

Reducing river pollution increases sediment quality

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Summary

Pollutants affecting the coastal zone mainly come from point and diffuse land-based sources. They are largely transported to the sea by rivers and adhere to clay and silt particles in the coastal zone. In the past these have had damaging effects on the environment. In the last few decades national and international efforts to reduce the levels of riverine pollutants have been successful. Subsequently, the concentrations in coastal sediments are lowered. The chemical compounds in the sediments of the Wadden Sea followed the same trend then those off the Dutch North Sea coast. However, a time lag between the cleaning of the rivers and the decrease of pollutants in the top layers of silty sediments of Wadden Sea, is observed. This is mainly caused by storms and bioturbation.

Spectacular improvement in the quality of the sediments of the North Sea and Wadden Sea, shows that meticulous cleaning of the sources of pollution, monitoring, international negotiations and cooperation benefit all the riparian countries.

The catchment areas of the rivers Rhine and Meuse - six riparian countries: Switzerland, France, Germany, Luxembourg, Belgium, the Netherlands, and the North Sea. (source: "Room for the River", Rijkswaterstaat)



1. Introduction

The coastal zones of the world are influenced by multiple pressures. Runoff of chemical compounds from land-based sources plays an important role in the degradation of coastal and marine ecosystems. This is especially true for the Netherlands where the four international rivers Rhine, Meuse, Scheldt and Ems flow into the North Sea. The circulation of the riverine water and the suspended particulate matter in the Dutch coastal zone is reflected by the salinity distribution, which is influenced by a generally anti-clockwise circulation pattern in the southern North Sea (Otto et al., 1990). The net transport of Rhine, Meuse and Scheldt waters is northwards along the continental coast causing a strong salinity gradient perpendicular to the coast (Figure 1).

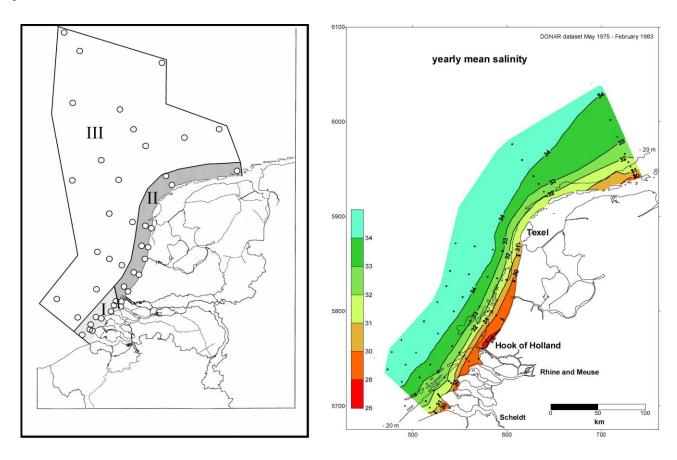


Figure 1a (left): Sampling location map of the Dutch coastal zone with three areas: Delta (I), Dutch coast (II), central North Sea (III). o = Sampling locations of the Rijkswaterstaat North Sea monitoring program;

Figure 1b (right): The annual average salinity distribution in the period 1975-1983. (source: Suylen and Duin, 2001, 2002)

2. Sources of pollution

For the Dutch coastal zone, in addition to riverine inputs, various other sources of chemical compounds can be distinguished (Wulffraat et al., 1993). Dissolved and particulate chemical compounds reach the area with the Atlantic waters flowing in via the Belgian coast, and from the United Kingdom. Offshore activities such as the oil and gas industry and shipping are another source. Atmospheric chemical compounds also affect the Dutch coastal zone (Bleeker et al., 2004).

Annual dredging of the waterways of the Rotterdam harbour reached a maximum in the beginning of the 1980s, due to deepening of the main navigation channel (Laane et al., 2006). The average annual amount of dredged material ranges from 15 – 20 million m³. The largest part of this material is clean and dumped offshore. Most of the dumped material moves northwards to the Wadden Sea, about a third remains on the marine dumping site and a smaller part returns to Rotterdam harbour (Sandeh, 2002, see Figure 2).

