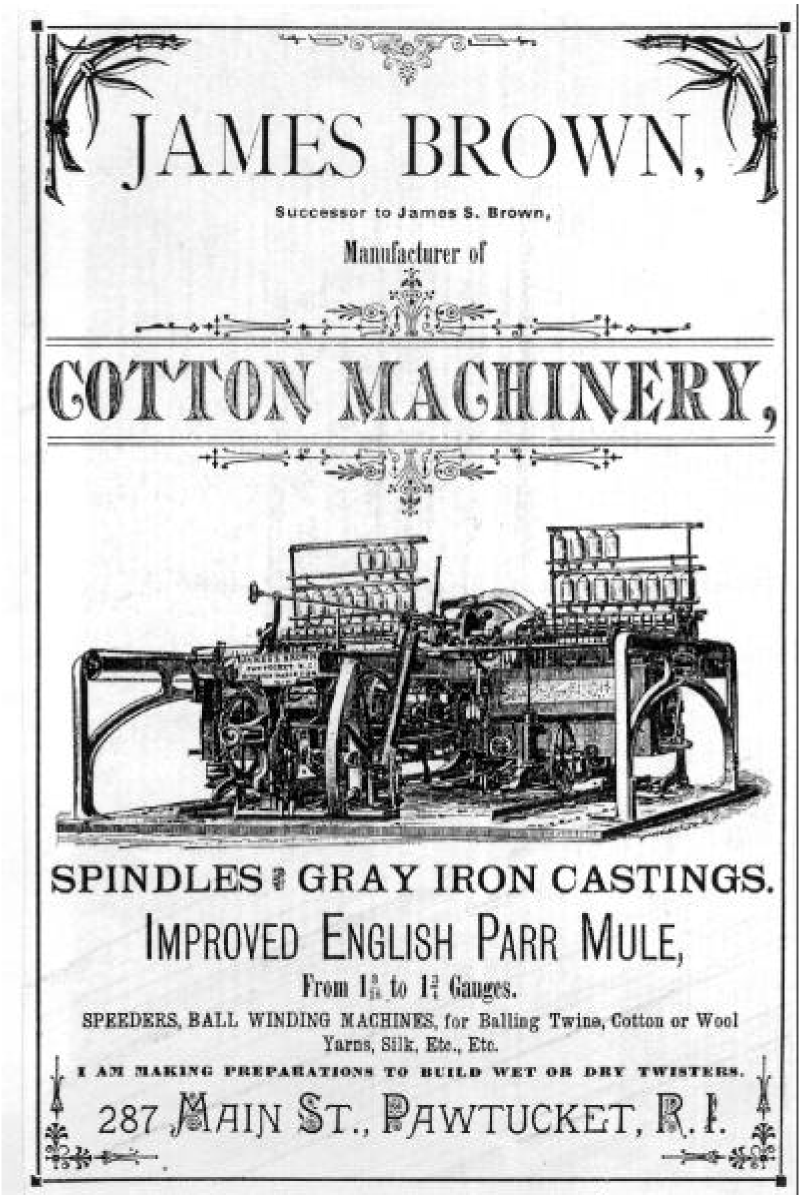
Metal Inputs
to Narraganset Bay



JAMES BROWN,

Successor to James S. Brown,

Manufacturer of

COTTON MACHINERY,

SPINDLES and GRAY IRON CASTINGS.

IMPROVED ENGLISH PARR MULE,

From $1\frac{3}{16}$ to $1\frac{3}{4}$ Gauges.

SPEEDERS, BALL WINDING MACHINES, for Balling Twine, Cotton or Wool Yarns, Silk, Etc., Etc.

I AM MAKING PREPARATIONS TO BUILD WET OR DRY TWISTERS.

287 MAIN ST., PAWTUCKET, R. I.

A History and Assessment
of Recent Conditions

Scott W. Nixon

Loan copies are available from the National Sea Grant Depository, Pell Library Building, University of Rhode Island Bay Campus, Narragansett, RI 02882-1197. Order RIU-T-95-001.

This publication is sponsored in part by Rhode Island Sea Grant, under NOAA Grant No. NA36RG0503. This publication is also sponsored by the R.I. Department of Environmental Management (DEM) Narragansett Bay Project under Grant No. CX812680 (refer to the Narragansett Bay Project report #NBP-91-66, *Recent Metal Inputs to Narragansett Bay*). The views expressed herein are those of the author and do not necessarily reflect the views of NOAA or any of its sub-agencies or of the R.I.DEM Narragansett Bay Project. The U.S. Government is authorized to produce and distribute reprints for governmental purposes notwithstanding any copyright notation that may appear hereon.

The proper citation for this report is:
Nixon, Scott W. 1995. *Metal Inputs to Narragansett Bay: A History and Assessment of Recent Conditions.* Rhode Island Sea Grant, Narragansett, R.I.

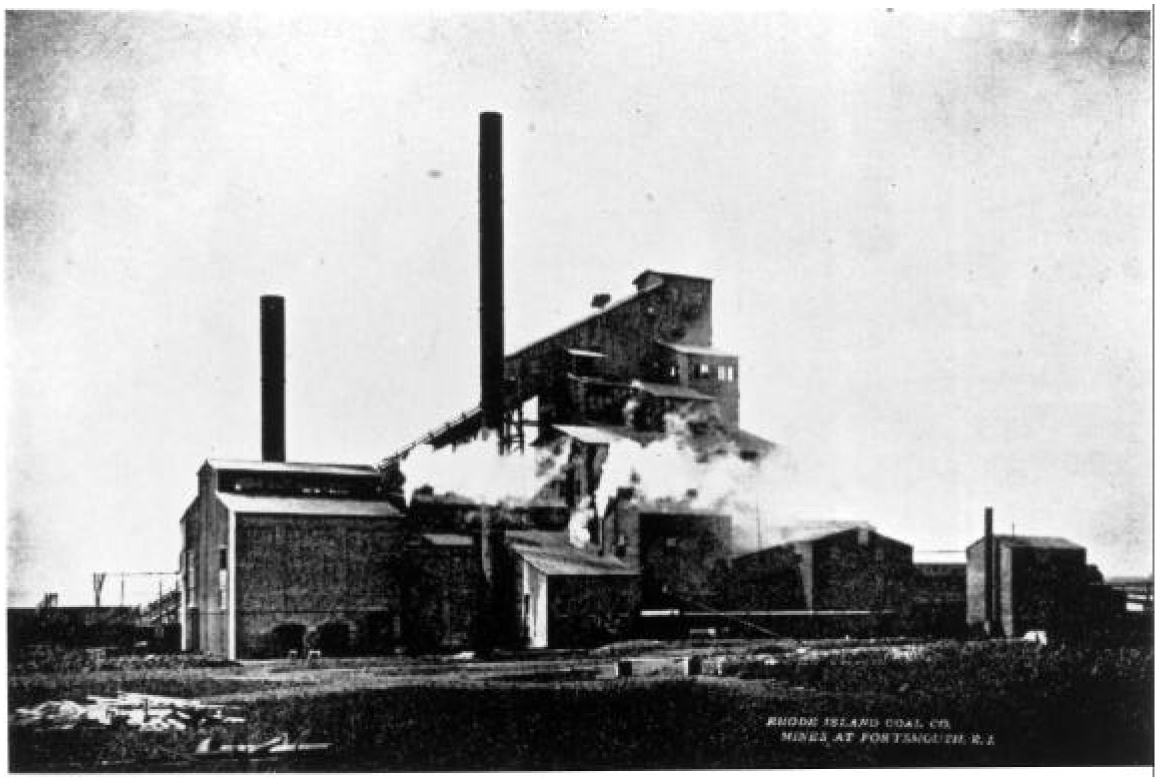
Metal Inputs to Narragansett Bay:
A History and Assessment of Recent Conditions

Scott W. Nixon
Graduate School of Oceanography
University of Rhode Island
Narragansett, RI 02882-1197

1995

SECTION I

A History of Metal Inputs to Narragansett Bay



Rhode Island Coal Company mine at Portsmouth, R.I.
Photo from the Wilfred Warren Collection, Jamestown, R.I.

A History of Metal Inputs to Narragansett Bay

Introduction

> In taking his first steps from the Stone Age into the Age of Metals, man crossed a great divide. But considering his long and skillful use of the other materials around him, that advance took place quite late in human history. In fact in some parts of the world it never took place at all.
>
> Robert Raymond (1984)
> *Out of the Fiery Furnace*

The oldest human remains yet found near Narragansett Bay are about 3300 years old. They were discovered on Conanicut Island, in shallow graves containing stone implements, including bowls and kettles made from steatite or soapstone. The early inhabitants buried carefully in those graves were archaic-period hunters, "who knew nothing of agriculture or pottery" (Simmons 1970). Without the ovens and bellows needed to achieve the high temperature (about 450°C) required to fire clay into pottery, it seems likely that there was little, if any, capability among these ancient people for smelting or working metals (Raymond 1984). While the water, sediments, and organisms of Narragansett Bay have always contained small amounts of metals that entered the bay with sea water and in river water that carried the weathering products of the drainage basin, the direct anthropogenic addition of metals to the bay came only after European discovery and settlement.

European Contact

There is some debate about the first European discovery of Narragansett Bay. It is possible that the Norse may once have had a presence and the Englishman John Cabot may have sailed close by in 1498. But there is no doubt that Giovanni da Verrazzano brought his ship *Dauphin* into what is now Newport Harbor on 21 April, 1524 (Wroth 1970). It may well be that with the dropping of the *Dauphine's* anchor and chain, the first anthropogenic metals entered the bay.

Verrazzano reported that the Narragansett natives already possessed "many sheets of worked copper which they prize more than gold" (Wroth 1970). The origin of this copper is unknown. It may have come from trade with tribes that had earlier contacts with Europeans or possibly from the Great Lakes region, where unique geological conditions had left large amounts of almost pure copper metal on the surface of the ground (Raymond 1984). In any case, the introduction of metals was rapid all along the Atlantic coast. Verrazzano gave the Narragansetts metal bells and trinkets , and by 1602 Bartholomew Gosnold "noted much copper in use by natives off the Massachusetts coast . . ." (Simmons 1970). A Narragansett Indian burial ground used around 1650 and lying adjacent to the prehistoric graves mentioned earlier was found to contain numerous and diverse metal objects (Table 1). All of these implements, however, were of European manufacture. In spite of the fact that the Narragansetts and other native tribes of the northeast had been pro-ducing a type of burnt clay pottery for perhaps a thousand years before Verrazzano arrived, metal smelting appears not to have been part of their culture (Fowler and Luther 1950). In his book describing *Indian New England Before the Mayflower*, Russell (1980) pointed out the irony that the great abundance of natural resources helpful for human use in this area included "none of the valuable metals on which early European explorers had so often set their hearts, except perhaps a very little copper, some of it brought by trade from the Great Lakes region."

While many of the Europeans were intent on gold and silver, the English in the New World had a genuine interest in iron, perhaps because a shortage of timber for the charcoal needed in smelting was already threatening the iron industry at home (Carroll 1973). Within twenty years of the founding of Boston in 1630, "the successful, sustained, and integrated production of cast and wrought iron was first achieved within the limits of the United States" at Hammersmith, Massachusetts, on the banks of the Saugus River (Hartley 1957). The iron making operation there failed by 1684, but the technology had already been transferred to Narragansett Bay, where Joseph Jenks, Jr. erected a forge at Pawtucket Falls about 1672 (Hartley 1957).

Various other iron making enterprises developed around Narragansett Bay through-out the eighteenth century, including works at Manville, in Cumberland, at Pawtucket Falls, in Warwick, and at Hope. In spite of these efforts, however, the low quality of the ore deposits in Cumberland and Cranston combined with a lack of skilled labor and compe-tition from iron makers in Pennsylvania and Europe to prevent the blast furnaces in Rhode Island from becoming nationally important (Kulik and Bonham 1978). Aside from some slag deposits on the sides of rivers and streams, these early ore extraction and smelting works probably added little to the natural input of metals to Narragansett Bay. It is possible that their greatest impact on metal pollution was from the burning of large amounts of wood. Accounts of ironworks in England in the late 1500s show that it took about 17 cords of wood to produce one ton of cast iron, and that a single establishment might use 22,000 cords of wood fuel each year (Carroll 1973).[1]

When combined with atmospheric emission factors for wood combustion (Nriagu and Pacyna 1988), these data suggest that

---

[1] Melosi (1985) reports that six cords of wood were required to produce one ton of iron in the early 1800s.

Table 1

Metal implements found in Narragansett Indian graves at a burial site on Conanicut Island. The site was used about 1650 (Simmons (1970).

Iron	Brass	Copper	Lead
lock	kettles	tube	musket balls
knives	ring		
musket	hawk bells		
buckle	spoons		
chisel	bell		
nails	belt hook		
hatchet	fish hook		
ax heads	arrowhead		
hooks	thimbles		
hoes			
bar			
box			
scissors			

the wood burning required for the production of one ton of iron may have released 4-13 g Cd, 45-90 g Cu, 45-135 g Ni, 90-225 g Pb, and 90-450 g Zn to the atmosphere. While some of the metals thus mobilized may have been deposited on the bay, it seems likely that most would have returned to forests and fields where they would have been retained in the vegetation and soil. The same reasoning suggests that the large campfires favored by the Narragansett Indians, as well as their practice of burning the forest undergrowth (Day 1953; Cronon 1983), probably had little impact on metal inputs to the bay.

Metal pollution in any significant extent would come as a by-product of the Indus-trial Revolution. And because that revolution first came to America along the Blackstone River, in the watershed of Narragansett Bay, it seems likely that this estuary has been subjected to metal pollution longer than any other on the western side of the Atlantic.

The Industrial Revolution

The beginning of the Industrial Revolution in America is commonly traced to Samuel Slater's introduction of the factory system in Moses Brown's textile mill at Pawtucket, RI. There, in December, 1790, the energy of the Blackstone River was used to spin cotton by water power for the first time in America (Coleman 1963). In spite of this early success, however, "the Rhode Island cotton industry developed so slowly that many years elapsed before a full-fledged factory system emerged" (Coleman 1963). It was not until the War of 1812 and the invention of the power loom and cotton cleaning machinery that the cotton industry began its remarkable expansion (Table 2).

The first fully developed factory system in America involving the use of water power and "the consolidation of all manufacturing processes from carding to finishing within a single mill" was actually achieved by Rowland Hazard in his woolen mill in South Kingstown, RI in 1813 or 1814 (Coleman 1963). Woolen manufacturing, however, would remain second in importance to the state's cotton industry, and it grew more slowly (Table 2). Woolen mills were largely concentrated in the southwest part of Rhode Island until about 1840, but by 1860, the woolen and cotton industries were both dominated by factories along the Blackstone River.

Growth of the Base Metals Industry

The development and dramatic growth of textile manufacturing in Rhode Island and throughout the Blackstone River valley is likely to have resulted in the first major increase in the addition of metals to Narragansett Bay. Some of the metals came from the mordants used in dyeing. Even before the first use of coal-tar dyestuffs in the late

1850s, vegetable dyes were used with iron, tin, copper, lead, aluminum, antimony, and especially

Table 2

Early growth of the Rhode Island textile industry (compiled from data in Coleman 1963).

	Cotton Industry			Woolen Industry	
	Capital[1]	Spindles	Workers	Capital[1]	Workers
1810		1,500			
1812		30,663			
1815		76,792			
1832	5,590	237,978	9,071		380
1840	7,324	518,217	12,086	685	961
1850	8,446		12,386	1,139	1,943
1860	12,032		15,739	3,000	4,200

[1] Thousands of dollars

chromium mordants (Whittaker 1926). But the link between textiles and metal pollution lay less in the manufacture of the yarn and cloth than in the manufacture of the new machinery needed to support the expansion of the textile industry.

While historians have paid much more attention to the role of Samuel Slater and his introduction of the Arkwright system, the successful mechanization of the textile industry also required an increasingly sophisticated base metals industry to design, cast, and fabricate the complex machines that were needed (Plate 1). In the earliest years, that capability was found in one remarkable family.

Machinery and Machine Tools

Shortly before Samuel Slater began to apply English carding and spinning technology in Brown's mill, the Wilkinson family also moved to Pawtucket. With this move,

> . . . Pawtucket Village became one of the most important industrial areas in the United States. The Wilkinsons, a family of skilled blacksmiths, forged anchors, molded and turned the large iron screws used in fish and linseed oil works, manufactured cannon . . . and cast and finished the iron work for early textile machinery set up in Providence and East Greenwich. David Wilkinson, who made the castings for Slater's first carding machines, went on to become the American inventor of the industrial lathe, to experiment with early steam power generation, and to build the power loom which eventually dominated the early nineteenth century textile industry.
>
> Kulik and Bonham (1978)

As early as 1793, fourteen years before Robert Fulton demonstrated the *Clermont*, David Wilkinson cast and assembled a steam engine that he used successfully to run a paddleboat on the Providence River. He continued to work on steam engineering "with striking success," and installed a steam engine at his own mill by 1810 (Kulik and Malone 1977). The demands of the growing textile industry quickly became too great for any one metal shop, however, and by 1810 at least 12 firms were manufacturing machinery in Rhode Island. By 1832, there were 10 foundries and 30 machine shops employing some 1250 men. Most of the metal industry was located adjacent to the cotton and woolen mills in the Blackstone and Pawtuxet valleys and in Providence. Larger metal working firms in North Providence, Cumberland, and Smithfield "became noted for the production of screws, nuts, bolts, muskets, edge tools, and, above all, textile machinery . . . In addition, there were several hundred petty ventures making simple articles ranging from reeds, bobbins, spools, pickers, shuttles, brushes, and temples for the textile industry to tacks, nails, utensils, firearms, shovels, plows, and tools for various other markets, local as well as external" (Coleman 1963). Further up the Blackstone

River, Worcester, MA had, by 1830, become a center for manufacturing pickers, cards, tacks, broad and satinet looms, napping, shearing, and brushing machines (Plate 2). The metals involved in the manufacture of textile machinery included bar and pig iron, steel, blocked tin, zinc, copper, and lead (Clark 1929a).

In contrast to many areas of the country, the Civil War "ushered in an age of industrial prosperity for Rhode Island" and

> . . . fueled the growth of precision manufacturing in Providence. The Burnside Rifle Works, Providence Tool Company, and Schubarth Company all profited handsomely from producing munitions for the Union army. The Corliss Company and Providence Steam Engine Company built marine engines and boilers for the navy. Builders' Iron Foundry cast heavy ordnance, including 11-inch and 13-inch Dahlgren guns.
>
> With the return of peace, the Burnside Rifle Works retooled to make locomotives and the Providence Machine Company devoted itself to making tools and sewing machines. The Providence Tool Company continued to manufacture guns, filling large contracts for the Turkish government. Brown & Sharpe, which as late as 1853 had only fourteen employees, became the world's largest manufacturer of machine tools. From a small workshop on the East Side it expanded to a large factory at the foot of Smith Hill, covering three blocks and employing 2,500 workers by 1900. The city's manufacturing jewelers did even better. By 1880, when jewelry overtook cotton manufacturing as the city's leading industry, upwards of 150 different firms made Providence the jewelry capital of the world. In Gorham Providence boasted the largest silver-plate manufacturer in the United States. The American Screw Company, Providence Tool Company, and Nicholson File Company also boasted the largest plants of their kind in the country.
>
> Gilkeson (1986)

Smelting

Small amounts of coal had been collected on Aquidneck Island at least since the mid 1700s, but in 1808, the first commercial deposits of anthracite coal were discovered and exploited near the shoreline of Narragansett Bay, at Portsmouth, Rhode Island (Bolhouse 1967, Plate 3). The coal proved difficult to burn in ordinary home and industrial furnaces, however, and it played little or no role in meeting the emerging energy needs of Providence and the Blackstone Valley. Instead, much of the coal was exported, used in local brick manufacture, or consumed by a copper smelter. The smelter was constructed adjacent to the coal mines in 1866 by the Taunton Copper Company to process ore imported from Cuba (Plate 3). During the 10-15 years that the smelter operated, it produced some 900 tons of copper each year (Garmon 1978) and doubtless released numerous metals into the atmosphere. If present day atmospheric emission factors are applied to the annual copper production, the smelter appears to have been a particularly strong source of arsenic, and it may also have emitted more cadmium, copper, and, perhaps, lead to the atmosphere than the total combustion of wood and coal in Providence during the same period (compare Tables 3 and 19). As emphasized later in this report, however, it is impossible to know how much of the emitted metals were actually deposited on the bay or found their way into the bay if they were deposited on the watershed. That the smelter produced some direct fluxes of metals into the bay is evident even today from slag deposits along the shore at Portsmouth.

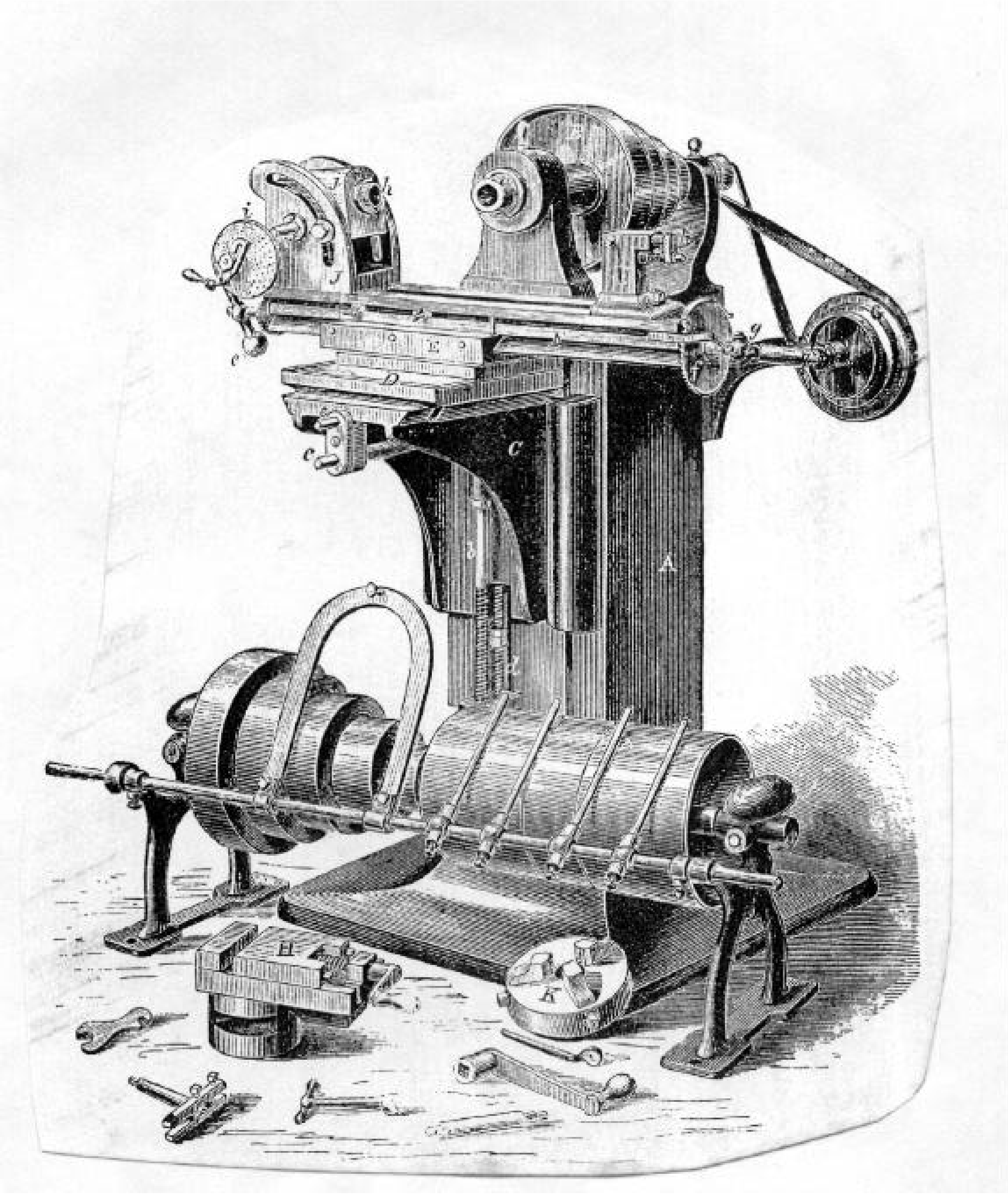


Plate 2 Brown and Sharpe Universal Milling Machine developed in Providence during the 1860s and 1870s. From an 1888 drawing reproduced by Kulik and Bonham

Steam Engines

The expansion and mechanization of the textile industry also stimulated the dramatic development of another major base metal industry. In 1827, Slater again pioneered and again benefited from Wilkinson skills and experiments by installing a steam engine in a large cotton mill constructed in Providence. The successful application of steam power freed the textile mills from the need to locate at sites with water power and thus allowed them to lower land costs and position themselves closer to markets, supplies, and transportation routes. In addition, "Machinery driven by an efficiently governed steam engine operated at a more constant and at a higher speed than that achieved in water power-mills. This improved the quality of yarn and cloth, increased productivity per unit of labor, and, except for the cost of fuel, reduced operating expenses" (Coleman 1963).

The benefits of steam power were so great that it quickly spread throughout all forms of manufacturing. After 1835, "the engine building segment of the base metal industry boomed," and by 1860, it made Providence the leading industrial center in southern New England (Coleman 1963). Again, while several firms produced engines, Providence became famous as a center for their manufacture because of one outstanding engineering talent. George H. Corliss emigrated to Providence in 1844 and soon invented a valve system that greatly increased the efficiency of the reciprocating engine. So important was this contribution that Coleman (1963) ranked it with that made by James Watt a century earlier. While manufacturers were slow to install the new engines, "establishment of the Corliss Works made Rhode Island the nation's leader in the building of steam engines" (Kulik and Bonham 1978) and by the Civil War, they were "a major segment of the Rhode Island base metal industry" (Coleman 1963). When the great Centennial Exposition of the United States opened in Philadelphia in 1876, it was a giant Corliss steam engine from Providence that was used to provide power in Machinery Hall (Plate 4).

Invention and Expansion

The industrial base of primary and fabricated metals that developed so early in the Narragansett Bay watershed continued to expand and diversify until recent years

Table 3

Estimated emission of metals to the atmosphere from the Taunton Copper Company smelter at Portsmouth, R. I. between 1866 and the early 1880s.

	Emission Factor, [a] gt^{-1}	Annual Emission,[b] tons
As	1000-1500	0.9-1.4
Cd	200- 400	0.2-0.4
Cu	1700-3600	1.5-3.2
Ni	900	0.8
Pb	1300-2600	1.2-2.3
Sn	50- 200	0.05-0.2
Zn	500-1000	0.5-0.9

[a] Nriagu and Pacyna (1988).

[b] Based on a mean annual production of 900 tons (Garmon 1978).

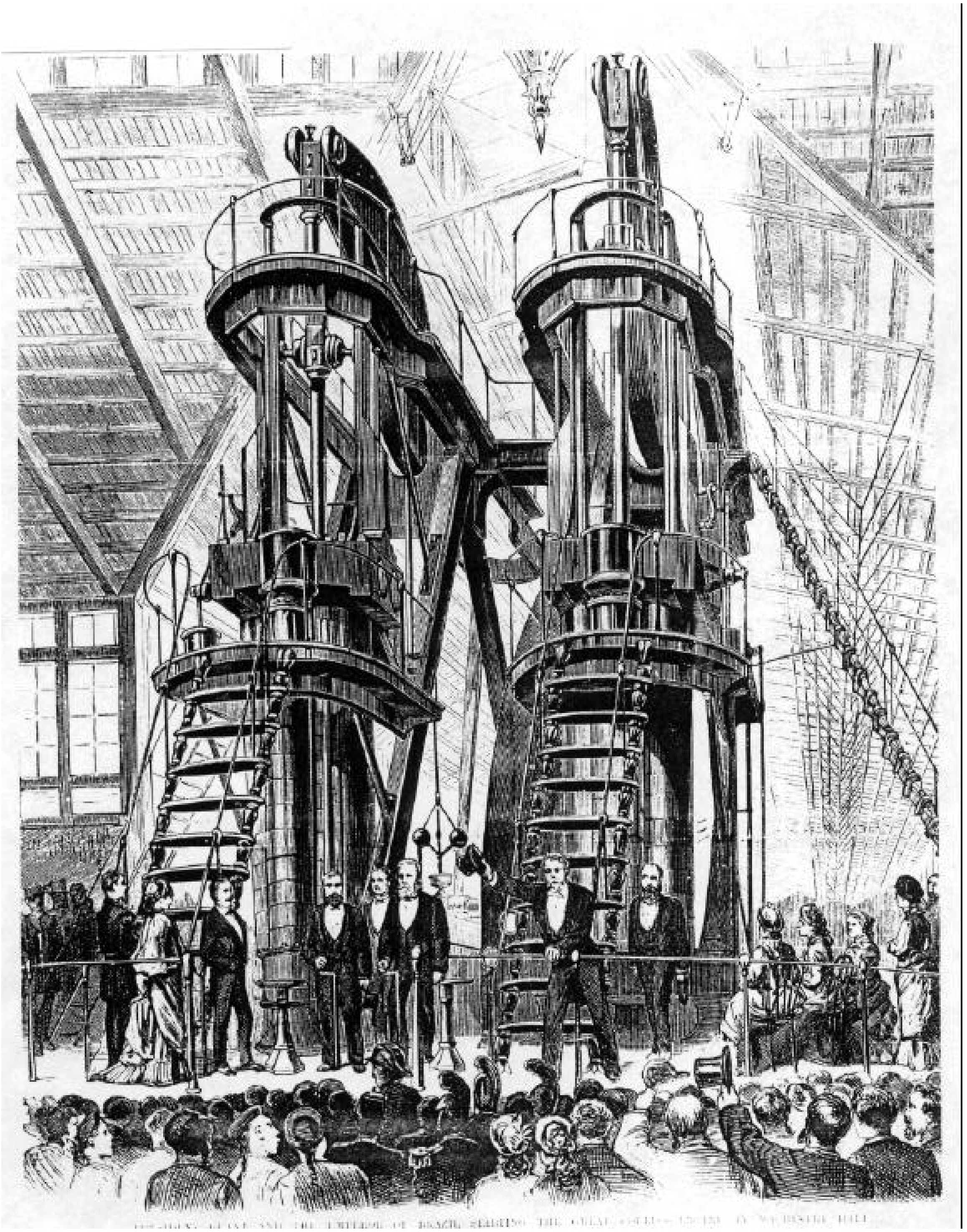


Plate 4: President Grant and the Emperor of Brazil starting the Corliss steam engine that provided the power for Machinery Hall at the 1876 Centennial Exposition of the United States in Phildelphia

(Fig. 1). While this growth was made possible by steam power and, later, by electrical power and the internal combustion engine, it was also driven by the increasing demand for metals in American society. Much of this demand, in turn, was stimulated by a great flowering of technological invention that revolutionized the United States between about 1850 and 1925 (Fig. 2). The development of the railroads, the mechanization of agriculture, the invention and spread of electrical distribution systems, the telegraph and the telephone, the introduction of canning and skyscrapers and automobiles, all contributed to an exponential growth in metal manufacturing.

