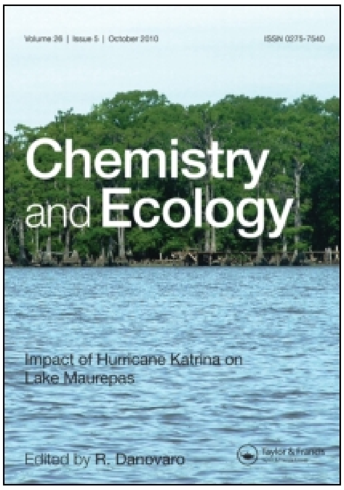


**Chemistry and Ecology**
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713455114

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**To cite this Article** Cantillo, Adriana Y. and O'connor, Thomas P.(1992) 'Trace Element Contaminants in Sediments From the Noaa National Status and Trends Programme Compared to Data From Throughout the World', Chemistry and Ecology, 7: 1, 31 — 50

**To link to this Article: DOI:** 10.1080/02757549208055431

**URL:** http://dx.doi.org/10.1080/02757549208055431

*Chemistry and Ecology*, 1992, Vol. 7, pp 31–50
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# TRACE ELEMENT CONTAMINANTS IN SEDIMENTS FROM THE NOAA NATIONAL STATUS AND TRENDS PROGRAMME COMPARED TO DATA FROM THROUGHOUT THE WORLD

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*(Received 22 August, 1991)*

The National Oceanic and Atmospheric Administration's (NOAA) National Status and Trends (NS&T) Programme has analyzed samples of surface sediment collected at almost 300 coastal and estuarine sites throughout the United States since 1984. The overall distributions for concentrations of each element are approximately lognormal allowing a definition of geometric means and of "high" concentrations as those exceeding the mean plus one standard deviation of the lognormal distribution. Those mean and "high" concentrations should be useful for comparing the NS&T data set and with other reports on sediment contamination. A world-wide data set, constructed from data in the literature, was summarized in an identical manner. It was found, after excluding locally extreme values, that the NS&T and world-wide data sets yielded very similar mean and "high" concentrations for Cu, Ni, Pb and Zn. Chromium was the sole element showing higher summary statistics in the NS&T data set, a result attributed to NS&T sampling in areas of naturally (not contaminated) elevated Cr content. It remains undetermined, however, why the world-wide data set, even with exclusion of "hot spots", yielded higher means and "highs" for Cd, Hg, As, and Ag.

KEY WORDS: Sediments, marine/estuarine, trace elements, mean concentrations, bibliography

## INTRODUCTION

Decisions on the use and allocation of resources in the nation's coastal and estuarine regions require reliable and continuous information on the status and trends of environmental quality in those areas. Beginning in 1984, NOAA undertook the task of providing this information through its National Status and Trends (NS&T) Programme for Marine Environmental Quality. The programme's objectives include defining the geographic distribution of contaminant concentrations in tissues of marine organisms and in sediments, and documenting biological responses to contamination. Samples have been collected since 1984 under the NS&T Benthic Surveillance Project and since 1986 under the Mussel Watch Project. At Benthic Surveillance sites, benthic fishes are collected and their livers excised and stored for subsequent chemical analysis. At Mussel Watch sites bivalve molluscs are collected for analysis. At all sites, in both projects, surface sediment samples have been collected. A series of NS&T reports (NOAA, 1987a, 1987b, 1988, and 1989) have presented and discussed contaminant concentrations in molluscs, fish, and sediments.

The laboratories that have undertaken Benthic Surveillance sampling and trace element analyses are the NOAA National Marine Fisheries Service laboratories in: Sandy Hook, NJ; Beaufort, NC; and Seattle, WA. The Mussel Watch trace element work has been performed by the Battelle laboratory in Sequim, WA; Texas A&M

University in College Station, TX; and the LaJolla, CA, Laboratory of Scientific Applications International Corporation.

## FIELD AND ANALYTICAL METHODS

Since the objective of the NS&T Programme is to quantify contamination over large spatial scales, sites at major point sources of contamination are avoided. The influence of any individual point source will probably not be seen in the NS&T data unless that source exerts a dominant influence on environmental quality over a relatively large area. On the other hand, the NS&T Programme will identify the combined influence of many point and non-point sources of contamination to an area.

