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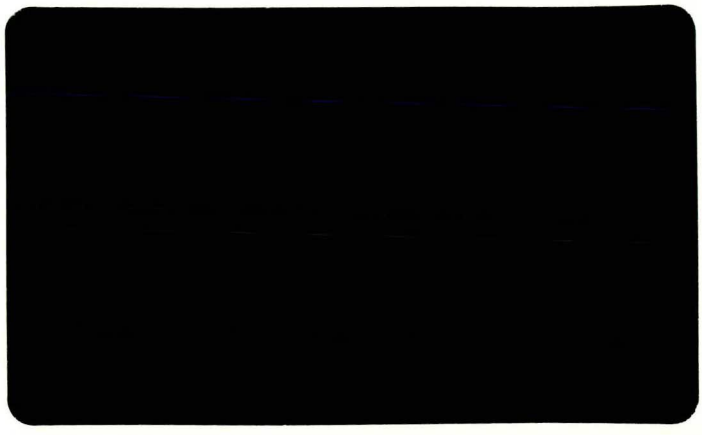

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AN EVALUATION OF DREDGING IN THE WESTERN SCHELDT (THE NETHERLANDS) THROUGH BIOASSAYS

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ABSTRACT

In order to evaluate the impact on biota of contaminated dredged materials from the eastern part of the Western Scheldt estuary, a flow-through solid phase bioassay was conducted. Although dredging and disposal may influence an ecosystem in various ways, only unconfined aqueous disposal was taken into consideration at this point. Furthermore, the experiments were focussed on the mobility of bioavailable contaminants (heavy metals, PCBs, PAHs and pesticides).

Three species of organisms were used:

- *Arenicola marina*, the lug worm, a heavily bioturbating species and
- *Macoma balthica*, the baltic tellin. Both species are deposit feeders and live in the sediments.
- *Mytilus edulis*, the blue mussel. This species is a filter feeder; direct contact of this species with the sediments was prevented.

The main experiment was conducted with 5 experimental systems (2.5 m^3 each) containing a layer of about 0.40 m of sediment (1 m^3) of Zandvliet, Bath, Valkenisse, Konijnenschor and Rotterdam.

Side experiments with the Zandvliet sediment were performed simultaneously in three additional systems. In these experiments we studied the ways bioavailability and mobility were influenced by bioturbation, water exchange rate and fractionation of sediment into a coarse and a fine grain size fraction.

Main experiment

Bioavailability for deposit feeders of contaminants in sediments from silts at Zandvliet and Bath was found to be higher than that of the sediment from an anticipated disposal area (Konijnenschor). In this respect sediments from the silt at Valkenisse are comparable to sediments from the disposal area and from the western Wadden Sea. Both sediments from Zandvliet and Bath released considerable amounts of bioavailable cadmium, and smaller amounts of

some PAH and PCB components to the waterphase. Sediments from Zandvliet and Rotterdam are somewhat comparable to each other in as much that they contain contaminants with a high bioavailability to deposit feeders, although the relative distribution of several contaminants is quite different.

Side experiments

Bioturbation

Bioturbation by the lug worm did not influence availability of contaminants to the baltic tellin or the blue mussel to any significant extent.

Water exchange rate

A 10 times higher water exchange rate (2% versus 20% per hr) resulted in a considerably reduced uptake of cadmium by the blue mussel and the baltic tellin. However, this did not have any effect on the bioaccumulation of PCBs, which proved to be equally elevated above blanks under both exchange conditions.

Fractionation of sediments

Fractionation of Zandvliet sediments into two grain size classes (one with larger and one with smaller grain sizes than 100 μm) lead to a highly contaminated fine fraction and a hardly contaminated coarse fraction. Remarkably, bioavailability of PCBs and heavy metals to the baltic tellin proved to be about the same for both fractions.

Ecotoxicological observations

No adverse effects were noticed in relation to the accumulation of contaminants in any of the exposed species. On the contrary, the best growth and best condition were observed in animals exposed to the most contaminated sediments (Zandvliet and Rotterdam). This finding has far reaching ecotoxicological implications because all three species tested are a prime foodsource for many others (including fishes and birds). Disposal of highly con-

taminated sediments, but rich in organic matter as well, might lead to an increased production of contaminated food for more sensitive valuable species. The lugworm accumulated much higher concentrations of PCBs, PAHs and pesticides than the other two species, but did not respond to heavy metals. For this reason, but also because of its wide occurrence in estuarine habitats and its importance as a foodsource for other species, it is proposed to use the lugworm as a bioassay test species to assess bio-availability of organic micropollutants and the baltic tellin as a bioassay test species for heavy metals.

1. GENERAL INTRODUCTION

Sediments in shallow coastal areas of industrialized countries retain an important fraction of the aquatic contaminant input. In such areas, deposition of particulate trace metals and organic contaminants takes place under conditions of low turbulence. Sedimentation and a more recent demand for deeper shipping channels has led to intensive dredging operations in both marine and freshwater harbor environments. Many sediments to be dredged may prove to be moderately contaminated and only some may be severely contaminated, as shown for a case of PCB contamination of sediments (figure 1).

Whether or not disposal of contaminated dredged materials will degrade the environment is a question strongly dependent on the choice of disposal alternatives and the management of both the disposal operation and the disposal site. In general, three disposal alternatives are considered: aquatic disposal, wetland creation, and upland disposal. In table 1 these alternatives are shown together with some implications and uses. It is clear the unconfined subaqueous (aquatic) disposal can effect large areas along the transportation route of the materials in question. Their fate might lay in deeper oceanic parts or in wetland and upland sites (figure 2). Dredging and the disposal alternative influence the physico-chemical conditions of the former sediment as well as the physical mobility of the contaminants. Apart from transport of contaminants to overlying waters, the potential for contaminants to accumulate in the atmosphere or groundwater may be either enhanced or decreased.

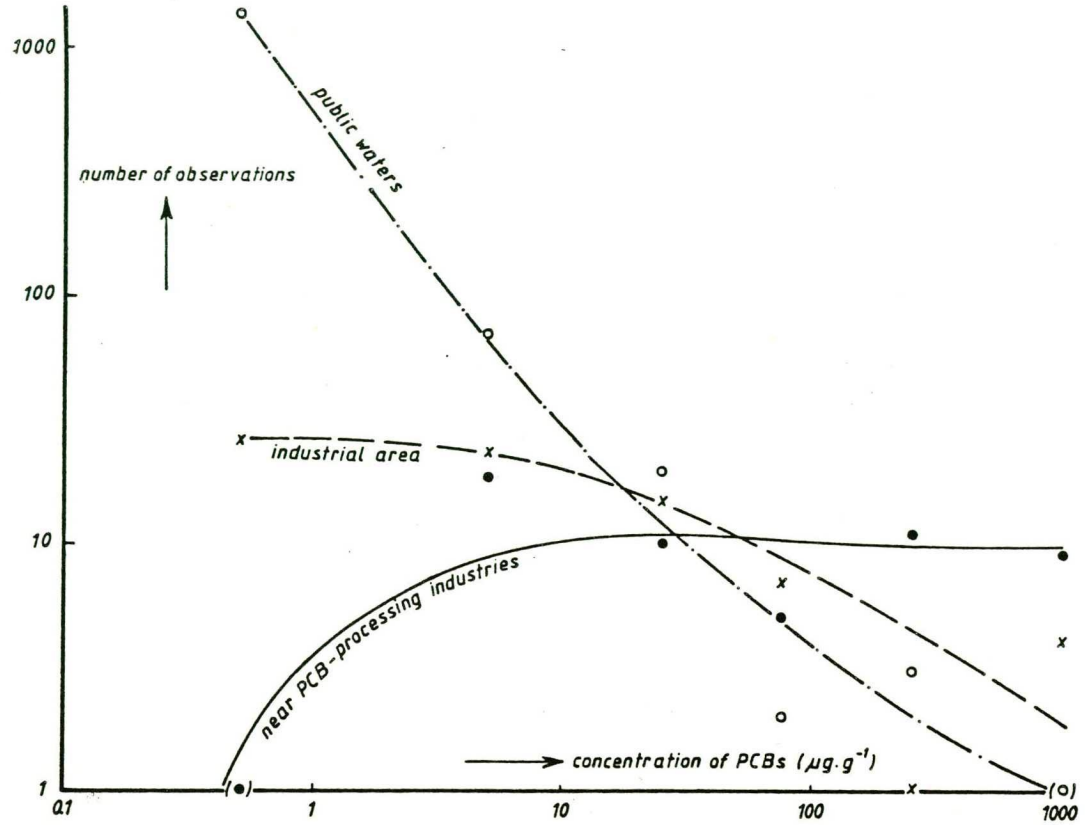

Figure 1 Concentrations of PCBs encountered in sediments related to the number of sediment analyses. Inventory of Japanese waters, 1971 (based on Fujiwara, 1975).

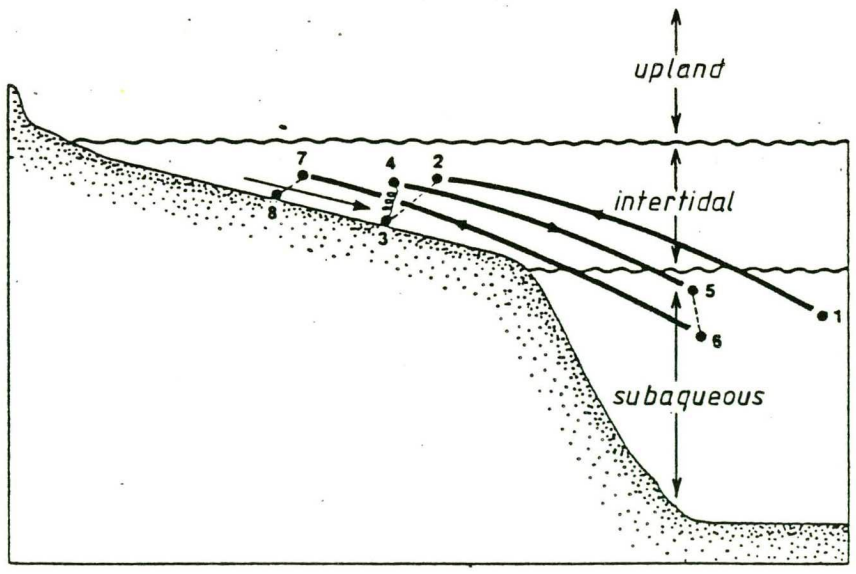

Figure 2 Transport of suspended matter in the Wadden Sea. After Hickel (1979).

Table 1 An outline of disposal alternatives in relation to habitat, use and perspectives.