The speed and extent of this growth is difficult to appreciate for those of us accustomed to living in a fully industrialized nation that has been preoccupied during recent years with a loss of basic manufacturing and the expansion of a service economy. A few examples may be helpful.

In 1829, ". . . the first locomotive had arrived in New York from Britain . . . It had been eagerly inspected, jacked up on blocks, wheels spinning briskly but futilely under steam, because there were no tracks for it to run on. Within a year the Americans were building their own steam locomotives, and tracks were snaking out across the land like iron tentacles" (Raymond 1984) (Fig. 3). In Providence, the Rhode Island Locomotive Works was established in 1865 and eventually reached an output of four steam locomotives per week. By 1889 the firm had produced 2,200 engines (Conley and Campbell 1982).

Once metal stoves were modified to burn coal, they became enormously popular for domestic heating and cooking. Providence was one of eight major centers of stove manufacturing, and by 1850 some 30,000 were made there each year (Clark 1929a).

The introduction of barbed wire and bale wire, as well as the telegraph, telephone, and electricity put a huge demand on the wire pulling industry. And in 1860, 58% of all the wire manufactured in the United States was produced along the Blackstone River, in Worcester, MA (Clark 1929a). Barbed wire was invented in 1874, when about 5 tons were manufactured. Annual production increased to 300 tons in 1875, 6000 tons in 1878 and over 75,000 tons by 1884 (Clark 1929b). The telegraph was invented in the 1840s, but technical difficulties delayed the achievement of a transcontinental line until 1861 (Hofstadter et al. 1959; Furnas 1969). By 1866, Western Union had put up some 76,000 miles of wire and then added over 850,000 miles more by the turn of the century (Fig. 4). The telephone was introduced to the public at the Philadelphia Centennial of 1876, and by 1880 it had become a common business tool (Furnas 1969). Over one million were in service by 1899. After the charge basis for calls was reformed in 1900, hundreds of thousands of telephones were manufactured and installed each year until the Great Depression of the 1930s (Fig. 4).

Plate 5 The Model T Ford Coupe, a major contributor to America's increasing use of metals. From the Mrs. John Howard Benson collection, Providence Public Library.

While Michael Faraday first demonstrated the principle of the electrical dynamo in 1831, it was not until Christmas of 1879 that Thomas Edison was able to demonstrate the first complete electric lighting system. By 1882, America's first city power station opened in New York with 85 customers and 400 electric lights (Raymond 1984). It was a modest beginning, but the impact of electrification on the consumption of copper would prove dramatic. According to V. S. Clark's (1929b) *History of Manufactures in the United States*, "In January, 1884, there were probably not more than two hundred miles of hard drawn copper wire in use in the country. In 1889 there were at least 50,000 miles." Beginning in 1895, the American Electrical Works (later the Washburn Wire Company) produced large amounts of copper wire on the shores of the Seekonk River.

Electric power quickly replaced steam as the immediate source of energy for manufacturing (Figs. 5 and 6) and greatly accelerated the consumption of coal. "Between 1902 and 1929, output of electrical power increased more than nineteen times . . . In 1914, only one-third of all industrial plants were electrified; by 1929, three-fourths were . . . In 1912, only one-sixth of all American families had electricity in their homes, but by 1927 about two-thirds had electrical power" (Melosi 1985). And with the availability of electric power came the invention and manufacture of all sorts of appliances to use it. Virtually all contained metals. According to Melosi (1985), "By 1925, 80 percent of the homes with electricity had electric irons, 37 percent had vacuum cleaners, and 25 percent had washing machines." The production of radio sets increased 20 fold between 1922 and 1925 and another 10 fold by 1947 (Fig. 7). Once television sets went into commercial production after World War II, their manufacture rose from 6000 units in 1946 to over 6,000,000 units in 1952 (Fig. 7).

And, of course, there was the invention of that great metal consumer, the automobile. Henry Ford delivered the first Model T in 1909. By 1913, when the first moving assembly line was installed, the Ford plant was producing a thousand cars a day. One of the reasons for the great success of the Model T ". . . at a time in history when most vehicles were still being made of wood, by hand, was Ford's grasp of metallurgy and the manufacturing advantages of precision-made metal parts. Ford was one of the first to appreciate and use the special properties of metals to be cast, forged, pressed or machined to any pattern, to tolerances which would make parts truly interchangeable, and thus capable of assembly by relatively unskilled workers" (Raymond 1984). By 1927, when the last Model T came off the assembly line, over 16,000,000 had been built, and the automobile was well on its way to becoming a dominant force in the economy and the environment (Plate 5, Fig. 8).

While the base metal industries in the Narragansett Bay watershed did not necessarily produce railroad track or very many automobiles, they did contribute in one way or another to most of these products as well as to thousands of others. As one of the major manufacturing centers in the country, Rhode Island and the Blackstone River Valley participated fully in the industrialization of the United States. While that process began earlier here than anywhere else in America, the environmental impact of industrialization, at least in terms of metal pollution, was probably not large until the mid 1800s. From the Civil War on, the development and spread of steam power, electrical power and the internal combustion engine supported an unprecedented time of industrial expansion, and from the Civil War until the turn of the century, Rhode Island was the most industrialized state in the country (Coleman 1963). "No single thing" wrote V. S. Clark (1929a) "better measures the industrial standing of a nation than its use of metal." Until very recently, the increase in metal pollution in Narragansett Bay reflected increasing wealth and power and a rising standard of living for those working in its watershed.

Precious Metals

In addition to an early development of the base metals industry, Providence can lay claim to being the birthplace of the manufacturing jewelry industry in the United States (Anonymous 1950). The industry developed along two paths pioneered by two brothers, Seril and Nehemiah Dodge. Seril Dodge began the manufacture of silver buckles in Providence in 1775 and the practice of high quality gold and silver work was expanded and carried on by Jabez Gorham after 1813 (Coleman 1963). Nehemiah Dodge opened a jewelry, clockmaking, and goldsmith shop in Providence in 1794 and specialized in the production of lower cost or costume jewelry. "By 1805, the industry employed some thirty artisans to make either fine jewelry or cheaper quality gold necklaces, rings, cases, knobs, chains, twists, keys, and seals" (Coleman 1963).

Jewelry manufacture employed 175 people by 1815, but went through two sharp depressions before beginning a sustained expansion after 1820. Between 1850 and 1860, the application of steam power and mechanization stimulated sharp growth, especially in costume jewelry (Table 4), and by 1880, Providence was "the jewelry capital of the world" (Gilkeson 1986). However, while a few large manufacturers emerged, such as Gorham and Son, the average firm employed only a dozen workers (Plate 6). The small size of most precious metal manufacturers prompted them to locate together in the central Providence business district where they could buy power from larger factories (Coleman 1963). The small size and central location of the industry is still characteristic.

In 1957, 56% of Rhode Island firms had fewer than ten employees and 88% had fewer than 50

Table 4

Early growth of the Rhode Island precious metal industry
(compiled from data in Coleman 1963).

	Number of Firms	Number of Employees[1]
1805		30
1810		100
1815		175
1832	27	280
1850	59	704
1860	91	2043

[1] See Figure 10 for more recent trends.

(Bride 1959). While employment in precious metal industries in the state did not continue to expand as it did in base metal industries (Fig. 9), the value added by manufacture in fine jewelry, silverware, and costume jewelry exceeds that of base metal manufacturers (Fig. 10).

Situated downstream from ". . . one of the greatest jewelry manufacturing centers in the world" (Anonymous 1950), it is ironical that Narragansett Bay now contains far more gold and silver than it did when the European explorers first came searching for precious metals.

The Emission of Metals

Metals may be emitted or released into the environment in association with a great variety of manufacturing activities, not all of which are confined to the primary and secondary metals industries. The production of cement, for example, releases chromium, nickel, lead, and zinc to the atmosphere (Nriagu and Pacyna 1988). While it is reasonable to assume that the dramatic and sustained growth of metals industries around Narragansett Bay involved increasing amounts of smelting, filing, grinding, etching, electroplating, etc., it is virtually impossible to reconstruct a quantitative history of the metals emissions that may have resulted. Such a calculation would require a detailed inventory of manufacturing activity and emission factors appropriate to past technologies. A great variability among industrial plants and their raw materials and products makes such an effort difficult for present conditions, let alone for those of a century ago (Nraigu 1979; Nraigu and Pacyna 1988). However, even a qualitative historical relationship between industrial growth and metal inputs to the bay should take into account some major changes influencing the disposal and fate of waste metals. The most important of these must certainly have altered dramatically the rate at which metals entered the bay from the atmosphere and in wastewater.

Roads, Roofs, and Runoff

While most of the metals emitted to the atmosphere from tribal campfires, colonial fireplaces, and early iron smelters were deposited on fields and forests, this was not the case in the rapidly developing urban and industrial centers. As vegetation and soil were replaced by roads, sidewalks, and roofs, pollutants deposited from the atmosphere in dry fall and in rain were no longer retained. Instead, they could be washed quickly into streams and rivers or directly into the bay in storm water runoff.

While it is often assumed that paved streets came with the automobile, the much greater efficiency of horse drawn wagons on paved surfaces (Table 5) as well as public

Table 5

Relative size of a wagon load that could be pulled with equal ease by a horse over level ground with various surfaces.[1]

Iron track	100
Asphalt[2]	60
High quality stone block[3]	30
Common stone block	20
Common macadam[4]	13
Cobblestone	10
Gravel	5

[1] Road Transportation Exhibit, National Museum of American History, Smithsonian Institution, Washington, DC.

[2] Compressed asphalt blocks were used in San Francisco as early as 1869, but became more common after 1880. The first asphalt streets in the U. S. were laid in New York in 1869. This material became popular after about 1905.

[3] Stone blocks were laid in New York in 1852 and became common after 1860 (see Plate 4).

[4] John Loudon Macadam devised a system in 1815 by which roads were formed from stone broken into 5 cm angular pieces, compacted into a layer about 20 cm thick, and elevated in the center for drainage.

sanitation and basic human comfort stimulated the early application of various kinds of paving materials on city streets (Melosi 1981). Cobblestones, for example, were first used in the United States in Pemaquid, Maine, about 1625. For many years they were the most common paving material used in most cities, though they obviously left much to be desired from the point of view of the draft horse (Table 5). Even asphalt, a paving material usually associated with the spread of automobiles, had already been used to cover almost one-third of urban streets by 1909, the year the Model T Ford was introduced (see Fig. 8) (Melosi 1981).

By 1878, the year the Providence City Engineer began to include such information in an annual report, the city had 17 km (about 10.4 miles) of streets paved, for the most part, with granite blocks (Table 6). The paving of Providence continued at a steady pace until about 1910 (Fig. 11, Plate 7) when construction accelerated. The increase may have been in response to the growing number of automobiles or to accommodate new mechanical street cleaning equipment (Melosi 1981). Street cleaning had emerged as a major urban concern for two reasons. First, there were still large numbers of horses in the cities, and their manure posed a constant nuisance:

> pulverized horse dung . . . blew into people's faces and the windows of their homes, and over the outdoor displays of merchants' wares. The coming of paved streets accelerated this problem, as wheels and hoofs ground sun-dried manure against the hard surfaces and amplified the amount of dust.
>
> (Tarr 1971)

Second, Providence had begun a major expansion of its combined storm water and sanitary sewer system beginning about 1890, and it was increasingly recognized that paved and cleaned streets reduced the amount of mud and silt entering the system and helped to alleviate the pollution effects of combined storm water discharge (Tarr 1979).

Unfortunately, it appears from more recent reports that until after World War II macadam roads were not included in the City Engineer's inventory of paved streets, so it is not possible to know how the total area of paved surface increased over time (Fig. 11, Table 6). The problem of assessing the likely impact of street paving on metal fluxes to the bay is further confounded because the various types of surfaces used have somewhat different retention capacities for different metals and, while the paving increased runoff, it also made more effective street cleaning possible. On balance, however, as the city became more "impermeable" and as more and more metals were emitted to the atmosphere, storm water runoff must have become an increasingly important pathway by which metals entered the bay.

Table 6

Paved streets in the city of Providence, R.I. as reported by the City Engineer.

			Paving Material, %		
Year	Length, km	Area*, m^2	Granite Block	Asphalt, etc.[a]	Other[b]
1878	17	152,700			
1880	22	203,000			
1884	31	334,400			
1896	57	521,500	74.5	8.4	17.1
1900	64	584,500	74.3	11.6	14.1
1905	67	624,800	75.8	12.2	13.3
1910	75	684,000	74.0	11.0	15.0
1915	106	969,700	59.6	29.4	11.0
1920	127	1,165,900	54.9	36.1	9.0
1925	140	1,534,450	43.5	48.8	7.7
1928	158	1,445,300	36.4	59.6	4.0
1943-44	191	1,744,500	10.8	66.0	23.2[c]
	(354)	(2,907,000)		(Bituminous macadam)	
	545	4,651,500			
1950	203	2,265,200	5.9	49.8	44.3[c]
	(331)	(2,670,500)		(Bituminous macadam)	
	(17)	(134,000)		(Waterbound macadam)	
	551	5,069,700			
1960	589	6,485,800		Includes macadam	
1971	595	6,530,700		Includes macadam	

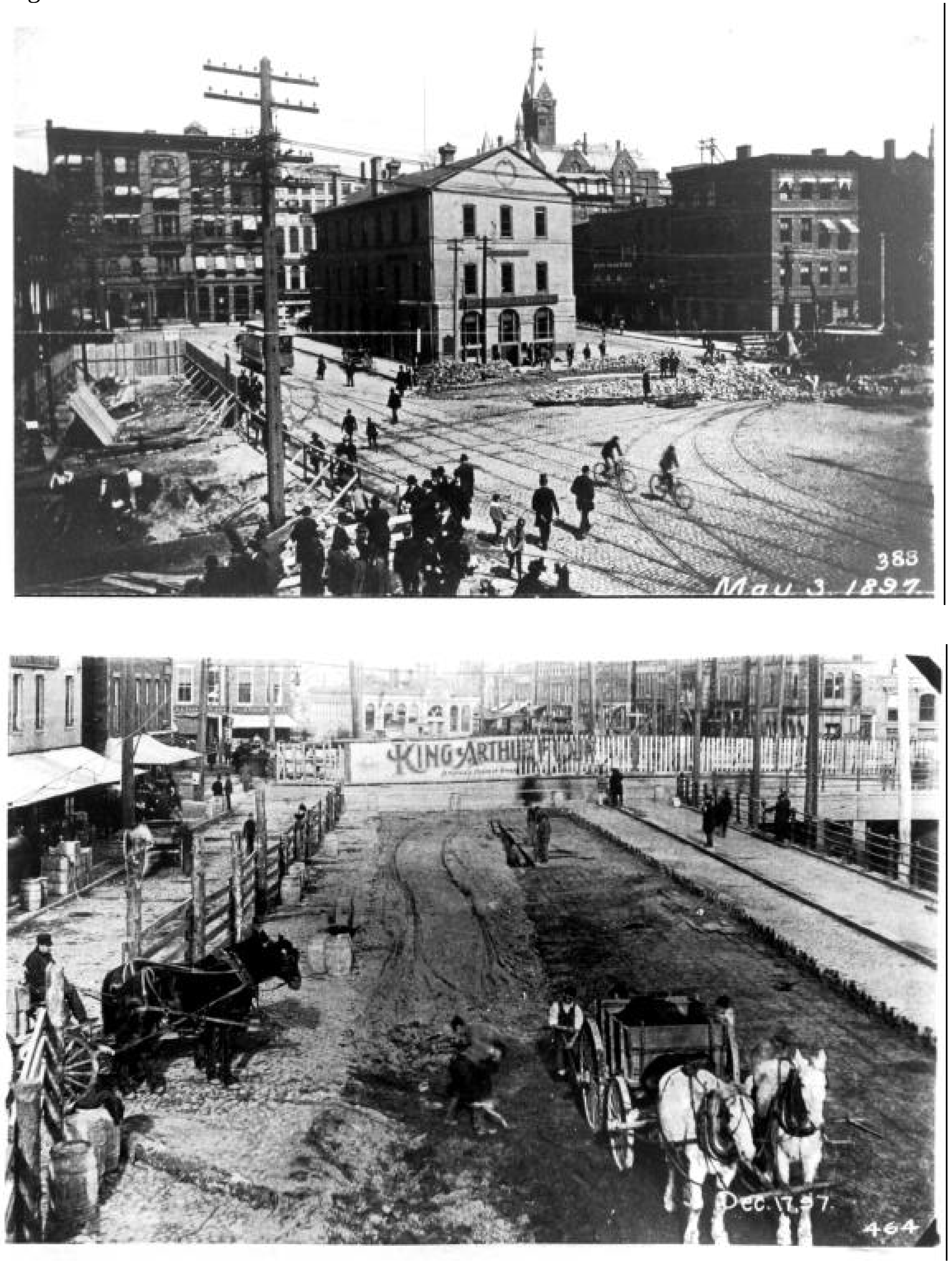
[a] Includes vulcanite and bitulithic.

[b] Includes cobble, wood block, brick.

[c] Includes bituminous concrete (1.3%), reinforced concrete (1.5%), colprovia (20%).

*In some years area was calculated from length using the City Engineer's working average road width of 9.1 m.

Plate 7 Paving streets in Providence with stone blocks in 1897. Sewage construction is also underway on the left of the upper photograph. Photos from the Providence City Engineer's office.



As part of the paving of streets, the city also constructed sidewalks. No separate account of their area was reported, but based on construction data for 1879-1884, sidewalks may have equaled about 18% of the street area.

The growth of industry in Providence was accompanied by a dramatic growth in the population of the city that averaged 3.3% y^{-1} between 1800 and 1910. To put this in some perspective, the 15 countries with the most rapid growth of population in 1987 had annual growth rates averaging just under 3.1%. Kenya, with the world's fastest growing population, is increasing at 3.9% y^{-1} (Brown et al. 1988). While the growth of the city was largely the result of immigration, the impact of such a rate of increase on the urban environment must have been felt strongly.

Between 1878 (the first year of the City Engineer's Report) and 1928 (the last year for which data were available before the Great Depression), building permits were issued for 33,355 structures. Assuming a modest 100 m^2 of ground covered per structure gives an increase in "impermeable" surface of 3.3 km^2. During the same period, the population of Providence increased by about 148,000, giving a ratio of people to buildings of 4.4. At the time the City Engineer began to report the number of building notices issued each year, the population of the city was already around 100,000. If the same ratio of buildings to people applied, an additional 23,800 structures covering 2.4 km^2 may be added to give a total of 5.7 km^2 of ground covered by buildings at about 1930. If the reported area of paved streets in 1928-30 (Table 6) is doubled to approximately account for macadamized surface (Fig. 11) and increased by 18% to include sidewalks, an additional 3.3 km^2 of hard surface had been constructed. The total "impermeable" surface that covered the city by 1930 then amounted to over 9 km^2 or 19% of the total area of Providence at that time. This estimate is certainly low because some of the structures included factories, warehouses, and stores much larger than 100 m^2. Even with 9 km^2 of "impermeable" surface, an annual rainfall of 1 m would produce 9 million m^3 of runoff, or an average of 25,000 m^3d^{-1}. This was equivalent to about 16% of the sewage being discharged from the city treatment plant at Fields Point.

Sewage Treatment [1]

Virtually all manufacturing facilities release some metals in waste water and cooling waters (Table 7). Before 1870 there was no city sewer system in Providence, and it can be assumed that manufacturers simply ran their untreated effluents directly into the nearest receiving water. The city began constructing sewers in 1871 and expanded the system

[1] This section is based largely on Annual Reports of the Providence City Engineer.

Table 7

Emission factors for the release of trace metals to receiving waters from various types of industrial activity, µg m^{-3} of effluent (Nriagu and Pacyna 1988).

	Smelting and Refining			Manufacturing Processes	
	Iron and Steel	Non-ferrous Metals	Steam Electric *Generating*	Metals	Chemicals
As		0.5-6.4	0.04-0.12	0.01-0.06	0.12-1.4
Cd		0.004-1.8	0.001-0.04	0.02-0.07	0.02-0.5
Cr		1.5-10	0.5-1.4	0.6-2.3	0.5-4.8
Cu		1.2-8.5	0.6-3.8	0.4-1.5	0.2-3.6
Hg		0.001-0.002	0-0.6	0-0.03	0.004-0.3
Mn	2.0-5.2	1.0-7.5	0.8-3.0	0.1-0.8	0.4-3.0
Ni		1.0-12	0.5-3.0	0.008-0.3	0.2-1.2
Pb	0.2-0.4	0.5-3.0	0.04-0.2	0.1-0.9	0.08-0.6
Sb		0.04-3.8	0-0.06	0.1-0.6	0.004-0.5
Zn	0.8-3.5	1.0-10	1.0-5.0	1.0-5.5	--

greatly beginning in 1890 (Fig. 12). Like other American cities embarking on sewer construction during the 1860s and 1870s, Providence chose to develop a combined system that would carry sanitary wastes and storm water in the same pipes (Plate 8) (Tarr 1979). It seemed a natural extension for industries to begin to tie into the same expanding system to remove their wastes.

At first, the sewer system simply collected wastewaters and runoff and carried them to various discharge points along the Moshassuck, Woonasquatucket, Seekonk, and upper Providence Rivers. The flow was unmetered and untreated. It was not until late in 1892 that interceptor sewers began to carry the sewage away from the city harbor area for discharge further down the Providence River at Fields Point (Plates 9-11). During the next four years, the effluent released at Fields Point was also unmetered and untreated and came from about 4.4 km^2 (1090 acres) of the higher parts of the city that could drain by gravity to the discharge site. On 26 December, 1896 the Ernest Street Pumping Station began pumping sewage to Fields Point from another 4.6 km^2 (1140 acres) of the most densely developed part of the city. Altogether, about 21% of the city area was now discharging its sewage untreated at Fields Point. Beginning in 1897 the amount of sewage released at Fields Point was measured, however, and it is possible to get some estimate of the contribution from manufacturing by comparing the average dry weekday flow with the average dry Sunday flow. Between 1897 and 1909 (the last year the data were summarized in this way), the average Sunday flow equaled 61% of the weekday flow, with an annual range from 55-67%. It seems clear that industries had been quick to take advantage of the expanding sewer system and that waste water from manufacturing made up about one-third of the increasing discharge into Narragansett Bay at Fields Point (Fig. 13). Since Providence had a combined system, increasing amounts of the metals and other pollutants in urban runoff were also discharged at Fields Point, though the largest amount of storm water runoff was released through numerous outfalls further up the Providence and Seekonk Rivers.

On 17 April, 1901 the first[1] treatment of the sewage began at Fields Point when chemical precipitation tanks were put into operation and lime was used to settle sludge from the effluent. It was reported that this process removed over 80% of the suspended albuminoid ammonia and 55% of the total organic matter before the effluent was discharged. By 1913, however, the annual reports from the treatment plant claimed only a 10% removal of total solids, though bleaching of the effluent had been added in 1912 that reduced the number of <u>E.</u> <u>coli</u> bacteria by 97%.

[1] When the pumping station went into operation, bar screens were used to remove objects larger than 1.9 cm from the effluent.

A remarkable feature of the treatment plant operation was that the treated effluent was discharged into the Providence River (at about 12 m below mean high water) only during ebb tides. Effluent generated during flood tides was held in large tanks. This practice continued until 1936, when the plant was converted to the activated sludge process and discharge became continuous.

A shortage of lime during World War I reduced chemical precipitation in 1918 and 1921 and eliminated it entirely in 1919 and 1920, though chlorination continued on a more or less regular basis throughout the war. By 1928 all of the dry flow sewage from the city was being treated and released at Fields Point, though many industries doubtless continued to discharge their manufacturing wastes directly into nearby waters.

With the introduction of chemical precipitation in 1901, the treatment plant began to produce large quantities of sludge. At first this material was dewatered and used as fill on the site, but after seven years the city was forced to find a new disposal area. On 8 July, 1908, the city scow *Pomeganset* took the first load of sludge down the river into Narragansett Bay and dumped it in deep water (about 38 m) south of Prudence Island. This dumpsite was used continuously until 1949, when the city began to burn the sludge.

During the 41 years that the Prudence Island dumpsite was used, sludge containing some 137,400 tons of dry solids was released in the middle of the bay. If the metal content of this material was even roughly similar to contemporary Providence sludge, the dumping added large amounts of metals to an undeveloped part of the bay (Table 8).

A particularly interesting feature of the dumping record is that it shows no correlation with the reported discharge of effluent from the Fields Point treatment plant or with the number of houses tied into the sewer system (Fig 14). Since the total amount of settleable solids reaching the treatment plant must have been increasing, it seems reasonable to conclude that increasing amounts of metal-rich particulate matter were being discharged into the Providence River from about 1910 until 1950, when new vacuum filtration equipment was fully in service and sludge was incinerated.

Several factors may have contributed to the declining efficiency at Fields Point, including shorter retention times for effluent in the settling tanks as the flow into the plant increased, and reductions in the amount of lime used for precipitation. For example, in 1909, 84 g m^{-3} were applied while only 16 g m^{-3} were used in 1932. The impact of lime shortages during World War I has already been mentioned and is evident in the dumping record (Fig. 14). The removal of solids per volume of effluent treated amounted to 345-400 g of dry matter m^{-3} during 1909-1911, but fell to about 110 g m^{-3} by the early 1920s. Part of this apparent reduction in efficiency may have been due to dilution. The amount of waste water treated at Fields Point rose in parallel with the consumption of water from the

Plate 9 Construction of sewers at Field's Point. From the 1891 Annual Report of the Providence City Engineer.

Plate 10 Excavating and plank driving machinery involved in sewer construction along the Providence River above Fields Point. From the 1894 Annual Report of the Providence City Engineer.

Plate 11 Outlet chamber for sewage discharge at Fields Point. From the 1893 annual Report of the Providence City Engineer.

Table 8

Estimated addition of various metals to Narragansett Bay at the Prudence Island sewage sludge dumpsite between 1908 and 1949.

	Sludge Composition[1]	Total Added[2]	Average
Annual	mg kg^{-1} dry wt	tons	**Error!**
Hg	1.5	0.21	0.005
Ag	1025	140	3.4
Cd*	100	13.7	0.33
As**	16	2.2	0.054
Cr	800	110	2.7
Cu	2700	370	9.0
Ni*	1000	137	3.3
Pb	1150	158	3.9
Zn	4600	632	15.4

[1] Analyses of sludge from Fields Point treatment plant reported in the 208 Water Quality Management Plan for Rhode Island (R. I. Statewide Planning 1979). Sewage sludges vary widely in their content of metals (National Research Council 1984, Straub 1989) and these values may underestimate the concentrations that would have been found in Providence during earlier times when manufacturers were less aware of pollution and used more wasteful technologies. The concentration of lead may be an exception, since some portion of this metal in the modern sludge came indirectly from the combustion of leaded gasoline and lead was not used as an additive in motor fuel until 1923.

[2] Based on a total addition of 137,340 metric tons of solids compiled from annual reports of the Providence City Engineer.

*As discussed on pages 86 and 94, the inputs of Ni and Cd are overestimated before about 1940 and 1925, respectively, by the assumptions used here.

**New York City sludge, 1983 analyses (National Research Council 1984).

Municipal supply system (Fig. 15), and the daily per capita consumption of water increased from 242 to 276 liters between 1909 and 1924, or about 2% y^{-1}. It is difficult to extrapolate this statistic into later years because from 1920 on, census data were used for the population of Providence rather than the earlier and more detailed estimates of the size of the city population actually served by the public water system that were maintained by the Superintendent of Health. A significant suburban population was also served by the Providence water supply. Use of the census data results in a per capita consumption that is 30-40 liters per day greater than the Health Department estimate, with an increase of about 1.7% y^{-1}. Some of the increased water use per capita was probably accompanied by an increase in the generation of particulate waste per capita and did not really provide any dilution of the sewage. Unfortunately, there appear to be no historical data on the concentrations of suspended matter in the raw effluent reaching the plant. It is also not known to what degree increased industrial use of water contributed to the rising per capita consumption.

If we accept a dilution correction of 2% y^{-1} as a rough working number and assume that the concentration of “settleable solids” in the sewage reaching the Fields Point plant (after correction for dilution) remained as it was during 1909-1911 (360 g dry weight m^{-3}), it is possible to calculate the potential production of sludge that would have been harvested if the plant had continued to operate with its earlier efficiency. The difference between this potential harvest and the amount of sludge dumped below Prudence Island represents a very rough lower estimate of the discharge of sludge-type particulate material into the Providence River (Fig. 16). If the metal contents of this material are assumed to be the same as that dumped in the lower bay (Table 8), it is evident that sludge-type material was a much larger source of metals to the Providence River than at the official sludge dumpsite (Table 9).