In the Benthic Surveillance Project, sediment samples were obtained with a specially constructed box corer or a standard Smith-McIntyre bottom grab. In the Mussel Watch Project, the samples were obtained with the box corer or with a Kynar-coated Van Veen grab sampler. Three grabs or cores were obtained at each of the three stations at a site. Composite samples were made from surface sediment in those three grabs or cores. Sediment analyses for a site consisted of organic analyses of three composites (one from each station), inorganic analyses of three separate composites, and grain-size and other measurements on a third set of three composites. In the Benthic Surveillance Project, a small corer was used to subsample the top 3 cm of each box core or grab sample for trace metal and other analyses. In the Mussel Watch Project, the sub-samples for all composites were surface skims from the top 1 cm of each box core or grab. Samples for analyses of major and trace elements were stored in Teflon jars or ziplock bags. A more detailed presentation of the sampling protocols is included in Shigenaka and Lauenstein (1988).

The elements measured in NS&T sediment samples are Al, Fe, Mn, Si, Sb, As, Cd, Cr, Cu, Pb, Hg, Ni, Se, Ag, Sn and Zn. Digestion in concentrated hydrofluoric acid and subsequent analysis by atomic absorption or, for some elements analysis by X-ray fluorescence, were used for quantification. Details of analytical methods employed in the Mussel Watch Project can be found in Battelle (1987) and the report of Texas A&M Geochemical and Environmental Research Group (Texas A&M, 1988).

Quality assurance (QA) protocols are an integral part of the NS&T Programme. QA efforts are designed to produce nationally uniform analytical results of known and accepted quality, thereby ensuring comparability among data sets. The QA Programme involves four major activities; interlaboratory comparisons of analytical methods, annual QA workshops, and development and required use of Standard Reference Materials, Interim Reference Materials, and control materials for marine sediments and tissues.

## NS&T SEDIMENT DATA

The NS&T sediment data base containing analytical results from 1426 composite samples from 266 sites and detailed statistical analysis of the data can be found in NOAA (1991). Contaminants in sediments are associated with particle surfaces, and differences in contaminant concentrations among sites can be the result of differences in particle size distributions. To compensate for this, sediment data have been adjusted by dividing the raw concentration in a composite by the fraction by weight of sediment particles which are less than 63$\mu$ in diameter (i.e., the fine-grained or silt and clay fraction). This is equivalent to assuming that no contaminants are associated with sand-sized particles and that the only effect of sand in a sample is to dilute its level of contamination. This method can lead to errors when the sediments involved are composed primarily of sand-sized or larger particles, so only sediments with greater than 80% fine-grained particles were considered in the overall assessment (NOAA, 1991).

## THE NATIONAL DISTRIBUTIOIN OF SEDIMENT CONTAMINATION

The distribution of trace metal concentrations among sites was skewed towards low values, and when plotted as logarithms the data were approximately normally distributed. This type of distribution provides a convenient method for defining a "high" concentration as the mean plus one standard deviation of the lognormal distribution. A perfect normal distribution would be one where 17% of the values exceed the mean plus one standard deviation, and that percentage is approximated by the distributioins of the logs of NS&T mean concentrations. Typical distributions of the logarithms of the mean concentrations for all NS&T sites are shown for silver and zinc in Figure 1. Table 1 is a list of geometric means and "high" concentrations for nine trace elements measured in the NS&T Programme and being considered in this paper.

The tendency for many chemicals to be high at the same site is reflected in a factor analysis (Table 2) that includes data supplied by Dr. Robert Hamill of the Geography Division of the US Census Bureau. Using 1980 census data he provided numbers of people residing within various distances of NS&T sites. The data used are the numbers of people within 20 km and demonstrate that most chemicals not only occur together but that their concentrations are related to human population levels.