The most polluted dredged material, originating from the more upstream part of the Rotterdam waterways is stored on land since 1983 (Stronkhorst, 2003), amongst others in the 'Slufter' (see CCC I-2-2).

Because the rivers are the primary source of pollutants, efforts to reduce coastal contaminants focus on river basin management.

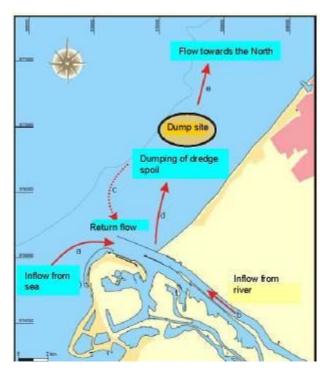


Figure 2: Map of the mouth and the outflow of the rivers Rhine and Meuse into the Dutch coastal waters. Indicated are the dumping site and arrows showing the flows of suspended particulate matter including the return flow to Rotterdam harbour. (source: Sandeh, 2002)

3. Reduction of the polluting load

There are a large number, more than 100,000 synthetic chemical compounds produced, many ending up in rivers and oceans (Laane, 2001). In the Netherlands, the riverine sources were significantly reduced during the 1980s (Figure 3: examples of the metal cadmium and organic contaminant polychlorinated bifenyls - PCBs). Most of this was directly from load reduction in river sediments and indirectly from dumped dredged material. The contribution from the Atlantic Ocean is more difficult to reduce and becomes relatively more important over time. However, the concentrations of these chemical compounds are relatively low and close to natural background levels.

PCBs also show a reduction, caused mainly by a large cutback in the river load. The contribution from the other three sources of contamination remains more or less the same. It is remarkable that the relatively large load reductions, factor 2-4, were realised in only a decade.

The results are based on regular monitoring, which began in the 1970s. Quality Controlled analysis of metals was possible from the 1980s, and some years later for persistent organic compounds like PCBs and PAHs (polycyclic aromatic hydrocarbons). It is, in principle, possible to analyze every chemical compound. However, this is very expensive and not cost-effective. Monitoring of relevant indicators, data management and exchange and reporting are essential tools and are undertaken or guided by the responsible Netherlands Ministry Infrastructure and the Environment (former Ministry of Transport, Public Works and Water Management) on a national and European level.

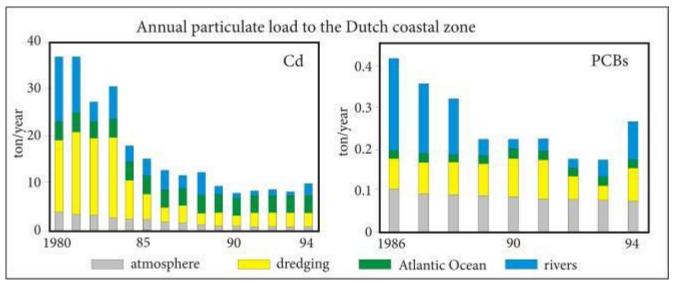


Figure 3: **Annual particulate load of cadmium (Cd) and PCBs** (polychlorinated biphenyls) from various sources to the Dutch coastal zone during the years 1980-1994. (source: Sonneveldt and Laane, 2000)

4. Dual track reduction policy: national and international

River basin management at national and international level has been successful in contributing to the clean-up of rivers. This has had a positive influence on the level of contamination in the lower lying deltaic states and its coastal sediments.

In the 1970s, the effect of enhanced concentrations of chemical compounds in European rivers (e.g. the Rhine) and coastal zones were visible (Laane et al., 2006). The oxygen concentration in the rivers was relatively low due to untreated wastewater discharges. The increase in concentrations of persistent organic compounds and their negative effects on for instance the reproduction of seals in the international Wadden Sea was the motive for political action.

The struggle to reduce contamination involved a dual track policy:

- National: cleaning the Netherlands' own sources of pollution (emission reduction) and confining the most polluted dredged material in man-made basins (e.g. the Slufter, see CCC I-2-2), and
- International: efforts to reduce the upstream emissions through long term, ministerial Rhine, Meuse, Scheldt-river basins negotiations.

