Spec. Publs int. Ass. Sediment. (1981) **5**, 429–450

Pathways of mud and particulate trace metals from rivers to the southern North Sea

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INTRODUCTION

One of the sources of trace metals in the North Sea are the rivers. The major rivers from Belgium, the Netherlands and Germany which influence the southern North Sea are shown in Fig. 1. Estuaries are 'normal' pathways by which the particulate and dissolved trace metals enter the marine environment. However, civil engineering activities have created two additional pathways which affect the transport of metals into the North Sea.

(a) The closing of river mouths and coastal lagoons has created large freshwater basins in which the river water, before it enters the marine environment, is subject to a variety of hydrodynamic and geochemical processes which affect the amount of trace metals entering the marine environment.

(b) The construction of deep harbours has also influenced the sedimentation of the suspended matter and its associated heavy metals. Part of the dredged material from the harbours is dumped on land and is thus permanently removed from the aquatic environment, while another part is transported to dumping sites in the North Sea.

To determine the input of trace metals into the marine environment, the geochemical and hydrodynamical processes associated with each of these three pathways should be known. Intensive research carried out over the past five years in the estuaries, the harbour areas and the freshwater basins has made it possible to construct a first tentative balance of the amount of trace metals entering the North Sea by way of these three pathways.

0141–3600/81/1205–0429 $02.00

THE RIVERS

General

The particulate metals in the rivers are carried by the suspended mud in the water, and so the transport of the particulate trace metals of a river is determined by its mud transport and the metal concentrations in the suspended mud.

In general the (suspended) mud content of the water (mud is defined as matter finer than 50 μm) shows some relation with the discharge of the river: low discharge corresponds to low mud concentrations, and high discharge to high suspended-matter concentrations. However, many individual factors are involved in the rate of erosion of the catchment area, the mud content of the water and hence the sediment transport by the river (Hinrich, 1974). This results in a considerable scatter in the data showing the relationship between the suspended mud concentrations and the river discharge. An example of this is given in Fig. 2 for the river Meuse, which shows the effects of the seasons (distinct difference between the winter months and the rest of the year), the influence of a high water wave (extremely high suspended mud concentrations during the front of a wave increasing discharge) and presumably the effects of agricultural activities (e.g. accelerated erosion due to harvesting and ploughing) during September, October and November. The relatively high mud contents during an increasing discharge have also been shown by Kreugel & Terwindt (1969) for discharges exceeding 800 m^3 sec^{-1} in the River Meuse.

The trace-metal concentrations in the suspended

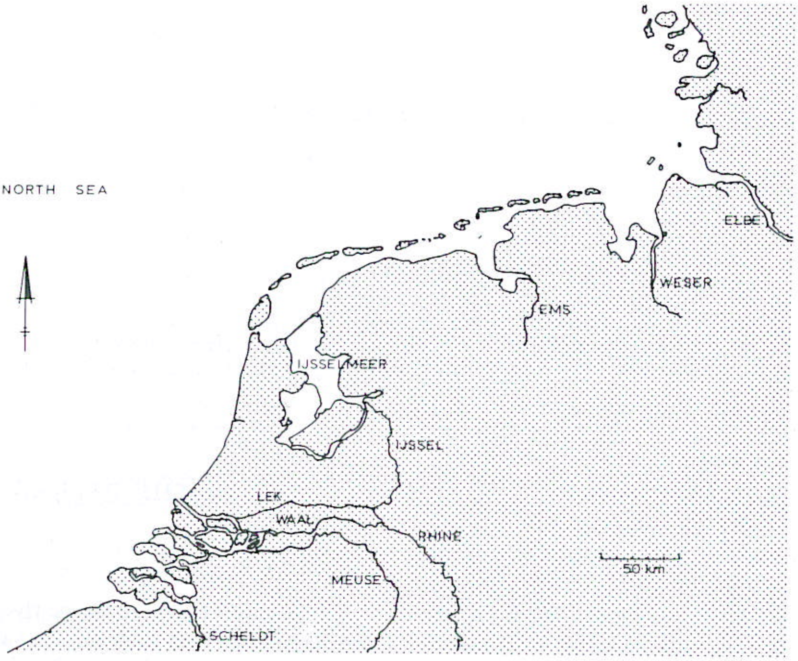


Fig. 1. Major rivers in Belgium, the Netherlands and Germany which influence the southern North Sea.

matter depend on the discharge of the river (Hellmann, 1970; Schleichert, 1975). With an increase in the discharge, the suspended mud concentrations increase but the metal concentrations in the suspended matter decrease. This relationship sometimes makes it possible to detect the influence of tributaries on the composition of suspended mud in the main river. An example has been given by Schleichert (1975) for the influence of the rivers Neckar and Main on the river Rhine.

In spring, most of the water in the Rhine is meltwater, so that as a consequence the suspended matter also mainly originates from its upper course. In the autumn period, precipitations in the upper course accumulate as snow, and the contribution of distributaries like the (polluted) rivers Main and Neckar increases. Since the cadmium concentrations in the river Neckar (and also other trace metals, with the exception of chromium) are much higher compared with the river Rhine, two different relationships are found between the discharge and particulate metal concentrations: one for the autumn period (Neckar influence) and one for the remainder of the year (Fig. 3). Also in the lower course of the Rhine, in the Nieuwe Merwede, particulate metal concentrations depend on the discharge (Fig. 3).

The decrease in particulate metal concentrations at high discharge is due to the contribution of contaminated eroded soil particles into the river system by the surface runoff. Also, during the high discharge the residence time of the sediment in the rivers is reduced because of the increased flow velocity, and consequently the particles have less time to pick up trace metals from polluted sources. Moreover, dissolved metal concentrations are much lower because of dilution by relatively uncontaminated water, so far less metal will be adsorbed. The above-mentioned phenomena make it difficult to determine the proper amount of mud transport and hence of metal transport. During high water waves large amounts of mud and metals may be transported. To determine these effects as well as the right transport quantities, frequent sampling of the river water is required. However, most data are based on weekly, bi-weekly or even monthly samplings, especially with regard to the trace metals, and often only individual measurements are available.

The river Scheldt

The river Scheldt, a relatively small river draining part of western Belgium, discharges into the Western Scheldt estuary (Fig. 1). Depending on the rainfall, the discharge rate may show considerable fluctua-

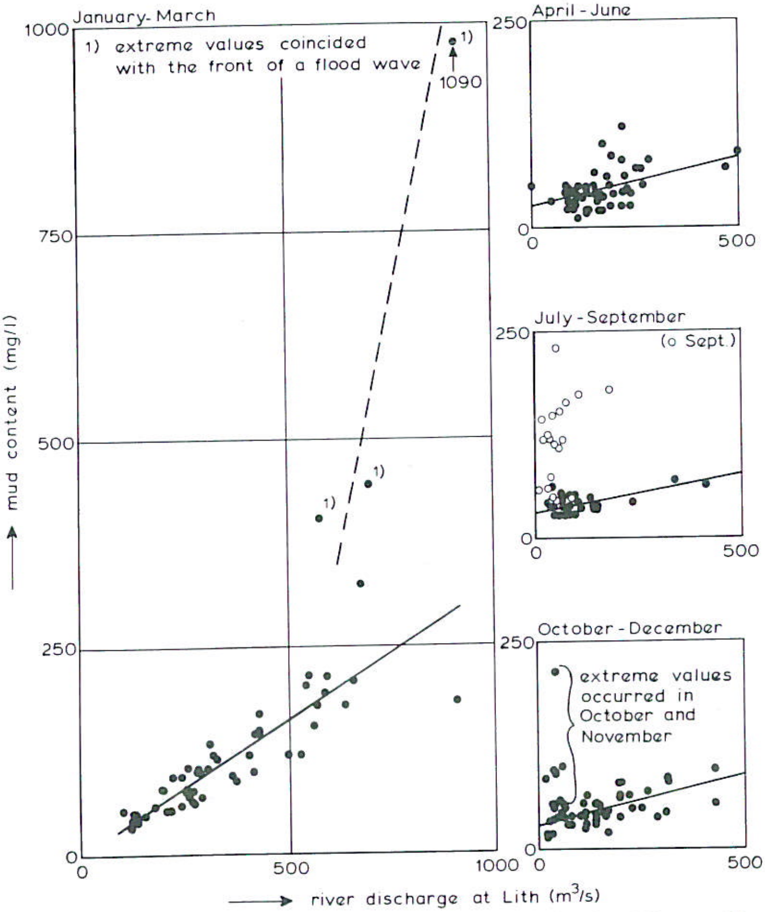


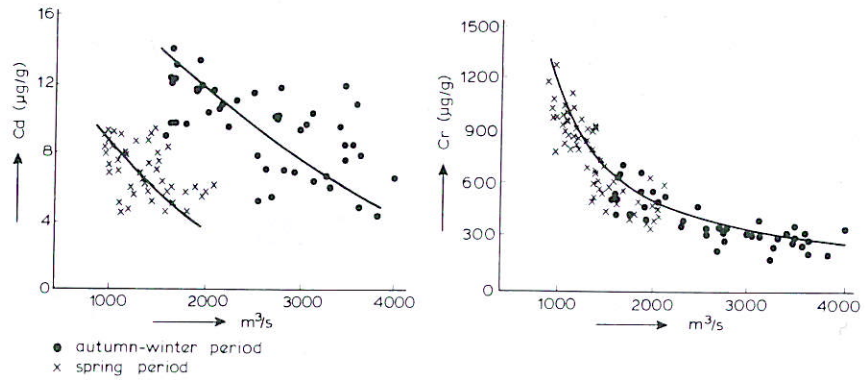
Fig. 2. Relationship between mud content and river discharge for the river Meuse at Ravenstein in 1971.

tions with time while the monthly mean discharge changes with the season. With the discharge of the water the concentration and the transport of suspended mud also vary with time. The annual mean discharges of water and mud amount to $3{\cdot}2 \times 10^9$ $m^3\,yr^{-1}$ and about 1×10^6 tons yr^{-1} respectively (Table 1). The latter figure is a rough estimate based on various data from the literature (McCave, 1973; Wollast *et al.*, 1973; Wollast, 1976; Terwindt, 1977; Eisma, 1978), ranging from 0·1 to $1{\cdot}41 \times 10^6$ tons yr^{-1}. Only a negligible amount of this mud supply seems to reach the North Sea as most of it is deposited in the estuary of the Western Scheldt. Metal concentrations in the suspended mud sampled in 1978 at Hoboken and those in sediments collected downstream from Antwerp are presented in Table 2.