DISPOSAL ALTERNATIVES

Subaqueous Disposal

1. Disposal not in conflict with important economic or ecological productivity
   - 1.1. Confined
     - 1.1.1. Mounded (isolated) in a stable fashion
     - 1.1.2. Confined in a depression and covered with a layer of uncontaminated material
   - 1.2. Unconfined - wide dispersal with apparent loss from the disposal area
2. Application to especially productive aquatic systems

Application to Intertidal Sites for Disposal or Productive Use

1. Habitat development
   - 1.1. Confined by boundary structure - resistant to erosion
   - 1.2. Unconfined - some erosion and some consolidation (i.e. mudflat, marsh - island complexes)
2. Nonbiological purposes
   - 2.1. Shoreline stabilization/modification (to combine subaquatic discharge with island development).
   - 2.2. Confined boundary structures

Upland Application

1. Long-term confinement for disposal purposes only
   - 1.1. Ponded (no drainage)
   - 1.2. Nonponded (managed to enhance dewatering)
2. Interim confinement for contaminated sediments for dewatering, consolidation, and possibly treatment prior to transport and reuse for productive purposes
3. Unconfined upland - not specifically intended for habitat development, but subject to biotic colonization with time
4. Habitat development - application for specific biological purposes
5. Agricultural soil amendment and land reclamation
6. Use for land fill and other construction/engineering purposes

Anthropogenic displacement of sediments nearly always disrupts an existing plant and wildlife community, and the resulting new surface aera might or might not be recolonized by organisms. Depending on the final disposition of the dredged material, these organisms can be the same species as those originally inhabiting the dredged site, or other species more typical for the selected disposal site. These other species may be aquatic, wetland, and terrestrial organisms as well. For a given species of organism, bioaccumulation depends upon the physico-chemical conditions of the sediment (redox potential, organic matter, etc.) as well as upon differences between species. Because it also has been shown that the potential for contaminants to bioaccumulate cannot be predicted from "bulk" chemical analysis of sediments, bioassay techniques were developed to assess environmental risks in relation to the disposal alternatives.

With reference to sediments and dredged materials, these bioassay techniques were mainly developed, applied, and evaluated by the U.S. Army Corps of Engineers (CE) Waterways Experiment Station (WES) and the Netherlands Organization for Applied Scientific Research (TNO) Division of Technology for Society (MT). Untill 1982, both organizations (the WES and MT-TNO) independently developed and/or applied procedures based on the disposal strategies that were locally confronting their sponsors. From early 1983, these institutes have cooperated with joint research projects under the auspices of the Memorandum of Understanding (MOU) between the US and the Netherlands specifically in the area concerning dredged material. In order to facilitate the maximum exchange of expertise, J.M. Marquenie of the Department of Biology, MT-TNO and Dr. J.W. Simmers, of the Ecosystem Simulation and Research Division of the WES collaborate on different projects.

In 1983 and again in 1984, J.M. Marquenie was stationed at the WES in the U.S.A. for a period of several weeks. In 1984 J.W. Simmers was stationed at MT-TNO in the Netherlands for two periods of about 6 weeks each. In these two periods in the Netherlands cooperative experiments were conducted in cooperation with Ir. E. Birnbaum of the Dutch Ministry of Transport and Public Works (DDMI) while Mr. B. Schrieken of MT-TNO was in charge of the technical installation and sampling coordination and tissue preparation. The project was sponsored by the Netherlands Ministry of Transport and Public Works.

Acknowledgements

Several others contributed to this project and deserve to be mentioned in some kind of chronological order.

- Prof. Dr. D.J. Zandee and coworkers (University of Utrecht) and G. Hoornsman (MT-TNO) for tissue preparation, ash-free dry weights and metal analysis in tissues and Dr. J. Gielen (MT-TNO) and coworkers for PCB, pesticide and PAH analysis, Ir. P. de Jong (MT-TNO) and coworkers for heavy metal analysis in sediments, Prof.dr. W.G. Mook (University of Groningen) and coworkers, for analysis of stable isotopes in some sediment samples, H. van het Groenewoud (MT-TNO) for nutrient analysis, K. de Jong for typing of the manuscript and G. van Velzen and coworkers for technical report production.

- Personnel from the firm Wielemakers and the firm Van de Wal who transported and mixed sediments according to laboratory standards fashion.

1.1. THE WESTERN SCHELDT

The Western Scheldt, connecting the Scheldt River with the North Sea, is situated in the southern part of the Netherlands. The estuary, about 70 km long and in average about 10 km wide, is characterized by a sandy stream bed with dominant erosion and sedimentation processes due to heavy tidal currents. These currents also are responsible for the formation of silts in between tidal flats, intertidal areas and wetlands with prolific wildlife along the banks. The Western Scheldt is the main shipping route to the Port of Antwerp in Belgium. Maintenance dredging is applied in areas were silts built up continuously. A more recent demand for deeper shipping channels will nog only lead to a single increased dredging operation, but is also expected to incrase maintenance dredging. This is due to a further disturbance of equilibrium between present dredging and disposal on the one hand and erosion and sedimentation on the other hand.

As an example maintenance dredging at the silt of Zandvliet amounts one to two million cubic meters annually at the present time; the expected increase after redeepening the channel amounts one million cubic meters.

Several of these silts are threatening shipping traffic, the three most eastern ones being Zandvliet, Bath and Valkenisse.
Dredged materials are disposed of at unconfined subaqueous, disposal sites, somewhat downstream along the banks.

1.2. PROBLEM RECOGNITION

Environmental concern arises from several points. Firstly, dredging and disposal may have adverse effects on the water quality (due to an increased load of suspended matter). Adverse effects due to sediment load are described by Maurer et al. (1981) and Attena et al. (1982) for benthic communities of crustaceans and by Maurer et al. (1982) for polychaeates. For the bivalve, *Spisula solidissima*, Robinson et al. (1984) state that concentrations of suspended matter over 100 $mg.l^{-1}$ are harmful.

Secondly, enhanced contact of anaerobic sediment particles with the superficial water, not only by dredging and disposal but also by the efflux of transportation water may decrease oxygen concentrations and increase concentrations of nutrients and easily leachable contaminants. Thirdly, a displacement of contaminated sediments to areas, less affected by pollution, might endanger wildlife in those areas, depending upon the ability of organisms to colonize the new substrate and the bioavailability of contaminants.

Both deepening the shipping channel and the expected increase in maintenance dredging apply to all three points of concern, especially the latter one, because of the capacity of nearby disposal areas is limited.

At the same time, sediments from the Western Scheldt estuary are rather contaminated (Wollast and Peters, 1978; Salomons and Forstner, 1984). Main sources for the contaminants are the river Rupel (with waste water from Brussels) discharging into the river Scheldt about 90 km upstream and the river Scheldt itself (carrying waste waters from Gendt and Antwerp). Downstream of Antwerp both salinity and oxygen concentration increase (Somville

and De Pauw, 1982). In this region, both adsorption to flocculated material and mobilization due to oxidation of organic matter and an increased salinity take place. Several interactions, including sediment transport through the estuary, result in a gradient in contaminant concentrations, not only in superficial water but also in the sediments. Contaminant specific gradients were also found in organisms exposed in the Western Scheldt estuary (de Kock and Marquenie, 1984; de Kock, 1983) or in plants growing on the banks (Beeftink et al., 1982).

Experimental evidence for the direct accumulation of heavy metals and PCBs from Western Scheldt sediments in organisms was obtained by translocating these sediments to the Eastern Scheldt and to the laboratory (Marquenie et al., 1983, Marquenie, 1985).

1.3. SCOPE OF WORK AND OBJECTIVES

This project serves to evaluate effects of dredging and disposal in the Western Scheldt estuary. It is till now restricted to the present situation of unconfined subaquaous disposal in the eastern part and contaminant biomobility of heavy metals (Cd, Zn, Hg and occassionally Cu), PCBs, and occassionally PAHs and pesticides. The biological effects of fractionation were included in the experiments because of the following: dredgers actually tent to overflow their barges, flushing the lighter materials and retaining the heavier materials for transportation and disposal and moreover fractionation of sediment in a more defined technical way seems feasible and promising from an environmental point of view. The objectives were defined as follows:

1. Assess bioavailability of heavy metals and PCBs to deposit feeders, of sediments from three silts: Zandvliet, Bath and Valkenisse, in comparison with sediments from a disposal site: Konijnenschor, a blank: Wadden Sea and a contaminated reference: Rotterdam Harbor.
2. Assess leaching of bioavailable contaminants from these sediments.
3. Investigate whether water exchange rate and bioturbation influence contaminant availability.

4. Delineate the effects of fractionation as a proposed technique in sediment management.
5. Indicate which PAHs and pesticides are of concern in relation to dredging and bioaccumulation.

2. MATERIALS AND METHODS

2.1. EXPERIMENTAL DESIGN

The experiments were performed at the TNO experimental outdoor test facility "Fort Harsens". The experimental units, circular polyester tanks with a volume of 2.5 m^3 and a height of 0.80 m, resemble a lower version of the MERL mesocosms (at Rhode Island) in order to facilitate light to penetrate to the bottom. Flowing sea water, with a reasonably good quality, can be

Table 2 Experimental design. The table provides a key to the treatment numbers in tables with experimental data. Presence of organisms *Arenicola marina* (A.m.), *Macoma balthica* (M.b.) and *Mytilus edulis* (M.e.) are indicated (+).

Treatment	Sediment	Remarks	Water [1] exchange	Organisms A.m.	M.b.	M.e.
1	-	raw water	17 ton.hr^{-1}	-	-	+
2	-	raw water	17 ton.hr^{-1}	-	-	+
3	Waddenzee	sand filter	2% hr^{-1}	+	+	+
4	Zandvliet (coarse fraction)	sand filter	2% hr^{-1}	-	+	-
5	Zandvliet (fine fraction)	sand filter	2% hr^{-1}	-	+	-
6	Zandvliet	sand filter	2% hr^{-1}	-	+	+
7	Zandvliet	sand filter	20% hr^{-1}	+	+	+
8	Zandvliet	sand filter	2% hr^{-1}	+	+	+
9	Bath	sand filter	2% hr^{-1}	+	+	+
10	Valkenisse	sand filter	2% hr^{-1}	+	+	+
11	Konijnenschor	sand filter	2% hr^{-1}	+	+	+
12	Rotterdam	sand filter	2% hr^{-1}	+	+	+
13	-	blanks (t=0)		+	+	+
14	-	blanks (t=0)		+	+	+

[1]) Percentage of total volume of water on top of sediments.

applied continuously, both filtered through an automatically self-purifying sandfilter (reflux) and/or directly in quantities up to 17 tons/hr. Temperature, salinity and oxygen are monitored continuously.

Experiments can be divided into: main experiment (2.1.1.) in order to reach objectives 1 and 2; side observations within the main experiment (2.1.2.; objective 5); and side experiments (2.1.3.; objectives 3 and 4).

Figure 3 provides a schematic drawing of the experimental facility "Fort Harsens". Figure 3 and table 2 together form an outline of the experimental design. The treatment numbers correspond to numbers in the tables in chapter 3 (results).

