Concentration of arsenic in blue mussels (*Mytilus edulis*) from the Belgian coast

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Summary
In the period 1986-1991 mussels were collected each year (except 1987) from three jetties along the Belgian coast.
A positive regression between the length of the mussels and the arsenic concentration was found. In the western and middle parts of the coast, arsenic levels increased significantly during the past five years (up to 25 % per year). There was also a decreasing concentration gradient from west to east. The reasons for these differences are not clear.
On the other hand, arsenic concentrations recorded in the present work are in the same range as those reported in other areas i.e. 4.5 to 12 mg/kg dry weight and can be considered as normal.

1. Introduction

Many marine organisms contain appreciable amounts of arsenic in their tissues. The major arsenicals in fish and crustaceans have been shown to be water soluble organic compounds (especially arsenobetaine), which are much less toxic than the inorganic salts (Shinagawa et al., 1983; Kaise et al., 1985). A limited number of studies have also indicated the prevalence (at least 90 %) of organic arsenicals in bivalves, although it still remains largely to be elucidated in what form those filter-feeding organisms deriving nutrition largely from phytoplankton, contain their arsenic burden (GESAMP, 1988). In Japan, *Mytilus edulis* was found to contain only 0.5 % inorganic arsenic (Shiomi et al., 1984).
Blue mussels are considered to be good indicator organisms for environmental pollution (Bryan, 1980; Szefer et al., 1985).
Within the framework of the Belgian monitoring programme on trace contaminants, the analysis of arsenic in fish and shellfish was included from 1984 onwards. This paper reports results with mussels for the five years' period 1986-1991. Data on fish and shrimp for the period 1984-1988 have already been published (De Clerck et al., 1990).

2. Material and methods

2.1. Mussel samples

Mussels were collected in September from three jetties along the coast situated at Nieuwpoort (west), Oostende (middle) and Blankenberge (east). Samples were taken every year except in 1987.
They were divided into four length classes : 20-29 mm, 30-39 mm, 40-49 mm and 50-59 mm. From each category, 40 to 50 specimens were taken. They were kept for 24 hrs in running sea water to eliminate impurities. The mussel flesh was removed from the shell, drained and homogenized.
All samples were kept at -28°C until analysis.
Table 1 reports the average dry matter content of the samples.

2.2. Determination of arsenic

1 g of soft mussel tissue was placed into a high pressure teflon lined acid digestion bomb, Parr 4746. After the addition of 2 ml

Table 1 **Average dry matter content of blue mussels (%)**

Sampling point	1986	1988	1989	1990	1991
Nieuwpoort	27.6	29.1	25.2	24.4	23.1
Oostende	22.1	27.9	29.3	26.6	26.5
Blankenberge	27.4	28.8	27.5	26.8	27.3
Total average	26.0	28.6	27.3	25.9	25.6

Table 2 Arsenic concentrations in blue mussel (mg/kg dry weight)

Sampling point	Length (mm)	1986	1988	1989	1990	1991
Nieuwpoort	20-29	4.55	6.29	7.90	10.21	11.67
	30-39	4.91	7.03	8.86	10.48	10.54
	40-49	5.30	7.02	9.49	9.12	10.30
	50-59	5.60	6.41	10.13	10.66	10.48
Oostende	20-29	6.36	7.70	6.76	9.06	8.99
	30-39	6.22	6.75	6.45	8.58	8.68
	40-49	5.70	6.35	5.87	9.74	8.07
	50-59	6.62	6.65	7.09	11.92	7.35
Blankenberge	20-29	5.50	6.87	6.52	7.14	5.89
	30-39	5.33	6.15	8.19	7.02	6.21
	40-49	6.28	6.55	7.31	8.46	6.08
	50-59	7.32	7.56	6.74	7.52	6.43

Table 3 Time trend analysis of arsenic concentrations (a)

Sampling point (b)	Correlation coefficient	Slope	Trend line equation (c)
Nieuwpoort	0.950***	1.23***	y = 1.23x + 4.94
Oostende	0.642**	0.57**	y = 0.57x + 5.96
Blankenberge	N.S.	N.S.	—

(a) ** = significant at the 99 % level;
*** = significant at the 99.9 % level;
N.S. = not significant
(b) n = 20 per sampling point
(c) y = concentration of arsenic
x = time in years

nitric acid the bomb was heated in an oven at 150°C for 1 hour. After cooling, clear solutions were diluted with water to 10 ml. Arsenic was determined by atomic absorption with graphite furnace, Zeeman background correction and Pd modifier (Hoenig et al., 1986).