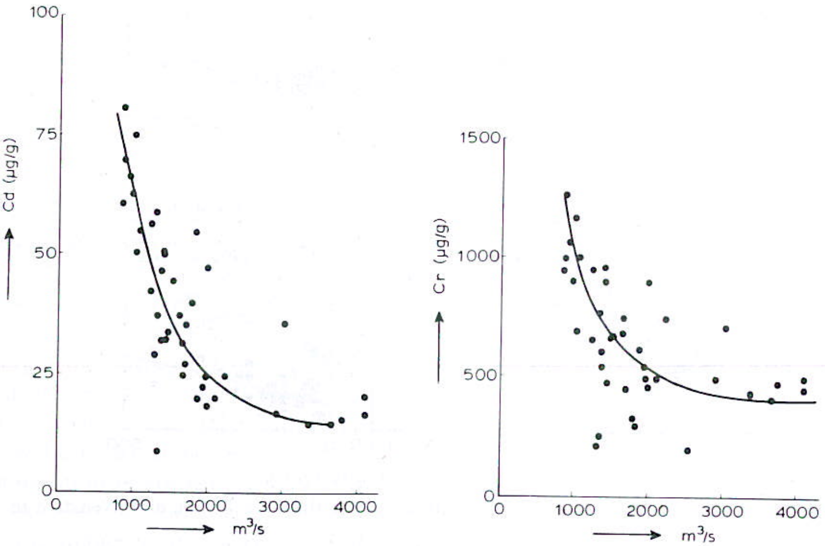
The rivers Rhine and Meuse

The rivers Rhine and Meuse influence the same areas in the Netherlands (Fig. 4). The river Meuse and the lower course of the river Rhine, as well as the entire delta area, have been subject to extensive interference and alteration by man, so that the many civil works affect the regime of the rivers Rhine and Meuse and the distribution of the water over the lower courses. The enclosure of the Zuiderzee (1932) and the Haringvliet (1979), especially, have had a great influence on the hydrology of the rivers Rhine and Meuse and the estuaries in the south-west of the Netherlands.

The distribution of the water of the Rhine is controlled by weirs in the lower Rhine and Lek (completed in 1970) and sluices in the closure dam of the Volkerak (completed in 1976) and the Haring-



A Rhine at Koblenz



B Rhine in the Netherlands (Nieuwe Merwede)

Fig. 3. The relationship between the water discharge and the metal concentrations in the suspended matter. (A) River Rhine at Koblenz (Schleichert, 1975). (B) River Rhine in the Netherlands: locality 3 of Fig. 4.

vliet (completed in 1970). The distribution is planned to comply with the so-called 'normal discharge scheme (NLP '70)' as given in Fig. 5. In fact, this makes the Rotterdam Waterway the main estuary of the rivers Rhine and Meuse at present. At high discharge most of the Rhine water enters the North Sea by way of the Haringvliet. Data on the discharge rates of water and mud of the rivers Rhine and Meuse are presented in Table 1.

The heavy metal pollution of the river Rhine is not a recent phenomenon, because early in the twentieth century the river Rhine could be regarded as heavily polluted with trace metals. Cadmium concentrations of 4·4 $\mu g\ g^{-1}$ are already 15 times the baseline level (0·3 $\mu g\ g^{-1}$), whereas the zinc concentrations are more than 1000 $\mu g\ g^{-1}$. Between 1920 and 1958 all trace-metal concentrations increased (Fig. 6), while between 1958 and 1979 this rise continued for cadmium only. Striking are the decreases for arsenic (after 1958) and for mercury (after 1973). In fact arsenic is close to the baseline level at present. Zinc and lead concentrations are also slightly decreasing.

The anthropogenic contribution to the metal concentrations in sediments from the river Rhine and in some of the other rivers studied exceeds background values by one order of magnitude (Salomons & de Groot, 1978). It can even be estimated that less than

Table 1. Water discharge, mud transport and some tentative data for particulate metal transport by the rivers Rhine, Meuse, Ems, Scheldt, Weser and Elbe. Me_t = total metal transport

River	Water discharge 10^9 m^3 yr^{-1}	Mud transport 10^6 tons yr^{-1}	Particulate metal transport* tons yr^{-1}						
			Cu	Ni	Zn	Pb	Cd	Cr	Me_t
Scheldt	3·2	1†	270	130	1430	400	55·6	390	2675
Meuse	10·3	0·7‡	144	70	2821	466	57	186	3743
Rhine	69·4	3·4§	884	391	4845	1547	120	1870	9657
Ems	3·2	0·07§	6	3	41	6	0·2	8	63
Weser	11	0·37§							
Elbe	22·5	0·8§	288	68	2000	208	14	296	2874

* Based on the most recent data from Table 2 and the annual mud transport. For the rivers Ems and Elbe no data on the suspended matter were available, therefore, the deposited sediment concentrations were used.

† Data on the annual silt discharge of the Scheldt range from 0·1 to $1{\cdot}42 \times 10^6$ tons yr^{-1} (McCave, 1973; Wollast *et al.*, 1973; Wollast, 1976; Eisma, 1978). Based on Wollast (1976) and Terwindt (1977) it is taken at approximately 1×10^6 tons yr^{-1}, although this figure seems rather high for this small river.

‡ Based on Terwindt (1977).

§ Hinrich (1971, 1974), Hellmann *et al.* (1977).

Table 2. Metal concentrations in bottom sediments and in suspended matter from a number of rivers

	Cu	Ni	Zn	Pb	Cd	Cr
Scheldt						
Bottom sediments 1974	165	66	1080	230	26·4	380
Bottom sediments 1978/79	180	61	1015	270	35·4	290
Suspended matter 1978	270	130	1430	400	55·6	390
Meuse						
Bottom sediments 1958	160	43	1520	380	28·5	215
Bottom sediments 1970	165	48	1430	325	24·2	325
Bottom sediments 1975	185	67	3050	475	41·5	330
Bottom sediments 1977	170	64	2630	440	40·4	255
Suspended matter 1977/78	205	100	4030	665	81·7	265
Rhine						
Bottom sediments 1922	68	36	1050	275	4·4	110
Bottom sediments 1958	295	54	2420	535	14	640
Bottom sediments 1970	325	62	1855	445	27	790
Bottom sediments 1977	285	76	1665	390	37·4	825
Suspended matter 1977/78	260	115	1425	455	35·4	550
Ems						
Bottom sediments 1964	95	38	504	66	2·6	115
Bottom sediments 1971	80	42	590	82	3·0	110
Elbe						
Bottom sediments	360	85	2500	260	16·9	370

The data on the bottom sediments have been corrected for differences in grain-size composition; the values refer to the calculated concentrations at 50 % < 16 μm (Salomons & Mook, 1977). Bottom sediments from the Scheldt were taken between the river Rupel and Antwerp; the suspended matter data refer to samples taken at Hoboken. Data on the suspended matter from the rivers Rhine and Meuse are from Rijkswaterstaat (1977–8). The bottom sediments from the rivers Rhine and Meuse were sampled in the Biesbosch area, those from the river Ems at the locality of Diele. The data for the river Elbe have been taken from Brummer & Lichtfuss, 1977. All data in μg g^{-1}.

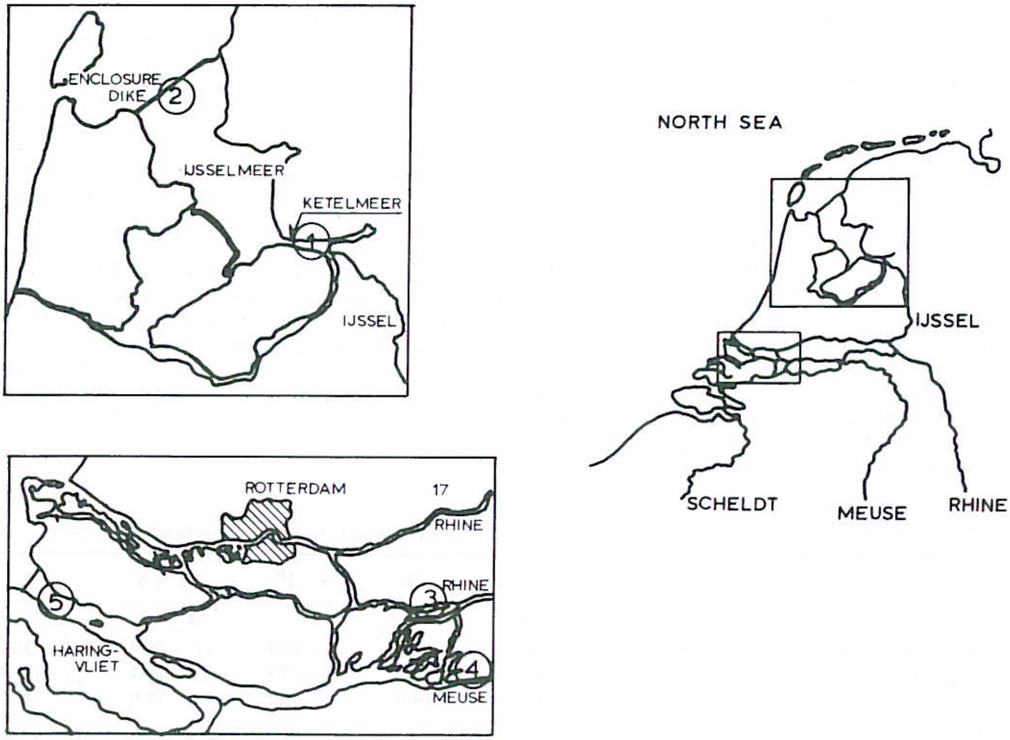


Fig. 4. Areas in the Netherlands influenced by the rivers Rhine and Meuse: IJsselmeer, Haringvliet and Rotterdam harbour.