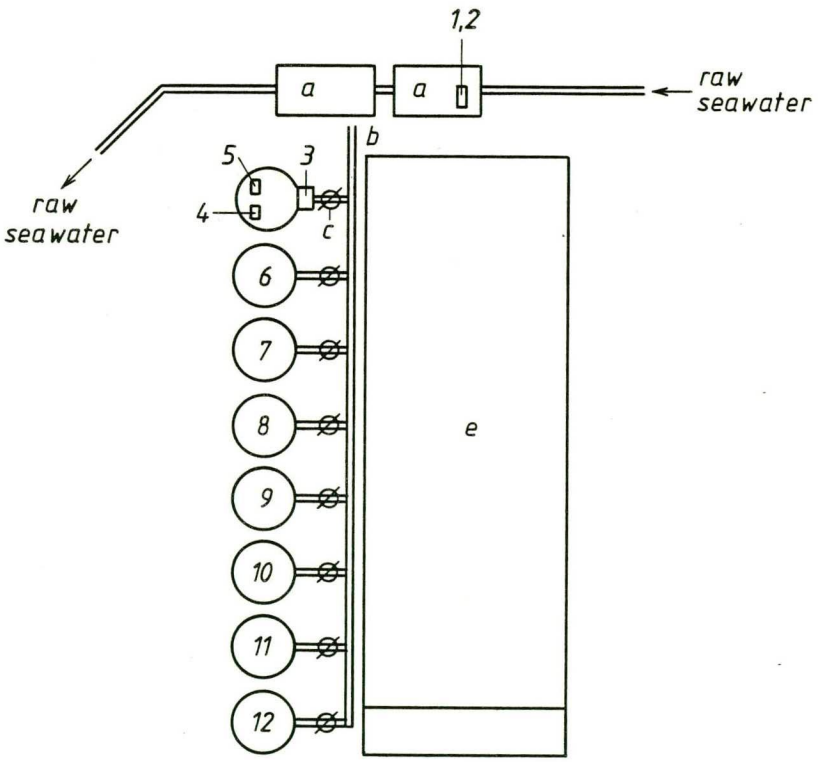

Figure 3 A schedule of the experimental facility Fort Harsens, Den Helder. Numbers refer to treatment numbers (table 2).

a = sand filter
b = filtered seawater
c = adjustable flow meter
d = circular tank with axial inflow and central outflow
e = corrosion laboratory building
3 = refers to treatment 3 in table 2. Small tank under inflow but above standing water level, overflowing in circular tank.

2.1.1. Main experiment
(treatments 1, 2, 3, 8, 9, 10, 11, 12)

Six different sediments were used. Four of these sediments came from the Western Scheldt, from three silts that need to be dredged frequently: Zandvliet, Bath and Valkenisse and from another location Konijnenschor, where dredged materials are being disposed and which is considered for further disposal. Two sediments did not come from the Western Scheldt: one from a relatively clean part of the Wadden Sea, and the other, a standard Rotterdam-dredged material, highly contaminated and also used in a previous experiment. The latter two sediments served both as a contaminated control and to test reproducibility of the bioassay. The sediments were translocated from the Western Scheldt to our experimental facility in Den Helder, carefully mixed per location in quantities of up to 3 m^3 and put into the 2.5 m^3 systems. After consolidation (10 days) natural but filtered (sandfilter) sea water was supplied continuously at an exchange rate of 2% of the superficial water per hour (about 32 litres per hour).

2.1.2. Side observations
(treatment 8)

It was decided to analyse the Zandvliet sediment and the organisms exposed to it for PAHs and for some pesticides which are not determined in the routine PCB procedure.

Furthermore, throughout the whole experiment, bioavailability of contaminants, the behaviour of organisms and the development of the systems were monitored daily. Growth of organisms during exposure and some physiological parameters at the end of the exposure period were determined as well.

2.1.3. Side experiments
(treatments 4, 5, 6, 7)

Three types of side experiments were conducted within the framework of the main experiment. For all these side experiments Zandvliet sediments were used. Two additional systems were provided with Zandvliet sediment. The influence of water exchange rate was studied in one of these systems (treatment 7) by maintaining an inflow of 20% of the superficial water per hr. instead of 2% as in all other systems. The influence of absence of heavy bioturbation was studied in the other system (treatment 6) by exclusion of the lugworm *A. arenicola* in that system. In a third additional system three smaller (200 l) containers were placed. One of these containers half way filled with Wadden Sea sediment and with the rim somewhat elevated above the waterlevel served as a blank. Filtered water was applied to this container (treatment 3), overflowing into the third additional system. The two other containers were placed below the water level. They served to investigate the influence of sediment fractionation on contaminant speciation and bioavailability.

Zandvliet sediment was fractionated by sieving. A fraction of particles smaller than 100 µm was obtained by vigorous shaking of Zandvliet sediment in nylon netting of the appropriate mess size in the 200 l container and in the transportation water. The fraction retained by the net was used in the other 200 l container. In this way leaching and subsequent loss of contaminants was prevented.

2.2. ORGANISMS

Deposit feeders, *Macoma balthica* and *Arenicola marina* were collected in the Wadden Sea, the former species in the central Wadden Sea, north of Wierum and the latter species in the western Wadden Sea, in the Mokbaai of the island of Texel. The filter feeder, *Mytilus edulis* was collected from a bouy in the North Sea. Although they were supposed to come from about 70 km offshore unfortunately, by some error, they originated from only about 5 km west of Egmond.

Organisms were introduced into the systems after 10 days of equilibration and consilidation. In the main experiment, including the blank, all three species were used in densities of about 50 *M. balthica* and 25 *A. marina* (100 g) per m^2; also 180 *M. edulis* per system were caged above the sediments.
These three species were also used in the side experiment on water exchange rate. The influence of bioturbation was studied in an additional system without *A. marina* as opposed to the main experiment with *A. marina*. The influence of fractionation was studied with *M. balthica* only, in a density of about 200 individuals per m^2.
Two more samples of *M. edulis* were exposed to the raw seawater on top of the sandfilter.

2.3. SAMPLING

Sediment samples for analysis were collected just prior to the stocking of organisms. Core samples (not analyzed) were collected at the end of the exposure period of 60 days. Water samples (filtrate and total) were collected in the middle of this period after 30 days of exposure of the organisms. These samples were only analyzed for nutrients.
Organisms were collected after 60 days of exposure. Some *M. edulis* were also collected midway during the experiment after 30 days; they were however stored but not analyzed. Two samples of each species were stored at -20°C at the beginning of the experiments, to serve as a background control.
M. balthica and *A. marina* were depurated overnight in order to empty guts and mantle cavity of sediment particles.

2.4. ANALYSIS

Prior to analysis all tissue samples were homogenized in the glassware in which they were to be stored. An Ultra Turrax was used to homogenize the tissues. The Ultra Turrax was modified by TNO and equipped with a solid titanium milling device. The homogenates were divided into subsamples for the various analytical procedures. Sediments were thoroughly mixed prior to analysis.

Dry and ash weights

About 1.0 g of wet homogenate was weighted accurately to four decimal points, dried for 16 hrs at 150°C, reweighed, ashed for 4 hours at 600°C, and reweighed again at standard conditions (desicator, 20°C). Percentage of water and ash-free dry material were recorded and used to convert the concentrations of contaminants from a wet weight, to a dry weight basis (sediments) or an ash-free dry weight basis (tissues). The percentage of ash-free dry material from sediments was used as an estimate of the percentage of organic matter.

Heavy metals

Concentrations of cadmium (Cd), zinc (Zn) and mercury (Hg) in sediments were determined with destructive neutron activation analysis (INAA) according to TNO procedure No. 2. Approximately 0.2-0.5 g of wet tissue was exposed to a neutron flux of 10^{13} neutrons.cm^{-2}.s^{-1}. After a wet destruction procedure and a separation on ion exchange resins, energies were measured: Cd at 336 KeV, Zn at 1115 KeV and Hg at 77 KeV.

For a first wide-range screening of elements in the dredged material one sample was selected for analysis by instrumental neutron activation analysis (INAA) according to TNO procedure No. 1.

Organochlorine contaminants

Concentrations of PCBs (polychlorinated biphenyls) and HCB (hexachorobenzene) were determined by gas chromatography. The insecticide Mirex proved to be absent and was used as an internal standard. Sediment samples (5 g, spiked with Mirex) were extracted, and the extracts were cleaned by chromatography over alumina and dried over sodium sulfate. Tissues (1-4 g) were treated in the same way, and proteins were enzymatically broken down.

After clean-up, the extracts were analyzed on a 50 m fused silica CP Sil 13B column with electron capture detection. Each series of 8 samples was accompanied by a blank and three standard samples with different concentrations of HCB and PCB isomers. The PCB isomers determined were those selected as indicators by a Dutch PCB analysis standardization committee on the basis of their occurrence and analytical accessibility.

The sensitivety of the electron capture detector was monitored by including one "GC standard solution" sample, containing all substances to be determined for every five real samples, blanks, and/or standards. Peak identification was based on retention times, and peak height was used for quantification. Any measurements outside the linear range of the detector were avoided. The recovery percentage of the procedure followed here is in the order of 80-90%.

Polycyclic aromatic hydrocarbons (PAHs)

Concentrations of PAHs were determined by high performance liquid chromatography (HPLC). Sediment samples (5 g) were spiked with dibenzo(a,h) anthracene a contaminant not occurring in the samples under study. Samples were extracted, and taken up in methanol. Tissues (2.5 g) were treated with sodium hydroxide, extracted and then cleaned over alumina. After clean-up the extracts were analyzed on a 250 x 4.6 mm reverse phase C18 column with a methanol-water gradient and fluorescence detection. A number of spiked, uncontaminated tissue samples were treated in the same way, as well as a number of blanks. The PAH components that were analyzed are a standard series of 21 components that is routinely analyzed at TNO laboratories.

Peak identification was based on retention times and supported by fluorescence behaviour of the components, and quantification was based on peak height. The recovery percentage of the procedure followed, ranged from 80-120% for tissues and from 80-100% for sediments.

Data Recording Procedure

All chromatographs are directly coupled to a CIS Lab Automation System, which, in return, is coupled to a Hewlett Packard HP-1000 computer. All appropriate data are filed on tape and can be reproduced off line.

3. RESULTS AND DISCUSSION

The results are summarized in several tables. In general no specific problems were encountered. Daily inspections showed that in order to maintain a constant exchange rate, waterflow had to be readjusted only in very few occasions. After about 40 days floating masses of algae were removed in order to prevent clogging of the drainage.
Apart from a single dead lug worm at the beginning of the experiment no mortalities were observed. However, it was noticed, that general burrowing and deposit activity of lugworms was much lesser in the sediments of Rotterdam, Zandvliet and Bath than in the others, probably due to more available food, because they also grew better.

3.1. SEDIMENT CHARACTERISTICS

3.1.1. The origin of the Western Scheldt sediments

Obviously, the sediments were collected at well defined locations. However, they were deposited after transport through currents from other locations and sources. The sediments in question might originate from:

- The river Scheldt directly through natural erosion, atmospheric deposition or from industrial discharges;
- The North Sea as geological deposits or freshly transported through the estuary due to the density flow along the bottom;
- The banks and wetlands by erosion of plant debris.