A similar exercise can be carried out for the eight years between the time untreated effluent began to be discharged at Fields Point and the beginning of active precipitation treatment and land disposal of sludge in 1901. Unfortunately, the lack of annual sewage discharge measurements before 1897 makes it necessary to estimate the flows for 1893 through 1896 using the ratio of sewage produced to water consumed (1.33 in 1897) and annual water consumption data for those years. The combination of estimated and measured flows suggests that, on average, some 17,972,000 m^3 y^{-1} of untreated effluent were discharged during the eight-year period. Before multiplying this by 360 g m^{-3}, the weight of dry sludge solids removed per unit volume when precipitation began, it may again be appropriate to consider that the raw sewage entering the plant in 1909 had already been diluted somewhat by rising per capita water consumption compared to the effluent of

Table 9

Calculated input of various metals from the Fields Point Sewage Treatment Plant to the Providence River in association with “settleable solids” not removed during treatment. Assumptions regarding metal concentrations in sludge type material are given in Table 8.

	Tons Per Year						
Metal	1915	1925	1930	1935[a]	1940[a]	1945	1949
Hg	0.007	0.012	0.013	0.013	0.014	0.024	0.025
Ag	4.5	8.4	8.8	9.0	9.8	16.3	17.4
Cd*	0.43	0.82	0.85	0.87	0.95	1.6	1.7
As	0.07	0.13	0.14	0.14	0.15	0.25	0.27
Cr	3.5	6.6	6.8	7.0	7.6	12.8	13.6
Cu	11.9	22.1	23.0	23.6	25.8	43.0	45.8
Ni*	4.6	8.1	8.4	8.8	9.5	15.8	17.2
Pb	5.0	9.4	9.8	10.0	11.0	18.3	19.4
Zn	20.4	37.8	39.3	40.2	44.1	73.3	78.2

[a] Dilution correction held constant at 1930 value during the Depression (1931-1939). See Text.

*As discussed on pages 86 and 94, the inputs of Ni and Cd are overestimated before about 1940 and 1925, respectively, by the assumptions used here.

1893 through 1900. While there was no trend in per capita water consumption during the earlier period, the mean value for those years was 16% lower than reported for 1909. To compensate for this, the estimated volume of sewage can be increased by a factor of 1.16.

When multiplied by the concentration of settleable solids (360 g m^{-3}), this gives an average discharge of sludge-type material equivalent to some 7500 tons dry weight y^{-1}. Applying the same assumption concerning the composition of this material used in Tables 8 and 9 reveals the very large amounts of metals that may have been released for the first time into the mid-reaches of the Providence River when the sewage effluent first began to be discharged at Fields Point (Table 10).

While there are obviously many assumptions in these calculations, and the resulting values are no more than very rough approximations, they may be useful in setting some probable lower limits on the extent of earlier metal inputs to the bay in sewage sludges. Additional metals would have been discharged in dissolved form and in association with finer solids. It is unclear whether an estimate of that historical input using recent metal concentration data is more or less credible than the estimate of inputs associated with larger sludge-type solids. The calculation is particularly tenuous because no measurements of metals in the Fields Point effluent were made before the plant converted to an activated sludge process or before new sludge collection equipment was added in 1948-1949 and 1958-1961. The latter improvements removed very large amounts of solids (see Fig. 17) amounting to 875 g dry weight m^{-3} of effluent treated in 1961. This retention efficiency was almost 2.5 times that of the early 1900s. This level of performance was not maintained, however, and the removal efficiency fell to 460 g m^{-3} in 1964 and to 350 g m^{-3} in 1970. By 1975, when total sludge production was no longer reported, a comparison of the concentrations of total suspended solids in sewage arriving and departing the treatment plant indicated a removal of only 34 g dry weight m^{-3} of effluent (R. I. Statewide Planning 1979). It seems likely that the situation was similar two years later, when particulate and dissolved metals were measured in the effluent. If so, then these measurements included most of the fine particulates collected as sludge in the late 1950s and throughout the 1960s. For this reason it is probably best to estimate the total release of metals from the treatment plant prior to 1950 by adding the earlier estimates of sludge discharge based on removal efficiencies of 1909-1910 to a calculation of dissolved and fine particulate metals discharged that is based on measurements of annual historical effluent flow and total metal concentrations in 1977. Using the higher sludge removal efficiencies of the 1960s would double count the particulate metals measured in the effluent in 1977.

The concentration measurements made in 1977 seem to be the oldest reliable data available. They were made a year before the Fields Point plant essentially ceased to

Table 10

Calculated input of various metals to the Providence River in association with settleable solids contained in sewage discharged at Fields Point during the eight years prior to the application of chemical precipitation treatment. Assumptions regarding metal concentrations in the sludge type material are given in Table 8.

	1893 through 1900	
Metal	Average Input, tons y^{-1}	Total Input, tons
Hg	0.01	0.09
Ag	7.7	61
Cd*	0.75	6.0
As	0.12	0.96
Cr	6.0	48.0
Cu	20.2	162
Ni*	7.5	60
Pb	8.6	69
Zn	34.5	276

*As discussed on pages 86 and 94, the inputs of these metals are overestimated by the assumptions used here.

Function as anything more than a chlorination facility because of improper maintenance (J. Fester, R. I. Dept. of Environmental Management, personal communication). Similar concentrations were found in 1982, after the plant was repaired and before the current industrial pretreatment program began to reduce metal loadings to Fields Point (Hoffman et. Al. 1984).

If the 1977 concentration data (Table 11) are applied to the effluent discharged between 1909 and 1950, and the results added to the calculated metal discharge associated with sludge material (Table 9), it is possible to get some sense of the potential total metal loading to the Providence River from the treatment plant and the probable importance of the declining efficiency of the plant in increasing greatly the discharge of metals like Cd and Pb that associate strongly with settleable material (Table 12). The total metal loading to the river, of course, also included inputs from storm water runoff, from industrial discharges not connected to the sewer system, from direct atmospheric deposition, and from upstream flows of the Blackstone River and other tributaries.

Since the sewage treatment system received both domestic and manufacturing waste, it may be of some interest to evaluate the potential contribution of the expanding human population (Fig. 14) to the metals discharged at Fields Point. This can be assessed in a very approximate way by assuming “a generous allowance” for food intake of 3000 kcal d^{-1} person $^{-1}$ (Kleiber 1961). At 5 kcal g^{-1}, this converts to an annual per capita ration of 220 kg dry weight. Assuming that this is 25% vegetable matter and 75% meat or animal products and that the metal contents of the food are comparable to those of “average” terrestrial plants and animals (Table 13), the amounts of metals released by one person at steady state appear small (Table 14). Even in 1915, when the population served by the Providence sewer system was 218,000, the total contribution of nutritional metals to the discharge from Fields Point was trivial (Table 14).

The Burning of Solid Wastes

After November of 1949, vacuum filtration equipment was put into operation at Fields Point to harvest sludge, and the material collected was burned on site in a new incinerator. The amount of sludge collected with the new equipment rose almost immediately to around the level harvested in 1910 and suggests that the previously calculated discharge of larger solids to the Providence River may have been approximately correct (Fig. 17). Sludge production increased dramatically again around 1960, when even better filters were installed (J. Fester, R. I. Dept. of Environmental Management, personal communication). If the additional, presumably finer particulate sludge collected after 1960 (Fig. 17) contained concentrations of metals assumed to be characteristic of the

Table 11

Concentrations of some metals in sewage effluent from the Fields Point Treatment Plant before initiation of an industrial pre-treatment program, g m^{-3}.

1977 Data[1]

Cd	Cr	Cu	Hg	Ni	Pb	Zn
0.004	0.15	0.98	0.001-0.005	0.89	0.01-0.10	1.55

1982 Data[2]

Cd	Cu	Ni	Pb
0.0012-0.0014	0.53-0.83	0.59-1.10	0.037-0.19

[1] Compiled from R. I. Dept. Environmental Management monthly and self-monitoring files by Olsen and Lee (1979). No data on Pb or Hg were reported from Fields Point; the values given here are ranges for seven other treatment plants discharging to Narragansett Bay.

[2] From Hoffman et al. (1984). Low and high ranges represent flow-weighted mean concentrations for storm events and dry weather conditions, respectively.

Table 12

Calculated inputs of various metals from the Fields Point Sewage Treatment Plant to the Providence River in addition to those associated with “settleable solids” (see Table 9). Concentration ranges for metals given in Table 11.

	Tons Per year of Dissolved and Fine Particulate Metals		
	1915	1930	1945
Cd**	0.14	0.20	0.26
% of total*	25	19	14
Cr	5.15	7.45	9.52
% of total*	60	52	43
Cu	33.7	48.8	62.3
% of total*	74	68	59
Hg	0.03-0.17	0.04-0.25	0.06-0.31
% of total*	81-96	75-95	71-93
Ni**	30.6	44.3	56.6
% of total*	0.34-3.43	0.49-4.96	0.63-6.34
Pb	0.34-3.43	0.49-4.96	0.63-6.34
% of total*	6.4-40	4.8-34	3.3-26
Zn	53.2	77.0	98.4
% of total*	72	66	57

*Metals dissolved and associated with fine grained particulates not removed as sludge divided by the sum of such metals plus the amount associated with sludge type materials released to the river (Table 9).

**As discussed on pages 86 and 94, the inputs of these metals are overestimated by the assumptions used here.

Table 13

Estimates of the content of some trace metals in terrestrial plants and animals, mg kg^{-1} dry weight (Bowen 1966).

	Plants	Animals
Cu	14	2.4
Cr	0.23	0.07
Mn	630	0.2
Ni	3	0.8
Pb	2.7	2.0
Zn	100	160

Table 14

Comparison of the potential release of metals in human waste with the total estimated release of metals from the Fields Point Sewage Treatment Plant in 1915.
Population served by the system was 218,000.

Discharged	Annual Per Capita Consumption of Metals, g	Nutritional Metals Released by Population of Providence, tons	Total Metals by Fields Point Sewage Treatment Plant, tons*
Cu	1.17	0.25	54.6
Cr	0.024	0.005	11.4
Mn	34.68	7.56	--
Ni**	0.30	0.06	38.5
Pb	0.48	0.10	9.2-12.3
Zn	31.90	6.95	89

*Sum of calculated release of dissolved and fine particulates (Table 12), non-precipitated sludge-type materials (Table 9) and sludge dumped below Prudence Island (Table 8).

**As discussed on page 86, the discharge of Ni from Fields Point is overestimated in 1915 by the assumptions used here.

Heavier sludge (Table 8), the particulate loading of metals from the Fields Point Treatment Plant to the Providence River may have been over twice that calculated in Table 9 (compare Figs. 16 and 17). In any case, it is evident that the improved efficiency of particulate removal and the switch from sludge disposal in the bay and river to sludge incineration must have reduced greatly the total input of metals such as Cd, Pb, and Ni (Table 12).

The disposal of sludge through incineration, followed by land fill of the ashes, did not eliminate the possibility that some of the metals contained in the sludge would find their way back into the bay through direct deposition from the atmosphere and in storm water runoff. It is impossible to evaluate this pathway in any exact way because there is a considerable range in emission factors for sludge incineration (Table 15), and neither the actual deposition of metals on the bay and watershed nor the retention of metals by the landscape are known. Calculation of the probable range of emission for various metals suggests, however, that the input of metals from sewage sludge was not large after 1960 (Table 15). An exception to this condition was the period during 1978 and 1979, when the Fields Point plant virtually ceased to operate as a treatment facility (J. Fester, personal communication).

Two smaller sewage treatment plants began operation on the Seekonk and Providence Rivers in 1952 and 1950, respectively, but both the Blackstone Valley and East Providence plants disposed of their sludge on land (J. Fester, personal communication).

Sludge was not the only waste material incinerated. Sometime before 1928, Providence began operating city incinerators to dispose of rubbish and garbage, though data on the amount of material burned appear not to have been reported in earlier years. Between 1951 and 1959, four times more garbage and almost three times more rubbish than sewage sludge was incinerated, though the increased efficiency of solids removal at Fields Point after 1960 quickly made sludge approximately equal in terms of the mass of material burned (Fig. 18).

It is as difficult to estimate the flux of metals from garbage and rubbish incineration through the atmosphere to the bay as it was for sewage sludge. On the basis of the potential emission of metals from refuse burning, however, it appears that this source must have been even smaller than the incinerated sludge (Table 16). Other larger cities in the Narragansett Bay watershed must also have used incineration as it became a popular alternative to swine feeding and reduction during the period between 1910 and 1920. From the late 1930s, however, “the number of incinerators declined sharply as they had greater competition from sanitary landfill operations” (Melosi 1981).

Passage of federal clean air legislation closed the Fields Point incinerator and forced Providence to switch to land-fill disposal of all solid wastes in 1970. The smaller towns around Narragansett Bay burned garbage and rubbish in open dumps and these must have

Table 15

Estimate of the emission of metals to the atmosphere from the incineration of sewage sludge at Fields Point (Fig. 17).

Metal	Emission Factor,[a] g t^{-1} sludge	Average Emission,[b] t y^{-1} (1950-1971)	Maximum Emission, (1960-1961, tons,)
Cd	1-12	0.02-0.25	0.05-0.6
Cr	50-150	1.0-3.1	2.6-7.4
Cu	10-60	0.2-1.2	0.5-3.0
Hg	5-20	0.1-0.4	0.2-1.0
Mn	50-100	1.0-2.1	2.5-5.0
Ni	10-50	0.2-1.0	0.5-2.5
Pb	80-100	1.7-2.1	4.0-5.0
Sn	5-20	0.1-0.4	0.2-1.0
Zn	50-150	1.0-3.1	2.5-7.4

[a] From Nriagu and Pacyna (1988).

[B] Interpolated values used for sludge incinerated in 1956, 1959; 1965, 1968, 1969.

Table 16

Estimate of the emission of metals to the atmosphere from the incineration of garbage and rubbish in Providence (see Fig. 18).

	Emission Factor,[a]	Average Emission,
Metal g t^{-1} waste	tons y^{-1} (1943-1971)	
Cd	0.4-10	0.02-0.50
Cr	0.7-7.0	0.03-0.35
Cu	7.0-14	0.35-0.70
Hg	1.0-15	0.05-0.75
Mn	1.8-9.0	0.09-0.50
Ni	0.7-3.0	0.03-0.15
Pb	10-20	0.50-1.0
Sn	1.0-10	0.05-0.50
Zn	20-60	1.0-3.0

[a] From Nriagu and Pacyna (1988).

Made some contribution to the metals deposited in the bay. The state's Clean Air Act of 1966 prohibited this practice, though only 10-15% of the towns had complied by the end of1967.[1] Most of the metals emitted from rural sites fell on soils and vegetation where they were probably retained with a high efficiency (Ragsdale and Berish 1988; K. Rahn, URI Center for Atmospheric Chemistry Studies, personal communication), though surprisingly little is known about the behavior of anthropogenic metals deposited on different land-scapes (Fortescue 1980). Overall, it seems likely that the input of metals into Narragansett Bay from waste incineration was small relative to the metals added directly to the bay in waste water effluents and sewage sludge.

The Burning of Fuels

The smelting and working of metals and the incineration of waste materials were not the only sources of metal emissions to the atmosphere. The release of certain metals during the burning of wood has already been mentioned, and while Indian campfires and colonial fireplaces were not numerous enough to be of any real consequence, the combustion of fuels in the rapidly growing cities of the nineteenth century was another matter.

In 1800, several decades before efficient, coal-burning metal stoves were manufactured in great numbers in Providence, the average New England household burned 30-40 cords of wood each year in open fireplaces.[2] With an average household of 5.7 persons (U. S. Bureau of the Census 1975), the 7600 people living in Providence at that time may have consumed some 40-50 thousand cords of fuel wood. The emission of metals from this source was approximately equal to those generated by garbage and rubbish incineration in the much larger Providence of 150 years later (compare Tables 17 and 16), but it seems likely that a lack of paved roads and other impermeable surfaces in the early city would have greatly reduced the washoff of any metals deposited from the atmosphere. By 1850, the average family consumption of firewood in America had declined to 18 cords per year (Melosi 1985), but a growing population in Providence may have increased the total firewood consumption to about 135,000 cords, with metal emissions perhaps three times those of 1800 (Table 17).

After about 1850, the potential role of fuel combustion in releasing metals to the atmosphere became much larger and more difficult to evaluate. It became larger because the growth of steam and, later, electric power produced an extraordinary increase in energy

---

[1] J. T. Kaull, "Dump Fires Still Pollute State's Air", Providence Journal Bulletin, 12/27/67, p. 1.

[2] Special exhibit, “Everyday Life in America 1780-1800”, National Museum of American History, Smithsonian Institution, Washington, DC , 1989.

Table 17

Estimated emission of metals to the atmosphere from firewood combustion in Providence about 1800.

	Emission Factor,[a]	Total
Emission,[b]		
Metal	mg kg^{-1}	tons y^{-1}
As	0.1-0.5	0.015-0.066
Cd	0.1-0.3(0.3)	0.015-0.039
Cr	trace	
Cu	1.0-2.0(19)	0.11-0.26(2.5)
Ni	1.0-3.0(5)	0.11-0.39(0.66)
Pb	2.0-5.0(7)	0.21-0.66(0.92)
Zn	2.0-10.0(117)	0.21-1.3(15)
Hg	0.1-0.5	0.015-0.066

[a] From Nriagu and Pacyna (1988). Values in ( ) are from Nriagu (1979). No explanation for the discrepancy was given.

[B] 40-50 x 10^{3} cords y^{-1} assuming 3.5 m^{3} per cord and a density of 0.75 g cm^{-3} gives 105-131 x 10^{3} tons of wood y^{-1}.

Consumption in the United States, much of which supported the great industrial expansion described earlier. While wood continued to make an important contribution to domestic heating, coal was the basis for manufacturing power. By 1885-1890, coal had become the major energy source in the United States (Melosi 1985) and it remained so until it was replaced by petroleum between 1950-1955 (Plate 12) (Fig. 19). Increasing amounts of the coal and oil were burned to produce electricity after Edison's success in the early 1900s (Fig. 20). While no data on the historical consumption of various energy sources appear to be available for the Narragansett Bay watershed, it may be reasonable to assume that per capita consumption rates from the country as a whole can be applied to this area. The purpose of making such an assumption is to allow the calculation of potential metal emissions to the atmosphere that may have been associated with the changing quantity and nature of energy consumption around the bay. Such a calculation can not provide anything more than a very rough approximation of the relative strength of this source of metals to the atmosphere, because the emission factors that have been published (for example, Nriagu and Pacyna 1988, Straub 1989) apply to recent technology and not to the releases of metals that accompanied earlier burning practices. While this may be a minor concern in assessing wood combustion in home fireplaces, it probably results in a serious underestimate of early metal emissions from coal burning and may be particularly important in considering the metals associated with larger soot and fly ash particles that would have been deposited from the atmosphere near combustion sites. In spite of this limitation, however, it is still helpful to develop at least a lower estimate of the potential importance of energy consumption in contributing to metal pollution. Since absolute fluxes can not be calculated, it is practical to limit the calculations to Providence so that the results can be compared with earlier estimates of metal emissions to the atmosphere from the incineration of sewage sludge, garbage, and rubbish as well as the discharge of metals directly to Narragansett Bay in sewage treatment plant sludge and effluents. Use of per capita energy consumption data from the entire country applied to the population of a northeastern industrial city like Providence probably overestimates the importance of wood fuel and underestimates the contribution of petroleum. Total energy consumption in Providence is probably also underestimated because of the city's above average industrial development. Declines in the population of the city after 1925 produced significant departures from the national trend in consumption (Table 18).

If each component of the changing mix of energy sources is multiplied by the appropriate range of emission factors for individual metals and the results summed by metal, it is possible to generate an historical trend in atmospheric metal emissions from fuel burning in the city (Table 19, Fig. 21). The combination of a declining population and a

Table 18

Estimated consumption of fuel wood, coal, and oil (including gasoline) in Providence, thousands of tons per year.[1]

	Fuel Wood	Coal	Petroleum	Total Energy,
	Joules y^{-1} x 10^{12}			
1850	212	14	0	4,449
1860	239	30	0.1	5,387
1880	334	155	4.8	11,029
1900	261	561	13	22,120
1915	218	1083	77	39,741
1925	198	1200	257	50,931
1930	167	991	302	46,093
1935	150	718	265	35,943
1940	146	849	352	43,690
1945	134	1066	463	54,862
1950	107	745	483	45,657
1955	80	562	530	41,777
1960	53	410	465	33,862
1970	21	398	467	33,103

[1] Computed from Historical Statistics of the United States, U. S. Bureau of the Census, Table of Calculated Consumption of Mineral Energy Fuels and Fuel Wood. Energy consumption converted to mass assuming 8.2 x 10^3 BTU lb^{-1} for wood, 12 x 10^3 and 13 x 10^3 BTU lb^{-1} for hard and soft coal, respectively, and 19 x 10^3 BTU lb^{-1} for petroleum. Total U.S. per capita consumption was multiplied by the Providence population for selected years.

Table 19

Estimated emission of various metals to the atmosphere from the combustion of fuel wood, coal, and oil (including gasoline) in Providence, tons per year.[1]

	Metal							
Year	As	Cd	Cr	Cu	Ni	Pb	Zn	Hg
1850	0.02-0.13	0.02-0.07	0.001-0.007	0.23-0.49	0.24-0.85	0.44-1.2	0.44-11	0.03-0.15
1860	0.03-0.18	0.03-0.09	0.003-0.015	0.28-0.63	0.30-1.2	0.51-1.5	0.53-20	0.04-0.21
1880	0.06-0.48	0.05-0.21	0.03-0.13	0.55-1.5	0.74-3.7	0.83-3.25	0.91-84	0.11-0.63
1900	0.14-1.2	0.08-0.36	0.07-0.34	1.1-3.4	1.6-10.2	1.1-7.0	1.4-286	0.31-1.8
1915	0.24-2.3	0.13-0.62	0.18-0.93	1.8-6.1	4.0-23	1.7-12	2.1-546	0.56-3.4
1925	0.27-2.6	0.15-0.71	0.38-1.1	2.0-7.2	7.7-39	7.8-20	2.5-606	0.62-3.7
1930	0.23-2.1	0.13-0.61	0.40-2.0	1.7-6.2	8.2-40	11-21	2.1-500	0.51-3.1
1935	0.16-1.6	0.10-0.45	0.33-1.7	1.3-4.7	6.9-32	11-18	1.7-364	0.37-2.2
1940	0.19-1.8	0.11-0.52	0.43-2.2	1.5-5.6	8.9-41	14-23	1.9-429	0.43-2.6
1945	0.23-2.3	0.15-0.66	0.57-2.8	1.8-7.0	11-53	13-25	2.3-539	0.54-3.3
1950	0.17-1.6	0.10-0.50	0.55-2.8	1.4-5.4	11-50	19-28	1.8-378	0.38-2.3
1955	0.13-1.3	0.09-0.41	0.59-2.9	1.1-4.6	12-51	21-28	1.5-286	0.29-1.7
1960	0.09-0.94	0.07-0.31	0.50-2.5	0.85-3.6	10-44	21-26	1.2-209	0.20-1.3
1970	0.09-0.90	0.07-0.29	0.51-2.5	0.80-3.4	10-43	24-29	1.1-203	0.19-1.2

[1] Fuel consumption from Table 18. Emission ranges from Nriagu and Pacyna (1988). Calculation of Pb emission from oil (including gasoline) is complicated because of the addition of lead to gasoline beginning in 1923. The petroleum category has been disaggregated here for 1925 through 1970 using U. S. per capita gasoline consumption, and motor fuel consumption was then multiplied by an emission factor of 97 mg kg^{-1} (Nriagu 1977). Non-motor fuel petroleum was multiplied by 2-6 mg Pb kg^{-1} (Nriagu and Pacyna 1988).

Shift in energy source from coal to oil produced significant declines in the potential emission of many metals after World War II. Nickel is an exception that is much more strongly emitted from oil burning than from coal, and the release of lead was strongly influenced by the growing consumption of leaded gasoline after 1923.

In general, it appears that fuel consumption and waste incineration were roughly comparable sources of chromium, copper, mercury, arsenic, and cadmium to the atmosphere over Providence during the decades when incineration was practiced, while nickel, lead, and zinc appear to have been much more strongly emitted by fuel combustion (Table 20). Atmospheric emissions of chromium and copper appear small relative to the discharge of these metals to Narragansett Bay from the Fields Point treatment plant, but the atmospheric emissions of nickel and cadmium may have been roughly comparable to those in waste water discharges and the emission to the atmosphere of lead, zinc, and mercury apparently exceeded the loading of these metals to the bay from Providence during various times. It should be emphasized again that the estimates of atmospheric emissions are low because they are based on current rather than historical emission factors, they do not include direct emissions from manufacturing processes, and they are biased by nationwide per capita energy consumption rates. The discharge of metals in wastewater is also under-estimated, especially in earlier years when many industries had not yet been connected to the treatment plant. The compositions of sewage sludge and effluent, like the atmospheric emission factors, were also based on recent analyses.

It is possible to gain some sense of the impact that changing technology and a growing concern with air pollution may have had on atmospheric emissions and metal deposition by reviewing some of the data on air quality in Providence and around Narragansett Bay. While recent concerns about air pollution have focused on acid rain and on gases such as ozone, carbon dioxide, nitrous oxide, and methane, the emphasis in earlier years was on smoke and the associated deposition of dust or soot. For example, the Smoke Prevention Association of America was organized in Chicago in 1907 (McCabe 1959). The particles of concern were usually between 10-100 µm in diameter (U. S. Public Health Service 1969) and settled from the atmosphere relatively near their points of emission compared to the much finer particles and gases that constitute present day air pollution problems.

As coal consumption increased in the late 1800s (Fig. 19, Table 18), dense smoke became an increasing nuisance and a public health issue. Providence appointed an Inspector of Smoke in 1904, and a stronger state law went into effect in 1913 that allowed only “light gray” smoke to be discharged for six minutes out of every hour, except between 5 and 7 a.m. when fires were being kindled (Providence Journal 2/4/13). It is not evident that this early legislation had much effect, however, since various later newspaper articles described the Providence Air Pollution Ordinance of 1947 as the first such regulation.[1]

Like numerous other cities, Providence began monitoring the amount of soot fall in 1930. Open "test jars" were placed on roofs at six locations around the city and the collected material weighed at regular intervals.[2] Unfortunately, most of these data have apparently been lost, but the surviving pieces still provide useful information and perspective. As expected, there was considerable variability among the monitoring sites (Table 21), and some tendency for deposition to be greater during winter (Fig. 22). It was the opinion of the Providence Public Service Engineer that the rate of soot fall declined significantly during the 1930s, but returned to high values during World War II when many industries and commercial consumers had to abandon oil and revert to soft coal as their energy source (Table 21, Mancini letter). By 1947, the Providence Department of Public Works reported that the dust fall data "show that the smoke situation in this city is greatly improving." In 1961, despite a Providence Journal editorial titled "Dustfall is Tremendous, and Expensive", the city-wide average deposition had declined even further, from 142 g m^{-2} y^{-1} in 1947 to about 85 g m^{-2} y^{-1}, and Providence could claim ". . . a world-wide reputation for keeping its skyline free of the thick, black smoke that hovers over many an industrial center."[3] Even allowing for an excess of pride in a local newspaper account, it is evident that soot conditions in Providence had improved and that the city enjoyed much cleaner air than some infamous "dirty" urban centers (Fig. 23). Much of this was due to a series of strong anti-smoke laws passed between 1949 and 1956, as well as to vigorous citizen support from the League of Women Voters and other groups of volunteer "smoke spotters". According to the Director of the Providence Division of Air Pollution, the emission of solids from the city into the atmosphere declined

---

[1] For example, S. O. Hale, "Air Pollution in City Decreases 100% in a Year, Engineer Reports", Providence Journal 3/16/47, p. 6. "The City of Providence, armed with an effective air pollution ordinance for the first time in its history, is shaking the soot from a thousand chimneys out of its hair with reasonable success." And L. M. Howard, "Providence Pioneers in Eliminating Smoke Pollution," Providence Sunday Journal 2/11/60, p. N62.

[2] According to a letter from Philip S. Mancini, Providence Public Service Engineer,
to B. Earl Clarke, M. D. , Providence Medical Association (23 May 1945, Providence Public Library Collection), the monitoring sites in Providence were the Biltmore Hotel,

the police station on Sessions Street, Roger Williams Park Casino, Brown University engineering building, the School for the Deaf, and Central High School.

[3] L. M. Howard, “Providence Pioneers in Eliminating Smoke Pollution”, Providence Sunday Journal, 12/11/60, p. N62.

Table 21

Soot Fall in Providence.[1]

	$g\ m^{-2}\ d^{-1}$		
	April 1930	April Mean, 1930-38	April 1945
Biltmore Hotel	2.78	0.83	5.46
Police Station	0.37	0.24	0.48
Brown University	0.64	0.29	1.42
Central High School	0.83	0.43	0.73
School for the Deaf	1.17	0.40	0.32
Roger Williams Park	0.32	0.26	0.29

[1] Letter from P. S. Mancini, Providence Public Service Engineer, to B. Earl Clarke, M.D., Providence Medical Association, 5/23/45. Providence Public Library collection. Data reported as tons $mi^{-2}\ y^{-1}$. Mr. Mancini suggested that the April, 1945 Biltmore Hotel data should be disregarded because the collection jar was too close to the chimney.

From about 0.7-0.9 g m^{-2} d^{-1} in 1948 to 0.4-0.5 g m^{-2} d^{-1} in 1960.[1] As a matter of interest, these emission rates were about twice the respective deposition rates each year.

The metal contents of the dust fall were never measured, but the greatest part of the material falling on the city was almost certainly fly ash from coal combustion. In 1958, the Chief of the Providence Division of Air Pollution noted that even hard coal sent over 60 kg of ash out of the smokestack for every ton burned if no control devices were used.[2] If the composition of the dust resembled average fly ash generated from western U. S. coal burned in a modern power plant, the deposition of metals associated with it would have been only a fraction of the total metal emissions calculated earlier (Table 22). The composition of modern fly ash varies widely, however, and the operation of particulate collectors on virtually all modern smoke stacks influences greatly the size and composition of the material emitted (National Research Council 1980, Smith 1987). Even the highest metal concentrations found in modern ash would have resulted in an atmospheric deposition on the city that was smaller than the estimated input of most metals to Narragansett Bay from the Fields Point sewage treatment plant (Table 22). Since only a fraction of the metals deposited on the city in soot would actually have entered the bay, and the sewage values are almost certainly low, the evidence suggests that the treatment plant was probably a much stronger source of metals to the bay than atmospheric deposition and urban runoff.