3. Results and discussion

The arsenic concentrations per sampling point, per length interval and per year are reported in Table 2. Figure 1 shows the evolution of the average concentrations during the five years' period.

A positive regression between the length and the arsenic content was found. The correlation coefficient for n = 60 was 0.224 and significant at the 95 % level. For this computation, all data were divided by the respective annual means to avoid the variability caused by the increase in concentrations in time (see further).

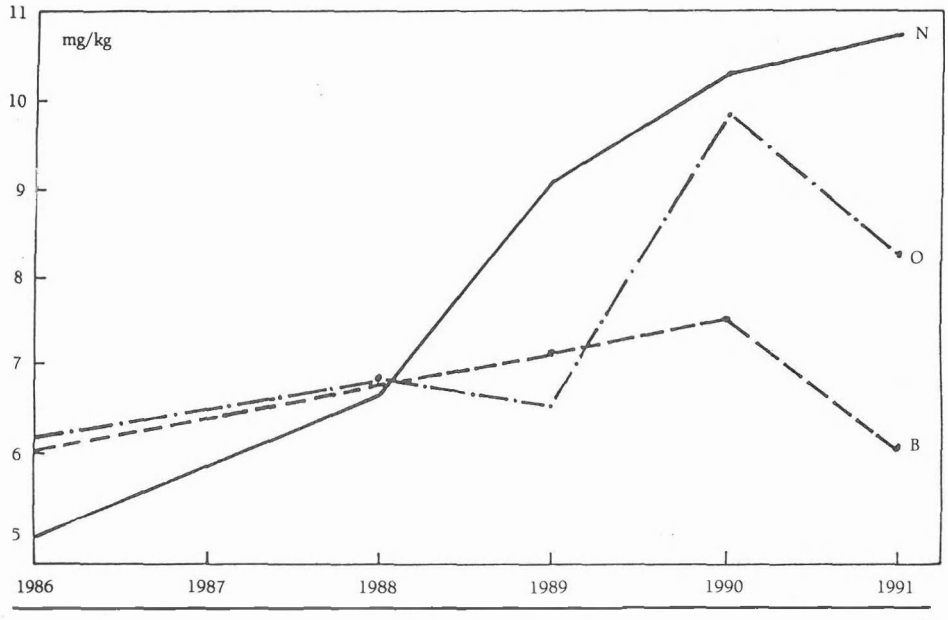
In previous work a similar positive relationship was found for cadmium. This was not so for other trace metals (Vyncke et al., 1988).

Touche et al. (1982) also found that the tissue burden of arsenic in blue mussels from Oregon, U.S.A. correlated positively with size.

The apparent increase in arsenic levels (Figure 1) in mussels from Nieuwpoort and Oostende was confirmed by statistical time trend analysis. The significance of the correlation coefficient and of the slope (difference from 0) were also tested (Table 3). Blankenberge was not significantly different. However during the four years' period 1986-1990, a significant increase was also observed there (r = 0.640** and slope = 0.36***). The annual average increase was about 25 % for Nieuwpoort and 10 % for Oostende.

Applying analysis of variance on the time -

Figure 1 **Evolution of average arsenic concentrations in mussels collected at Nieuwpoort (N), Oostende (O) and Blankenberge (B)**



corrected data from the three sampling points (see above), a significant difference at the 95 % level was found, indicating a decreasing concentration gradient from west to east.

Arsenic enters the marine environment mainly from the atmosphere, especially due to the burning of fossil fuels and the smelting of ores. More local inputs from rivers are caused by pesticide residues, liquid waste from metal foundries, and a variety of industrial and domestic sources such as antifouling paints (GESAMP, 1988; ICES, 1988b).

