Chemical Stressor Indicators

CHAPTER 9:
LEGACY CONTAMINANTS
BACKGROUND
- Exposure to metals, polychlorinated biphenyls (PCBs), and pesticides causes a variety of human health issues (especially through consumption of contaminated fish and shellfish) and reduces environmental quality particularly in benthic habitats. Because of their long-term effect, metals, PCBs, and pesticides are referred to as legacy contaminants and are considered chemical stressors. Industrial manufacturing processes are major sources of these contaminants, with transport to the ecosystem by atmospheric deposition, river runoff, and wastewater discharges. While the Estuary Program recognizes the impact of metals and PCBs on freshwater ecosystems, this chapter focuses on the estuarine portions of the Narragansett Bay Watershed.

KEY FINDINGS
- **Status:** Although significant reductions have been made, the Seekonk River, Providence River and Taunton River sediments and some Upper Bay sediments have high concentrations of many contaminants (particularly mercury) and may still pose a human health risk through the bioaccumulation of these contaminants in locally harvested seafood.

- **Trends:** Generally, legacy contaminant concentrations have decreased dramatically in the last 40 to 50 years due to intense regulation and removal programs (pre-treatment and upgrades at wastewater treatment facilities) instituted in the 1970s. Sediment cores show similar patterns—rapid increases in deposition in the late 1800s and early 1900s, followed by rapid declines after the 1950s.
Introduction

Metals, polychlorinated biphenyls (PCBs), and pesticides are released into the environment through numerous sources including industrial practices, incomplete combustion, and insect control practices. Atmospheric deposition of these contaminants involves the release of small particles into the atmosphere that fall onto nearby surfaces or are captured in precipitation (rain, snow) and delivered to the ground. Many of these contaminants do not dissolve in water and instead collect in sediments, and they can be resuspended when those sediments are disturbed. When organisms come into contact with these contaminants, through ingestion or resuspension, the pollutants can bind to their fatty tissues. These contaminants then bioaccumulate—meaning that they are retained in the tissue of the organisms and passed up the food web to the apex predators, a process known as biomagnification. Additionally, the half-life of many of these compounds in the environment is quite long, making them persist for decades beyond when they were initially released (Nixon 1995). Because of their long-term effect on human and environmental quality, metals, PCBs, and pesticides are referred to as legacy contaminants and are considered chemical stressors.

Legacy contaminants are strongly associated with manufacturing practices globally and are concentrated near urban centers. They decrease with distance from the discharge points, although pesticides tend to be more ubiquitously distributed (Valiela 2006). The major sources of these contaminants to estuaries are atmospheric deposition, river runoff, and wastewater discharges (Nixon 1995, Valiela 2006). Prior to upgrades to wastewater treatment facilities (which reduced the amount of metals and contaminants in wastewater) and the creation of pre-treatment programs (which prevent the contaminants from reaching the facilities), industrial wastewater was a significant source of contaminants (Nixon 1995). Even though contamination tends to be concentrated near urban centers, evidence of metals, PCBs, and pesticides have been found in remote areas, such as the waters off Antarctica (Valiela 2006).

Legacy contaminants behave differently in water, sediments, or animal tissue, and how these contaminants react to salinity or temperature or affinity to particles affects how they impact the food web. Metals such as mercury that undergo methylation (the process by which a methyl group binds to a metal) tend to be lipophilic (combining with or dissolving in lipids and fats) and are more likely to enter animal cells and be stored in fatty tissue. On the other hand, lead sulfides and phosphates are permanent sinks of lead in soils and sediments and are not readily brought into the food chain (Valiela 2006). PCBs tend to resist degradation, attach to particles (including sediments), and are lipophilic, giving them a long-term persistence in the environment and food web. Similar behavior is observed for many pesticides. All metals, PCBs, and pesticides that are lipophilic tend to biomagnify and increase tissue concentration in organisms further up the food chain.

A variety of human health issues can result from legacy contaminants depending on level of exposure and how the chemical contacts the body (inhalation, ingestion, manual handling). Acute exposure tends to have short-term effects, mostly gastrointestinal and irritations (lung, skin). However, chronic exposure has caused genetic mutations, cancer, neurotoxicity, and endocrine disruption (Tchounwou et al. 2012, OSHA 2016). Mercury and lead, in particular, are known to cause developmental issues in unborn babies and small children. Many organizations and state health agencies have worked to educate the public on how they can reduce their exposure to heavy metals and PCBs in their diets and environment (e.g., Rhode Island Department of Health and Massachusetts Department of Public Health).