This dual track, integrated policy approach had positive effects in two areas by reducing:

- The contaminant load by rivers, dredging and atmospheric deposition and
- The pollution in the North Sea and Wadden Sea.

The lowering riverine contaminant load was established in the Netherlands at first through stepwise gentlemen-agreements between government and industry, and later by legislative enforcements. Internationally integrated river basin management was established for the river Rhine in the 1950s by the five riparian Rhine states: Switzerland, Germany, France, Luxembourg and the Netherlands. These countries on the initiative of the Netherlands, founded the International Commission for the Protection of the Rhine (ICPR). The main ICPR objectives and tasks are to:

- Improve the chemical and ecological state of the Rhine and taking into account the state of the North Sea;
- Develop a holistic flood prevention and protection approach, which includes ecological requirements;
- Support the coordinated implementation of European regulations, such as the Water Framework Directive and the Flood Directive in the watershed of the Rhine.

Important landmarks in the history of the ICPR are the 1976 signature of the Convention on the Protection of the Rhine against Chemical Pollution, followed by the 1987 Ministers' approval of the implementation of the Rhine Action Programme (RAP), see website ICPR.

Similar integrated river basin measures were carried out for the Meuse and Scheldt catchments.

In 2000, the EU Water Framework Directive came into force. A uniform Europe-wide policy was established, with the aim of maintaining and restoring the ecological status of the waters in Europe through integrated river basin management. This Directive covers the ecological status of all inland waters and coastal waters up to one mile offshore; for chemicals the limit is twelve miles from the coast (see website: EU Water Framework Directive).

A series of Ministerial Conferences on the Protection of the North Sea focused on reducing the loads of chemical compounds. The first conference took place in 1984 and the most recent one (Bergen, Norway, 2010) within the framework of the OSPAR Commission (see website OSPAR Com). OSPAR is the mechanism by which fifteen Governments of the western coasts and catchments of Europe, together with the European Community, cooperate to protect the marine environment of the North-East Atlantic. One of the important milestones was reached in 1987, when the countries meeting at the Second Ministerial Conference agreed to reduce the inputs of potentially dangerous substances to the North Sea by approximately 50% over the period 1985 – 1995. This Conference also agreed to eliminate the incineration of dangerous wastes and disposal of sewage sludge at sea. These were important achievements, because before then, each country had set its own objectives (Laane et al., 2005).

5. Improved coastal sediment quality

As a consequence of the lower chemical loads in the rivers, over time the concentration of pollutants adsorbed to the fine grained near-shore marine and coastal sediments is reduced. Cd, PCBs and PAHs are representative for the 150 contaminants, which are regularly monitored in air, rivers and coastal waters.

The Dutch North Sea coast

The concentration of Cd (Figures: 4a and b): the highest concentrations are found in the coastal zone, with decreasing concentrations offshore and to the north.

Time series graphs show that the concentrations have decreased over the past 25-30 years (Figures: 4b and 5b). The concentrations of cadmium in the three different parts of the Dutch coastal zone are, from 1990 onwards, close to or below the national eco-toxicological target (negligible risk concentration) of 0.8 mg/kg.

A similar picture of decreasing concentration is presented for PAHs (the six of Borneff); the monitoring started in 1986 (Figures: 5a and b). Only at a few locations in the Dutch coastal zone the PAH concentrations are above the Dutch maximum permissible risk concentration of 600 µg/kg.

Similar reductions are observed for most other analysed contaminants and the level of most contaminants is below the EU-target values and national toxicological objectives, with exception at some sites for the group of TBT (Tributyl tin compounds), PAHs, Zinc, Mercury, mineral oil and HCBs (hexachlorobenzene) (Hegeman and Laane, 2008).

This relatively rapid and large decrease in offshore contaminants appears to be related to mixing and resuspension of the fine sediment fraction during storm conditions. Over a period of about 2 years, the heavily contaminated silt particles ($<63 \mu m$) in the surface sediments are replaced by less contaminated riverine particulate matter (Sonneveldt and Laane, 2000).