## NATURAL AND "HIGH" CONCENTRATIONS

Because the factor analysis is based on correlations among chemical concentrations that are lognormally distributed, strong correlations and therefore common loadings will be found for chemicals that occur together at high concentrations. Because they exist naturally, all trace elements will be found in sediments at some concentration. For most elements these natural levels are not among the "high" concentrations. Some chemicals, though, are entirely separated from the main group in the factor analysis, while others have relatively high loadings on factors beyond the first.

High concentrations of chromium and nickel are found mainly on the West Coast north of Point Conception. Those elements are enriched in the rocks of that area (USGS, 1981), so their high concentrations in sediments of the region cannot be attributed to human activities and, in that sense, they are not contaminants. In addition to chromium and nickel, there is a strong component to the cadmium and arsenic concentrations that is not associated with proximity to population centres. High arsenic concentrations are found in the northwest coast of the US in association with chromium and nickel. It also appears as the sole "high" level chemical at some sites in the southeast coast of the US. The reasons for "high" cadmium concentrations away from cities are not evident. Nevertheless, sites having high concentrations of arsenic, cadmium, chromium or nickel should not be considered contaminated (i.e., influenced by human activity) unless concentrations of other elements are also high.

## WORLD-WIDE LITERATURE SURVEY

The geometric means and "high" concentrations serve as convenient markers for characterizing the coastal and estuarine United States. The "high" values were used in NOAA (1991) and by O'Connor (1990) to identify areas in the coastal United States where sediments were most affected by human activities. If these markers truly do represent mean and "high" conditions for chemical concentrations in sediments, they should also be applicable outside the NS&T data base from which they were derived.

To test their general applicability, we used a data base on trace metal concentrations in surficial coastal sediments throughout the world that was prepared from values published in the scientific literature (Cantillo, 1982) and subsequently expanded in 1991. The areas of interest were estuaries, lagoons, fjords, harbours, bays, tidal mud flats and other coastal areas. The data base does not include river, lake or deep sea sediments, or data on sands. Two electronic bibliographic data bases were searched for papers reporting total concentrations of any of the elements determined as part of the NS&T Programme. The result of the initial computer searches was expanded manually as the compilation of papers and data progressed.

In total, 226 papers (cited separately in this report) were used in the compilation of the data base, representing more than 60,000 data points. Availability of data varied among the 226 papers, reflecting compromises between journal requirements and amount of data available. If the amount of data was reasonably small, data tables were usually provided. Otherwise, only the mean and range of the data for each element were reported. In some cases data were extracted from graphical presentations. If only a concentration range was reported, the average of the high and the low values was used as the mean. When the range is large, the mean of the extremes is effectively half the high value. The geometric mean may have been a better choice, but in papers where means were provided they, too, were arithmetic means. Since these arithmetic means were, presumably, based on all data, they would not be as biased toward high values as means based on the extreme values of a range but, nevertheless, any arithmetic mean must be biased toward high values.

Data were from sediment samples that had been solubilized using a variety of acid dissolution procedures, such as nitric-perchloric-hydrofluoric acid mixtures. Only papers reporting the use of methodology resulting in a measure of total element content in sediments were considered. By far the most commonly-used instruments were atomic absorption spectrophotometers, including cold vapour, graphite furnace and flame techniques. It was noticed that, with time, an increasing number of papers reported the use of standard or certified reference materials.

## COMPARISON OF NS&T AND WORLD-WIDE DATA

For nine elements, there were at least 30 papers reporting sediment concentrations

