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ARTICLE *in* ENVIRONMENTAL SCIENCE AND TECHNOLOGY · FEBRUARY 2004

Impact Factor: 5.33 · DOI: 10.1021/es040347s · Source: PubMed

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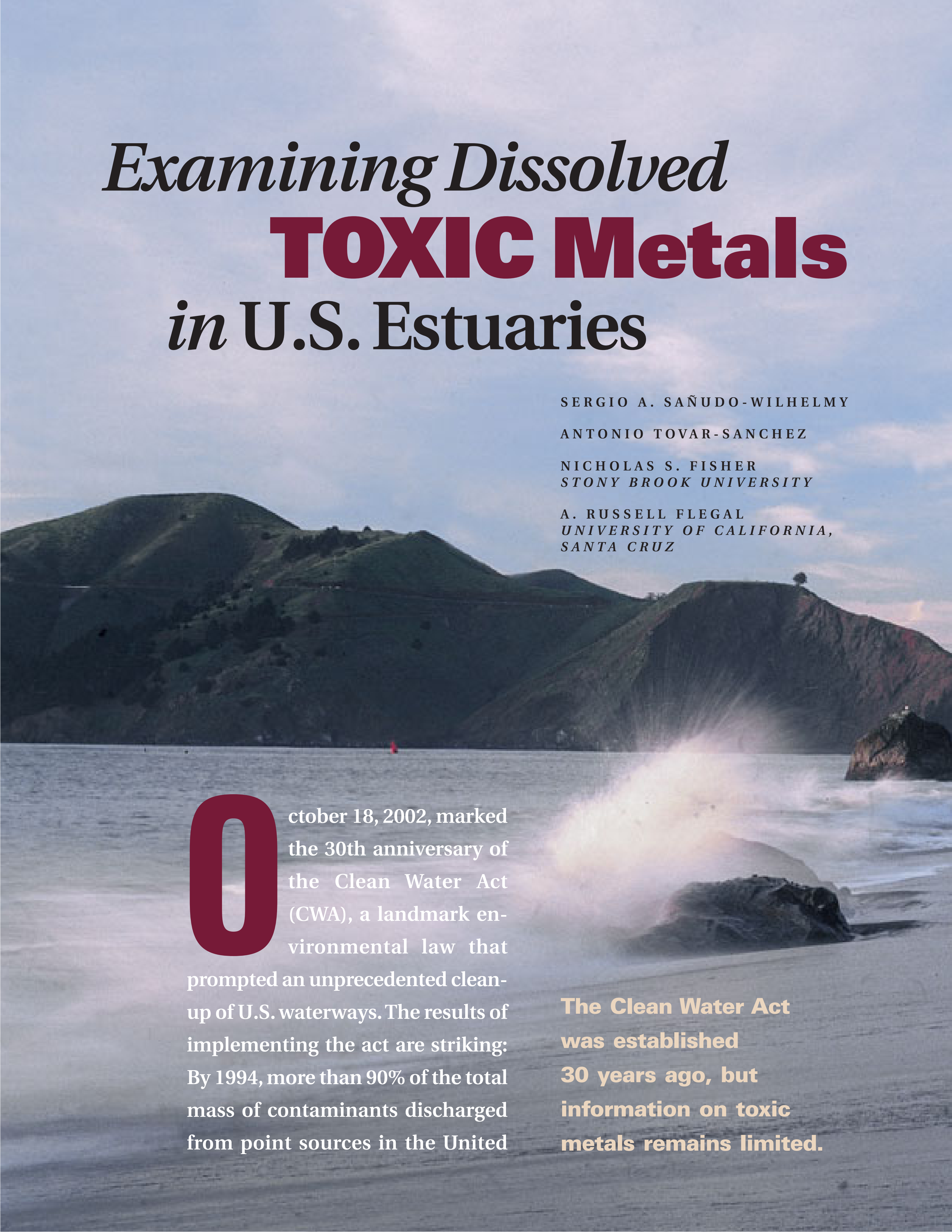

