

Chapter 5

TOXIC CONTAMINANTS



The Abagadasset River's sinuous channel gently winds through western Merrymeeting Bay's tidal marshes. Such scenes hint little at the environmental legacy caused by historical and ongoing contamination. Photo: Slade Moore and John Sowles.

Toxic contaminants are common in drainages with large watersheds, historical industrial use, and sizeable population centers. In the Kennebec Estuary, some contaminant groups warrant concern for public health, ecological functioning, and biodiversity. What conditions make some species, including humans, more vulnerable than others to excessive contaminant uptake? How well have contaminant reduction efforts succeeded? What challenges persist?

Introduction

Since the enactment of the Clean Water Act of 1972, sewage management, pollution treatment, and non-point source pollution reduction have dramatically ameliorated many of the water quality problems for which the Kennebec and Androscoggin Rivers were notorious. Also in 1972, the ban on use of the pesticide DDT (dichlorodiphenyltrichloroethane) in the United States represented an important step in limiting the intentional release of known toxic contaminants into the environment. Yet 36 years after these landmark events, elevated levels of toxic contaminants in the Kennebec Estuary persist. Estuaries often incur a disproportionately high exposure to chemical contamination as a result of their proximity to industry and population centers (Jones et al. 2001). Additionally, physical and chemical conditions in estuaries facilitate the filtering and accumulation of contaminants introduced higher up in the watershed (Larsen and Gaudette 1995; Chester 2000; Applied Biomonitoring 2005). In the Kennebec River, this exposure potential is compounded by the large number of historically permitted contaminant discharges (NOAA 1994).

As our awareness of toxicological risk grows, the list of contaminants identified as warranting ecological and public health concern continues to expand. These contaminants encompass a wide variety of elements and compounds linked to industry, waste disposal, and the routines of everyday life for most people in Maine; many of them are presently considered ubiquitous throughout the physical environment and the biota it supports—including humans.

Of long-standing concern are contaminants that bind to proteins or fats, such as mercury and polychlorinated biphenyls (PCBs). Uptake of these compounds can occur anywhere in the food chain as organisms interact with surrounding water and sediment. Because some contaminants accumulate faster than they can be metabolized, concentrations in individual organisms increase over time in a process known as bioaccumulation. Concentrations in animals at lower trophic (feeding) levels, such as small invertebrates, might not exceed thresholds beyond which the individual's health or survival is compromised. However, each time a predator consumes a prey organism residing at a lower trophic level, the prey's contaminant burden is transferred to the predator. This process is repeated many times during a predator's life, eventually leaving the predator with a higher concentration of contaminants than its prey. As the predator-prey dynamic unfolds at increasingly higher trophic levels, contaminant concentrations magnify considerably. Consequently, apex predators such as eagles, seals, and sharks can incur the greatest risk of impaired health and reproduction due to contaminants in the environment (Jarman et al. 1996; Atwell et al. 1998). When animals lower in the food chain demonstrate elevated concentrations of highly potent toxic contaminants, it is a signal that top consumers, including humans, may be at risk of compromised health (Jones et al. 2001).

Researchers have experimentally demonstrated health effects of many contaminants as they relate to developmental, reproductive, behavioral, and organ function. Yet for most contaminants, uncertainty remains regarding thresholds for unhealthy concentrations in water, soil, air, and animal tissues. Even less well understood is how interactions between two or more contaminants may influence effects on health. Some combinations of contaminants, such as methylmercury and PCBs, interact to produce more dramatic health effects than each individual contaminant would in isolation (Grandjean et al. 2001; Stewart et al. 2003).

Identifying and Monitoring Contaminant Risk

Several monitoring efforts and short-term assessments have shed light on contaminants in the Kennebec Estuary. Monitoring has particular value in providing data that can be tracked over time and can thus be used to identify and address long-standing and emerging ecological and public health concerns. One such effort is Gulfwatch, an international program that since the early 1990s has used the blue mussel (*Mytilus edulis*) as a sentinel species for identifying and tracking contaminants of concern in the nearshore Gulf of Maine (Jones et al. 2001). Other efforts include the MDEP's Surface Water Ambient Toxics (SWAT) program, which the Maine Legislature established in 1993 to determine the nature, scope, and severity of toxic contamination in Maine's surface waters and recreational fisheries (MDEP 2008). In the Kennebec Estuary, most SWAT monitoring has focused on measuring concentrations of persistent and bioaccumulative toxic contaminants in animal tissues.

Concentrations of contaminants in fish and shellfish determined by MDEP programs are used by the MDCDCP, which assesses public health risks associated with human consumption of fish and establishes fish tissue action levels (FTALs) for each contaminant under consideration (Smith and Frohberg 2008). FTALs are maximum contaminant concentrations in fish tissue that can be safely consumed at a rate of one 8-oz. (250 g) meal per week. Should tissue contaminant concentrations for a species exceed an established FTAL the recommended consumption rate is adjusted to avoid toxic effects (Smith and Frohberg 2008). In the discussions that follow, we put fish tissue contaminant concentrations in the context of the most current FTALs to indicate whether these benchmarks have been exceeded. We also provide USEPA screening levels for subsistence consumption. Today's consumption patterns suggest that people of the Kennebec Estuary do not eat in sufficient quantities to qualify as subsistence consumption (i.e., four meals per week for an average lifetime), but we provide subsistence screening levels to illustrate the shift in resource-use potentials (in this case, use of wild foods) from pre-industrial conditions to those of the present.



A solitary great egret (*Ardea alba*) prowls the expanses of a high salt marsh. Biomagnification of persistent contaminants represents the greatest threat to species occupying the highest levels of the food chain. Photo: Slade Moore.

Much less common than contaminant monitoring to protect public health is research directed at linking levels of contamination to potential health effects in wildlife (Evers and Reaman 1998). In conjunction with the MDEP SWAT program, sporadic monitoring efforts have attempted to determine the nature of contamination in some wildlife species (Goodale 2008; MDEP 2008). One recent program focused on characterizing wildlife contaminant uptake and contamination hot spots in major river drainages throughout Maine (Goodale 2008). Previous studies have demonstrated the presence of elevated levels of contaminants in the Kennebec Estuary's sediments and water (Larsen and Gaudette 1995; Larsen et al. 1997; Applied Biomonitoring 2005), but in the sections that follow we focus on research that demonstrated the biological availability of toxic contaminants in fish and wildlife by directly measuring body burdens of contaminants in the estuary's organisms.

Contaminants of Particular Concern

Mercury

When mercury is present in biologically available forms at sufficient concentrations, it can impair the growth, behavior, reproduction, and survival of organisms (USEPA 2001*b*). Mercury occurs naturally in the environment, although increased mercury levels over time have been linked to anthropogenic activities (USEPA 2001*b*). Releases of mercury directly into surface waters have been attributed to activities associated with pulp and paper mills, wastewater treatment, leather tanning, electroplating, and chemical manufacturing (USEPA 2001*b*). Disposal of solid waste and incinerator ash and the application of sludge, fertilizers, and fungicides are direct sources of mercury to soils (USEPA 2001*b*). Mercury in sediments can also be mobilized after disturbances such as dredging and flooding (USEPA 2001*b*). Atmospheric deposition of mercury to surface waters and soils originates largely from coal-fired industrial plants and incinerators (USEPA 2001*b*). Almost half of the atmospheric mercury deposition in the northeastern United States is thought to originate from within the region, with about 30% coming from the United States outside the region and 23% contributed by the global atmospheric reservoir (Pilgrim et al. 2000).