Narragansett Bay was one of the first estuaries in the United States to be intensively polluted by metals discharged to waters and released to the atmosphere from fuel combustion and industrial practices (Nixon 1995, Nixon and Fulweiler 2012). Starting in the late 1700s, Fall River and greater Providence were home to large industries of cotton textile and woolen production, machinery production, jewelry makers, and metals finisher manufacturers (Rhode Island Historical Preservation Commission 1981). The majority of these industries were originally located within the Narragansett Bay Watershed near major streams and rivers that provided both power and convenient discharge. As environmental regulations were not in place or were limited at this time, millions of pounds of various legacy pollutants made their way into Narragansett Bay. The metals-based manufacturing and subsequent pollution in Narragansett Bay became most prevalent in the mid-1800s (Corbin 1989, Nixon 1995).

Metals and PCBs have historically been used extensively in industrial and mill operations, particularly during the 1800s and early 1900s. Cadmium, copper, and lead were all emitted during the smelting process, removing the metals from ore (Nixon 1995). These emissions were usually atmospheric, but some may have been discharged directly as waste into receiving waters. Mercury was used in the
textile industry, as were chromate (chromium) and other chemicals used to produce dye. Lead was also emitted heavily by coal combustion and automobile emissions, prior to the reduction of lead in gasoline in the 1970s. PCBs had many industrial uses, including in insulating material, fire-resistant material, and coolant fluid (Valiela 2006). Production of PCBs was banned in the 1970s.

Manufacturing changed with time, from machine shops to textile processors and jewelry makers. Each shift in industry or produced goods changed the legacy contaminants that were emitted. Concurrently, the amount of impervious surface increased, particularly after the automobile was introduced in the early 1900s (Nixon 1995). The increased amount of impervious surface created easy avenues for metals to discharge directly into the Bay (see “Impervious Cover” chapter). Around this same time, wastewater treatment included chemical precipitation, creating contaminant-laden sludge. This sludge was initially dumped into receiving waters or incinerated (Nixon 1995). These processes released the legacy contaminants into the Bay’s ecosystem. It was not until later in the twentieth century that pre-treatment programs were enacted to reduce the amount of legacy contaminants coming into the treatment facilities (see below). Records of atmospheric emissions and sediment cores show similar patterns—rapid increases in deposition in the late 1800s and early 1900s, followed by rapid declines after the 1950s (Bricker 1993, Nixon 1995).

Most of the practices that delivered heavy metals, PCBs, and pesticides to the Narragansett Bay Watershed are waning. Industrial and mill operations are no longer the main economic resource in Rhode Island or Massachusetts. Some of the chemicals have been banned, and alternatives used, or the use of those chemicals has fallen out of favor (such as chromate dyes). While new emissions and discharges are lower than they were in the past, these legacy contaminants still affect the environment.

Researchers at the University of Rhode Island during the 1980s and 1990s analyzed the depositional history of metals and PCBs in sediment (Corbin 1989, Nixon 1991a, Latimer and Quinn 1996, Hartmann et al. 2004a and 2004b). These assessments found a north-south gradient in Narragansett Bay. Concentrations in surface sediments were greatest in the north near Providence and decreased southward in the Bay. Nixon (1991b) found that metal inputs to Narragansett Bay were greatest during wet years, owing to atmospheric deposition and overland flow delivering metals to the Bay. Latimer and Quinn (1996) found that the sediments in the Providence River Estuary accumulated PCBs at a greater rate than those in the rest of the Bay, a pattern later confirmed by Hartmann and colleagues (2004a and 2004b). They established that the major contaminant source to Narragansett Bay was the Providence River Estuary, and that the Taunton River appeared to have little impact on the western sections of the Bay (Hartmann et al. 2004a, 2004b).