Apart from morphological methods (for instance microscopic examination for diatom species) an analytical chemical method is frequently used wich discriminates between stable isotopes of carbon, but also of oxygen (Salomons and Mook, 1981). Carbon occurs in nature with mass numbers 12, 13 and 14. The last isotope is radioactive and also used for dating purposes. The other two isotopen are stable but differ slightly in physico-chemical and biological behaviour. About 98.9% of all carbon has a mass of 12. Plants accumulate with some preference ^{12}C above ^{13}C in their tissues. This leads to another ratio in carbon from plants than in carbon in the atmosphere. This ratio R

is known to be specific for different environmental compartments (including organisms). Moreover, within plants a general difference exists between two groups of species that use different physiological pathways (C3 and C4 route of assimilation). Desert plants and halophytes (salt tolerant) use the C4 route. For practical reasons the ratio R is expressed as a relative deviation (delta) from a standard in parts per thousand:

$$(\text{delta}) = \frac{(R \text{ sample} - R \text{ standard})}{R \text{ standard}}$$

Values range from +5 to zero for carbonate and from -12 to -15 for plants using the C4 pathway and from -25 to -30 for plants using the C3 pathway.

Delta values of sediments from the Western Scheldt were assessed for both carbonate and organic matter (Table 3). Based on organic matter, it can be concluded that the materials from Zandvliet and Bath originated for about 88% from the Scheldt river and for about 12% from the North Sea. Sediments from Valkenisse might originate for 94% from the North Sea if a contribution of plants from banks is excluded. However, the results for Konijnenschor indicate a substantial contribution of the wetland in this respect. This means that the influence of marine material might be over-estimated in this case. Results for carbonate are somewhat inconclusive, probably due to several sources, including industrial effluents possible neutralized with limestone, involved.

3.1.2. Composition of sediments

The general composition of sediments is shown in table 4. This table also includes some extra samples collected at Zandvliet (Z1 to Z6), in order to assess the local variation in sediment composition and sediment contamination at that location. Water content (%w) was used to convert contaminant concentrations in raw samples to dry weight. Particle seize distribution of these sediments is given in table 5 and for some sediments shown in figure 4. From this table, (treatments 4, 5 and 8) a mixing ratio coarse/fine (a) was calculated as follows:

$$\text{Zandvliet (total)} = \frac{a\ (\text{Zandvliet coarse}) + (\text{Zandvliet fine})}{a + 1}$$

Table 3 Relative occurrence (°/oo) of stable isotopes in sediment samples.

Sediments	Organic matter σ 13	Carbonate σ 13	Carbonate σ 18
4 Zandvliet (coarse)	- 26.51	+ 0.91	- 1.86
5 Zandvliet (fine)	- 23.56	+ 2.70	- 2.54
8 Zandvliet	- 27.08	+ 0.68	- 2.35
9 Bath	- 27.73	+ 0.82	- 1.70
10 Valkenisse	- 19.54	+ 1.45	- 1.81
11 Konijnenschor	- 15.54	+ 0.82	- 1.73
Reference values			
Scheldt river	- 28.20	- 1.71	- 2.77
North Sea	- 19.00	+ 2.00	
Vegetation on banks	- 12.00		

Table 4 Sample composition. Percentage of water (%w) and organic matter (%a) in raw sediment samples and total oxidazable carbon (%c) and carbonate ($\%CO_3$) based on dry weights.

Treatment	% w	% a	% C	% CO_3
1				
2				
3				
4	24.0	1.04	0.1	4.6
5	62.9	4.08	2.0	11.2
6				
7				
8	37.8	3.49	0.5	5.5
9	25.7	0.95	0.5	4.3
10	25.7	0.80	< 0.1	4.0
11	28.8	1.58	0.4	3.1
12	69.7	4.61	-	-
13				
14				
Z1	17.0	0.62	< 0.1	2.0
Z2	26.6	1.46	0.4	6.1
Z3	20.2	0.81	< 0.1	4.0
Z4	38.2	4.87	2.2	11.4
Z5	20.6	0.78	< 0.1	4.2
Z6	45.9	5.12	2.7	11.1

- = not analyzed.

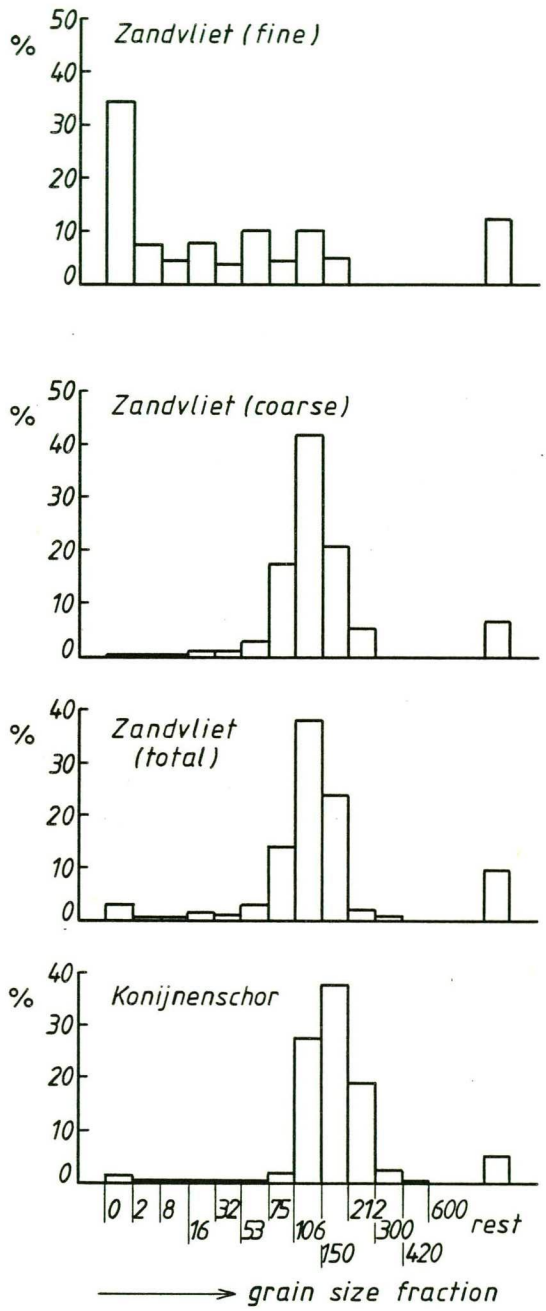

Figure 4 Particle size distribution in some sediment samples.

Table 5 Particle size distribution of sediments. The rest fraction consists of organic matter, carbonates and salts.

Treatment	Particle size upper boundery (µm)												
	2	8	16	32	53	75	106	150	212	300	420	600	rest
4	0.9	0.3	0.2	1.1	0.9	3.2	17.1	42.3	20.9	5.3	0.4	0.1	7.2
5	33.9	7.5	4.6	8.1	3.5	10.3	4.2	10.0	4.7	0.4	0.1	0.1	12.4
8	3.0	0.7	0.6	1.6	1.1	3.2	13.7	38.7	24.0	2.3	0.8	0.2	9.8
9	0.8	0.1	0.1	0.2	0.3	0.8	5.4	39.5	38.0	6.4	1.4	0.2	6.6
10	0.8	0.2	0.1	0.2	0.3.	0.5	7.6	46.9	31.7	5.5	0.3	0.0	5.8
11	1.4	0.4	0.2	0.3	0.4	0.2	2.0	28.0	38.2	19.5	2.9	0.7	5.7
Z1	0.4	0.1	0.1	0.1	0.1	0.5	2.5	27.6	62.1	3.1	0.1	0.0	3.3
Z2	4.3	0.9	0.5	0.2	0.3	2.9	19.3	40.5	17.9	4.5	0.2	0.0	8.5
Z3	0.3	0.0	0.0	0.2	0.8	1.3	15.7	61.3	14.5	0.5	0.0	0.0	5.3
Z4	18.2	4.8	3.4	6.5	1.4	4.8	14.4	16.7	3.2	1.6	0.6	0.4	21.3
Z5	0.5	0.1	0.1	0.3	1.0	1.1	16.5	62.0	11.6	0.9	0.1	0.2	5.7
Z6	19.06	5.2	3.8	5.2	1.0	5.0	15.8	15.7	3.7	0.8	0.2	0.1	23.1

Table 6 Concentrations of heavy metals in sediments ($\mu g.g^{-1}$ dry weight).

Treatment	Cd	Cu	Zn	Hg	Cr	As
4	0.23		41	0.10		
5	6.0		510	1.6		
8	0.60		120	0.34		
9	0.18		38	0.024		
10	0.041		27	0.0092		
11	0.061		20	0.0083		
12	7.3		720	2.1		
15	10.8	112	-	2.24	206	27.7
Z1	0.090		22	0.0055		
Z2	0.30		73	0.12		
Z3	0.11		33	0.0092		
Z4	1.9		330	1.2		
Z5	0.11		32	0.056		
Z6	2.8		420	1.1		

Table 7 Concentrations of PCBs, HCB and DDE in sediments ($\mu g.kg^{-1}$ dry weight).

Treatment	PCB component													
	15	28	52	49	44	70	101	87	153	138	180	HCB	opDDE	ppDDE
4	1.8	<1.3	<1.3	<1.3	2.5	1.3	1.3	<1.3	2.0	1.7	<1.3	4.6	<1.3	<1.3
5	39	7.3	12	8.1	8.1	20	21	7.3	30	30	20	<2.7	7.0	5.1
8	7.7	2.1	3.5	2.6	4.3	6.0	6.8	2.4	9.7	8.7	5.1	5.6	2.3	<1.6
9	1.6	<1.3	<1.3	<1.3	2.8	<1.3	<1.3	<1.3	2.0	1.7	<1.3	1.6	<1.3	<1.3
10	<1.3	<1.3	<1.3	<1.3	1.5	<1.3	<1.3	<1.3	<1.3	<1.3	<1.3	<1.3	<1.3	<1.3
11	1.8	<1.4	<1.4	<1.4	7.3	<1.4	<1.4	<1.4	<1.4	<1.4	<1.4	<1.4	<1.4	<1.4
12	270	51	37	33	32	41	38	9.9	38	31	18	14	22	8.9
15		56					32		43	36	19			

For the particle classes 0-75 µm and 150-300 µm the values of "a" were averaged in order to obtain a mean mixing ratio of 12.02 ± 4.16 (n=7). Exclusion of size classes larger than 32 µm increased the ratio to 14.49 and at the same time decreased the standard deviation to 3.14 (n=4). It is felt that the latter value is more accurate, and is preferred to be used to compare with contaminants mixing ratio's (3.1.3.). This comparison leads to the conclusion that about 50% of the contaminants occurs in the fraction < 100 µm comprising about 7% of the total mass.

3.1.3. Concentrations of contaminants in sediments

Table 6 provides the concentrations of heavy metals (Cd, Zn and Hg) in the sediments based on dry weights. Table 7 does the same for PCBs, HCB and DDE. Concentrations of PAHs and pesticides are reported elsewhere. Concentrations in Rotterdam Sediment as determined before (1983) are reported as treatment 15.

