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Comparison of Results of Mussel Watch Programs of the United States and France with Worldwide Mussel Watch Studies

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As part of the Global Ocean Observing System (GOOS), the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program compiled the World Mussel Watch (WMW) data base with results from the analyses of marine or estuarine mussels or oysters as far back in time as possible. Here we compare WMW data with results from two long-term Mussel Watch Programs, the Réseau National d'Observation de la Qualité du Mulieu Marin (RNO) Mussel Watch in France and NS&T Program in the United States. The medians and 85th percentiles for Cr, Ni, Cu, Zn, As, Se, Ag, Cd, Hg and Pb in mussels and oysters were calculated for the WMW, NS&T and RNO Mussel Watch programs. While there was generally good agreement for medians among all three data sets, the upper ends of the WMW concentrations tend to be higher than their NS&T and RNO counterparts. This probably reflects the fact that the latter two programs emphasize collection of mollusks at representative sites rather than within small areas of extreme contamination such as near waste discharges. © 1998 Elsevier Science Ltd. All rights reserved

In response to increased awareness of possible global environmental changes, the Global Ocean Observing System (GOOS) was created under the auspices of the United Nations (Anderson, 1997) as a framework to put national data into a global context. Mussel Watch programs have been used to assess coastal environmental pollution since they were proposed in the 1970s by Goldberg *et al.* (1978). In the spirit of GOOS, the National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program undertook the task of providing the global context for these types of data by compiling results of Mussel Watch programs throughout the world.

World Mussel Watch Data

A worldwide literature search for reports with results from chemical analyses of soft parts of marine or estuarine mussels or oysters yielded more than 300

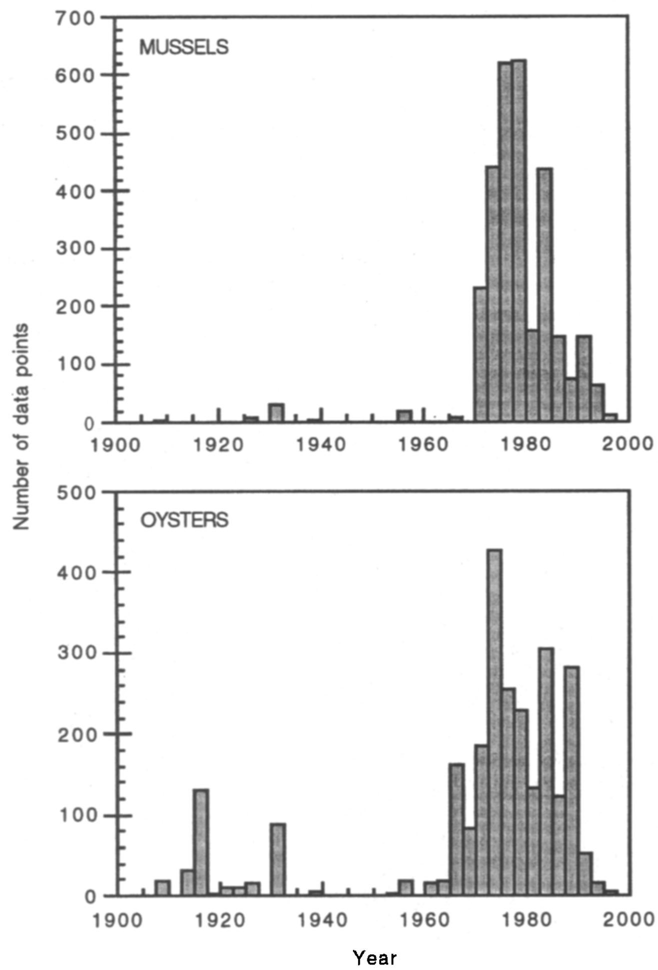

Fig. 1 Histogram of years for which data are available for mussels and oysters in the WMW database.

