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# Trace Metals in Mussels (*Mytilus edulis*) From the Waddenzee, Coastal North Sea and the Estuaries of Ems, Western and Eastern Scheldt

J. B. Luten, W. Bouquet, M. M. Burggraaf, A. B. Rauchbaar, and J. Rus

TNO-CIVO Food Technology Institute, Institute for Fishery Products TNO, P.O. Box 183, 1970 AD IJmuiden, The Netherlands

It is well known that mussels (*Mytilus edulis*) are good accumulators of trace metals from the aquatic environment in which they live. Mussels are therefore considered to be a suitable tool for the monitoring of trace metal levels in the aquatic environment (Schultz-Baldes 1973; Phillips 1976; Goldberg et al 1978; Meeus-Verdinne et al 1983). So far very few data are available on the trace metal content of mussels from (un)polluted areas in the Netherlands. In 1971/1972 the mercury content of mussels from the Dutch coast has been measured (de Wolf 1972).
Since 1979 the trace metal content of mussels in the Netherlands has been monitored as part of the Joint Monitoring programme for the assessment of the level of marine pollution and the effectiveness of measures taken for its reduction. The Joint Monitoring Programme includes the coordinated monitoring activities of the Conventions of Oslo (1972) and Paris (1974).
The results of the Dutch contribution are given in this paper.

## MATERIALS AND METHODS

In the period 1979-1983 mussels (*Mytilus edulis*) were collected each year in April and October at nine sampling locations distributed over the Waddenzee, coastal North Sea and the estuaries of Ems, Western and Eastern Scheldt (figure 1). Samples were collected by the Netherlands Institute for Fishery Investigations and the Public Works Department. The spawning period of mussels in Dutch waters is in April. Mussels collected in October are considered to be taken in the pre-spawning period. A mussel sample consisted of at least 50 individuals (length 3-7 cm). The individual animals were carefully opened by cutting the aductor muscle. The shell cavity liquor was discarded and the entire remaining shell contents were collected, homogenized and stored at -20°C.
Mercury was determined by cold-vapor atomic absorption spectrometry. After wet digestion of one gram of wet material by 5.0 ml of 70% nitric acid, in a teflon-lined stainless steel decomposition vessel for at least 3 hours at 150°C, the mercury was reduced to elementary mercury by means of $SnCl_2$. The volatile elemental mercury was pre-concentrated on a crumpled gold wire by purging hydrogen through the reduction vessel. The gold wire was then heated to ∿320°C within 30 seconds to evaporate the mercury which was transferred to a HGM-2300 mercury meter by means of a stream of hydrogen.