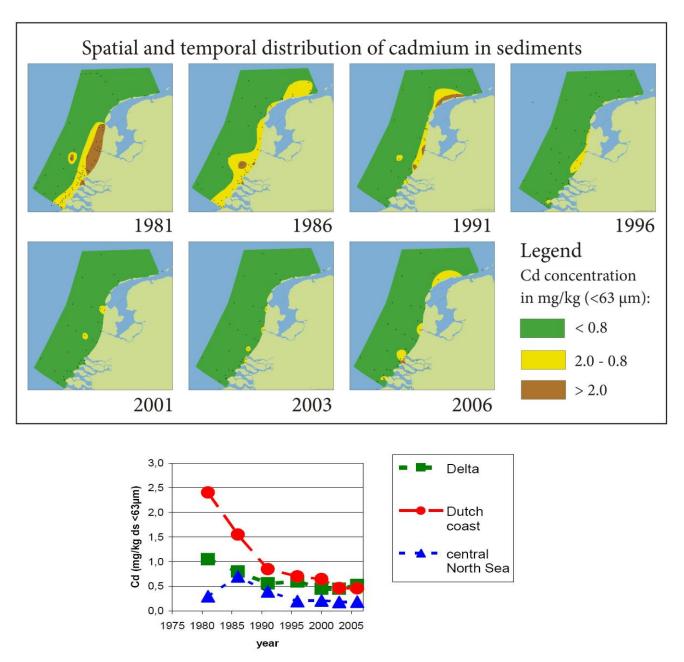


Figure 4: The Dutch North Sea coastal zone: Geographical distribution (4a) and long-term trend: 1981 – 2006 (4b) of the concentration of cadmium (mgCd/kg) in the surface sediments (grain size fraction <63µm) in three areas in the Dutch coastal zone. (source: Hegeman and Laane, 2008).

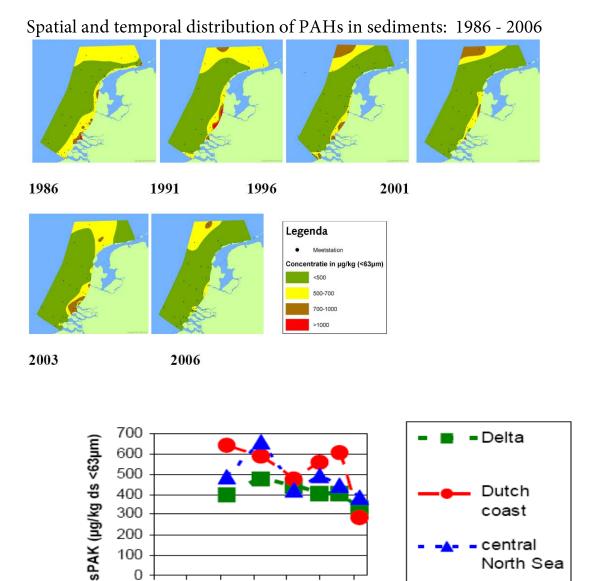


Figure 5: The Dutch North Sea coastal zone: Geographical distribution (a) and long-term trend: 1986 - 2006 (b) of the concentration of PAHs (the six polycyclic aromatic hydrocarbons = six of Borneff - µgr/kg) in the surface sediments (grain size fraction <63µm) in three areas in the Dutch coastal zone. (source: Hegeman and Laane, 2008)

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The Wadden Sea

The Wadden Sea is a tidal flat area in the northern part of the Netherlands (see Figure 6). The area is influenced by inputs from the river Rhine/IJssel, either directly though sluices in the closure dam that separates the Wadden Sea from Lake IJsselmeer, or indirectly from the rivers Rhine and Meuse, that enter the North Sea at Hoek van Holland and – mixed with

the coastal seawater - travel north along the Dutch coast. The Wadden Sea is a sediment sink for silty, fine-grained sediments. As a result of riverine inputs there is a gradient in contaminant concentrations from west to east. Because of the relatively large surface area and the organic coating on particles, contaminants are mostly contained in the fine fraction of the sediment ($<63 \mu m$).

The history of contamination in the Wadden Sea comes from a series of tidal flat sediment cores extracted from the western Wadden Sea (Kramer et al., 1989; Kramer et al., 1991). The results for some trace elements of the Mokbaai sampling location (red dot Figure 6), a sheltered intertidal area, are shown in Figure 7.