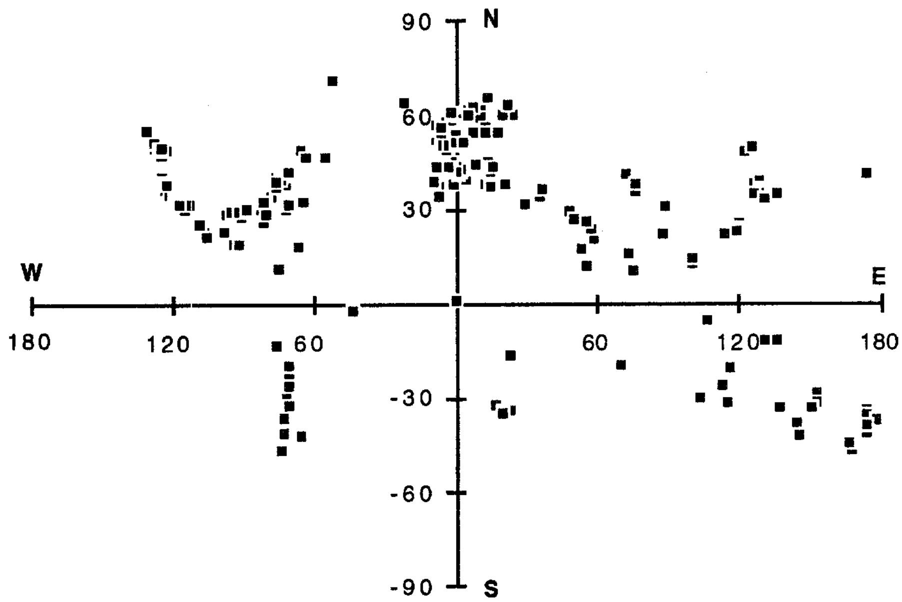

Fig. 2 Geographical distribution of all sampling locations in the WMW database.

citations. Most of the reports were based on quantifying contamination, but some of the oldest were on the nutritional use of oysters in the treatment of anemia. Results for the trace elements Cr, Ni, Cu, Zn, As, Se, Ag, Cd, Sn, Sb, Hg, Ti, and Pb in mussels and oysters were entered into four files to create the World Mussel Watch (WMW) data base.

The trace organic contamination data were much fewer than for trace elements and are difficult to compare among sources. Often, insufficient analytical methodology detail was available in citations to judge to what degree one set of data could be compared to another. There have also been changes in analytical methodology for the analysis of trace organic contaminants in the environment over the last two decades so comparison of older data with more recent data even from the same source may not be valid. It was decided,

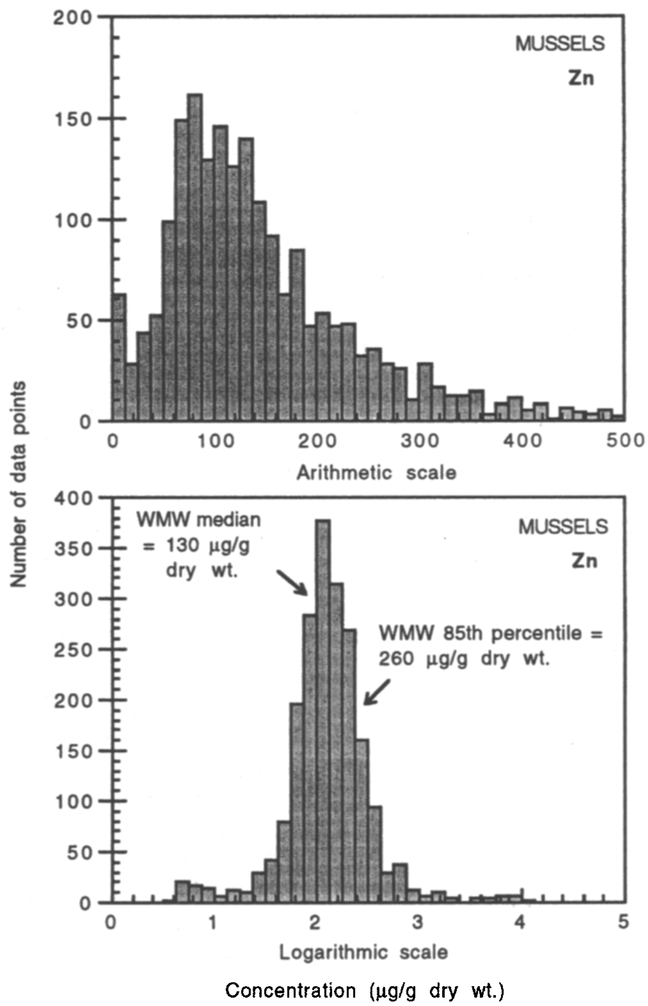

Fig. 3 Distribution of zinc in mussels on arithmetic and logarithmic scales. (WMW mean is the mean of the logarithmic values and the WMW high is the mean plus one standard deviation of the logarithmic values. All data used in calculations. Some high concentrations not shown.)

