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P. Kelderman [1], Yang Xuedong[1] and W. M. E. Drossaert[2]

Sediment pollution with respect to heavy metals and organic micropollutants in the city canals of Delft (The Netherlands)

– Assessment of a data base of 188 sediment stations

ABSTRACT

This paper gives an overview of a long-year study on the quality characteristics of dredged sediments in the city of Delft (The Netherlands). An inventory at 188 stations showed that especially the inner city canals and an industrial area have been polluted strongly with heavy metals and organic micropollutants. For the inner city sediments, much of the pollution can be attributed to transfer from the river Rhine. The industrial area, on the other hand, shows a legacy of past industrial activities related to metal processing. Finally, the more remote city areas are characterised by near-background concentrations with respect to sediment heavy metals and organic micro-pollutants contents.

INTRODUCTION

The city of Delft (The Netherlands; see Fig. 1) has many problems in the management of polluted dredged materials from its waterways [1]. In the Dutch system of sediment quality classes (see Table 1; cf. [2]), dredged sediments of classes 1-2 generally may be disposed of into the environment without many restrictions. Class 3-4 sediments, on the other hand, are of unacceptable quality and need (for class 4: with highest urgency) sanitation, *viz.* dredging, disposal in special storage reservoirs and, if possible, sediment clean-up measures. This will increase the processing cost by a factor of at least 2. Earlier papers [1, 3] have made a general inventory of the sediment pollution sources and possible abatement measures in Delft. It was shown that especially the inner city canal sediments have been strongly polluted with heavy metals and organic micro-pollutants. Water turbulence caused by shipping traffic plays a dominant role in the supply of these sediments [4].

This paper presents a detailed overview of the sediment quality characteristics in Delft, in the form of a data base comprising 188 stations and, in total, 75 sediment parameters (for details, see [5]). Statistical evaluation of the data base parameters is used to attain a better insight into the various pollution sources for, on the one hand, the relatively polluted sites in the inner city and an industrial area and, on the other, the relatively unpolluted outer city districts.

In total 188 sediment stations were monitored between the years 1991 and 2000 (see Fig. 1 [5]); 54 of the stations were monitored 2-3 times, usually in different years. Of the 188 sta-

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[1] *UNESCO-IHE Institute for Water Education, PO Box 3015, 2601 DA Delft, The Netherland. E mail first author: kld@ihe.nl*

[2] *De Straat Milieuadviseurs, PO Box 270, 2600 AG Delft, The Netherlands.*

tions, 21 stations were located within the inner city (see Fig. 1). The sediment samples, between 20 and 50 cm thickness, were all analysed by a certified laboratory (for details, see [3]). Statistical analysis involved mutual correlations between the parameters (parametric or non-parametric, dependent on the "normality" of the data set involved) as well as statistical comparison with possible pollution sources such as from the river Rhine. A more in-depth statistical evaluation, involving Factor and Cluster Analysis, has been presented elsewhere [6].

Table 1. Definitions of pollution classes (for the year 1997)* with respect to the contents of the heavy metals chromium, nickel, copper, zinc, cadmium, mercury, lead, of arsenic and of organic micro-pollutants for polluted sediments in The Netherlands. Class limits are given with respect to a "standard sediment" with 10% organic matter and 25% clay (< 2μm) contents; corrections are necessary for "non-standard sediments" [cf. 2].

Parameter (all as mg/kg, except Σ DDT and Σ PAH)	**Class 1**	**Class 2**	**Class 3**	**Class 4**
Cr	< 380			≥ 380
Ni	< 35	35 - < 45	45 - <210	≥ 210
Cu	< 35	35 - < 90	90 - < 190	≥ 190
Zn	< 480	480 - < 720		≥ 720
Cd	< 2	2 - < 7.5	7.5 - <12	≥ 12
Hg	< 0.5	0.5 - < 1.5	1.5 - < 10	≥ 10
Pb	< 530			≥ 530
As	< 55			≥ 55
Σ DDT	< 10	10 – 20	21 – 4000	> 4000
Σ PCB	< 100	100 – 200	201 – 1000	> 1000
Σ PAH	<1	1 - < 10	10 - < 40	≥ 40
Mineral oil	< 1000	1000 - < 3000	3000 - < 5000	≥ 5000

Σ DDT (as μg/kg) expresses the sum of all DDT, DDD and DDE congeners.
Σ PCB (as μg/kg) expresses the sum of the polychlorinated biphenyls: PCB-28, 52, 101, 118, 138, 153 and 180.
Σ PAH is the sum of the polycyclic aromatic hydrocarbons: fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(ghi)perylene, indeno(1,2,3-cd)pyrene, naphthalene, chrysene, phenanthrene, anthracene and benzo(a)anthracene.
* For reasons of consistency with earlier literature (see *e.g.* [1,3]), the 1997 class limits will be used here; somewhat different class definition are in use in The Netherlands today.

DESCRIPTION OF THE AREA

Delft is a city situated in the SW Netherlands, close to the mouth of the river Rhine (see Fig. 1). It has a population of *ca.* 95,000, of which some 13,000 are permanent residents of the historical Delft inner city.