Beginning in the 1950s, there was increasing dissatisfaction with the admittedly crude dustfall collectors:

> There is no question that, within a given city, dustfall tends to increase with intensity of human activity . . . However, trying to extract detailed information from small fluctuations in dustfall appears to be an exercise in futility.
>
> U. S. Public Health Service (1969)

As a result, a more sophisticated national network of air quality sampling stations was established that used “high volume” (19-24 liters s^{-1}) samplers that drew air across a 20 x 25 cm glass fiber filter that was reported to capture virtually all particulates greater than 0.3 μm in diameter (U. S. Public Health Service 1962). After early trials that ended in 1957, the samplers were kept at various heights above the ground in vertically mounted shelters to limit the effect of wind. They were operated for 24 h every other week on a day selected at random for each week.

---

[1] L. H. Howard, “Pollution Cut in Half Since ‘48, Official Says”, Providence Journal, 12/12/60, p. 12.

[2] ”Providence Air Much Cleaner After Decade of Hard Work”, interview with

Genaro G. Constantino, Providence Journal, 10/19/58, p. N52.

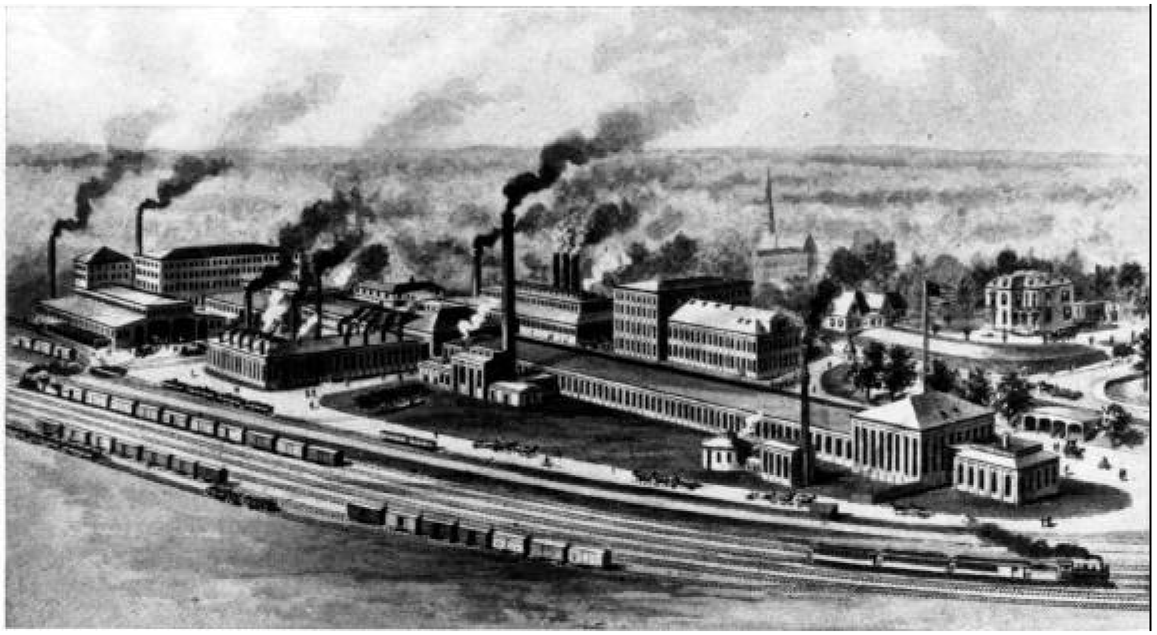


Plate 12 Heavy coal smoke discharged from the original Corliss Steam-Engine Works In Providence when it was under the ownership of the American and British Manufacturing Company. From *Rhode Island Industries Catalogued and Illustrated*, Providence, 1904.

Table 22

Comparison of the estimated metal deposition on Providence associated with fly ash or soot and the estimated emission of metals to the atmosphere over the city from fuel combustion during 1935. The estimated release of metals from the city sewage treatment plant is also shown.

	Fly Ash Composition,[a] μg g^{-1}	Atmospheric Deposition,[b] tons	Emission to the Atmosphere,[c] tons	Release in Sewage Effluent,[d] tons
As	9(2.8-6300)	0.06(42)	0.16-1.6	›0.3
Cd	1(0.7-130)	0.006(0.78)	0.10-0.45	1.5
Cr	54(10-690)	0.34(4.3)	0.33-1.7	18
Cu	63(14-1000)	0.40(6.3)	1.3 -4.7	87
Ni	34(10-4300)	0.22(28)	6.9 -32	60
Pb	48(7-279)	0.31(1.8)	11-18	18
Zn	72(36-1333)	0.46(8.5)	1.7 -364	142

[a] Average and range summarized by the National Research Council (1980).

[b] Assuming the mean soot fall for 1930-38 (Figs. 22 and 23) and a city area of 49 km^2 (Hoffman et al. 1983). Values were calculated using average and (maximum) of the range in composition.

[c] See Table 19, includes emissions from fuel wood, coal, and petroleum (including gasoline) for 1930-1940..

[d] Estimated discharge from the Fields Point Treatment plant (Table 20).

Providence and a number of other towns and cities in Rhode Island participated in the national network, and a few stations have remained in operation for many years. The data from these long-term stations show a decreasing trend that suggests that the concentrations of particulates in the air around Narragansett Bay may now be about one-half the values characteristic of the early 1960s (Fig. 24). Earlier data showing still higher concentrations of particulates in Providence were reported by the U. S. Public Health Service (1958, 1962), but the sampling stations were different:

	arithmetic mean, μg m^{-3}
1954	92
1955	153
1957	115

	geometric mean, μg m^{-3}
1957	112
1958	124
1959	90
1960	109
1961	74

Significant relative declines in particulate emissions around 1960 are also evident in the Providence Smoke Inspector's reports of the duration of smoke emission in excess of that allowed on the Ringelmann scale:[1]

1958 = 60 h 26 min
1959 = 53 h 53 min
1960 = 32 h 30 min
1961 = 36 h 39 min

Direct measurements of some of the metals associated with the particulates were also carried out in some years, and these data can be used to make a very rough calculation of the approximate deposition of metals from the atmosphere (Table 23). The results suggest fluxes of metals from the atmosphere in 1958 that are considerably higher than those calculated from the dustfall data during the 1930s (Table 22). While both estimates are obviously very uncertain, the apparent increases for copper, nickel, lead, and especially zinc, are not consistent with changes in the calculated local emissions of these metals to the atmosphere between 1935 and 1960 (Table 20). It seems likely that the use of average modern fly ash composition seriously underestimated the metal content of the earlier soot, though it is also possible that distant sources contributed to the high concentrations of metals in the air in Providence. The calculated metal depositions in 1958 are considerably lower than the estimated local emissions and, with the exception of lead, much lower than the estimated releases of metals from the Fields Point sewage treatment plant (Table 20). The calculated deposition of lead from the atmosphere in 1958 was 1.5-3 times greater than the estimated discharge from the treatment plant, though it would have been only some

---

[1] Interview with Genaro G. Costantino, Chief of the Providence Air Pollution

Control Unit, by C. M. Burbank, Providence Journal Bulletin, 6/5/62, p. 8

Table 23

Calculated deposition of some metals from the atmosphere over Providence based on concentration measurements in the air during 1958. The estimated emission of metals to the atmosphere from incineration and fuel combustion is also shown.

	Air Concentration,[a] μg m^{-3}	Calculated Wet Deposition, 1958[b] mg m^{-2}	Total Wet Deposition, 1958[c] tons	Emission to the Atmosphere,[d] tons
Cr	0.03	8-16	0.4-0.8	5
Cu	0.05	21-42	1.0-2.0	4
Ni	0.07	24-47	1.2-2.3	28
Pb	0.93	100-200	4.9-9.8	28
Zn	0.84	366-734	18-36	108

[a] Annual geometric mean of biweekly measurements at Providence Police Headquarters on Fountain Street at LaSalle Square, 13.7 m above the ground, U. S. Public Health Service (1962). Mean urban air concentrations for the U. S. between 1957-1961 were: Cr = 0.02, Cu = 0.04, Ni = 0.03, Pb = 0.6, Zn = 0.48 μg m^{-3}.

[b] Assumed scavenging ratios of 100-200 for Pb, 300-600 for Cr and Ni, and 400-800 for Cu and Zn (K. Rahn, U.R.I. Center for Atmospheric Chemistry Studies, personal communication); air density of 1.2 kg m^{-3}; 1958 rainfall of 131 cm (Pilson 1989). Dry deposition is very much smaller than wet under present conditions (K. Rahn, personal communication).

[c] City area = 49 km^2 (Hoffman et al. 1983). Using the long-term (1905-1987) mean Providence rainfall of 106 cm (Pilson 1989) would reduce the calculated deposition by about 20%.

[d] Mid-point of emission ranges for 1960, Tables 15, 16, 19 and 20 and Fig. 21 for assumptions and ranges.

25-50% of the discharge if sludge had still been released to the bay as it was in 1950 (Table 20). With the possible exception of lead, it still seems reasonable to conclude that the discharge of metals from sewage treatment plants to Narragansett Bay exceeded the probable input from atmospheric deposition and storm washoff.

Under present conditions, important sources of many of the metals deposited from the atmosphere onto Narragansett Bay lie far outside the watershed (K. Rahn, U.R. I. Center for Atmospheric Studies, personal communication), but during the earlier years when wood and coal burning were important, more metals may have been associated with the larger particulates that were emitted and deposited locally. The past importance of local sources is reflected in the variability of soot fall (Table 20) and in the contrast in particulate concentrations in the air at different sites around the bay (Fig. 24). As recently as 1940, coal combustion accounted for over 98% of the particulate emissions to the atmosphere (36% from industry, 35% from electric utilities, 27% from commercial and residential sources) while it provided only 42% of the energy consumed (Moore 1966). Fly ash collectors were about 75% effective in 1940 (Moore 1966), but the Narragansett Electric Company apparently did not install any fly ash arrestors until 1950, and their largest stack was not scheduled to have an ash arrestor in operation until 1954 (Providence Journal, 6/17/49, p. 1). At least as late as 1949, the standard procedure at the utility was to blow smoke and fly ash out of the stacks each night, "to get rid of waste particles and allow proper generation of steam." One consequence was local newspaper coverage such as "Scourge of Narragansett Electric's Smoke Stacks Covers Wide Area" that appeared on the front page of The Providence Journal on 17 June, 1949.

The historical importance of atmospheric emission, deposition, and washoff as a pathway by which metals entered Narragansett Bay probably increased dramatically for most metals between about 1900 and 1930, when the amount of impermeable surface was already significant and increasing rapidly (Table 6, Fig. 11), and coal combustion more than doubled in response to increases in population and industrial power demands (Table 18). Emissions of metals strongly associated with coal burning (Cu, Zn, As, Hg) peaked in the Providence area around 1925-1930. While this reflects the fact that the population of the city declined after 1925, it is also due to the increasing importance of petroleum in the United States as a whole after a general decline in total fuel consumption during the Depression (Fig. 19). Since stack emission controls were non-existent or very inefficient during the early 1900s, and larger particulates deposited in urban areas adjacent to the bay would have a greater probability of being washed into the bay than more recent and widely dispersed fine-particle pollutants, it seems likely that the period of maximum deposition and flux to the bay in storm water runoff was also around 1925-1930 for these metals. Arsenic and mercury are also released in association with sludge incineration, and this source was significant in Providence in the late 1950s (Fig. 17, Tables 15 and 20). The emissions of nickel and chromium continued to increase along with oil consumption, and the apparent decline after 1955 in Providence was due to the loss of population from the city. The total emission of nickel and chromium in the air shed of the bay probably continued to increase, at least until federal clean air legislation of the 1960s. Chromium was also emitted strongly during sludge incineration in the late 1950s (Fig. 17, Tables 15 and 20). It appears that the atmospheric emission and deposition of cadmium peaked with sludge incineration in 1960, since this was such a strong source from Providence (Table 21), but the overall trend in the watershed probably followed coal burning. The emission of lead was dominated after 1923 by the consumption of leaded gasoline, and the stability of emission from Providence (Table 21, Fig. 21) is due to the declining population after 1925. With the exception of a brief reversal during World War II, the emission of lead to the atmosphere around the bay must have increased continuously between 1923 and 1974, when unleaded fuel was required for all new automobiles.

Comparison of Reconstructed Inputs with the Sedimentary Record

Because many metals enter Narragansett Bay in particulate form or associate to some degree with particles after entering the bay, it is possible to learn something of the pollution history of the area by examining the concentrations of metals associated with sediments that were deposited on the bottom of the bay at various times in the past. Since most sediments appear to settle relatively quickly once they enter the bay, the concentrations of many pollutants in the sediments may be influenced strongly by local, as well as regional, sources (Bender et al. 1989).

In principle, particles lying at the sediment-water interface represent recent conditions and those buried at greater depths reflect conditions during progressively earlier times. In practice, however, sediments are eroded, resuspended, disturbed by human activities, and mixed by benthic animals, and it is often difficult to establish the age of the material at any given depth with a high degree of certainty (Santschi 1980; Bricker Urso et al. 1989). It is also possible that the geochemical behavior of particular metals may have an impact on the concentration profiles that develop within the sediment (Bender et al. 1989). For example, iron and manganese become soluble under reducing conditions, while lead is more soluble in oxidized sediment.

In spite of these difficulties, a pollution history derived from analyses of sediment cores rests on measurements of materials actually taken from the bay. For this reason, it offers direct evidence of a kind seldom available in an historical assessment of anthropogenic activities and metal inputs. Since Narragansett Bay has been the focus of several investigations that attempted to develop the history of metal inputs based on analyses of the sediments, we have a rare opportunity to compare and combine the two approaches. Unfortunately, the most extensive studies of metal pollution in subtidal and intertidal sediments around the bay are now only in the final stages of analysis and interpretation (John King, Graduate School of Oceanography, URI and Suzanne Bricker, Graduate School of Oceanography, URI, respectively, personal communication), and it would be inappropriate to present more than a few general observations from this work before the authors have a chance to publish their own detailed conclusions.

Subtidal Sediments

The first historical profiles of metals in the sediments of Narragansett Bay were derived from a core collected in 1974 in the upper bay, south of Rumstick Neck, under about 7 m of water[1] (Goldberg et al. 1977). Several features of the profiles are particularly interesting (Fig. 25). First, manganese shows a relatively uniform distribution with depth (age) that reflects the fact that natural weathering processes in the watershed, rather than pollution, are the major source of this metal (Graham et al. 1976). It may also be important that manganese is remobilized in the reducing sediments that lie more than a few cm below the sediment-water interface, with the result that there is a large flux of this metal in dissolved form to the overlying water during summer (Nixon et al. 1989). The monotonous background concentration may simply represent that fraction of the total manganese not lost during diagenesis.

Second, except for nickel, the pollutant metals Goldberg et al. (1977) measured show a sharp increase beginning at about 35-40 cm depth. According to the ^{210}Pb-based chronology developed for this core by Goldberg and his coworkers, the sediments found at 35-37 cm were deposited between about 1887-1899, a time corresponding remarkably well with the increasing discharge of untreated sewage at Fields Point that began in 1892. The plateaus in zinc and copper concentrations and the declines in lead and chromium concentrations above about 26-27 cm also correspond well with marked reductions in the calculated discharges of these metals at Fields Point after 1950 (Fig. 26), the date assigned to this layer by Goldberg et al. (1977). The calculated reduction was most dramatic for lead and the observed decline in the concentration of this metal was also the most pronounced.

---

[1] Located at 41° 41.4'N, 71° 18.7'W (Bender et al. 1989).

It is not clear why the concentrations of pollutant metals increase again above about 18 cm, a depth corresponding roughly with the mid 1950s, or why the concentration of lead would not have continued to increase with the consumption of leaded gasoline until 1974. Bender et al. (1989) have interpreted the essentially uniform concentrations of these metals between about 10-15 cm and the surface as being due to bioturbation, but there may be more information in the profiles. Since bioturbation was presumably also operating when the deeper sediments reflecting large changes in metal concentrations were laid down, it seems at least equally reasonable to conclude that the flux of most metals to the bottom at this site remained approximately constant between about 1960 and 1974 (Fig. 25).

Nickel is an interesting exception in almost every respect (Fig. 25). The concentration of this metal in the sediments shows essentially no response to the introduction of sewage discharge at Fields Point, in spite of a significant calculated input (Fig. 26). This discrepancy is a useful reminder that while the amounts of sewage effluent and sludge were either measured or estimated from historical data, the calculated fluxes of the metals they contained were necessarily based on modern analyses (Tables 8 and 11). While the United States began using nickel in some coins beginning in 1865, large scale smelting and refining of nickel ores only became feasible after 1890 (Ware 1965). Until about 1940, relatively little nickel was used in the United States compared to many other metals that were exploited intensively beginning in the late 1800s and early 1900s (Fig. 27), and it seems likely that sewage effluents and sludges before World War II contained far less nickel than recent samples would suggest. As a result, the calculated discharge of nickel at Fields Point and the addition of nickel in sludge dumped below Prudence Island were probably overestimated for the years before 1940 (Tables 8, 9, 10, and 12, Fig. 26).

The approximately 50% increase in nickel above about 18 cm (1955 according to Goldberg et al. 1977) is probably evident because improved sludge removal at Fields Point and incineration of the sludge after 1950 would have had relatively little effect on the total amount of nickel released from the plant compared to many other metals (Fig. 26). Nickel consumption also remained high (Fig. 27), and atmospheric emissions rose throughout the watershed as oil replaced coal as the major primary fuel (Fig. 21). That the increase in nickel concentration was not obscured by biomixing lends support to the earlier conclusion that the relative constancy of lead, copper, chromium, and zinc fluxes during this period may have been real.

Despite the apparent agreement between the major features of the Goldberg et al. (1977) core and the calculated release of metals at Fields Point, some inconsistencies are also challenging. For example, once the sediment metal concentrations began increasing rapidly at about 35 cm, they continued to do so until they truncated sharply some 60 years later at 27-28 cm (Fig. 25). The calculated metal loadings, however, increased rapidly during 1892-1901, fell sharply for about 7 years, increased rapidly once again over the next decade, then rose only modestly over the next 30 years (Fig. 26). With the sedimentation rate reported by Goldberg et al. (1977), the seven year hiatus in sludge release would have impacted a layer of sediment only 1.2 cm thick. Such a thin layer could easily be missed or obscured in taking subsamples for metals analyses or by bioturbation. It is not clear if bioturbation alone could be responsible for other differences between the shape of the calculated metal loadings curve and the concentration profiles in the sediments.

Santschi (1980) criticized the chronology established by Goldberg et al. (1977) because they had not accounted for the influence of bioturbation on the distribution of ^{210}Pb. The dating of the core was further complicated by a shell layer between 15 and 26 cm that Goldberg et al. (1977) attributed to a hurricane that passed over the bay in 1954. In his reevaluation of the dating of the core, Santschi (1980) associated the shell layer with another hurricane in 1893. The strongest hurricanes that struck Narragansett Bay, however, occurred in 1815 and 1938. Moreover, it is at least equally probable that the shell layer was deposited as part of oyster culture operations that leased much of the bottom in this part of the bay (Olsen et al. 1984). While the yield of oysters from the bay peaked strongly in 1912, the industry was active from the mid 1800s until about 1940, and it is impossible to know when or how often during that time shell might have been placed on the bottom (Kochiss 1974). After considering Santschi's (1980) arguments, Goldberg and Koide (1980) concluded that their original dating was still preferable, while admitting that ". . . the data can be manipulated to satisfy other chronologies."

Partly in response to the uncertainties and complications of the Goldberg core, Santschi et al. (1984) collected and analyzed an additional core from nearby Ohio Ledge in the upper bay.[1] Their core, collected in 1980, was dated using a more elaborate procedure that attempted to model the effects of bioturbation on the distributions of ^{234}Th and ^{239,240}Pu, as well as ^{210}Pb.

The Santschi core penetrated to a greater depth in the sediment than the Goldberg core and revealed sharp decreases in the concentrations of many of the metals below about 90 cm (Fig. 28). This discontinuity may reflect an abrupt change in sediment type or grain size, but no description of the core was provided in the published report. With the exception of manganese, which is much lower at Ohio Ledge, the concentrations of metals are similar in the top 25-30 cm of the two cores (Fig. 29). Below about 40 cm, however,

---

[1] Located at 41° 40.7'N, 71° 20.5'W (Bender et al. 1989).

the sediments from Ohio Ledge show increases in copper, zinc, chromium, and especially lead, that were not evident in the Goldberg core (Figs. 28 and 29).

Using the sedimentation rate of 0.5 cm y^{-1} calculated by Santschi et al. (1984), the marked increases in copper and zinc above about 35-40 cm at Ohio Ledge occurred in 1900-1910. Given the various uncertainties in estimating sedimentation rates, this is probably not significantly different from the 1890s date that Goldberg et al. (1977) assigned to the increase in metal concentrations in their core. In both cases, the increases were probably brought about by the discharge of sewage at Fields Points beginning in 1892 (Figs. 13 and 26). As in the Goldberg core, increases in nickel in the sediments at Ohio Ledge were more recent than for the other pollutant metals that were measured and date from around 1940.

The profile of lead concentrations is particularly interesting at Ohio Ledge because of the steep increase between about 75 and 65 cm depth (1830 to 1850), a prolonged decline from about 60 cm to 35 cm (1860 to 1910), and the lack of a clear trend above about 30 cm (1920 to 1980). Santschi et al. (1984) speculated that the early enrichment of lead and, to a lesser degree, copper and other metals, might reflect inputs from the Taunton Copper Company smelter that operated on the upper East Passage of Narragansett Bay between 1866 and the early 1880s (Garmon 1978). However, based on calculations of the relative strength of the smelter and the sewage discharged at Fields Point as sources of copper and lead, it is difficult to accept this explanation. While the smelter may have released 2-3 tons of copper and 1-2 tons of lead to the atmosphere each year while it operated (Table 3), some 40-70 tons of copper and 10-15 tons of lead were probably discharged directly into the Providence River at Fields Point each year between 1890 and 1910 (Fig. 26). Yet the sediments deposited at Ohio Ledge between about 1850 and 1870 contain more copper and lead than do those deposited around 1900 (Fig. 28).

An alternative explanation is that the high metal concentrations at depth may be associated with sediments dredged from the more polluted Providence or Seekonk Rivers and dumped in the upper bay sometime during the 1830s-1850s. Dredging in the Providence River by the U. S. Army Corps of Engineers was authorized at least as early as 1852 (USACE 1977), and it is clear from early navigational charts that the City of Providence had begun an active dredging program much earlier. While Ohio Ledge has not been used as an official dumpsite in recent years (Seavey and Pratt 1979), it is possible that early projects simply carried dredged materials below Conimicut Point or that "short-dumping" may have taken place. The presence of dredged material could have been indicated by ^{210}Pb measurements, but no samples were analyzed from the Ohio Ledge core in the questionable layer between about 50 and 75 cm (Santschi et al. 1984).

The lack of significant nickel enrichment in the deeper sediment is consistent with dredge spoil disposal, since this metal would not have been elevated in the more polluted harbor sediments during the first half of the 1800s (Fig. 27). Mixing of the dredged spoil with relatively cleaner upper bay sediments that settled on top of the spoil might account for the gradual reduction in metal concentrations between about 55 and 35 cm (Fig. 28).

Regardless of whether or not the dredge spoil hypothesis is correct, it is surprising that the Santschi core shows almost no trend in lead concentrations above 30 cm depth or the 1920s. Lead was added to gasoline beginning in 1923, and between that time and 1974, when federal legislation required unleaded gasoline for new automobiles, the calculated emission of lead to the atmosphere from the R. I. automobile fleet alone increased from about 100 tons to almost 1000 tons y^{-1} (S. Bricker, personal communication). My own estimate of lead emissions to the atmosphere from combustion in the City of Providence increased 3-4 fold between 1920 and 1960 (Fig. 21). And the estimated release of lead from the Fields Point sewage treatment plant decreased sharply around 1950 (Fig. 26). In addition, the East Providence and Blackstone Valley sewage treatment plants began operation in the early 1950s. It seems remarkable that these large changes in the potential input of lead to the bay are not reflected in the Ohio Ledge core. Perhaps an increasing input of lead from urban storm water runoff and a marked decline in input from sewage treatment plants left such an imprint in the sediments that bioturbation could provide the appearance of relatively constant loading during recent decades.

An important factor that may contribute to the difficulty of resolving the sedimentary record in the upper bay is the low sedimentation rate characteristic of the area (J. King, Graduate School of Oceanography, URI, personal communication). Fortunately, the results of metals analyses in sediments from an area of rapid accumulation in the Seekonk River have recently become available (Corbin 1989). These data show dramatic increases in copper, lead, and silver that began sometime after the early 1860s and terminated abruptly during the late 1880s to early 1890s (Fig. 30). This pattern is consistent with the development of the Providence sewer system beginning in 1871 that collected effluents from throughout the city and released them without treatment at numerous locations in the Seekonk and upper Providence Rivers (Fig. 12). In 1892, interceptor sewers began to collect those effluents and carry increasing volumes south for discharge below the city at Fields Point (Fig. 13). Continued industrial development upstream along the Blackstone River apparently balanced this local sewage diversion, however, with the result that the loadings of lead, copper, and silver to the Seekonk River may have remained relatively constant until the 1920s or 1930s.

The increase in silver in the sediments between about 200 and 175 cm also corresponds well with the sharp increase in silver production in the United States after 1860 (Fig. 27). Before the discovery of the Comstock Load in Nevada in 1859, the production of silver in the United States was insignificant, but by 1871 this country had become the major world producer, and it remained so until 1900 (Ryan 1965).

In contrast to the other pollutant metals, chromium concentrations appear to have increased early during the first half of the 1800s (Fig. 30). This may reflect the importance of this metal as a dye and mordant in textile manufacturing along the Blackstone River (Whittaker 1926). While chromium is best known today as a decorative plating, its primary use until the early 1900s was in the manufacture of chemicals and pigments. Chromite deposits discovered in Maryland and Pennsylvania around 1827 were the major world source of this metal until Turkish ores were discovered in 1848. These deposits could have provided easily accessible raw material for the rapidly expanding cotton and woolen mills in Rhode Island and Massachusetts (Table 2). The first successful plant to produce potassium bichromate or "chrome yellow" on a large scale for American textile mills and pigment manufacturers went into operation in 1845 (Holliday 1965).

The long decline in chromium concentrations above about 110 cm or 1920 appears to mirror the fate of the New England textile industry as it lost ground to the southern states. The number of active spindles in New England fell dramatically and continuously after 1920 (Dunwell 1978), and "Between the end of the First World War and the outbreak of the Second, over a fifth of the Rhode Island textile workers lost their jobs . . . " (Coleman 1969).

At the other extreme, cadmium concentrations in the Seekonk River sediments are markedly enriched only above about 100 cm, or the mid 1920s (Fig. 30). This is not consistent with the calculated loadings of cadmium in sewage effluents and sludge (Fig. 26), or the appearance of the other pollutant metals in earlier (deeper) sediments. As with nickel, an explanation may lie in the development of the technology involved with this metal. For 60 years after its discovery in 1817, the only major use of cadmium was in sulfide form as a paint pigment. In the late 1800s, new uses in plating steel, in low melting alloys, and in chemical reagents were developed, but consumption was small.

Only after 1919, when M. J. Udy developed a successful electroplating process, did demand accelerate (Schroeder 1965). Large amounts of cadmium were used during World War II in the manufacture of bearings designed to carry heavy loads, though that application declined sharply after the war. Once again, the evidence from the sediments suggests that the application of metal analyses from recent sewage effluents and sludges led to an overestimate of cadmium fluxes to the Providence River and the bay before the 1920s (Tables 8, 9, 10, and 12, Fig. 26).

In contrast to the cores collected from the upper bay, Seekonk River profiles show dramatic declines in the concentrations of copper and lead in the near-surface sediments (Fig. 30). It may be that these trends are evident because of the higher sedimentation rate in the Seekonk (over 1 cm y^{-1}, Corbin 1989) and, perhaps, because of less vigorous mixing of the sediments by benthic infauna (J. King, personal communication). The decline in copper began during the early 1960s and appears to predate the achievement in 1973-74 of full secondary treatment at the Blackstone Valley District Commission sewage treatment plant. Since this plant discharges directly into the Seekonk River, it may exert a strong influence on pollution levels in the river even though it releases less than one half the flow discharged at Fields Point. In any case, there is no reason to believe that there were changes in the operation of the treatment plant at Fields Point that would have reduced the discharge of copper or lead in 1960 or 1970, respectively.

A possible explanation for the reduction in copper and lead pollution during this period may be a decline in manufacturing involving base metals. Expressed in constant dollars, the value added by primary metals manufacturing and machinery manufacturing declined steeply in Rhode Island after the mid 1960s. (Fig. 10). Presumably, much of the base metals industry along the Blackstone River experienced similar declines that would have reduced the amounts of copper and lead carried down the river to the Seekonk. This interpretation is supported by the observation that the concentration of silver in the sediments of the Seekonk did not decline along with copper and lead (Fig. 30), and the precious metals industry in Rhode Island remained vigorous during most of the 1960s and 1970s (Fig. 10).

Manufacturing in the Providence area releases much more copper than lead in its wastewater (Tables 7 and 11), and the reduction of lead in gasoline after 1974 may have played a dominant role in bringing about the more recent decline in lead concentrations in the sediments compared to copper. Improved wastewater treatment at the Blackstone Valley plant after 1973-74 may also have contributed to a reduction in the discharge of metals associated with particles.

Other cores in addition to the three discussed here have also been collected from the bottom of Narragansett Bay and analyzed for metals (Goldberg et al. 1977, Santschi et al. 1984, Bender et al. 1989). While these data have proven useful in describing variations in the concentrations of metals in different parts of the bay, they add little, if anything, to the analysis of historical loadings. A very much expanded treatment of the cores recently analyzed by Corbin (1989) and others as part of the Narragansett Bay Project will doubtless add much to the preliminary assessment developed here.