Clearly, sediments from Valkenisse and Konijnenschor are hardly contaminated. Surprisingly in sediments from Bath no significant contamination was detected apart from a minor increase in cadmium. This observation contrasts with observations only a few years before, indicating moderate to severe contamination with cadmium and PCBs at Bath. However, those sediment samples were collected in the intertidal zone along the banks and consisted of much finer grain size material, rich in organic matter. Sediments from Zandvliet are clearly contaminated with heavy metals and PCBs (table 6 and 7), PAHs (table 15) an some pesticides (table 17). However for heavy metals and the lower chlorinated PCBs the concentrations are about 10 times lower than in the Rotterdam sediment. For the higher chlorinated PCBs, the concentrations are about 4 times as low. Fractionation by sieving over a 100 µm net of the Zandvliet sediment lead to two fractions: a coarse one (69% of the particle larger than 106 µm) and a fine one (72% of the particles smaller than 106 µm). The analytical data show that a large proportion of the contaminants is associated with the smaller grain size fraction. For PCBs and mercury the concentrations in the fine fraction are 15 times as high as in the coarse fraction. For cadmium the concentrations are 25 times as high. For zinc this ratio is somewhat less (about 12) and remarably for HCB the reverse behaviour was noticed (higher in the coarse fraction).

Table 8 Coefficient of lineair correlation between heavy metals, particle size and organic matter (n = 11).

	16	32	Org.M.	Cd	Zn
% < 16 µm (16)	-				
% < 32 µm (32)	0.999	-			
Organic matter (Org. M)	0.94	0.94	-		
Cd	0.97	0.96	0.91	-	
Zn	0.99	0.99	0.97	0.94	-
Hg	0.99	0.99	0.96	0.93	0.99

Table 9 Estimates of coefficients of exponential regression analysis. Between brackets the 95% confidence interval.

	P1	P2
Cd		
32	0.0028 (0.0011 - 0.046)	1.91 (1.76 - 2.06)
Org.m.	0.0023 (0.0002 - 0.044)	3.24 (2.87 - 3.61)
Zn		
32	27.7 (14.6 - 40.8)	0.74 (0.61 - 0.86)
Org.m.	21.0 (2.5 - 40.0)	1.33 (0.95 - 1.71)
Hg		
32	0.090 (0.0009 - 0.18)	0.74 (0.50 - 0.98)
Org.m.	0.076 (-0.12 - 0.27)	1.27 (0.21 - 2.34)

A deviating behaviour of HCB in relation to other contaminants was also noticed in uptake by *M. edulis* along the Dutch coast in 1979/1980 (De Kock and Marquenie, 1981). Mussels exposed in the tidal zone exhibited much higher concentration of all contaminants except for HCB than those from 3 km offshore. The same difference applies to mussels exposed just below and at the bottom of the Nieuwe Waterweg where the Rhine flows into the North Sea.

In order to evaluate the geochemical behaviour of the contaminants in the Western Scheldt in relation to sediment speciation we analyzed the data statistically. Table 8 shows the coefficients of linear correlation between heavy metals, percentages of the fractions smaller than 16 and 32 μm, and organic matter (based on dry weight) in sediments from the Western Scheldt. Konijnenschor was excluded because organic matter did partly originate from the banks and not from the river Scheldt or the North Sea. Clearly all coefficients are highly significant. However, a further analysis shows that the regression are not lineair but generally fit exponential curves:

$$\text{Metal} = p1 * \exp(p2 * \text{Var}) + p3.$$

In which metal concentration were related to the variables: fraction smaller than 16 and 32 μm and organic matter respectively and p1, p2 and p3 are regression coefficients. The results for the fraction 32 μm and organic matter are tabulated in table 9.

From this table it is concluded that the metal concentration relates somewhat more precise to the fraction of 32 μm than to organic matter content, although it was also noticed that the standard error from the squared residuals is much smaller for organic matter.
The data do not sustain a conclusion concerning the way metals occur in the sediments because grain size and organic matter are closely interrelated. Some results are shown for zinc in figure 5 and for mercury is figure 6.

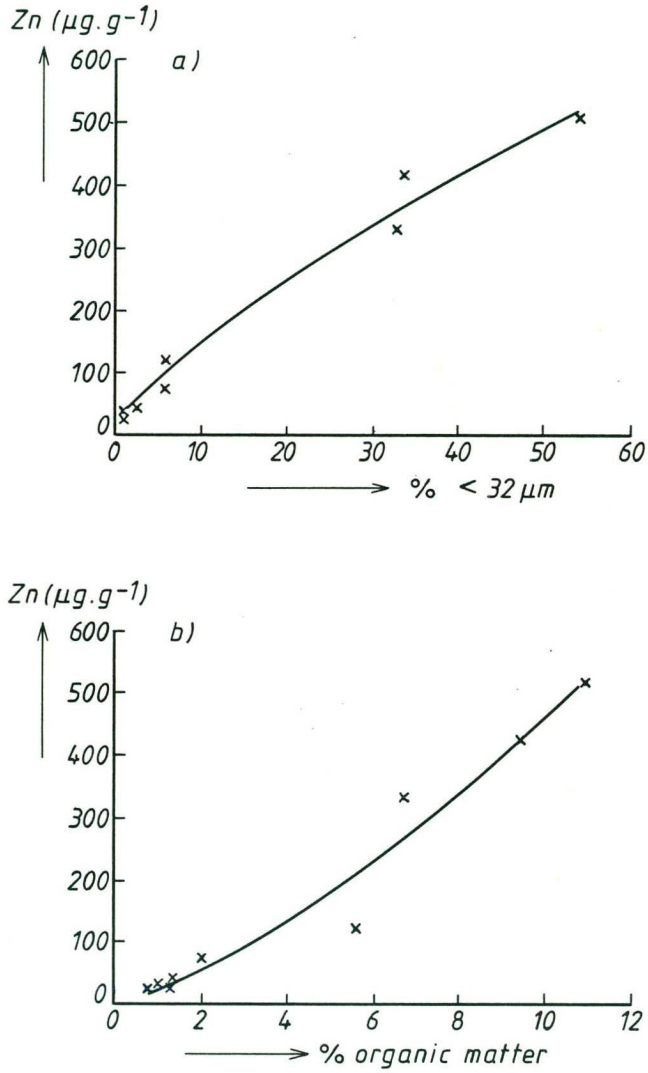

Figure 5 Zinc (µg.g^{-1} dry weight) related to % < 32 µm (a) and % organic matter (b). Sediments from Zandvliet, Bath and Valkenisse.

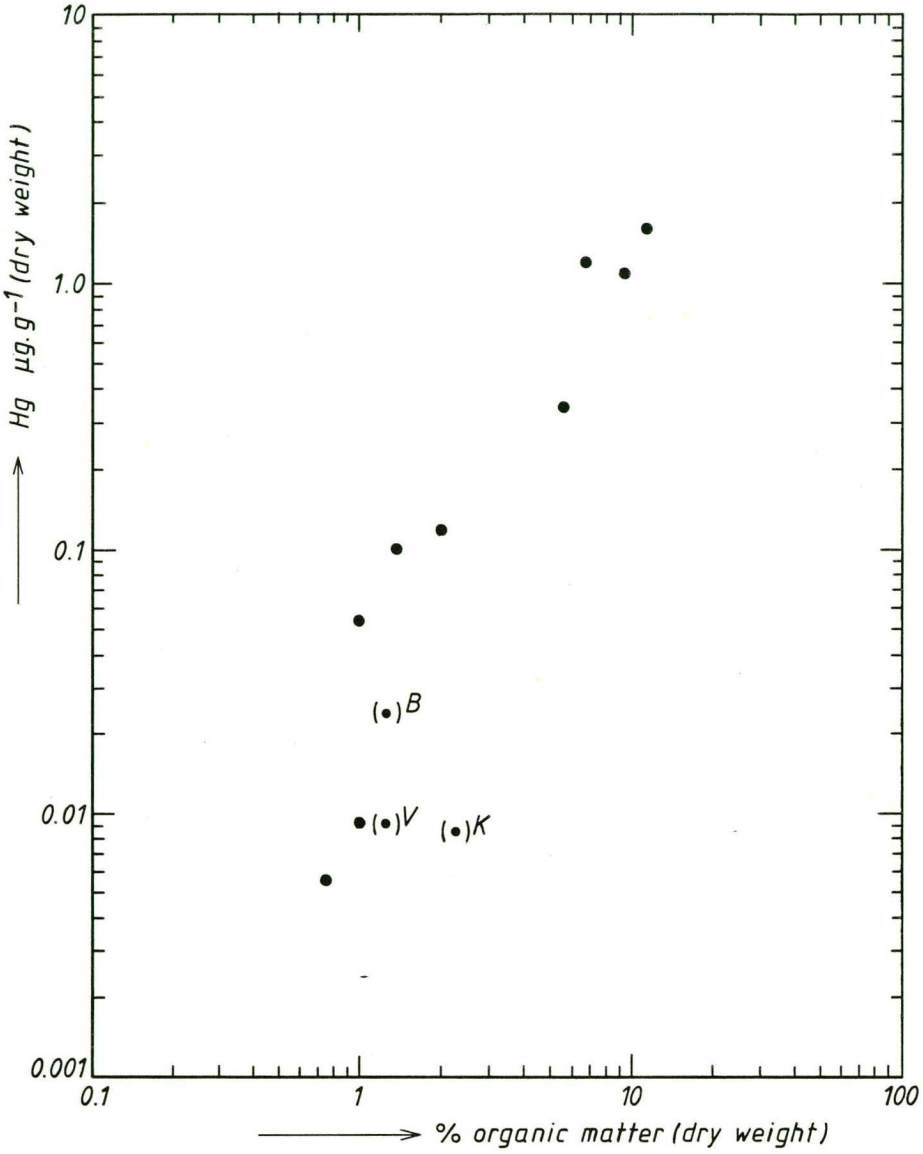

Figure 6 Mercury (µg.g^{-1} dry weight) related to % organic matter (dry weight). Sediments from Zandvliet (total, coarse and fine fraction, Z1-Z6), Bath (B), Valkenisse (V) and Konijnenschor (K).

3.2. WATER QUALITY

Water samples were collected in the middle of the experiment and stored frozen (-20°C) till needed. The samples were analyzed for nutrients in order to assess the risks for eutrophication. Leaching of other contaminants was already covered through analysis of suspended mussels. The results are shown in table 10. Data are reported in $\mu mol.l^{-1}$. It is remarked that ammonium data might not be accurate due to storing. Compared to the concentrations in the inflowing water (treatment 3) nutrients are trapped in the systems apparently due to biological activity. This is most obvious in systems containing sediments poor in nutrients. Nitrate trapping appeared in all treatments. In treatment 11 all nutrients investigated were involved. At the same time nutrients may leach from the sediments. This accounts strongly for sediments from Zandvliet and Rotterdam.

Table 10 Concentrations of nutrients in water samples, total (t) and 0.45 µm filtrate water (f) in $\mu Mol.l^{-1}$.