1% of the cadmium found in sediments from the river Rhine originates from natural sources.

The zinc concentrations in the sediments of the river Meuse are higher than those in the Rhine; they show a large increase between 1958 and 1979. The chromium levels in sediments from the Meuse are lower compared with the Rhine; cadmium levels, however, are slightly higher. In Table 1, a tentative estimate is given of the total amount of particulate metals transported by the various rivers entering the southern North Sea, showing that the rivers Rhine and Meuse taken together account for 70% of the total metal transport.

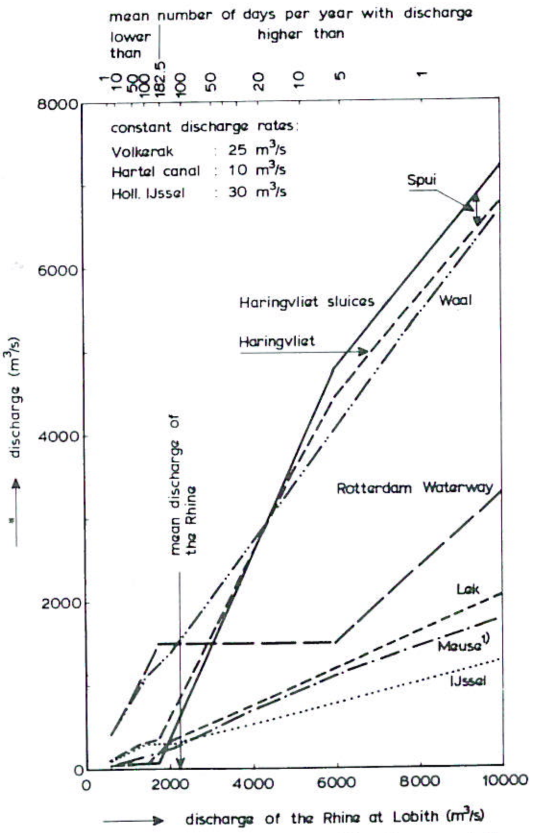
The rivers Ems, Weser and Elbe

The rivers Ems and Weser are relatively small rivers which drain the northern part of western Germany, with the Elbe, draining water and suspended mud from areas up to Czechoslovakia, being more important. Data on mud and water discharges of these rivers are obtained from Hinrich (1971, 1974) and Hellmann *et al.* (1977) and presented in Table 1. Metal concentrations in the sediments from the river Ems are given in Table 2. Zinc and lead concentrations increased slightly between 1964 and 1975. Metal concentrations in the river Ems are low compared with other rivers influencing the southern North Sea. Metal concentrations in the river Elbe have been studied in detail by Lichtfuss & Brümmer (1977), and some results are presented in Table 2. The total amount of particulate metals transported by the river Elbe is about equal to that of the river Scheldt.

PROCESSES AFFECTING THE TRANSPORT OF SEDIMENT AND METALS BY RIVERS TO THE NORTH SEA

Estuaries

In the estuaries the fresh river water and the salt sea-water meet and are mixed by the strong tidal motion and the drift currents generated by the wind. The mixing ratio can often be determined from the salinity data. The hydrodynamics of an estuary, in



1) 50% discharge of the Meuse at the coinciding discharge of the Rhine at Lobith

Fig. 5. Distribution of the discharges of the Rhine and Meuse according to the discharge scheme of the weirs in the lower Rhine and the Haringvliet sluices (NLP 70).

general, are very complex because of the effects of such independent variables as tide, river discharge and wind and the density differences between water masses in the estuary caused by differences in salinity and temperature. In this system also marine and fluvial suspended matter meet and are mixed by the hydrodynamic forces. The mixing ratio of marine and fluvial suspended mud (and also of the particulate metals) cannot simply be obtained from the mud concentration in the river and the sea respectively, because settling (differential) processes occur. These processes can be caused by different phenomena which do not necessarily affect the fluvial and marine mud transports in the same way.

Knowledge of the mixing ratios of marine to fluvial material in the suspended matter, deposited sediments and in dredged material is essential for an understanding of the transport processes as well as for making a balance of fluvial mud and particulate metals in the estuarine environment. Studies using radioactive or activable tracers provide answers to the movement of the sediments over only relatively short time periods. More useful for long-term transport studies are natural tracers. Several natural tracers (e.g. natural differences in composition between two or more sediment sources) are available in principle for tracing argillaceous sediments:

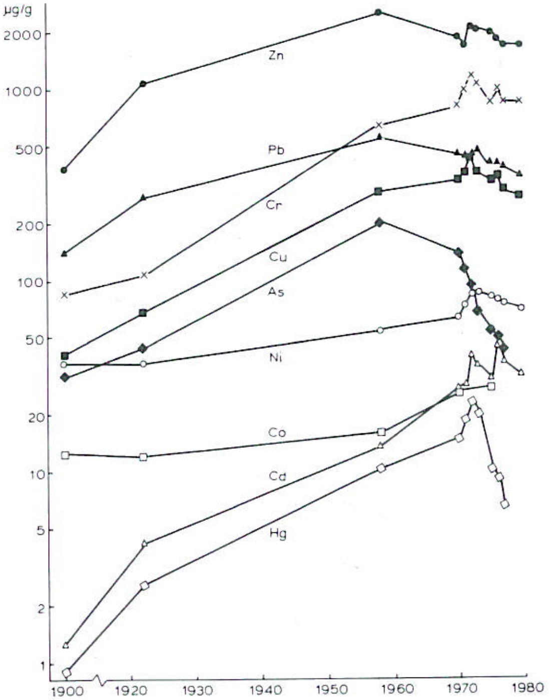


Fig. 6. History of heavy metal pollution of the river Rhine. Metal concentrations in the deposited sediments. All data corrected for grain-size differences.

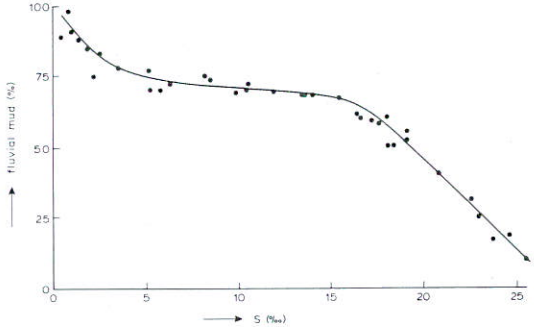


Fig. 7. Relationship between the percentage of fluvial matter in suspended matter from the Scheldt estuary and salinity.

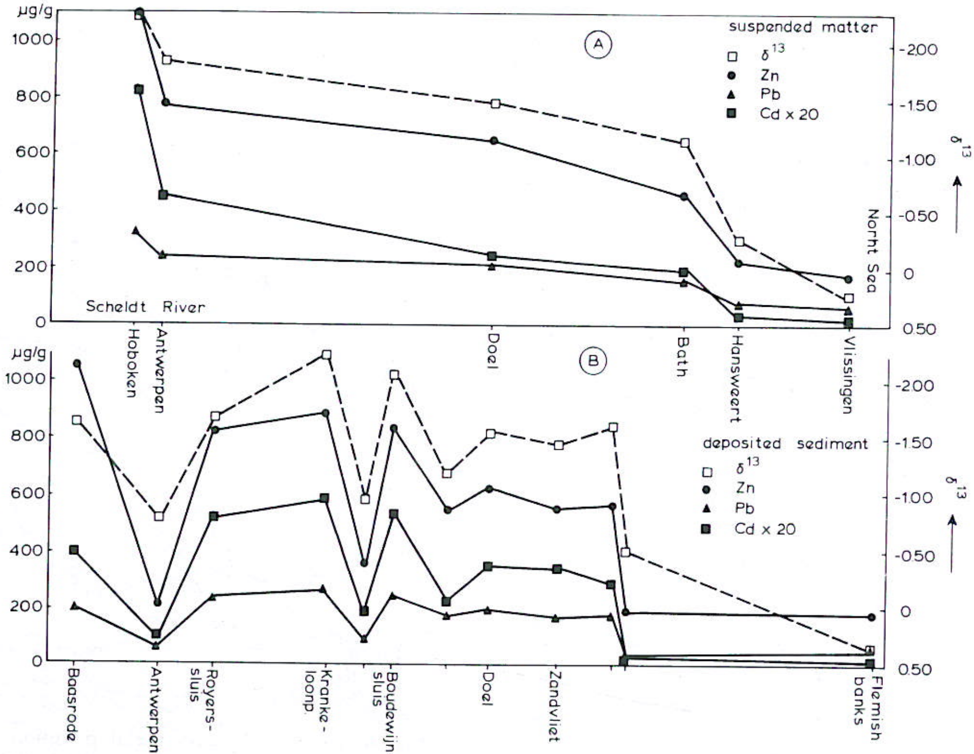


Fig. 8. (A) Metal concentrations and isotopic composition of the carbonates in suspended matter from the Scheldt estuary. (B) Metal concentrations and isotopic composition of the carbonates in deposited sediments from the Scheldt estuary.

differences in mineralogy; differences in chemical composition; and differences in the isotopic composition. The first two differences, in most cases, refer to the differences in the overall composition between two sediment sources; the third refers to differences in the isotopic composition of individual components (e.g. clays) making up the sediment. A difference in composition between two sediment sources may be used as a natural tracer provided that the tracer shows conservative behaviour, i.e. the amount of tracer per unit weight of sediment in a specific source (river or sea) is not subject to variations in time, either during transport or after deposition (Salomons *et al.*, 1978). Useful tracers for the estuaries along the southern North Sea are the isotopic composition of the carbonates and clay minerals and, in a few cases, the multi-element method (as determined with neutron activation analysis) (Salomons, 1975; Salomons *et al.*, 1975; Salomons & Mook, 1977).

The ratio of marine to fluvial mud in the suspended matter is not a simple function of the salinity. The relationship between salinity and the percentage of fluvial mud in the suspended matter for the Scheldt estuary shows three distinct ranges (Fig. 7). With low salinities the percentage of fluvial mud decreases relatively quickly to about 70 %, and stays more or less constant between 5 and 15 ‰. The amount of fluvial mud decreases again with salinities of 15 ‰ and higher. This second decrease coincides with the outflow of the Scheldt in the Western Scheldt.