Intertidal Saltmarsh Sediments

Before leaving the subject of sediment metal profiles, however, another set of cores ought to be mentioned. In contrast to the subtidal sediments analyzed in all of the other studies, Suzanne Bricker collected a series of cores from five intertidal salt marshes from the Seekonk River to the lower bay (Bricker Urso et al. 1989; Bricker 1990). While the marshes are thought to offer the advantage of less mixing and disturbance of the sediment once it is in place, they also provide an additional complexity in that metals are deposited on the marsh surface directly from the atmosphere as well as in association with the sediments deposited from the water when the marsh is inundated during high tides and storms (McCaffrey and Thomson 1980). It may also be the case that the profiles of metals that develop in the intertidal sediments are more strongly influenced by the geochemical behavior of the various metals than are those characteristic of the sediments on the floor of the bay (Bender et al. 1989).

The profiles of metals differ somewhat in character as well as in concentration among the marsh sites (Bricker Urso et al. 1989), but a consideration of one core from Conimicut Point, at the mouth of the Providence River, can give some sense of the additional perspective that the Bricker cores provide as well as the difficulties they present. While the ^{210}Pb-derived accretion rate at the Conimicut Point marsh (0.47 cm y^{-1}) is similar to sedimentation rates on the bottom of the upper bay (Santschi et al. 1984), increases in the concentrations of pollutant metals in the marsh peat are much less abrupt than in the subtidal muds (Fig. 31). They also begin earlier. For example, lead concentrations increase continuously from the early 1800s, copper concentrations from about 1865 (approximately 55 cm), chromium from about 1845 (approximately 65 cm), and zinc from about 1855 (approximately 60 cm).

These distributions do not seem consistent with the development of the Providence sewer system and the calculated abrupt changes in the discharge of metals at Fields Point beginning in the early 1890s (Figs. 12, 13, 26). Perhaps the early enrichment in the marsh was caused primarily by increases in the deposition of metals from the atmosphere. The calculated emissions of lead, copper, and especially zinc, to the atmosphere over Providence increased significantly long before sewage effluents were released at Fields Point (Fig. 21).

There is also a suggestion in the profiles that the flux of metals to the marsh increased with the initiation of sewage discharge at Fields Point in 1892, a date corresponding to a depth of about 43 cm (Fig. 31). While a change in the rate of increase in concentration is evident at about this depth for lead, copper, chromium, and silver, it is most dramatic for zinc. This is consistent with modern analyses that have shown much higher concentrations of zinc than other metals in the effluent and sludge from Fields Point (Tables 8 and 11).

While I have not been able to find data on the emission of silver to the atmosphere, it seems reasonable to asume that it must have been very small relative to that of the more common metals. This is consistent with the lack of excess or pollutant silver in the marsh sediments below about 40 cm and the hypothesis that the atmosphere was the major pathway by which lead, copper and zinc reached the marsh before construction of the Fields Point outfall (Fig. 31). Silver was enriched in the sediments of the Seekonk River long before the early 1890s (Fig. 30), but it appears that little of this precious metal reached Narragansett Bay until the wastewater from Providence was carried south for discharge at Fields Point.

The Conimicut Point marsh may also have captured a record of other important changes. The long decline in chromium concentrations beginning in the early 1930s is similar to that seen a decade earlier in the Seekonk sediments, and may also reflect the loss of textile manufacturing in Rhode Island. Lead concentrations increased markedly after the mid 1920s (about 26 cm) as the consumption of leaded gasoline grew after 1923 (see Fig. 8). It is tempting to attribute the sharp drop in lead concentrations above about 18 or 19 cm (1942-1945) to gasoline rationing during World War II (see Fig. 8), but the decrease seems far too large and persistent, as well as too late. Moreover, there are also declines in copper and zinc at about the same time that can not be associated with gasoline consumption (Fig. 31). It is more likely that these decreases were due to the installation of new sludge presses at the Fields Point treatment plant after the war. As noted earlier, the largest calculated reduction from the treatment plant at this time was for lead, and this is consistent with the marsh record (see Fig. 26). The fact that silver concentrations continued to increase after the war may be due to vigorous growth of the jewelry industry in the late 1940s and the 1950s (Bride 1959, Fig. 9).

The History of Metal Inputs to Narragansett Bay—A Summary

> A romantic philosopher once said that the Historian is the Prophet in Reverse. When we become historians, we are seduced by the prophet's temptations—to pretend to be wiser than we really are, and to underestimate the probability of the unexpected . . . Our past is only a little less uncertain than our future, and, like the future, it is always changing, always revealing and concealing.
>
> Daniel Boorstin(1987)
> *Hidden History*

The purpose of this historical assessment has been to provide a perspective for considering the more quantitative information on recent metal inputs to the bay that will be presented in a second report (Section II). There is a disconcerting tendency in human nature to assume that problems begin at precisely the moment we first become aware of them, but it is clear that human activities involving metals around Narragansett Bay have been of sufficient scale to pollute the environment since at least the early 1800s. There is good reason to believe that the release of copper, lead, zinc, and probably other base metals to the Seekonk and upper Providence Rivers was already significant by 1850 and that it increased markedly during and following the Civil War. Chromium, a metal used directly in textile manufacturing, appears to have preceded other metals as an early pollutant in the Seekonk River. The technology for dealing with cadmium and nickel developed slowly, and these metals appear to have entered the bay in large quantities only since the 1920s and 1940s, respectively. Of the precious metals, only silver has been measured in the bay, and the evidence suggests that the input of this metal was not significant until after the discovery of large silver deposits in the western United States.

Prior to the development of a city sewer system beginning in 1870, it seems reasonable to assume that untreated industrial effluents entered the urban portions of the rivers and streams individually, through numerous small discharge pipes. It is possible that a significant fraction of the metals they carried accumulated in the sediments behind dams, in ponds and coves, and in stream beds and lower salinity reaches of the tributary estuaries that entered the Seekonk and Providence Rivers. After 1870, the capture of effluents from increasing numbers of individual manufacturing establishments in the sewer system and the discharge of ever larger volumes of effluent without any treatment at a smaller number of locations around Providence Harbor probably resulted in much greater fluxes of metals directly into the Seekonk and upper Providence Rivers. Contemporary descriptions obviously did not contain data on metals, but they make it clear that there was severe pollution in the upper Providence River evidenced by discoloration of the water, odors, fish kills, and debris.

The construction of still larger interceptor sewers was intended to relieve these problems by carrying all of the effluents further down the Providence River for discharge at Fields Point. After late 1892, and especially after 1897, the increasing diversion of the city's sewage to Fields Point decreased local metal inputs to the upper Providence and Seekonk Rivers, though with corresponding increases in the fluxes to the lower Providence River and upper Narragansett Bay. However, the fact that metals in the sediments of the Seekonk River remained relatively constant may indicate that the diversion of metals from Providence was balanced by increasing inputs of metals from upstream sources along the Blackstone River.

The introduction of sludge removal at Fields Point in 1901 must have reduced dramatically the input of virtually all metals from this source, and the practice from 1901-1936 of releasing effluent only during ebb tides further reduced metal loadings to the upper Providence and Seekonk Rivers from the treatment plant. With the introduction of sewage sludge disposal south of Prudence Island in 1908, large quantities of metals were added directly to Narragansett Bay proper for the first time. The fates of the sludge and the various metals it presumably contained are not known, but it seems certain that there must have been some redistribution around the bay during the 41 years that the practice continued, and in the years since. Declining efficiency of the sludge removal process at the treatment plant also released increasing amounts of what was almost certainly metal-rich sludge directly into the Providence River between about 1910 and 1950. Except during the first years of treatment plant operation, it seems likely that more sludge-type material was released into the river than was dumped at the official site in the bay. The installation of new sludge collection equipment at Fields Point in late 1949 and substitution the same year of sludge incineration for dumping below Prudence Island must have greatly reduced the input of metals to the Providence River.

It is probable that the late 1940s and early 1950s were the years of maximum base metal discharges from Fields Point. There has been no trend in the volume of effluent released from the treatment plant since the early 1950s, and it seems unlikely that metal concentrations in the effluent increased sufficiently to offset the effective removal of metals in sludge after 1950. The value added by manufacture in primary metals industries in Providence increased until the mid 1960s, however, and it is possible that this activity was reflected in at least somewhat higher metal concentrations in the sewage. Expansion of the precious metals industries, especially costume jewelry, appears to have increased the input of silver to the upper bay after World War II. Two other treatment plants discharging to the Seekonk and Providence Rivers began operation in the early 1950s, but their combined input was less than that at Fields Point, and they used land disposal for sludge. It is reasonable to assume that they further reduced the total metal loadings to the rivers. Changes in the loadings of metals from the treatment plants during more recent years will be discussed in the next section.

The historical role of atmospheric deposition of metals as an input to Narragansett Bay is even more difficult to evaluate because of the great variety of sources of emissions to the atmosphere, great uncertainties in applying emission factors derived from modern factories and power generation facilities to antique technologies, uncertainties regarding the relative contributions at different times of local and far-field sources of emissions, and a general lack of information regarding the behavior and fate of metals once they are deposited on different land use types and at various distances from the bay. This assessment has assumed that the greatest exchange of metals from land to the atmosphere and back to the land took place in urban industrial areas like Providence, and that local sources were more important than distant ones, at least during the years before 1950-1960, when the emission of larger particulates was a major pollution problem. Calculations of the potential metal emissions from fuel combustion and the incineration of refuse and sewage sludge using upper estimates of recent emission factors suggest that the fluxes of nickel, zinc, lead, mercury, and, perhaps, arsenic and cadmium into the atmosphere from Providence may have been comparable to, or larger than, the estimated release of these metals from the city in sewage effluent and sludge. For copper and chromium, atmospheric emissions were probably minor, even during periods of intensive coal burning.

Evidence from intertidal sediments at the mouth of the Providence River suggests that the deposition of many base metals from the atmosphere increased measurably during the first half of the 1800s. Before the construction of the sewage outfall at Fields Point, deposition from the atmosphere was probably the major pathway of entry for metals into Narragansett Bay proper, though wastewater outfalls in the upper Providence and Seekonk Rivers were more important in these areas. Metals not emitted in large quantities to the atmosphere, such as silver, were probably not added in any significant quantity to the waters below Conimicut Point until after the opening of Fields Point.

If we accept that atmospheric deposition and urban runoff were probably not major sources of copper, chromium, or silver entering Narragansett Bay, the problem remains to speculate on the fate of the potentially large amounts of the other metals emitted to the atmosphere. No historical metal deposition measurements exist, but if soot falling on the city in the 1930s contained the same amounts of metals as average modern fly ash, metal deposition was only 0.1% to 20% of the maximum emission, depending on the element. If the highest concentrations of metals found in modern fly ash are applied to the soot-fall data, metal depositions would have exceeded the calculated local emissions for all metals examined except lead and zinc.

Measurements of the concentrations of some particulate metals in the air in Providence in 1958 make it possible to arrive at very rough estimates of the deposition rates at that time. For chromium, copper, and nickel, the fluxes calculated using these data are 1.2-10 times greater than those derived from the 1930s soot fall and average fly ash com-position, but still much lower than estimated using the extreme upper concentrations of these elements in fly ash. For lead and zinc, the deposition calculated from the air concentration measurements is very much higher (16-32 and 39-78 fold, respectively) than estimated from the soot fall and both the average and the extreme upper values for metals in fly ash. Even for these elements, however, the estimated release in sewage effluent and sludge during the 1950s is greater than the calculated atmospheric deposition. Comparison of the estimated metals loading from Fields Point in 1950 and 1960 with the calculated deposition from the atmosphere in 1958 suggests the following:

	Atmospheric Deposition as a Percent of Fields Point Treatment Plant Discharge	
	1950	1960
Cr	2-4%	5-10%
Cu	1-2	2-4
Ni	1.7-3.2	2.4-4.7
Pb	22-44	163-326
Zn	6-12	22-42

The potential relative contribution from atmospheric deposition increased after 1950 because of the operation of new sludge harvesting equipment and the substitution of sludge incineration for dumping in the bay.

Since only a fraction of the metals actually deposited from the atmosphere would have been carried into Narragansett Bay, it seems likely that the history of metal inputs to the bay since 1892 has largely been determined by factors influencing industrial waste water generation, disposal and treatment, rather than by atmospheric emissions, deposition, and urban runoff. Lead is probably an exception, since it was so strongly emitted from gasoline combustion, a source not confined to Providence and other urban centers, and lead emissions continued to increase until 1975. This is not to say that the Fields Point treatment plant has always been the largest source of all the other metals added to the bay. There are no historical data on earlier direct industrial discharges of metals, and the historical fluxes of metals entering Narragansett Bay from the

Blackstone, Pawtuxet, Taunton, and other rivers are unknown. Without these data or even a basis to arrive at crude estimates for these terms, it is impossible to develop metals budgets for earlier years. However, since the metals carried by the rivers would have entered from urban industrial areas similar in many respects to Providence, it seems likely that the changing burden of metals in the fresh water drainage from the watershed would have been the product of the same kinds of sources we have examined in detail for Providence.

This conclusion does not agree completely with the extensive recent study of metal inputs to Narragansett Bay that reported that urban runoff accounted for 5% of the copper, 56% of the zinc, and 65% of the lead entering the bay in 1982 (Hoffman and Quinn 1984). While the conclusions regarding copper and lead are consistent with the historical assessment, it is difficult to find a local atmospheric source large enough to account for the zinc found in the study of urban runoff. One explanation is that most of the zinc may have come from crankcase oil leaked onto highways, rather than from atmospheric deposition. Hoffman and Quinn (1984) calculated that this source could have provided virtually all of the zinc they measured. If so, the modern evidence is consistent with the historical, though it must be admitted that both rest heavily on numbers that are too loosely constrained and on assumptions that are only a little less uncertain than the future.

Metals are not obvious pollutants, and the techniques necessary for obtaining reliable measurements of their concentrations in coastal waters have only been available for the past 15-20 years. During the first 150 years or more that pollutant metals were accumulating in Narragansett Bay and its tributaries there was little awareness that these byproducts of industrial growth and social prosperity were entering the bay or that they might come to pose a threat to the organisms living there. Until very recently, the impacts of pollution control measures on the input of metals to the bay have been inadvertent or accidental consequences of attempts to alleviate other forms of pollution, including smoke, organic loading, and bacterial contamination. Specific attempts to reduce the emission of at least some metals to the atmosphere began in the 1950s and 1960s, but legislative requirements for industrial pretreatment of wastewaters to reduce metal emissions did not come until the 1980s.

The history of metal pollution in Narragansett Bay has been the product of circumstances large and small, of the geology of the watershed and the genius of individuals. And, like any history, it has proven to be full of surprises and unexpected connections. If the glaciers had not left the gradient of the Blackstone River so steep that it yielded abundant water power near the coast, the capital accumulated in Moses

Brown's maritime shipping ventures and the secrets of English spinning technology carried in Samuel Slater's memory might never have come together at Pawtucket Falls. And without the genius of David Wilkinson, the metal machinery needed to move from Slater's memory to the realization of an American textile industry might have been forged on some other river flowing to some other bay. Without such links from ice to iron, our history and our sediments would tell a very different story.

There have been times during this reconstruction when the methodical records of long-dead City Engineers, the results of modern chemical analyses, and the development of the technology involving a particular metal have come together with what is known of local industry and economic conditions to suggest a trend in the input of a metal that is so consistent with the distributions of that metal at various depths in the sediments of the bay that it seems we must be very close to an understanding of at least part of the history of metal inputs. But Daniel Boorstin's careful words given at the start of this section leave a haunting message. Testing various kinds of information against each other has helped to catch some of the errors that plague any one line of evidence, but there can be no doubt that the account developed here remains imperfect and incomplete. It could not be otherwise, for the history of metal pollution captures much of the history of social progress, of technological innovation, of war and peace, and of economic growth and decline, all mixed with the complex array of chemical properties that characterize the metals themselves and the various physical forces that determine the movements of water and sediments in the bay. Nevertheless, the effort has set some limits and developed some constraints against which present pollution inputs can be compared. And there have been other rewards as well:

> . . . history is that impossible thing: the attempt to give an account, with incomplete knowledge, of actions themselves undertaken with incomplete knowledge. So that it teaches us no short-cuts to Salvation, no recipe for a New World, only the dogged and patient art of making do . . . by for ever attempting to explain we may come, not to an Explanation, but to a knowledge of the limits of our power to explain.
>
> Graham Swift (1983)
> *Waterland*

ACKNOWLEDGMENTS

This review was made possible by support from the Narragansett Bay Project (a joint effort of the R. I. Department of Environmental Management and the U.S. Environmental Protection Agency) and the R. I. Sea Grant College Program (NOAA). I also acknowledge, with much appreciation, the help of Suzanne Bricker, John King, Virginia Lee, Jenny Martin, Andrew Milliken, Ken Morse, Ken Rahn, Steve Silvia, Dolores Smith, and Wendy Thibodeau, and the patience of Caroline Karp.

References

Anonymous. 1950. Costume Jewelry Manufacturing in Rhode Island. Prepared by R.I. Department of Employment Security, Providence. 30 p.

Bender, M., D. Kester, D. Cullen, J. Quinn, W. King, D. Phelps, and C. Hunt. 1989. Trace metal pollutants in Nasrragansett Bay waters, sediments and shellfish. Report to the Narragansett Bay Project, R. I. Dept. Environmental Management. 75 p. with appendices.

Bolhouse, G. 1967. Incidents through the years at the Portsmouth coal mines, Vol. 1, 1808-1870. *Newport History* 125(40), Part 1:1-21.

Boorstin, D. J. 1987. *Hidden History: Exploring Our Secret Past.* New York: Harper & Row, Publishers, Inc.

Bowen, H. J. M. 1966. *Trace Elements in Biochemistry.* New York: Academic Press.

Bricker, S. 1990. The History of Metals Pollution in Narragansett Bay as Recorded by Salt Marsh Sediments. Ph.D. Thesis in Oceanography, University of Rhode Island, Narragansett, RI.

Bricker Urso, S., S. W. Nixon, J. K. Cochran, D. J. Hirshberg, and C. Hunt. 1989. Accretion rates and sediment accumulation in Rhode Island salt marshes. *Estuaries* 12(4):300-317.

Bride, T. H. 1959. A Study of Employment Patterns in the Rhode Island Jewelry Industry. R. I. Department of Employment Security, Providence. Mimeo. 50 p.

Brown, L. R., W. U. Chandler, A. Durning, C. Flavin, L. Heise, J. Jacobson, S. Postel, C. P. Shea, L. Starke, and E. C. Wolf. 1988. *State of the World 1988.* A Worldwatch Institute Report on Progress Toward a Sustainable Society. New York: W. W. Norton & Company. 237 p.

Carroll, C. F. 1973. *The Timber Economy of Puritan New England***.** Providence, Rhode Island: Brown University Press. 221 p.

Clark, V. S. 1929a. *History of Manufactures in the United States, Vol. IV: 1983-1928*. New York: McGraw Hill.

Clark, V. S. 1929b. *History of Manufactures in the United States, Vol. I: 1607-1860*. New York: McGraw Hill.

Coleman, P. 1963. *The Transformation of Rhode Island, 1790-1860*. Providence, Rhode Island: Brown University Press. 314 p.

Conley, P. T. and P. R. Campbell. 1982. *Providence—A Pictorial History*. Norfolk, Virginia: The Donning Company. 232 p.

Corbin, J. M. 1989. *Recent and Historical Accumulation of Trace Metal Contaminants in the Sediment of Narragansett Bay, Rhode Island*. M. S. Thesis in Oceanography, University of Rhode Island, Kingston, RI.

Cronon, W. 1983. *Changes in the Land*. New York: Hill and Wang. 241 p.

Day, G. M. 1953. The Indian as an ecological factor in the northeastern forest. *Ecology* 34:329-346.

Dunwell, S. 1978. *The Run of the Mill*. Boston, Massachusetts: David R. Godine, Publisher, Inc. 299 p.

Fortescue, J. A. C. 1980. *Environmental Geochemistry: A Holistic Approach*. Ecological Studies 35. :New York: Springer-Verlag. 347 p.

Fowler, W. S. and H. A. Luther. 1950. Cultural sequence at the Potter Pond site. *Bull. Massachusetts Archaeological Society* XI(4):91-102.

Furnas, J. C. 1969. *The Americans: A Social History of the United States: 1587-1914*. New York: G. P. Putnam's Sons.

Garmon, J. E. 1978. *A History of Portsmouth, Rhode Island 1638-1978*. Newport, Rhode Island: Franklin Printing House. 54 p.

Gilkeson, Jr., J. S. 1986. *Middle Class Providence, 1820-1940*. Princeton, New Jersey: Princeton University Press. 380 p.

Goldberg, E.D., E. Gamble, T. T. Griffin, and M. Koide. 1977. Pollution history of Narragansett Bay as recorded in its sediments. *Estuarine and Coastal Marine Science* 5:549-561.

Goldberg, E. D and M. Koide. 1980. A revised estimate foir trace metal fluxes to Narragansett Bay: a reply. *Estuarine and Coastal Marine Science* 11(1):119.

Graham, W. F., M. L. Bender, and G. P. Klinkhammer. 1976. Manganese in Narragansett Bay. *Limnol. Oceanogr*. 21:665-673.

Hartley, E. N. 1957. *Ironworks on the Saugus*. Norman, Oklahoma: University of Oklahoma Press.

Hofstadter, R., W. Miller, and D. Aaron. 1959. *The American Republic, Vol. Two: Since 1865*. Englewood Cliffs, New Jersey: Prentice-Hall, Inc.

Hoffman, E. J., G. L. Mills, J. S. Latimer, and J G. Quinn. 1983. Annual input of petroleum hydrocarbons to the coastal environment via urban runoff. *Can. J. Fish. Aquat. Sci.* 40(Suppl. 2):41-53.

Hoffman, E. J., C. S. Carey, G. L. Mills and J. G. Quinn. 1984. The Magnitude and Effect of Wet Weather Pollutant Inputs to a Municipal Wastewater Treatment Facility Served by a Combined Stormwater-sewage Collection System, p. 241-287. In: Hoffman, E. J. and J. G. Quinn, *Hydrocarbons and Other Pollutants in Urban Runoff and Combined Sewer Overflows.* Report submitted to Water Research June, 1984.

Hoffman, E. J. and J. G. Quinn. 1984. *Hydrocarbons and Other Pollutants in Urban Runoff and Combined Sewer Overflows*. Report submitted to NOAA, Ocean Assessments Division. September 1980-April 1984.

Holliday, R. W. 1965. Chromium, p. 211-255. In: *Mineral Facts and Problems*. 1965 edition. U.S. Bureau of Mines. Bulletin No. 630.

Kleiber, M. 1961. *The Fire of Life: An Introduction to Animal Energetics.* New York: John Wiley & Sons, Inc. 454 p.

Kochiss, J. M. 1974. *Oystering from New York to Boston*., Vol. VII. Middletown, Connecticut: Wesleyan University Press. 251 p.

Kulik, G. and J. C. Bonham. 1978. *Rhode Island: An Inventory of Historic Engineering and Industrial Sites*. Washington, DC: U. S. Department of the Interior, Historic American Engineering Record. 296 p.

Kulik, G. and P. M. Malone. 1977. *The Wilkinson Mill.* Pawtucket, Rhode Island: The American Society of Mechanical Engineers.

McCabe, L. C. 1959. Technical Aspects—the 1950 Assessment, pp. 25-28. In: *Proceedings National Conference on Air Pollution 1958*. U. S. Public Health Service, Pub. No. 654. Washington, DC: U. S. Government Printing Office.

McCaffrey, R. J. and J. Thomson. 1980. A record of the accumulation of sediment and trace metals in a Connecticut salt marsh, p. 165-136. In: B. Saltzman (ed.), *Advances in Geophysics, Estuarine Physics and Chemistry: Studies in Long Island Sound*, Vol. 22. New York: Academic Press.

Melosi, M. V. 1981. *Garbage in the Cities: Refuse, Reform, and the Environment, 1880-1980*. College Station, Texas: Texas A&M University Press. 268 p.

Melosi, M. V. 1985. *Coping with Abundance: Energy and Environment in Industrial America.* Philadelphia, Pennsylvania: Temple University Press. 355 p.

Moore, W. W. 1966. Reduction in ambient air concentration of fly ash—present and future prospects, pp. 170-178. In: *Proceedings Third National Conference on Air Pollution.* U. S. Public Health Service, Pub. No. 1649. Washington, DC: U. S. Government Printing Office.

Mote, R. H. 1949. Cadmium, p. 184-191. In: *Minerals Yearbook*. U. S. Bureau of Mines.

National Research Council. 1980. *Trace -Element Geochemistry of Coal Resource Development Related to Environmental Quality and Health.*. U. S. National Committee for Geochemistry. Washington, DC: National Academy Press. 153 p.

National Research Council. 1984. *Ocean Disposal Systems for Sewage Sludge and Effluent.*. Washington, DC: National Academy Press. 126 p.

Nixon, S. W. 1991. Recent Metal Inputs to Narragansett Bay. Final report to the Naragansett Bay Project, R. I. Department of Environmental Management.

Nriagu, J. O. 1979. Global inventory of natural and anthropogenic emissions of trace metals to the atmosphere. *Nature* 279:409-411.

Nriagu, J. O. and J. M. Pacyna. 1988. Quantitative assessment of worldwide contamination of air, water and soils by trace metals. *Nature* 333:134-139.

Olsen, S. and V. Lee. 1979. A summary and preliminary evaluation of data pertaining to water quality in upper Narragansett Bay. Coastal Resources Center, University of Rhode Island, Narragansett, RI.

Olsen, S., D. D. Robadue, Jr., and V. Lee. 1980. *An Interpretive Atlas of Narragansett Bay*. Coastal Resources Center, Graduate School of Oceanography, University of Rhode Island, Marine Bulletin 40.

Pilson, M. E. Q. 1989. *Aspects of Climate Around Narragansett Bay*. Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Ragsdale, H. L. and C. W. Berish. 1988. Trace metals in the atmosphere, forest floor, soil, and vegetation, p. 367-380. In: W. T. Swank and D. A. Crossley, Jr. (eds.). *Forest Hydrology and Ecology at Coweeta*. Ecological Studies 66. New York: Springer-Verlag.

Raymond, R. 1984. *Out of the Fiery Furnace: The Impact of Metals on the History of Mankind.* University Park and London: The Pennsylvania State University Press. 274 p.

R. I. Department of Health. 1975. Summary of measured Rhode Island air quality 1957-1973 in comparison to national ambient air quality standards. Mimeo report. 22 p.

R. I. Department of Health. Annual air quality data summary issued for calendar years.

R. I. Statewide Planning. 1979. Draft 208 Water Quality Management Plan and Environmental Impact Statement for Rhode Island.

Russell, H. S. 1980. *Indian New England Before the Mayflower*. Hanover, New Hampshire: University Press of New England.

Ryan, J. P. 1965. Silver, p. 809-821. In: *Mineral Facts and Problems*. U. S. Bureau of Mines. Bulletin No. 630.

Santschi, P. H. 1980. A revised estimate for trace metal fluxes to Narragansett Bay: a comment. *Estuarine and Coastal Marine Science* 11(1):115-118.

Santschi, P. H., S. Nixon, M. Pilson, and C. Hunt. 1984. Accumulation of sediments, trace metals (Pb, Cu) and total hydrocarbons in Narragansett Bay, Rhode Island. *Estuarine, Coastal and Shelf Science* 19(4):427-449.

Schroeder, H. J. 1965. Cadmium, p. 165-173. In: *Mineral Facts and Problems*. U. S. Bureau of Mines. Bulletin No. 630.

Simmons, W. S. 1970. *Cautantowwit's House–An Indian Burial Ground on the Island of Cananicut in Narragansett Bay*. Providence, Rhode Island: Brown University Press.

Smith, I. M. 1987. Trace elements from coal combustion: emissions. London, UK. International Energy Agency OECD. 87 p.

Stern, A. C. 1959. Changes in identity and quality of pollutants, past, present, and future, pp. 86-94. In: *Proceedings National Conference on Air Pollution 1958*. U. S. Public Health Service, Pub. No. 654. Washington, DC: U. S. Government Printing Office.

Straub, C. P. 1989. *Practical Handbook of Environmental Control*. Boca Raton, Florida: CRC Press, Inc.

Swift, G. 1983. *Waterland*. New York: Washington Square Press. 270 p.

Tarr, J. A. 1971. Urban Pollution - Many long years ago. *American Heritage*. p. 65-69 and 106.

Tarr, J. A. 1979. The separate vs. combined sewer problem - a case study in urban technology design choice. *J. Urban History* 5(3):308-339.

U. S. Army Corps of Engineers. 1977. *Water Resources Development.* Rhode Island.

U. S. Bureau of the Census. 1975. *Historical Statistics of the United States from Colonial Times to 1970.* Washington, DC: U. S. Department of Commerce.

U. S. Public Health Service. 1958. *Air Pollution Measurements of the National Air Sampling Netword: Analyses of Suspended Particulates 1953-1957.* Washington, DC: U. S. Dept. of Health, Education, and Welfare.

U. S. Public Health Service. 1962. *Air Pollution Measurements of the National Air Sampling Netword: Analyses of Suspended Particulates 1957-1961*. Washington, DC: U. S. Dept. of Health, Education, and Welfare.

U. S. Public Health Service. 1969. National air quality criteria advisory committee. Air quality criteria for particulate matter. Pub. No. AP-49.

Ware, G. C. 1965. Nickel, p. 607-620. In: *Mineral Facts and Problems*. U. S. Bureau of Mines, U. S. Dept. of the Interior. Bulletin No. 630.

Water Supply Board. 1988. Annual Report of the City of Providence, Rhode Island for the year ended June 30, 1984.

Whittaker, C. M. 1926. *Dyeing with coal-tar dyestuffs*. 2nd Edition. New York: Van Nostrand Company. 248 p.

Wroth, L. C. 1970. *The Voyages of Giovanni da Verrazzano: 1524-1528*. New Haven and London: Yale University Press.