Treatment	NO_2^-		NO_3^-		NH_4^+		$PO_4^=$		Si	
	t	f	t	f	t	f	t	f	t	f
1										
2										
3	2.6	2.6	13.5	18.0	37.0	38.7	7.8	7.9	3.7	5.3
4	0.6	0.5	2.3	2.4	44.0	24.6	6.5	5.4	2.7	2.6
5										
6	0.6	0.6	0.8	2.3	33.5	26.4	>13.3	5.4	15.4	11.7
7	2.5	2.1	12.2	9.6	28.2	22.9	7.1	6.9	4.3	3.2
8	0.4	0.3	2.5	2.6	42.3	21.1	>13.3	>13.3	26.6	18.6
9	0.7	0.5	2.2	2.4	28.2	21.1	9.2	7.1	1.6	2.6
10	0.8	0.4	2.1	1.0	37.0	19.4	2.9	3.6	4.1	3.2
11	0.5	0.6	0.9	2.3	19.4	22.9	3.6	4.1	3.2	4.2
12	2.9	2.6	12.1	10.6	26.4	21.1	>13.3	>13.3	13.3	13.2
13										
14										

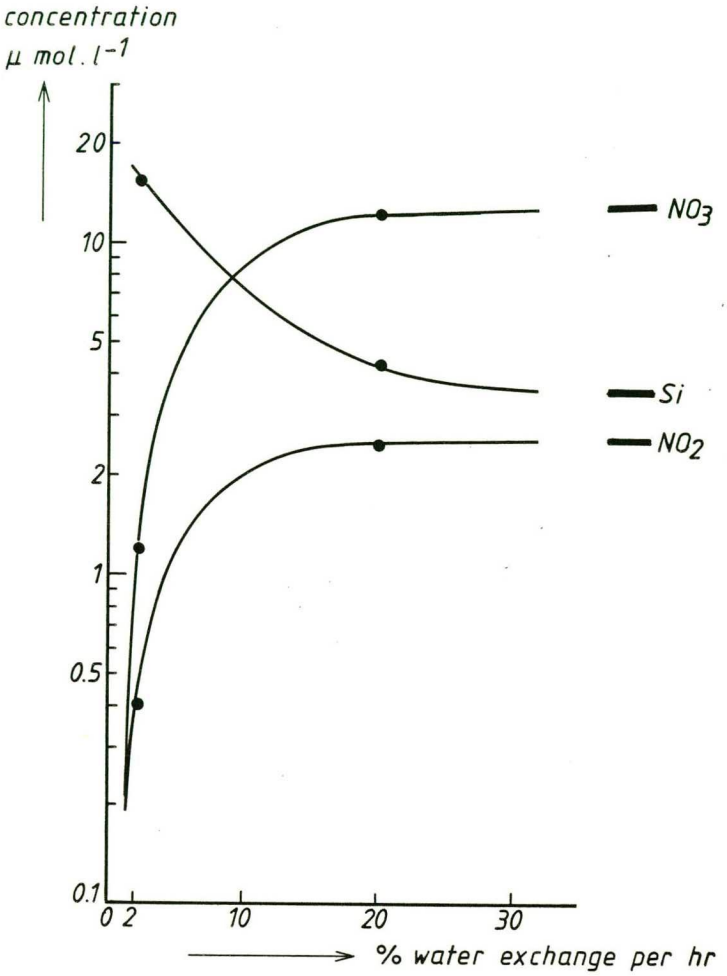

Figure 7 Expected concentration of nutrients in the Zandvliet system in relation to water exchange rate. Indicated are the concentrations in the inflowing water in the right hand margin.

In order to visualise the leaching of silicum and the trapping of nitrite and nitrate, the concentrations in treatments 7, 8 and 3 were used to plot against water exchange rate (figure 7).

From this graph it can be deducted that maximum leaching of silicum from the sediment occurs at exchange rates in the order of 20% per hr and over. Apparently nitrates and nitrites do not leach significantly from this sediment.

These deductions correlate well with the observations in relation to bioturbation. Bioturbation enhances leaching of silicium but does not influence nitrate and nitrite.

It can be concluded that subaquous disposal of sediments of Zandvliet and Rotterdam might lead to eutrophication problems. In case of Zandvliet low availability of nitrogen might limit severe eutrofication problems. For the sediments from Konijnenschor and Valkenisse silicium might be a limiting factor in bioproduction.

3.3. ORGANISMS

General information upon the tissue homogenates is provided in table 11. The table shows for each species the number of individuals that were pooled into one sample, the water content of the homogenate and the percentage of ash-free dry material, the calculated mean wet weight and ash-free dry weight per individual. Percentages of ash weight are not reported though they were low and constant for each species, except for the blanks of *A. marina*, which were not depurated.
For *M. edulis* it can be seen that growth was best in the raw water. These animals approximately doubled their weight in 60 days. On the contrary, directly exposure to the filtrate water resulted in a weight loss of about 50%. A weight loss was further observed above sediments from Konijnenschor, Bath and Zandvliet (20% water exchange). Both first systems were already characterized by low silicum concentrations and therefore a limited algal production. Weight reduction in the one Zandvliet is explained by the high exchange rate with filtered water because at lower exchange rates reasonable growth occurred. Almost the same applies for *M. balthica*. Direct exposure to filtrate water leads to a weight loss of about 50% despite of the low density of animals and the natural unpolluted sediment.

Table 11 Sample composition. The number of individual organisms (n) pooled into one sample, percentages of water (%w) and ash-free dry tissue (%a), the calculated mean wet (w) the ash-free dry weight (a) (in gr), per organisms and the relative increase of a in % (Δa).

Treatment	*M. edulis*						*M. balthica*						*A. marina*					
	n	%W	%a	w	a	Δa	n	%W	%a	w	a	Δa	n	%W	%a	w	a	Δa
1	59	79.0	19.1	2.13	0.407	118												
2	66	79.1	18.8	2.01	0.378	102												
3	65	83.8	14.2	0.70	0.099	-47	12	93.3	5.1	0.274	0.014	-49	7	87.3	10.7	3.83	0.41	12
4							82	86.9	10.0	0.263	0.026	-5.5						
							106	83.7	12.7	0.245	0.031	13						
6	60	81.2	16.7	1.33	0.222	19	97	83.7	12.3	0.238	0.029	5.5						
7	69	84.2	13.8	1.29	0.178	-4.8	79	87.7	9.7	0.295	0.029	5.5	25	82.3	12.7	4.07	0.52	42
8	70	83.1	14.9	1.53	0.228	22	70	89.1	8.6	0.259	0.022	-20	27	84.8	12.2	4.44	0.54	48
9	60	85.6	12.2	1.20	0.146	-22	96	87.1	9.9	0.240	0.024	-13	27	84.5	11.5	3.29	0.38	4.1
10	60	83.8	14.1	1.52	0.214	14	118	83.1	12.8	0.284	0.036	31	30	84.7	11.2	2.48	0.29	-21
11	60	84.0	14.0	1.14	0.160	-14	82	81.9	12.7	0.223	0.028	1.8	17	84.2	11.5	2.05	0.24	-34
12	60	84.7	13.1	1.52	0.199	6.4	92	83.3	14.3	0.473	0.068	147	32	86.0	11.5	3.94	0.45	23
13	60	82.9	14.9	1.29	0.192	-	167	87.6	8.8	0.306	0.027	-	75	76.4	10.2	3.32	0.34	-
14	60	83.7	14.0	1.30	0.182	-	164	87.1	9.3	0.300	0.028	-	74	78.2	11.2	3.52	0.39	-

On the contrary, animals exposed to the highly contaminated sediment from Rotterdam almost increased their weight with a factor 2.5. Except for Valkenisse, with a weight increase of about a factor 1.3 and Zandvliet (treatment 8) with a decrease of 0.85, the weights more or less stayed constant. The data for *M. balthica* support observations that this organisms is not restricted to deposit feeding alone, but also depends upon foodsources in the superficial water.

A. marina, is a real deposit feeder. This can be concluded from the observed growth in the filtered water compartment with Wadden Sea sediment, where the other two species lost about 50% of their weight. A significant weight increase was further observed in the contaminated Zandvliet and Rotterdam sediments. A weight decrease was observed in sediments from Valkenisse and Konijnenschor, both poor in organic matter. In general it can be concluded that the best growth occurs in and above the most contaminated sediments, due to equally available foodsources or developing diatoms and algae in connection with leaching of nutrients.

Disposal of sediments form Zandvliet and Bath at Konijnenschor might therefore lead to an increased biological produktion and change the existing ecosystem. Whether such an increase in productivity is wanted lays beyond the scope of this report.

However, these effects will be related to contaminant mobility in the next chapter.

3.3.1. Bioavailability of heavy metals

Table 12 shows the concentrations of cadmium, zinc and mercury, and additionally to this program copper in the organisms prior and after subjection to the treatments. It should be remarked at this point that the blanks (treatments 13 and 14) originate from area's influenced by the highly polluted river Rhine. *M. edulis*, as mentioned before, was collected close to Egmond. *M. balthica* (Central Dutch Wadden Sea) and *A. marina* (Mokbaai at Texel), were collected from locations highly valued as some of the ultimate natural and ecological conservation resources available to the Netherlands. At the same time, however, these locations are in discussion because of the failure of seals to reporduce, probably due to certain chemicals (e.g. PCBs,

Table 12 Concentrations of cadmium (Cd), copper Cu), zinc (Zn) and mercury (Hg) in organisms in $\mu g.g^{-1}$ (ash-free dry weight).

Treatment	*M. edulis*				*M. balthica*				*A. marina*			
	Cd	Cu	Zn	Hg	Cd	Cu	Zn	Hg	Cd	Cu	Zn	Hg
1	1.3	12	140	0.28								
2	1.1	12	130	0.28								
3	2.1	13	200	0.52					1.6	14	89	0.51
4					1.2	48	750	0.48				
5					1.3	49	690	0.44				
6	8.3	11	140	0.73	2.5	64	690	0.69				
7	2.0	15	140	0.67	1.4	51	960	0.61	1.6	15	95	0.31
8	6.4	9.9	130	0.60	2.5	73	1100	0.61	1.6	11	75	0.56
9	3.7	9.2	180	0.43	1.3	56	1100	0.47	1.3	8.2	83	0.32
10	1.7	13	120	0.41	0.63	35	710	0.31	1.4	14	77	0.29
11	1.8	14	120	0.39	0.61	43	710	0.43	1.8	9.7	95	0.30
12	1.8	14	120	1.06								
13	1.4	11	510	0.57	0.51	54	1100	0.54	1.8	16	120	0.34
14	1.6	9.4	440	0.29	0.22	49	850	0.52	2.2	15	140	0.39

Reinders, 1979) accumulating in the foodchain. Under no circumstances the concentrations in the blanks should be regarded as environmentally safety levels.
A comparison of the treatment data with the internal control (treatment 3), however, is also difficult to make for *M. edulis* and *M. balthica*. Both species lost about 50% of weight. Recent experiments (not yet published) indicated that mussels (*Dreissena polymorpha*) might fail to accumulate and eliminate some heavy metals in absence of food. This can be seen most clearly for cadmium, zinc and mercury by comparing treatment 1 and 2 with 3. In both treatments the water was the same but food and other particles were absent in treatment 3, resulting in a decrease of weight and an increase in metal concentrations.

Main experiment

Obviously, bioavailability to all three species tested in sediment from Konijnenschor (the disposal site, treatment 11) relates well within sediment from Valkenisse (treatment 10).