Figure 6: **Western part of the Dutch Wadden Sea** showing the islands, tidal inlets, tidal channels and tidal flats. The red dot identifies the sampling location = Mokbaai. (photo: NASA)



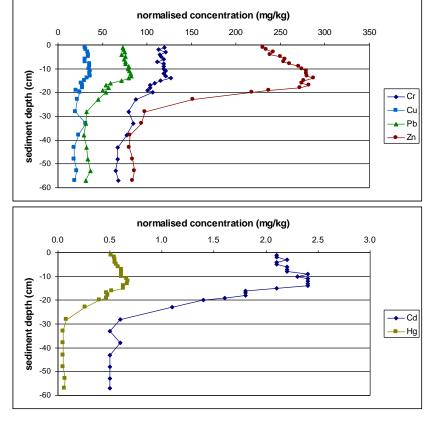


Figure 7: **Vertical profiles** of chromium (Cr), copper (Cu), lead (Pb) and zinc (Zn) and cadmium (Cd) and mercury (Hg), in sediments of the Mokbaai core (SRS normalised). (source: Kramer et al., 1991)

All profiles showed uniform background concentrations below the 30 cm sediment depth. Then a clear increase in concentration was observed reaching a maximum at 14 cm sediment depth, the result of anthropogenic inputs. The top layers showed a more uniform distribution, with a slight downward trend for most elements. Furthermore, the results of the radiotracer studies provided congruent sedimentation rates: $^{210}\text{Pb} \cong 6.5 \text{ mm/yr}$, $^{137}\text{Cs} \cong 6 \text{ mm/yr}$, meaning that the contaminant concentrations peaked at about 1963–1965.

Interpreting these results proved difficult. Looking at pollution history of the rivers Rhine and Meuse, the maximum pollutant load occurred towards the beginning of the 1980s.

Moreover, the ¹³⁴Cs distribution in the sediment (Figure 8) showed a clear presence up to 13 cm sediment depth and a nearly homogeneous distribution in the top 7 cm. ¹³⁴Cs is short-lived (half-life = 2.06 yr) and all ¹³⁴Cs found must be from the Chernobyl accident in April 1986, less than 2 years before the sampling took place. With the calculated average sedimentation rate the radiotracer would have been present only in the very top layers. So why did it appear lower down? It appeared that the surface sediments are very dynamic. Beside the effects of storms on the clearing of surface sediments, bioturbation will play an important role. The Wadden Sea is very rich in benthic organisms, including burrowing worms and clams. (see photo: Figure 9: Benthic activities on the tidal flats of the Wadden Sea).

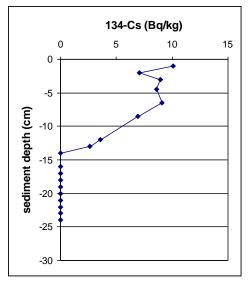


Figure 8: **Vertical profile** of ¹³⁴Cs in the Mokbaai core. (source: Kramer et al., 1991)

Using a simple mathematical model to simulate the biological/physical mixing with three bioturbation layers (WADSEDI, Wadden Sea Sediment, Van Veen et al., 1989) it was possible to demonstrate that the distribution of chemical compounds (fixed to sediment particles) are more or less homogeneously spread in the top layers of the sediment. The depth is determined by the borrowing depth of the organisms present. Reliable sediment pollution history is attainable from sediment layers below the reach of burrowing organisms, in this case 14 cm (Kramer et al., 1991). Thus, although the results from the top layers should be evaluated with caution, a reduction in contamination of the Wadden Sea sediments is slowly taking place, due to mixing with the more contaminated sub-layers.

Sediment cores of the other Wadden locations showed similar vertical distribution of contaminants including clear signals of bioturbation and its bio-mixing effects (Kramer et al., 1989). Other, physical processes, such as the mixing of the surface sediments due to wave action as described for the North Sea coastal zone, may also have played a role in the more exposed Wadden locations.

