotton crops until its gradual ban from 1982 to 1986 (Seleh 1991). It was the most widely used pesticide in North America in the 1970s as a replacement for the banned DDT. It was extensively used in the subtropics, and now has spread atmospherically to temperate and Arctic regions (Kidd et al. 1995; Li et al. 2001). Toxaphene was not reported as regularly as other organochlorines until quite recently because of practical problems related to quantifying the over 1000 constituent compounds (Korytar et al. 2003). CHBs were produced in quantities similar to PCBs (de Geus et al. 1999).

Studies found that both sexes of harbour porpoises collected between 1989 and 1991 from the Gulf of Maine had high levels of CHBs in the blubber (Westgate et al. 1997). Tuerk et al. (2005) reported similar CHB levels in juvenile, female and male Atlantic white-sided dolphins stranded on Cape Cod between 1993 and 2000. More research is needed in the Gulf of Maine as there is no information on the bioaccumulation or toxicity of these compounds on the more sedentary organisms lower in the food chain.

Chlorobenzenes (CBs)

Hexachlorobenzene (HCB) was used in industry and agriculture, mainly as a seed dressing for several crops to prevent fungal disease from the late 1940s to 1970s. HCB is the only CB found in measurable quantities in environmental samples of the Gulf of Maine. Sediment concentrations of HCB in the Gulf of Maine were lowest at the mouth of the Merrimack River, Mass., intermediate at the mouth of the Kennebec River, Maine and highest in Salem Harbor, Mass. (Hauge 1988). HCB was rarely detected (>2.4 ng per g dry) in the Gulf of Maine mussel tissues in the 1990s and 2000s (Jones et al. in prep). White-sided dolphin and pilot whale blubber samples collected from strandings near Cape Cod in the early to mid 1990s contained high concentrations of HCB in blubber (0.18±0.16 and 0.21±0.18 ppm wet weight respectively; Weisbrod et al. 2001). There is no obvious trend of HCB concentrations in harbour porpoises collected between 1973-77 (Gaskin et al. 1983) and 1989-91(Westgate et al. 1997). Low levels of HCB were found in the blubber of harbour seals with an almost twofold decrease observed between 1991 and 2001 (Shaw et al. 2005). A long-term study in the Arctic of HCB levels in seabird eggs indicates a decline in HCB levels between 1975 and 2003 (Braune 2007). HCB, therefore, is present throughout the Gulf of Maine but at much lower concentrations than the other organochlorines and its presence is declining.

Hexachlorocyclohexane (HCHs)

HCH was used as an agricultural insecticide in North America beginning in the 1940s (Brooks 1974). Technical grade HCH, which contained a high proportion (60 to 70%) of the α -HCH isomer, was banned for production and use in the United States and Canada in 1976. However, lindane (98% γ -HCH isomer) is still imported by the United States and used as a pesticide on seed crops and as a scabicide in humans (US EPA 2002).

HCHs are another group of organochlorines that has spread north into temperate and Arctic regions because of evaporation from soils in the south, slow degradation rates and more favourable air to water partitioning at cooler temperatures (Wania et al. 1999; Bidleman et al. 2007). Atmospheric concentrations of HCHs are higher in the centre of the North American continent (Ma et al. 2004) and HCHs have been transported to the North Atlantic (Lohmann et al. 2009) and Arctic Ocean (Bidleman et al. 2007).

Lindane (γ -HCH) was the only form of HCH detected in the Gulf of Maine mussel tissues (Jones et al. in prep). Lindane was detected in 7% of 728 samples at levels just above the detection limit of >1.5 ng per g dry weight from Massachusetts to Nova Scotia and was primarily detected in two sampling years of 1994 and 1995. This is an interesting finding as the more persistent forms of HCH (α -HCH and β -HCH) are predominant at higher trophic levels in the Gulf of Maine. Apeti et al. (2010) found very low values in canned mussels from the 1940s but found higher levels (ppb) of both γ -HCH and α -HCH isomers at Birch Cove, Maine, between 1986 and 2007. Four out of 14 Mussel Watch sites sampled in the Gulf of Maine by National Oceanic and Atmospheric Administration's National Status and Trends (NOAA NS&T) between 1986 and 2003 had significant decreasing trends of lindane whereas the remaining stations had indeterminate trends (O'Connor and Lauenstein 2006). Similarly, 30% of the 47 NOAA Mussel Watch sites sampled along the eastern seaboard from Florida to Maine had significant decreasing trends of lindane consistent with its restricted use. Annual median concentrations declined tenfold at these sites from the late 1980s to the early 2000s (O'Connor and Lauenstein 2006).

Humpback whale blubber samples collected from the Gulf of Maine during the summers of 2005/2006 were found to contain 0.011 ± 0.002 (0.004-0.014) ppm wet weight total HCH (Elfes et al. 2010). Harbour porpoise collected from the Gulf of Maine and Bay of Fundy between 1989 and 1991 contained 0.20 ± 0.12 ppm total HCH wet weight in the blubber and the amount of α -HCH was four times that of the β -HCH and γ -HCH isomers combined (Westgate et al. 1997). This is surprising because technical HCH, of which α -HCH is a major constituent, has been banned in North America since 1976, thus the α -HCH measured in Gulf of Maine harbour porpoises most likely came atmospherically from the Asian continent. Lindane was listed in 2006 as a regulated substance in North America (US EPA 2012a).

Chlordanes (CHLs)

Technical chlordane is a manufactured pesticide that is a mixture of more than 140 compounds, of which trans- and cis-chlordane, heptachlor and trans- and cis-nonachlor are major components (Dearth and Hites 1991). Heptachlor is the most toxic component of the chlordane group, and its rapid breakdown product heptachor epoxide has been used as a pesticide in its own right. The presence of CHLs in the Gulf of Maine reflects the widespread historical application of this extremely persistent pesticide on agricultural and urban soils for about 40 years, including lawns and golf courses on the New England east coast (Phillips and Birchard 1991). It was used as a pesticide for seed crops, such as corn, from 1948 to the mid 1970s in North America.

The sediments of Boston Harbor, Merrimack River, Kennebec River and Portland Harbor had low ppb CHL concentrations (dry weight) in the 1980s (Figure 4; Ray et al. 1983; Hauge 1988). In a later study of Portland Harbor and Casco Bay in 1990, sediment CHL values were found to decrease towards the outer bay indicating either urban or riverine contamination of this harbor (Kennicutt et al. 1994). The associated polychaetes (worms) and clams from the 1980 Portland Harbor study had CHL values in the low ppb levels on a wet weight basis (Ray et al. 1983). Chlordane levels determined in mussels from around the Gulf of Maine between 1993 and 2008 were composed of the alpha-chlordane, trans-nonachlor, heptachlor and heptachlor epoxide isomers and were also in the low ppb range (Jones et al. in prep). Highest chlordane values for mussels were obtained along the Massachusetts shoreline indicating an urban influence (Table 2). Weisbrod et al. (2001) reported CHLs at ppm levels in white-sided dolphins and pilot whale blubber from stranded animals in the Cape Cod area from the early to mid 1990s (Figure 4), whereas their presumed prey, mackerel, herring and squid, contained only ppb CHL concentrations in whole ground organisms. The average CHLs measured in farmed Atlantic salmon fillets (with skin) from Maine and Canada ranged from about 7.5 to 24.5 ppb wet (Shaw et al. 2006). Humpback whales sampled in the Gulf of Maine in 2005/6 had lower ppm CHLs levels in blubber than either dolphins or pilot whales, as expected because they can feed lower in the food chain (Elfes et al. 2010). Blubber samples taken from stranded harbour seals from the Gulf of Maine and New England coast between 2001 and 2002 also ranged in the low ppm CHLs (Shaw et al. 2005). The above CHL values taken together illustrate how organochlorines, such as the chlordane group, bioaccumulate in the higher trophic levels of the Gulf of Maine (Figure 4).

Casco Bay was revisited in 1990 and sediment chlordane concentrations were found to be unchanged from the early 1980s in the immediate area of Portland Harbor (Ray et al. 1983; Kennicutt et al. 1994). CHL levels in mussels were found to be increasing over the 1993-2008 period at Merrimac River, Massachusetts and Limekiln Bay, New Brunswick; however, no trends were observed at the other 16 sites around the Gulf of Maine with six or more years sampled (Jones



Figure 4: Illustration of the biomagnification of chlordane in the Gulf of Maine food web. Chlordane is transferred from the environment (sea water and sediments) to marine organisms; proportionately more is absorbed by the smaller organisms (e.g., plankton and meiobenthos) and subsequently biomagnified in the food web as shown.

et al. in prep). Over the longer term, Lauenstein (1995) compared 51 stations of the NOAA Mussel Watch program taken in 1992 with reanalyzed archived samples from 1972 and found that 46 of these sites had decreased levels of CHL. More recent Mussel Watch data from the entire eastern seaboard of United States indicates that 35 of 47 sampling sites had a significant decreasing trend in CHLs between 1986 and 2003, with the remaining sites indeterminate (O'Connor and Lauenstein 2006). Apeti et al. (2010) found no trace of CHLs in Maine mussels canned in the 1940s, whereas cis-chlordane and trans-nonachlor values from the NOAA NS&T Mussel Watch for Birch Cove, Maine, appear to have declined between the late 1980s and mid 1990s with no further decline up to 2007. There was no change observed in chlordane concentrations associated with the blubber of harbour porpoises stranded in the Bay of Fundy and approaches between the

1970s (Gaskin et al. 1983) and those sampled between 1989 and 1991 (Westgate et al. 1997). However, Lake et al. (1995) found an apparent twofold drop in cis-Chlordane and trans-nonachlor values in harbour seals from 1980 to 1990-92 on the New England coast. The lack of temporal trends in mussel CHL values since the 1990s throughout the Gulf of Maine (Jones et al. in prep) is consistent with CHL values reported for harbour seals collected between 1991 and 2001 (Shaw et al. 2005). This lack of change in recent CHL concentrations in mussel and seal studies in the Gulf of Maine is supported by the results of a time series analysis of 17 biota types in the Arctic with greater than six years of observations where only two species had decreasing concentrations (Riget et al. 2010).

It is estimated that 25 to 50% of the chlordane applied in the United States since the 1940s remains unaltered in the environment with unknown ecological consequences for the northern hemisphere (Shaw et al. 2005).