Sediments from Zandvliet showed a higher availability, with Bath as an intermediate. For *M. edulis* cadmium and mercury concentrations are clearly increased indicating a leaching of highly bioavailable metal forms. Sediment from Rotterdam was about 10 times more contaminated with cadmium, however, no leaching of bioavailable forms was indicated by a sensitive organism as *M. edulis*. With respect to copper and zinc no indications were found for leaching of bioavailable forms.

For cadmium and mercury in *M. balthica* the same applies as for *M. edulis*. Unfortunately data for Rotterdam sediments are lacking. As a substitute data from a previous experiment are presented (table 13). However, the organisms in the present experiment were not analyses by TNO and cannot be compared very well. Reanalysis by TNO of some samples even revealed a substantial difference, probable due to different techniques applied (AAS versus NAA).

Table 13 Concentrations of heavy metals in *M. balthica* ($\mu g.g^{-1}$ ash-free dry weight).

Treatment	Metal					
	Cd	Cu	Zn	Hg	As	Cr
Compare to treatment 13 and 14	0.020	31.1	-	0.19	15.4	0.54
Compare to treatment 12	0.48	38.0	-	0.40	16.8	2.74

Copper in *M. balthica* does not compare to copper in *M. edulis*. However, these organisms compare well with regards to cadmium and mercury accumulation.

As expected, the lugworm, *A. marina*, does not reflect concentration differences in sediments to a large extent, confirming that polychaeates are generally known as poor indicators of metal pollution (De Kock and Marquenie, 1982; Bryan, 1976; Simmers and Marquenie, 1984).

Side experiments

Bioturbation

Bioturbation (treatment 8 versus 6) did not enhance leaching of bioavailable metal forms.

Water exchange rate

An increase in water exchange rate from 2% to 20% reduced uptake of cadmium in both *M. edulis* and *M. balthica*. No descriptive model can be proposed at the present time due to the number of parameters involved simultaneously (uptake, growth, water exchange and leaching). An estimate might be obtained by analyzing stored water samples.

Fractionation

Despite a high difference in concentrations in two fractions coarse and fine substrate, concentrations in the exposed *M. balthica* were the same. A feasable explanation can be based on observations of Mirmand et al. (1982) that *M. balthica* accumulates metals from the interstitial water. Other observations, Marquenie (1985), indicate that only ionic cadmium is bioavailable. It can be easily seen from this point that both coarse and fine fractions, once united into a continuous, steady state situation of desorbing and adsorbing will both end up at the same ionic concentration in the interstitial water.

3.3.2. Bioavailability of PCBs, HCB and DDE

Main experiment

Concentrations of PCBs, HCB and DDE are shown in table 14. The same remarks made under 3.3.1. concerning the blanks, apply to these organic contaminants.

In general much lesser differentiation is noted for *M. edulis* than for the other species, indicating low leaching rates. However, some uptake can be noticed. Sediment from Rotterdam most clearly releases lower chlorinated PCBs and DDE. Concentrations of HCB, on the contrary, remain low. Concentrations in mussels above Konijnenschor (disposal site, treatment 11) and Bath (treatment 9) sediments are roughly comparable, except for PCB-180 (higher at Bath) and HCB (lower at Bath). Somewhat lower levels were found in treatment 10 (Valkenisse, downstream of Bath and Konijnenschor). This does not account for PCB 15 (peak can be disturbed by Ionol) and PCB 49 and 70 (peaks can be distrubed by chlorostyrenes). Except for the above mentioned PCB components, concentrations in mussels from treatment 10 are comparable to or lower than from the intrinsic blank (treatment 3). Concentrations in the Zandvliet systems are not increased compared to downstream locations. These observations are in contrast to the other two species tested. For both *M. balthica* and *A. marina* it is obvious that bioavailability of PCBs, HCB and DDE in sediments from Konijnenschor is much lesser than in that from the Wadden Sea (treatment 13 and 3). Concentrations in organsisms from and ex-

posed to Wadden Sea sediments, in this report defined as environmentally critical levels, are highly comparable to those exposed to sediment from Valkenisse. Further upstream, at Bath and Zandvliet bioavailability increases with distance. For *A. marina*, which proved to be a better accumulator of organic contaminants than *M. balthica*, the concentrations in the tissues at Zandvliet are 4-5 times as high as at Valkenisse and 5-10 times as high as at Konijnenschor. Some concern should be mentioned regarding the occurrence of PCBs in the Wadden Sea. If for instance PCBs will prove to be cause of reproduction failure in seals living in the Dutch Wadden Sea the concentrations in *M. balthica* in treatment 13 should be regarded as critical, harmfull levels. Remarks concerning an influence of food on accumulation and elimination of heavy metals, however, do not apply to PCBs.

Side experiments

Bioturbation

It should be noticed that the concentrations in *M. edulis* and *M. balthica* in absence of *A. marina* (no bioturbation, treatment 6) are somewhat higher than with *A. marina* (treatment 8). This might be due to improved oxidation of the top layer, not continuously covered by new deposits, and a subsequent release of former absorbed contaminants. However, the differences were not regarded as serious thus no further efforts were undertaken at this point to reveal the cause.

Water exchange rate

Remarkably, no significant, clear differences were observed in relation to water exchange rate (treatments 7 and 8). The only explanation we have for this observation is that uptake of these contaminants is a very efficient proces and not comparable to the uptake of heavy metals.

Fractionation

Fractionation resulted in an about 15 times more contaminated fine fraction (treatment 5) compared to the coarse fraction (treatment 4). Exposure of *M. balthica* to both fractions did not result in such a wide difference in concentrations in the tissues. For the higher chlorinated PCBs there was, as for heavy metals no difference at all. For the lower chlorinated PCBs and

DDE the difference was less than a factor of 2. Therefore, the concentrations in the tissues do not, or to a very small extent, relate to concentrations in the sediments.
Attention should be given to HCB. Concentrations in *M.balthica* exposed to the coarse fraction, which proved to more contaminated than the fine one, also accumulated more HCB in their tissues. Furthermore, *M. balthica*, but also *A. marina* exposed to sediments from Rotterdam, with much higher concentrations accumulated the lowest concentrations of all treatments in their tissues. Based on these observations, and some other observations, with *M. edulis*, as mentioned before, it is concluded that HCB is primarely associated with sandy materials but that its bioavailability and leaching to superficial waters is reduced by organic matter. This conclusion is supported by the facts that HCB in *M. edulis* above Rotterdam sediment is also very low and that bioavailability of HCB to all three species in relation to Zandvliet sediments is generally lower than to sediments from downstream locations.

3.3.3. Bioavailability of PAHs and pesticides

Because no recent biological information was available to indicate whether PAHs and pesticides would pose an environmental problem connected to dredging in the Western Scheldt, sediments and organisms after exposure in treatment 8 (Zandvliet) and prior exposure (treatment 13) were analysed for PAHs and "drins" (aldrin, dieldrin and endrin).
Results for PAHs are shown in table 15. In general, the concentrations in the sediment proved to be high. Unfortunately, no significant conclusions can be drawn with regard to leaching because data from the other systems are lacking. However, based on observations with PCBs and experiences in the USA (Marquenie et al., 1984) strong indications are found that phenanthrene, pyrene, benzo(a)anthracene, benzo(b)fluoranthene and benzo(a)pyrene leach from the sediment and become bioavailable for *M. edulis*.
Results for *M. balthica* and *A. marina* prove to be highly comparable although concentrations in *A. marina* are about twice as high. According to the present results with *A. marina* the observations can be grouped into 8 catagories, based on concentration factors (concentration sediment/concentration tissue) (table 16).

Table 14 Concentrations of PCBs, HCB and DDE in organisms ($\mu g.kg^{-1}$ ash-free dry weight). Included are data from a previous experiment (*M. balthica*, treatment 15).

Treatment	PCB component 15	28	52	49	44	70	101	87	153	138	180	HCB	opDDE	ppDDE
M. edulis														
1	36	20	32	25	24	35	73	18	110	83	14	24	32	18
3	14	12	23	13	17	47	68	16	125	92	9.2	15	32	18
6	27	16	25	27	19	49	66	19	116	98	19	17	24	14
7	29	11	20	14	17	45	61	17	110	84	17	21	22	13
8	27	12	23	17	20	44	58	15	98	76	13	18	20	11
9	27	16	35	21	21	< 8.2	70	18	110	87	14	21	27	15
10	70	8.5	14	23	13	32	41	9.9	74	56	7.8	39	16	9.2
11	26	14	63	19	23	< 7.1	62	14	100	77	7.9	48	28	14
12	41	44	66	60	40	93	89	18	110	110	16	19	43	18
13	41	30	50	36	32	41	66	17	93	67	11	37	33	26
14	39	26	44	33	32	36	61	15	89	65	10	38	29	15
M. balthica														
3	41	22	18	< 8.0	30	< 8.0	58	14	78	71	19	44	42	13
4	18	17	30	19	28	30	82	26	110	95	35	63	26	17
5	65	27	58	36	31	38	98	31	110	91	32	31	31	20
6	150	61	89	79	210	160	220	61	270	240	76	220	63	33
7	150	57	120	72	49	170	260	75	280	230	74	68	89	53
8	99	44	92	67	135	63	160	50	180	150	50	45	52	35
9	89	43	96	62	51	140	195	60	230	190	68	49	65	35
10	58	21	30	21	19	52	85	30	98	80	19	41	27	14
11	22	8.7	< 7.9	< 7.9	20	22	34	7.9	50	41	13	48	11.8	< 7.9
12	150	190	270	230	150	200	230	53	220	140	48	28	120	43
13	60	22	18	< 11	34	32	57	16	89	76	20	50	26	16
15	-	112	197	-	-	-	189	-	188	121	58	-	-	-
A. marina														
3	55	21	33	26	37	33	79	19	140	99	34	32	36	20
7	440	100	220	130	120	140	370	110	470	150	160	45	120	100
8	250	88	190	120	98	120	310	99	420	330	130	75	100	82
9	140	35	92	57	52	130	170	50	230	180	130	34	57	43
10	140	20	46	30	26	28	95	27	150	110	33	52	35	24
11	31	8.7	12	< 8.7	18	17	24	< 8.7	49	36	12	37	8.7	<8.7
12	200	250	360	300	290	380	380	80	410	240	86	35	190	86
13	96	17	23	160	57	45	66	17	130	97	29	50	29	15

Table 15 Concentrations in $\mu g.kg^{-1}$ of PAH components in a sediment (dry weight) and in organisms ash-free dry weight prior to stocking (p) and after 60 days of exposure (e), (treatment 8).