In general the fluvial mud content in the bottom sediments does not compare with that in the suspended matter (Fig. 8). The isotopic composition of the carbonate fraction (representative of the amount of fluvial mud) in the suspended matter decreases in the seaward direction; however, the isotopic composition of the carbonates in the bottom sediments shows an erratic behaviour. High values, indicating the presence of marine mud, are found at two localities in the estuary. The pattern of the heavy-metal concentrations in the bottom sediments is identical to that of the isotopic composition of the carbonates; e.g. low metal concentrations (influence of marine mud) correlate with high values for the isotopic composition of the carbonates.

Table 3. The calculated and observed trace-metal concentrations at Leerort (Ems estuary) and in the Haringvliet (Rhine–Meuse estuary)

	Concentration (μg g⁻¹)			
	Leerort		Haringvliet	
Metal	Calculated	Measured	Calculated	Measured
Cadmium	0·7	0·8	5·0	5·0
Chromium	86	97	195	224
Copper	17·1	22·6	82	99
Nickel	28·0	33·3	25·8	30·8
Lead	49·0	58·6	177	213
Zinc	196	220	675	870

The mixing of relatively uncontaminated marine sediments with contaminated fluvial sediments explains to a large extent the decrease in metal concentrations in bottom sediments as observed in the Rhine–Meuse, Ems, Elbe and Scheldt estuaries (Salomons & Mook, 1977; Müller and Förstner, 1975) (see also Figs 8 and 11). However, superimposed on this physical process of mixing, chemical processes such as adsorption, precipitation and mobilization may occur. To determine whether these processes affect the composition of the particulate metals, it is essential to know the mixing ratio of marine to fluvial sediments. If that mixing ratio and

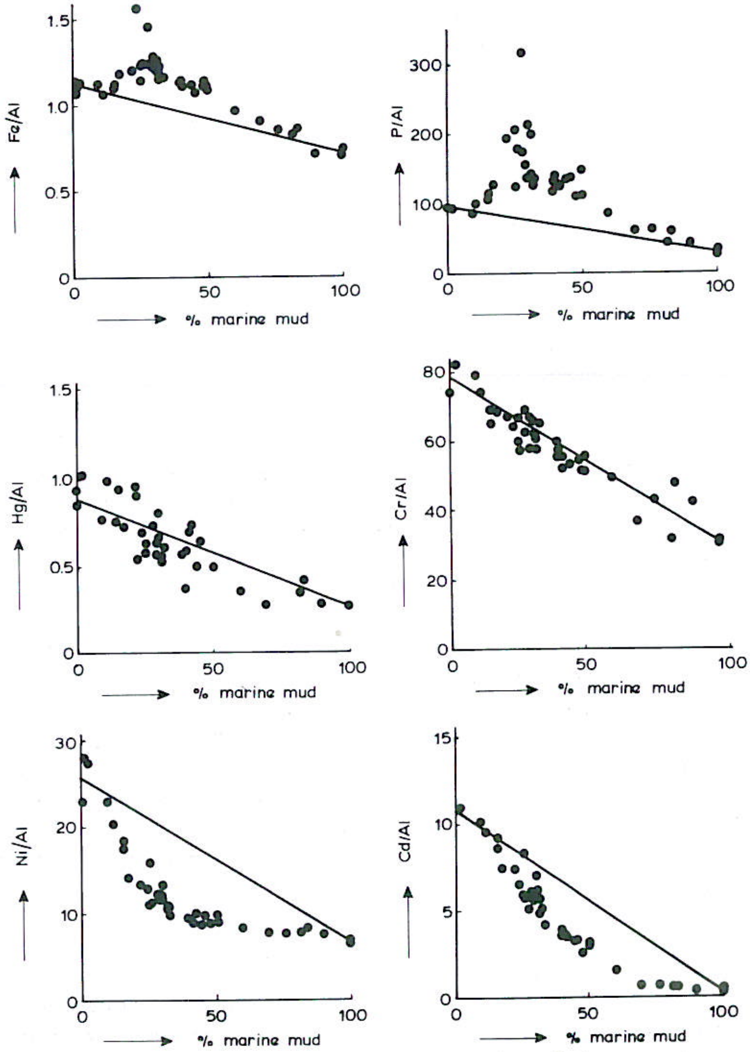


Fig. 9. The relationship between trace element to aluminium ratio and the percentage marine mud in the suspended matter in the Scheldt estuary. The straight line is the theoretical mixing curve connecting the fluvial and marine end-members.

the relevant metal concentrations are known, it is possible to calculate the trace-metal concentrations in the estuarine deposits for trace metals behaving conservatively. If so, the computed concentrations should be equal to those actually measured. Values that exceed those actually observed would point to mobilization processes; lower values, on the other hand, would indicate precipitation or adsorption processes.

Computed and measured metal concentrations for estuarine deposits from the Rhine–Meuse and Ems estuaries are presented in Table 3 (Salomons & Mook, 1977). The computed and measured values are very similar, showing that the mixing of marine and fluvial sediments is the main cause for the observed decrease in metal concentrations. However, the computed values tend to be slightly lower, which can be explained by the possible occurrence of precipitation–adsorption processes. Evidence for adsorption–precipitation processes has also been given by Duinker & Nolting (1977) for the Rhine–Meuse estuary. The dissolved trace metals show a drastic decrease in their concentrations over the range 0–5 ‰ salinity.

More detailed investigations have been carried out in the Scheldt estuary. Not only the deposited sediments, but also the suspended matter were studied. To determine the mixing ratio of marine to fluvial sediments, the stable isotopic composition of the carbonates was used. Results on the bottom sediments, presented in Fig. 8, have already been discussed. The behaviour of 24 trace elements during estuarine mixing was studied by analysing the suspended matter and determining the ratio of marine to fluvial sediments, and some of the results are presented in Fig. 9, with the straight line giving the theoretical mixing curve for the suspended matter. Positive deviations from the mixing curve (addition processes) are found for Fe and P (shown in Fig. 9), but also for V, As, Eu, Tb and Mn. Conservative mixing was observed for Yb, Ta, Rb, Th, Cs, Hg and Cr. For the elements Co, Pb, Zn, Cu, Sb, Cd and Ni negative deviations from the mixing curve were found, indicating the transfer of these elements from the particulate phase to the estuarine waters.

These results, together with those for the rivers Rhine, Ems and Elbe, show that the mixing of marine and fluvial sediments is the dominant process which affects the particulate trace-metal concentrations. However superimposed on the mixing process, addition and/or removal processes, depending on the metal and on environmental conditions, are taking place.

The differences in behaviour of trace metals in the Scheldt and in the Rhine–Meuse estuary show the need for more research into their estuarine behaviour. The influence of fluvial sediments on the composition of the sediments deposited close to the mouths of the estuaries appears to be limited. In the Ems estuary at Leerort, situated above the mean salinity limit at high water slack, already 90 % of the sediments are derived from the marine environment. In the Western Scheldt, the influence of River Scheldt sediments can be detected only down to Hansweert, whereas in the Rotterdam Harbour area, at least downstream of the Oude Maas, most of the deposited sediments originate from the North Sea.

Harbour areas

Important harbour areas for sea-going vessels along the European coast of the southern North Sea are the harbours of Antwerp on the Scheldt, Rotterdam on the Rotterdam Waterway, IJmuiden (Amsterdam), Emden on the Ems–Dollard estuary, Wilhelmshaven on the Jade estuary, Bremerhaven on the Weser estuary and Hamburg on the Elbe estuary. In these areas artificial navigation channels and harbour basins have been dredged, thereby disturbing natural conditions. These new situations can only be maintained by considerable maintenance dredging, as the channels and harbour basins act as sediment traps. The amount of sediment removed by dredging is considerable as can be seen from the total amount removed each year from the harbours given in Table 4. This 75–80 × 10^6 tons is already twice the total mud budget of the North Sea. Part of this spoil is permanently removed from the system (applied as landfill), but most of it is dumped at selected dumping sites in the North Sea and so it is not lost from the aquatic system.

The artificial deepening of the estuaries in the navigation channels and in front of jetties at the bank locally reduces the natural current velocities. This decreases the capacities both of the suspended sediment transport and the sediment transport along the bottom, so siltation of the dredged areas occurs. Theoretically, the following formula can be derived, which can be used to estimate the silting rate of suspended sediment in those areas:

$$\Lambda\Upsilon_s = wc_1A\left\{1-\left(1+\frac{18}{C_2}\log\frac{h_1}{h_2}\right)\frac{u_2}{u_1}\right\}$$

Table 4. Mean annual maintenance dredging of harbours along the coast of the southern North Sea in the period around 1975

Harbour	Mean annual maintenance dredging (approximate values in 10^6 m^3 yr^{-1})
Dutch harbours at the Western Scheldt estuary	2·5 (Data Rijkswaterstaat)
Antwerp	10 (Wollast, 1976)
Rotterdam	21 (van Oostrum, 1978)
Scheveningen	0·2 (Data Rijkswaterstaat)
IJmuiden	2·5 (Data Rijkswaterstaat)
Emden/Delfzijl	15 (Data Rijkswaterstaat)
Wilhelmshaven	?
Bremen/Bremerhaven	10 (Niebuhr *et al.*, 1961; D'Angremond *et al.*, 1977)
Hamburg/Cuxhaven	11 (D'Agremond *et al.*, 1977)
London	0·7 (D'Angremond *et al.*, 1977)
Hull	4·8 (D'Angremond *et al.*, 1977)

where

$\Delta\Upsilon_s$ = silting rate of suspended sediment in a dredged area;

w = settling velocity of sediment grains in stagnant water;

c_1 = sediment concentration (weight) of the water upstream of the deepened area;

A = horizontal area of the deepened part of the estuary;

C_2 = roughness coefficient of Chézy of the deepened area;

h_1/h_2 = ratio of water depths upstream and at the deepened area respectively;

u_2/u_1 = ratio of flow velocities upstream and at the deepened area respectively, which can be estimated by:

$u_2/u_1 = h_1/h_2$ (flow perpendicular to a dredged channel)

or

$u_2/u_1 = F_1/F_2$ (flow parallel to a dredged channel) where F_1/F_2 is the ratio of the cross-sections of the estuary at the dredged region before and after the dredging of the channel.