Mirex

Mirex is another organochlorine pesticide that was used extensively between 1958 and 1978 as both a fire retardant and a pesticide to control an epidemic of fire ants, originating from Europe, in the southeastern United States. Mirex was never used in agriculture in Canada. Mirex was found in the sediments of Casco Bay in 1991 in the low ppb range, with no indication of any concentration gradient out from Portland Harbor (Kennicutt et al. 1994). Mirex was not detected by the NOAA NS&T mussel monitoring along the coast of Maine between 1965 and 1970 (Butler 1973). Weisbrod et al. (2001) was unable to detect Mirex in mackerel, herring or squid in the Gulf of Maine, however white-sided dolphin and pilot whales were found to contain ppb levels in blubber, between the early and mid 1990s. Lake et al. (1995) reported Mirex at ppb levels in harbour seal blubber along the coast of Massachusetts in 1980 and around Long Island, NY, in 1990-92. Shaw et al. (2005) found ppb Mirex concentrations in blubber of female, male and yearling harbour seals along the Gulf of Maine coast between 2001 and 2002. Westgate et al. (1997) reported higher ppb Mirex levels in the blubber of male and female harbour porpoise collected between 1989 and 1991 from Grand Manan and Jeffreys Ledge in the Gulf of Maine. Stein et al. (1992) found ppb Mirex levels in the blubber of three harbour porpoises in 1991 from the Boston and Boothbay Harbor regions.

This is an example of an organochlorine used selectively in the southwestern United States over 30 years ago that is presently widespread at ppb levels in the higher trophic levels of the Gulf of Maine.

Aldrin/Dieldrin/Endrin

Aldrin, endrin and dieldrin are another organochlorine group widely used as pesticides in Canada and the United States for crops like corn and cotton from the 1950s to 1970s. Aldrin breaks down rapidly in the environment to dieldrin



by both microbial activity and exposure to sunlight. Dieldrin is known to bind tightly with soil particles and evaporate slowly over decades. Thus, dieldrin is another very stable and persistent organochlorine compound that is transported atmospherically towards the poles (Barrie et al. 1992).

Dieldrin and its stereoisomer endrin have been quantified in the sediments of Casco Bay, Maine, at low ppb levels, on a dry weight basis, with no evidence that a gradient existed seawards (Kennicutt et al. 1994). Dieldrin and its homologues were not detected in Maine mussels canned in the 1940s, which is consistent with known usage, but were at low ppb levels dry weight during the period of the NOAA Mussel Watch program of 1986 to 2006 (Apeti et al. 2010). Soft-shelled clams were found to have 20 ppb wet weight levels of dieldrin in the late 1960s (Butler 1973). Dieldrin was detected in 16% (126/728) of the mussel sites sampled by the Gulf of Maine monitoring committee between 1993 and 2008, with eight positive sites in Massachusetts and one positive site in both Maine and Nova Scotia (Table 2, Jones et al. in prep). The highest site median was in Boston Harbor (1.3 ppb wet), followed by Portland, Maine (1.0 ppb).

Mackerel, herring and squid caught in the mid 1990s, in the southern Gulf of Maine were found to contain low ppb dieldrin levels (Weisbrod et al. 2001). These researchers also report low ppm levels of dieldrin in the blubber of white-sided dolphins and pilot whales from the Gulf of Maine in the early to mid 1990s. Shaw et al. (2005) found considerably lower concentrations of dieldrin (ppb) in the blubber of harbour seals between 1991 and 2001/2002 for the New England coastline, including the Gulf of Maine. The male harbour seals had three times the contamination of females, since females transfer dieldrin to their offspring while pregnant and nursing. The Bay of Fundy harbour porpoises in the late 1960s were found to have low ppm dieldrin levels in blubber, with male concentrations being seven times that of females (Gaskin et al. 1979). Stein et al. (1992) found comparable levels in the blubber of three porpoises from Boston and Boothbay Harbors of 1.66±0.93 ppm dieldrin in 1991. Dieldrin levels in eggs of seabirds nesting in the Bay of Fundy during the 1970s were in the low to mid ppb wet weight and found to have no relation to trophic habitats from benthic-foraging eiders to fisheating cormorants (Pearce et al. 1979).

Observations from the Gulfwatch program (Jones et al. in prep) showed that dieldrin levels in bivalves dropped tenfold from the 1960s (Butler 1973); however, no temporal trends were observed for dieldrin concentrations over the 1993 to 2008 period monitored extensively by Gulfwatch. However, the NOAA Mussel Watch program in the Gulf of Maine had 4 out of 14 sites with significant decreasing trends between 1986 and 2003 (O'Connor and Lauenstein 2006). Shaw et al. (2005) found no difference between dieldrin levels in harbour seals between 1991 and 2001/2002; however, their 1991 sample size of three seals was small. A resampling of porpoise blubber in the 1980s, from Grand Manan and Jeffreys Ledge, found lower levels than reported in an earlier study in the 1960s (Gaskin et al. 1983; Westgate et al. 1997). These marine mammal data are consistent with the decline in dieldrin concentrations in shellfish in the Gulf of Maine following the mid-1970s ban of this insecticide. Thus, dieldrin concentrations in bivalves and the blubber of harbour porpoise have decreased since the 1960s, following the North American ban on crop applications but appear to have levelled out since this time due to its environmental persistence.

3.3 ORGANOBROMINE COMPOUNDS

Organobromine compounds are organic compounds that contain carbon bonded to bromine. They are the most common organohalides naturally present in marine organisms although bromide is only 0.3% of the concentration of chloride in seawater. The application of concern here is the use of polybrominated diphenyl ethers (PBDEs) as fire retardants. PBDEs are a synthetic category of brominated aromatic compounds developed for their flame resistant properties for use in the production of domestic and commercial products such as polyurethane foam in upholstery, plastics and electronics. PBDEs are widespread persistent compounds that are bioaccumulated in the marine food chain and known to have a variety of toxic effects (see Shaw and and Kannan 2009).

PBDEs have not been measured in seawater in the Gulf of Maine but are reported in the pptr range elsewhere in marine waters (see Shaw and Kannan 2009). PBDEs were quantified at 2.5 ppb, on a dry weight basis, in sediments sampled off Massachusetts. In the Gulf of Maine there is clear evidence for food chain magnification with 21 to 143 ppb on a lipid basis, measured in blue mussels, 71 to 91 ppb in herring, 2340 ppb in herring gull eggs, 80 to 3827 ppb in harbour seal blubber, 610 to 2410 ppb in white-sided porpoise blubber and 8627 ppb in bald eagle eggs (Goodale et al. 2008; Kimbrough et al. 2009; Shaw et al. 2008; 2009; Tuerk et al. 2005). PBDE levels in herring gull eggs from colonies in the Bay of Fundy were low, at mid ppb wet weight, compared to inland Canadian colonies, which can be attributed to a reduced source due to lower human population densities (Chen et al. 2012). PBDEs have increased exponentially in marine life and humans since their introduction in the 1970s (Shaw and Kannan 2009).

Two other brominated compounds, tetrabromobisphenyl A (TBBPA) and hexabromocyclo-dodecane (HBCD), are currently unregulated alternatives to PBDEs as flame retardents. HBCD was present at ppb lipid in most fish specimens analyzed from the Gulf of Maine (Shaw et al. 2009). Low HBCD levels (ppb wet weight) were measured in eggs from herring gull colonies in the Bay of Fundy (Chen et al. 2012). Atlantic white-sided dolphins, stranded along the United States east coast, had concentrations ranging between 3 to 340 ppb HBCD in blubber lipid between 1993 and 2004 (Peck et al. 2008). However, TBBPA and HBCD levels in marine organisms are two to three orders-of-magnitude lower than PBDEs. Human adipose tissue and human milk samples from New England, including Massachusetts, ranged from ppb to ppm in fat (Johnson-Restrepo et al. 2005; 2007). These levels in North Americans are 10 to100 times higher than for human populations elsewhere and are thought to be derived 75% from dust venting from furniture and carpets and 25% from consuming sea food.

3.4 PERFLUORINATED COMPOUNDS (PFCS)

Polyfluoroalkyl substances are synthetic chemicals that have been produced since the late 1940s for a wide variety of applications such as polyurethane production, vinyl polymerization, fire-fighting foams, oils, surfactants, coating solutions for fabrics, etc. (Prevedouros et al. 2006). They degrade in the environment to persistent perfluoroalkyl sulfonates (PFSAs) and perfluorocaboxylates (PFCAs). Perfluorinated compounds (PFCs) are distinguished by having all the hydrogens on their carbon chain replaced by fluorine and possess at least one different atom or functional group. This structure makes PFCs extremely persistent toxic contaminants in the environment, with no evidence for biodegradation (Giesy and Kannan 2002). They are distributed worldwide and known to be at elevated concentrations in the top marine predators (Tomy et al. 2004; Houde et al. 2006; Shaw et al. 2009).

There is limited information available on PFCs for the Gulf of Maine region. Very low levels of 0.1 to 0.3 pptr PFCs were measured in seawater from the North Atlantic off Nova Scotia and Newfoundland (Yamashita et al. 2005). Low levels of both PFSAs and PFCAs (ppb wet weight) were found in herring gull eggs from two colonies in the Bay of Fundy in 2008 (Gebbink et al. 2011). Highest values in this study were found inland near major urbanized areas. PFCs were analyzed in the livers of harbour seals collected between the Gulf of Maine and New York between 2000 and 2007 (Shaw et al. 2009). Levels were in the low ppb wet weight range with no significant geographical differences found between the Gulf of Maine and New England regions. There was little indication that levels had declined since the voluntary phase-out of PFSAs compounds in 2000 by the 3M Company.

3.5 ORGANOPHOSPHATES (OPS)

An organophosphate is a general term for esters of phosphoric acid. OPs are used as pesticides, herbicides, nerve gas, plasticizers and flame-retardants.

Organophosphate pesticides

Certain organophosphates, with the ability to incapacitate the nervous system of insects and crustaceans, have replaced organochlorines as pesticides due to their

rapid degradation in the environment. Diazinon (*O*,*O*-Diethyl *O*-[4-methyl-6-(propan-2-yl)pyrimidin-2-yl] phosphorothioate) is an OP pesticide that was used extensively in North America during the 1970s and early 1980s for domestic and agricultural purposes. Diazinon was measured in four of seven rivers flowing into the Gulf of Maine as recently as 1999 and 2000 in the low ppb range (Kolpin et al. 2002).

Azamethiphos (S-6-chloro-2,3-dihydro-2-oxo-1,3-oxazolo[4,5-*b*]pyridin-3-ylmethyl *O*,*O*-dimethyl phosphorothioate) is currently the only pesticide registered for "sea lice" removal from salmon grown in open sea pens in Canada. It is, therefore, a chemical of concern to east coast lobster fishermen. Azamethiphos has a short half-life in nature and therefore would have a limited effect on wild crustaceans (Ernst et al. 2001; Jackman et al. 2001). These properties make it difficult to quantify in seawater following aquaculture applications, so rhodamine dye was used to trace seawater as it dispersed from the cages (Ernst et al. 2001).