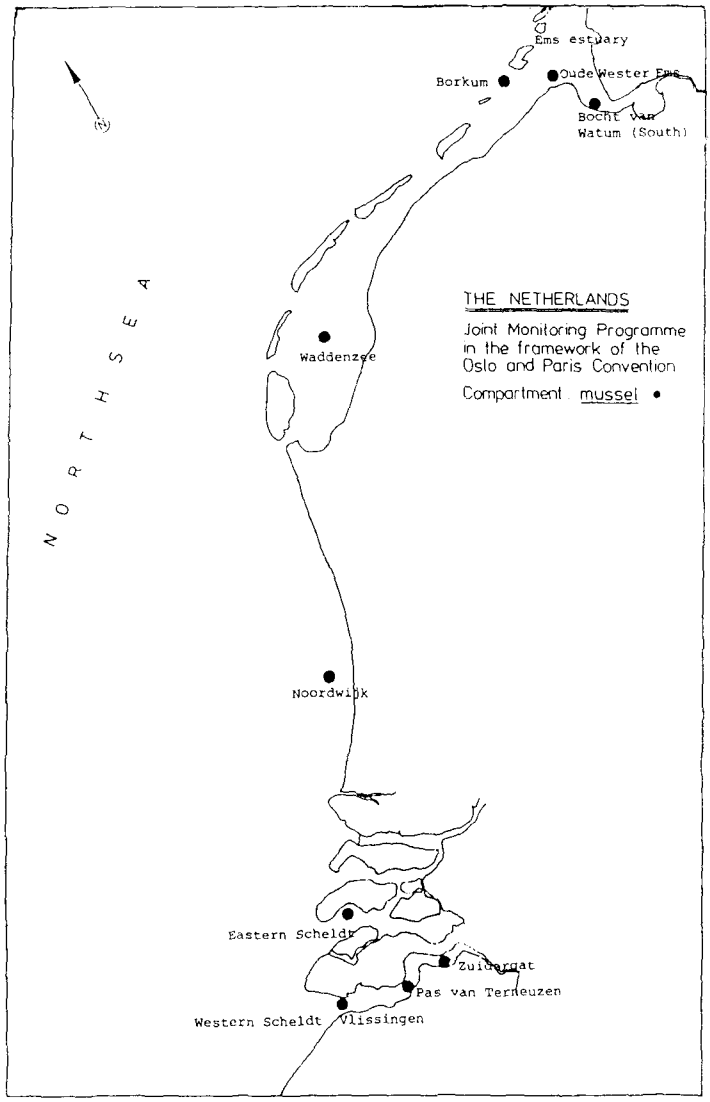


Figure 1. Sampling locations.

Lead and cadmium were determined by differential pulse anodic stripping voltammetry (DPASV). Portions of 5 gram of each sample were mixed with 1 ml of 70% nitric acid and 1 ml of 25% magnesium nitrate solution. Bidistilled water was added if necessary to ensure good mixing. The samples were dried at 150°C on a hot plate and put into a muffle furnace heated to 150°C.

The temperature was slowly increased to a final temperature of 450°C, and the samples were ashed overnight. If necessary the samples were reashed at 450°C for 1 hour after the addition of 1 ml of 70% nitric acid and drying.

The residue of each sample was dissolved with 0.5 ml of 37% hydrochloric acid, then 5 ml of bidistilled water was added and the solution was gently heated on a hot plate. The solution was transferred to a polarographic cell and made up to approx. 12 ml by adding 1 ml of 10%/10% sodium acetate/acetic acid-buffer-solution and bidistilled water. The buffer solution in addition to the sodium acetate and acetic acid contained 20% of citric acid. The pH was adjusted to 3.0 ± 0.2 by adding ammonia (25%).

The DPASV measurements were carried out with the polarographic Analyzer 348 B (EG&G/PAR) and a Model 303 static mercury drop electrode. The polarographic conditions were as follows:

| | |
|---|---|
| initial potential | : -0.85 V (vs Ag/AgCl) |
| final potential | : -0.35 V (vs Ag/AgCl) |
| pulse height | : 50 mV |
| deposition time | : 120 seconds |
| equilibrium time | : 30 seconds |
| drop time | : 0.3 seconds |
| scan increment | : 2 mV |
| drop mode | : large. |

The concentration of lead and cadmium were determined by standard addition method.

Copper and zinc were determined by graphite furnace atomic absorption spectrometry (GFAAS) respectively flame atomic absorption spectrometry (FAAS).
One gram portions of the samples were decomposed in a similar way as used for mercury. After destruction the nitric acid solution was evaporated until dry. Then 0.5 ml of 37% hydrochloric acid and 1 ml bidistilled water were added. The solution was transferred to a polyethylene bottle and made up to 25 ml with bidistilled water. The concentrations of copper and zinc were determined by standard addition method.

## RESULTS AND DISCUSSION

The data on the content of mercury, lead, cadmium, copper and zinc in mussels from the nine sampling locations during the five-year period are given in Table 1 - 5. The median values over the whole sampling period are presented in Table 6.

About 25% of all examined mussel samples have a mercury content exceeding 0.5 mg/kg dry matter. The highest mercury content (1.15 mg/kg dry matter) was observed in the Ems-Dollard estuary. The lowest mercury content (<0.13 mg/kg dry matter) was found in a sample from the Western Scheldt (Vlissingen). The median mercury content of mussels (0.51 mg/kg dry matter) from two sampling