This shows the dependence of the silting rate on the dimensions of the dredged area (A, h_1/h_2 and/or F_1/F_2), the sediment characteristics (w), and the natural sediment transport (c_1).

The siltation of the harbour basins along the estuaries, however, is governed by a different mechanism. In general, the flow velocities in these basins are far lower than those outside and so are the capacities of sediment transport. This means that a larger part of the suspended sediment that penetrates into a basin will settle there. The silting rate of a harbour basin depends upon the volume of water which is exchanged with each tide and the sediment concentration of the water which is penetrating. The water exchange is determined by: (a) the tidal volume of the harbour basin; (b) the horizontal exchange in the harbour entrance by the flow in the estuary; and (c) the vertical exchange, caused by density differences between the water in the basin and in the estuary. The first quantity is simply given by the area of the harbour times the tidal range. The second quantity is determined by a more complex mechanism.

A passing flow in front of the harbour entrance generates an eddy at that point. Silty water from the passing flow is exchanged with relatively clean water from the eddy by diffusion, with the mud penetrating in the slowly flowing eddy slowly settling, etc. This horizontal exchange depends on the flow velocity in the estuary and on the size and shape of the harbour entrance. Both the exchange by tidal volume and the horizontal exchange mainly affect the harbour at and near the entrance. The latter particularly is an effective exchange mechanism that affects the whole harbour basin. The density differences generating this type of exchange flow are caused by the salinity of the water in front of the basin fluctuating with the tide. Around high water slack the water in front of the basin will be more saline than the water in the basin and consequently the denser estuary water will penetrate along the bottom into the basin, replacing the same amount of water which is flowing out at the surface. Around low water slack the opposite occurs. In both cases harbour water, of which the suspended sediment has partly settled in the basin, is replaced by water with a higher mud content from the estuary, with the density-induced exchange velocities being proportional to the square root of the density differences and the water depth. The exchanged volume per tide can be expressed as:

$$V_e = F * hb * 0{\cdot}45\sqrt{\left(\frac{\Delta\rho}{\rho}gh\right)} * T,$$

where

V_e = density-induced exchanged volume per tide;

f = empirical coefficient a.o. depending on the harbour layout;

hb = cross-sectional area of the harbour entrance;

$0{\cdot}45\sqrt{\frac{\Delta\rho}{\rho}gh}$ = initial velocity of lock exchange flow (Yih, 1965; Abraham & Eysink, 1971);

$\Delta\rho$ = characterizing density difference, e.g. maximum density difference during a tidal period;

ρ = density of water;

T = tidal period.

So the mechanism of vertical exchange is most effective in deep harbours with a wide entrance in the area of a salt-water wedge which is moving forth and back with the tide.

In addition, however, there are two other mechanisms that enhance the siltation of harbour basins in the area of the salt-water wedge. First, in the transition region where fresh water mixes with saline water, mud particles start to flocculate. This increases the effective fall velocities of the silt particles and so stimulates the settling of mud in relatively calm regions like the harbour basin. Secondly, the density difference between fresh water and seawater distorts the flow-velocity profiles and causes the water to circulate in the transition zone with a residual inflow along the bottom and a residual outflow at the surface. The salt penetrating in this way along the bottom is balanced by entrainment of water from the lower layer into the upper layer, where it is discharged to the sea. Mud particles also follow this path, although they may sink back into the lower layer after they have been entrained into the upper layer. So, in the transition zone between fresh and salt water a circulation of suspended sediment is possible, which in general causes it to accumulate, as often is found in increased sediment concentrations of the water in the transition zone (Postma, 1960, 1967b; Wollast *et al.*, 1973; Peters & Sterling, 1976). This increases the sediment contents of the exchanged water volumes and so the siltation of the harbour basins. The sediment settled in the artificial channels and basins is regularly removed by maintenance dredging, the spoil being partly brought to land and partly returned to the aquatic system by dumping it at sea or at particular dumping sites in the estuary. During the dumping process a large part of the fine sediment is washed out and entrained by the local currents. The rest of the spoil sinks to the bottom where, in general, it is gradually eroded and redistributed. In fact, the natural transport of sediment is temporarily interrupted by settling in the man-made settling basins and sediment is, at least partly, brought back again into the aquatic system for further transportation by nature by the dumping of maintenance dredging material in the estuary or at sea.

Harbour of Rotterdam: case study

The history of the harbour of Rotterdam, situated along today's main branches of the Rhine delta, gives a good impression of the present importance of the above-described processes in the transport of mud to the southern North Sea. The Rotterdam Waterway is only relatively young; it is a man-made branch of the Rhine delta which was dug in 1868. During the next three decades it took a great deal of effort to achieve a stable access channel which was satisfactory for the harbour of Rotterdam. This situation with a stable profile of the Rotterdam Waterway lasted from about 1897 to 1909 but then gradually changed due to harbour extensions (such as the first and second Petrol harbours and the Maas-, Rijn-, Waal- and Eem-harbour basins) and improvements of the Rotterdam Waterway until a second equilibrium (including the effect of maintenance dredging) was reached in the period from about 1950 to 1958. Since then, considerable new harbour extensions (such as the Botlek and Europoort basins) and improvements of the fairway to Rotterdam have led to further changes in the estuary. During these developments the storage area increased with the increasing harbour area, and the hydraulic resistance against the penetration of the tide and the salt sea-water decreased with the increasing water depth in the Rotterdam Waterway. This resulted in a considerable increase of the tidal range in Rotterdam, and further inland in a drastic increase of the tidal volumes, the salt penetration and, which is particularly important for the subject under consideration, the maintenance dredging (Table 4). Since the closure of the Haringvliet in November 1970, the mean tidal volume of the Rotterdam Waterway has decreased from about 210×10^6 m^3 (mean ebb volume plus mean flood volume at Hook of Holland) to about 170×10^6 m^3, which also has had an unfavourable effect with regard to the maintenance dredging (Haring, 1978). The amount of spoil from the basins increased from $0{\cdot}3$–$0{\cdot}4\times10^6$

Table 5. Some characteristic figures of the Rotterdam Waterway in relation to the historical development of the Rotterdam harbour

Year	1900	1923	1955	1962	1974
Area of harbour basins (10^4 m^2)	60	570	850	1600	2100
Cross-sectional area of KMR 1030 at Hook of Holland (m^2 below LW)	3200	3625	5100		7900
Sum of mean ebb and flood volumes at KMR 1030 (10^6 m^3)	112	138	173	196	170*
Mean tidal range at Rotterdam (m)	1·45	1·50	1·70	1·75	1·72
Mean annual maintenance dredging of the basins (10^6 m^3)†		0·3–0·4	2–2·5	4–4·5	7·5–8

* On 2 November 1970 the Haringvliet was closed off from the sea, which reduced the mean tidal volume of the Rotterdam Waterway at the Hook of Holland from approximately 200×10^6 m^3 to about 170×10^6 m^3.

† Based on Terwindt (1971) and van de Ridder (1978).

m^3 in 1923 to 7·7–8 × 10^6 m^3 in 1974; a 20- to 25-fold increase in about 50 years of harbour development.

At present the total annual maintenance dredging amounts to about 21×10^6 m^3, of which approximately 70 % is mud. Expressed in tons of dry matter per year, this corresponds with about 10×10^6 tons yr^{-1}, which is about 5 times the amount supplied by the river. More detailed information is shown in Table 5, which gives the data of maintenance dredging as provided by the Public Works Department of Rotterdam and by the Rijkswaterstaat.

The siltation of the fairway and harbour basins does not occur gradually but shows a seasonal effect with, in general, the highest silting rates being during the winter (van de Ridder, 1978; Ottevanger, 1978; Van Bochove & Nederlof, 1979) (see Fig. 10). The seasonal effects in river discharges and meteorological conditions (storm periods) play a role in this phenomenon, and there are indications that particularly stormy weather has a great influence on the supply of sediment that settles in the harbour area (Terwindt, 1977). It is obvious that the harbour basins and fairways act as important sinks for marine sediments. According to the results of stable isotope studies the contribution of marine sediments in the spoil decreases in a landward direction. In the Europoort area the amount of marine mud is estimated to be about 90 %, in the Botlek area about 60 % and in the most easterly harbours about 25 %.

The spoil of the maintenance dredging is partly dumped on land, but the greater part is dumped at sea at a site (Loswal Noord) 5 km offshore northwest of the mouth of the Rotterdam Waterway. Since 1970 a mean annual discharge of about 14·5 × 10^6 m^3 of spoil has been released there, representing about 7 × 10^6 tons of dry sediment per year. At least half to three-quarters of this spoil is mud (Terwindt, 1971, 1978; Van Oostrum, 1978), which has meant a mean annual mud discharge here of 4–5 × 10^6 tons (dry matter). In the past few years this amount of spoil dumped at sea has been increased to about 17·5 × 10^6 m^3 yr^{-1}, containing 5–6·5 × 10^6 tons of dry mud. A large proportion of this muddy part of the spoil is resuspended during the dumping process as well as by wave and current action on the bottom (Terwindt, 1978). In fact, it is an important source of (reactivated) mud in the North Sea.

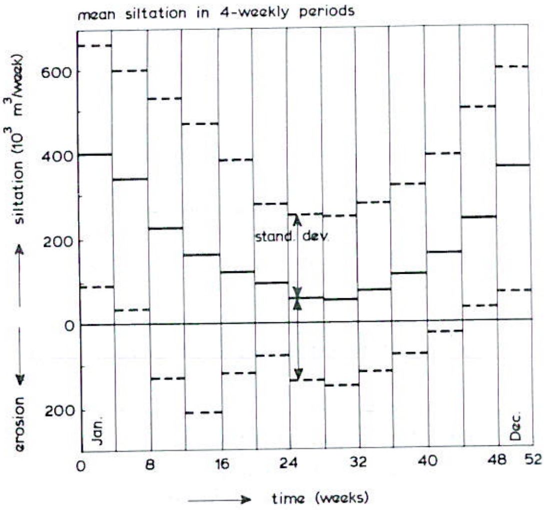


Fig. 10. Seasonal effect in siltation of the Rotterdam harbours (after van Bochove & Nederlof, 1979).