Organophosphate flame retardants

There has been renewed interest in OPs as flame retardants by chemical companies because of the recent human health concerns expressed by various American state governments about the possible toxic effects of organobromines used as flame retardants and plasticizers in household goods. California passed legislation to ban the use of polybrominated diphenyl ethers (PBDEs) as flame retardants in 2003, followed by eight other states and the European Union such that the only American manufacturer, Chemtura, voluntarily phased out production by 2004 (Levcik and Weil 2006; Stapleton et al. 2011). The OP replacements are man-made compounds that do not occur naturally, although other OPs make up the important building blocks for life, such as RNA and DNA. The chlorinated organophosphates, such as tris(2-chloroethyl) phosphate (TCEP) and tris(2-chloroisopropyl) phosphate (TCPP) are favoured as flame retardants in polyurethane foam, etc., whereas the non-chlorinated OPs, such as tris(2-butoxyethyl phosphate (TBEP), are mostly used as plasticizers.

Neither flame retardants nor plasticizers are chemically bonded so they are eventually emitted from the polyurethane or plastic surfaces on dust particles or volatized directly to the air. Studies on soil contamination have confirmed the importance of the atmospheric transport and deposition route of OPs by measuring their presence in field soils remote from urban and industrial centres (Fries and Mihajlovic 2011). However, sewage treatment plant outfalls are thought to be the biggest source of OPs escaping to the environment with the chlorinated OPs passing through the treatment process unscathed (Meyer and Bester 2004; Marklund et al. 2005). Two flame retardants (TCEP and TCPP) and one plasticizer (TBEP), have also been quantified in five of seven Massachusetts rivers sampled at >0.04 to 0.07 ppb and >01 to 0.16 ppb, and >0.2 to 0.62 ppm respectively (Barnes et al. 2002; Kolpin et al. 2002). There are presently no measurements of OPs for seawater in the Gulf of Maine; however, comparable freshwater levels to the Massachusetts rivers were found in the River Elbe, Germany, and they contributed 3 to 28 pptr TCPP into the German Bight, North Sea (Bollmann et al. 2012). OPs have not been quantified in the Gulf of Maine marine food chain.

3.6 PYRETHRUM AND PYRETHROIDS

Pyrethroids are among the most toxic of pesticides (Fairchild et al. 2010). Pyrethroids are synthetic forms of the natural pyrethrin neurotoxins extracted from the chrysanthemum plant, which has been in use as a pesticide since the late 1800s. Pyrethroids were developed in the 1960s to replace DDT and other chlorinated pesticides in agriculture for insect control. Agricultural use can result in exposure of near-shore fauna to drift of pesticide spray or surface runoff from cultivated fields adjacent to coastal waters. However, pyrethrins and pyrethroids are relatively short lived in the environment, being degraded by both sunlight and microbial activity.

Two pyrethroids, cypermethrin and deltamethrin, are chemicals of concern, particularly to the lobster fishery, because of their extreme toxicity and their possible application to control "sea lice" in salmon aquaculture. Cypermethrin is approved for marine use in the United States but not in Canada. Deltamethrin was permitted in New Brunswick on an emergency and experimental basis in 2009 and 2010 after a sea lice outbreak; as of 2011 it had not been used in Maine. Both cypermethrin and deltamethrin are applied to fish pens enclosed within an impervious tarp for approximately an hour at a specified concentration then allowed to disperse to the surrounding seawater. It is difficult to quantify these pyrethroids in seawater as they are applied at extremely low concentrations of 2 to 5 ppb and rapidly disperse from the sea pens, depending on the location and coastal currents. Cypermethrin and deltamethrin are known to have a half-life of hours in water. However, both pesticides have the potential to adsorb to organic particles in the water column and settle to the bottom where they can be more persistent. However, experimental studies suggest that these pesticides are not available for uptake by bottom-living organisms unless they partition into overlying or interstitial waters (Clark et al. 1987).

Fish farming is confined to relatively sheltered embayments in the cooler northern waters of the Gulf of Maine, from Boothbay Harbor north. Activities are concentrated in Machias and Cobscook Bays, Maine and Passamaquoddy Bay, New Brunswick, with smaller operations west in Annapolis Basin and southwestern Nova Scotia.

3.7 PHARMACEUTICALS

The pharmaceuticals, as human and animal medicines are known, are often referred to as "emerging" categories of environmental contaminants. In fact, certain pharmaceuticals, such as a blood pressure lowering substance, were first detected thirty years ago in treated wastewater in the United States (Garrison et al. 1976). New analytical techniques developed in the last fifteen years have enabled trace quantities of polar compounds to be detected (Fent et al. 2006). There are currently estimated to be 3000 pharmaceuticals in use in Europe (Fent et al. 2006). Sewage treatment effluents have been well documented as a major source of pharmaceuticals to surface and ground waters that ultimately reach the sea (Halling-Sorensen et al. 1998; Metcalfe et al. 2004; Gros et al. 2007; Kasprzyk-Hordern et al. 2008; Schultz et al. 2010). Sewage treatment plants are unable to efficiently remove most pharmaceuticals with the exception of analgesics and anti-inflammatories (Fent et al. 2006). Many treatment plants around the Gulf of Maine are primary treatment (solids) at best. A number of these chemicals are also used as veterinary pharmaceuticals to improve growth and health in livestock and poultry operations, and these also end up in surface runoff from manure spread on fields and from farm effluent. Pharmaceuticals are also used in the aquaculture of fish species in sea pens.

The pharmaceuticals are an exceptionally chemically-diverse group of drugs best described here by usage, such as analgesics and non-steroidal anti-inflammatory drugs, beta-blockers, blood lipid lowering agents, cancer therapeutics, neuroactive compounds, synthetic steroidal hormones, antidiabetics, antiepileptics , X-ray contrast drugs, antiacids, broncodilators, antibiotics, biocides, stimulants and others. There has been little attention paid to pharmaceuticals entering the Gulf of Maine with the exception of the continental United States Geological Survey of 1999 and 2000, which included several rivers in Massachusetts.

Analgesics

A widely used analgesic, acetaminophen, and an analgesic/anti-inflammatory, ibuprofen, were found in Massachusetts rivers draining into the Gulf of Maine (Kolpin et al. 2002).

Synthetic steroidal hormones

Steroidal estrogens, used for human birth control pills, have been measured in Massachusetts rivers draining into the Gulf of Maine (Kolpin et al. 2002).

Antibiotics

The United States Geological Survey examined 139 waterways across the nation for chemical contaminants and found measureable quantities of 8 out of 29 antibiotics analyzed in Gulf of Maine rivers (Kolpin et al. 2002).



Biocides

Avermectin biocides are applied in the food of farmed fish to remove ectoparasitic copepods, known as "sea lice". They are semi-synthetic chemicals derived, in part, from a precursor produced by the bacteria *Streptomyces avermitilis*. Ivermectin was used without the sanction of the manufacturer as an "off-label" treatment in the 1990s and has been replaced by emamectin benzoate in 1999 as an Emergency Drug Release from Health Canada. Both chemicals have low water solubility and are readily adsorbed onto particulate material. Ivermectin and emamectin benzoate are difficult to measure in seawater because of this extremely low solubility in a very diffusive environment. They are found, however, concentrated in the sediments under or close to fish sea pens, depending on local current speed. Sediment concentrations result from the settling of uneaten food pellets, fish feces and particulates. Mussels were shown to accumulate low levels of emamectin benzoate beneath or close to Scottish aquaculture operations within the first week of use; however, this dissipated by four months time (Telfer et al. 2006).

3.8 METALS AND ORGANOMETALS

Butyltin compounds

Butyltins were introduced as antifouling agents for ship hulls and fishing gear in the late 1960s as a more effective replacement for copper-containing treatments. Tributyltin (TBT), a synthesized compound, was the primary toxic ingredient in marine paints used to prevent fouling by benthic organisms. Butyltins are the sum of tributyltin and its breakdown products of monobutyltin, dibutyltin (DBT) and tetrabutyltin. Butyltins and other organotins can also be introduced into the marine environment through their use as stabilizers in PVC piping, silicone resins and polyurethane forms. TBT has a very low water solubility, which means that it will bind strongly with particulate material in the water column and settle to the bottom sediments. There is no concern that TBT will be transported atmospherically because of its low vapour pressure.

The highest concentrations of butyltins occur in harbours where boating and shipping activities occur. Butyltins were analyzed in Maine coastal seawater collected in 1991 between Portland Harbor and Boothbay Harbor, four years after its restricted use in antifouling paint. Seawater concentrations ranged between the detection limit of ~1 to 20 ppb as tin, with the parent compound TBT dominating (Larson et al. 1997). Butyltins were quantified also in sediments from Portland Harbor, Maine, in 1990 and found to have a range of concentrations of 24 to 693 ppb (TBT) and 15 to 453 ppb (DBT) as tin on a dry weight basis, with the highest concentrations found near shipyards (Wade et al. 2008). Dogwhelks, *Nucella lapillus*, were found to have ppb (dry weight) TBT levels in Saint John Harbour in both 1995 and 2008 (Prouse and Ellis 1997; Titley-O'Neal et al. 2011a). In general, small coastal cetaceans (e.g., porpoises and dolphins) have higher concentrations

of TBT in their liver compared to oceanic species and cetaceans have higher concentrations of TBT in industrialized as compared with developing nation coastal waters (Tanabe 1999).

Tributyltin concentrations in Casco Bay sediments declined dramatically over a ten-year period following the introduction of paint restrictions on small boats in 1988, such that only two sites had levels above 15 ppb tin dry weight (maximum 60 ppb) in 2001 compared to seventeen sites scattered throughout the bay in 1991 (Wade et al. 2008). A similar decline was observed with oyster data from Galveston Bay between 1986 and 1994 (Jackson et al. 1998) and near Miami between 1988 and 1994 (Cantillo et al. 1997). The NOAA mollusc monitoring group reported only decreasing trends for butyltins at ~25% of the 196 sites sampled for the entire United States between 1986 and 1996 (O'Connor 1998). There is no comparable local information on marine mammals; however, butyltins have declined in Pacific sea otters and cetaceans since restrictions were introduced in the late 1980s (Murata et al. 2008; Tanabe 1999).

Mercury and methylmercury

Mercury is a natural element of the earth's crust that can be toxic to life at high concentrations. Mercury, in its many forms, is very mobile in the marine environment with methylmercury being the most toxic and only biomagnified form in aquatic food chains (Sunderland et al. 2012). Past anthropogenic inputs to the marine environment were through pesticide and pharmaceutical use and from industrial sites such as gold mining, chlor-alkali production and pulp and paper plants (Sunderland and Chmura 2000a). Mercury also has a long history of being introduced into the aquatic environment from the combustion of wood, coal and other petroleum products, which has proven to be more difficult to control and legislate than other sources (Sunderland and Chmura 2000b).