TABLE 1

Countries or territories represented in the WMW database.

Argentina	Hong Kong	Peru
Australia	Iceland	Poland
Bahrain	India	Portugal
Belgium	Indonesia	Puerto Rico
Bermuda	Ireland	Russia
Brazil	Isle of Man	Scotland
Canada	Italy	Singapore
Chile	Japan	South Africa
Columbia	Korea	Spain
Croatia	Kuwait	Sweden
Denmark	Lebanon	Taiwan
Egypt	Mexico	Thailand
England	Monaco	The Netherlands
Finland	Morocco	United States
France	New Zealand	Wales
Germany	Northern Ireland	Yugoslavia
Greece	Norway	
Greenland	Oman	

therefore, not to include the trace organic contaminant levels in the WMW.

Results of the analysis of trace metals in more than 60 species of mussels and oysters, mostly Mytiloids and Ostreoids, were entered into four files to create the WMW database: TM, DRY, TM WET, TM RANGES DRY and TM RANGES WET. Data from citations containing trace metal (TM) concentrations on a dry weight basis were entered into the TM DRY file. If there was sufficient information in the citation to convert wet weight results to dry weight, the concentrations were converted and added to the TM DRY file. The TM WET file contains only those concentrations reported on a wet weight basis without information on wet–dry ratios. In some citations, only the mean, low, and high concentrations were reported and these are contained in the TM RANGES DRY or TM RANGES WET files. In addition to the trace metal data, each file contains information on data source, latitude and longitude of sampling sites, sampling location name, and country.

Values which were listed as below a specific detection limit were listed at that level (e.g. <5.0 was listed as 5.0). In cases where mollusks were transplanted, only concentrations measured prior to transplanting were used. No attempt was made to obtain approximate concentrations from graphical data displays although this may be possible in many cases. If no sampling date was listed in the data source, the publication year of the citation was used. If a period of time was listed, then the time half-way between the two stated dates was used. Cantillo (1997) includes a summary of each data source in terms of: bibliographical citation; country; year of sampling or publication; number of sites sampled; species sampled; a short description of the analytical methodology; a notation of the use of reference materials or participation in intercomparison exercises; collection and analysis of other species or other types of samples such as sediment or seawater; and country, author, species and analyte indices.

The data of the NOAA NS&T and the Réseau National d'Observation de la Qualité du Milieu Marin (RNO) Mussel Watch efforts were not added to the