Through chemical and biological processes, inorganic mercury converts to a much more biologically available and potentially toxic form known as methylmercury, which is assimilated by phytoplankton and zooplankton and is then subject to accumulation and biomagnification throughout the food chain (Boudou and Ribeyre 1985; CCME 2003). For high trophic level consumers such as piscivorous fish, birds, and mammals, feeding is the most important route of methylmercury uptake and these animals are thought to contain a very high proportion (90–100%) of total mercury concentrations in methylmercury form (CCME 2003).

At least trace amounts of methylmercury are found in humans due to the contaminant's presence throughout the environment, especially in foods such as fish and shellfish. Human exposure to methylmercury at elevated levels is linked to impaired neurological development. Impacts on cognition, memory, attention, language, and fine motor and visual–spatial skills have been observed in children exposed to excessive methylmercury levels (USEPA 2008). In wildlife, elevated levels of methylmercury cause neurological and reproductive health effects (Wolfe et al. 1998).

Polychlorinated Biphenyls (PCBs)

PCBs are a group of chemicals that include 209 individual compounds (known as congeners) with varying harmful effects. Before 1979, PCBs were used in the manufacture of capacitors, transformers, plasticizers, surface coatings, inks, adhesives, pesticide extenders, and carbonless duplicating paper (USEPA 1999). PCBs are persistent in the environment; as a result, they are subject to bioaccumulation in receptor organisms and to biomagnification as they accumulate in higher trophic level predators (Hoffman et al.

1996). The use of PCBs was banned in the United States in 1979 (Rice et al. 2003), but they are still widely detected in wildlife.

Little toxicity information is available for many of the specific PCB congeners (USEPA 1999). Dietary PCB exposures in humans have been linked to developmental effects and the EPA has classified PCBs as a probable human carcinogen (ATSDR 1997; USEPA 1999). Documented PCB effects on wildlife include wasting, immune dysfunction, reduced reproduction, and liver damage (Hoffman et al. 1996). As PCBs are not produced or used in the United States today, the major source of exposure is the redistribution of PCBs already present in soil and water (ATSDR 1997). Some of the highest concentrations of PCBs are found in fish (ATSDR 1997) and fish-eating birds (Goodale 2008).

Dioxins

Dioxins are a class of chemical contaminants that are formed during combustion processes such as waste incineration, forest fires, and backyard trash burning, as well as during herbicide and pesticide manufacture (WHO 2007; NIH 2008). The chemical name for dioxin is 2,3,7,8-tetrachlorodibenzo para dioxin (TCDD), but the term “dioxins” also includes structurally and chemically related polychlorinated dibenzo-para-dioxins and polychlorinated dibenzofurans. Additionally, PCBs with dioxin-like properties (coplanar PCBs) are often grouped with dioxins. Over 400 dioxin-related compounds have been identified; less than 10% of these are considered significantly toxic, with TCDD being the most toxic form (WHO 2007). In Maine, historical releases of dioxins related to paper pulp bleaching represent an important source of the contaminant in riverine environments.

Dioxins have been detected worldwide in nearly all media tested, with the highest levels being observed in soils, sediments, and food such as dairy products, meat, fish, and shellfish (WHO 2007). All people have some body burden of dioxin related to background exposure, but it is not commonly thought that most background levels pose a significant health risk (WHO 2007). Because of their lipophilic tendencies, dioxins occur in greater amounts in organisms with greater amounts of fat (Smith and Frohberg 2008). Owing to their chemical stability, dioxins also have a half-life in the body estimated to be 7 to 11 years (WHO 2007). As a result, dioxins are subject to bioaccumulation and biomagnification as they ascend trophic levels (WHO 2007). In humans, long-term exposure to elevated dioxin levels has been linked to impaired nervous system development and impaired function of the immune, endocrine, and reproductive systems (WHO 2007; NIH 2008). Chronic TCDD exposure is associated with several types of cancer (WHO 2007; NIH 2008). In sufficient amounts, dioxin can impair the development, growth, and survival of fish and wildlife (Janz and Bellward 1996).

According to MDEP, Maine paper mills monitored for dioxin discharges have all come into compliance with the “no-discharge” provision of 38 MRS Section 420, thereby dramatically reducing direct, intentional inputs of dioxins in surface waters. As a result, MDEP has observed a trend of declining dioxin concentrations in some fish species in the Androscoggin, Kennebec, and Penobscot Rivers (MDEP 2007). However, fish sampled from some locations do not follow the pattern of decreasing levels, and dioxin concentrations have also been observed to vary greatly from year to year (MDEP 2007). With limitations of discharges to surface waters, dioxin emissions into the atmosphere now make up the greatest source of release. The New Hampshire Department of Environmental Services estimated that medical waste incineration, combustion from wood-fired boilers, and residential trash burning were responsible for more than 60% of total dioxin emissions in New Hampshire (NHDES 2001). In 2001, MDEP (2001*a*) estimated that about half of dioxin emissions are associated with burning trash and wood for heating; wood-fired commercial boilers and waste incinerators made up the balance of emission sources. Despite reductions in atmospheric emissions and discharges to surface waters from previously important source categories, New Hampshire authorities suggested that dioxin is still produced at levels of concern and continues to accumulate in the environment (NHDES 2001).

Organochlorine Pesticides (OCs)

Used primarily for insect control, organochlorine pesticides (OCs) are extremely persistent in the environment and bioaccumulate in wildlife (Blus 2003). Of the OCs, the particularly notorious DDT was previously in wide use to control mosquitoes, forest pests, and agricultural pests in Maine (Lichter et al. 2006). Under conditions of short-term exposure, DDT is not considered to be acutely toxic and in some regions is used directly on clothes and in soap to control pest species (WHO 1997). It is, however, designated as a probable carcinogen and there is growing evidence that DDT may disrupt reproductive and endocrine function in humans (USEPA 1987; WHO 2005). High DDT levels adversely influence avian and mammalian reproduction by eggshell thinning, infertility, and embryo-fetal toxicity (ATSDR 1994).

Other Contaminants of Concern

A wide range of other contaminants are being evaluated for their potential effect on public health and ecological function. Though sometimes referred to as “emerging” contaminants, they do not all represent classes of chemical compounds that were recently introduced into the environment. Rather, most are the subject of recent investigations resulting from emerging concerns because the latest monitoring data indicate they are relatively ubiquitous in surface waters and in some cases in the tissues of organisms tested, including humans. In a recent study by the U.S. Geological Survey, 95 of these contaminants were identified from sampling sites across 36 states (New Hampshire was the closest site to Maine) (Kolpin et al. 2002).

Polybrominated diphenyl ethers (PBDEs) are one such class of contaminant; these brominated flame retardants are used in plastic resins and textiles (Environment Canada 2004). An assessment by Environment Canada (2004) found no evidence that current levels of PBDEs in the environment harm human health, but the rapid increase in PBDE levels in the environment over the last several years has nevertheless prompted concern. In wildlife, PBDEs have been observed to damage behavioral, kidney, liver, and thyroid functions, and may also represent a carcinogenic threat, although tissue concentrations in experiments that induce these conditions generally exceed those observed in field studies (Darnerud 2003). Despite uncertainties, the use of some forms of PBDEs has already been discontinued, including Deca, which is banned in Maine.