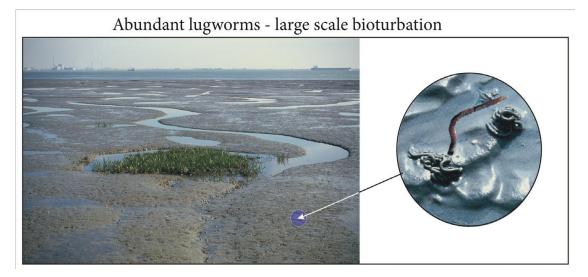


Figure 9: **Abundant lugworms** (Arenicola marina) in the freshly accumulated sediments of the Wadden Sea intertidal flats, indicating large scale bioturbation activities, which also contribute to the retardation of the "cleaning-up" process. (photo: //beeldbank.rws.nl, Rijkswaterstaat)

Biological effects of pollution and bioassays

Laane et al. (2006) summarised the observed biological effects of chemical compounds in the Dutch coastal zone and estuaries. The first documented observations began 40 years ago, with lethal effects on organisms, such as the harbour seal (*Phoca vitulina*) population in the Wadden Sea. The seal population showed a sharp decline, which was correlated with increased PCB concentrations (Reijnders, 1980; 1982). In the last decade the PCB load to the western Wadden Sea has dropped to such a level, that it no longer negatively affects the seal reproduction.



(photo: Harry van Reeken)

(photo: Dick Vethaak)

Figure 10: Serious warning during the 1980s Cleaning-up was needed:

Foam of massive dying algae along the Dutch coast, related to high level of nitrogen and phosphate in Dutch coastal waters. **Liver cancer** in the North Sea flounder (Platichthys flesus L.).

Vethaak (1985) was the first to observe that 40% of dab (*Limanda limanda*) and flounders (*Platichthys flesus*) aged over 3 years, were affected with liver carcinogen nodules, lymphocystis nodules and pseudo-tumours in skin tissue. (see Dick Vethaak 1996 – Figure 10). In large mesocosm experiments with dredged material from the Rotterdam harbours he correlated the occurrence of these nodules with the presence of PAHs in the surface sediments (Vethaak, 1993). Since 1996 the concentration of the PAHs has dropped (Figure 5b) to 400 μg.kg⁻¹ PAHs and the occurrence of liver nodules is now at a natural level. Although the concentration of PAHs in surface sediments can reach twice the target value (Hegeman and Laane, 2008) strong adsorption onto the silt/clay fraction may cause a smaller bioavailability than previously expected (Jonker, 2004).

Alternatives to monitoring the 100,000 chemical compounds have been introduced by integrated pollution monitoring techniques. Examples are: fish as water quality monitor in laboratories, mussel surveillance in the river Rhine and bioassays with benthic (bottom dwelling) organisms in contaminated dredged Rotterdam harbour material (Stronkhorst, 2003). In



Figure 11: **The whelk** (Buccinum undatum), a member of the North Sea marine ecological web, threatened with extinction by anti-fouling agent from boats through imposex.

(thata. @ Hans Hillowaert / CC-RY-SA-30)

bioassays, various organisms are used to determine effects of chemical compounds. No significant temporal or spatial differences were observed in the surface sediments from the harbour basins and the coastal zone. Although at certain locations the concentrations in the surface sediments are above the national target, no toxicological effects could be measured (Stronkhorst, 2003).

The one exception is TBT (tributyltin), which is an anti-fouling agent for ships' hulls. The concentrations of TBT are still above target level (Van Gils and Friocourt, 2008). TBT affects the sex organs of snails (imposex), such as the whelk (figure 11). It can cause the extinction of entire populations, threatening the common whelk (*Buccinum undatum*) at various locations along the Dutch coast in the North Sea.

6. Conclusions

Cleaning the rivers is feasible in a relative short period of time (several decades) if riparian countries cooperate at a river basin level. The Ministerial Conferences are supported by research efforts focusing on regular monitoring, standards and target setting, data management and exchange and reporting. At the same time sources of pollution in the Netherlands were addressed, first through gentlemen agreements then by legislation and enforcement.

The large-scale reduction of contamination in rivers is reflected in the reduction of the level of contamination in the coastal zone. The relative long time lag between the cleaning of the rivers and the reduction of pollution in the accumulating Wadden Sea sediments is caused by mixing of sediments due to bioturbation and storms.

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