Table 1. The content of mercury (mg/kg dry matter) in mussels at different sampling locations over the period 1979-1983

| Sampling location \ Sampling period | 1979 | | 1980 | | 1981 | | 1982 | | 1983 | |
|---|---|---|---|---|---|---|---|---|---|---|
| | April | October | April | October | April | October | April | October | April | October |
| Ems Dollard-Borkum | - | 1.35 | 0.16 | - | 0.26 | - | 0.37 | 0.22 | 0.26 | 0.24 |
| Ems Dollard-Oude Wester Eems | - | - | 0.16 | 0.33 | 0.40 | - | 0.33 | 0.24 | 0.36 | - |
| Ems Dollard-Bocht van Watum Zuid | - | - | 1.28 | - | 0.71 | 0.36 | 0.53 | 0.20 | 0.38 | 0.18 |
| Eastern Scheldt | 0.42 | 0.25 | 0.18 | 0.21 | 0.32 | 0.26 | 0.39 | 0.15 | 0.33 | 0.40 |
| Western Scheldt-Vlissingen | 0.28 | 0.57 | 0.37 | 0.18 | 0.45 | 0.25 | 0.40 | <0.13 | 0.29 | 0.25 |
| Western Scheldt-Pas van Terneuzen | 0.62 | 0.39 | 0.37 | 0.64 | 0.97 | 0.70 | 0.57 | 0.34 | 0.47 | 0.35 |
| Western Scheldt-Zuidergat | 0.36 | 0.46 | 0.40 | 0.42 | 0.81 | 0.99 | 0.56 | 0.36 | 0.51 | 0.68 |
| Coastal North Sea-Noordwijk | 0.13 | 0.53 | 0.39 | 0.15 | 0.71 | 0.23 | 0.30 | 0.16 | 0.20 | 0.29 |
| Waddenzee | 0.20 | 0.30 | 0.38 | 0.20 | 0.57 | 0.18 | 0.53 | 0.17 | 0.81 | 0.20 |

Table 2. The content of lead (mg/kg dry matter) in mussels at different sampling locations over the period 1980-1983

| Sampling location \ Sampling period | 1980 | | 1981 | | 1982 | | 1983 | |
|---|---|---|---|---|---|---|---|---|
| | April | October | April | October | April | October | April | October |
| Ems Dollard-Borkum | 2.1 | - | 3.6 | - | 3.2 | 1.6 | 4.3 | 2.5 |
| Ems Dollard-Oude Wester Eems | 2.2 | 4.0 | 2.8 | - | 3.1 | 1.5 | 3.5 | - |
| Ems Dollard-Bocht van Watum Zuid | 4.1 | - | 5.1 | 3.0 | 3.2 | 1.9 | 3.7 | 2.9 |
| Eastern Scheldt | 4.8 | 2.3 | 4.1 | 2.2 | 3.8 | 1.3 | 3.2 | 4.4 |
| Western Scheldt-Vlissingen | 7.4 | 5.7 | 7.7 | 5.9 | 5.3 | 2.4 | 10.9 | 6.2 |
| Western Scheldt-Pas van Terneuzen | 6.7 | 12.9 | 9.7 | 4.6 | 8.4 | 2.4 | 4.7 | 3.5 |
| Western Scheldt-Zuidergat | 5.6 | 7.6 | 6.4 | 5.2 | 8.4 | 3.5 | 5.5 | 4.5 |
| Coastal North Sea-Noordwijk | 4.3 | 2.9 | 5.1 | 3.0 | 4.7 | 2.1 | 4.8 | 3.6 |
| Waddenzee | 4.2 | 3.4 | 4.3 | 2.7 | 2.9 | 2.1 | 3.7 | 2.5 |

Table 3. The content of cadmium (mg/kg dry matter) in mussels at different sampling locations over the period 1979-1983