Examining Dissolved TOXIC Metals *in* U.S. Estuaries

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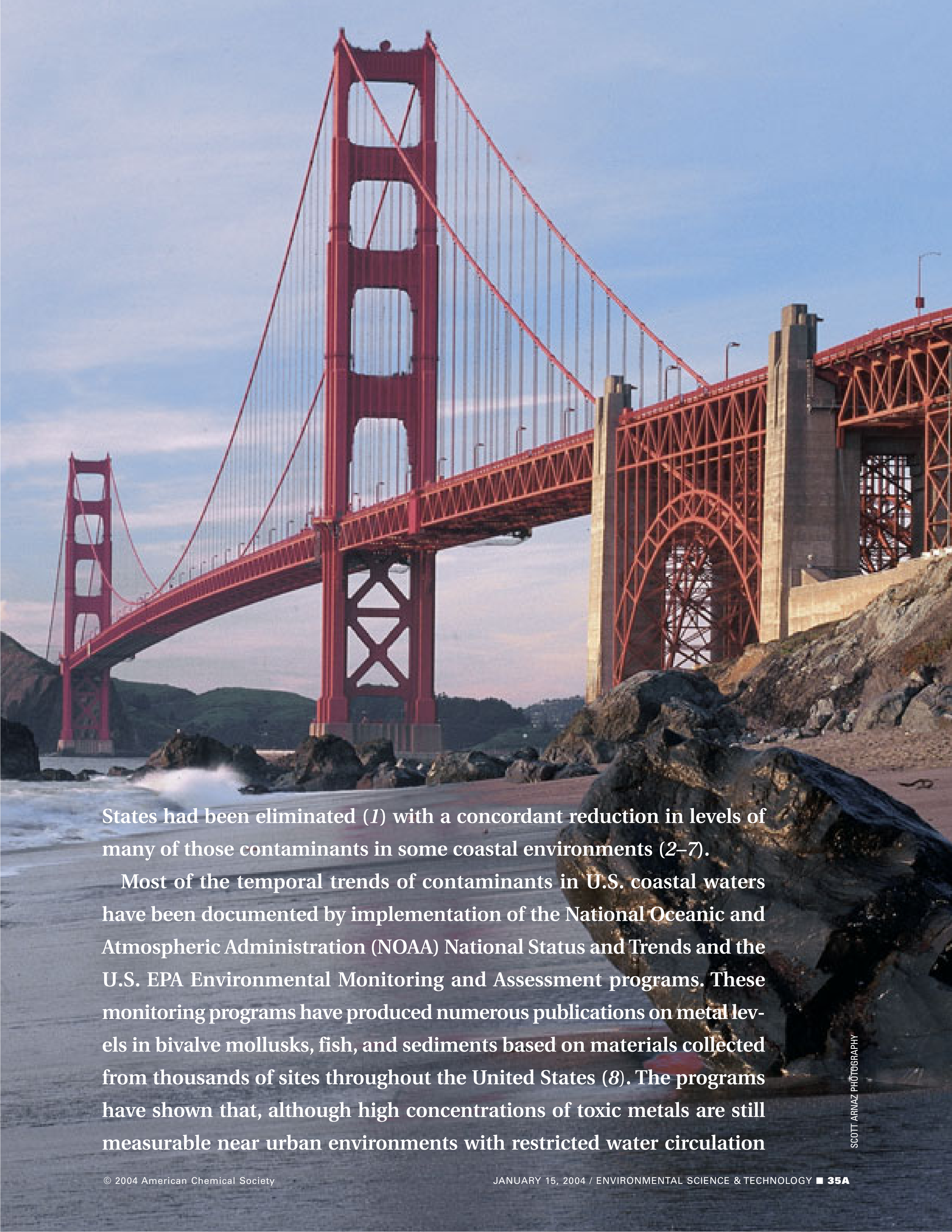
October 18, 2002, marked the 30th anniversary of the Clean Water Act (CWA), a landmark environmental law that prompted an unprecedented cleanup of U.S. waterways. The results of implementing the act are striking: By 1994, more than 90% of the total mass of contaminants discharged from point sources in the United

The Clean Water Act was established 30 years ago, but information on toxic metals remains limited.



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States had been eliminated (*1*) with a concordant reduction in levels of many of those contaminants in some coastal environments (*2–7*).