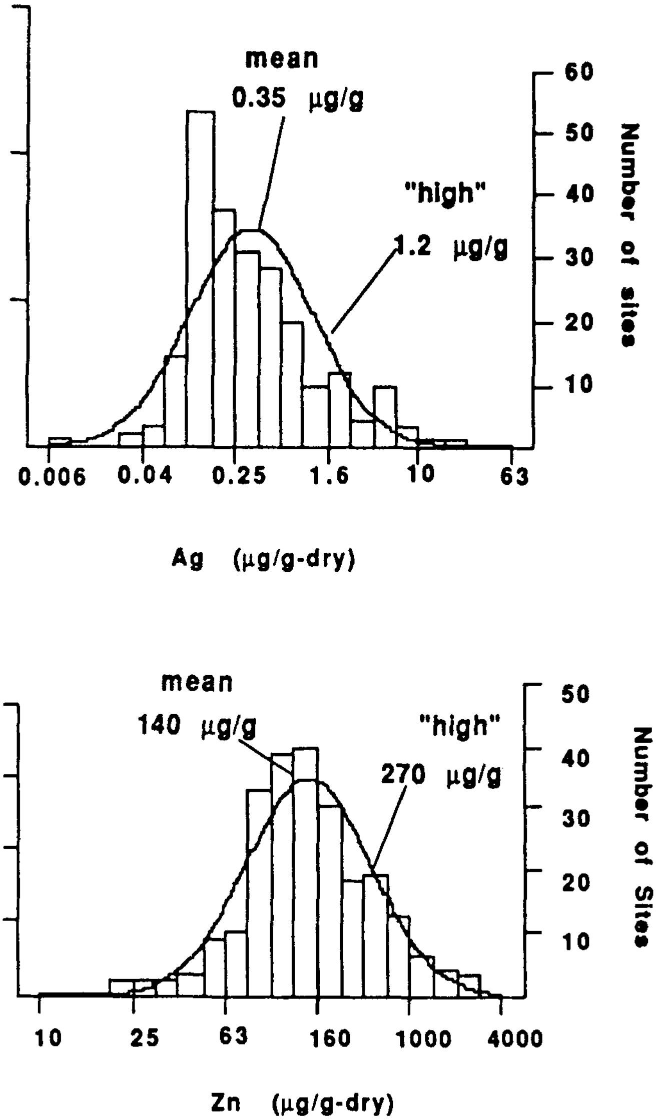


**Figure 1** Examples of distributions (note logarithmic scales) of mean concentrations at NS&T sites.

and those world-wide data were treated exactly as were the NS&T data to yield geometric means and "high" concentrations. The results (Table I and Figure 2) fall into three categories; first the elements copper, nickel, lead and zinc where the world-wide mean concentrations are similar to the NS&T means but the "highs" are higher, second the elements silver, arsenic, cadmium and mercury where both the world-wide mean and the "high" are higher, and a third category unique to chromium where the NS&T mean and "high" are higher than for the world-wide data.

**Table 1** Geometric mean and "high" concentrations from logarithmic distribution of site means. The "high" concentrations are derived from the mean plus one standard deviation. The NS&T statistics are based on analyses of samples containing at least 20% fine-grained sediment and have been adjusted for their sand content. World-wide data set I is based on literature survey (and World-wide data set II is the same as "I" with the exclusion of "hot spot" data (see text). In all cases, "n" is the number of data points defining the distribution. Elements are separated into three groups on the basis of extent of similarity between NS&T and World-wide statistics.

| | *NS&T* | | |
|---|---|---|---|
| *Element* | *Geometric mean* | *"high"* | *n* |
| Cu | 35 | 84 | (233) |
| Ni | 34 | 69 | (233) |
| Pb | 43 | 89 | (233) |
| Zn | 140 | 270 | (233) |
| Ag | 0.35 | 1.2 | (233) |
| As | 13 | 24 | (233) |
| Cd | 0.48 | 1.2 | (233) |
| Hg | 0.17 | 0.49 | (233) |
| Cr | 110 μg/g | 230 μg/g | (233) |

| | *World-wide (I)* | | |
|---|---|---|---|
| *Element* | *Geometric mean* | *"high"* | *n* |
| Cu | 32 | 130 | (241) |
| Ni | 27 | 90 | (163) |
| Pb | 43 | 184 | (227) |
| Zn | 110 | 450 | (238) |
| Ag | 1.3 | 6.4 | (32) |
| As | 30 | 151 | (52) |
| Cd | 1.5 | 7.6 | (179) |
| Hg | 0.82 | 7.7 | (95) |
| Cr | 65 μg/g | 200 μg/g | (163) |

| | *World-wide (II)* | | |
|---|---|---|---|
| *Element* | *Geometric mean* | *"high"* | *n* |
| Cu | 28 | 93 | (213) |
| Ni | 26 | 92 | (144) |
| Pb | 34 | 110 | (205) |
| Zn | 91 | 290 | (216) |
| Ag | 1.1 | 4.9 | (30) |
| As | 25 | 90 | (45) |
| Cd | 1.1 | 4.3 | (154) |
| Hg | 0.63 | 4.7 | (84) |
| Cr | 61 μg/g | 170 μg/g | (151) |