Perfluorochemicals (PFCs) are used as stain repellents, cleaning agents, floor polish, fire-fighting foam, and during the photographic process. Of the PFC congeners, perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) are thought to warrant the greatest potential health concern because of their widespread presence, persistence, and tendency for bioaccumulation. The USEPA advises that PFOs could potentially pose a risk to human health over the long term and has cooperated with industry to phase out this group of contaminants (USEPA 2000*a*).

Other contaminants of emerging concern include nanomaterials, hormones, prions, and certain classes of pharmaceuticals and personal care products (PPCPs) (USEPA 2007*b*). In Maine and elsewhere, some efforts are underway to assess and reduce the risk of these contaminants. For some classes of chemicals, such as PPCPs, the contribution to environmental levels through direct disposal versus excretion and bathing is not known. As a result, the positive impacts of limiting direct disposal of PPCPs are uncertain (USEPA 2007*b*).

Contaminants In Wild-Caught Foods

Contaminants of Ongoing Human Health Concern

Mercury

Relative to other Gulf of Maine coastal areas, the Kennebec Estuary does not appear to represent a hot spot for mercury in wildlife (W. Goodale, BioDiversity Research Institute, personal communication), but monitoring in the system has demonstrated elevated concentrations of mercury in the tissues of several estuarine species that are regularly consumed (MDEP 1996; MDEP 1997; MDEP 2008). One of these, the blue mussel, is ubiquitous in Maine’s rocky intertidal and shallow subtidal areas where the species is harvested for personal and commercial uses. Throughout the Gulf of Maine, wet-weight mercury concentrations in Gulfwatch-sampled mussels in the late 1990s were below the 0.20 parts per million (ppm) MDCDCP FTAL for developmental health risk. However, several of these sites (excluding the Kennebec Estuary) exceeded the more stringent USEPA screening value (0.04 ppm) for subsistence use, which assumes that at least four meals per week are consumed. More recent (2006–2007) Kennebec data indicated that mean mercury concentrations in mussel samples (Fig. 5-1) remained lower than the Maine FTAL and USEPA screening value for mercury.

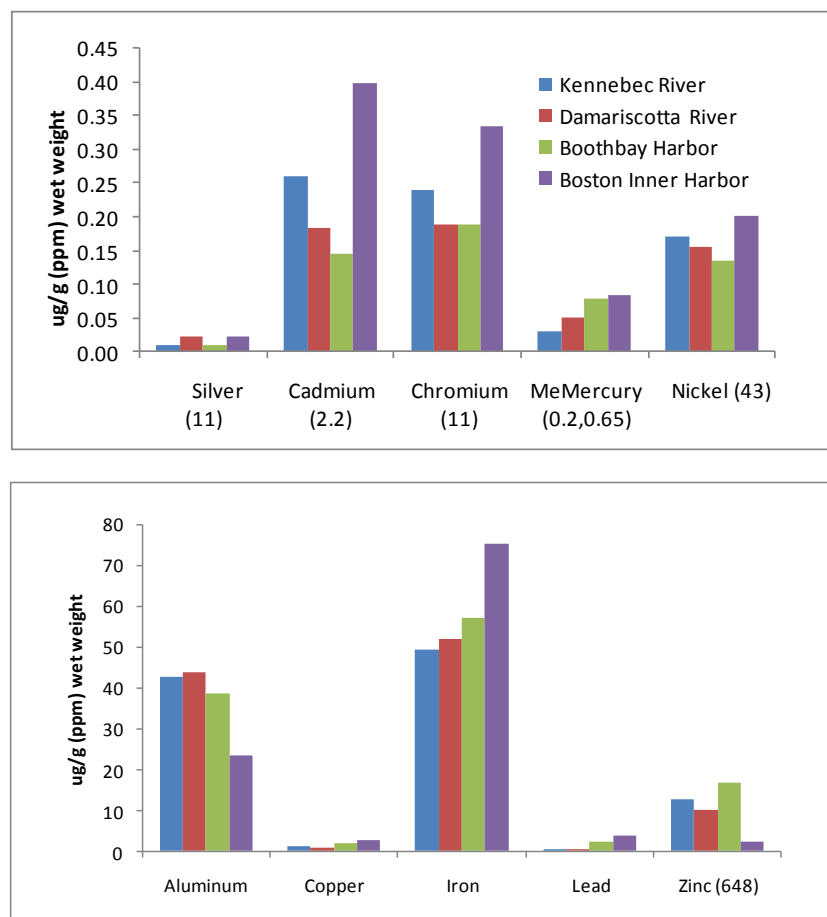


Figure 5-1. Overview of metal concentrations in blue mussel tissue from several Gulfwatch sites. Values in parentheses indicate Maine CDC Fish Tissue Action Levels (FTALs). Aluminum, copper and iron have no assigned FTALs. No FTAL has been developed for lead as it is assumed that there is no safe level. Adapted from Jones et al. (2001) and Gulfwatch (unpublished data).

Another species, American lobster (*Homarus americanus*), represents the backbone of Maine's commercial fishing industry, accounting for nearly 80% (\$279,599,292) of the total landed value of all fisheries in the state (MDMR 2008*b*). Lobster meat tested in the mid-1990s reached or exceeded the USEPA mercury concentration screening level for subsistence consumption (USEPA 2000*b*) and approached the MDCDCP FTAL (MDEP 1997; MDEP 2008) (Fig. 5-2). However, as is the case for several other species (eel, smelt, smallmouth bass, and bluefish), the mercury data for Kennebec lobster meat are somewhat dated, which raises the question of whether they reflect current conditions in the estuary. More recent data in the region are available for lobster tomalley, but not from the Kennebec; despite being designated "Kennebec" tomalley as a result of USEPA sampling conventions, these tissues were actually collected west of Sebasco (MDEP 2008; J. Stahlnecker, MDEP, personal communication), which may be outside the Kennebec Estuary's influence. Concentrations of mercury in lobster tomalley are typically about half those found in lobster muscle tissue; this suggests that mercury concentrations in lobster meat may exceed the subsistence consumption screening level. However, of the species discussed here, lobster of any sort (meat or tomalley) is probably least likely to be eaten in sufficient quantities to qualify as subsistence consumption.

Due to their long residence time in freshwaters and piscivorous feeding habitats (Smith and Tighe 2002), eels are thought to be among the most highly contaminated fish species in Maine rivers (MDEP 2001*b*). Mercury concentrations in Kennebec Estuary eels from Bowdoinham exceeded Maine CDCP mercury FTALs for developmental and adult health as well as the USEPA mercury subsistence consumption screening value (B. Mower, MDEP SWAT program unpublished data; Fig. 5-2). Commercial landings of older eels have declined substantially since the late 1970s peak, although some local harvest still occurs.