	Sediment	*M. edulis*		*M. balthica*		*A. marina*	
		p	e	p	e	p	e
1 phenanthrene	800	11	22	125	520	250	1200
2 anthrancene	63	13	15	11	130	18	390
3 fluoranthene	400	20	17	80	800	250	1600
4 pyrene	370	17	230	88	1500	200	3100
5 3.6-dimethylphenanthrene	<16	15	<23	<13	78	46	210
6 triphenylene	<56	18	<74	38	440	98	1200
7 benzo(b)fluorene	53	<8.1	<30	<16	200	<11	<14
8 benzo(a)anthracene	200	48	81	50	840	62	2000
9 chrysene	130	41	<43	45	410	84	980
10 benzo(e)fluoranthene	310	52	<130	<68	67	15	2400
11 benzo(j)fluoranthene	<450	<150	<58	<30	<55	<210	<260
12 perylene	100	10	8.7	26	61	65	<3.0
13 benzo(b)fluoranthene	230	30	43	73	360	180	- [1]
14 benzo(k)fluoranthene	110	17	24	34	140	61	370
15 benzo(a)pyrene	180	8.1	16	20	230	43	570
16 dibenzo(a,j)anthracene	71	<15	<58	<30	<57	<23	<25
17 dibenzo(a,i)pyrene	<35	<14	<55	<28	87	<20	<81
18 benzo(g,h,i)perylene	<66	<22	<8.7	<43	<84	<30	780
19 indeno(1,2,3-c,d)pyrene	24	<14	<55	55	<55	<20	<25
20 3-methylcholanthrene	<9.3	<8.1	<30	<16	<31	<11	<13
21 anthanthrene	60	3.4	<13	<6.8	13.8	<3.9	33

[1]) Peak disturbed.

Table 16 Categories of PAHs based on concentration factors.

Concentration factor	Component
< ½	perylene, benzo(b)fluorene, dibenzo(a,j)-anthrancene, indeno (1,2,3-c,d)pyrene
½ - 2	phenanthrene, anthanthrene
3 - 4	fluoranthene, benzo(b)f., benzo(k)f., benzo(a)-pyrene
6	anthrancene
8 - 10	pyrene, 3,6-dimethylphenanthrene, benzo(a)-anthranthene, chrysene, benzo(e)pyrene
20	triphenylene
unknown	benzo(j)fluoranthene, dibenzo(a,i)pyrene, 3-methyl-cholanthrene
contradictionary to M.b.	benzo(g,h,i)perylene

It should be remarked that these concentration factors only apply to this Zandvliet sediment because bioavailability might be quite different for other sediments (Simmers and Marquenie, 1984). In general, the concentrations in *A. marina*, should be considered high, from an ecotoxicological point of view. Because concentration factors for earthworms (although also variable, depending on soil composition in a confined upland disposal site for dredging materials) proved to be much lower (Marquenie and Simmers, 1984) from an ecotoxicological point of view upland disposal might be a better alternative for these materials.

Table 17 Concentrations of pesticides in $\mu g.kg^{-1}$ in a sediment (Zandvliet) and in organisms prior to stocking (p) (treatment 13) and after 60 days of exposure (e) (treatment 8). For concentrations of DDE and HCB in other treatments, see also table 7 and 14.

	Sediment [1]	Organisms [2]					
		M. edulis		*M. balthica*		*A. marina*	
		p	e	p	e	p	e
lindane	0.18	9.6	5.2	9.7	8.5	6.5	4.8
aldrin	0.24	<6.7	<6.7	<11	<12	<9.8	<8.2
dieldrin	<1.6	22.3	19	16	76	11	73
endrin	<1.6	<6.7	<6.7	<11	<12	<9.8	<8.2
op DDE	2.3	33	20	26	52	29	100
pp DDE	<1.6	26	11	16	35	15	82
HCB	5.6	37	18	50	45	50	75

[1]) Based on dry weight.

[2]) Based on ash-free dry weight.

Concentrations of pesticides are shown in table 17. This table also includes concentrations of HCB and DDE as reported before (table 7 and 14). These same drawbacks as for PAHs account for conclusions from these data. However, none of the pesticides seem to leach from the sediment. From the "drins", only dieldrin was assessable in the tissues of the organisms and exposure to the sediment leads to an increase. Form the other pesticides only DDE, as mentioned before, might pose an environmental problem.

3.4. REPRODUCABILITY OF THE BIOASSAY AND AN EXTRAPOLATION TO OTHER ORGANISMS

As indicated in subchapter 3.3.1., the Rotterdam sediment was also used in a previous experiment. Linking the results from both experiments will yield information on reproducability of the bioassay, which is generally a point of discussion and offer the unique possibility to extrapolate the results to other species of organisms. Based on the extrapolation an ultimate choice can be made for the most useful bioassay organisms. It is noted however, that the water exchange rates in the previous experiment were somewhat higher (about 10% per hr).

Table 18 summarizes some data already presented before and adds some new data.

From this table the following conclusions can be drawn:

- Sediment data, although derived from two different laboratories (TNO, this experiment and IB/WL, an other large contracting laboratory, the previous experiment) compare very well.
- Heavy metals in tissues are not comparable, although *M. balthica* were collected at the same location for both experiments. Tissues in the previous experiment were analysed by TNO, for this experiment by the RUU. Reanalysis by TNO of a few samples did not confirm any of the data sets. Therefore care should be taken in using the reported concentrations in an absolute sense.
- PCBs in tissues do compare very well, both in absolute numbers as related to each other. Somewhat higher numbers after exposure to the Rotterdam sediment (this experiment) might well be caused by the lower water exchange rate, bioturbation, or by a further oxidation of the sediment surface, rendering PCBs more bioavailable.
- *Mytilus edulis* proved to be a usefull bioassay organism in several respects. Firstly, it is used worldwide in biomonitroing, leading to an extensive data bank for comparison. Secondly it responded reliable to the waterquality in the sense of eutrophication and productivity. Thirdly, it indicated leaching of bioavailable contaminants, which can not be predicted based on concentrations in the sediment. It is proposed to use this organism in cases where a wide dispersal of materials to be disposed off is anticipated.
- For *Macoma balthica* almost the same is true, although it relates to both sediment and superficial water. Care should be taken therefore in choosing water exchange rates. Its value in evaluating bioavailability of organic contaminants, however, is lesser than *Arenicola marina* due to a lower differentiation and lower concentration factors.
- *Arenicola marina* proved to be extremely useful in assessing bioavailability of organic contaminants, however not for metals. Because uptake was also irrespective of water exchange rates much simpler test condi-

Table 18 A comparison of experimental data for Wadden Sea (M3 and M4) and Rotterdam (other data), S1, M1, M3, *C. edule* and *N. diversicolor* are based on a previous experiment (Anonymous, 1984).

Parameter	Sediment	Rotterdam	*Macoma balthica*		*Macoma balthica*		*M. edulis*	*A. marina*	*C. edule*	*N.diversicolor*
	S1	S2	M1	M2	M3	M4				
Cd	10.8	7.3	0.48	-	0.020	0.365	1.8	-	1.49	0.262
Cu	112	-	38.0	-	31.1	51.5	14	-	14.3	14.0
Zn	-	720	-	-	-	975	120	-	-	-
Hg	2.24	2.1	0.396	-	0.193	0.53	1.06	-	1.04	0.219
Cr	206	-	2.74	-	0.536	-	-	-	11.8	2.27
As	27.7	-	16.8	-	15.4	-	-	-	14.4	9.56
PCB15	-	270	-	150	-	60	41	200	-	-
PCB28	56	51	112	190	<21	22	44	250	48	96
PCB52	-	37	197	270	<30	18	66	360	62	34
PCB49	-	33	-	230	-	<11	60	300	-	-
PCB44	-	32	-	150	-	34	40	290	-	-
PCB70	-	41	-	200	-	32	93	380	-	-
PCB101	32	38	189	230	60	57	89	380	100	<34
PCB87	-	9.9	-	53	-	16	18	80	-	-
PCB153	43	38	188	220	94	89	110	410	131	164
PCB138	36	31	121	140	65	76	110	240	91	106
PCB180	19	18	58	48	28	20	16	86	42	52
HCB	-	14	-	28	-	50	19	35	-	-
opDDE	-	22	-	120	-	26	43	190	-	-
ppDDE	-	8.9	-	43	-	16	18	86	-	-

tions can be used for this organism. Its worldwide occurrence, its dominance in estuarine habitats and its importance as a foodsource for many other organisms, including fishes and birds, and the results obtained in this experiment favor this organism as a new bioassay test species.

- *Cerastoderma edule* and *Nereis diversicolor* are so called in-betweens. Both species inhabit sediments like the former two species. However, both species also depend much more on the superficial water quality. This means that they indicate everything a bit, but nothing real well. It is proposed to use these species only in special occasions: for regions where these species occur and the former three species are not only lacking completely but also their future occurrence on the new substrate is not anticipated.

3.5. ECOTOXICOLOGICAL CONSIDERATIONS

It was noted that the growth and conditions of organisms exposed to the most polluted sediments (Zandvliet and Rotterdam) were the best among all sediments tested. At the same time these organisms also accumulated the greatest amounts of contaminants in there tissues. It must be concluded at this point that the availability of food to the bioassay organisms suppresses toxic effect or that toxic effects do not occur at the concentrations we were dealing with. Further experiments are strongly needed to reveal this bias. Apart from possible toxic effects on the test species, other effects can be anticipated in nature. Most of the worldwide ecological disasters, recently summarized by Marquenie (1984) concerned mass mortality of birds or reproduction failure in birds and seals. In all cases reported, the contaminant reached its target through the foodchain without clear effects on the food organism itself. Translocating a contaminated but also eutrophic sediment to places with lesser bioavailable contamination might increase productivity in such area's of highly contaminated foodsources. This leads inevitably to the following conclusions with regard to effects in the foodchain:

- If no difference is found in productivity and bioavailability between dredged material and the material at the disposal site, disposal of this material will not change the local situation.

- If a substantially lower productivity and or mortality of test organisms, is found in the material to be dredged, even in case of a high bioavailability of contaminants disposal of this material might also not change the local environmental risk.
- Extreme care should be taken in cases of increased productivity in dredged material with the same or a higher bioavailability of contaminants, because adverse effect might show up the foodchain. Many dredged material fall into this category.

4. CONCLUSIONS

Based on the results of the bioassays and the considerations in the last subchapter the following general conclusions are drawn:

1. Sediments from Valkenisse and the Wadden Sea are comparable with respect to productivity and bioavailability of contaminants.
2. Sediments from Konijnenschor, the anticipated disposal site, showed the lowest bioavailability of contaminants of all sediments tested. Productivity, however, was also low. With respect to bioavailability of heavy metals, this location is approached closely by Valkenisse and Bath (except for cadmium). Productivity at Valkenisse was also low. No strong objections were found against disposal of dredged materials from Valkenisse at Konijnenschor. For Bath, because of the somewhat increased bioavailability and productivity, further experiments and analysis should be taken into consideration.
3. Sediments from Zandvliet are rather contaminated with respect to bioavailability of sediments and leachages, and eutrophication. Upland or confined disposal might be a better alternative especially with respect to the organic contaminants. Other types of bioassays are needed in relation to evaluate intertidal or upland disposal.
4. Sediments from Rotterdam displayed the highest bioavailability of contaminants.
5. Actual water exchange rates in the Western Scheldt are important for a further evaluation of disposal sites with respect to some contaminants.
6. Bioturbation should also be taken into consideration, especially in relation to eutrophication.
7. Fractionation, although diminishing the total amounts of contaminants for bulk disposal has no direct effect on bioavailability and will therefore not solve the environmental problem completely. Further experiments are anticipated.

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