The low ends of the world-wide data sets extend to lower values than the NS&T data because the NS&T set contains no samples with more than 80% sand and data for all other samples are adjusted (i.e., raised) to being on a 100% fine-grained basis. While sandy samples were avoided in constructing the world-wide dataset, such samples would have been included when means for an area were presented and when no

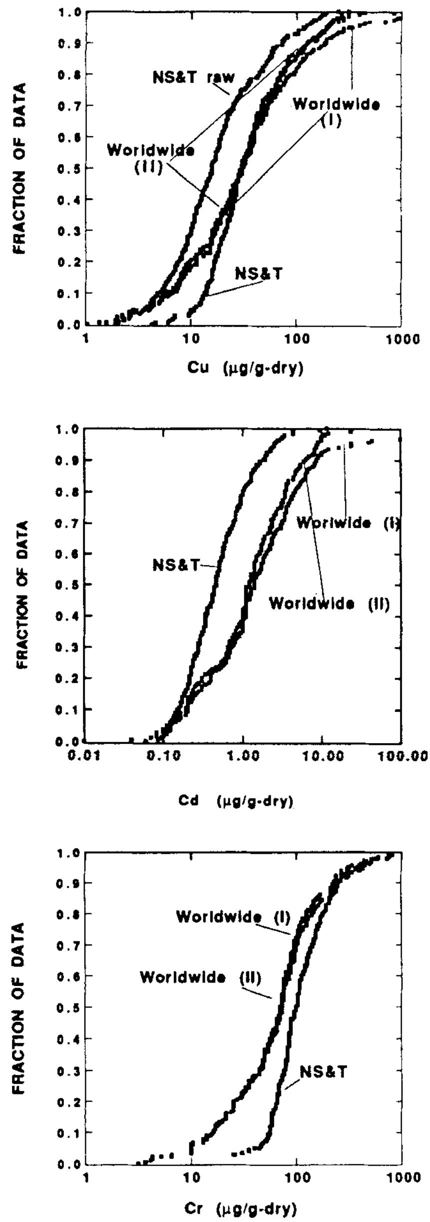


**Figure 2** Cumulative distribution plots (note logarithmic scale) for Cu, Cd, and Cr comparing that of NS&T data to NS&T raw data, and to World-wide data with (I) and without inclusion of "hot spots" (II). The Cu distributions are like those for Ni, Pb, and Zn where the World-wide datasets show lower low values than the NS&T data (unless raw NS&T data are used) and where the exclusion of "hot spots" from the World-wide data set brings the high end into compliance with the NS&T data. The Cd distributions are like those for Hg, Ag, and As where the World-wide data show higher means and higher highs than NS&T, regardless of inclusion of "hot spots". The Cr plot is unique in showing higher concentrations for NS&T relative to the World-wide data.

indications of grain-size were given. None of the world-wide data were adjusted for particle size.

The cumulative data plot in Figure 2 includes the cumulative distribution all NS&T data in their raw form. It demonstrates that the NS&T data can be made to match the low end of world-wide data if the NS&T concentrations are used without consideration of particle size. Not making the grain-size adjustment, on the other hand, increases differences between the NS&T and world-wide data sets at the mid- and upper-ends of the concentration distributions. The more interesting observation is that, except for chromium, the upper ends of the world-wide data sets exceed even the grain-size adjusted NS&T values. It is most likely that the higher concentrations in the world-wide data are from muddy sediments, so it is appropriate that they be compared to the (particle-sized adjusted) NS&T data but, still, the highest values are the world-wide values.