Arsenic is present in sea water predominantly as arsenate. Marine biota in lower trophic level (bacteria, plankton) however can transform inorganic arsenicals into organic ones (GESAMP, 1988; ICES, 1988b).

Due to the relatively short distances between the sampling points (about 20 km), differences in atmospheric arsenic loads are unlikely to be the cause of the higher concentrations westwards. It would thus be worthwhile to investigate the origin of the direct arsenic inputs to the marine environment via the river Yser and/or the coastal waters west of Nieuwpoort (e.g. the Dunkirk industrial area) and the reasons for the significant increase in arsenic levels over the past five years.

The arsenic concentrations recorded in the present work are in the same range as those reported in other areas.

In the Ems-estuary and in the Western Scheldt, mean values of 5.0 and 6.6 mg/kg dry weight respectively were reported (Akkerman et al., 1990).

In the Eastern Scheldt levels varied from 1.4 to 4.6 mg/kg wet weight, with an average of 2.5 mg/kg, corresponding to approximately 10 mg/kg dry weight (Vos et al., 1986).

In Japan blue mussels contained between 1.7 and 3.1 with an average of 2.4 mg/kg wet weight, corresponding also to about 10 mg/kg dry weight (Shiomi et al., 1984).

During a baseline survey coordinated by ICES in 1985 in areas covering the Dutch coast, the sea areas around Denmark, several sites in the Baltic Sea and several locations in the U.S.A., arsenic concentrations ranged from 1.6 to 12.8 mg/kg dry weight with the median value at 4.0 mg/kg (ICES, 1988a).

In a similar exercise, carried out in 1990, values in the Scheldt estuary and the Wadden Sea ranged from 3.8 to 13.6 mg/kg with

a median value of 8.6 mg/kg (ICES, unpublished results). These levels do not present a health hazard (GESAMP, 1988). Finally, it should be stressed that the arsenic levels found in mussels were markedly lower than those in fish and shrimp caught off the Belgian coast in about the same period. On average cod (*Gadus morhua*) and flounder (*Platichthys flesus*) contained twice as much arsenic and shrimp (*Crangon crangon*) three times the concentration in mussels. With these species however, no clear time trend could be observed (De Clerck et al., 1990).

Conclusion.

Arsenic levels in blue mussels from the Belgian coast increased over the past five years except in the eastern part. There was also an eastward decreasing gradient. The reasons for this situation are not clear. However, the range of concentrations can be considered as normal at the present time.

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Samenvatting

Arseenconcentraties in mosselen (Mytilus edulis) van de Belgische kust

In de periode 1986-1991 werden ieder jaar (uitgenomen in 1987) mosselen op drie strandhoofden langs de Belgische kust bemonsterd.
Een positieve regressie tussen de lengte van de mosselen en het arseengehalte werd gevonden. In het Westelijke en Centraal gedeelte van de kust stegen de arseenconcentraties significant gedurende de afgelopen vijf jaren (tot 25 % per jaar). Een dalende concentratiegradiënt van West naar Oost werd tevens vastgesteld. De redenen voor deze verschillen zijn niet duidelijk. Anderzijds waren de tijdens dit onderzoek waargenomen arseengehalten van dezelfde grootteorde als deze voor andere gebieden vermeld, nl. 4,5 tot 12 mg/kg droog gewicht en kunnen als normaal worden beschouwd.

Résumé

Concentrations d'arsenic dans les moules (Mytilus edulis) de la côte belge

Dans la période 1986-1991 des moules ont été récoltées chaque année (excepté en 1987) sur trois brise-lames le long de la côte belge.
Une régression positive entre la longueur des moules et le taux d'arsenic a été trouvée.
Dans les parties occidentale et centrale de la côte, les concentrations d'arsenic ont augmenté d'une manière significative pendant les cinq dernières années (jusqu'à 25 % par an). Un gradient de concentration diminuant d'ouest en est a également été constaté. Une explication de ce phénomène n'a pu être trouvée.
D'autre part, les niveaux d'arsenic observés dans la présente étude se situent dans la même fourchette que celle rapportée pour d'autres régions, soit 4,5 à 12 mg/kg de poids sec. Ils peuvent être considérés comme étant normaux.