SECTION II

Recent Metal Inputs to Narragansett Bay

Ion exchange cylinders – part of the modern pretreatment system for removing copper and lead from wastewater at Victory Finishing Technologies, an electroplating facility in Providence, R.I.
Photo by John Motta, Narragansett Bay Commission

Recent Metal Inputs to Narragansett Bay

Introduction

It is useful to begin this inventory of metal inputs to Narragansett Bay with a few comments about our perception of the amount of fresh water that enters the bay. This is true not just because much of the metal flux to the bay comes in rivers and streams, but also because the difficulty of developing a complete accounting of fresh water input emphasizes the even more serious limits to our knowledge of the inputs of anthropogenic pollutants such as metals.

The earliest estimate I have found of the total fresh water flow into Narragansett Bay was provided by Wehe (1953), who calculated that the flow averaged 76 m^3 s^{-1} for the 22 years from 1930 through 1951. He arrived at this value by applying the ratio of gauged flow to gauged watershed area of the Blackstone River to his estimate of the entire drainage area of the bay. Such an indirect approach was necessary because the fresh water flow from about 58% of the drainage area had never been measured. The ratio of gauged to ungauged flow has varied somewhat in subsequent years as gauges have been added, deleted, or moved, and the ratio is also sensitive to the choice of boundaries for the drainage basin. According to Pilson (1985), the largest drainage area reported in recent publications (4836 km^2) is 18% greater than the smallest.

Unfortunately, Wehe's calculations were contained in an obscure unpublished report, while the measured flows appeared annually in official reports from the U.S. Geological Survey. In spite of the fact that there was a general awareness among those concerned with Narragansett Bay that a portion of the watershed was not gauged, the measured flow began to appear more frequently in the literature, often with little or no qualification about how small a fraction of the total it might represent. A general perception began to form that the input of fresh water to the bay was less than 50 m^3 s^{-1} (Hess and White 1974, Kremer and Nixon 1978, Olsen et al. 1980).

In 1985, as part of his efforts to determine the flushing rate of Narraganset Bay, Pilson expanded and refined Wehe's approach of 30 years earlier and concluded that the more recent "long-term" total input of fresh water to the bay was on the order of 105 m^3 s^{-1}, with less than 50% of the drainage area measured. This revision then prompted an even more extensive analysis by the U.S.G.S. (Ries 1989) that agreed with Pilson's (1985) long-term average input and assigned an uncertainty of ± 10% to the long-term annual input estimate, and ± 20% to the calculated inflow for any particular month. The U.S.G.S. reported that 63% of the watershed was monitored by continuously recording stream gauges.

Assuming that the long-term average input of fresh water is now approximately correct, our working estimates have gone from about 75 to less than 50 to about 100 $m^3 s^{-1}$ in the past 30 years. And a "correct" value has been available only in the last five years. Since large proportions of many of the metals that enter the bay are carried by surface water, the errors in our knowledge of fresh water flow set a minimum uncertainty on our estimates of metal fluxes. In addition are errors associated with the calculation of long-term fluxes from a relatively small number of measurements of metal concentrations in the fresh water, the lack of any sampling in many fresh water sources, and errors involved with the chemical analyses of the metals themselves. Moreover, in contrast to the water budget, anthropogenic discharges from sewage treatment plants, industries, and storm water runoff are important terms in the total input inventory for many metals.

Metal Inputs

In principle, developing an inventory of the inputs of various metals to Narragansett Bay is a straightforward exercise in accounting. In practice, however, it involves an often frustrating accumulation of uncertainty. The problem is particularly difficult with Narragansett Bay because there has never been a systematic attempt to measure the inputs of any metal. In fact, with the notable exception of the discharge from the Fields Point Sewage Treatment Plant as it has recently been operated by the Narragansett Bay Commission, none of the major inputs of metals to the bay has been the subject of a regular and effective monitoring program.

Because of these constraints, it must be admitted at the outset that the inventory developed in the following sections will be very approximate and incomplete. However, I think it is useful to present the results in spite of their imperfections. Many of the major sources of a number of important metals are "reasonably" well described, and the extent of uncertainty in various estimates may stimulate a program of more thorough measurements.

Since the fluxes of metals carried by rivers and deposited from the atmosphere vary with rainfall and river flow, I have made estimates of the inputs in the driest (92 cm y^{-1}), wettest (171 cm y^{-1}), and average (115 cm y^{-1}) precipitation years for the period 1964-1987 (Pilson 1989). Other inputs from publicly owned sewage treatment works (POTWs), combined sewer outfalls (CSOs), and urban runoff must also vary in wet and dry years, but data were not available to quantify the relationships.

Deposition from the Atmosphere

The only direct measurements of the deposition of metals from the atmosphere near Narragansett Bay were made in downtown Providence during a one week period in March of 1982 (Latimer 1984). Unfortunately, it was a dry week with only 4 mm of precipitation that fell largely as snow flurries. However, an extensive series of measurements of the concentrations of many metals in precipitation and in the atmosphere near the ground has been collected on a regular basis by Professor Kenneth Rahn of the Center for Atmospheric Chemistry Studies at the University of Rhode Island (Rahn 1990). He has used the data resulting from measurements made on 198 occasions between February, 1985 and September, 1987 to calculate the estimated wet and dry deposition of numerous metals per unit area during a year with 1.2 m of precipitation (K. Rahn, personal communication 1989). In keeping with the framework adopted for this report, I have pro-rated his calculated fluxes to dry, wet, and average precipitation years and multiplied the results by the area of Narragansett Bay to estimate the total input of each metal directly from the atmosphere to the surface of the bay (Table 1).

Unfortunately, deposition rates calculated from measurements of metal concentrations in the air are sensitive to the choice of an appropriate "wash-out" factor for each metal, and these are known only approximately, perhaps within a factor of about 2 (see Section I, Table 23). I have assumed that the "wash-out" factors remain constant regardless of rainfall, but this may not be correct. There is also some error introduced by extrapolating fluxes calculated from metal concentrations measured in air collected adjacent to the lower West Passage of Narragansett Bay, at the Graduate School of Oceanography, to more urban areas of the bay. The deposition measurements made under essentially dry conditions in Providence were much larger than the dry deposition fluxes calculated from lower bay concentration data (Table 2), and the results of air quality monitoring by the R. I. Department of Health (1986) suggest that concentrations of total particulates and particulate lead may be 2.9 and 8 times larger, respectively, in Providence than at rural sites (Table 3).

There is a great variability in the deposition rates reported for various metals at different coastal sites around the world (Hoffman and Quinn 1984, Nixon and Lee 1986, Windom 1986), and it is difficult to use comparative data to estimate the degree by which the calculations provided in Table 1 may underestimate the total input of any one metal from the atmosphere to Narragansett Bay. For example, the measured deposition of Zn at five coastal sites summarized by Windom (1986) ranged from 3.4 (South Atlantic Bight) to 81 (New York Bight) mg m^{-2} y^{-1} compared to the 11 mg m^{-2} y^{-1}

calculated for Narragansett Bay during a year with average precipitation. Extensive measurements of metal deposition at the mouth of Delaware Bay during 1982 and 1983 showed a Zn flux of 7.5 mg m^{-2} y^{-1} (adjusted to 115 cmy y^{-1} precipitation), but much lower fluxes of Cu, Pb, Fe, and Mn than calculated for lower Narragansett Bay (Church et al. 1984).

Table 1

Estimated deposition of various metals from the atmosphere directly onto the surface of Narragansett Bay (328 km^2)[a]. The estimates were calculated by Prof. Kenneth Rahn (U.R.I. Center for Atmospheric Chemistry Studies) using his measurements of metal concentrations in precipitation and on particles in air sampled at the Graduate School of Oceanography on 198 occasions during 1985-1987. The estimates are for total wet and dry deposition. Units are metric tons.

Year[b]	Average Year[b]		Dry Year[b]	Wet
	mg m^{-2} y^{-1}	tons y^{-1}	tons y^{-1}	tons y^{-1}
As	<0.13	<0.04	<0.03	<0.06
Cr	0.36	0.12	0.10	0.18
Ni	1.6	0.52	0.42	0.77
Cu*	2.8	0.92	0.74	1.4
Mn	3.9	1.3	1.0	1.9
Zn	10.9	3.6	2.9	5.4
Pb	17.6	5.8	4.6	8.6
Fe	148	49	39	73
Al	466	153	122	228

---

[a] Bay surface area from Pilson (1985) does not include the Sakonnet River. Ratios of dry/wet deposition for the average year were: As = 0.044, Cr = 0.096, Ni = 0.73, Mn = 0.89, Zn = 0.44, Pb = 0.035, Fe = 1.41, Al =3.3

*Cu taken as 30% of Zn.

[b] Average year precipitation = 115 cm; dry year = 92 cm; wet year = 171 cm based on data compiled by Pilson (1989) for 1964-1987.

Table 2

Comparison of the measured deposition of various metals from the atmosphere in downtown Providence[1] during one dry week in 1982 (Latimer 1984) with the average dry deposition calculated from metal concentration measurements made adjacent to the lower West Passage of Narragansett Bay between 1985 and 1987 (K. Rahn, URI Center for Atmospheric Studies).

	$\mu g\ m^{-2}\ d^{-1}$	
	Providence	Lower West Passage
Cd	4.3	--
Cu	59	2.9
Pb	269	1.7
Zn	381	9.5
Mn	61	4.3
Fe	453	248

---

[1] Near Rt. 195 and the Narragansett Electric power generation facility.

Table 3

Concentrations of total particulates and particulate lead in the air at various locations in Rhode Island during 1986. Units are µg m^{-3}. Data from R. I. Dept. of Health (1986).

	Total Particulates[1]	Particulate Lead[2]
Providence		
mean for 6 stations (range)	46(43-53)	
mean for 3 stations (range)		0.13 (0.1-0.16)
Pawtucket	42	
East Providence (2 stations)	35-38	
Cranston	38	0.09
Newport	31	
West Greenwich (rural)	18	0.02

[1] Annual geometric mean.

[2] Annual arithmetic mean.

The one short-term set of deposition measurements obtained by Latimer (1984) in downtown Providence produced metal fluxes that were lower than those calculated from metal concentration data collected in the city in 1958 (Section I, Table 23), but that is hardly surprising considering the reductions in metal emissions that have taken place. The reliability of the earlier metal concentration measurements is also a potential problem.

	Calculated Deposition for 1958 (Section I)	Measured Deposition in 1982 (Latimer 1984)
	ug m^{-2} d^{-1}	
Cu	57-115	59
Pb	274-548	269
Zn	1000-2000	381

Unfortunately, there are no earlier estimates of atmospheric deposition in the lower bay to compare with Rahn's calculations. If the calculated deposition in Providence in 1958 applied to the bay as a whole, the flux of metals to the bay would have been very much higher than at present (Table 4).

Inputs from Rivers

Of the 14 river and stream systems identified by the U.S.G.S. as contributing significant amounts of fresh water to Narragansett Bay, the three largest—the Taunton, Blackstone, and Pawtuxet Rivers— drain 75% of the land area of the watershed (Ries 1989). The Taunton River supplies 34% of the long-term average fresh water flow, the Blackstone 28%, and the Pawtuxet 13%. The concentrations of metals in the free-flowing portion of the Taunton River appear to have been measured on only two occasions, both of which occurred in 1986 as part of the SINBADD program (Pilson and Hunt 1989). Total metal concentrations in the Blackstone and Pawtuxet Rivers have been measured twice each year since 1978, usually in March and September, by the U.S.G.S., and both rivers have received additional short-term study by various groups from the University of Rhode Island (Table 5). While the URI studies have differed in their purposes, with some collecting samples in transects along the rivers, in dry or wet weather, during individual storm events, at fixed time intervals, etc., they have all collected samples just above the point where the rivers first meet salt water (Fig. 1). The U.S.G.S. samples are collected about 13 km and 5 km above these points in the Blackstone and Pawtuxet Rivers, respectively. Some of the URI studies have also included measurements of metals in the Woonasquatucket and Moshassuck, two smaller urban rivers that pass through Providence (Fig. 1), and provide about 3.2% and 1.3%, respectively, of the long-term fresh water flow (Ries 1989). The more recent SPRAY sampling(Table 5) and a storm sampling program by R. Wright and

Table 4

Comparison of the calculated deposition of various metals from the atmosphere to the surface of Narragansett Bay (328 km^2) based on concentrations of metals in the air in Providence during 1958 and in precipitation and on particles in the air at the Graduate School of Oceanography, on the lower bay, during 1985-1987. The concentration data for both periods have been applied to an annual precipitation of 115 cm, the average for 1964-1987 (Pilson 1989).

	Deposition, metric tons y^{-1}	
	1958[a]	1985-1987[b]
Cr	2.3-4.6	0.1
Cu	5.8-11	0.9
Ni	6.7-13	0.5
Pb	29-58	6
Zn	105-210	4

[a] Modified from Section I, Table 23.

[b] From Table 1.

colleagues at U.R.I. also collected water from the Ten Mile River (Fig. 1) and this tributary was also analyzed for metals on some occasions in 1984 by the Rumford River Laboratories (1985) under contract to the Massachusetts Department of Environmental Quality. The Ten Mile River samples were all collected above Omega Pond, however, and the amount of each metal removed in the pond is not known. Because of this uncertainty, and the fact that only 3.4% of the average fresh water input is involved, I have not attempted to analyze the metal flux and river flow relationship of this system. Overall, there appear to be sufficient data available to provide a reasonably credible estimate of the input of at least some metals from the Blackstone, Pawtuxet, Woonasquatucket and Moshassuck Rivers. These systems provide a total of about 45% of the fresh water inflow to Narragansett Bay (Ries 1989). The calculation of input from the largest river, the Taunton, rests on only two sets of measure-ments and can only be described as a "best guess." Moreover, it is not known how much of each metal that is delivered to Mt. Hope Bay by the Taunton River actually enters Narragansett Bay proper. A large fraction of at least some metals must be retained in the sediments of Mt. Hope Bay, and a small portion must pass into the Sakonnet River (Fig. 1). As far as I am aware, no metal analyses have been reported for the rivers and streams that contribute the remaining 20% or so of the fresh water input to the bay.

Of the various sampling programs summarized in Table 5, only the one carried out by Hunt (1982) was designed to provide an estimate of metal fluxes from the rivers over time periods longer than a day or over the course of an individual storm. Hunt approached the problem of determining an annual flux by measuring the concentrations of metals in the Blackstone and Pawtuxet Rivers every other week during the eight months between 25 January, 1980 and 26 September, 1980. He then multiplied the measured concentrations by the average river flow on the day of sampling as reported by the U.S.G.S. to produce an estimate of the daily metal flux on each day of sampling. These fluxes were then averaged and multiplied by 365 to give an estimate of the annual input from each river (Table 6).

Since these unpublished values are the only long-term river flux estimates that have been available for the past decade and they have contributed significantly to several metal budgets that have been prepared for Narragansett Bay (e.g., Hoffman and Quinn 1984, Santschi et al. 1984, Nixon et al. 1986), they deserve some further comment. The U.S.G.S. river flow data used by Hunt came from gauges upstream of the points where the metal samples were collected and where the rivers actually first enter salt water. As a result, some adjustment may be necessary to account for runoff that entered the rivers below the gauges. Correction factors based on the ratio of total watershed area to gauged area of 1.17 and 1.15 have been reported for the Blackstone and the Pawtuxet, respectively, by Pilson

Table 6

Estimates of the fluxes of metals into the Seekonk and Providence Rivers from the Blackstone and Pawtuxet Rivers, respectively, during 1980 as reported by Hunt (1982) and after adjustment for ungauged river flow using factors provided by Pilson and Hunt (1989). Units are metric tons.

	Blackstone River	Pawtuxet River
Fe	592	207
adjusted	(691)	(238)
Mn	88	44
adjusted	(103)	(51)
Cu	12	4
adjusted	(14)	(4.6)
Pb	7.2	1.2
adjusted	(8.4)	(1.4)
Cd	1.7	0.25
adjusted	(2.0)	(0.29)

and Hunt (1989) and I have applied them to Hunt's original estimates in Table 6. It is also worth noting that river flows during the eight month period studied by Hunt were low. For example, for the Blackstone River the mean February-September flows in 1980 were only 78% of the mean for comparable months during 1963-1984 (Pilson 1989). Precipitation averaged over the Narragansett Bay drainage basin amounted to only 96 cm in 1980 com-pared to the long-term mean of 120 cm (Pilson 1989). Since regression analyses described in the next paragraph show that the flux of most metals in the rivers appears to be determined largely by flow, it seems reasonable to assume that the daily average fluxes calculated from 1980 river flow data would lead to a significant underestimate of metal inputs during average flow conditions.

Aside from these considerations, the averaging of daily flux estimates is only one of a number of approaches for dealing with the common problem of estimating the long-term (monthly, annual) flux of materials carried by rivers (Dolan et al. 1981, Bierman et al. 1988). Because the sampling programs applied to the rivers entering Narragansett Bay have provided estimates of metal fluxes under a wide range of flow conditions, but at irregular and relatively infrequent intervals, I have used a standard regression technique to relate the daily flux of each metal to the average flow of the river on the day of sampling. The U.S.G.S. flow data were increased by the factors provided by Pilson and Hunt (1989) to compensate for ungauged flow and linear regressions performed on the logarithmic transforms of flux and flow data. It was then possible to use the regressions or "rating curves" with daily river flow measurements during dry (1980), wet (1972), and "average" (1973) years to calculate the daily metal fluxes that might result. The calculated fluxes were corrected for the bias introduced by the logarithmic transformations (Ferguson 1986) and summed to provide estimates of the monthly and annual inputs of the various metals. The regression format also made it possible to account for differences in river flow when comparing the results of the different sampling programs (Figs. 2 and 3).

It is evident from the analyses that the fluxes of all of the metals examined, with the notable exception of nickel in the Pawtuxet River and in the small urban rivers, were governed largely by variation in river flow (Figs. 2 and 3; Table 7). In general, the corre-lations were stronger for dissolved and total metals than for the particulate portion, and in virtually every case the flux of metals was more strongly correlated with flow in the Blackstone River than in the Pawtuxet River or the two smaller systems (Table 7). In both the Blackstone and the Pawtuxet, total metal fluxes computed from the concentration measurements reported by the U.S.G.S. tended to be greater than those obtained from the more recent SINBADD and SPRAY programs (Figs. 2 and 3). The discrepancy is particularly clear for lead, and it may be that the U.S.G.S. data set is elevated because it

Table 7

Slope, intercept, and r^2 values for regression equations relating log metal flux (kg d^{-1}) to log river flow (m^3 d^{-1}, corrected for ungauged watershed area) used to estimate monthly and annual metal inputs to Narragansett Bay from rivers (see Figs. 2-5). Data for dissolved (< 0.4 µm) (d), particulate (p), and total metals (t) from SINBADD and SPRAY sampling programs; others from U.S.G.S. (see Table 5).

Metal	Slope	Intercept	r^2	Slope	Intercept	r^2
		Blackstone			Pawtuxet	
Cd (d)	1.20	-7.40	0.94	0.40	-2.67	0.37
Cd (p)	1.17	-7.77	0.87	0.71	-5.33	0.56
Cd (t)	1.09	-6.64	0.75	0.42	-2.71	0.40
Cr (d)**	0.96	-5.74	0.98	0.59	-4.05	0.89
Cr (p)**	1.30	-7.87	0.90	0.97	-6.21	0.64
Cr (t)**	1.15	-6.60	0.95	0.72	-4.48	0.82
Cu (d)	0.82	-4.07	0.96	0.64	-3.12	0.90
Cu (p)	1.26	-7.30	0.94	1.10	-6.36	0.64
Cu (t)	0.94	-4.69	0.97	0.72	-3.44	0.90
Pb (d)	1.00	-6.04	0.98	1.00	-6.25	0.78
Pb (p)	0.90	-5.15	0.88	0.88	-5.14	0.86
Pb (t)	0.93	-5.15	0.93	0.88	-4.98	0.91
Ni (d)	0.67	-2.98	0.86	0.10	0.38	0.11
Ni (p)	0.97	-6.03	0.65	0.56	-3.75	0.27
Ni (t)	0.70	-3.07	0.85	0.11	0.35	0.13
Zn (t)	1.08	-4.79	0.80	1.08	-4.90	0.61
As (t)	0.69	-3.90	0.79	0.96	-5.66	0.70
Mn (t)	0.99	-3.92	0.92	0.60	-1.39	0.74
Ag (d)*	1.03	-7.46	1.00	0.72	-5.51	0.73
Ag (p)	1.27	-8.94	0.95	1.09	-7.59	0.57
Ag (t)	1.16	-7.96	0.98	0.94	-6.47	0.65
		Woonasquatucket			Moshassuck	
Cd (d)	0.43	-3.81	0.32	0.40	-2.67	0.37
Cd (p)	0.62	-5.46	0.35	0.71	-5.33	0.56
Cd (t)	0.45	-3.82	0.34	0.42	-2.71	0.40
Cr (d)**	0.79	-4.31	0.69	0.59	-4.05	0.89
Cr (p)**	0.61	2.08	1.00	0.97	-6.21	0.64
Cr (t)**	0.49	-2.68	0.51	0.72	-4.48	0.82
Cu (d)	0.92	-5.10	0.73	0.64	-3.12	0.90
Cu (p)	0.46	-3.02	0.24	1.10	-6.36	0.64
Cu (t)	0.76	-4.05	0.61	0.72	-3.44	0.90

Table 7 (continued)

Metal	Slope	Intercept	r^2	Slope	Intercept	r^2
		Woonasquatucket			Moshassuck	
Pb (d)	1.01	-6.01	0.53	1.00	-6.25	0.78
Pb (p)	0.39	-2.38	0.26	0.88	-5.14	0.86
Pb (t)	0.56	-3.11	0.28	0.88	-4.98	0.91
Ni (d)	0.46	-2.45	0.39	0.10	0.38	0.11
Ni (p)	0.67	-4.77	0.33	0.56	-3.75	0.27
Ni (t)	0.46	-2.45	0.39	0.11	0.35	0.13
Ag (d)	1.48	-9.98	0.98	0.72	-5.51	0.73
Ag (p)	0.45	-4.85	0.27	1.09	-7.59	0.57
Ag (t)	1.61	-10.60	0.93	0.94	-6.47	0.65
		Taunton[***]				
Cd (d)	1.32	-8.90	--			
Cd (p)	1.73	-11.79	--			
Cd (t)	1.44	-9.48	--			
Cr (d)	0.61	-3.84	--			
Cr (p)	0.07	-0.64	--			
Cr (t)	0.34	-1.93	--			
Cu (d)	0.80	-4.28	--			
Cu (p)	1.34	-8.51	--			
Cu (t)	0.85	-4.54	--			
Pb (d)	1.12	-6.76	--			
Pb (p)	--	--	--			
Pb (t)	2.02	-12.0	--			
Ni (d)	0.71	-3.91	--			
Ni (p)	--	--	--			
Ni (t)	0.54	-2.84	--			
Ag (d)	--	--	--			
Ag (p)	--	--	--			
Ag (t)	--	--	--			

---

[*] Dissolved Ag values were often set at the lowest detectable concentration of 0.05 μg kg^{-1}. See text.

[**] While regressions for other metals entering the Seekonk and Providence Rivers were based on about 20 measurements, only three measurements of Cr were available from SINBADD on the Woonasquatucket River and four for the other rivers. The SPRAY program did not include Cr.

[***] Taunton "regressions" based on two measurements only.

contains older measurements. Presumably, both rivers are now carrying lower concentrations of metals, especially lead, than they did in the late 1970s and early 1980s when the U.S.G.S. began sampling (Table 5). Unfortunately, at twice each year, their sampling is not frequent enough to be appropriate for a trend analysis. It is also possible that the concentrations of metals are higher at the U.S.G.S. sampling sites than at the mouths of the rivers or that the analytical procedures used by the U.S.G.S. produce higher values than those used by the metals laboratory at the Marine Ecosystems Research Laboratory (MERL) at URI, the analytical facility for the SINBADD and SPRAY studies. There is a notorious variability in the analysis of metals among different laboratories, and there has never been an intercalibration among the four different laboratories producing the data shown in Figs. 2 and 3.

Because their samples were collected upstream of the SINBADD and SPRAY samples and during a period of over 10 years, I have only used the U.S.G.S. regressions to calculate annual loadings of As, Mn, and Zn, metals that were not included in the SINBADD and SPRAY programs. Because the fluxes of other metals reported by the U.S.G.S. appear high relative to those measured by SINBADD-SPRAY (Figs. 2 and 3), the annual fluxes of As, Mn, and Zn are reported here as being "less than" the values computed from the regressions based on the U.S.G.S. data (Tables 8 and 9). Annual fluxes of other pollutant metals have been calculated from regressions based on a combined data set from the SINBADD and SPRAY sampling programs (Tables 8, 9, 10). These data are generally consistent, the samples were collected recently (1985-1987) at points where the rivers enter tidal salt water, samples were collected in all seasons under a wide range of flow conditions, and the sampling and analyses were carried out by the same laboratory (URI-MERL). Coefficients of variation (%) for mean bias-corrected residuals of the daily metal fluxes calculated from the SINBADD-SPRAY regressions were as follows for an "average flow" year:

	Blackstone	Pawtuxet
Cd	63	46
Cr	11	11
Cu	25	22
Ni	31	22
Pb	37	17

Since particulate and dissolved (< 0.4µm) metals were analyzed separately, it was also possible to estimate the contribution made by each fraction to the annual flux under different flow conditions (Tables 11 and 12). The slopes of the flux vs. flow

Table 8

Estimates of the annual flux of various metals into the Seekonk River from the Blackstone River during years with low, high, and average rainfall. Data from SINBADD and SPRAY sampling programs unless noted. Flow data used in regressions were corrected for ungauged watershed areas according to Pilson and Hunt (1989). Units are metric tons.

	Dry Year	Wet Year	Average Year
As (USGS)[1]	<0.9	<1.6	<1.3
Fe (Hunt) [2]	690	--	--
Mn (Hunt)[2]	103	--	--
Mn (USGS)[1]	<64	<151	<109
Cd	0.6	1.5	1.0
Cr (SINBADD)	1.2	3.3	2.3
Cu	4.9	11.0	8.2
Ni	6.4	11.5	9.4
Pb	1.6	3.5	2.6
Zn (USGS)[1]	<30	<76	<53
Ag[*]	<0.07	<0.18	<0.12

[1] An assessment of the USGS metal sampling program in east coast rivers published after this report was originally submitted concluded that the USGS analyses overestimated concentrations of Cd, Cu, Pb, and Zn (Windom et al. 1991). No assessment of As or Mn analyses was given.

[2] C. Hunt (1982), concentrations measured in 1980. Flow used by Hunt has been corrected here to include ungauged portions of the watershed as reported by Pilson and Hunt (1989).

[*] Dissolved Ag concentrations in the Blackstone were frequently below the detection level of 0.05 µg kg^{-1}. When this was the case, a value of 0.05 was added to the particulate concentration used in the total metal flux vs. flow regression. Therefore, the actual total Ag flux was less than computed from the regression.

Table 9

Estimates of the annual flux of various metals into the Providence River from the Pawtuxet River during years with low, high, and average rainfall. Data from SINBADD and SPRAY sampling programs unless noted. Flow data used in regressions were corrected for ungauged watershed areas according to Pilson and Hunt (1989). Units are metric tons.

	Dry Year	Wet Year	Average Year
As (USGS)[1]	<0.4	<0.8	<0.7
Fe (Hunt)[2]	238	--	--
Mn (Hunt)[2]	51	--	--
Mn (USGS)[1]	<49	<74	<72
Cd	0.22	0.32	0.29
Cr (SINBADD)	0.20	0.37	0.32
Cu	2.0	3.8	3.3
Ni	3.5	3.9	3.8
Pb	0.52	1.1	0.95
Zn (USGS)[1]	<11	<27	<21
Ag[*]	<0.05	<0.11	<0.09

---

[1] An assessment of the USGS metal sampling program in east coast rivers published after this report was originally submitted concluded that the USGS analyses overestimated concentrations of Cd, Cu, Pb, and Zn (Windom et al. 1991). No assessment of As or Mn analyses was given.

[2] C. Hunt (unpublished), concentrations measured in 1980. Flow used by Hunt has been corrected here to include ungauged portions of the watershed as reported by Pilson and Hunt (1989).

[*] While dissolved Ag concentrations in the Pawtuxet were not below detection levels (0.05 µg kg^{-1}) as frequently as in the Blackstone, the regression for this river also contained enough detection level readings that the computed fluxes are overestimates.

Table 10

Estimates of the annual flux of various metals into the Providence River from the Woonasquatucket and Moshassuck Rivers during years with low, high, and average rainfall. Data from SINBADD and SPRAY sampling programs. Units are metric tons.

	Dry Year	Wet Year	Average Year
		Woonasquatucket River	
Cd	0.01	0.02	0.02
Cr (SINBADD)*	18	29	25
Cu	0.28	0.54	0.44
Ni	0.34	0.52	0.46
Pb	0.22	0.37	0.32
Ag	0.01	0.04	0.03
		Moshassuck River	
Cd	0.21	0.30	0.28
Cr (SINBADD)*	0.18	0.34	0.29
Cu	1.8	3.5	3.0
Ni	3.5	3.8	3.8
Pb	0.46	1.0	0.84
Ag	0.04	0.10	0.08

*Regressions derived from only three and four measurements of concentration (See Table 7).

Table 11
Estimated fraction (%) of the total flux of various metals from the Blackstone River that is in dissolved form during years with low and high rainfall and during months with the lowest and highest river flow in a year with average rainfall.

	Dry Year	Wet Year	Average February	Average
October				
Cd	81	81	82	78
Cr	49	42	42	58
Cu	73	65	65	81
Ni	95	94	93	97
Pb	31	33	33	29

Ag has not been partitioned because the dissolved fraction was usually below the detection level of 0.05 µg kg^{-1}.

Table 12

Estimated fraction (%) of the total flux of various metals from the Pawtuxet River that is in dissolved form during years with low and high rainfall and during months with the lowest and highest river flow in a year with average rainfall.