The spoil from the westerly harbours, which is less contaminated because of low admixtures of fluvial sediments (Fig. 11) is dumped at sea (Table 6), while the most severely contaminated spoil from the easterly harbour areas with high admixtures of fluvial sediment (Fig. 11) is dumped on land. So the landfill

Table 6. Dredging data of the Rotterdam harbour area. Based on data supplied by Rotterdam (period 1970–5, see also Stuurgroep Berging Baggerspecie (1978) and Rijkswaterstaat (period 1974–8)

	Maintenance dredging	
Location	Quantity $(10^6\ m^3\ yr^{-1})$	Dumped at sea Quantity $(10^6\ m^3\ yr^{-1})$
Fairways		
Euro channel	1·1	
Maas channel	1·5	1·5
Maas entrance	4·5	4·5
Caland and Beer channels (entrance)	4·5	4·5
Rotterdam Waterway KMR 1000–1033	2·8	2·8
Adjacent river area	0·9	
Sub-total	15·3	13·3
Harbours		
Europoort	1·2	1·2
Botlek	3·9	1·8
Other harbours	2·4	
Total quantity	22·8	16·3

areas act as important sinks for fluvial sediments and their associated pollutants. However, not only 'fluvial' metals end up in the landfill areas, but also relatively large amounts of 'marine metals' are put on land as well (Fig. 12).

Using the approximate mixing ratios of marine to fluvial sediments in the Rotterdam harbour area, the amount of dredged material and its metal concentrations, the amount of metal of fluvial origin which ends up in the landfill areas and which is dumped at Loswal Noord has been calculated. The results for 1977 are shown in Table 7. The balance shown in Table 7, however, is subject to a number of uncertainties. First, the survey of the Rotterdam Harbour was conducted in September 1977. The preceding period was one of decreased siltations; moreover, the major period of siltation is in winter (Fig. 10). And as the supply of mud is mainly from the sea, the percentage of marine mud in the dredged material may therefore be higher and thus, by using the data of September, the amount of fluvial mud transported to the North Sea (Loswal Noord) may be overestimated.

A second uncertainty concerns the ratio of marine to fluvial sediments in the major siltation area (Europoort/Maasmond). The natural tracers are rather insensitive at low admixtures of either marine to fluvial, or fluvial to marine sediments. The best estimate of 10% fluvial sediments in the dredged material from the Europoort/Maasmond may be out by 5%, and this may result in a large uncertainty about the metal balance. Additional data on the dredged material as well as further research into natural tracers are therefore required to produce a reasonably clear and accurate picture.

Freshwater basins

Two freshwater basins have been created by man within the delta area of the rivers Rhine and Meuse, which affect the amounts of mud and of particulate and dissolved metals entering the North Sea. The first basin, the IJsselmeer, has existed since 1932 when the former Zuiderzee was closed off from the Wadden Sea, while the second is the Haringvliet basin which was created in November 1970 (Fig. 4).

After the creation of the IJsselmeer the tidal motion has disappeared completely and the basin gradually became fresh by flushing with Rhine water supplied by the river IJssel and discharging through the sluices in the Enclosure dyke. Basically the water in this basin was practically stagnant and had a long residence time. Then the area of the basin gradually diminished as a consequence of the reclamation of polders, causing a decrease in the residence time of the water. At present, the mean residence time is still considerable: about 6 months. Primarily most of the suspended matter from the river IJssel settles close to the river mouth (the Keteldiep area); consequently, the deposits there have very high concentrations of trace metals (Table 8). Part of the suspended matter is directly transported into the lake itself, the bottom sediments of which are easily stirred up during stormy periods and transported by wind-induced drift currents. This mechanism of diffusive sediment transport is possible due to the restricted depth and the great surface area of the basin, allowing the wind energy to create sufficient turbulence in the water to stir up the bottom sediments. Fig. 13 shows the relationship between the wave height and the suspended-matter concentration. The amount of mud brought into suspension during storms is several times the yearly supply by the river IJssel. This and the long residence time of the water in the lake indicates that the sediment transports in the lake will be strongly affected by diffusion processes. Due to this the bottom sediments, with low metal concentrations, are mixed intensively with the recent contaminated suspended matter. As a consequence

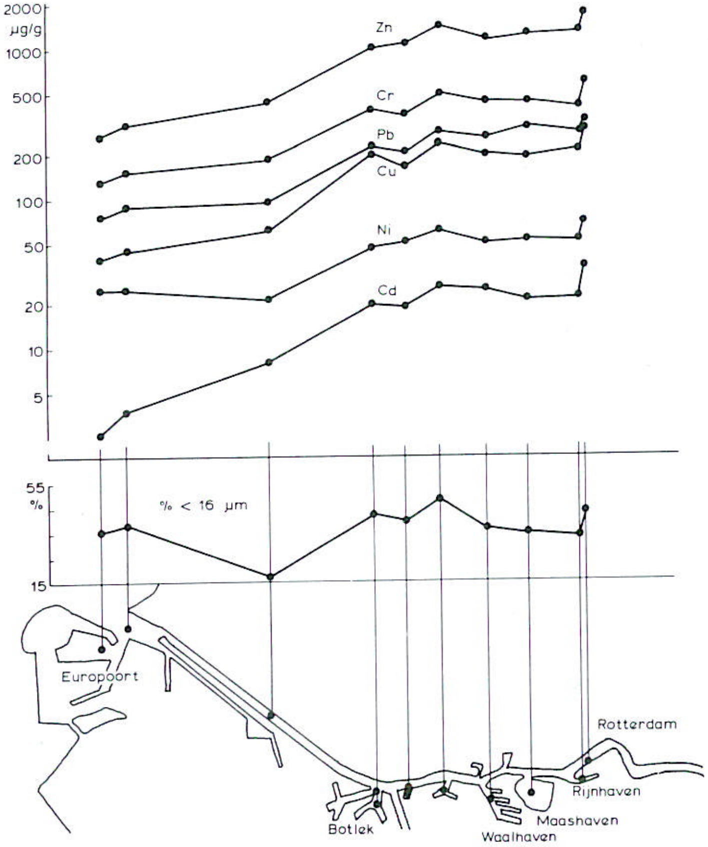


Fig. 11. Mean metal concentrations in dredged material from the Rotterdam harbour. The lower curve represents the mean grain-size composition (percentage < 16 µm) of the samples analysed. This survey was conducted in September 1977, and the data are based on about 150 sediment samples.

hardly any gradients in the metal concentrations in the bottom sediments are found (intensive reworking during storm) and, due to the admixture of relatively uncontaminated bottom sediments, the metal concentrations in the suspended matter at the Enclosure dyke are lower compared with the Ketelmeer (Table 9).

Apart from physical mixing processes, three more processes affect metal concentrations in the IJsselmeer, all directly or indirectly related to the high nutrient load of the river IJssel. The high phosphorus input into the IJsselmeer and the relatively long residence time (6 months) cause massive algal blooms. As a consequence, large amounts of organic matter are produced, part of which accumulates in the bottom deposits. Also the large amount of phosphorus taken up by algae becomes part of the sediment. The phosphorus balance of the lake,

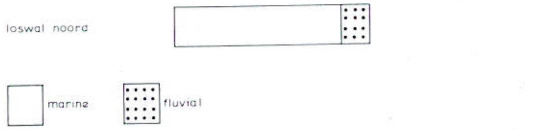


Fig. 12. Tentative metal balance for the Rotterdam harbour. (Total amounts of trace metals (Cu+Cr+Pb+Zn+Ni+Cd) are given.)

Table 7. Tentative balance of trace metals of fluvial origin in dredged material from the Rotterdam harbours (1977) (in tons (1000 kg))

	Zn	Cu	Cr	Pb	Cd	Ni	Me_t
Landfill							
Rijn-, Maas-Waalhaven	462	73	156	105	8	20	823
Eem-, 2nd Petroleum-, 1 Petroleum harbour, Botlek	807	150	318	175	16·3	40	1507
Total amount fluvial metals to landfill	1269	223	474	280	24·3	60	2330
North Sea							
Europoort	229	33	103	62	3·2	16	446
Botlek	233	45	92	54	4·4	12	440
Total amount of fluvial metals to the North Sea	462	78	195	116	7·6	28	887

Table 8. Metal concentrations in sediments from the IJsselmeer. All concentrations in $\mu g\ g^{-1}$ and corrected for grain size differences by using the calculated data at 50 % < 16 μm (Salomons & de Groot, 1978)

	Zn	Cu	Ni	Pb	Cd	Cr
Ketelmeer 1977	1960	250	67	320	34	570
IJsselmeer 1977	430	40	30	73	2·8	94
IJsselmeer 1933	133	19	39	39	0·4	88

therefore, can be used to estimate the amount of organic matter which accumulates in the bottom deposits (Postma, 1967a). In 1977 and 1978 the accumulation of organic matter was estimated at about 160,000 tons yr^{-1}, while Postma (1967a) had calculated for the period 1932–65 a mean accumulation of 120,000 tons yr^{-1}. Although metals are accumulated by algae, the metal concentrations in the algae are lower compared with those in the inorganic suspended matter (Salomons & Mook, 1980). Therefore the addition of newly formed organic matter to the suspended matter and to the bottom sediments causes a decrease in metal concentrations (dilution effect).

The uptake of carbon dioxide by algae and carbon dioxide exchange between the surface water and the atmosphere are responsible for the pH increase in the lake from 7·3 (river value) to more than 9 (in summer) in the northern part of the lake. At these high pH values, calcium carbonate precipitates. The pH of the lake in winter reaches values of only 8, and no precipitation of calcium carbonate takes place. In fact the dissolved calcium concentrations in the lake show distinct seasonal cycles. The production of calcium carbonate in the lake was estimated at 400,000 tons yr^{-1} for 1977 and 1978 (Salomons & Mook, 1980). Because metal concentrations in calcium carbonate are low, its addition to the suspended matter causes an apparent decrease in metal concentrations (dilution effect).