An important point in resolving this discrepancy is that highest values in the world-wide dataset are the result of sampling known "hot spots". This type of site was avoided in NS&T programme because they are small zones of high contamination that are not representative of their surroundings. Some of the data in the world-wide data set are from papers where the authors specifically indicate that they have sampled near discharge points or in industrial zones or in otherwise uniquely contaminated locations. The papers identified in Table 3 are ones where the authors specifically indicated that they were sampling "hot spots". When data from those papers are deleted from the world-wide data set, the high ends of the overall distributions are decreased and, for copper, nickel, lead and zinc, conform to the high end of the NS&T distribution (Table 2, Figure 2).



**Table 2** Results of factor analysis on correlation matrix among means of grain size-adjusted concentrations of NS&T sediment data. Loadings >0.4 on each factor by each chemical are indicated in parentheses. No chemical has a loading >0.4 on any factor beyond factor four. The percentage of overall variation attributable to each factor is also indicated. The variable POP20 is the number of people living within 20 km of the site centre (R. Hamill, US Census Bureau, 1990, personal communication).

| | | | |
|---|---|---|---|
| Factor 1 (49% of overall variation) | | | |
| Pb (0.90) | Cu (0.90) | Hg (0.87) | Zn (0.84) |
| Sn (0.76) | POP20 (0.69) | Cd (0.65) | Ag (0.67) |
| As (0.45) | Cr (0.41) | | |
| Factor 2 (12% of overall variation) | | | |
| As (−0.42) | Cr (−0.59) | Ni (−0.81) | |
| Factor 3 (6% of overall variation) | | | |
| Cd (−0.48) | As (−0.47) | | |
| Factor 4 (4% of overall variation) | | | |
| POP20 (0.42) | Zn (−0.43) | | |

In the case of one element, chromium adjusting for the grain-size effect has overcompensated for differences between the data sets. The cumulative distribution plot of the NS&T data lies to the right of the world-wide data, regardless of inclusion of "hot spot" data). This might be due to the fact that the NS&T sites include some where chromium is mineralogically enriched. Using raw NS&T data would have yielded a closer overall agreement with the world-wide data, except at the upper end where the higher values would then be from the world-wide data.

For silver, arsenic, cadmium and mercury, more than half the values from the world-wide data sets exceed the "high" value in the NS&T data. For those elements, removing "hot spot" data from the world-wide data set did not yield compliance with NS&T data. For silver and arsenic the number of data points, 32 and 52 respectively, are perhaps too small for a valid comparison with NS&T data. For cadmium and mercury, however, the numbers of points are 95 and 179, respectively, and the difference between world-wide and NS&T data sets begs explanation.

There is a continuing Quality Assurance component to the NS&T programme that does quantify the extent of agreement among laboratories for analyses of common samples. There is no way to check those laboratories against laboratories contributing to the world-wide data sets. On the other hand, there is certainly no basis for claiming that the world-wide data are erroneously high. While, on the basis of specific information provided by authors, "hot spot" data could be excluded from the world-wide data set, it is possible, for cadmium and mercury, that a large majority of the remaining data were from samples collected near contaminant sources. Due to tragic episodes of Minamata and *Itai-itai* disease from human exposure to extreme levels of mercury and cadmium, respectively, those elements, of the nine discussed in this paper, are of highest public concern. That concern could have lead to investigations of mercury and cadmium concentrations in poorly-flushed industrial or urban areas. Data from such studies would qualify for the "hot spot" category but, without explicit indications on the part of authors, we cannot objectively exclude them from the world-wide data set.

Another possible reason for the concentrations of cadmium, mercury, silver and arsenic being higher than is consistent with the NS&T data is that analyses in the past may well have higher detection limits than is the case now. That, in itself, could raise the overall concentration distributions in two ways. First, when no chemical was detected, analysts could substitute concentrations corresponding to their detection limits. Second, to avoid futile analyses, there could have been a tendency for investigators not to sample areas away from direct contaminant sources.



## *Conclusions*

The NOAA NS&T programme has generated a data set on chemical concentrations in fine-grained sediment collected at 233 sites throughout the coastal and estuarine United States. While that was done to compare the 233 sites, the NS&T data set should be applicable on a wider spatial scale and it has been compared, here, with a data set based on mean concentrations taken from 226 papers on sediment concentrations of Ag, As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn. This latter data set, like the NS&T data, is based on samples from estuarine and coastal areas (not open ocean or fresh water).