	Dry Year	Wet Year	Average February	Average
October				
Cd	89	87	85	92
Cr	46	41	40	45
Cu	77	72	70	81
Ni	87	90	92	85
Pb	29	34	36	28

Ag has not been partitioned because the dissolved fraction was often below the detection level of 0.05 $\mu g\ kg^{-1}$.

regressions for dissolved and particulate metals are sufficiently similar that there appears to be only a modest increase in the relative transport of particulate metals in high flow years or during wet compared to dry months. The Blackstone and Pawtuxet Rivers contribute most of their Ni, Cd, and Cu in dissolved form; most of their Pb is delivered in association with particles; and Cr is distributed about equally between particulate and dissolved fractions (Tables 11 and 12).

Since the flux of metals is determined so strongly by the discharge of the rivers, there is a marked seasonality in the calculated input of metals from this source (Figs. 4 and 5). The effective metal loading on Narragansett Bay may be less variable, however, because the residence time of water in the bay is shorter when fresh water inflow is high. Using Pilson's (1985) relationship between total freshwater input and flushing rate, the monthly average residence time of water during the "average" flow year for this report (1973) varied from 17 days in December to 36 days in October. If the average flushing rate (days) for each month is multiplied by the computed combined metal inputs from the Blackstone and Pawtuxet Rivers (kg d^{-1}), it is possible to gain some appreciation for the interaction of input and flushing rate on the relative loading of metals from the rivers (Fig. 6). For example, while the computed input of Cu from the two rivers varied by a factor of 4.8 between the months with lowest and highest river flow, the flushing-corrected loading rate only varied by a factor of 2.3. The importance of this interaction in setting ambient concentrations of metals in the bay will vary depending on the conservative or non-conservative behavior of individual metals and the influence of temperature and other factors on that behavior (Santschi et al. 1980, Bender et al. 1989).

The weaker relationships between metal fluxes and river flows in the two small urban rivers (Table 7) are not surprising, and it is tempting to dismiss the uncertainty involved because the combined flow of the Woonasquatucket and Moshassuck is only about 16% of the Blackstone (Ries 1989). Unfortunately, however, the regressions suggest that their combined metal inputs may amount to 30-45% of the flux from the Blackstone and exceed that of the Pawtuxet (Table 13). It seems clear that it will require a more intensive sampling than provided by the SINBADD or SPRAY programs to adequately characterize the metal inputs from these sources. The problem may be particularly important for chromium, since the very limited sampling for this metal during the SINBADD program showed a potential for very high fluxes from the Woonasquatucket (Table 10). However, as emphasized in the tables summarizing metal inputs from the rivers, chromium has only been measured a few times in any of the rivers, and data from the U.S.G.S. and from Quinn et al. (1985, 1987) show generally higher concentrations in the Blackstone and Pawtuxet Rivers than reported

Table 13
Estimated flux of various metals into Narragansett Bay proper (i.e., excluding Mt. Hope Bay) from rivers and streams during a year with average flow. Many of the values are quite uncertain, and the reader is advised to review the preceding text and footnotes carefully. Units are metric tons.

	Blackstone	Pawtuxet	Woonasquatucket Plus Moshassuck	Others[a]	Total
As[b]	<1.3	<0.7	<0.52[c]	1.0	<3.5
Fe[d]	>690	>238	>113[e]	528	>1569
Mn[d]	>103	>51	>17[e]	79	>250
Cd	1.0	0.29	0.30	0.77	2.4
Cr[f]	2.3	0.32	>0.29[g]	1.8	>4.7[h]
Cu	8.2	3.3	3.4	6.3	21
Ni	9.4	3.8	4.3	7.2	25
Pb	2.6	0.95	1.2	2.0	6.8
Zn[b]	<53	<21	<21[c]	41	<136
Ag	<0.12	<0.09	<0.11	0.09	<0.41

[a] Metal concentrations in other fresh water inflows have not been measured. The estimates provided here are 76.6% of the calculated loading from the Blackstone River. The average long-term input of fresh water from these sources is equal to the same percentage of the average annual long-term discharge of the Blackstone (Ries 1989). The actual inputs probably fall somewhere between zero and the values given here.

[b] These metals were only measured by the USGS upstream of the final dams over which the rivers enter salt water. As discussed in text, the metal concentrations found by the USGS were always higher than measured by other investigators sampling at the dams.

[c] The measured flux of pollutant metals in these two rivers is equal to approximately 40% of the flux from the Blackstone River. Since As and Zn have not been measured in the Woonasquatucket and Moshassuck Rivers, the estimated flux given here is 40% of the flux of these metals from the Blackstone River.

[d] As discussed in text, these metals were only measured by Hunt (1982) during a dry year. Fluxes during the "average flow" year would, presumably, have been greater. The flow of water from the Blackstone during an average year is 1.7 times the dry year flow.

[e] These metals have not been measured in the Woonasquatucket or Moshassuck Rivers. The values here are 16.4% of the reported flux from the Blackstone River. The combined long-term average annual flow of these two rivers is equal to the same percentage of the long-term Blackstone River flow (Ries 1989).

[f] As emphasized earlier, the data base for Cr consists of only 3-4 measurements.

[g] If the very high calculated flux from the Woonasquatucket is accepted (Table 10), the total would be 25 tons.

[h] If note f (above) is followed, the overall total would exceed 30 tons.

by SINBADD (Pilson and Hunt 1989) (Figs. 3 and 4). For all these reasons, my estimates of chromium input from the rivers are particularly uncertain.

As noted earlier, there are other small rivers and streams that enter Narragansett Bay proper and contribute about 20% of the total fresh water flow. With the exception of the Ten Mile River, these systems are ungauged and appear never to have been monitored for metals. Like the Ten Mile, many of them do not have a clear fall line separating free flowing fresh water from salt, and some flow into ponds that may act as settling basins retaining some of the metals carried by the streams. Some of these systems drain largely undeveloped land and probably have low concentrations of pollutant metals, while others flow through urban areas and probably carry a heavy burden of pollutants. In the absence of data, I have set metal fluxes from these combined sources as being equal to 76.6% of the calculated input from the Blackstone River (Table 13). Their combined long-term average flow amounts to this fraction of the Blackstone flow, and the Blackstone provides the best combination of a large drainage area with a wide mix of land uses and a "reasonably" well-characterized flux of most of the pollutant metals. It is evident that these unmeasured sources could contribute significantly to the total flux of metals into Narragansett Bay from surface fresh water flow (Table 13). If the Woonasquatucket and Moshassuck Rivers are combined with the "unmeasured sources", it appears that 40-50% of the estimated flux of metals into Narragansett Bay proper from rivers and streams may come from sources that are either unmeasured or very uncertain.

The situation in Mt. Hope Bay is much more difficult, since the "rating curve" for the Taunton River is based on only two sets of measurements and fewer metals were sampled (Table 5). The very preliminary estimates from these data suggest that the inputs of cadmium, chromium, copper, and nickel from the Taunton are only 5-20% of those from the fresh water sources entering Narragansett Bay proper, while lead may be about the same (Table 14). Characterizations of inputs from the Taunton River should be a high priority for any future monitoring program.

The Influence of Storms

Neither the SINBADD nor the SPRAY programs were intended to sample the rivers during or immediately following rain storms. By sampling at all seasons, however, they were able to capture a very wide range in flow conditions. For example, in the Blackstone River, metal concentrations were measured when flow rates ranged from less than 5×10^5 m^3 d^{-1} to over 7×10^6 m^3 d^{-1} and in the Pawtuxet River, from less than 3×10^5 m^3 d^{-1} to over 3×10^6 m^3 d^{-1}. These flow conditions span most of the range found by R. Wright and colleagues in their study of the effect of storms on metals in the rivers (Figs. 2 and 3).

Table 14

Estimates of the annual flux of various metals into Mt. Hope Bay from the Taunton River during years with low, high, and average flow. Flux and flow regressions were based on only two sampling measurements from the SINBADD sampling program. Flow data used in regressions were corrected for ungauged watershed areas according to Pilson and Hunt (1989). Units are metric tons.

	Dry Year	Wet Year	Average Year
Cd	0.08	0.41	0.33
Cr	0.50	0.73	0.71
Cu	1.7	4.5	4.0
Ni	0.09	1.7	1.6
Pb	1.0	10.5	7.1

While the "instantaneous" flux of metals in the rivers during and after individual storms is obviously influenced by factors in addition to flow, the combined data from the three storms studied by Wright and colleagues show a strong relationship between the flux of metals and the discharge of water in the Blackstone and, to a lesser degree, in the Pawtuxet Rivers (Fig. 7, Table 15). The flux of cadmium in the Pawtuxet was a notable exception. This metal was not a function of flow during storms and was only weakly correlated with flow in the SINBADD and SPRAY data (Table 7). Cadmium also presented problems in the Pawtuxet for Latimer et al. (1988) in their attempt to develop mass balances for metals in the river during low flow. On average, over 100 times more cadmium was discharged from the Pawtuxet than they could account for in their measured inputs to the river. The puzzling behavior of cadmium in the Pawtuxet River deserves further study.

A comparison of the regressions generated from the storm sampling data and the SINBADD-SPRAY data for the Blackstone River shows similar fluxes of cadmium and copper, but a much higher flux of lead during storms (Fig. 2, Table 16). This is consistent with earlier conclusions based on land use and measurements of metal runoff during storms, that urban runoff was potentially the major source of lead, but only a minor contributor of copper, to Narragansett Bay (Hoffman 1987). It is not clear, however, which regression serves as the better tool for estimating the annual input of lead from the river. In this report, I have relied on the SINBADD-SPRAY data for lead in order to be consistent with most of the other pollutant metals and because they were collected over a longer period of time. The storm-based regressions probably represent an upper bound to the estimate of lead input from rivers.

Some Implications of the Metal Flux-River Flow Relationship

The observation that the amounts of most metals carried by the rivers entering Narragansett Bay increase with increasing river flow (Figs. 2 and 3; Tables 7 and 15) does not necessarily mean that the concentrations of metals do not decrease at higher flow rates. For some metals, such as dissolved Cu and Ni, there is clear evidence of dilution, while for others, such as Pb or particulate Ni, concentrations remain relatively constant across a wide range in flow (Fig. 8). In most of the cases where the concentration declines with flow, however, the decline is small relative to the increase in flow, with the result that the total flux increases. For example, during the SINBADD and SPRAY programs, dissolved Cu concentrations in the Blackstone River varied by a factor of about 2.2, while river flow varied over 17 fold (Fig. 8).

This behavior suggests that the source of most metals entering the rivers must increase with the input of water. While it seems unlikely that industrial discharges of metals, either

Table 15

Slope, intercept, and r^2 values for regressions relating the log of metal flux (kg d^{-1}) to the log of river flow (m^3 d^{-1}) using data collected during three rainstorms* at the mouths of the Blackstone and Pawtuxet Rivers by R. Wright and colleagues at the University of Rhode Island.

	Blackstone			Pawtuxet		
	Slope	Intercept	r^2	Slope	Intercept	r^2
Cd (d)	0.87	-5.43	0.83	0.49	-3.09	0.18
Cd (p)	1.21	-7.94	0.69	1.09	-7.12	0.37
Cd (t)	1.01	-6.15	0.87	0.61	-3.65	0.25
Cu (d)	0.71	-3.27	0.87	0.84	-4.28	0.69
Cu (p)	1.20	-6.70	0.79	1.33	-7.38	0.64
Cu (t)	0.89	-4.25	0.92	0.99	-4.86	0.69
Pb (d)	1.26	-7.24	0.74	1.29	-7.46	0.60
Pb (p)	1.17	-6.50	0.76	1.65	-9.45	0.79
Pb (t)	1.24	-6.67	0.83	1.56	-8.61	0.84

---

*Oct. 1988, May 1989, June 1989

Table 16

Comparison of estimates of the annual flux of various metals into the Seekonk River from the Blackstone River using regressions developed from data collected during storms (R. Wright, personal communication) and regressions developed with data collected during a regular monitoring program (SINBADD and SPRAY). Units are metric tons.

	Dry Year	Wet Year	Average Year
Cd (storms)	0.4	1.1	0.8
Cd (SINBADD-SPRAY)	0.6	1.5	1.0
Cu (storms)	6.1	13	9.9
Cu (SINBADD-SPRAY)	4.9	11	8.2
Pb (storms)	3.6	10	6.8
Pb (SINBADD-SPRAY)	1.6	3.5	2.6

directly to the rivers or to sewage treatment plants, would necessarily increase with runoff, a recent study by Hoffman et al. (1988) has shown that the release of some pollutants, including Pb, from the Fields Point POTW increased during storms due to hydraulic surges and the loss of sludge. However, while their analysis suggested that the discharge of Pb could increase by at least 50% during an average rainstorm, they found little or no influence of storms on the release of Cd, Cu, or Ni. Moreover, the influence of storms on the release of metals from POTWs should be relatively evenly distributed throughout the year, following the distribution of precipitation (Pilson 1989) and urban runoff, while the source for the rivers must have a strong seasonality that follows total surface runoff and groundwater flow.

The sources of metals carried into the rivers by surface and groundwater flows could include deposition from the atmosphere, disposal of metals on natural or paved surfaces (for example, Zn in crankcase oil leaked onto highways as reported by Hoffman and Quinn 1984), and the leaching of metals contained in landfills.

As a first step in evaluating the potential importance of these sources, I have used the deposition calculations from Table 1, along with measurements of Cd deposition on lower Delaware Bay (Church et al. 1984), to estimate the total flux of metals from the atmosphere to the watershed of the Blackstone River. With the notable (and not surprising) exception of Pb, the flux of metals from the atmosphere is small relative to the mass of metals discharged by the river (Table 17). This is in marked contrast to the Delaware River, where Church et al. (1988) found that inputs of Cd, Cu, Ni, Zn and Fe from the atmosphere to the watershed exceeded the fluxes of these metals in the river by factors of 1.4, 5.9, 7.7, 46, and 206, respectively. Only Mn transport by the Delaware exceeded atmospheric deposition (by a factor of 53).

A major difference between the Delaware and the Blackstone Rivers is that there is very little industrial activity along the Delaware above Trenton, N. J., where the metal fluxes in the river were measured, while the Blackstone receives major sewage and industrial loadings upstream of the sampling station used to measure fluxes (Quinn et al. 1987, Lewis and Brubaker, undated). Assuming that point source discharges from POTWs and industries provide most of the metals carried by the Blackstone during dry weather, low flow conditions, I have extrapolated the calculated transports of metals during October of the "average flow year" (Fig. 4) to a full year for comparison with the annual fluxes calculated from the metal flux vs. river flow regressions. The results suggest that the source or sources of metals supporting the "excess" fluxes during higher flows are larger than those providing the flux of metals under base flow conditions

(Table 18). Since the "excess" fluxes (excepting Pb) are also still larger than the estimated fluxes from atmospheric deposition

Table 17

Comparison of the estimated deposition of various metals from the atmosphere to the watershed of the Blackstone River[1] with the flux of metals from the river into Narragansett Bay.

	Estimated Deposition[2]	Estimated River Flux[3]
	tons y^{-1}	
As	<0.16	<1.3
Cd	0.26*	1.0
Cr	0.44	2.3
Cu	3.4	8.2
Ni	2.0	9.4
Pb	21	2.6
Zn	13	<53
Mn	4.8	<109
Fe	180	>690

[1] 1222 km^2 (Ries 1989).

[2] See Table 1, average year.

[3] See Table 8, average year.

*Calculated using deposition measurements from lower Delaware Bay (Church et al. 1984) pro-rated to average year rainfall for Narragansett Bay.

Table 18

Comparison of the estimated deposition of various metals from the atmosphere to the watershed of the Blackstone River[1] with the flux of metals from the river into Narragansett Bay that is in excess of base flow transport[2] in a year with average precipitation.

	Estimated Deposition	Base Flow Flux	Excess
	Flux[3] metric tons y^{-1}		
As	<0.16	<0.61	<0.69
Cd	0.26[*]	0.29	0.71
Cr	0.44	0.58	1.7
Cu	3.4	2.8	5.4
Ni	2.0	4.3	5.1
Pb	21	0.9	1.7
Zn	13	<17.6	<35.4
Mn	4.8	<34.4	<68.6

[1] 1222 km^2 (Ries 1989).

[2] October flux, average year (Fig. 4).

[3] Excess = Annual flux - (October flux x 12), average year.

[*] Calculated using deposition measurements from lower Delaware Bay (Church et al. 1984) pro-rated to average year rainfall for Narragansett Bay.

(Table 18), it seems probable that other sources are also important in contributing to the metals carried by the Blackstone. As mentioned earlier, highway and parking lot runoff may be major sources of Pb and Zn (Hoffman and Quinn 1984), but used crankcase oil and gasoline do not contain large concentrations of Cd, Cu, or Mn (Latimer 1984).

While the major source of Mn in the Blackstone, as in the Delaware, appears to be natural weathering of the rocks in the watershed (Graham et al. 1976), additional anthro-pogenic sources contribute significantly to the As, Cd, Cr, Cu, and Ni carried by the Blackstone. These sources could enrich storm water surface runoff through processes not involving atmospheric deposition or automobiles, though it is not clear how this might be achieved. It is also possible that there may be significant flows of metal-enriched groundwater into the Blackstone. Latimer et al. (1988) found very much elevated concentrations of Cd, Cu, and Ni in groundwater samples near a sanitary landfill in Cranston, R.I., and it seems likely that similar sites could be found along the Blackstone. Given the evidence available thus far, it would seem important to make some further assessment of the impact of industrial and urban landfills on the metal budgets of the Blackstone and other rivers entering Narragansett Bay. It is also important to obtain direct measurements of the metal fluxes from the atmosphere at several locations in the bay watershed rather than relying on calculated deposition rates from a site on the lower bay. It is possible that atmospheric deposition may be the largest source of metals entering Narragansett Bay through the Blackstone River.

Before leaving the subject of rivers, it is worth noting that, within the range of flows covered by the data, there is no evidence of stream bed erosion. While the slopes of the particulate metal flux regressions tend to be steeper than for dissolved metals (Tables 7 and 15), there were no clear "steps" or breaks in the data indicating that threshold current speeds necessary for sediment erosion and resuspension had been attained (Figs. 2, 3, 7). It is certainly possible that river bed and bank erosion occur during extreme flows, but it is difficult to assess the contributions of such events to the inventory of metal inputs without additional study.

Inputs from Direct Industrial Discharges

With the possible exception of three electric power generating facilities, there appear to be no discharges of industrial waste waters directly to Narragansett Bay that are of consequence in the inventory of metal inputs. There are numerous direct discharges to the tributaries that enter the bay, but they contribute Cu, Pb, and Zn loads that are small relative to the metals entering the bay from publicly owned sewage treatment plants

(POTWs) and urban runoff (Hoffman and Quinn 1984, Hoffman 1987). Direct industrial discharges may contribute a significant portion of the Ag (Hoffman and Quinn 1984) as well as Cr, Cu, and Pb carried by the fresh waters entering the bay (Table 19), but these inputs are included in the calculated loadings from the rivers and streams. The great majority of industrial discharges will be included in the next section of this report that covers the assessment of metal releases from publicly owned sewage treatment plants.

As noted by Hoffman (1987), the large flows of cooling water used by electric power generating plants on the Providence River and in Mt. Hope Bay could provide a large input of metals even if there is only a modest increase in metal concentrations as water passes through the plants. In most cases, however, it is impossible to evaluate the addition because the concentrations of metals in the influent water are not measured or reported. For the largest power plant discharging into Mt. Hope Bay, Hoffman and Quinn (1984) appear to have obtained data on the actual increase in some metals (Hoffman 1987), and their work suggests that the plant may add almost as much Ni to Mt. Hope Bay as the input from the Taunton River (Table 20).

Publicly Owned Sewage Treatment Plants

There are 32 sewage treatment facilities that collect and discharge wastewaters within the Narragansett Bay drainage basin. Twenty-two of these discharge their effluents into rivers and streams, and their contribution to the total metal input to the bay has already been included in the calculation of metal loadings from rivers. Nine of the remaining plants discharge directly into the tidal waters of Narragansett Bay proper, and one discharges into Mt. Hope Bay (Table 21, Fig. 1).

Since the mid 1970s, treatment plants have been required to report the concentrations of various metals in their effluents, and it is often assumed that these data provide adequate information for calculating their discharges of metals to the bay. Unfortunately, this is not the case.

In a recent discussion of metal inputs from sewage treatment plants, Brubaker and Byrne (1989) correctly concluded that ". . . it is impossible to determine the level of discharge to Narragansett Bay." There are several contributing factors. Since metal loadings arise from the activities of numerous industries and manufacturing plants, there is no simple flux-flow relationship for POTW effluents as found for river inputs, and the concentrations of metals vary from day to day. For example, composite daily samples of the effluent from the Fields Point treatment plant, the largest that discharges to the bay, showed the following concentrations of metals on three consecutive days (SPRAY data, Doering et al. 1988):

Table 19

Comparison of the calculated inputs of various metals to Narragansett Bay (including Mt. Hope Bay) from rivers and streams with the direct discharge of metals reported by industries in the watershed.

Discharges	Estimated Input from Rivers and Streams[1]	Direct Industrial in the watershed[2]
	metric tons y^{-1}	
Cd	2.7	0.09
Cr	>5.4	13
Cu	25	15
Ni	27	0.04
Pb	14	8.0
Zn	<136	7.3

[1] Sum of Tables 13 and 14 for "average year".

[2] Hoffman (1987) for 1983. Data from R. I. and Mass., not including power plants.

Table 20

Comparison of some estimated inputs of metals to Mt. Hope Bay.
(49.2 km^2, Pilson 1985)

	Electric Power Plant Cooling Water[1]	Taunton River[2]	Direct Atmospheric Deposition[3]
	metric tons y^{-1}		
Cu	0.17	4.0	0.14
Ni	1.3	1.6	0.08
Zn	0.29	--	0.54

[1] Hoffman and Quinn (1984).

[2] From Table 14, average year.

[3] From Table 1, average year.

Table 21

Volume of effluent discharged directly into Narragansett Bay by publicly owned sewage treatment plants. Data for R. I. plants from R. I. Department of Environmental Management, RIPDES files.

	1986 Average Daily Flows	
	10^3 m^3 d^{-1}	Percent
Seekonk River		
Blackstone Valley District Commission (Bucklin Point)	78	22
Providence River		
Narragansett Bay Commission (Fields Point)	195	56
East Providence	22	6
West Passage		
East Greenwich	3	<1
Quonset	3	<1
East Passage		
Warren	6	2
Bristol	8	2
Jamestown	2	<1
Newport	32	9
Mt. Hope Bay[1]		
Fall River	73	21
	349	

---

[1] From Hoffman (1987).

		mg m^{-3}			
		Cd	Cu	Ni	Pb
8	March, 1987	2.15	118	249	11.3
9	March	2.59	309	300	19.9
10	March	0.70	281	255	20.2
9	August, 1987	2.33	283	138	41.1
10	August	1.10	167	77	28.8
11	August	1.32	178	357	23.3

In three-day sequential sampling on six occasions throughout an annual cycle at three treatment plants, coefficients of variation for concentrations of Cd, Cu, Pb, and Ni in effluent ranged from 35% to 67% (Table 22). Most smaller treatment plants are sampled for metals only a few times each year, and not all of the pollutant metals of interest are included in the analyses. Moreover, even when an analysis is reported, the results frequently appear as "less than" the allowable discharge concentration or "less than" the lowest detection limit of the analytical facility. While this type of report may be adequate for enforcement monitoring, it is not very useful for calculating mass loadings of metals or other pollutants.

Even for the Narragansett Bay Commission plant at Fields Point, a facility that is monitored frequently (weekly-monthly) with respect to metal discharge by an excellent "in house" analytical laboratory, there are difficulties in assessing the actual flux of metals to the bay. As pointed out by Brubaker and Byrne (1989), the standard data set on metals in sewage treatment plant effluents maintained by the R. I. Department of Environmental Management is the R. I. Pollution Discharge Elimination System (RIPDES) file, a continuation of the older National Pollution Discharge Elimination System (NPDES). For the Fields Point treatment plant and a few others that measure their effluent frequently, the discharge monitoring report (DMR) entered in the RIPDES file for each month contains only the highest concentration observed during that month. Moreover, flow data are not reported for the time during which the DMR concentrations are measured, and fluxes are calculated from the average daily flow for each month (Brubaker and Byrne 1989). Again, this may be appropriate for permit enforcement, but the practice almost certainly leads to a significant overestimate of the metals actually released from the treatment plants.

I have used the SPRAY data collected by Doering et al. (1988) on metal concentra-tions in composite daily samples of effluent collected on 18 occasions over an annual cycle in 1986-1987 from the three largest treatment plants that discharge directly into Narragansett Bay proper to calculate estimates of the mean concentrations of Cd, Cu, Pb, and Zn in each

Table 22

Concentrations of some metals in effluents from the Narragansett Bay Commission sewage treatment plant at Fields Point, the Blackstone Valley District Commission plant at Bucklin Point, and the East Providence treatment plant. Daily composite samples were analyzed for three consecutive days at six different times during 1986 and 1987 (Doering et al. 1988).

	Total Concentration (# of samples) mean ± 1 S.D., µg kg^{-1}			
	Cd	Cu	Pb	Ni
Fields Point	(18) 1.9 ± 0.7	(18)[a] 327 ± 205	(18) 26 ± 9.2	(18) 279 ± 149
		(15) 247 ± 101		
c.v., %	37	41-63	35	53
Bucklin Point	(5) 1.6± 0.7	(17) 52 ± 22	(17) 21 ± 5.9	(17) 96 ± 49
c.v., %	44	42	28	51
East Providence	(5) 0.7 ± 0.4	(16) 33 ± 21	(17) 10 ± 6.7	(18) 92 ± 40
c.v., %	57	64	67	43

% Dissolved (< 0.4 µm)

	Cd	Cu	Pb	Ni
Fields Point	67 ± 15	67 ± 10	25 ± 18	85 ± 7
		69 ± 10		
Bucklin Point	60 ± 37	44 ± 17	43 ± 27	92 ± 4
East Providence	68 ± 18	79 ± 26	74 ± 23	93 ± 5

[a] Samples collected on 24, 25, 26 July, 1987 had very high Cu concentrations of 700-770 µg kg^{-1}. This sampling was scheduled in an attempt to coincide with anticipated cleaning of electroplating tanks by the jewelry industry. The lower value based on 15 samples excludes these data.

of the effluents (Table 22). Multiplying these means by the average daily flow of effluent from each treatment plant for 1986 (Table 21) provides estimates of metal fluxes that are considerably lower than those calculated from the RIPDES files (Table 23). The discrepancy is large and particularly serious for the Fields Point plant, since it is such a major source of many metals. And the discrepancy, at least for Fields Point, must be due to differences in sampling and reporting rather than to analytical problems, because the Fields Point laboratory analyzed the SPRAY samples from the treatment plant (Doering et al. 1988). Unfortunately, the SPRAY program did not include other metals, such as Ag, Cr, Zn, or Hg, that are reported for these plants in the RIPDES files.

As in the rivers, the SPRAY data show that most of the Ni from treatment plant effluents is in dissolved form (Table 22). Copper was also similar, except at the Bucklin Point plant, where particulate Cu was more abundant than in the other effluents or in the rivers. Cadmium was also somewhat more particle-bound in the sewage effluents, but Pb tended to be a bit more soluble than in the rivers (compare Tables 11 and 12 with Table 22).

The problem of assessing the metal inputs from treatment plants is also compounded by the fact that the discharges from many, perhaps all, of the plants have been changing as a result of improvements in plant operations and/or the implementation of industrial pretreat-ment programs that have reduced the flows of metals into the plants (Brubaker 1986, Brubaker and Byrne 1989). While pretreatment programs were to be fully implemented by 1984, there have been numerous delays and problems in enforcement (Brubaker and Byrne 1989, Sutinen and Lee 1990). Nevertheless, the Narragansett Bay Commission reports that between 1981 and 1989 the following reductions in metal flows to the Fields Point treatment plant were achieved:

Ag — 81%
Ni — 84%
Cd — 87%
Cr — 90%
Cu — 95%
Pb — 81%
Zn — 72%
Hg — 35%

With the exception of Zn and Hg, the fluxes of metals from the Fields Point facility to the bay, as calculated from RIPDES data, showed equivalent or only somewhat smaller (˜10%) declines when comparing the same years. For unknown reasons, the export of Zn from the plant declined only 49% and Hg increased 32% in 1989 compared to 1981.

The decline in metal discharges from Fields Point has not been consistent, though the overall reduction in the discharge of Cu, Ni, Zn and Pb since 1982 has been dramatic (Fig. 9). Chromium and silver discharges remained relatively low, if erratic, after 1982, while Cd appears to have declined sharply only in 1989 (Fig. 9).

Table 23

Comparison of metal inputs from publicly owned sewage treatment plants discharging into the Seekonk and Providence Rivers as reported in RIPDES files for 1986 and 1987 and as calcuated using concentration measurements from the 1986-1987 SPRAY program (Doering et al. 1988) and mean flows from RIPDES files for 1986. Numbers of samples are shown in ( ). Values are metric tons per year ± 1 S.D.

	Cd	Cu	Ni	Pb
Narragansett Bay Commission, Fields Point				
RIPDES 1986 (12)	0.62 ± 0.60	36.9 ± 17.0	42.5 ± 18.0	6.1 ± 4.8
RIPDES 1987 (12)	0.66 ± 0.45	33.8 ± 15.9	37.8 ± 13.9	5.7 ± 3.1
SPRAY 1986-1987 (18)	0.14 ± 0.05	23.3 ± 14.6	19.9 ± 10.6	1.8 ± 0.6
		17.6 ± 7.2*		
Blackstone Valley Commission, Bucklin Point				
RIPDES 1986 (4)		3.13 ± 0.84	5.89 ± 2.39	
RIPDES 1987 (4)		3.55 ± 3.30	5.87 ± 3.18	
SPRAY 1986-1987 (16,18)	1.49 ± 0.63	2.74 ± 1.40		
East Providence				
RIPDES 1986 (7)	0.01 ± 0.01	0.21 ± 0.06	0.41 ± 0.08	0.08 ± 0.04
RIPDES 1987 (12)	0.02 ± 0.02	0.25 ± 0.15	0.91 ± 0.32	0.13 ± 0.09
SPRAY 1986-1987 (5,16,18,17)	0.006± 0.003	0.27 ± 0.17	0.72 ± 0.32	0.08 ± 0.05

*Does not include samples from July 1987. The higher SPRAY value should be compared with the RIPDES data.