In the early 1990s, Jeon and Oviatt (1991) reviewed the biological effects of pollutants on organisms within Narragansett Bay and compared them to field and laboratory experiments. This review was part of the same body of work described above. They, too, found that benthic diversity had a north-south gradient (see “Benthic Habitat” chapter) and that pollution was a driving factor in the gradient. Their review showed that toxic pollutants have declined in Narragansett Bay, particularly in the 1980s as noted by Nixon (1991b), and levels of contaminants in the Providence River Estuary were below proposed FDA alert levels (which were never formally adopted; Bender et al. 1989).

More recently, research has focused on surficial sediment. Murray and colleagues (2007) conducted an assessment of metals concentrations in surface sediments, finding that a strong spatial gradient still existed. The legacy contaminants measured included chromium, copper, mercury, nickel, lead, and zinc. Using copper as an example, the surface sediment pattern essentially showed an exponential decrease in sediment contamination from north to south in the Bay and that, in the upper reaches of the Bay, contaminant levels often exceeded sediment quality guidelines (SQG) otherwise known as Effects Range Median (ERM) and Effects Range Low (ERL) (Long et al. 1995) (Figure 1). This detailed mapping effort also pinpointed localized contaminant hotspots. The sediments in the Seekonk River and Providence River were highly contaminated with copper, those in the Upper Bay and Greenwich Bay were moderately contaminated, and those in the lower portions of the Bay were relatively clean (Figure 2).

Similarly, Murray and colleagues (2007) examined mercury concentrations in surface sediments, also finding a strong spatial gradient (Figure 3). The surface sediment pattern for mercury is similar to copper with an exponential decrease in sediment contamination from north to south in the Bay with additional localized highly contaminated hotspots in Greenwich Bay and Bristol Harbor (Figure 4). Nixon and Fulweiler (2012) reported on the findings by Murray and colleagues noting the strong spatial gradients in the upper portions of the Bay. They also noted that while the north-south gradient is clear,
there is a need to further examine the distribution of legacy contaminants from the Taunton River and Mount Hope Bay.

Murray and colleagues then worked with researchers from Roger Williams University and conducted additional sediment grab samples for mercury throughout Narragansett Bay (Taylor et al. 2012). The potential areas of high mercury exposure included the upper reaches of the Bay, including the Taunton River, Seekonk River, and Providence River (Figure 5). The results of the spatial analysis of land use and mercury levels demonstrated that the percent of total mercury that was methylmercury was significantly related to population density. This organic form of mercury is much more bioavailable and toxic than inorganic mercury. In addition, this study showed an association between higher levels of mercury in surface sediments and higher levels of mercury in the tissues of finfish harvested in the same areas (Taylor et al. 2012).

The bioavailability of other legacy contaminants (e.g., copper, lead, and cadmium) warrants further investigation. Studies of divalent trace metals that can produce toxicity in the marine environment indicate that they are often bound up as insoluble sulfides in anoxic sediments (Di Toro et al. 1990, 1991) and are only bioavailable when the total molar concentration of these metals exceeds that of the sulfide in the sediment. As an indicator of bioavailability, the Estuary Program’s partners used the molar ratio of simultaneously extracted metals (SEM) to acid volatile sulfide (AVS) concentrations found in sediments by dissolving surface sediment samples in strong acid for the divalent metals cadmium, copper, nickel, zinc, and lead (Di Toro et al. 1990, 1991). Ratios higher than 1.0 indicate potential bioavailability because the concentration of metals exceeds the concentration of sulfides. The excess metals are not bound as insoluble metal sulfide compounds, are likely to be found in solution, and are therefore potentially bioavailable and toxic. Using this method.
Figure 2. A map of surface sediment copper concentrations in parts per million (ppm) dry sediment. Symbols indicate the location of the sample: squares were from the Seekonk River, circles from the Providence River, triangles from the Upper Bay, diamonds from the West Passage, x’s from the East Passage, and plus signs from Greenwich Bay. The map reveals regions of particular concern with red color for values above the ERM level (270 ppm), orange and yellow colors for values between the ERM and ERL level, and green color for values below the ERL (34 ppm). Source: Murray et al. 2007
developed by Di Toro and colleagues, studies done in Narragansett Bay of the ratio of these trace metals to sulfide indicated that few sites were likely to have excess metals that would be bioavailable and could impact the biota, particularly near contaminant sources (Figure 6; Laliberti and King unpublished data). However, if contaminated and sulfide-rich sediments are disturbed during major storms, or by dredging activities, then the sulfides can oxidize and release toxic metals to the water column producing biological impacts. The potential biological impacts of such disturbance events would need to be monitored.