Most of the temporal trends of contaminants in U.S. coastal waters have been documented by implementation of the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends and the U.S. EPA Environmental Monitoring and Assessment programs. These monitoring programs have produced numerous publications on metal levels in bivalve mollusks, fish, and sediments based on materials collected from thousands of sites throughout the United States (*8*). The programs have shown that, although high concentrations of toxic metals are still measurable near urban environments with restricted water circulation

(*2*, *4*, *6*), levels of some toxic metals (cadmium, arsenic, copper, and selenium) have declined since the law was implemented in the 1970s. However, these recent improvements in coastal water quality should not provide us with a false sense of security. Levels of some contaminants in some urban estuaries have remained persistently high, despite order-of-magnitude reductions in point source inputs (*7*, *9–11*).

Although the EPA and NOAA monitoring programs have provided extensive data on contaminants in biological tissues and sediments, there is inadequate information on the levels of toxic metals in the dissolved phase, the fraction that passes through a filter with a pore size of 0.45 µm, in U.S. estuaries. Without measurements of all metal pools (water column, sediments, and seston), the transfer pathways and fluxes among different reservoirs of toxic metals in U.S. coastal waters remain poorly understood. Therefore, it has been difficult to determine if, for example, point versus nonpoint sources or anthropogenic versus natural processes are responsible for the high levels of toxic metals in certain estuaries.

PHOTODISC

Almost 70% of the articles on dissolved metals in the NOAA and EPA programs are from 5 estuaries.

Few good examples

Although the levels of dissolved cadmium, copper, nickel, and zinc in the water column of the Hudson River estuary in New York state have declined since the 1970s, they are still some of the highest ever reported in the United States, and mercury levels in this estuary continue to exceed current state and federal water quality standards (*5*, *12*). Furthermore, the ultimate source of all of those toxic metals remains largely unknown, in contrast to, for example, the extensively documented release of PCBs from historic industrial waste deposits located upriver (*13*).

Studies on the cycling of dissolved toxic metals in the Hudson River estuary are extremely limited. In the past 25 years, only 1 peer-reviewed scientific report on mercury and lead levels in the Hudson's water column (*5*) and 2 articles on other dissolved metals have been published (*12, 14*). With the marked reduction in point source discharges to U.S. estuaries, high levels of toxic metals are now attributed primarily to nonpoint sources. However, because of the incomplete metal data for the Hudson, it is almost impossible to establish the relative importance of several nonpoint sources on metal levels, such as surface runoff, groundwater seepage, diagenetic remobilization from contaminated sediments, and atmospheric deposition.

The paucity of peer-reviewed scientific information on the health of the Hudson River estuary is characteristic of essentially all U.S. estuaries (see Figure 1 and the Supporting Information, which includes a complete list of all publications). We have limited our consideration of this data deficiency to peer-reviewed articles because they represent an accepted standard of scholarship. Moreover, non-peer-reviewed reports on contaminant metal levels in natural waters may be unreliable for establishing dissolved water quality trends (*15*). We further limited our review to articles published from 1975 to 2002, because any research published before the development and application of trace metal cleanup techniques may not meet current standards for accurate analyses of trace element concentrations. Moreover, because levels of toxic metals in sediments and organisms are well established through government monitoring programs, we have restricted our assessment of the literature to reports of the traditionally defined "total dissolved" metal pool. Finally, dissolved metal levels are currently used to set and measure compliance with water quality standards (*16*).