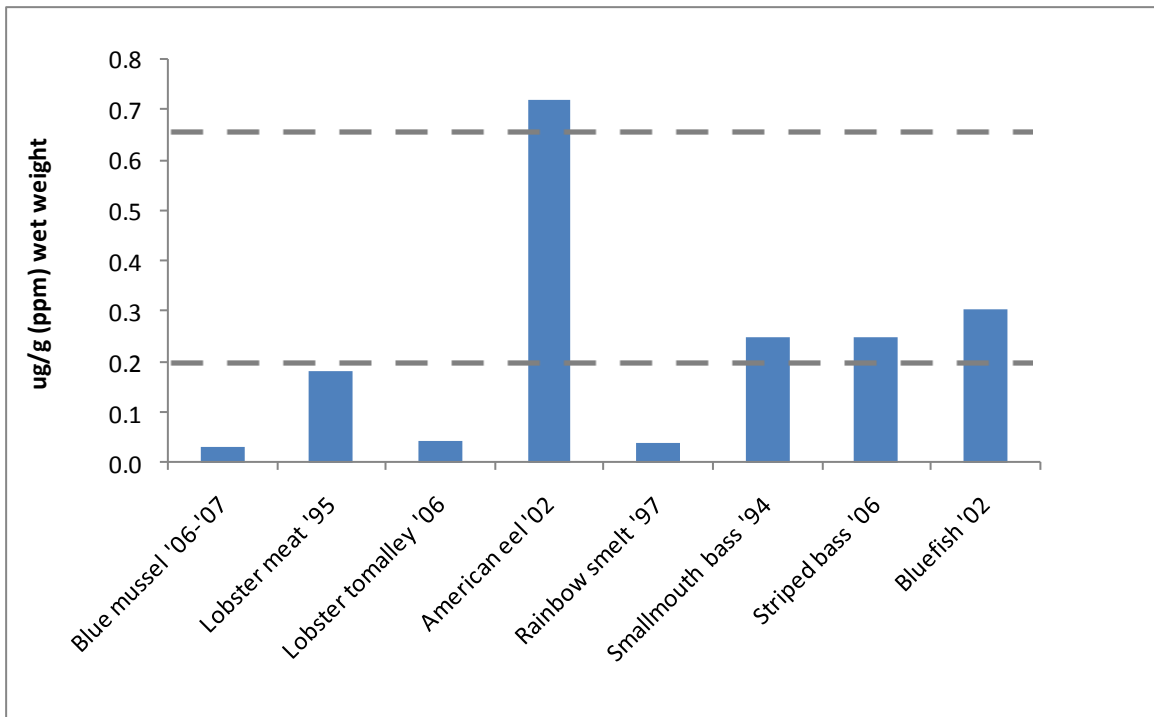


Figure 5-2. Methylmercury concentrations in Kennebec Estuary fish and invertebrate tissues (sampling years indicated). Dashed lines at 0.20 and 0.65 ppm represent Maine CDC Fish Tissue Action Levels for reproductive/developmental health and cancer, respectively. Tomalley data were collected from the Sebasco area, west of the Kennebec Estuary. Adapted from Gulfwatch (unpublished data), MDEP SWAT program data (MDEP 1996, 1997, 2008).

Rainbow smelt mercury concentrations reached the USEPA subsistence consumption screening value, and samples of smallmouth bass, striped bass, and bluefish all exceeded the Maine CDCP mercury FTALs for developmental and adult health effects as well as USEPA subsistence consumption screening value for mercury (MDEP 1996; MDEP 1997; MDEP 2001*b*). Despite their use as a food fish (albeit limited) and for lobster bait, alewife from the Kennebec Estuary have not been sampled for mercury analyses.

Polychlorinated Biphenyls (PCBs)

Lobster tomalley, American eel, rainbow smelt, and smallmouth bass from the Kennebec Estuary each exceeded Maine PCB tissue action levels for reproductive-developmental health and cancer-related risk. Fig. 5-3 indicates total PCB concentrations in species harvested commercially and recreationally that are monitored by MDEP and MCDCP (B. Mower, MDEP SWAT program, unpublished data; Jones et al. 2001; MDEP 2008). Because there is no longer a sampling site for lobster in the Kennebec River, recent PCB data for lobster meat and lobster tomalley were not available. In the case of eels, tissue concentrations were more than 8 times the FTALs for reproductive-developmental abnormalities and 34 times the FTALs for cancer risk. Striped bass and bluefish concentrations of total PCBs demonstrated considerable variability, but consistently exceeded Maine action levels. Striped bass total PCBs tissue concentrations were as much as 4 times the reproductive-developmental action level and 15 times the

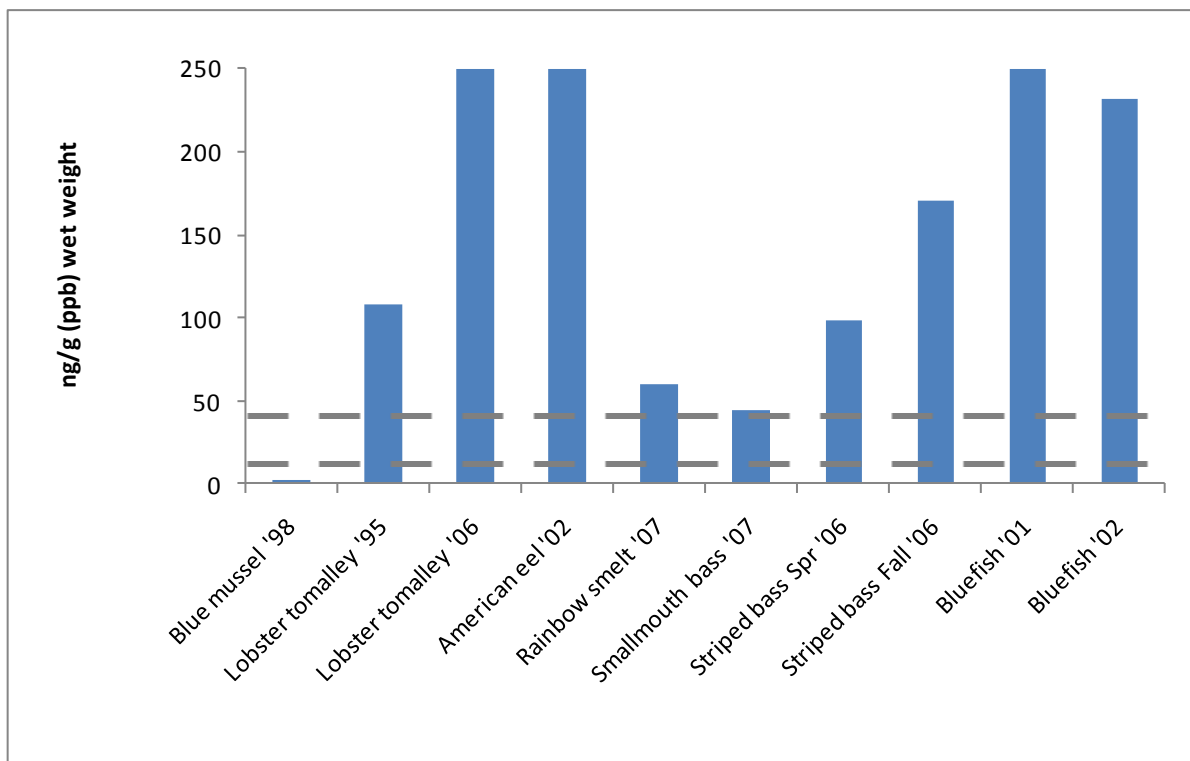


Figure 5-3. Total PCB concentrations (95th percentile upper confidence limit) in Kennebec Estuary fish and invertebrate tissues (sampling years indicated). Dashed lines at 11 and 43 ppb represent Maine CDC Fish Tissue Action Levels for cancer and non-cancer related human health risks, respectively. Tomalley data were obtained from the Sebasco area west of the Kennebec Estuary. Bluefish and striped bass data for two time periods are provided to demonstrate variability. Adapted from MDEP SWAT program, unpublished data, Jones et al. (2001), MDEP (2008).

cancer action level. PCB concentrations in bluefish were as much as 6 times the level associated with reproductive developmental abnormalities and 25 times the cancer-related risk level. Kennebec alewife have not been analyzed for total PCBs. Samples of this species from the nearby Sheepscot drainage (2002) had total PCBs concentrations of 63 parts per billion (ppb), which exceeded both MDCDCP action levels for PCBs.