A critically important study was completed recently by Taylor and Williamson (2017) on mercury contamination and its transfer up the food web to targeted fish species that could be consumed by humans (black sea bass, bluefish, scup, striped bass, summer flounder, tautog, and winter flounder). This study provided strong evidence that about half of the anglers and their families harvesting coastal fish from the Narragansett Bay system and adjacent waters experienced mercury exposures that exceeded the USEPA reference dose and were at risk of impacts from mercury neurotoxicity. Taylor and colleagues (2012) and Taylor and Williamson (2017) have clearly shown that mercury found in contaminated sediment is bioavailable to marine organisms, and it bioaccumulates and biomagnifies sufficiently to pose a human health risk.

Pesticides have been used extensively since the mid-1900s for control of disease-carrying insects and for insect pest control in both agricultural and developed areas. In Narragansett Bay, using a core from Apponaug Cove in Greenwich Bay, Hartmann and colleagues (2005) determined that DDT and chlordane were present in sediments starting in the late 1940s to early 1950s. DDT concentrations in sediment cores peaked in the early 1970s, around the time of their ban in 1972, and chlordane appeared to be declining (Hartmann et al. 2005). Chlordane was banned in 1988, twelve years before the sediment cores were collected. Sedimentation rates and bioturbation made it difficult for Hartmann

![Mercury vs. Latitude - Surface Sediment Grab Samples](image.png)

**Figure 3.** The gradient of surface sediment mercury concentrations in parts per million (ppm) dry sediment. Symbols indicate the location of the sample: squares were from the Seekonk River, circles from the Providence River, triangles from the Upper Bay, diamonds from the West Passage, x’s from the East Passage, and plus signs from Greenwich Bay. The gradient shows an exponential north-to-south decrease in contamination using the sediment quality guidelines (SQGs) ERM and ERL (Long et al. 1995). Source: Murray et al. 2007
Figure 4. A map of surface sediment mercury concentrations in parts per million (ppm) dry sediment. Symbols indicate the location of the sample: squares were from the Seekonk River, circles from the Providence River, triangles from the Upper Bay, diamonds from the West Passage, x’s from the East Passage, and plus signs from Greenwich Bay. The map reveals regions of particular concern with red color for values above the ERM level (0.71 ppm), orange and yellow colors for values between the ERM and ERL level, and green color for values below the ERL (0.15 ppm). Source: Murray et al. 2007
Figure 5. (A) Total surface sediment mercury (Hg) concentrations in parts per million (ppm) dry weight. Note that the ERM = 0.71 ppm and the ERL= 0.15 ppm, so most of the Bay was highly to moderately contaminated with mercury. (B) The surface sediment concentration of methylmercury in ppm dry weight. (C) The percent of total mercury that was methylmercury. Areas with the dark shades (high percent) were areas of particular concern. Source: Taylor et al. 2012
and colleagues (2005) to determine if chlordane peaked near its ban date, much like DDT.

It should also be noted that the Estuary Program decided not to utilize data and studies of metals from water column samples. In the 1980s, researchers at the University of Rhode Island compiled (Kester et al. 1987) and analyzed (Bender et al. 1989) trace metal concentration data from Narragansett Bay waters and determined that concentrations were higher in the upper Bay and decreased down bay. In general, even the highest concentrations did not exceed water quality criteria, and the waters of the Bay were not likely to produce biological impacts. More recent research (Kozelka and Bruland 1998) confirmed the north-south gradient for copper, zinc, cadmium, and lead—metals that can be toxic when present in high concentrations in the environment. This work also showed that these metals in the “dissolved” form were generally bound to other materials (e.g., organic matter) and were not particularly bioavailable and unlikely to cause toxicity. The limited studies of metals in the water column suggest that the waters of Narragansett Bay in recent decades are relatively clean and unlikely to cause adverse effects. However, studies of legacy contaminants from water samples are expensive and produce data that is noisy and difficult to interpret in terms of status and trends. On the other hand, studies of legacy contaminants in sediments (surface samples and dated sediment cores) and organisms (e.g., mussel tissue) are integrated over longer time frames, and are therefore less variable and better suited for determining environmental status and trends.