Mercury is transported via particulate matter and is concentrated in depositional areas in the Gulf of Maine: tidal flats, salt marshes, and deep basins (Dalziel et al. 2010; Hung and Chmura 2006; Sunderland et al. 2012). The mercury content of Bay of Fundy fine-grain sediments ranged between 0.02 and 0.09 ppm dry weight with highest values in Saint John Harbour, New Brunswick (Loring 1979; Loring et al.1996; Ray and MacKnight 1984). The sediments in Passamaquoddy Bay, New Brunswick ranged from 0.01 to 0.1 ppm dry weight (Sunderland et al. 2004).

Phytoplankton (25-63 µm size range) sampled at the approaches to the Bay of Fundy between 2000 and 2002 contain a median of 2.8 (range 1.9–4.3) ppb total mercury wet weight (Harding et al. in prep.). Mercury levels in filter-feeding blue mussels collected by Gulfwatch in the Gulf of Maine ranged between 0.04 and 0.60 with a median of 0.17 ppm total mercury wet weight from 51 locations around the Gulf of Maine between 2003 and 2008 (Jones et al. in prep). Copepods, predominantly filter-feeding *Calanus*, from surface plankton tows in



the Quoddy region, contained 4.3±2.2 ppb total mercury wet weight, whereas, the mercury content of one of their predators, herring, increased from 5.1 ± 1.0 ppb wet as yearlings to 14.9±4.8 ppb wet by age eight (Braune 1987a). Zitko et al. (1971) measured methylmercury in a variety of fish fillets from the Bay of Fundy and approaches, ranging from alewife (50 ppb wet weight) to witch flounder (30-400 ppb wet weight). Mercury levels within the Gulf of Maine marine bird community indicate clear trophic relationships, with fish-eating cormorants, petrels, guillemots and puffins having higher concentrations than bottom-foraging eider ducks and scavenger-feeding herring gulls (Pearce et al. 1979; Braune 1987a; Elliott et al. 1992; Goodale et al. 2008). Guillemots, eiders, herring gulls and cormorants in the Bay of Fundy have muscle total mercury content of 0.113, 0.153, 0.101 and 0.606 ppm wet weight, which reflect their higher trophic position (Braune 1987b). Harbour seals taken near Grand Manan and Deer Island, N.B., in 1971 had median total mercury concentrations in muscle tissue of 0.38 (0.16-0.59) ppm wet weight (Gaskin et al. 1973). Lake et al. (1995) report higher total mercury levels in harbour seal livers collected along the Gulf coast off Massachusetts in 1980 at 38.5±7.86 (31.6-49.3) ppm total mercury wet weight. Total mercury was measured in the muscle tissue of 146 porpoises tangled in fishing nets from the Bay of Fundy and adjacent waters in the 1970s with annual mean concentrations varying between 0.51 and 1.69 ppm wet weight (Gaskin et al. 1979). The above studies illustrate the biomagnification of mercury as methylmercury in the Gulf of Maine ecosystem (Chen et al. 2008).

No mercury temporal trends were found in herring gull eggs collected at two Bay of Fundy breeding colonies between 1972 and 2008 after adjustment for dietary shifts (Burgess et al. 2013). Likewise, no temporal trends or differences between the male and female porpoises were observed between 1969 and 1977. Furthermore, Stein et al. (1992) got similar values for porpoises in the Gulf of Maine collected in 1991. Atmospheric levels of mercury have tripled over the industrial age (Mason et al 1994; Mason and Fitzgerald 1996) but this increase has not been evident in the upper trophic levels in the ocean (Miller et al. 1972; Scott 1977; Kraepiel et al. 2003).

3.9 TRACE ELEMENTS

Trace elements are natural components of seawater, sediment and biota, through the weathering of the earth's crustal material; however their concentrations have increased in certain environmental compartments by industry and mining wastes, agriculture runoff and domestic sewage discharges (Weinstein and Moran 2004). Trace metals are associated primarily with fine particulate material that settles in less turbulent conditions (Weinstein and Moran 2004). Higher concentrations, therefore, are found in sedimentary regions, such as tidal flats and salt marshes (Daoust et al. 1996), or in the deeper marine basins offshore (Weinstein and Moran 2004). The transport of metals atmospherically has received attention only recently with Canadian and American sources of lead being distinguishable by their isotopic ratios (Knowlton and Moran 2010).

Trace metals have been measured in sediments at various locations around the Gulf of Maine since the late 1970s (Larson 1992; Gottholm and Turgeon 1992; Loring 1979). Sediment studies of the late 1980s found that Boston and Salem Harbors, Massachusetts, had consistently the highest values of lead, cadmium, copper, chromium, nickel and zinc in the Gulf of Maine. The mid-Maine coastal embayments have been studied from the 1980s and been found to have moderate enrichment of arsenic, cadmium, chromium, copper, lead, mercury, nickel and zinc over pre-colonial conditions (Wade et al. 2008; Larsen and Gaudette 2010). The St. Croix estuary and Passamaquoddy Bay study results suggest that only cadmium and perhaps lead and zinc may be above the natural levels of the pre-colonial period due to human activity (Loring et al. 1998).

The seven trace metals analyzed in blue mussels by Gulfwatch between 1993 and 2008 were present at a wide range of concentrations from just above detection level to relatively elevated levels (Table 3). Median mercury and lead

		AVERAGE-ALL								
		CHEMICALS	Hg	Ag	Cd	Pb	Ni	Zn	Cr	Cu
Mass.	Low	12%	17%	7%	33%	0%	20%	0%	13%	7%
	Medium	57%	75%	27%	60%	40%	67%	73%	67%	47%
	High	31%	8%	67%	7%	60%	13%	27%	20%	47%
	Low	0%	0%	10%	Q 0/	0%	8%	0%	8%	8%
19.11.		770	0 /0	72 /0	0 /0	070	0 /0	0 /0	0 /0	5.001
	Medium	42%	30%	42%	25%	/5%	33%	58%	1/%	58%
	High	48%	70%	17%	67%	25%	58%	42%	75%	33%
									_	
Maine	Low	38%	29%	39%	39%	44%	39%	50%	39%	28%
	Medium	50%	57%	56%	44%	39%	50%	33%	56%	67%
	High	12%	14%	6%	17%	17%	11%	17%	6%	6%
N.B.	Low	38%	50%	38%	25%	63%	38%	13%	38%	38%
	Medium	41%	17%	38%	50%	38%	38%	75%	38%	38%
	High	20%	33%	25%	25%	0%	13%	13%	25%	25%
				_						
N.S.	Low	40%	60%	45%	45%	36%	18%	45%	18%	55%
	Medium	47%	40%	45%	27%	55%	55%	36%	82%	36%
	High	13%	0%	9%	27%	9%	27%	18%	0%	9%

Table 3: Percentage of Gulf of Maine Gulfwatch sites within each state/province with low, medium and high trace metal concentrations in blue mussels (Jones et al. in prep). Contaminants were grouped into three categories—low, medium and high—using cluster analysis, so that "low" and "high" means low and high relative to the other levels.

Chemical symbols: mercury (Hg), silver (Ag), cadmium (Cd), lead (Pb), nickel (Ni), zinc (Zn), chromium (Cr) and copper (Cu).



concentrations in mussels were higher in the Gulf of Maine compared to those of the much broader coverage of the American continental Mussel Watch program. For other trace metal comparisons, Gulf of Maine Gulfwatch measurements were slightly lower or the same magnitude as those reported in the broader Mussel Watch Program. Mercury and lead concentrations also differed within the Gulf of Maine jurisdictions with mercury levels highest in New Hampshire and lead levels highest in Massachusetts. Temporal trend analysis of the Gulf of Maine mussels showed little change in metals at the 38 locations with more than four sampling periods over the 18-year period (Tables 4 and 5).

Most trace metals appear to be regulated in seabird tissues from the Bay of Fundy, with the exception of selenium and cadmium in Leach's storm-petrels and lead in herring gulls where elevated levels were found (Elliott et al. 1992).

Table 4: Significant increasing/decreasing trends for contaminants in blue mussel tissue at Gulfwatch sites that have been sampled for n > = 6 years (Jones et al. in prep).

SITE	TRACE METALS	TREND DIRECTION	SITE	ORGANIC CHEMICALS	TREND DIRECTION
Massachusetts			Massachusetts		
Merrimack River	Cr	-	Merrimack River	Chlordane	+
Sandwich	Ag	-		Dieldrin	+
New Hampshire		Sandwich		DDT	-
Dover Point	Cr	-		LMW PAHs	+
	Hg	-	New Hampshire		
	Pb	-	Schiller Station	DDT	-
	Zn	-		PAH	+
Hampton/Seabrook Harbor	Cr	-		Pesticides	-
	Pb	-	Maine		
Little Harbor	Pb	-	Clarks Cove	PCB	-
Schiller Station	Cd	-	Portland Harbor	Dieldrin	-
Maine			Kennebec River	PCB	-
Clarks Cove	Cr	-	New Brunswick		
	Pb	-	Limekiln Bay	Chlordane	+
Kennebec River	Hg	+		DDT	+
New Brunswick			Nova Scotia		
Nova Scotia			Apple River	DDT	-
Apple River	Ag	+			
Digby	Cu	-			
Yarmouth	Ag	-			

Chemical symbols: Ag (silver), Cd (cadmium), Cr (chromium), Cu (copper), Hg (mercury), Pb (lead) and Zn (zinc).

Table 5: Increasing/decreasing trends for contaminants in blue mussel tissue at Gulfwatch sites that have been sampled for n = 4 or 5 years (Jones et al. in prep).

SITE		TREND	SITE		TREND
n = 5	CONTAMINANT	DIR.	n = 4	CONTAMINANT	DIR.
Massachusetts			Massachusetts		
Brewster Island	DDT	-	Duxbury	Cd	-
Boston inner harbor	Ag	-		DDT	+
	Dieldrin	-		РАН	+
Marblehead	Dieldrin	+		Pest	+
Maine			lpswich	Ni	+
Boothbay Harbor	Cr	-	Plymouth – Manomet	Ag	-
	Cu	-	Point	LWPAH	+
	DDT	-	New Hampshire		
	Pb	-	Pierce Island	Pb	_
	Pest	-		Pest	-
Brave Boat Harbor	DDT	+	South Mill Pond	Cd	+
	Pest	+		Pb	+
Cobscook Bay	Chl	-		Zn	+
Dalmariscotta	Ag	+			
	DDT	-			
	Pest	-			
Presumpscot River	PCB	+			
Royal River	DDT	+			
	PAH	+			
	Pest	+			
New Brunswick					
Tin Can Beach	Ag	-			
	LWPAH	+			
New Hampshire					
North Mill Pond	Ag	+			
	Chl	-			
	HWPAH	+			
	РАН	+			
Nova Scotia					
Argyle	Cd	-			
	Cr	_			
	Ni	_			
	Zn	_			
Spechts Cove	РАН	+			

Chemical symbols: Ag (silver), Cd (cadmium), Cr (chromium), Cu (copper), Hg (mercury), Ni (nickel), Pb (lead) and Zn (zinc). Pest is the sum of organochlorine pesticides, ChI is the sum of all chlordanes, PCB is polychlorinated biphenyls, PAH is polyaromatic hydrocarbons, LWPAH is low weight PAHs, HWPAH is high weight PAHs.