Some of the earliest metals discharge data for the Fields Point plant were summarized by Olsen and Lee (1979) from R. I. Department of Environmental Management permit and "self-monitoring files", presumably for the period 1976-1978. They show discharges of:

Cu = 88.6 t y^{-1}
Cr = 13.6 "
Ni = 80 "
Zn = 140 "

If these values are correct, there were already significant declines in the amounts of Cr, and especially Zn, released from Fields Point by 1981, though Cu and Ni showed little change between about 1977 and 1981 (Fig. 9). While the amount of lead in the Fields Point effluent declined by about 80% between 1981 and 1989 (Fig. 9), concentrations of lead in the air in Providence in 1981 had already declined by about 50% from their value in 1978, three years after unleaded gasoline was required for new automobiles (R. I. Department of Health, annual reports). However, since storm water runoff contributed only about 9% to the lead discharged by the plant, at least during 1982 (Hoffman et al. 1988), it does not seem that the removal of lead from gasoline would necessarily have produced a marked drop in lead entering and leaving the treatment plant.

It is also likely that changing economic conditions have played a role in reducing the amounts of metals reaching the treatment plants. With the exception of costume jewelry, the value added by manufacturing in other major metal consuming industries in Rhode Island declined during the late 1970s and early 1980s (Section I).

Because of the dramatic declines in many of the metals discharged by the treatment plants, their contribution to the total input of metals to the bay will depend on the time when the inventory is taken. I have chosen to use the mean discharges during 1986 and 1987 because data on metal concentrations in the effluents from all of the treatment plants dis-charging directly to the bay were analyzed on four occasions during 1986 by the SINBADD program (Pilson and Hunt 1989) and the effluents from the three plants discharging directly to the Seekonk and Providence Rivers were also analyzed on 18 days in 1986 and 1987 by the SPRAY program (Doering et al. 1988). As a practical matter, however, I have had to supplement the 1986-1987 data with some earlier measurements in order to assemble even the imperfect inventory summarized in Table 24. In some cases, such as Cd, there were no data reported in the RIPDES file for the Bucklin Point plant in 1986 or 1987, and the flux for 1985 was anomalously high (2.4 t y^{-1}). It seemed more representative to report the mean flux for 1980 through 1984. Similarly, the RIPDES Cr flux from Bucklin Point was very large in 1986 (12 t y^{-1}), and

I again calculated a mean for 1980 through 1984. Lead data have not been reported for Bucklin Point since 1984.

The most striking feature of the inventory of direct metal fluxes from treatment plants is the dominance of one source. In 1986-1987, the Narragansett Bay Commission facility at Fields Point accounted for the following proportions of the metals released from treatment plants directly into the bay:

Ag — 45%
Cd — 50%
Cr — 51%
Cu — 88%
Ni — 85%
Pb — 51%
Zn — 93%

Except for Cd (which declines to 28%), these proportions are roughly the same regardless of whether the RIPDES or SPRAY data are used to calculate the fluxes (Table 24).

Urban Runoff

There is probably no item in the inventory of pollutant inputs to the bay that has aroused more recent concern or been surrounded by more confusion than urban storm water runoff. Much of the concern is due to an intensive study of this source of pollution to Narragansett Bay that was carried out by Hoffman and Quinn (1984) between 1980 and 1984. Their measurements and calculations suggested that urban runoff could provide about 48% of the petroleum hydrocarbons, 44% of the high molecular weight PAHs, 56% of the Zn, and 65% of the Pb entering "waters of the Narragansett Bay *basin*." I have added emphasis to the term "basin" because their assessment was based on multiplying measured fluxes of each pollutant per unit area from each of four specific land use types by the total amount of each land use type in the watershed of the bay and summing the results. This was a very reasonable and practical approach to the problem, but it is important to remember that the subject of their inventory was the entire drainage basin of the bay, including tributaries, and not just the tidal salt waters of Narragansett Bay itself. For this reason, their assessment of industrial discharges and sewage treatment plants also included outfalls located along the tributaries to the bay as well as those emptying directly into the bay. Rivers were not considered as inputs, but as unquantified transfers within the larger system of interest. The Hoffman and Quinn (1984) study did not address the question of how much of the pollutants potentially generated by urban runoff actually reached the various tributaries or how much of the pollutants entering the tributaries from runoff and point source discharges actually reached Narragansett Bay.

The results of some more recent comparisons between metal inputs to the Blackstone and Pawtuxet Rivers and the fluxes of metals from these systems to the Seekonk and Providence River portions of Narragansett Bay have been reported by Quinn et al. (1987) and Latimer et al. (1988), respectively. During three low-flow studies, the masses of Cd, Cr, Cu, Ni, and Ag discharged by the Blackstone River at Pawtucket, R. I. averaged between about 40% (Cr) and 70% (Ni) of the measured inputs (including flow from upstream) between the Rhode Island—Massachusetts state line and the dam at Pawtucket. Virtually all of the Pb was exported. This contrasts markedly with the low-flow mass balance of the Pawtuxet River, where the export of Cd, Cr, Cu, Pb, and Ni averaged three (Ni) to 100 (Cr) times greater than the measured inputs on the three occasions studied by Latimer et al. (1988).

Until more extensive mass balance studies on the watersheds and rivers have been completed during wet weather conditions, it will be difficult to know how large a contribution urban runoff actually makes to the input of metals to the bay. By including the measured fluxes of metals at the mouths of the larger rivers and extrapolating those fluxes on a flow-weighted basis to "all" the fresh water entering the bay, it seems likely that much of the influence of urban runoff has been captured in this inventory. That there is some additional metals input due to runoff is evident from the steeper slopes of the river flux vs. flow regressions for Cu and Pb derived from Wright's storm measurements (Table 15 compared to Table 7), but the storm regressions produce only a modest increase in Cu even if they are applied to an entire year of flow measurements (Table 16). The 2.6 fold higher annual Pb flux computed for the Blackstone River in an average flow year using the Wright storm regression (Table 16) is not surprising considering the large predicted flux of lead in urban runoff, and a similar increase might have been found for Zn if that metal had been measured in the wet weather sampling (Hoffman and Quinn 1984). Since the storm regression would apply for only a fraction of the year, however, the actual increase in Pb and (perhaps) Zn above that predicted using the SINBADD—SPRAY regressions would be considerably less than suggested in Table 16. At this point, I think the most prudent course is to identify urban runoff as an unquantified source that may be found to make a significant additional contribution to the inputs of Pb, Zn, and perhaps some other metals.

The finding that metal fluxes from the rivers increase with flow (Figs. 2 and 3) certainly indicates that non-point sources may be very important in contributing to the total amounts of various metals carried into Narragansett Bay. In fact, as shown for the Blackstone River in Table 18, these sources appear to be larger than the point sources that maintain the base flow concentrations of metals.

Rhode Island Sound

Narragansett Bay is characterized by strong gradients in the concentrations of most of the metals that have been examined, from high values in the water, sediments, and organisms of the Providence River and upper bay to low values near the mouth (Bender et al. 1989). Because of these gradients and the large fluxes of metals that enter the upper bay, we do not normally think of the relatively unpolluted waters offshore as a potential additional source. Nevertheless, the waters of Rhode Island Sound contain dissolved and particulate metals that are carried into the bay twice each day with the flooding tide. The behavior and fate of these metals once they enter the bay are not known, but even if most or all of them are exported during the ebb tide, the Sound contributes to the total inventory of metal inputs.

No attempt has ever been made to obtain a direct measure of the flux of metals into (or out of) any of the passages of Narragansett Bay, and it would probably not be practical to do so. A very rough approximation of the flux due to tidal pumping can be obtained by multiplying the mean tidal volume that enters the bay by the metal concentrations in the bottom waters of Rhode Island Sound. As part of their hypsographic analysis of the bay, Chinman and Nixon (1985) computed the volume at mean high and mean low tide. The difference between the two is equal to 390×10^6 m^3 or, at two tides per day, a flow of 284.7×10^9 m^3 y^{-1}. This is about 90 times the annual input of fresh water (Pilson 1985). The tidal prism of the Sakonnet River (Fig. 1) accounts for 14% of the total, leaving 244.8×10^9 m^3 y^{-1} of tidal flow in the East and West Passages of Narragansett Bay proper, assuming that little of the tide enters Mt. Hope Bay through the Sakonnet passage.

Unfortunately, there are few measurements of metals in Rhode Island Sound. The SINBADD program (Pilson and Hunt 1989) did collect offshore samples in October and November of 1985 and April and May of 1986, but total metals from these stations were only analyzed for the first two cruises. Chromium was only measured on the first cruise and only the dissolved fraction was reported. Based on the data that are available from the bottom waters at SINBADD stations 21 and 22, I have assumed offshore total metal concentrations of:

Ag = 3 µg m^{-3}
Cd = 20 "
Cr = 110 "
Cu = 350 "
Ni = 415 "
Pb = 130 "

When combined with the tidal flow volume, these concentrations produce fluxes into the bay of:

Ag = 0.73 metric tons y^{-1}
Cd = 4.9 " "
Cr = 27 " "
Cu = 86 " "
Ni = 100 " "
Pb = 32 " "

These values are larger than all of the other metal inputs combined, and virtually all of the reviewers of an earlier draft of this report objected to them on the grounds that much, most, or all of the metals entering the bay from offshore with the tide are exported again on the ebb. Some reviewers thought metals entering from offshore would not react or accumulate in the bay, and therefore should not be included in the inventory of inputs, though they did not subject metals from anthropogenic activities or land drainage to this same standard. Regardless of their fate or effect, however, it is inescapable that large amounts of metals enter the bay with the tidal prism and, in the strictest sense, they are part of a complete accounting of metal inputs.

It is also appropriate (perhaps more appropriate) to take a more conservative approach to evaluating the offshore source by estimating the mass of each metal associated with the volume of offshore bottom water actually involved in circulation exchanges in the bay. This approach was used earlier to calculate the input of nutrients from offshore (Nixon and Pilson 1984), and recent calculations of the flushing rate of the bay as a function of fresh water input make it easy to arrive at a first-order estimate.

For the period between April 1972 and August 1973, Officer and Kester (1991) report an average flushing rate of 1512 m^{3} s^{-1}. Subtracting the mean river flow during the same period of 150 m^{3} s^{-1} gives an average sea water exchange of 1362 m^{3} s^{-1}. Multiplying this flow by the bottom water metal concentrations used previously gives a flux into the bay of:

Ag = 0.13 metric tons y^{-1}
Cd = 0.86 " "
Cr = 4.7 " "
Cu = 15 " "

Ni = 18 " "
Pb = 5.6 " "

These much lower estimates of the potential input from offshore are, with the exception of Cd, still large and comparable to the loadings from rivers. However, it also remains true that we know little about the fate or effects of these metals, and they may, for the most part, return to the sea.

Summary of Metal Inputs

Having emphasized the uncertainty in each of the terms of this inventory, I am a bit uncomfortable with the notion of an overall summary, a final accounting that adds up the result. Impatient readers may turn to such a table without "wasting time" on the details, and the numbers in summary tables begin to assume a look of certainty that belies their often ambiguous origins. Equivocating academics are not much help, however, to those who must get on with the job of issuing permits and planning for the future. How much copper comes into Narragansett Bay? There is an "answer" in Table 25. For Mt. Hope Bay, the "answer" is in Table 26. But, at best, the answers are only first approximations with an undefined uncertainty around them.

On the other hand, some findings seem reasonably clear. For example, the input of all the metals examined is larger in wet years than in dry, and the fluxes of most metals from sewage treatment plants have declined markedly in recent years. With the exception of lead, direct deposition from the atmosphere onto the surface of the bay does not contribute much to the metal inputs (Table 27), though the atmosphere may be an important source for many metals carried into the bay by rivers. Most of the cadmium entering the bay from land comes from sources upstream along the tributaries, rather than from effluents discharged directly into the bay, while almost all of the silver comes from the Bucklin Point and Fields Point sewage treatment plants (Tables 25 and 27). For lead and zinc, the two metals thought to be carried in large quantity by urban runoff, land drainage appears to provide more of the total input than direct sewage discharge, while direct and upstream sources were about equally important in supplying chromium, copper, and nickle. Marked reductions in the discharge of these metals, as well as zinc, from the Fields Point treatment plant have almost certainly now made rivers and upstream sources the major input of all the metals considered here, except perhaps silver. Because of the large contribution that runoff and (perhaps) unidentified runoff-related point sources make to the metals carried by the rivers (Table 18), reductions in the discharge of metals by upstream POTWs may achieve only a modest reduction in the metal loading to the bay.

Because Providence and the Blackstone River Valley have had a long and continuing history of intensive industrialization (Section I), it is often assumed that this area is particularly polluted by metals and that the inputs of metals to Narragansett Bay are notably

Table 27

Relative contribution (%) of various sources to the input of metals to Narragansett Bay proper during 1985-1987. Values in ( ) include the estimated input from Rhode Island Sound. Data from Table 25.

	Cd	Cr	Cu	Ni	Pb	Zn	Ag
Atmospheric Depositon	1.7 (1.3)	1.1 (0.7)	1.6 (1.2)	0.9 (0.7)	33 (24)	1.5	?
Rivers and Upstream Sources	81 (63)	51 (34)	44 (34)	50 (37)	45 (33)	69	9 (9)
Direct Sewage Discharge	17 (13 (89)	48 (32)	54 (41)	49 (36)	22 (16)	30	91
Offshore	(23) (3)	(34)	(24)	(27)	(27)	?	

large, even when compared to other urban coastal waters. It is difficult to know if this is true because few detailed inventories have been prepared for bays and estuaries. The small sample of comparative input data that I have been able to find does not isolate Narragansett Bay, at least at the levels of metal input characteristic of the mid 1980s (Table 28). In the recent past, however, the situation must have been quite different. During the 1950s, the deposition of lead and zinc from the atmosphere onto the bay may have exceeded or roughly equaled the total present input from the atmosphere, rivers, and sewage (Tables 4 and 25). The discharge of copper to the bay from the Fields Point treatment plant was probably about 15 times larger in 1950 than it is today (Table 29).

Remarkable reductions in the inputs of metals to Narragansett Bay have been achieved over the past 40 years, and especially over the past 8-10 years. The evidence for these reductions is in the sediments (Section I) and in the monitoring reports of the Department of Environmental Management. The reductions have come about as a result of air and water pollution control legislation as well as changes in the economy of the bay watershed (Section I for an assessment up to about 1975). Among the most important factors have been the shift from wood and coal to oil and natural gas as fuels of choice (Section I, Figs. 19, 21 and 23), the development and installation of devices to reduce stack emissions to the atmosphere during the 1950s and 1960s (Section I, Figs. 23 and 24), the removal of lead from gasoline in 1974, the termination of sewage sludge dumping in the bay in 1950 (Section I, Fig. 14), the initiation of industrial and municipal discharge permits under the NPDES section of the 1972 Clean Water Act, the construction and upgrading of sewage treatment plants during the 1950s, 1960s, and 1970s, the implementation during the early 1980s of the National Pretreatment Program, and, finally, the loss of primary metals manufacturing industry since the late 1960s (Section I, Fig. 10).

It is also true, of course, that legislation has no effect if it is is not enforced and that sewage treatment plants only function as well as they are staffed and maintained. The growing strength and scientific sophistication of Save the Bay and other environmental groups, the professionalism of the Department of Environmental Management and the Narragansett Bay Commission, and the cooperation of enlightened metal industries have all contributed to bringing Narragansett Bay back to the point where the direct anthropogenic discharge of most metals to the bay is now less than the input from the coastal ocean (Table 25). For metals such as copper, chromium, and lead, the current inputs are almost certainly lower than they have been for over a century.

Table 28

Comparison of estimated volume specific inputs (mg m^{-3} y^{-1}) of various metals to different estuarine and coastal marine systems.

	Fe	Mn	Cd	Cr	Cu	Pb	Ni	Zn
Narragansett Bay[1]	>740	>117	<1.4	>4.3	~21	<6.7	<23	~92
Mt. Hope Bay[1]			<3	<6.5	<25	<55	<12	
Chesapeake Bay[2]	2748		3.6	13	11	9.1		38
Delaware Bay[3]	2755	510	0.6		7.5		9	37
Hudson Estuary[4]			16	210	352	290	175	972
San Francicso Bay[5]				~10	~14	~13	~9.3	~31
Baltic Sea[6]	17		0.02		0.14	0.27	0.13	1.2

[1] This report, volumes from Chinman and Nixon (1985), inputs from atmospheric deposition, rivers, and POTWs.

[2] Bieri et al. (1982) data for 1980 from atmospheric deposition, rivers, and POTWs. Volume from Cronin (1971).

[3] Church (1986) data for 1981-1982. Inputs do not appear to include direct industrial discharges or POTWs. Volume from NOAA (1985).

[4] Muller et al. (1982) data for 1975-1980 for Hudson-Raritan system, including atmospheric deposition, rivers, POTWs, urban runoff, and landfill leachate.

[5] Aquatic Habitat Institute (unpublished) preliminary assessment for inputs from atmospheric deposition, rivers, industry, and POTWs. There are also large potential inputs from urban and non-urban runoff not included in the values reported here. Volume from NOAA (1985).

[6] Brügmann (1986) data for 1980-1983. Inputs from atmospheric deposition, rivers, POTWs, industry, and sediments.

Table 29

Comparison of the estimated historical and recent inputs of various metals to Narragansett Bay from the sewage treatment plant at Fields Point.
Values are metric tons per year.

	Year					
	1900[a]	1925[a]	1950[a]	1986[b]	1988[b]	1993[c]
Cd	<1.3	<1.4	1.9	0.6 (0.14)	0.6	<0.1
Cr	13	17	22	2.3	1.2	1.4
Cu	54	76	104	35 (18-23)	6.8	6.4
Pb	15	16	22	5.9 (1.8)	3.3	0.9
Ni	<31	<51	71	40 (20)	23	15
Zn	90	125	171	54	25	6.6
Ag				1.8		0.4

[a] From Section I, Table 20.

[b] From Table 24. Values in ( ) are from SPRAY concentration measurements; others are from RIPDES files.

[c] RIPDES files reported by Save the Bay (1993) Wastewater Treatment Plant Performance Survey. These values are the mean of samples collected approximately weekly.

ACKNOWLEDGMENTS

This review was made possible by support from the Narragansett Bay Project (a joint effort of the R. I. Department of Environmental Management and the U. S. Environmental Protection Agency) and the R. I. Sea Grant College Program (NOAA). Steve Granger and Andrew Milliken assisted greatly with data analysis and the production of graphics. Carlton Hunt, Jim Quinn, Ken Rahn, Jon Sutinen, and Ray Wright made unpublished data available to me, and I thank them. I also acknowledge, with much appreciation, the stimulating conversations concerning metal inputs to the bay that I have had with Suzanne Bricker, Caroline Karp, John King, Virginia Lee, Michael Pilson, Jim Quinn,and Ken Rahn. A number of others have also helped during various phases of this project, including Charles Roman and Jenny Martin of the Narragansett Bay Project, Connie Carey and Chris Deacutis at the R. I. Department of Environmental Management, Peter Doering at the Marine Ecosystems Research Laboratory, and Alan Desbonnet at the Coastal Resources Center. Dolores Smith produced the final manuscript and assisted with editing.

REFERENCES

Aquatic Habitat Institute. Undated. Internal draft. Status and trends report on pollutants in the San Francisco Bay estuary. Unpublished draft report to the San Francisco Estuary Project.

Bender, M., D. Kester, D. Cullen, J. Quinn, W. King, D. Phelps, and C. Hunt. 1989. Trace metal pollutants in Nasrragansett Bay waters, sediments and shellfish. Report to the Narragansett Bay Project, R. I. Dept. Environmental Management. 75 p. with appendices.

Bieri, R., O. Bricker, R. Byrne, R. Diaz, G. Helz, J. Hill, R. Huggett, R. Kerhin, M. Nichols, E. Reinharz, L. Schaffner, D. Wilding, and C. Strobel. 1982. Toxic substances, p. 263-375. In: *Chesapeake Bay Program Technical Studies: A Synthesis*. Washington, DC: U. S. Environmental Protection Agency.

Bierman, V. J., Jr., S. D. Preston, and S. E. Silliman. 1988. Development of estimation methods for tributary loading rates of toxic chemicals. Water Resources Research Center Tech. Rept. No. 183. Purdue University, Lafayette, IN.

Brubaker, K. L. 1986. Down the drain: Toxic pollution and the status of pretreatment in Rhode Island. A report from Save the Bay, Providence, RI.

Brubaker, K. L. and J. H. Byrne. 1989. Zero tolerance: Reducing toxic pollution in Narragansett Bay. A report from Save the Bay, Providence, RI.

Brügmann, L. 1986. The influence of coastal zone processes on mass balances for trace metals in the Baltic Sea. *Rapp. P.-v. Réun. Cons. int. Explor. Mer*. 186:329-342.

Chinman, R. A. and S. W. Nixon. 1985. Depth-area-volume relationships in Narragansett Bay. Mar. Tech. Report No. 87, The University of Rhode Island, Graduate School of Oceanography, Narragansett, RI. 64 pp.

Church, T. M. 1986. Biogeochemical factors influencing the residence time of micro-constituents in a large tidal estuary, Delaware Bay. *Marine Chemistry* 18:393-406.

Church, T. M., J. M. Tramontano, J. R. Scudlark, T. D. Jickells, J. J. Tokos, Jr., A. H. Knap, and J. N. Galloway. 1984. The wet deposition of trace metals to the western Atlantic Ocean at the mid-Atlantic coast and on Bermuda. *Atmospheric Environment* 18 (12):2657-2664.

Church, T. M., J. M. Tramontano, J. R. Scudlark, and, S. L. Murray. 1988. Trace metals in the waters of the Delaware Estuary, p. 93-115. In: S. K. Majumdar, E. W. Miller and L. E. Sage (eds.), *Ecology and Restoration of the Delaware River Basin***.** The Pennsylvania Academy of Science.

Cronin, W. B. 1971. Volumetric, areal, and tidal statistics of the Chesapeake Bay and its tributaries. Chesapeake Bay Institute, Special Report No. 20. Reference 71-2. The Johns Hopkins University, Baltimore, MD. 135 p.

Doering, P. H., L. Weber, W. M. Warren, G. Hoffman, K. Schweitzer, M. E. Q. Pilson, C. A. Oviatt, J. D. Cullen, and C. W. Brown. 1988. Monitoring of the Providence and Seekonk Rivers for trace metals and associated parameters. Data report on SPRAY cruises to the Narragansett Bay Project, R. I. Dept. of Environmental Management from the Marine Ecosystems Research Laboratory, Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Dolan, D. M., A. K. Yui, and R. D. Geist. 1981. Evaluation of river load estimation methods for total phosphorus. *J. Great Lakes Res.* 7(3):207-214.

Ferguson, R. I. 1986. River loads underestimated by rating curves. *Water Resources Research* 22(1):74-76.

Graham, W. F., M. L. Bender, and G. P. Klinkhammer. 1976. Manganese in Narragansett Bay. *Limnol. Oceanogr*. 21:665-673.

Hess, K. and F. White. 1974. A numerical tidal model of Narragansett Bay. URI Sea Grant Tech. Rept. No. 20. University of Rhode Island, Kingston, RI.

Hoffman, E. J. 1987. Pollution Inputs to Narragansett, p. 31-69. In: *Narragansett Bay: Issues, Resources, Status and Managemen*t. NOAA Estuary of the Month Seminar Series No. 1.

Hoffman, E. J., C. G. Carey, G. L. Mills and J. G. Quinn. 1988. Urban runoff inputs to a municipal wastewater treatment facility. Unpublished manuscript. Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Hoffman, E. J. and J. G. Quinn. 1984. Hydrocarbons and other pollutants in urban runoff and combined sewer overflows. Report submitted to NOAA, Oceans Assessments Division, September 1980-April 1984.

Hunt, C. D. 1982. Metal inputs to Upper Narragansett Bay via point and nonpoint sources. Unpublished report from the Marine Ecosystems Research Laboratory, Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Kremer, J. N. and S. W. Nixon. 1978. *A Coastal Marine Ecosystem: Simulation and Analysis*. Ecological Studies, Vol. 24. Springer-Verlag, Heidelberg. 210 pp.

Latimer, J. S. 1984. Characterization of the sources of hydrocarbons in urban runoff from relationships of organic distributions and metal content. M.S. Thesis, University of Rhode Island, Graduate School of Oceanography, Narragansett, RI.

Latimer, J. S., C. G. Carey, E. J. Hoffman, and J. G. Quinn. 1988. Water quality in the Pawtuxet River: Metal monitoring and geochemistry. *Water Resources Bull.* 24(4):791-800.

Lewis, M. F. and K. L. Brubaker. Undated. Bring Back the Blackstone. A report from Save the Bay, Providence, RI.

Mueller, J. A., T. A. Gerrish, and M. C. Casey. 1982. Contaminant inputs to the Hudson-Raritan Estuary. NOAA Tech. Memorandum OMPA-21. 192 p. + Appendices.

National Oceanic and Atmospheric Administration. 1985. *National Estuarine Inventory Data Atlas*. Volume 1: Physical and Hydrologic Characteristics. Washington, DC: U. S. Department of Commerce.

Nixon, S. W. 1990. A History of Metal Inputs to Narragansett Bay. Unpublished report submitted to the Narragansett Bay Project, R. I. Dept. of Environmental Management.

Nixon, S. W., C. D. Hunt, and B. L. Nowicki. 1986. The retention of nutrients (C, N, P), heavy metals (Mn, Cd, Pb, Cu), and petroleum hydrocarbons in Narragansett Bay, p. 99-122. In: J. M. Martin and P. Lasserre (eds.), *Biogeochemical Processes at the Land-Sea Boundary.* Amsterdam: Elsevier Press.

Nixon, S. W. and V. Lee. 1986. *Wetlands and Water Quality*. U.S. Army Corps of

Engineers, Technical Report Y-86-2.

Nixon, S. W. and M. E. Q. Pilson. 1984. Estuarine total system metabolism and organic exchange calculated from nutrient ratios: An example from Narragansett Bay, p. 261-290. In: V. S. Kennedy (ed.), *The Estuary as a Filter*. New York: Academic Press.

Officer, C. B. and D. R. Kester. 1991. On estimating the non-advective tidal exchanges and advective gravitational circulation exchanges in an estuary. *Est. Coast. Mar. Sci.* 32:99-103.

Olsen, S. and V. Lee. 1979. A summary and preliminary evaluation of data pertaining to water quality in upper Narragansett Bay. Coastal Resources Center, University of Rhode Island, Narragansett, RI. 189 p.

Olsen, S., D. D. Robadue, Jr., and V. Lee. 1980. *An Interpretive Atlas of Narragansett Bay*. Coastal Resources Center, Graduate School of Oceanography, University of Rhode Island, Marine Bulletin 40.

Pilson, M. E. Q. 1985. On the residence time of water in Narragansett Bay. *Estuaries* 8(1):2-14.

Pilson, M. E. Q. 1989. Aspects of Climate Around Narragansett Bay. Unpublished report Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Pilson, M. E. Q. and C. D. Hunt. 1989. Water quality survey of Narragansett Bay—A summary of results from the SINBADD cruises 1985-1986. Report to the Narragansett Bay Project, R. I. Dept. of Environmental Management from the Marine Ecosystems Research Laboratory, Graduate School of Oceanography, University of Rhode Island, Narragansett, RI.

Quinn, J. G., E. J. Hoffman, J. S. Latimer, and C. G. Carey. 1985. A study of the water quality of the Pawtuxet River: Chemical monitoring and computer modeling of pollutants. Volume 1: Chemical monitroing of pollutants in the Pawtuxet River. A report submitted to the Division of Water Resources, Dept. of Environmental Management, Providence, Rhode Island. 249 p.

Quinn, J. G., J. S. Latimer, C. G. Carey, J. T. Ellis, and J. Zheng. 1987. Development of a one dimensional water quality model for the Blackstone River: Part 1: Chemical monitoring of pollutants in the Blackstone River. Final report to the Narragansett Bay Project, R. I. Dept. of Environmental Management.

Rahn, K. A. 1990. Will emissions from new power plants in Rhode Island affect Narragansett Bay? *Maritimes* 34:1-5.

R. I. Department of Health. 1986. Air quality data summary. Issued annually by the Rhode Island Dept. of Environmental Management

Ries, K. G., III 1989. Estimation of surface-water runoff to Narragansett Bay, Rhode Island and Massachusetts. U. S. Geological Survey Water Resources Investigations Report, Boston, MA.

Santschi, P. H., Y.-H. Li, and S. Carson. 1980. The fate of trace metals in Narragansett Bay, Rhode Island: Radiotracer experiments in microcosms. *Estuarine and Coastal Marine Science* 10:635-654.

Santschi, P. H., S. Nixon, M. Pilson, and C. Hunt. 1984. Accumulation of sediments, trace metals (Pb, Cu) and total hydrocarbons in Narragansett Bay, Rhode Island. *Estuarine, Coastal and Shelf Science* 19(4):427-449.

Sutinen, J. G. and S.-G. Lee. 1990. Regulatory compliance and enforcement: Industrial wastewater pretreatment programs in Rhode Island. Dept. of Resource Economics, University of Rhode Island, Kingston, RI. A report submitted to the Narragansett Bay Project, R. I. Dept. of Environmental Management.

Wehe, T. J. 1953. Run-off data, p. 30-32. In: C. J. Fish (ed.), *Physical Oceanography of Narragansett Bay—Rhode Island Sound*. Inshore survey project. Final Report. URI GSO Unnumbered special reports.

Windom, H. L.. 1986. The importance of atmospherically transported trace metals to coastal marine waters in relation to river transport, p.194-202. In: *Estuarine Processes: An Application to the Tagus Estuary*. Lisbon, Portugal: National Commission of the Environment, Proceedings of a UNESCO/IOC/CNA workshop held in Lisbon, Portugal 13-16 December 1982.

Windom, H. L., J. T. Byrd, R. G. Smith, Jr., and F. Huan. 1991. Inadequacy of NASQAN data for assessing metal trends in the nation's rivers. *Environ. Sci. Technol.* 25(6):1137-1142.