Dioxins

In 1985, Androscoggin River fish analyzed by MDEP were found to have elevated concentrations of dioxin (MDEP 2007), leading to Maine’s first fish consumption advisory that same year. Since then, MDEP has documented elevated dioxin concentrations in fish and shellfish of the Kennebec Estuary (Fig. 5-4). Early to mid-1990s data for Phippsburg soft-shelled clams, lobster meat, and lobster tomalley, eel from Richmond, and rainbow smelt from Hallowell all had dioxins concentrations high enough to exceed MDCDCP’s new FTAL (0.4 parts per trillion, or ppt) for developmental and reproductive health (MDEP 2007). Lobster tomalley concentrations of dioxins from those same samples were 12 times the 1.5 ppt state action level for cancer risk (MDEP 2007). Paired dioxin and coplanar PCB concentrations provide a more accurate assessment of risk, but were not available for most species. In the case of smallmouth bass they provided a total toxic equivalent value that slightly exceeded the dioxin FTAL for reproductive-

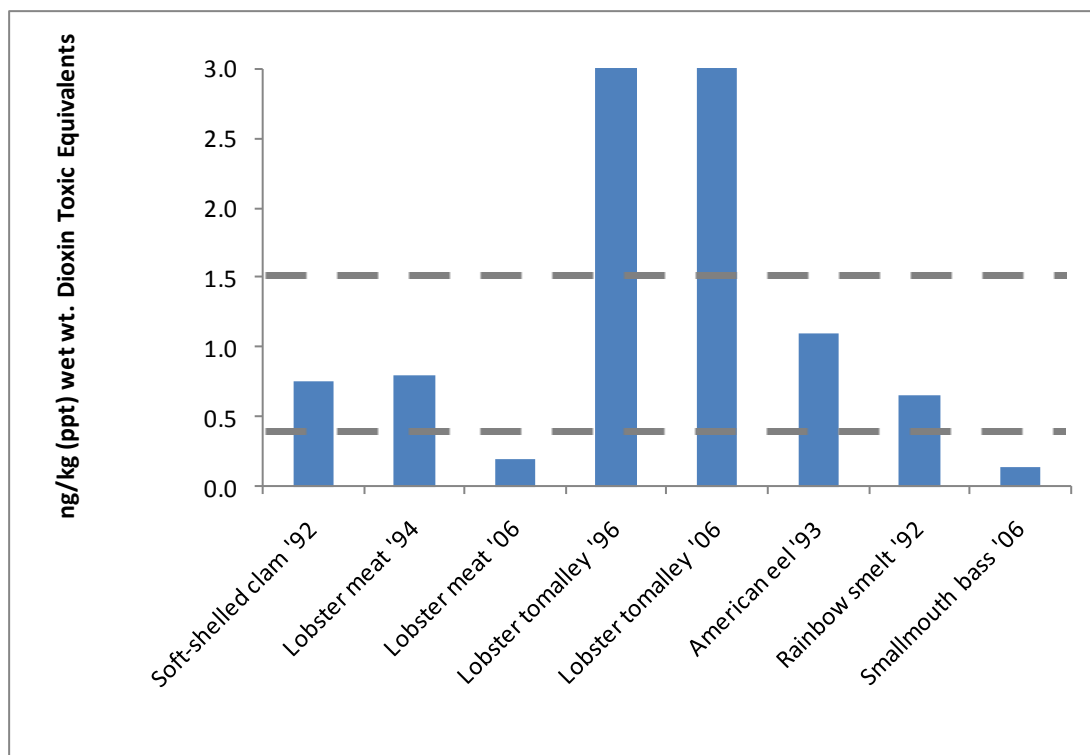


Figure 5-4. Mean dioxin concentrations (95th percentile upper confidence limit) in fish and invertebrate tissues sampled (year indicated) from the Kennebec Estuary. Dashed lines at 0.4 and 1.5 ppm represent Maine CDC Fish Tissue Action Levels for developmental/reproductive health and cancer, respectively. 2006 lobster data were obtained from the Sebasco area west of the Kennebec Estuary and may not reflect Kennebec conditions. Adapted from Mower (2006) and Maine SWAT program, unpublished data.

developmental effects (MDEP 2007). Concentrations of dioxins in lobster meat or tomalley more recent than 1994 data were unavailable for the Kennebec Estuary. We provide 2006 Sebasco data here (Fig. 5-4), which indicated an exceedance in lobster tomalley but not lobster meat. Again, we stress that although

referred to as “Kennebec” tomalley due to USEPA sampling conventions, these Sebasco region data may not reflect conditions in the Kennebec Estuary.

Most of the data presented here predate the legislatively forced reduction of dioxin releases at mills on the Kennebec. Since dioxin abatement, DEP observed declining concentrations of dioxins in some Kennebec fish, most notably smallmouth bass. However, the lack of more current sampling data for all but smallmouth bass hinders attempts to assess whether other species addressed here have also experienced reduced uptake of dioxins. A lack of coplanar PCB data for most target species may also hinder assessments of current contaminant-associated health risk. For alewife, striped bass, and bluefish, no dioxin data were available because MDEP considers these species to have insufficient exposure to riverine dioxins (B. Mower, MDEP, personal communication).

Consumption Advisories

In all of Maine’s inland water bodies, a statewide consumption advisory for fish is currently in effect due to elevated methylmercury concentrations (MCDCP 2008). In addition to mercury, exceedances of MCDCP FTALs for PCBs and dioxin-like contaminants have also prompted advisories. Under these advisories, children less than eight years old and women who are pregnant, nursing, or of reproductive age are advised to avoid eating freshwater fish from Maine’s inland waters. Exceptions are brook trout and landlocked salmon, for which one meal per month is considered safe. MCDCP considers it safe for all other adults and children over the age of eight to consume two meals per month of freshwater fish or one meal per week of brook trout or landlocked salmon.