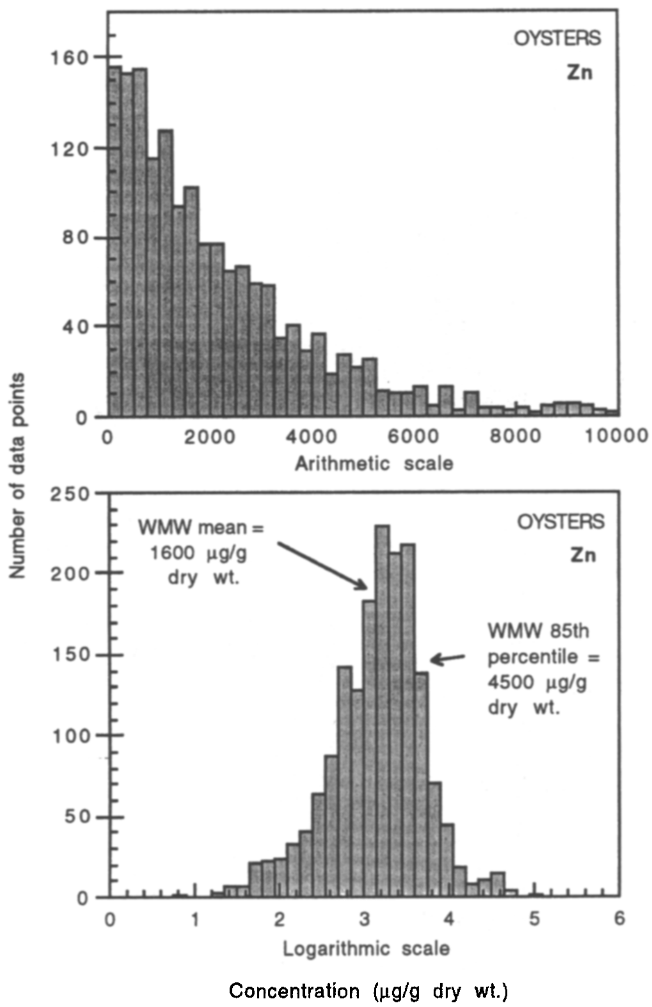

Fig. 4 Distribution of zinc in oysters on arithmetic and logarithmic scales. (WMW mean is the mean of the logarithmic values and the WMW high is the mean plus one standard deviation of the logarithmic values. All data used in calculations. Some high concentrations note shown.)

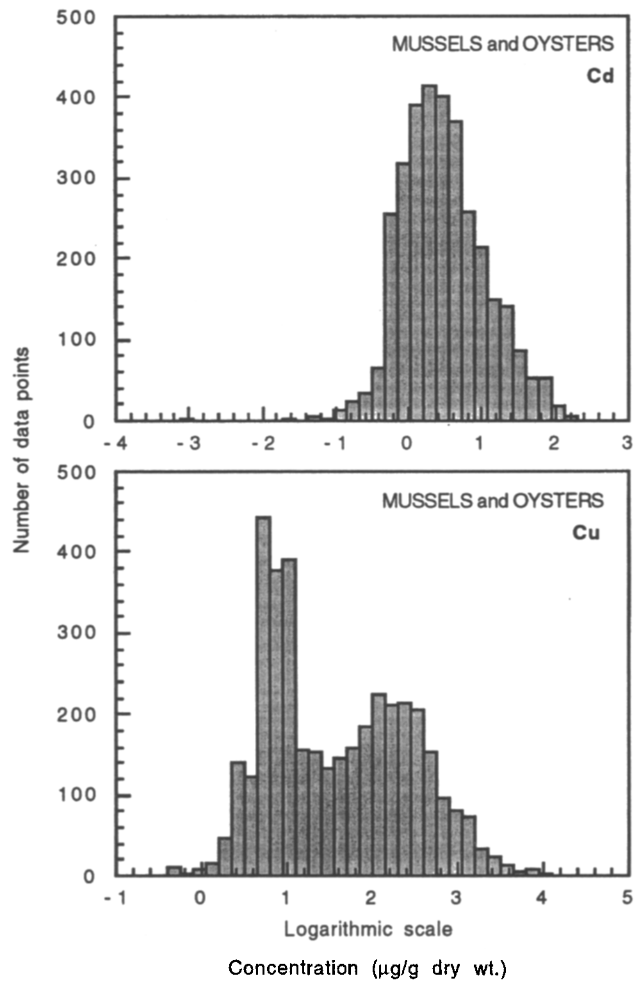

Fig. 5 Distribution of cadmium and copper in musselsand oysters on arithmetic and logarithmic scales. (Oysters and mussels differ in their ability to concentrate some elements such as Cu.)

WMW database. The results of these programs were treated separately since the numnber of data points in each is larger than that of the WMW data. The NS&T Mussel Watch data can be obtained from the Program Office by contacting the Manager, NS&T Program, NOAA/NOS/ORCA 21, 1305 East West Hwy, Silver Spring, MD 20910, USA, or from the Internet at: http://www-orca.nos.noaa.gov, the RNO data can be obtained by contacting D. Claisse, IFREMER, BP 1105, 44311 Nantes Cedex 03, France.

WMW Database Statistics

Time and geographical distribution

Data found for samples collected before 1940 (Fig. 1) were mostly from studies of the use of oysters in the treatment of anemia. No data were found for the decade of the 1940s and few prior to 1970. The large increase in data in the 1970s reflects the beginning of the environmental movement and the inception of 'Mussel Watch' monitoring by Goldberg *et al.* (1978). The distribution of data by latitude and longitude shows that data were collected throughout the world (Fig. 2). However, in the listing of countries (Table 1), the sparcity of data from the continents of South America and Africa is noted.

Statistics per element

Oysters and mussels are not equal in their ability to concentrate trace elements (O'Connor, 1993). The trace elements Ag, Cu, and Zn are enriched more than 10-fold in the oyster *C. virginica* relative to the mussel *M. edulis*. Data for other metals do not show so strong a species effect. Nonetheless, there were many species of mussels and oysters used in the compilation, so it was decided to simplify the initial evaluation of the WMW data base by treating the mussel and oyster data separately.