3.10 MISCELLANEOUS SUBSTANCES

Artificial sweeteners

Artificial sweeteners are used as sugar substitutes in food, drinks and drugs. These substances therefore pass with the urine and feces into the sewage treatment systems unaltered and were found to pass sewage treatment plants (Scheurer et al. 2009). It follows that certain sweeteners are released in considerable quantities into receiving waters of rivers, lakes and marine waters. There are no measurements available on sweeteners within the Gulf of Maine.

Stimulants

Caffeine is an alkaloid present in many species of plants and consumed as a beverage in coffee, tea and soft drinks and in medicines. Nicotine is a toxic component of tobacco. The caffeine and nicotine breakdown product cotinine was measured in the rivers draining into the Gulf of Maine from Massachusetts (Kolpin et al. 2002). The Deer Island sewage treatment plant effluent emptying into Boston Harbor had caffeine levels of 6.7 ppb (Siegener and Chen 2002). In the same study, caffeine measured in Boston Harbor seawater ranged between 0.14 to 1.6 ppb, and decreased towards Massachusetts Bay (5.2-71 pptr). Cotinine values ranged between <0.4 to 0.07 ppb in Boston Harbor seawater.

Disinfectants

Triclosan (polychloro phenoxy phenol), a commonly used disinfectant in toiletries and underclothing, was detected in five of seven Massachusetts rivers sampled (Barnes et al. 2002; Kolpin et al. 2002).

Aircraft deicing fluid

The corrosion inhibitor, 5-methyl-iH-benzotriazle (5-MEBT), in aircraft deicing fluid was measured in the Charles River, Boston (Barnes et al. 2002; Kolpin et al. 2002).

Insect deterrent

N,N-Diethyl-meta-toluamide, a biting-insect deterrent know as DEET, has been measured in Massachusetts rivers flowing into the Gulf of Maine (Barnes et al. 2002; Kolpin et al. 2002).

4. Impacts

The anticipated impacts of the known contaminants in the Gulf of Maine are summarized below (see Table 6).

Table 6: Anticipated impacts of the contaminant categories found in the Gulf of Maine.

CONTAMINANT GROUP	PERSISTENT	ATMOSPHERICALLY TRANSPORTED	BIOMAGNIFIED	TOXIC	CONCERN LOCAL*	CONCERN GLOBAL
Polycyclic aromatic hydrocarbons	No	Yes	No	Yes/No	Yes	Yes
Organochlorines	Yes	Yes	Yes	Very toxic	Yes	Yes
Perfluorinated compounds	Yes	Probably	Yes/No	Very toxic	Yes	Yes
Organobromines	Yes	Yes	Yes	Very toxic	Yes	Yes
Organophosphates	No	Yes	No	Very toxic	Yes	No
Pyrethroids	No	No	No	Very toxic	Yes	No
Antibiotics	No	No	No	No	Yes	Yes
Pharmceuticals	No	No	No	No	Yes	No
Steroidal hormones	No	No	No	No	Yes	No
Organometals	Yes	Yes/No	Yes/No	Very toxic	Yes	Yes
Trace elements	Yes	Yes	No	Yes/No	Yes	No
Sweeteners	Yes	No	No	No	No	No
Stimulants	No	No	No	No	No	No
Disinfectants	No	No	No	Yes	No	No
Deicing fluid	No	No	No	Yes	Yes	No
Insect deterrent	No	No	No	No	No	No

* Gulf of Maine

4.1 POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)

PAHs are of particular concern because of their mutagenic, carcinogenic and endocrine-disrupting effects on organisms and threat to human health (Bostrom et al. 2002; Santodonato 1997; Villeneuve et al. 2002). PAHs are bioaccumulated by benthic organisms (Pruell et al. 1986; Jones et al. in prep); however they are not known to biomagnify in aquatic food webs but rather have reduced concentrations further up the food chain due to the more efficient metabolic degradation in higher trophic organisms (Kayal and Connell 1995; Nakata et al. 2003; Wan et al. 2007). Nonetheless, coastal tissue levels of PAHs are close to levels that can impair reproduction, growth and cause larval mortality in salmon (Heintz et al. 1999). There is growing evidence that the lowest marine trophic level, the phytoplankton, might be at the greatest risk to the various contaminants including PAHs (Djomo et al. 2004; Echeveste et al. 2010a; 2010b). There are currently no published American or Canadian federal human tolerances for total PAH concentrations in edible seafood. However, Canada has issued interim sediment quality guide-lines and probable effects levels for individual PAHs (CCME 1999) which are approached or exceeded in many of the coastal bays in the Gulf of Maine.

4.2 ORGANOCHLORINES

Polychlorinated biphenyls (PCBs)

The United States Agency for Toxic Substances and Disease Registry considers that PCBs are probable human carcinogens (ATSDR 2000). PCBs are enzyme disruptors (interfere with biochemical functioning of the cell) in marine mammals and have been attributed with a population decline of seals in Europe (Chiu et al. 2011). PCBs are an ever-present and persistent pollutant that bioac-cumulate readily to five orders-of-magnitude in marine food chains (Oehme et al. 1996; Harding et al. 1997; Skarphedinsdottir et al. 2010; Sobek et al. 2010). The concentrations in sediments are thought to be below levels that produce toxic effects in benthic organisms (370 ng PCB/g dry sediment; Long et al. 1995). However, in experimental studies in which polychaete worms were first exposed to New Jersey sediments and then fed to lobsters, it was found that the coplanar PCBs 77 and 126 (compounds that exhibit dioxin-like toxicity) became enriched in both these trophic levels (Pruell et al. 1986). This is of concern because coplanar PCBs are highly toxic and present at orders-of-magnitude higher concentrations than ambient dioxin levels (Metcalfe and Haffner 1995).

The United States Environmental Protection Agency (EPA) published guidance values for limiting total PCB fish consumption by recreational fishers as follows: 5.9 to 12.0 ng/g wet weight due to of possible cancerous effects and 23 to 47ng/g wet weight for non-cancerous toxic effects, assuming four 8-ounce meals/month (US EPA 2000). The EPA Environmental Monitoring and Assessment Program's National Coastal Assessment survey of demersal fishes between 1999 and 2001, which included the Gulf of Maine, found that PCBs can exceed this guidance in larger fish (Harvey et al. 2008). However, all of these PCB values are below the current tolerance levels recognized for human consumption of seafood (2.0 ppm, see Table 8).

Polychlorinated bornanes (CHBs)

Of the many CHB congeners, only a few bioaccumulate in the marine food chain (Oehme et al. 1996). CHBs are highly carcinogenic to laboratory mammals and this has caused concern for human health, particularly those members of society that consume fish products (Seleh 1991; de Geus et al. 1999).

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs)

PCDDs are the most toxic, man-made chemicals unintentionally released into the environment. Marine organisms take up PCDDs and PCDFs from the water, sediment and their prey (Pruell et al. 2000). Biodilution (decreasing concentrations through the food chain) was observed in a marine food chain of phytoplankton/ seston, zooplankton, crab, shrimp, scallop, six species of fish, and gulls (Wan et al. 2005). The discrepancy between the behaviour of PCBs and PCDD/PCDFs in aquatic food webs appears to be due to the ability of organisms to metabolize the latter compounds (Pruell et al 2000).

DDT and its Metabolites (total DDT)

DDT is implicated for its endocrine disrupting properties in developing embryos (Guillette et al. 1994). It is known to be estrogenic and a potent anti-androgen (male hormone) thought to affect male reproductive health in humans and other animals (Gray et al. 2001; Toppari et al. 1996). DDT is biomagnified a thousand-fold between predator and prey in both white-sided dolphins and pilot whales in the Gulf of Maine if herring and mackerel were their chief food source, respective-ly (Weisbrod et al. 2001). Of the total DDT in the dolphin and whale, 93 and 86% respectively was in a degraded form, of which 79 and 77% was DDE. Nevertheless DDE has comparable toxicity to DDT.

Chlorobenzenes (CBs)

HCB is known to cause cancer in experimental animals, thus Environment Canada lists it as toxic in the Canadian Environmental Protection Act (CEPA). Hexachlorobenzene (HCB) is bioaccumulated at the higher trophic levels in the Gulf of Maine.

Hexachlorocyclohexane (HCHs)

Lindane (γ -HCH) is a toxin that primarily affects the nervous system and may be carcinogenic. HCHs are bioaccumulated in the marine food chain but not to the extent of other organochlorines with higher octanol-water partitioning coefficients (log K_{ow}).

Chlordane (CHLs)

Chlordane is a persistent and neurotoxic substance that is transported long distances through the atmosphere (Bidleman et al. 2002). Chlordane is highly toxic to birds in their food and freshwater fish at ppb water levels in 96-hour LC50 tests (US EPA 1986). It is estimated that 25 to 50% of CHL applied in the United States since the 1940s still remains unaltered in the environment (Shaw et al. 2005). It readily biomagnifies in aquatic food chains (Figure 4), although its compositional pattern is known to differ among different trophic levels and species (Norstrom et al. 1988; Kawano et al. 1988).

Mirex

Mirex is carcinogenic in experimental animals and listed as a toxic, persistent and bioaccumulative compound (Shaw et al. 2005). Mirex was found to be highly toxic to shrimps and crabs in the Gulf of Mexico. In the late 1990s, Mirex was routinely found in the plasma and milk of Inuit women from northern Quebec; this toxic organochlorine is passed to infants from their mother's breast milk (Muckle et al. 2001).

Aldrin/Dieldrin

Aldrin breaks down quickly to dieldrin within organisms and in the environment. Dieldrin is a known neurotoxin and can cause cancer of the liver in mammals. Dieldrin is a very stable and persistent organochlorine compound that is transported atmospherically towards the pole in the northern hemisphere by the repeated processes of evaporation and precipitation (Barrie et al. 1992). Dieldrin is another lipophilic organochlorine compound well known to biomagnify through the marine food chain (Weisbrod et al. 2001).

4.3 ORGANOBROMINES

PBDEs are known to be persistent, toxic and bioaccumulate in fatty tissues, like their predecessors the polybrominated biphenyls (PBBs), particularly in aquatic food webs (Shaw and Kannan 2009). PBDEs are known endocrine disrupters, altering thyroid hormone production and causing developmental and reproductive abnormalities in a wide variety of wildlife (Talsness 2008). Current PBDE levels in North Americans are 10 to 100 times higher than human populations elsewhere, which is not surprising given that 95% of the world's production of PBDE was used in the United States and Canada (Shaw and Kannan 2009). PBDEs will remain in the environment for decades to come, because of the highly refractory nature of this category of compounds.