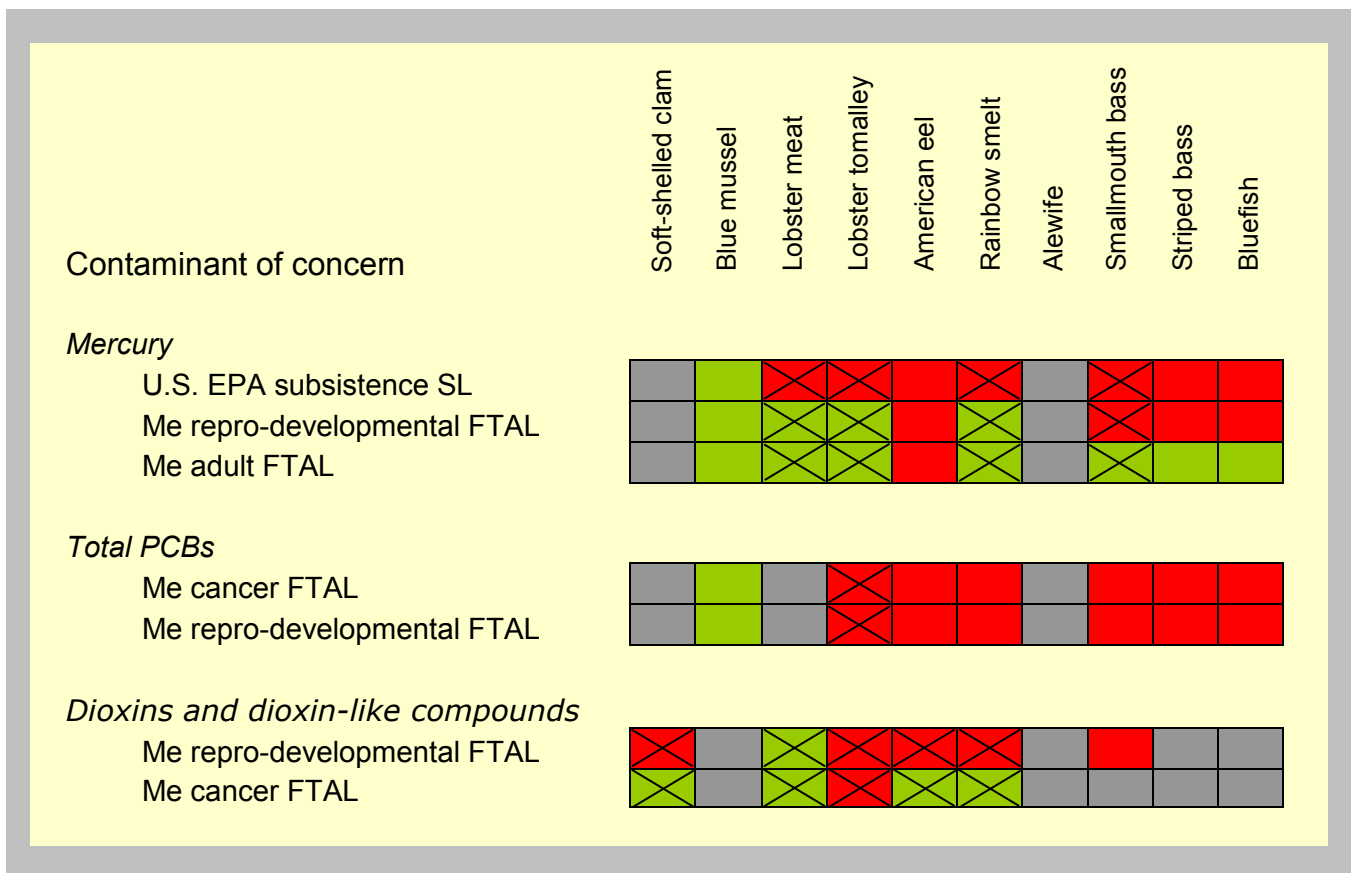


Figure 5-5. Summary of exceedances of Maine CDCP fish tissue action levels and U.S. EPA subsistence fishing screening levels as they relate to several contaminant types in Kennebec Estuary species. Grey = no data, green = no observed exceedance, red = action or screening criteria threshold met or exceeded. Hatched areas denote data less current than 2002. Adapted from Maine DEP SWAT and Dioxin Program data.

To address the variations in concentrations of contaminants, portions of some watersheds have been identified as warranting a more localized approach to managing risk. For the tidal Kennebec River between Augusta and the downstream extent of Merrymeeting Bay (the Chops), the MDCDCP (2008) advisory states “do not eat any fish from these waters,” a warning prompted by elevated concentrations of mercury, PCBs, and dioxins in fish analyzed by the MDEP SWAT and dioxin monitoring programs for that river segment (Fig. 5-5). Most species tested, regardless of where in the estuary they were caught, exceeded the USEPA mercury screening level for subsistence consumption and several exceeded the MDCDCP mercury FTAL for reproductive and developmental health. FTALS for total PCBs and dioxins were also exceeded by several species. These exceedances confirm that sufficiently elevated levels of contaminants are present in the Kennebec Estuary to warrant concern, but for some species of local economic and consumption importance, data are unavailable or dated (Fig. 5-5).

The presence of elevated contaminant concentrations in tissues of wild food products does not, in and of itself, constitute a risk to human consumers. The magnitude of contaminant concentration exceedances, seasonal availability of the species, observed human consumption patterns, and beneficial nutritional properties of the species under consideration are among the many factors considered during the MDCDCP risk evaluation process. Factors that can confound this process include a lack of current sampling data, uncertainty regarding human consumption patterns, and a poor understanding of how different contaminants—including many whose properties have not been adequately assessed—can interact in combination.

Rainbow smelt provide an example of how interacting factors are addressed in risk characterizations. Smelt in the estuary were among several species that recently demonstrated total PCB concentrations exceeding MDCDCP action levels for reproductive-developmental abnormalities and cancer risk (Fig. 5-5). Dioxins in smelt tissues also exceeded the FTAL for reproductive-developmental risk, but data supporting that determination were collected in 1992 and may no longer represent current dioxin body burdens in smelt, given marked reductions in dioxin releases from Kennebec River paper mills. The MDCDCP advises against eating any fish from the Augusta-Chops river segment (where smelt represent a traditional winter fishery of cultural and economic importance), but statewide outreach materials from the agency promote a limited amount of smelt consumption due to the health benefits associated with this species.

Other ocean-going fish species harvested in the Kennebec Estuary are also subject to advisories. Striped bass and bluefish both have mercury and PCB tissue concentrations exceeding FTALs. In response, the MDCDCP advisory for these species adjusts the one meal per week FTAL-based consumption rate down to two meals per month. For most other ocean fish and shellfish, MDCDCP advises that women of reproductive age and young children should not eat more than two meals per week, except for lobster tomalley, which they state should never be eaten by anyone. By implication, lobster meat falls under the heading of “all other fish and shellfish” that can be safely consumed on a weekly basis even by the most sensitive target population. However, Kennebec Estuary lobster meat samples from the 1990s had concentrations of dioxins that would have exceeded the newly implemented 0.4 ppt FTAL. Although MDEP has observed a general trend of declining dioxin concentrations, the pattern of decline is not universal for all sampling locations and species (MDEP 2007). More recent lobster sampling data from the Kennebec Estuary would allow state authorities to better confirm a safe human consumption rate for this important species.

One area of potential confusion arises from the migratory habitats of the species under consideration and the wording of advisories. The Augusta–Chops advisory states “do not eat any fish from these waters,” which indicates that tissue contaminant concentrations from that river segment are so high that they are unsafe at any consumption rate. Yet several species traditionally valued by harvesters using the Augusta–Chops river segment are migratory (e.g., eel, smelt, alewife). These species may occur anywhere



A bank-side angler hooks a non-native white catfish (*Ameiurus catus*) destined for the table. Despite Maine CDCP warnings against fish consumption in Merrymeeting Bay, the Cathance River landing—where no fish consumption advisories are posted—remains a popular destination for “meat” fishers. Photo: Slade Moore.

between the estuary mouth and freshwater segments of the river (Wippelhauser 2008). In effect, the advisory warns against eating these species while they are using the Augusta-Chops river segment, but consumption of what are possibly the same fish is addressed differently while they use portions of the river either upstream or downstream of that segment.