The most commonly determined elements in mussels and oysters were Cu, Zn, Cd, Pb, Hg, Cr, and Ni. Examples of the Zn data distribution with time and histograms are shown in Figs 3 and 4. The Cr, Ni, Cu, Zn, Cd, and Pb data for mussels and oysters were found to be log normal. The As data also appear to be log normal although the distribution may be bi-modal. There are some Cu and Zn data dating back to the turn of the century. The levels found back then were not the highest found in the literature, although the specimens analyzed were usually 'green' oysters with known high levels of Cu. No data were found for Se in oysters and the very little data found for Se in mussels appear to be log normal within a small concentration range. The Ag and Hg data distributions do not clearly

TABLE 2

Comparison of WMW, NS&T, and RNO median and 85 percentile concentrations ($\mu g\ g^{-1}$ dry wt)[a,b].

		Mussels			Oysters		
		Median	85th percentile	n^c	Median	85th percentile	n^c
Cr	WMW	1.6	6.5	343	2.5	10	705
	NS&T	1.8	3.1	1854	0.55	1.2	1909
	RNO	–	–	–	–	–	–
Ni	WMW	2.2	5.0	776	2.2	4.7	320
	NS&T	1.9	3.5	1851	1.8	3.2	1940
	RNO	–	–	–	–	–	–
Cu	WMW	7.9	21	1877	160	680	1960
	NS&T	9.1	12	1854	120	280	1943
	RNO	7.1	9.1	3295	130	320	2796
Zn	WMW	130	260	2058	1600	4500	1726
	NS&T	130	190	1855	2100	4300	1944
	RNO	110	180	3297	2100	3500	2804
As	WMW	7.1	16	189	5.7	14	276
	NS&T	9.6	15	1855	7.9	18	1943
	RNO	–	–	–	–	–	–
Se	WMW	2.2	3.9	95	–	–	–
	NS&T	2.5	3.6	1830	2.7	3.9	1930
	RNO	–	–	–	–	–	–
Ag	WMW	0.25	1.0	440	1.3	2.6	250
	NS&T	0.15	0.72	640	2.1	5.1	813
	RNO	–	–	–	–	–	–
Cd	WMW	2.0	7.5	1982	4.1	21	1293
	NS&T	2.2	4.8	1854	3.2	6.0	1943
	RNO	0.95	1.9	3273	2.3	6.0	2793
Hg	WMW	0.32	0.99	942	0.27	0.70	541
	NS&T	0.12	0.26	1832	0.09	0.19	1926
	RNO	0.12	0.22	3154	0.20	0.32	2718
Pb	WMW	5.0	20	1804	2.5	8.6	947
	NS&T	1.8	5.1	1852	0.47	0.85	1927
	RNO	2.3	4.5	3265	1.4	2.4	2764

[a]The 85th percentile concentration is the one below which are the concentrations of 85% of the samples. The median concentration is the 50th percentile.[b]All data used in calculations. NS&T data cover 1986 through 1995. RNO data cover 1979 through 1994.[c]n = Non-zero values in the data sets.

TABLE 3

Comparison of WMW, NS&T, and RNO median and 85 percentile combined mussel and oyster concentrations (μg g^{-1} dry wt)[a].

		Median	85th percentile	*n*
Cr	WMW	2.1	9.5	1048
	NS&T	1.1	2.5	3763
	RNO	–	–	–
Ni	WMW	2.2	5.0	1096
	NS&T	1.9	3.4	3791
	RNO	–	–	–
As	WMW	6.3	15	465
	NS&T	9.1	16	3798
	RNO	–	–	–
Cd	WMW	2.6	12	3275
	NS&T	2.6	5.5	3797
	RNO	1.4	3.7	6066
Hg	WMW	0.30	0.87	1482
	NS&T	0.11	0.23	3758
	RNO	0.15	0.28	5872
Pb	WMW	4.0	16	2751
	NS&T	0.76	3.2	3779
	RNO	1.8	3.7	6029

[a]Footnotes as in Table 2.

show log normal distributions. Low levels and analytical methodology could contribute to this data distribution pattern. Most of the data found for Sn resulted from measurements of tributyltin rather than elemental Sn and few data were found for Sb and TI. No conclusions could be drawn for these three elements.