4.4 PERFLUORINATED COMPOUNDS (PFCS)

PFCs are persistant, bioaccumulative and toxic man-made compounds that are distributed globally (Giesy and Kannan 2002; Prevedouros et al. 2006). PFCs have been associated with mortality, carcinogenicity, thyroid, pancreatic and developmental effects (Lau et al. 2007). PFCs have been quantified in the breast milk of Massachusetts women at pptr levels but these values did not exceed the calculated daily infant intake recommended from a risk assessment (Tao et al. 2008). PFCs are both lipophobic (poorly absorbed in fat) and hydrophobic yet some compounds are known to bioaccumulate in marine food chains (Tomy et al. 2004; Houde et al. 2006).

4.5 ORGANOPHOSPHATES (OPS)

Organophosphate pesticides

OP pesticides are used to control insect and crustacean pests by disabling their nervous systems. Diazinon, used agriculturally, is also a powerful neurotoxin for rainbow trout and has a high toxicity, particularly to birds.

Ernst et al. (2001) compared the toxicity of azamethiphos for a broad range of marine phyla, from bacteria to protistes to echinoderms to polychaetes to crustaceans to fish, and found that the crustaceans were the most susceptible. The toxicity of azamethiphos to planktonic lobster larvae has been well researched (Burridge et al. 1999; Burridge et al. 2000). Azamethiphos is also acutely toxic to adult lobster at 25 ppb over 15 to 20 minutes, which is a much lower concentration than the 300 ppb applied to fish pens for control of "sea lice" infestations of salmon (Burridge et al. 2000). The effect of azamethiphos on adult female lobsters was found to be most adverse from June through September, which coincides with the moulting, breeding and larval release period in nature (Burridge et al. 2005). It was found that repeated applications of azamethiphos to female lobsters in the lab to hundredfold lower concentrations than used in the sea pens (10 ppb) resulted in 43 to 100% mortality (Burridge et al. 2008).

Plume studies in the Bay of Fundy following a realistic experimental release of azamethiphos and rhodamine tracer dye from salmon pens demonstrated that the dispersing water was not toxic to a benthic crustacean after 20 minutes with few fatalities earlier (Ernst et al. 2001). A benthic crustacean (amphipod) was found to have a lethal dose for 50% of the individuals after a 10-day sediment exposure to approximately 200 ppb azamethiphos (Mayor et al. 2008). Dispersion and toxicity studies have shown that azamethiphos use in salmon pens presents a low to medium risk to the surrounding environment due to its water solubility and low persistence, with an estimated half-life of nine days (Burridge et al. 1999; Jackman et al. 2001).

Diazinon and azamethiphos do not bioaccumulate in aquatic food chains and have low persistence in the environment.

Organophosphate flame retardants

OPs have not been quantified in the marine food chain, although the physicalchemical properties of TCPP and TDCP indicate the possibility of their bioaccumulation in marine organisms (Reemtsma et al. 2008). Support for this prediction is provided by measurements on Baltic herring collected in 2007 with levels at 42-150 ppb TCPP and 2-3.4 ppb TCEP in lipid, although TBEP was below the detection limit (Sundkvist et al. 2010). Organophosphate flame retardants are carcinogenic, toxic, and environmentally persistent (WHO 1998).



4.6 PYRETHROIDS

Pyrethroids are very toxic synthetic insecticides with marine crustaceans being far more susceptible than fish. Pyrethroids are rapidly metabolized in nature and therefore not accumulated within the food web. The toxicity to planktonic lobster has been well documented for cypermethrin (Burridge et al. 2000). Deltamethrin was found to be extremely toxic to lobster larvae, sand shrimp and a benthic amphipod (Fairchild et al. 2010). Plume studies at aquaculture sites, using the amphipod *Eohaustorius estuarius* as the indicator organism, found that almost all the cypermethrin water samples collected up to five hours post release were toxic (Ernst 2001).

There have been a number of studies demonstrating the adverse effects of cypermethrin on planktonic species at concentration levels well below the fish pen application level of 5 μ g/L (Medina et al. 2002; Willis and Ling 2004). However, in the case of holoplanktonic organisms, such as copepods, it is reasoned that the short duration and finite area adversely affected by cypermethrin would be recolonized in a relatively short time from neighbouring waters (Barata et al. 2002; Medina et al. 2004). However, the meroplanktonic larvae of many benthic organisms, such as lobster, crabs and shrimps, seek the surface waters during a relatively brief time and represent an entire year class. Lobster release their larvae into the surface waters for about a month, temperature dependent, preferably at the heads of bays to take advantage of the warmer waters but this places them in optimal aquaculture sites.

There are few studies on the effects of aquaculture pesticides on bottom-living organisms underneath the fish pens. Cypermethrin has a lower water solubility and therefore is expected to adsorb to particulate material (Maund et al. 2002) and settle to the bottom. The benthic amphipod *Corophium volutator* was subjected to a 10-day whole sediment bioassays and found to have a LC_{50} (lethal concentration in 50 percent of test organisms) of 5 µg cypermethrin/kg wet sediment (Mayor et al. 2008). Nevertheless, the direct observation of adverse effects of these aquaculture pesticides on benthic species and therefore community structure in coastal regions has not been studied.

4.7 PHARMACEUTICALS

The acute or immediate toxicity of most pharmaceuticals are fairly well known in lower trophic level aquatic organisms (Fent et al. 2006). However, environmental concentrations are 1000 to 10 million times lower than those used in shortterm toxicity tests. Very little is known about the chronic or long-term effects of pharmaceuticals to aquatic life and to fish in particular. Even less is known about potential interactive effects of mixtures of these contaminants. Pharmaceuticals do not generally pose a risk of bioaccumulation in the aquatic food chain with the possible exception of the analgesic diclofenac and carbamazepine in fish (Schwaiger et al. 2004). A review of the known contaminants entering the Bay of Fundy from sewage treatment plants is available (Kidd and Mercer 2012).

Analgesics

Ibuprofen has the highest acute toxicity to algae and zooplankters at 8-10 ppm compared to fish at >100 ppm (Fent et al. 2006).

Synthetic and natural steroidal hormones

The steroidal estrogens of human origin are known endocrine-disrupting chemicals that bind with estrogen receptors of wild species with the potential to have an effect at extremely low concentrations. The synthetic 17α -ethynylestradiol has been found to be an order-of-magnitude more potent at inducing a vitellogenin response (egg protein production) in juvenile female trout (Thorpe et al. 2003). The steroidal estrogens, altogether, have an accumulative effect. It is well established that male fish subjected to estrogens in freshwater take on secondary female characteristics that results in reproductive impairment (Mills and Chichester 2005). More recently, Kidd et al. (2007) have demonstrated that a three-year exposure of minnows to parts per trillion concentrations of ethynylestradiol (ng/L) in an experimental lake resulted in the near extinction of this population. As we have seen earlier, treated sewage water ends up in our groundwater and marine coastal waters, which is of concern for municipalities, human consumption and marine fish. The presence of androgens in the water is known to cause pheromonal responses of young male salmon at concentrations as low as 0.003 pptr of testosterone (Moore and Scott 1991). This response of immature males could result in biochemical, as well as behavioural changes, that would be energetically wasteful and increase their risk to predation (Kolodziej et al. 2003).

Antibiotics

Microorganisms are developing an increasing resistance to antibiotics used in human and veterinary medicine and from large-scale livestock operations and aquaculture (Tenover and Hughes 1996; Reboucas et al. 2011; Beleneva 2011). Rhodes et al. (2000) have provided evidence for a connection between human and aquaculture environments in Northern England via tetracycline resistanceencoding plasmid exchange between *Aeromonas* species and *Escherichia coli*. The increasing presence of antibiotic resistant bacteria has the potential to become a problem for food production and a human health hazard. Oxytetracycline resistant species of *Pychrobacter* were found to be able to grow in Bay of Fundy sediments within 100 m of salmon pens at concentrations up to 160 μ g/ml (Friars 2002). Antibiotics are known to have toxic effects on phytoplankton, particularly cyanobacteria; however, effects in zooplankton are minimal (Lanzky and Halling-Sorensen 1997; Guo and Chen 2012).



Biocides

Avermectins affect neural transmission particularly in crustaceans (Burridge and Haya 1993; Mayor et al. 2008), although they are also toxic to fish at higher concentrations (Palmer et al. 1987). Ingestion of emamectin benzoate by lobsters has been found to induce premature moulting in lobsters (Waddy et al. 2002), which would interfere with the timing of mating and larval release. Emamectin benzoate is not expected to bioaccumulate in marine food chains. Avermeetins can concentrate in sediment underneath or adjacent to sea pens due to the settling of uneaten food pellets, fish feces and particulates, particularly if the currents are weak. Emamectin benzoate is known to be persistent in anaerobic soils, with a half-life of 427 days, which indicates a potential for accumulation in the anoxic sediments often present near sea pens (see Burridge et al. 2010). An individual sea pen has a limited impact, being confined to the sediments and fauna in the immediate area (SEPA 1999). In bays with a high density of fish pens, sheltered from weather-related, resuspension events, and with reduced dispersion from ocean currents, such as in the Quoddy region, there would be an increased probability of environmental harm from avermectins to the benthic or bottomliving community. The benthic community would likely be selectively reduced of the more sensitive crustacean fauna such as shrimp, crabs, and lobsters; however, more studies are required to resolve these issues.

4.8 METALS AND ORGANOMETALS

Butyltin compounds

The persistence of TBT in water is low with a half-life of a few days due to its low solubility, whereas it may remain in sediments for years. Tributyltin is toxic to wide range of organisms from invertebrates to fish (Hall and Pickney 1985). TBT, as the common oxygenated form bis (tributyltin) oxide, is toxic to marine phytoplankton at 0.3 to 1 ppb in seawater (US EPA 1985). Tributyltin is also extremely toxic to the larvae of crustaceans with growth cessation in lobster at 1 ppb in seawater (US EPA 1985). It is known to be immunotoxic, hepatotoxic and neurotoxic in fish and marine mammals (Kannan et al. 1997; Nakata et al. 2002). TBT has been found to adversely affect feral oyster growth in bays (Alzieu et al. 1986) and induce masculine characteristics in female marine snails (Stickle et al. 1990; Titley-O'Neal et al. 2011b). Sixty-six percent of the female dogwhelks in Saint John Harbour, Bay of Fundy, N.B., exhibited male sexual characteristics (Prouse 1996). Furthermore, dogwhelks were noticeably absent from many other Maritime harbours with seemingly ideal habitat (Prouse 1996).

Although tributyltin bioaccumulates 2- to 11-thousand fold in invertebrates, fish and marine mammals, it does not appear to biomagnify in marine food chains due to differing degrees of biodegradation in various organism types and at different trophic levels (Tanabe 1999; Murata et al. 2008).