The third process affecting metal concentrations is also related to the pH increase, which causes an adsorption of some dissolved trace metals. The adsorption process is important for cadmium, chromium and zinc, because the adsorption of these three metals is strongly dependent on the pH over the range observed in the lake. On the other hand, the adsorption of copper is not strongly dependent on the pH between 7 and 9 and therefore adsorption plays a minor role in the removal of copper from solution (Fig. 14). A comparison between dissolved and particulate metal concentrations in the mouth of the river IJssel (Ketelmeer) and the Enclosure dyke shows the extent of these various (physical and chemical) processes. The IJsselmeer not only acts as a sink for particulate metals but also for dissolved metals (Tables 9 and 10).

The lake not only acts as a sink for sediment transported by the river IJssel into the lake but, due to the biogeochemical processes, an internal sediment production takes place which is twice as high as the input of sediment by the river. A rough mud balance can be obtained by using the mud supply by the IJssel and the mud discharge of the sluices at Den Oever and Kornwerderzand (Table 11).

The freshwater basin Hollands Diep–Haringvliet

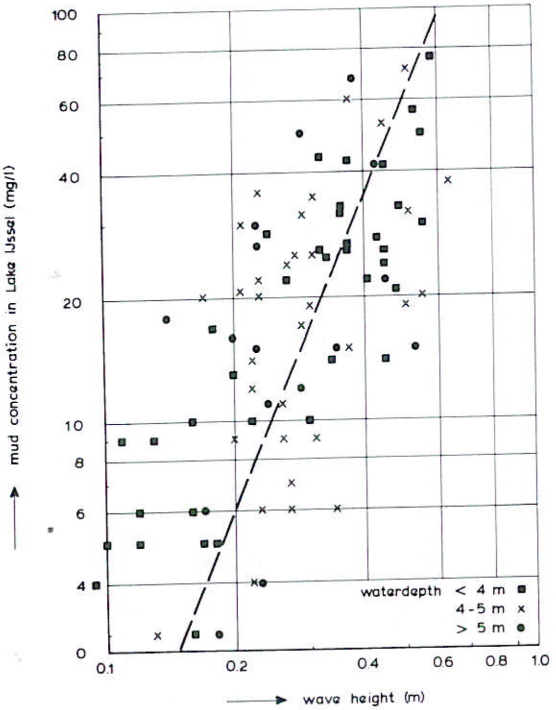


Fig. 13. Relationship between waves (wind) and the mud content of the water of the IJsselmeer.

was created by the closure of the Volkerak in 1969 and the Haringvliet in 1970. Since then the tidal motion, now penetrating mainly via the Rotterdam Waterway, Oude Maas, Spui and Dordtse Kil, has been strongly reduced. Basically, three phases can be distinguished in this new situation (Fig. 5).

(1) During Rhine discharges lower than 1730 m^3 sec^{-1} the discharge through the Harvingvliet sluices is negligible. The waters of the rivers Meuse and Rhine are discharged to the sea through the Dordtse Kil and Spui. The tidal range in the basin is only 0·1–0·2 m. The flow velocities in this situation are very weak, and wind-induced currents may dominate.

(2) With Rhine discharges increasing from 1730 m^3 sec^{-1} up to 6000 m^3 sec^{-1} the gate opening of the Haringvliet sluices gradually increases until its maximum. The discharges vary with the tide at sea, resulting in a tidal range in the basin which slowly increases from about 0·2 m at low Rhine discharges to several decimetres up to a maximum of roughly 0·5 m with increasing flow velocities.

(3) During Rhine discharges exceeding 6000 m^3 sec^{-1} the gates of the Haringvliet sluices are fully opened at low tide at sea. The tidal range and the motion of water then approaches the former situation. These conditions, however, occur on average only 6 days per year.

Thus the tidal range and the flow velocities in the basin are generally low. The basin is much deeper than the IJsselmeer which strongly reduces the effect of wind on the mud transports in this freshwater basin. The effect of the discharge of the Rhine is in this respect more important. It appears that in the new situation, after the closure of the Haringvliet in November 1970, mud is deposited in the Nieuwe Merwede and Amer near the Biesbosch during low Rhine discharges. During high Rhine discharges part of this may be eroded again and, with other mud, may be partly redeposited further downstream, in particular in the Hollands Diep (Haring, 1978; Ferguson *et al.*, 1976; Terwindt, 1977). An approximate mud budget for the Hollands Diep–Haringvliet basin is presented in Table 12 (see also Fig. 15), showing a siltation of about $1{\cdot}8 \times 10^6$ tons of mud per year.

Processes affecting trace metals in the Haringvliet are the same as in the IJsselmeer. However, the differences in the hydrodynamic conditions cause some notable variations in their extent. The residence time of the water is much shorter (weeks) compared

Table 9. Metal concentrations ($\mu g\ g^{-1}$) in the suspended matter from the freshwater basins. The numbers in parentheses refer to the localities shown in Fig. 4. Data are based on 3–4 weekly surveys conducted in the freshwater basins in 1977 and 1978

	Zn	Cu	Ni	Pb	Cd	Cr
IJsselmeer						
Ketelmeer (1)	2160	283	94	428	47·7	644
Enclosure dike (2)	751	62	56	158	5·6	184
Haringvliet						
Nieuw Merwede (3)	1743	294	88	489	44·5	765
Amer (4)	3152	204	88	537	40·3	315
Haringvlietdam (5)	1869	172	77	470	21·1	518

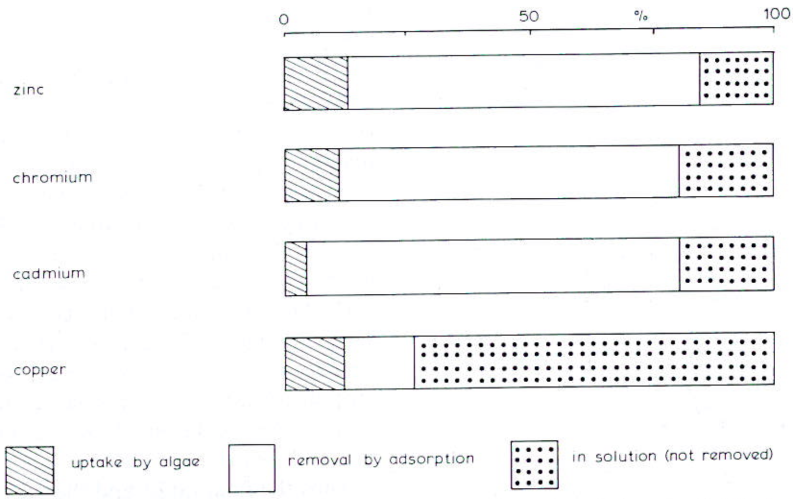


Fig. 14. Fate of dissolved trace metals in the IJsselmeer (based on Salomons & Mook, 1980).

Table 10. Dissolved metal concentrations ($\mu g\ l^{-1}$) in the freshwater basins. The numbers in parentheses refer to the localities shown in Fig. 4. Data are based on 3–4 weekly surveys conducted in the freshwater basins in 1977 and 1978

	Zn	Cu	Ni	Pb	Cd	Cr
IJsselmeer						
Ketelmeer (1)	40	5·5	6·0	1·5	0·8	2·3
Enclosure dike (2)	4·3	2·7	5·3	1·6	0·1	0·3
Haringvliet						
Nieuw Merwede (3)	70	4·6	6·1	1·3	1·6	2·7
Amer (4)	41	2·9	14	3·1	0·2	0·6
Haringvlietdam (5)	25	2·8	5·0	3·4	0·6	1·1

Table 11. Approximate silt budget of the IJsselmeer (exclusive of Markermeer) and the water discharge of the river IJssel and through the sluices in the Enclosure dyke

	Sediment ($\times 10^3$ ton yr^{-1})	Water* ($\times 10^6\ m^3$)
IJssel†	330	9600
Organic matter‡	160§	
Carbonates	440	
Sluices Enclosure dike¶		−12430**
Den Oever	−180	
Kornwerderzand	−105	

* Water discharge data based on Rijkswaterstaat and Rijksdienst IJsselmeerpolders (1976). Mean values over the period 1969–74.

† Based on mean river discharges according to plan S 300 and mean silt contents at Kampen (period 1972–8; 11 to 14 samples per quarter) (Rijkswaterstaat, 1972–8).

‡ Approximately 120,000 tons yr^{-1} according to Postma (1976b) during the period 1932–65.

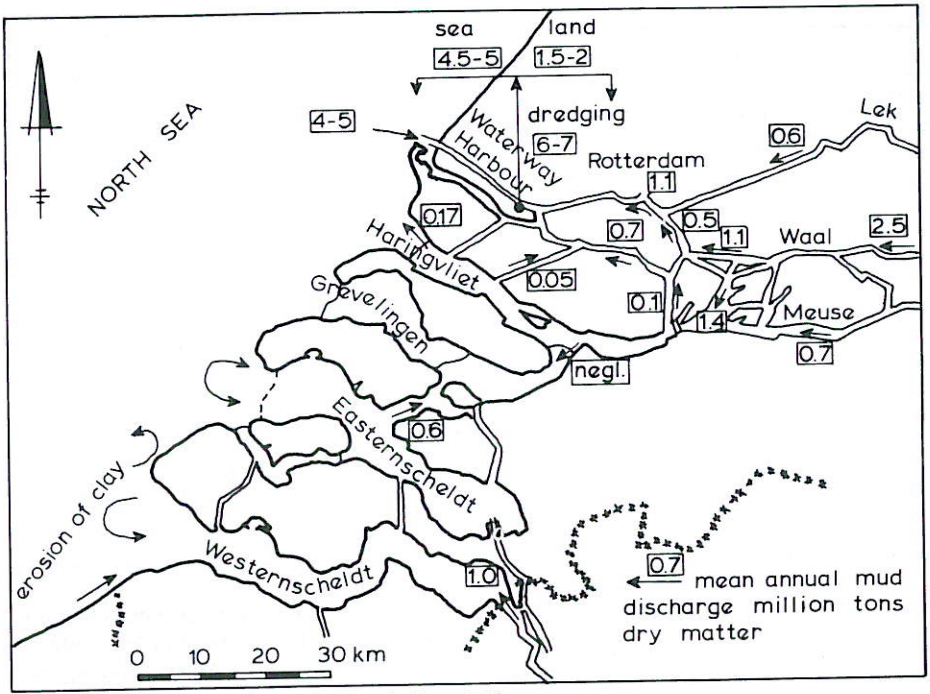
¶ Based on discharge data and silt content of the period 1973–7; three water samples per quarter (Rijkswaterstaat, 1972–8).