In this chapter, the Narragansett Bay Estuary Program reports on the available data concerning legacy contaminants in Narragansett Bay. These data include results of sediment core and mussel tissue analysis for key metals (cadmium, chromium, copper, lead, and mercury), PCBs, and pesticides. While the Estuary Program recognizes the impact of metals and PCBs on freshwater ecosystems, this chapter focuses on the estuarine portions of the Watershed. The results presented in the Status and Trends section of this chapter will be discussed in the context of previous work (above) and how legacy contaminant bioavailability may be altered by climate change.

**Methods**

To examine legacy contaminants, the Narragansett Bay Estuary Program worked with its partners to analyze recent data in Narragansett Bay involving dated sediment cores and tissue from blue mussels (Mytilus edulis).

**Figure 6.** The molar ratio of simultaneously extracted metals (SEM) to acid volatile sulfide (AVS) for cadmium, copper, lead, nickel, and zinc (following Di Toro et al. 1990, 1991). Values lower than 1.0 indicate that metals are bound up as insoluble metal sulfide compounds and are not likely to be bioavailable and toxic. Conversely, values higher than 1.0 indicate the potential for bioavailability and toxicity. Source: Laliberte and King unpublished data

**DATED SEDIMENT CORES**

Several dated sediment cores in various sections of Narragansett Bay were analyzed to provide an historical analysis of legacy contaminant concentrations. A dated sediment core from the Seekonk River (Corbin 1989, King unpublished data) was used to measure metal concentrations for specific legacy contaminants (copper, lead, cadmium, and chromium) and the age model used is based on dating done by
Cs137, Pb210, pollen stratigraphy, and radiocarbon dating. ERM values for these metals were compared to metal concentration results from the 1770s to modern-day results. A high-resolution record from a dated sediment core near the Field’s Point sewage treatment plant in the Providence River was examined for trends in trace metals (copper and lead) and PCBs (Cantwell and King unpublished data). The age model was based on Cs137 and Pb210 from the 1940s to 2015, and a change in sediment type near the base that indicated dredging in 1941. Lastly, a dated sediment core at Field’s Point in the Providence River was examined for chlorinated pesticides and results, from every three to four years from 1941 to 2015, were summed for trans-chlordane, cis-chlordane, and 4,4’-DDE (Cantwell and King unpublished data).

**MUSSEL DATA**

Two approaches were utilized to analyze metals and PCB concentrations in blue mussel tissue. First, using an earlier USEPA study by Phelps and Galloway (1979) as a model, the Narragansett Bay Commission (NBC) designed a study, unpublished, to measure legacy contaminants. Blue mussels were collected from Fort Getty, Jamestown (Lower West Passage) during the fall seasons of 2008, 2009, and 2012. As a control, a portion of each set of mussels collected were immediately put on ice and frozen and brought to the NBC laboratory for analysis. Remaining mussels were then deployed in cages at Conimicut Point (Upper Bay), in approximately the same location as used by Phelps and Galloway (1979), for a four-week time period. Mussels were then collected and analyzed for metals concentrations.

Second, a research project evaluated the use of indigenous mussel populations as sentinel organisms for indicating levels of pollutants in coastal marine waters (Goldberg et al. 1978). The National Oceanic and Atmospheric Administration (NOAA) adapted this research effort and created the Mussel Watch program. This program has sites throughout the United States and collects blue and ribbed mussels as well as surface sediment samples to analyze concentration levels of 25 different heavy metals and 98 types of PCBs. Mussel Watch data for concentration of metals and PCBs in Narragansett Bay were collected yearly from 1986 to 1999, and then every other year until 2011. The Estuary Program analyzed Mussel Watch data from three sites within Narragansett Bay—Dutch Island (Lower West Passage), Dyer Island (Middle East Passage), and Patience Island (Upper West Passage).

**Figure 7.** Metal concentrations (dry weight ppm) from a dated sediment core from the Seekonk River (Corbin 1989, King unpublished data). The age model is based on Cs137, Pb210, pollen stratigraphy and radiocarbon dating. The red lines show the ERM values for the respective metals (Long et al. 1995).
Both approaches analyzed the same metals: cadmium, chromium, copper, and lead. Total PCB concentration trends were also analyzed in the Mussel Watch program. All metals data from the Mussel Watch program were based on one sample per metal, so no standard deviations could be calculated. PCB data included the total concentrations of all congeners tested in the Mussel Watch program.