A lack of systematic posting of fish consumption advisories at fishing areas and launch sites may also add to confusion regarding what fish are safe to eat at a given location. MCDCP does not distribute signage or advisory materials at boat launches or other public access points; instead, they target the most sensitive populations (women of childbearing-age and young children) through other means (E. Frohberg, Maine CDCP, personal communication). As a stopgap, the Maine Toxics Action Coalition posts advisory materials, but coverage is incomplete. Additionally, some posted signage warns against consumption of freshwater fish in the Augusta-Chops river segment, which is less inclusive than the MCDCP advisory that, by implication, also addresses diadromous species.

Wild Foods and Ecosystem Services

Elevated contaminant levels in the Kennebec Estuary limit the amount of fish and shellfish that can be safely consumed by the public. This limitation undermines the ecosystem's ability to deliver provisioning services that were historically important to local people. In regions where the majority of nutritional sustenance originates from local sources, disruption of ecosystem provisioning services that support the availability of nutritious wild foods can impact aspects of public well-being ranging from health to security (Corvalan et al. 2005). In Maine, consumption restrictions for Kennebec Estuary-caught fish may not seem particularly alarming given that, even in the absence of fish consumption advisories, most species discussed in this report would probably not represent a substantial contribution to the average person's diet. Yet some segments of local commercial and recreational harvesters, as well as cultural and ethnic groups, can represent subpopulations that exploit fish from Maine estuaries in quantities that exceed the norm.

Although subsistence consumption of fish from the estuary is currently not likely, human resource-use patterns change over time and as a result, future dietary preferences, needs and sources are difficult to predict. Currently, at least half of the "healthy fish" choices in state promotional materials (MCDCP 2005) represent species that are locally overexploited, depleted, or too costly for many households to regularly consume, which may limit availability of these local food products. Elevated contaminant levels discussed in this chapter further erode options for sustainable, healthy dietary use of local fish.

Ecological Concerns

Knowledge of contaminant-induced impacts to fish and wildlife health requires an understanding of the toxic potency of a contaminant, its concentration in the organism's tissues (USACE 2008), and how that contaminant interacts with other contaminants. Few data are available to fully characterize how contaminant concentrations in the tissues of wild organisms influence the health of the individual. Predicting the population-level effects of contaminants is an even more complex problem (Bridges et al. 1996). In general, our lack of understanding is such that currently, only the most obvious toxicological effects (e.g., reduced productivity or survival) provide a strong inferential link between the individual organism and cumulative effects at the population or ecosystem level (USACE 2008). Even for species studied in the Kennebec Estuary, data to support characterizations of contaminant-related health effects based on tissue concentrations are lacking. In the following sections we occasionally present findings of research targeting organisms that can be considered more or less as surrogates, although interpretations based on these comparisons are made with caution because sensitivity to the same contaminant can differ among species. For instance, research indicates that wild aquatic birds such as the common tern are highly exposed to dioxins in the environment, but are up to 250 times less sensitive to these compounds than the typical avian research surrogate, the domestic chicken (*Gallus gallus*) (Karchner et al. 2006). Recent studies providing local data and information for this report section were available primarily through the efforts of MDEP, Gulfwatch, and the BioDiversity Research Institute (BRI).

Contaminants of Ongoing Ecological Research Interest

Mercury

Reported effects of methylmercury concentrations observed in the fish and shellfish species monitored by MDEP and the Gulfwatch program and presented in this report are not available in the scientific literature. For the sake of comparison, several surrogate organisms are provided. For a blue mussel surrogate we used soft-shell clams, which are arguably subject to different levels of contact with contaminated media than *Mytilus*. Soft-shell clam methylmercury concentrations as low as 25 ppm were reported to cause changes in immune system characteristics (Gorbi et al. 2001), but these concentrations were far higher than those measured in mussel tissues collected from the mouth of the Kennebec. Noel-Lambot and Bouquegneau (1977) found no increase in mortality of European eels with methylmercury tissue concentrations of 0.2 ppm, but this level was considerably exceeded by eels from the Kennebec Estuary, leaving the question of eel health responses to elevated tissue concentrations of mercury largely unanswered. Largemouth bass were used as a surrogate for mid-level piscivores such as smallmouth bass, striped bass, and bluefish. A marked shift in endocrine function was observed in largemouth bass with methylmercury concentrations of 1.23 ppm, which is considerably higher than any concentration found in Kennebec Estuary fish. Although mercury concentrations observed in most Kennebec fish and shellfish did not meet or exceed mercury benchmarks for effects that are reported in the scientific literature, it should be noted that the dearth of research in this area leaves much to be learned and consequently, profound uncertainty.

Estuarine and marine bird eggs recently analyzed for contaminants in Maine had the lowest mercury concentrations of those analyzed in all habitat types (Goodale 2008). Evers et al. (2005) attributed the higher concentration of mercury in inland waters versus marine environments to sulfur-reducing bacteria in fresh waters that convert mercury from inorganic to organic forms that enter the food web more readily. Lower mercury concentrations in estuarine and nearshore marine waters may also result from the fact that these waters are connected to more open systems which rapidly refresh (and therefore dilute) contaminated waters (Evers et al. 2005). The mouth of the Kennebec River is not considered a mercury hot spot relative to other sites in Maine, but researchers found that concentrations of the contaminant in eggs of piping plover, herring gull, and common eider were nevertheless higher at the mouth of the Kennebec than the regional mean. Despite the inclusion of samples representing two high-trophic level predators in the Kennebec Estuary—the double-crested cormorant (*Phalacrocorax auritus*) and osprey (*Pandion haliaetus*)—no eggs exceeded the lowest observed adverse effect concentration of 1.3 ppm for mercury in common loons (*Gavia immer*), a commonly used surrogate for other piscivorous bird species (Goodale 2008). Barr (1996) documented lowered productivity of adult loons eating whole fish with 0.3 ppm mercury concentrations and no productivity when methylmercury concentrations in prey fish exceeded 0.4 ppm. As noted above, observed incidences of methylmercury concentrations higher than 0.3 ppm were uncommon in fish of the Kennebec Estuary, being found in only two species, American eel and bluefish, although concentrations in striped bass and smallmouth bass were close (0.25 ppm).

PCBs

Concentrations of PCBs in fish tissue that have been reported to trigger adverse health effects range widely, sometimes spanning several orders of magnitude. The spectrum of PCB concentration thresholds associated with induced effects probably reflects differences in toxic potencies of different PCB combinations and inter-species differences in sensitivity to PCBs. If predictions based on tissue residues do not focus on distinct species or congener mixtures, assessments of toxicity risk to fish will include a high degree of uncertainty. For this reason, we refrain from using the results of tissue-residue effects studies because these studies either did not precisely target species of interest in the Kennebec Estuary or focused on congener combinations other than those observed in the estuary.