Mercury and methylmercury

Mercury is likewise very persistent in sediments as evidence shows for Passamaquoddy Bay, New Brunswick (Sunderland et al. 2010). Mercury is extremely toxic when it is converted to methylmercury in estuarine and coastal sediments by sulfate reducing bacteria (Compeau and Bartha 1985) and little understood processes in the marine water column (Topping and Davis 1981; Monperrus et al. 2007). Most sediment values measured in the Gulf of Maine are well below the threshold effects levels of 0.13 ppm and a probable effects level of 0.7 ppm dry weight derived for marine biota (MacDonald et al. 1996). Mercury toxicity levels in seafood are exacerbated by the biomagnification of methylmercury in aquatic food webs (Bargagli et al. 1998; Campbell et al. 2005; Hammerschmidt and Fitzgerald 2006) such that the terminal predators can contain unacceptable quantities for human consumption (Sunderland 2007). Methylmercury is a strong neurotoxin to humans (Grandjean et al. 1997). The tunas, swordfish, sharks and fish-eating mammals at the top of the marine food chain attain the highest levels of methylmercury; however, these levels do not appear to be detrimental to these predators themselves and there is some evidence that they have evolved selenium sequestration to counter the neurologic effects of methylmercury (Palmisano et al. 1995; Ikemoto et al. 2004; Branco et al. 2007).

4.9 TRACE ELEMENTS

Certain trace metals are essential for the healthy growth of plants and animals, such as copper, zinc, cadmium and iron but concentrations above certain thresholds can be toxic. Many trace elements are toxic to organisms such as arsenic, lead, mercury and selenium. It is well known that the methylated forms of arsenic and mercury are the most toxic. Thus, knowing the total concentration (all forms) of a potentially toxic trace element may be insufficient to assess its toxicity. Marine fish, in general, are equivalent to or 10 to 100 times less sensitive to toxic metals compared to invertebrates that have been studied, chiefly crustaceans and molluscs (Taylor et al. 1985). Moulting and reproduction in crustaceans are adversely affected by certain trace metals (Fingerman et al. 1996). Many trace metals are accumulated in organisms both directly from seawater and from their food, but they are not biomagnified in marine food chains because they are efficiently excreted (Wang 2002). Selenium can be an exception to this in certain trophic conditionis (Stewart et al. 2004).

Sediment studies of the late 1980s found that lead and chromium values were above the median biological effects range established by Long and Morgan (1990) in several bays and harbours of the Gulf of Maine. Measurements taken by the Gulfwatch program found that the Mussel Watch continent-wide mercury level of concern (85th percentile, 0.296 ppm dry weight) was exceeded in four urbanized regions: Penobscot Bay, Casco Bay, Great Bay estuary, and in and near Boston



Harbor (Jones et al. in prep). Elevated levels of mercury and lead at some locations remain a concern. Nevertheless, most trace metals do not present an environmental concern in the Gulf of Maine.

4.10 MISCELLANEOUS CONTAMINANTS

Stimulants

Nicotine is toxic to insects (Tomlin 2005) and thereby to marine crustaceans. The present concentrations of caffeine and nicotine in seawater do not appear to be of immediate concern; however studies of toxicity and biodegradation in marine invertebrates are needed.

Disinfectants

Triclosan is of concern because it may increase bacterial resistance to antibiotics in the environment (Dann and Hontela 2011). Apart from its antibacterial properties (>0.21 ppb), triclosan also inhibits photosynthesis in diatoms (>0.42 ppb) that are essential for productive spring blooms in boreal marine shelf ecosystems (Ricart et al. 2010).

Aircraft deicing fluid

In a study of sensitivity to 5-MEBT (aircraft deicing fluid), the marine bacteria *Vibrio fischeri* was most sensitive at a concentration of 7 ppm (15 min EC_{50}), followed by flathead minnows at 38 to 65 ppm (96hr LC_{50}) and the daphnid, *Ceriodaphnia dubia* at 102 to 108 ppm (48hr LC_{50}) (Cornell et al. 2000). In a similar study, the IC25, the point where 25 percent of experimental algae cells stop dividing, of a green alga, *Selenastrum capricornutum*, was 23 ppm (Cancilla et al. 2003).

Insect deterrent

DEET has been shown to be toxic to the freshwater zooplankter *Daphnia magna* at an LC_{50} 48hr of 160mg/L (Seo et al. 2005), which is well above the concentrations observed in Massachusetts river water of >0.04 and 0.1 µg /L range (Barnes et al. 2002; Kolpin et al. 2002).

5. Actions and Responses

5.1 LEGISLATION

Major pieces of legislation to restrict complex mixtures of chemical contaminants from entering U.S. and Canadian waters from industrial effluents, agricultural runoff, mining seepage and municipal wastewater treatment plant discharges were enacted in the late twentieth century as the United States *Toxic Substances* Control Act (1976) and the Canadian Environmental Protection Act (1999) and Fisheries Act (1972).¹ The EPA and Environment Canada use the Toxic Substances Control Act and the Fisheries Act, respectively, to impose guidelines and regulations, testing requirements, reporting and recording of chemical mixtures or substances to protect human health and the environment. These laws are enforced both by monitoring for select deleterious substances and by biological toxicity testing on effluents with standard organisms, usually rainbow trout and Cladocera (crustaceans). The EPA and individual states register pesticides under the Federal Insecticide, Fungicide and Rodenticide Act and establish tolerances for pesticides in food under the Federal Food, Drug and Cosmetic Act. In Canada, pesticide use is overseen by the Pest Control Products Act and the Food and Drug Act, both administered by Health Canada. The EPA establishes standards of wastewater release in surface waters under the Clean Water Act. The United States Pollution Prevention Act established a policy of pollution prevention, wherever possible, at source, as in Canada. The proliferation of synthetic chemicals and their ofteninadvertent introduction to the environment has caused rising concern by the public, medical profession and in the scientific community for both human and ecosystem health. This has led to the regulation of organic contaminants by both national and international agencies (Table 7). In 2001, the United Nations Environmental Programme Governing Council banned the use of 12 organic compounds that are resistant to degradation, bioaccumulate, are toxic, and are subject to long-range atmospheric transport (Stockholm Convention 2012). The so-called "dirty dozen" are aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, polychlorinated biphenyls, polychlorinated dibenzop-dioxins, polychlorinated dibenzofurans and polychlorinated bornanes (UNEP 2012). It has been suggested that carcinogenic polycyclic aromatic hydrocarbons, brominated flame retardants and butyltin should be included. The Organisation for Economic Co-operation and Development (OECD) and the United Nations, through the International Maritime Organization (IMO), also evaluate and classify the hazards of industrial chemicals that are shipped worldwide under international agreements, such as MARPOL 72/78.

¹ This discussion of legislation reflects the situation as of May 2012.

TUDIE 7. ACIONS and responses related to containing in the Guil of Maine.	Table 7:	Actions and	responses	related to	contaminants	in the	Gulf of Maine. ²
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CONTAMINANT	RESPONSE
Polyaromatic hydrocarbons (PAHs)	 Regulated by the <i>Oil Pollution Act</i> (U.S.) and the <i>Canada Shipping Act</i> within the 200 nautical mile Exclusive Economic Zone. Regulated by Canadian <i>Fisheries Act</i> Section 36 and Canadian interim sediment quality guidelines and probable effects levels (CCME 1999). Regulated by the International Convention for the Prevention of Pollution from ships (MARPOL) and ratified by Canada and the United States.
Polychlorinated biphenyls (PCBs)	 PCBs voluntarily restricted in North America in the 1970s, followed by legislative restrictions in United States and Canada in 1976-77. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004.
Polychlorinated bornanes (CHBs)	 Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004. Germany is the only country to set an acceptable limit of 0.1 ppm (lipid) in food for human consumption.
Dioxins and difurans (PCDDs and PCDFs)	 Pulp and paper regulations introduced by Canada in 1972 and 1992 to reduce release into the environment. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004. The Canadian tissue residue guidelines for the protection of mammalian and avian consumers of aquatic food were established in 2001 (CCME 2001).
DDT and its metabolites	 Banned in Canada in 1970 and in the United States in 1972. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004 but limited use in disease vector control in tropics continues to this day.
Chlorobenzenes (CBs)	 Agricultural usage was discontinued in Canada in 1972 and in the United States in 1982. Environment Canada lists it as toxic in the <i>Canadian Environmental Protection Act</i>. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004.
Hexachlorocyclohexanes (HCHs)	 Canadian manufacturing discontinued in 1972, followed by a spray ban in Canada and the United States in 1976. Lindane currently restricted to minor use as an alternate backup treatment for lice and scabies in humans. Banned globally at Stockholm International Convention on Persistent Organic Pollutants in 2004.
Chlordanes (CHLs)	 Use in the United States discontinued in 1986 except for treatment of fire ants which was terminated in 1995. Use for crops in Canada discontinued in 1976 and registration discontinued in 1985. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004.
Mirex	 Banned in the United States and Canada in 1978. Banned globally at the Stockholm International convention on Persistent Organic Pollutants in 2004.
Aldrin/Dieldrin	 Banned in Canada and the United States in 1974 except for controlling termites and totally banned in 1987. Registered use in Canada limited after 1970 except for termite control, which ended in 1984. Banned globally at the Stockholm International Convention on Persistent Organic Pollutants in 2004.
Organobromines (PBDEs)	 Discontinued use of Penta-BDE in the United States and other industrially developed nations by 2007. Octa-BDE also discontinued; Deca-BDE still in use. Penta and Octa BDE will soon be added to the list of banned chemicals included in the Stockholm International Convention on Persistent Organic Pollutants

 $^{^{\}scriptscriptstyle 2}$ This table reflects the situation as of May 2012.