**Status and Trends**

**DATED SEDIMENT CORES**

A dated sediment core from the Seekonk River showed the 300-year history of trace metal inputs for copper, lead, cadmium, and chromium (Figure 7; Corbin 1989, King unpublished data). This record reflected the rise during the industrial age in the Providence area followed by the decline in response to strict environmental regulations (Clean Air and Clean Water Acts) in the 1970s. After 1990, contaminant levels decreased below Sediment Quality Guideline (SQG) levels that would likely be associated with biological impacts. These trends have been described by Corbin (1989) and Nixon (1995) as reflecting industrial usage and improvements in environmental regulation. These sediments were contaminated based on regulatory standards, but levels have fallen in recent decades to levels at or below SQG levels at which biological impacts are likely to be observed (ERM).

A very high-resolution record from a core near the Field’s Point sewage treatment plant showed trends in trace metals and PCBs that revealed improvements achieved by environmental regulation (Figure 8; Cantwell and King unpublished data). At this site, SQGs were achieved around 2000 for lead and only recently, in 2015, for copper and PCBs.

Several chlorinated pesticides measured in the Field’s Point core provide insights into their long-term use and persistence (Figure 9; Cantwell and King unpublished data). Here, summed concentrations of chlordane and ∑DDT rapidly increased from 1940 to maximum values in the mid-1950s, reflecting their high volume of usage. Declines in the late 1950s onward reflect limitations placed on the use of DDT as concerns over its usage increased, with an eventual ban in 1972. Chlordane usage continued until its ban, as seen by the decline in concentration in 1988. Measurable concentrations were present at the surface of the core, likely from land-based sources.

![Figure 8. Metals concentrations (ppm) and PCBs concentrations (ppb) from a dated sediment core from Field’s Point in the Providence River (Cantwell and King unpublished data). The age model is based on Cs137, Pb210, and a change in sediment type near the base that indicates dredging in 1941. The red lines indicate the ERM values for the legacy contaminants (Long et al. 1995).](image-url)
residuals of both pesticides continuing to enter the Bay, testifying to their persistence.

**MUSSEL TISSUE**

The following results were developed by the Narragansett Bay Commission (NBC) from an updated study by Phelps and Galloway (1979). Metals concentrations measured in mussels deployed at Conimicut Point (Upper Bay) in each of the three more recent NBC study years were compared to samples collected in 1976 (Phelps and Galloway 1979) and analyzed for statistical differences using ANOVAs (Figure 10). The 2008, 2009, and 2012 concentrations in mussel tissue for all metals analyzed in all three study years were significantly lower when compared to the 1976 samples, with the exception of chromium in 2009. These data suggest a trend of decreasing metals concentrations in the Upper Bay in response to water quality improvements over the 30-year time interval. Nickel showed the greatest percent reduction with an average decrease of 88 percent, while copper had the least decrease of fourteen percent. It is also interesting to note that the more recent concentrations of cadmium, nickel, and zinc in the study mussels from Conimicut Point were lower than those from Jamestown North in 1976, a site further down the Bay.

Current (2008, 2009, 2011, and 2012) concentrations of metals and total PCBs from NBC and the NOAA Mussel Watch data are included in Table 1. Cadmium, chromium, mercury, and lead were among the metals with the lowest concentrations in mussels and copper had the highest.

Of the five metals analyzed during the study period (1976 to 2012), cadmium and lead had a decreasing trend (Figure 11). Between 1976 and 1986, metal concentrations decreased dramatically except copper (Figure 11). Mercury concentrations appear to have decreased at most sites within the last decade (Figure 12). No sites appear to have consistently increasing concentrations in any of the metals.

Total PCB concentration in mussel tissue was highest at both Dutch and Dyer Islands in the late 1980s followed by a decline (Figure 13). Concentrations at Patience Island may reflect a decreasing trend since the mid- to late 1990s.

**Discussion**

Radiometrically dated sediment cores from areas of the Bay with high sedimentation rates and limited mixing by deep bioturbation can provide useful information on both the status and trends in environmental quality over the time frames of several decades to hundreds of years. Records from the upper reaches of the Bay and from coves can typically provide records of environmental quality with a resolution of a few years. This approach provides very useful information on the impacts of historical land use and industrial changes within the Bay system and of environmental regulatory actions and infrastructure upgrades.