Total PCB concentrations in eider, osprey, herring gull, cormorant, and plover eggs from the mouth of the Kennebec were higher than the regional mean, but no samples exceeded the 8,000–20,000 ppb observed effects level for terns and other wild bird species (Goodale 2008). The only sample in recent research efforts that exceeded these thresholds was one bald eagle from Boothbay (Goodale 2008). Previous eagle contaminant observations in Maine also demonstrated considerably higher PCB levels in eagles nesting at coastal sites than those at inland sites (Welch 1994). In general, recent assessments suggest a decrease statewide in PCB levels in wildlife since 1977 (Goodale 2008). However, no eagle eggs from the Kennebec Estuary have recently been sampled so current PCB body burdens in local eagles are unknown.

Dioxins

Vertebrates are considered to exhibit greater sensitivity to dioxins and dioxin-like compounds such as TCDD than invertebrates, which lack the aryl hydrocarbon receptor that induces toxic effects (Larchner et al. 2006). MDEP reported concentrations of dioxins in Kennebec Estuary finfish of less than 1.5 pptr (MDEP 2007). Lake trout (*Salvelinus namaycush*) with dioxin muscle tissue concentrations exceeding 500 pptr did not show reduced survival after 77 days of monitoring (Walker et al. 1994). Given the much lower concentrations observed in Kennebec fish at similar trophic levels (smallmouth bass = 0.14 pptr), even the most contaminated species in the estuary is unlikely succumb to acute, dioxin-induced mortality.

Elsewhere, researchers found that the survival of adult lake trout eggs was not influenced by muscle tissue dioxin concentrations of 310 pptr (Walker et al. 1994). Again, these concentrations far exceed those recorded in Kennebec Estuary fish. Though they are limited in number, toxicological experiments involving extremely high dioxin concentrations suggest that Kennebec Estuary finfish are probably not subject to a heightened incidence of acute responses ending in mortality or to certain types of reproductive dysfunction due to dioxin exposure. Unfortunately, little more can be said because characterizations of more subtle responses are lacking and no recent data from the Kennebec Estuary were available to assess concentrations of dioxins and dioxin-like compounds in non-fish vertebrates such as birds. Likewise, despite dramatic reductions in dioxin discharges in Maine and trends that indicate lower concentrations in some taxa, the lack of recent dioxin sampling for the range of at-risk species—especially apex predators—impedes confident characterizations of local ecological risk.

Other Contaminants of Concern

In 1968, eggshells of eagles from Swan Island (Perkins Township) in Merrymeeting Bay and other locations along the Kennebec River had the highest concentrations of DDE and DDD residues (byproducts of DDT breakdown) recorded from sites in Maine, Wisconsin, and Florida (Krantz et al. 1970). After the 1972 ban on DDT use in the United States, recovery of the Maine eagle population was highly protracted—in 1975 only a single pair of nesting eagles was observed in the state and fledgling survival was negligible for over a decade afterward (Lichter et al. 2006). By 1990 a sustained recovery had begun and more recently organochlorine residues from eggs of all bird species recently sampled by BRI were significantly below published effects thresholds (Goodale 2008).

Avian ingestion of spent lead shot associated with waterfowl hunting was an issue investigated for decades before hunters were legally required to replace lead shot with non-toxic steel varieties. Between 1976 and 1980, Longcore et al. (1982) sampled Merrymeeting Bay sediments and waterfowl gizzards to quantify the incidence of lead shot. The authors estimated a mean of $99,932 \pm 40,391$ pellets/ha ($246,832 \pm 99,766$ pellets/acre) and found that 7% of American black ducks (*Anas rubripes*) sampled had ingested lead shot. Among ducks that ingested lead shot, 73% had only one well-eroded pellet in their gizzards. Under experimental conditions, 20% (16 of 80) of black ducks that ingested one lead pellet experienced lead-induced mortality (Longcore et al. 1974). However, it was thought that the consumption by wild black ducks of mollusk shells, which are rich in calcium carbonate, might act to ameliorate lead toxicity (Longcore et al. 1974). Although lead shot toxicity in Merrymeeting waterfowl was not documented, the

risk to longer-lived, higher trophic level predators feeding on waterbirds was nevertheless a concern (J. Longcore, personal communication). Since the enactment of lead shot prohibitions, there is probably little if any new lead entering the upper estuary. However, a lack of follow-up studies leaves open the question of whether previously spent lead ammunition remains accessible to bottom-feeding ducks in quantities sufficient to warrant concern.

Brominated flame retardants are a class of compounds generating more recent concern. PBDE concentrations in bird eggs tended to be higher in coastal Maine between Mount Desert Island and the Isles of Shoals (Goodale 2008). However, concentrations of PBDEs in bird eggs from the lower Kennebec Estuary were less than the regional mean. Assessments of ecological risk based on Kennebec Estuary PBDE concentrations in bird eggs are hindered by a lack of wildlife health benchmarks for this contaminant and tissue residues linked to observed effects.

Emerging awareness of perfluorochemicals has also prompted limited research in the Kennebec. Recent research by BRI found that PFOS concentrations tend to be higher in the southern coastal region than elsewhere along Maine's coast (Goodale 2008). At the mouth of the Kennebec River, PFOS was observed at greater than mean concentrations in eggs of herring gull, cormorant, eider, osprey, and piping plover (Goodale 2008). Among these species, the eggs of all but herring gulls had PFOS concentrations that were above the lowest observed adverse effects level of 100 ng/g wet weight for leghorn chickens (Goodale 2008), although use of that species as a benchmark warrants prudence due to the potential for differential sensitivity to PFOS among distinct wildlife species.

Conclusions

Despite considerable gains made since implementation of the Clean Water Act, fish and wildlife of the Kennebec Estuary are nevertheless subject to the uptake of toxic contaminants from both past and contemporary sources. The fate and effects of these contaminants as they move through the ecosystem are largely unknown; however, for some chemical groups compelling evidence warrants concern for public health, biodiversity, and ecological functioning and services. Efforts to reduce or prohibit intentional releases of specific contaminants have met with some notable success. But despite reduction efforts, atmospheric deposition and substrate/soil releases associated with historical pollution are ongoing sources of contaminants to the Kennebec Estuary.

Characterizing the scope of the problem remains a daunting challenge. Tissue contaminant sampling is infrequent or lacking for several key species of Kennebec fish and shellfish that traditionally supported economic and food-provisioning ecosystem services. The lack of current information for some of these species hinders efforts to confidently characterize human health risk and the ecological fate of individual contaminants in the estuary. The lack of research describing the health effects of differing contaminant concentrations on distinct species remains a source of uncertainty. Nationwide, emerging awareness of the risks associated with some groups of household chemicals (e.g., pharmaceuticals and personal care products) is a positive development that warrants local research, monitoring, and abatement efforts.

Toxic contamination represents challenges to ecosystem recovery and restoration that are rarely acknowledged by those outside the field of toxicological assessment and research. As a result, toxic chemicals are rarely featured in the ongoing dialogue of restoration professionals. Unseen and the subject of little public discussion, the contamination of our fish and wildlife and the implications of that contamination to food provisioning, public health, and local ecology are issues that have lacked resonance with the general public. Toxic contaminants present a particularly difficult restoration problem. However, there are

many ways to make progress, from implementation of increased monitoring that would better characterize risk to abatement of ongoing, intentional discharges. Above all else, raising awareness among restoration professionals and the general public is both the greatest challenge and the greatest opportunity for progress.