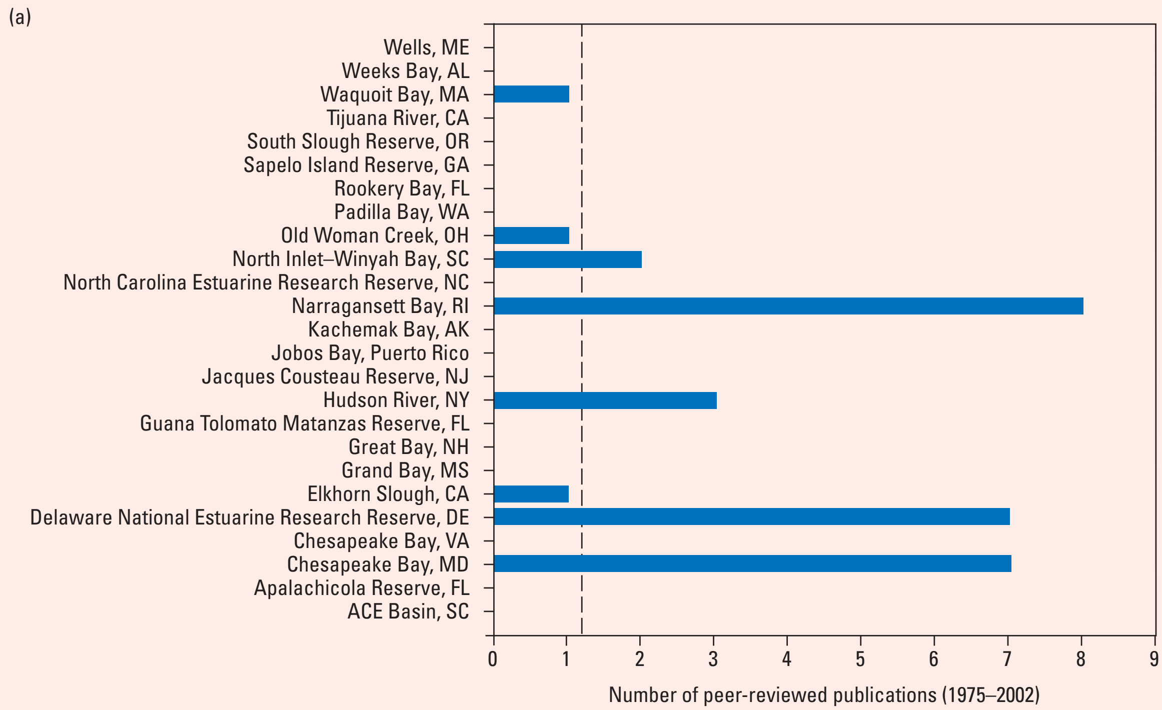
The deficiency of dissolved trace metal data for estuaries is illustrated by our results of a bibliographic search of estuaries listed in the NOAA National Estuarine Research Reserve System (25 estuaries) and in the EPA National Estuarine Program (28 estuaries), using the previously defined criteria. Dissolved trace metal concentrations have been reported for other estuaries not listed under the EPA and NOAA programs (*17*). However, our investigation focuses on the estuaries listed in those programs because they are across the United States in variable biogeographic regions and are considered of national importance.