Dated sediment cores showed a consistent pattern of legacy contamination—an increase in metals and PCB concentrations from the start of the Industrial Revolution through the early to mid-1900s, and then a decline as manufacturing practices shifted (Figures 7 and 8). Currently, metals and PCB concentrations appear to be at or near their respective ERMs (Figures 7 and 8), indicating that Bay sediments at and near the surface are getting cleaner. These findings are similar to previous findings (Nixon 1995, Hartmann et al. 2004 and 2005, Nixon and Fulweiler 2012).

In general, the mussel data from NBC’s study and NOAA’s Mussel Watch program indicate that metal and PCB concentrations were decreasing or remaining similar throughout the study period (1976 to 2012) (Figures 10 through 13). This trend was
Figure 10. Comparison of metals concentrations in Conimicut Point mussels from 2008, 2009, and 2012 to results from the mussels placed in the Conimicut Point in 1976 (Phelps and Galloway 1979). Error bars were not included on lead for Phelps and Galloway data since a range (10.8 to 15.5) was given in the paper and not a standard deviation. The letters indicate ANOVA tests of significance on the average metal concentrations: different letters indicate significance levels greater than 0.05; similar letters indicate no significant difference between time periods.

Table 1. Mussel tissue average concentration for metals and total PCBs for each sample site from 2008 to 2012. NBC data (Conimicut Point 2008, 2009, 2012) include standard deviations of mussel tissue samples. Patience, Dutch, and Dyer Islands sites from 2011 are from NOAA Mussel Watch.

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<td>Cadmium (Cd)</td>
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<td>Copper (Cu)</td>
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<tr>
<td>Lead (Pb)</td>
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<td>0.08</td>
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<td>Total PCBs (ppb)</td>
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Figure 11. Concentrations (ppm) of cadmium, chromium, copper, and lead in mussel tissue. Conimicut Point values are from NBC's study comparing Phelps and Galloway data from 1976 to data collected in 2008, 2009, and 2012 (see text). Patience, Dutch, and Dyer Islands sites are from NOAA Mussel Watch.
Figure 12. Mercury concentrations (ppm) in mussel tissue from NOAA Mussel Watch.

Figure 13. Total PCB concentrations (ppb) in mussel tissue from NOAA Mussel Watch.
expected since the loadings of these contaminants have been decreasing through the late twentieth and early twenty-first centuries (Nixon 1995, Nixon and Fulweiler 2012). These conclusions also compare well with a National Estuarine Research Reserve Mussel Watch trend analysis using mussel tissue collected from sites throughout the US (Lauenstein and Cantillo 2002, O’Connor and Lauenstein 2005).

Influent loadings of both copper and chromium to the Field’s Point wastewater treatment facility have decreased by more than 98 and 97 percent, respectively, since 1981 (NBC 2015), although the decreases observed in the mussel data were not as great. There are several reasons these differences may exist. While both copper and chromium were used extensively in industries prevalent in Rhode Island for decades, the decrease in these types of manufacturers has reduced copper and chromium loading into the Field’s Point plant greatly. Copper, however, still has a more prevalent presence within the waters of the Bay from sources such as copper piping used for water distribution and anti-fouling methods used in the marine industry.

An important point of distinction is the total mercury findings from the Mussel Watch program (Figure 12) compared to the total mercury findings from Murray and colleagues and Taylor and colleagues (Figures 3 through 5; Murray et al. 2007, Taylor et al. 2012). Mercury levels have generally declined since 1985 in the Mussel Watch data (Figure 12). However, in surficial sediment, mercury levels are elevated in most of the Bay with the mercury commonly in the bioavailable form of methylmercury (Taylor et al. 2012). Mercury values have decreased in sediments but still tend to considerably exceed ERM values in the upper reaches of the Bay (Cantwell et al. 2007). Therefore, mercury contamination poses an ongoing significant human health risk in the Narragansett Bay system (Taylor and Williamson 2017). Evaluations of other legacy contaminants that tend to be found in more available forms, taking a similar approach to that of Taylor and colleagues (2012) and Taylor and Williamson (2017), are warranted. ERM values can provide a useful indicator for focusing these studies.