For all nine metals, the low ends of the concentration distributions are lower for the world-wide data set than for the NS&T data set. That difference is due to the fact that the NS&T data do not include samples with more than 80% sand (particles $>63\mu$) and all other concentrations have been raised in inverse proportion to their fine-sediment fraction. Low concentrations were thus excluded from the NS&T data set to allow comparisons that were not affected by grain-size differences among NS&T sites. Such low concentrations could be part of the world-wide data set because, while known sand-based samples were excluded, not all papers provided grain-size differences among NS&T sites. Such low concentrations could be part of

**Table 3** Data in world-wide data set excluded as "hot spot" data in compiling World-wide (II).

| *Source* | *Site description* |
|---|---|
| Bloom and Ayling (1977) | near metallurgic plant and Hobart sewage outfall |
| Boboti *et al.* (1985) | " . . . highly polluted sediments . . into Piraeus harbour" |
| Boyden *et al.* (1979) | ". . grossly polluted" mining area of Cornwall |
| Crecelius *et al.* (1975) | copper smelting and chlor-alkali plant near Tacoma |
| Cronin *et al.* (1974) | industrial parts of Baltimore harbour |
| Donazzolo *et al.* (1984) | industrial part of Venice |
| Harbison (1984) | near Pirie lead smelter |
| Kiorboe *et al.* (1983) | near a facility that once discharged Hg |
| Langston (1980) | mining area "As levels among highest in U.K." |
| Meyerson *et al.* (1981) | industrial part of Newark Bay |
| Panda *et al.* (1990) | chlor-alkali plant |
| Pavoni *et al.* (1987) | industrial part of Venice |
| Pruell *et al.* (1990) | industrialized harbour of New Bedford |
| Roy and Crawford (1984) | lead-zinc smelter |
| Samant *et al.* (1990) | Belledune "contaminated with Cu, Zn, Pb, and Cd" |
| Sasamal *et al.* (1987) | chlor-alkali plant |
| Shaw *et al.* (1988) | chlor-alkali plant |
| Skei *et al.* (1972) | zinc, aluminium, and phosphate mining/processing |
| Stenner and Nickless (1975) | extreme levels at La Rabida |
| Stoffers *et al.* (1986) | Wellington harbour history of pollutant discharges |
| Taylor (1986) | Liverpool history of pollutant discharges |
| Tiller *et al.* (1989) | near Pirie lead smelter |
| Villa and Johnson (1974) | industrial parts of Baltimore harbour |
| Voutsinou-Taliadouri and Varnavas (1985) | iron-nickel alloy smelter |
| Ward and Young (1981) | near Pirie lead smelter |
| Watling *et al.* (1974) | sewage sludge dumpsite |
| Yim (1976) | mining area of Cornwall |



the world-wide data set because, while known sand-based samples were excluded not all papers provided grain-size information, and, furthermore, data were not adjusted for the grain-size effect.

Except for chromium, the "high" concentrations (sum of geometric mean and standard deviation) in the world-wide data always exceeded those in the NS&T data. The higher concentrations are in the world-wide data set because that data set includes concentrations measured in sediments from small but highly contaminated locations near the ends of discharge pipes or industrialized embayments. Such "hot spots" were deliberately excluded from the NS&T sampling grid which attempted to sample only sites that were representative of their general surroundings. Where information provided by authors of papers in the world-wide data set clearly indicated that "hot spots" had been sampled, that data could be excluded. When such a reduced world-wide data set was compared with the NS&T data there was excellent agreement for geometric means and "high" concentrations for copper, nickel, lead and zinc. However, even with exclusion of explicitly identified "hot spot" data in the world-wide data set, silver, arsenic, cadmium and mercury concentrations were higher than in the NS&T data set.

This difference remains unresolved. For silver and arsenic the world-wide data set is small and more data may tend to lower the overall concentration distributions. For cadmium and mercury, however, there are already about 100 or more values in the world-wide data set. Conceivably, in the case of these two metals, there is a strong inclination to sample near contaminant sources.

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