** Apart from the river IJssel, polders and some small streams contribute water to the lake.

with the IJsselmeer. No massive algal blooms occur and the pH reaches values of about 8 only in the western part of the basin, thus causing a smaller removal of dissolved trace metals (Table 10). As the depth of the Haringvliet is greater than that of the shallow IJsselmeer, the resuspension of (relatively unpolluted) bottom sediment, therefore, is less frequent, causing only small admixtures of bottom sediments to the suspended matter. Particulate metal concentrations, therefore, stay high in the Haringvliet, which acts mainly as a sink for suspended matter and its associated pollutants but less so for the dissolved trace metals.

TENTATIVE MUD AND METAL BALANCE FOR THE RIVERS RHINE AND MEUSE

The data for the rivers Rhine and Meuse, which account for a large part of the metals transported by the rivers in Belgium, the Netherlands and Germany



Based on Terwindt (1977) and Tables 1, 11 and 12

Fig. 15. Mean annual discharge of mud in the Dutch delta area. (Lek, Waal and Meuse data are based on Terwindt, 1977, all other data in Tables 6 and 12).

Table 12. Approximate silt budget of the Hollands Diep–Haringvliet basin

	Major mean silt discharges and productions ($\times 10^3$ tons yr^{-1})
Nieuwe Merwede	1400*
Meuse	700*
Dordtse Kil	−320†
Volkerak sluices	
Spui	
Haringvliet sluices	
Siltation	1780 10^3 tons yr^{-1}

* Terwindt (1967, 1977).

† Based on quarterly mean discharge and silt content data of the Rijkswaterstaat during the period 1973–7 (generally 3–7 water samples per location per quarter). These data did not allow a reliable estimate of the separate mean annual discharges as the silt discharges of the Dordtse Kil, the Spui and of the Haringvliet sluices distinctly depend on the discharges of the Rhine and during the period considered the discharges of the Meuse and the Rhine did not exceed the average discharges. However, the sum of these silt discharges, also taking into account the NLP 70 scheme (see Fig. 3), seems to be rather constant over a rather wide range of discharge conditions at an average of about 320,000 tons yr^{-1}. For average discharge conditions, this total silt discharge can be subdivided as follows: Dordtse Kil: 100,000 tons yr^{-1}, Volkerak sluices: 5000 tons yr^{-1}, Spui: 40,000 tons yr^{-1}, Haringvliet sluices: 175,000 tons yr^{-1}.

in the direction of the North Sea (Table 1), can be used to show the relative importance of the three pathways for the input of trace metals and mud into the southern North Sea.

Data on the amount of trace metals transported by the rivers Rhine and Meuse have been obtained from Rijkswaterstaat (1977–8). The weekly measurements of both dissolved and particulate trace metals were simply averaged, because insufficient data are available to estimate the influence of high water waves on metal transport. The amounts, in tons yr^{-1}, are presented in Table 13. The calculations on the input and accumulation of trace metals in the freshwater basins are based on data presented in Tables 9 and 10. The water discharge is estimated from the NLP '70 scheme (Fig. 5) and the mud discharge (particulate metal transport) are based on Tables 11 and 12 (see also Fig. 15). To facilitate the reading of the data, they are given as a percentage of the total metal transport by the rivers Rhine and Meuse.

Table 13. Preliminary metal balance for the Netherlands

	Cu	Ni	Zn	Pb	Cd	Cr	Me_t
Input (tons yr^{-1})							
Rhine	1138	835	8380	1548	173	2383	14,457
Meuse	77	51	1671	159	22	80	2060
Total	1215	886	10,051	1707	195	2463	16,517
Accumulation (in percentage of input by Rhine and Meuse)							
Haringvliet basin	43	27	49	53	51	46	48
IJsselmeer	7	1	8	5	9	7	7
Landfill	18	7	13	16	13	19	14
Total	68	35	70	74	73	72	69
Output							
Haringvliet basin	7	20	8	4	8	5	8
IJsselmeer	4	10	3	4	2	2	3
Loswal Noord	6	3	5	7	4	8	5
Total	17	33	16	15	14	15	16
Not accounted for (Any other direct transport by the estuary to the North Sea)	15	32	14	11	13	13	15

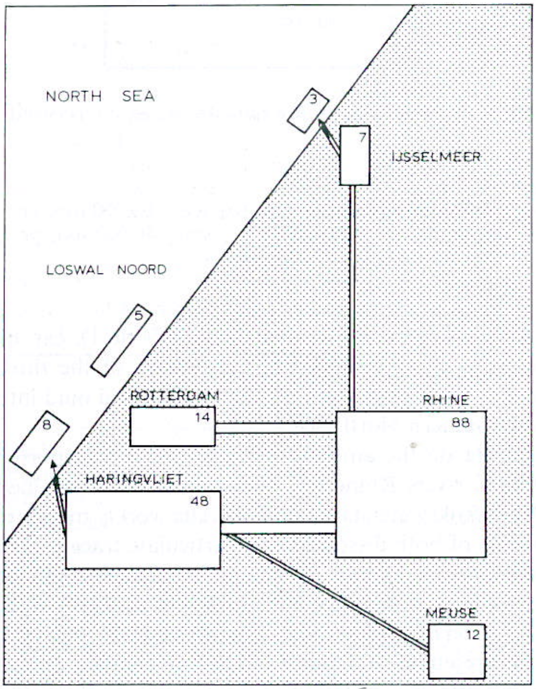


Fig. 16. Metal balance for the rivers Rhine and Meuse. Transport of metals (Cu + Cd + Cr + Pb + Ni + Zn) by the rivers Rhine and Meuse is 100 %. The major sinks and the transport to the southern North Sea are shown. Insufficient data were available on the direct transport of trace metals through the Rhine estuary to the North Sea; it is probably the major part of the 15 % not accounted for.

Concentrations of trace metals in dredged material from the Rotterdam harbour were obtained during a survey in September 1977. Data on the amount of dredged material used for landfill and transported to the North Sea have been obtained from the municipality of Rotterdam and from the Rijkswaterstaat. These data, together with the approximate ratios of marine to fluvial sediments in dredged material (see 'Harbour of Rotterdam; case study') were used to calculate the amount of trace metals of fluvial origin which were transported to the landfill areas or to the North Sea. Insufficient data were available on the direct transport of trace metals through the Rhine estuary to the North Sea, so this part of the metal balance could not be directly calculated; it is probably the major part of 'not accounted for' in Table 13.

The trace metals (with the exception of nickel) behave more or less similarly. Therefore, it is possible to make a schematic diagram showing the major sinks of trace metals from the rivers Rhine and Meuse and the output to the North Sea by using the total metal transport quantities (Fig. 16). Of the total amount of trace metals transported by the rivers Rhine and Meuse, 48 % accumulates in the Haringvliet. For the individual metals (with the exception of nickel), this amount varies between 43 and 53 %. More than two-thirds of the trace metals entering the Netherlands through the rivers Rhine and Meuse accumulate in the freshwater basins or end up in the landfill areas (dredged material), with less than one-third entering the North Sea. The freshwater basins account for 11 % of the input into the North Sea and the dredging activities for 5 %. Not accounted for, which may be partly the direct transport through the estuary to the North Sea, is about 15 %.

CONCLUSIONS

The two additional pathways (dredging and freshwater basins) determine to a large extent the amount of trace metals entering the North Sea through the rivers Rhine and Meuse. Together they account for about 70 % of the metal load transported by the rivers in Belgium, the Netherlands and Germany to the North Sea.

The freshwater basins act as sinks for particulate and for dissolved trace metals. Relatively small amounts of suspended matter escape in the freshwater basins and enter the North Sea. Due to the mixing of bottom sediments (relatively uncontaminated) with fluvial (highly contaminated) suspended matter in the freshwater basins, the metal concentrations in the suspended matter entering the North Sea are lower compared with the river. This process is, due to its shallowness, important in the IJsselmeer but less so in the Haringvliet. The pH increase in the freshwater basins, due to algal growth and CO_2 exchange with the atmosphere, is responsible for the adsorption of trace metals on the suspended matter. Also the uptake of metals by algae causes a removal of dissolved trace metals from solution. The long residence time of the water in the IJsselmeer and the massive algal blooms in this lake are responsible for the fact that the removal of dissolved metals in the IJsselmeer is greater when compared with the Haringvliet.

The creation of artificial deep navigation channels and the construction of harbour basins cause an increased settling of suspended matter. In the case of the Rotterdam harbour the supply of mud originates mainly from the North Sea. But as part of the dredged material is used for landfill, both fluvial and marine mud and their associated trace metals are removed from the aquatic environment. The major part of the dredged material is transported to the North Sea, and the dumping sites act as important point sources for sediments in the North Sea. The total quantity of sediments dredged along the southern North Sea coast is twice the mud budget of the North Sea.

The result of the two additional pathways with regard to pollutants entering the North Sea has been that two-thirds of the trace-metal load of the rivers Rhine and Meuse accumulate in the Netherlands. This situation may be regarded as beneficial for the North Sea, but poses problems for the management of the inland waters and the landfill areas in the Netherlands.

ACKNOWLEDGMENTS

Part of this work has been made possible by several investigations carried out on behalf of the Rijkswaterstaat and the municipality of Rotterdam. In addition these two organizations provided us with unpublished data. The investigations carried out in the IJsselmeer were partly supported by a grant from the EEC (contract No. 199-77-1 ENV N).

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