| Sampling location \ Sampling period | 1979 | | 1980 | | 1981 | | 1982 | | 1983 | |
|---|---|---|---|---|---|---|---|---|---|---|
| | April | October | April | October | April | October | April | October | April | October |
| Ems Dollard-Borkum | - | 1.5 | 1.8 | - | 3.8 | - | 1.3 | 0.76 | 1.4 | 0.82 |
| Ems Dollard-Oude Wester Eems | - | - | 1.8 | 1.8 | 5.3 | - | 1.3 | 0.77 | 1.4 | - |
| Ems Dollard-Bocht van Watum Zuid | - | - | 3.8 | - | 3.7 | 0.72 | 1.8 | 1.3 | 1.5 | 0.65 |
| Eastern Scheldt | 2.1 | 0.58 | 2.3 | 1.6 | 3.2 | 0.68 | 1.6 | 0.62 | 1.2 | 0.98 |
| Western Scheldt-Vlissingen | 8.5 | 15.9 | 4.3 | 4.6 | 7.7 | 2.3 | 3.7 | 2.3 | 2.6 | 2.6 |
| Western Scheldt-Pas van Terneuzen | 19.4 | 2.4 | 20.9 | 47.7 | 41.9 | 17.7 | 18.6 | 14.2 | 14.7 | 9.9 |
| Western Scheldt-Zuidergat | 21.4 | 14.4 | 53.2 | 88.9 | 52.4 | 28.4 | 42.2 | 18.6 | 28.0 | 29.7 |
| Coastal North Sea-Noordwijk | 7.4 | 1.6 | 6.5 | 2.6 | 4.7 | 1.3 | 1.6 | 1.1 | 1.6 | 2.3 |
| Waddenzee | 2.2 | 0.50 | 2.3 | 0.60 | 5.2 | 0.64 | 1.2 | 0.52 | 0.88 | 0.67 |



Table 4. The content of copper (mg/kg dry matter) in mussels at different sampling locations over the period 1980-1983

| Sampling location \ Sampling period | 1980 | | 1981 | | 1982 | | 1983 | |
|---|---|---|---|---|---|---|---|---|
| | April | October | April | October | April | October | April | October |
| Ems Dollard-Borkum | 21.9 | - | 28.9 | - | 6.1 | 5.4 | 6.6 | 5.2 |
| Ems Dollard-Oude Wester Eems | 18.2 | 19.6 | 12.2 | - | 4.8 | 5.0 | 8.2 | - |
| Ems Dollard-Bocht van Watum Zuid | 20.8 | - | 15.9 | 6.0 | 6.6 | 8.0 | 7.2 | 7.1 |
| Eastern Scheldt | 20.5 | 15.7 | 10.2 | 14.9 | 4.9 | 7.7 | 10.6 | 9.2 |
| Western Scheldt-Vlissingen | 19.8 | 18.5 | 11.5 | 9.2 | 4.5 | 6.4 | 8.9 | 7.5 |
| Western Scheldt-Pas van Terneuzen | 28.4 | 25.8 | 12.9 | 9.5 | 5.9 | 7.4 | 14.7 | 8.2 |
| Western Scheldt-Zuidergat | 29.0 | 25.0 | 14.5 | 12.8 | 7.1 | 9.0 | 13.1 | 10.1 |
| Coastal North Sea-Noordwijk | 17.5 | 15.8 | 8.9 | 10.2 | 5.0 | 6.8 | 7.9 | 8.0 |
| Waddenzee | 27.8 | 19.0 | 15.2 | 7.3 | 4.5 | 6.4 | 6.9 | 10.0 |

Table 5. The content of zinc (mg/kg dry matter) in mussels at different sampling locations over the period 1980-1983

| Sampling location \ Sampling period | 1980 | | 1981 | | 1982 | | 1983 | |
|---|---|---|---|---|---|---|---|---|
| | April | October | April | October | April | October | April | October |
| Ems Dollard-Borkum | 83 | - | 99 | - | 117 | 87 | 151 | 106 |
| Ems Dollard-Oude Wester Eems | 107 | 53 | 101 | - | 115 | 71 | 145 | - |
| Ems Dollard-Bocht van Watum Zuid | 168 | - | 124 | 90 | 132 | 100 | 119 | 82 |
| Eastern Scheldt | 84 | 50 | 121 | 68 | 125 | 57 | 140 | 150 |
| Western Scheldt-Vlissingen | 142 | 88 | 224 | 113 | 120 | 100 | 172 | 169 |
| Western Scheldt-Pas van Terneuzen | 119 | 194 | 290 | 215 | 186 | 227 | 280 | 269 |
| Western Scheldt-Zuidergat | 121 | 236 | 234 | 213 | 211 | 216 | 297 | 284 |
| Coastal North Sea-Noordwijk | 234 | 81 | 148 | 149 | 121 | 104 | 318 | 201 |
| Waddenzee | 76 | 75 | 114 | 45 | 100 | 87 | 99 | 80 |