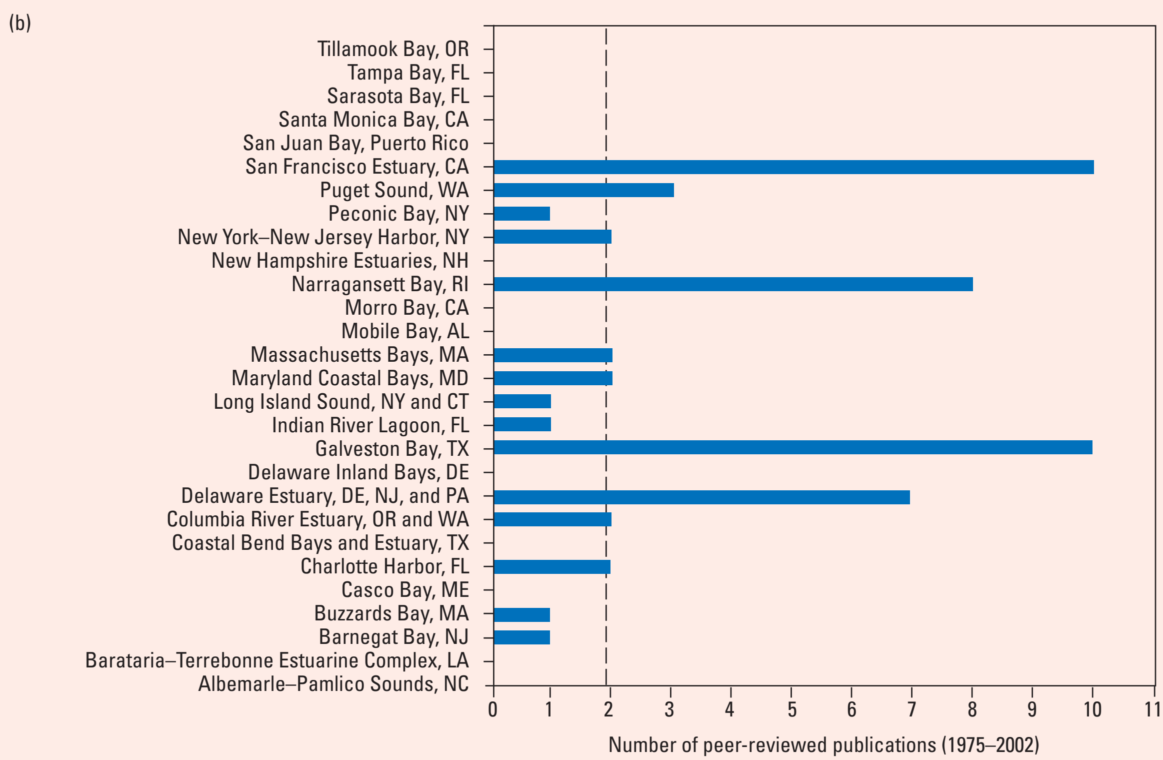
Despite the importance of these estuaries, since 1975 a total of only 83 articles have been published on dissolved metal cycling for all of those listed in the NOAA system (30 articles) and in the EPA program (53 articles). On average, only 1 article about dissolved metals in each of the NOAA estuaries and only 2 articles on the EPA estuarine program have been published over the past 27 years. The number of publications generated for 5 estuaries (Delaware, Chesapeake Bay, Galveston Bay, Narragansett Bay, and San Francisco Bay) biases those averages. In fact, almost 70% of the articles on dissolved metals in the NOAA and EPA programs are from those 5 estuaries.

FIGURE 1

Peer-reviewed articles published on dissolved trace metal concentrations in U.S. and Puerto Rican estuaries from 1975 to 2002

The data sets for both (a) NOAA programs and (b) EPA programs were obtained by using the Web of Science and the Scirus for Scientific Information Only search engines. The dotted line represents the average number of publications for the 27 years of the literature survey. Full citations are provided in the Supporting Information.





In contrast, there is not a single peer-reviewed article on dissolved trace metals in 68% and 46% of the estuaries listed in the NOAA and EPA programs, respectively. Furthermore, levels of mercury, one of the most toxic metals found in the environment, have been measured in just 6 of the 51 estuaries: Chesapeake Bay, Galveston Bay, Hudson River estuary, Long Island Sound, Maryland Coastal Bays, and North Inlet–Winyah Bay.


USDA

Levels of mercury have been measured in just 6 of the 51 estuaries.

Another 30 years?

Consequently, the benefits of investing tens of billions of U.S. taxpayer dollars over the past three decades to comply with the CWA cannot be quantified with any assurance for many contaminants in U.S. estuaries (*18*, *19*). The lack of peer-reviewed scientific reports on current levels of contaminants in those waters over that period precludes statistically valid measures of changes in contaminant levels over the past decade.

One exception is the San Francisco Bay Regional Monitoring Program. Established in 1989, the program has provided data to resolve subtle changes in dissolved metal levels on a decadal scale, in spite of the relatively large seasonal and annual variation (*7*). In the absence of that kind of information, it is impossible to predict how changes in climate, human population, and land-use dynamics will affect the future levels of many aquatic contaminants in the United States.

Thus, while the NOAA and EPA monitoring programs have produced numerous publications on metal levels in sediments and bio-indicators since the 1980s (*2*, *4*, *6*, *8*), similar long-term, systematic studies of the biogeochemical cycling and fate of aquatic contaminants in the water column of estuaries are needed. Until those studies are instituted, quantifying the real impact of the CWA on water quality in the United States will be difficult. Moreover, without that information, the efficacy of the multibillion dollar responses to those mandates will remain largely unknown. Finally, the limited peer-reviewed information available on dissolved metal levels in U.S. estuaries suggests that it will be very difficult for government agencies like EPA to issue new environmental regulations based on peer review, as was recently proposed by the White House (*20*).

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Note: Supporting information for this article is available at http://pubs.acs.org/est.