Comparison with long-term mussel watch programs

O'Connor and Beliaeff (1995) summarized NS&T data using geometric means and geometric means plus one standard deviation to define 'high' concentrations. This was based on the log normal nature of the data distributions and the fact that the concentration range beyond one standard deviation from the mean contained about 17% of the data. A simpler way to summarize the data is to use medians and 85th percentiles. These in turn are similar for the NS&T data and do not require the assumption that the data be log normal. Table 2 lists the medians and 85th percentile concentrations ('highs') for trace metals from the WMW, NS&T, and RNO data sets.

There was generally good agreement for medians among all three data sets. The RNO medians and 'highs' appear to be the lower of the three sets of data. Beliaeff *et al.* (1997) compared the NS&T and RNO data in detail and explained differences in national medians in terms of different patterns of metal use. The WMW 'highs' tend to be higher than their NS&T and RNO counterparts. This is probably due to the fact that the latter two programs emphasize collection of mollusks at representative sites rather than within small areas of extreme contamination such as near waste discharges. Within the WMW compilation, on the other hand, some data came from programs specifically designed to sample 'hot spots' and these are evident at the upper end of the concentration distributions.

TABLE 4

Concentrations of trace metals in mussels and oysters that are indicative of contamination (μg g^{-1} dry wt).

Metal	Concentration
Mussels and oysters	
Cr	2.5
Ni	3.4
As	16
Cd	3.7
Hg	0.23
Pb	3.2
Mussels	
Cu	10
Zn	200
Ag	0.75
Oysters	
Cu	300
Zn	4000
As	5

Compilation of all data for mussels and oysters

As previously discussed, mussels and oysters are not equal in their ability to concentrate trace elements. For example, the WMW concentration histograms for Cd and Cu show a bi-modal distribution for Cu due to species difference between oysters and mussels for Cu uptake but no such difference for Cd (Fig. 4). The differences for Cr, Ni, As, Se, Cd, Hg and Pb were not sufficiently high as to prevent the combination of the mussel and oyster data and the mussels and oysters in each of the WMW, NS&T and RNO databases were combined and the medians and 85th percentiles calculated (Table 3). No Cr, Ni or As data were available from RNO. There were insufficient Se data in the WMW database to use meaningfully. The agreements among the medians of the three programs for some elements are very close. Some differences such as the high median and 85th percentile for Pb and high 85th percentile for Cd in the WMW remain. Lower Pb levels in the NS&T data can be explained by the history of use of these metals (Beliaeff *et al.*, 1998). The main conclusions remain in that differences among medians for the three data sets are relatively small while the WMW data set has higher 'highs'.

Conclusion

There were few data from common locations and several years so no determinations of worldwide temporal trends have been made. Few data were found prior to the 1970s. During the 1970s, there was a significant increase in the number of citations reporting the use of mussels and oysters to determine contamination level and monitor coastal areas. Many of these studies included highly contaminated sites, resulting in very high trace metal values. The high values reported during the 1970s may be the result of increased awareness of coastal contamination and evaluation of contaminates sites, not necessarily of increased pollution levels.

In the absence of human activity, there would be measurable concentrations of trace elements in mussels and oysters. We have no independent basis for declaring any specific concentration as a natural limit, but to a first approximation, we could use the lowest of the 85th percentiles from the NS&T or RNO Mussel Watch programs as indicative of contamination (i.e. elevated by human activity). The WMW 85th percentiles include data from heavily contaminated sites and thus are not comparable to NS&T and RNO percentiles. These concentrations (Table 4) in any mussel or oyster could denote contamination. There are mollusks with lower concentrations that are contaminated. Conversely, there are sites where local mineralogy causes mollusks to have greater concentrations of one or another of these trace elements and are not contaminated. Nonetheless, as a first approximation, these limits can be used by GOOS for classifying concentrations reported by monitoring programs from around the world. Even without classifying, new data can be put into their worldwide context by comparing them with the data summarized here or by inserting them into the WMW database.

The author wishes to thank the chemists that produced the data, over many years, which were used to compile the WMW database, the Staff of the NOAA Central Library for their assistance in locating numerous citations. T. O'Connor for his suggestions, and D. Harris for scanning, proofreading, and his many, many trips to the library.

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