Table 6. The median content (mg/kg dry matter) of mercury, lead, cadmium, copper and zinc in mussels at different sampling locations over the period 1979/1980-1983

| Sampling location | Mercury | Lead | Cadmium | Copper | Zinc |
|---|---|---|---|---|---|
| Ems Dollard-Borkum | 0.26 | 3.2 | 1.4 | 6.6 | 106 |
| Ems Dollard-Oude Wester Eems | 0.33 | 3.1 | 1.8 | 12.2 | 107 |
| Ems Dollard-Bocht van Watum Zuid | 0.38 | 3.2 | 1.5 | 7.2 | 119 |
| Eastern Scheldt | 0.32 | 3.8 | 1.6 | 10.6 | 121 |
| Western Scheldt-Vlissingen | 0.29 | 6.2 | 4.3 | 9.2 | 142 |
| Western Scheldt-Pas van Terneuzen | 0.57 | 6.7 | 19.4 | 12.9 | 227 |
| Western Scheldt-Zuidergat | 0.51 | 5.6 | 29.7 | 13.1 | 234 |
| Coastal North Sea-Noordwijk | 0.29 | 4.3 | 2.3 | 8.9 | 149 |
| Waddenzee | 0.30 | 3.4 | 0.88 | 10.0 | 87 |

stations of the Western Scheldt (Pas van Terneuzen and Zuidergat) was about 2 times the median value (0.29 mg/kg dry matter) of all mercury data in mussels from the other seven sampling locations.

The lead contents in mussels from Ems-Dollard, Eastern Scheldt and Waddenzee did not significantly differ from each other over the sampling period. The median lead content varied from 3.1 - 3.8 mg/kg dry matter. The median lead content in mussels from the coastal North Sea (Noordwijk) is somewhat higher (4.3 mg/kg dry matter). Mussels from the Western Scheldt however contained significantly more lead than mussels from the other sampling locations. Their median lead content varied from 5.6 - 6.7 mg/kg dry matter.
The highest lead content (12.9 mg/kg dry matter) has been found in a mussel sample from the Western Scheldt. The lowest lead content (1.4 mg/kg dry matter) was present in a sample from the Eastern Scheldt.
It seems that the lead content in spawning mussels (April) is higher than the lead content in mussels sampled in the foregoing pre-spawning period (October).

From Table 3 it is obvious that the accumulation of cadmium in mussels from the Western Scheldt sampling locations is much higher than in mussels from the other sampling locations. The median cadmium content ranges from 4.3 - 29.7 mg/kg dry matter in mussels from the various sampling locations in the Western Scheldt. The highest cadmium content (89 mg/kg dry matter) has been obtained in a mussel sample from Zuidergat. The median cadmium content in mussels from the Waddenzee (0.88 mg/kg dry matter) is more than 30 times lower than the median value of cadmium in mussels from the Zuidergat location.
The median cadmium content in mussels from the North Sea (2.3 mg/kg dry matter) is somewhat higher than the various median cadmium values in mussels from the Ems Dollard estuary (1.4 - 1.8 mg/kg dry matter), Eastern Scheldt (1.6 mg/kg dry matter) and Waddenzee (0.88 mg/kg dry matter).

In the Western Scheldt estuary the cadmium results show a downward gradient from Zuidergat towards the open sea (location Vlissingen).
As observed with lead, the cadmium content in most of the mussel samples taken in the April period is higher than in samples in the foregoing October sampling period.

The copper content of mussels samples in 1982 and 1983 is generally lower than the copper content in the sampling period 1980/1981. The highest copper content (29.0 mg/kg dry matter) has been found in mussels from the Western Scheldt. A mussel sample from the Western Scheldt (Vlissingen) and Eastern Scheldt contained the lowest copper content (4.5 mg/kg dry matter).
There is no significant difference between the copper content of the various sampling locations due to the relatively large fluctuation of the copper content of mussels within each sampling station.

The median zinc content of mussels from the Waddenzee (87 mg/kg dry matter) is 2 - 3 times lower that the median zinc content in mussels from two sampling locations of the Western Scheldt (Pas van Terneuzen (227 mg/kg dry matter) and Zuidergat (234 mg/kg dry matter)). The median zinc content in mussels from Eastern Scheldt and Ems Dollard estuary ranges from 106 - 121 mg/kg dry matter.

From the results it is concluded that the trace metal content in mussels from the Western Scheldt is higher than the content of trace metals in mussels from the other investigated sampling locations.

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