Two aspects of climate change—temperature and precipitation—may have effects on legacy contaminants. Increased temperature may enhance volatility and partitioning of persistent organic pollutants (such as PCBs) from the water to the atmosphere, which could reduce exposure to aquatic biota (Noyes et al. 2009). In terms of the effect of temperature on metabolism, the literature presents several possibilities. Increases in temperature can increase metabolism, which raises the concentration of contaminants within an organism (Schiedek et al. 2007, Sokolova and Lannig 2008, Noyes et al. 2009). Temperature increases may also increase toxicity of certain contaminants, which may affect biomagnification (Noyes et al. 2009). Degradation of contaminants may increase with increasing temperature, which makes the contaminants unavailable to biota (Whitehead et al. 2009).

Precipitation is another aspect of climate change that may change the availability of metals and PCBs. Erosion and runoff may allow more bioavailable metals to reach the water column. Winter/spring precipitation is expected to increase in the Narragansett Bay Watershed by approximately 30 percent by the year 2100, and the frequency of extreme precipitation events is also expected to increase significantly (see “Precipitation” chapter). The combination of these factors is expected to increase the remobilization of contaminated sediments in the upper reaches of the Bay. Under changing hydrology conditions, increases in methylmercury production have been noted (Whitehead et al. 2009). Precipitation can alter salinity regimes in coastal waters, and then affect the chemical itself through oxidation state changes, or enhanced/reduced toxicity based on salinity levels (Noyes et al. 2009). Combined with temperature changes, the interactive effects of metals, PCBs, and pesticides are very complex and need to be understood for the specific location or organisms of interest.

In general, the water and sediment data from Narragansett Bay indicate that the water is relatively clean with respect to legacy contaminants throughout the Bay system (Bender et al. 1989), whereas contaminated sediments are found in surface sediments in the upper reaches of the Bay system (Corbin 1989, Murray et al. 2007, Cantwell et al. 2007, Taylor et al. 2012) and highly contaminated sediments are found at depth in sediment cores in the coves and upper reaches of the Bay system (Corbin 1989, Cantwell et al. 2007, King et al. 2008). The general spatial pattern of legacy contaminants can be described as clean water and contaminated sediments in the upper reaches of the Bay system, and clean water and relatively clean sediments in the mid to lower Bay. Temporal trends indicate that most legacy contaminants have decreased significantly from maximum levels in the 1950 to 1970 interval to values that are either at or below ERM values today.
Data Gaps and Research Needs

- The concentration of legacy contaminants, including mercury, in estuarine and freshwater fish and shellfish is a data gap. More studies using an approach similar to that used by Taylor et al. (2012) and Taylor and Williamson (2017) for mercury are needed to determine the human health risk posed by the uptake of legacy contaminants by fish and other human-consumed biota (e.g., shellfish). Future work would be to expand the state monitoring programs to include estuarine and near-shore fish (i.e., Taylor’s work) to create a holistic assessment of mercury in commercially and recreationally important species throughout the Bay. Other legacy contaminants that need to be assessed include, at a minimum, PCBs, pesticides, and cadmium.

- The concentration of legacy contaminants in river sediments within the Narragansett Bay Watershed is a data gap that can contribute to delays in pursing riverine restoration actions. Studies like Cantwell et al. (2014) need to be conducted to assess the amount of contaminants in the sediments and water column before and after dam removals.

- Brayton Power Plant maintained metals-monitoring data in quahogs (Mercenaria mercenaria) that could be incorporated into the status and trends analyses. Given Brayton Power Plant’s shut down, it is unlikely this monitoring program will continue. Adding a Mussel Watch monitoring station to Mount Hope Bay would be useful in tracking legacy contaminants in that region.

- These results are framed around a north-to-south gradient, with the study sites reflecting that preference. However, sediment contaminant maps have pinpointed localized hotspots throughout the Bay—such as near the East Greenwich Wastewater Treatment Facility in Greenwich Bay—that warrant further research (Figures 2 and 4).

- The climate change section of this chapter showed that there is little knowledge of how these legacy contaminants will behave under a changing climate. While release into the environment is decreasing, these contaminants may still pose health risks due to relic deposits in sediments. Understanding how climate change will affect mobility and toxicity of these contaminants both directly and indirectly is important to inform human and environmental risk assessments.

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