5. Actions and Responses

CONTAMINANT	RESPONSE
Perfluorinated compounds (PFCs)	 PFOS-based compound production voluntarily stopped by a major corporation (3M 2000). New law formulated to regulate PFOS (US EPA 2002). PFOS and POSF banned by the Stockholm Convention on Persistent Organic Pollutants in 2009. PFOA – EPA requested 8 manufacturers voluntarily cease production (2006) (http://www.epa.gov/opptintr/pfoa/pubs/pfoastewardship.htm) PFCAs continue to be produced worldwide.
Organphosphate pesticides	 OPs generally banned for residential use by EPA in 2001 but used for fruit and vegetable agriculture and for pest control in public places. Diazinon banned in the United States in 2004 for all domestic and non-agricultural uses such as sod farms and golf courses. Agricultural use of azamethiphos to be restricted in fruit and vegetable farming (EPA in 2001). Azamethiphos was used for "sea lice" in finfish aquaculture in Canada until 2005 but in 2009 reinstated on an emergency status (Health Canada, Pest Management Regulatory Agency, 2009).
Pyrethrum and Pyrethroids	 Pyrethrum was allowed temporary registration for salmon aquaculture in the 1990s. Cypermethrin is used in Maine aquaculture operations for "sea lice" control but it is not registered for use in aquaculture in Canada. Deltamethrin was given emergency registration in Canada in 2009 (Health Canada, Pest Management Regulatory Agency, 2009).
Avermectins	 Ivermectin was used as an "off-label" treatment in the 1990s regulated by Health Canada under veterinary prescription. Emamectin benzoate was regulated until 2009 under Health Canada's Emergency Drug Release program; in June 2009 it was registered for use under the <i>Food and Drugs Act</i>. Emamectin benzoate is regulated by the United States Food and Drug Administration (FDA) under an Investigational New Animal Drug (INAD) exemption.
Pharmaceuticals	 FDA guidance for environmental assessment necessary if any human drug is expected to exceed 1 ppb wet weight in the aquatic environment (FDA-CDER 1998). FDA have required environmental assessments of veterinary drugs since 1980 (Boxall et al. 2003).
Organotin compounds	 The US Organotin Anti-fouling Paint Act of 1988 and similar Canadian legislation in 1989 regulated tributyltin application to boats less than 25m in length. The International Maritime Organization proposed to ban organotin antifouling paints in 2003 and ban their presence on ships by 2008 (http://www.imo.org/OurWork/Environment/Ati-foulingSystems?Documents/FOULING2003.pdf).
Methylmercury	 Canada has permissible limits for total mercury in seafood for human consumption. For swordfish, tuna (fresh or frozen), marlin, escolar, shark and orange roughy, the limit is 1 ug/g wet weight). For all other species, the limit is of 0.5 µg/g wet weight. The FDA in the United States has an action level of 1 µg/g wet weight for methylmercury. There is basically little difference between the two countries because most fish have more than 90% of their mercury content in the methylated form. Northeastern United States and the Maritime Provinces issued human mercury consumption advication in fish in 1008 (NESCALIM 1008).



Human Health and Seafood Consumption

As a result of the potential health risks associated with the consumption of contaminated seafood, the Canadian Food Inspection Agency (CFIA) and the United States Food and Drug Agency (FDA) have implemented tolerance levels for human consumption of certain toxic substances in seafood (Tables 8 and 9). These agencies monitor seafood on a regular basis and if the action or tolerance levels are exceeded, legal action is taken to remove these products from the market. There is no review mechanism associated with this regulatory monitoring.

Table 8: Action or tolerance levels for recognized toxic substances in seafood (µg/g wet weight, ppm)

CONTAMINANT	CANADA ^a	UNITED STATES ^b
Aldrin/dieldrin	>0.1	>0.1
Chlordane (CHLs)	>0.1	>0.3
Chloredecone	>0.1	>0.3(0.4)°
Endrin	>0.1	>0.3
DDT and its metabolites	>5.0	>5.0
Heptachlor/heptachlor epoxide	>0.1	>0.3
Hexachlorobenzene (HCB)	>0.1	>0.3
γ-Hexachlorocyclohexane (Lindane)	>0.1	—
Polychlorinated biphenyls (PCBs)	>2.0	>2.0 ^d
Polychlorinated bornanes (CHBs)	>0.1	>5.0
Polychlorinated dibenzo-p-dioxins (PCDDs)	>0.00002	—
Polycyclic aromatic hydrocarbons (PAHs)	>0.003 ^e	
Methylmercury (MeHg)	>0.5	>1.0
Mirex	>0.1	>0.1

^a Source: CFIA 2005.

^b Source: FDA 2011.

^c Action level for crustaceans, such as shrimp and lobster. Level for molluscs, such as mussels, in brackets.

^d Tolerance not action level.

^e Toxic equivalents of benzo(a)pyrene.

Table 9: Legal Limits (Canada) and guidance levels (United States) for hazardous metals in fish and fisheries products (μ g/g wet weight) (from CFIA 2005 and FDA 2011).

POLLUTANT	CANADA	UNITED STATES
Arsenic (As)	>3.5	>76(86) ^a
Cadmium (Cd)		>3(4) ^a
Chromium (Cr)		>12(13) ^a
Lead (Pb)	>0.5	>1.5(1.7) ^a
Nickel (Ni)		>70(80) ^a

^a Values for bivalve molluscs such as decapods and mussels (in brackets).

5.2 RESEARCH AND MONITORING

The following organizations have had the most extensive monitoring programs in the Gulf of Maine region.

Gulfwatch

Gulfwatch is a chemical contaminants monitoring program organized and administered by the Gulf of Maine Council on the Marine Environment (Gulfwatch 2012). Since 1993, Gulfwatch has measured contaminants in blue mussels (*Mytilus edulis*) at 64 locations to assess the types and concentration of contaminants in coastal waters of the Gulf of Maine. It is one of the few monitoring programs and the only one in the Gulf of Maine to be coordinated across international borders. Gulfwatch is coordinated and conducted by scientists and managers from agencies and universities around the Gulf. The program operates under the guidance of the Gulf of Maine Council's Gulfwatch Contaminants Monitoring Subcommittee and has been supported variously with funding from the Gulf of Maine Council on the Marine Environment, the U.S. Environmental Protection Agency and Environment Canada. Gulfwatch measures 12 low-molecular-weight PAHs and 12 high-molecular-weight PAHs, 22 PCBs, 16 chlorinated pesticides, and 9 metals at 38 sites along the coast of Massachusetts, New Hampshire, Maine, New Brunswick, and Nova Scotia.

Mussel Watch

Mussel Watch represents the longest-running continuous contaminant monitoring program in American coastal and Great Lakes waters. The project was developed to analyze chemical and biological contaminant trends in sediments and bivalve tissues collected at over 300 coastal sites from 1986 to present. Attributes or variables monitored include sediment and bivalve tissue chemistry for over 100 organic and inorganic contaminants; bivalve histology; and *Clostridium perfringens* (pathogen) concentrations. This project regularly quantifies PAHs, PCBs, DDTs and its metabolites, CHLs, other chlorinated pesticides, TBT and its metabolites and toxic trace elements at a total of 12 locations within the Gulf of Maine, of which 3 are located close to Gulfwatch sampling sites.

Environmental Protection Agency National Coastal Condition

The Environmental Protection Agency's National Coastal Condition program measured contaminants in sediments of the Gulf of Maine between 2000 and 2006 and in 2010 and reports the results regularly through their Coastal Condition reports (US EPA 2012b).

Environment Canada

Environment Canada has measured contaminants in eggs of nesting seabirds in the Bay of Fundy since 1972 as part of a Canada-wide program initiated in 1968

(Environment Canada 2003; Burgess et al. 2013). Monitoring includes Atlantic puffin, double-crested cormorant, herring gull, eider duck, and Leach's storm-petrel, which each feed in different food webs in the marine environment.

Massachusetts Water Resources Authority

Massachusetts Water Resources Authority has been measuring freshwater variables in runoff since 1991.

National Atmospheric Deposition Program, Mercury Deposition Network

The Mercury Deposition Network has been measuring atmospheric mercury deposition in the eastern United States and Canada since 1996.

Biodiversity Research Institute

The Biodiversity Research Insitute in Maine has been measuring mercury in selected bird species in the northeastern United States since 1995.

6. Indicator Summary

THERE ARE TOO MANY CHEMICAL INDICATORS OF CONTAMINATION TO PRACTIcally list in a table so they have been grouped into a variety of contaminants, contaminant categories and combined effect of contaminants. There are several other approaches that could be taken, such as using environmental indicator categories, e.g., atmosphere, rain, seawater, sediments and biota. Contaminant concentrations in the air, rain and seawater are generally so low that the analytical detection becomes a challenge and too expensive for regular monitoring programs. Sediments and biota are more suitable for contaminant monitoring because concentrations are usually higher. Analyses of sediments provide an ideal indication of local pollution and can also provide a stratographic record of past discharges. Biota, particularly widespread species, are also excellent indicators of contamination. Higher trophic level organisms, such as the larger fish, seals and porpoises, are the preferred monitors for contaminants that bioaccumulate, such as methylmercury, organochlorines, organobromines, etc. However, these predators tend to be wide-ranging so a stationary organism, such as the mussel, will give a better spatial and temporal picture of local contaminant sources and overall environmental quality in the Gulf of Maine.

INDICATOR	DPSIR FRAMEWORK	STATUS	TREND
Quantity of toxic contaminants released into the Gulf of Maine	Pressure	Unknown – Many sources of toxic contaminants are not tracked.	Unknown – Cannot determine trend with existing information.
Number of contaminants	Pressure	Poor – Number of contaminants is in the thousands, a challenge for monitoring and responding to impacts.	Worsening – The number of contaminants is increasing, which will likely result in further impacts on the environment.
Concentration of toxic contaminants in marine waters and sediments of the Gulf of Maine relative to more remote (pristine) locations, and where available, national standards and guidelines.	State	Fair – In most areas, except for industrialized harbours, concentrations are similar to more remote locations. There are few national standards and guidelines for toxic contaminants in marine waters.	Unknown – There is limited information on temporal trends of contaminants in sediments and no temporal data on contaminants in seawater.
Concentration of toxic contaminants in marine organisms of the Gulf of Maine relative to more remote (pristine) locations, and where available, national standards and guidelines.	State	Fair – Some organisms have concentrations higher than background levels. Concentrations generally do not exceed national food guidelines. There are limited national standards and guidelines for toxic contaminants.	No trend (limited information) – Available information on a limited number of species shows no clear overall trend.
Presence of contaminants whose use has been banned	State	Fair – Presence of banned contaminants in sediments and organisms remains a concern.	Improving – Levels of banned contaminants such as PCBs, DDTs, HCHs, CHLs and Dieldrin have stabilized or decreased.
Presence of emerging contaminants (e.g., pharmaceuticals, OP flame retardants)	State	Poor – Emerging contaminants have been detected near urban centres of the Gulf of Maine.	Worsening – More emerging contaminants are being detected; treatment plants are ineffective at removing most pharmaceuticals.
Sub-lethal and/or lethal health effects in marine organisms directly attributed to toxic contaminants.	Impacts	Unknown – Lethal and sub-lethal impacts have been observed in some species; total impacts on the marine environment due to toxic contaminants are unknown.	Unknown – The health effects of contaminants on marine organisms and the ecosystem as a whole are largely unknown; skin lesions and sex changes in fish detected in polluted harbours.
Number of seafood consumption advisories or market restrictions due to toxic contaminants.	Impacts	Fair – In recent years, areas of the U.S. northeast have been under fish consumption advisories due to elevated levels of toxic contaminants. Canada has also issued seafood consumption advisories for areas of the Gulf of Maine.	No trend – No clear trend in the number of seafood consumption advisories.
Number of banned or regulated chemicals and substances	Response	Fair – There is an extensive management regime to deal with major toxic contaminants.	Worsening – The number and variety of toxic contaminants is increasing more quickly than monitoring and management efforts can accommodate.

Categories for Status: Unknown, Poor, Fair, Good. Categories for Trend: Unknown, No trend, Worsening, Improving.

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