The Delft inner city canal system has a total length of 6.5 km with an average water depth of 1.1 m. This canal system is surrounded by the Rijn-Schie canal (in direct connection with the river Rhine), with which it has five open connections. To refresh the stagnant canal waters, Rijn-Schie canal water is pumped continuously into and through the inner city canal system, and is finally pumped out into the Rijn-Schie canal again, via a pumping station in the North (see Fig. 1).

In and around Delft, industrial as well as agricultural activities take place. The main industrial activities within Delft have been concentrated in a special industrial area close to the Rijn-Schie canal. Close to Delft lies the largest area (*viz.* 100 km^2) of greenhouses in The Netherlands, with extensive cultivation of vegetables and flowers on the, dominantly, clay-loamy soils of the region.

With respect to sewage management in the region, at present more than 99% of the point discharges (domestic and industrial) have been connected to the sewer system and are being treated in sewage treatment plants; 90% of the latter discharge their effluents outside the Delft area.

Fig.1. The city of Delft in The Netherlands with the 188 sediment monitoring stations. Areas A, B, C and D represent the inner city (PS = pumping station), an industrial area, and a residential and recreational area, respectively. (N.B. Numbering of the monitoring stations is consistent with that in the original data base [5]).

PRIORITY POLLUTANTS IN THE SEDIMENTS OF THE DELFT CANALS

As mentioned before, 54 out of the 188 sediment stations were sampled 2-3 times over the period 1991-2000. Using the Wilcoxon test [7], no time trends for the sediment pollutants contents were found at these sites. This observation seems logical because the vertical sediment cores will probably have had different thicknesses (between 20 and 50 cm) for the different sampling events, and may also originate from slightly different positions. Therefore it was decided, in these cases, to take the average values over the sampling dates. However, for the evaluation at the four different regions A-D (see later), the multiple monitoring results were considered as individual, separate data sets.

Fig. 2 presents the sediment contents for four heavy metals and four organic micropollutants. The data have been subdivided according to the class 1 - 4 limits mentioned in Table 1. Since no corrections have been applied for “non-standard sediment” conditions, the classes have been indicated as *e.g.* “class 4_{ns}”.

Heavy metals

- Copper (range: 3.5 - 950 mg/kg); see Fig. 2A. Cu is the most contributing contaminant to the sediment pollution in Delft. In total 54 out of 188 stations showed values ≥ 90 mg/kg of which 21 (100%) were located in the inner city. Even sediment contents ≥190 mg/kg (class 4ns) were frequently found, viz. in 24 stations, 11 of which in the inner city. Pollution hotspots were mainly clustered in or around the inner city, in the industrial area and in close proximity to the Rijn-Schie canal (see Fig. 2A). Intermediate Cu contents between 35 and 90 mg/kg can be observed in large parts of the Delft outer city. Lowest values are found in the south-east residential area of Delft (C), as well as in the north-easterly recreational area (D). The two apparent hotspots for copper (and other) pollution, viz. the Delft inner city and the industrial area, will be discussed later.
- **Zinc** (range: 20 - 4000 mg/kg); see Fig. 2B. In total 25 out of the 188 stations were found to be in class 4_{ns}, with Zn contents ≥ 720 mg/kg; of these 11 were located in the inner city. Just as for copper, the lowest values were observed in the SW residential and NE recreational areas.
- **Mercury** (range: 0.04 - 6.4 mg/kg); see Fig. 2C. In total 15 stations, of which 9 in the inner city, showed values exceeding the class 3_{ns} limits (≥ 1.5 mg/kg). Lowest Hg contents can be observed in virtually the whole eastern outer city, and in the NE recreational area.
- **Lead** (range: 0.3 - 1630 mg/kg); see Fig. 2 D. Overall, only 3 stations, all in the inner city, showed Pb contents > 530 mg/kg (class $4_{ns.}$). No elevated Pb levels were found near highways and busy roads. Lead has been used extensively as an anti-knocking agent in car petrol, but since the gradual ban on its use, overall Pb pollution levels have decreased sharply. Lowest Pb sediment contents can be found in the same areas as above.

For the remainder of the heavy metals*, i.e.* chromium, nickel, cadmium, as well as for arsenic, the limits for classes $(3+4)_{ns}$ were virtually never exceeded (see [5]).

Organic micropollutants

- **PAHs** (range: 0.1 - 330 mg/kg); see Fig. 2E. After copper, the polycyclic aromatic hydrocarbons were found to be the main contributors to the sediment pollution in Delft. Of the 188 stations, 31 had contents ≥ 10 mg PAHs/kg (class 3_{ns} and 4_{ns}); 13 of these were located in the inner city. The industrial site B shows moderate pollution with respect to PAHs, with 6 stations in class $3+4_{ns}$. It could be expected that elevated PAHs contents (related with automobile exhaust gases) would be present near highways and busy roads; however such a trend was not observed. This can probably be ascribed to the ever increasing use of catalysts in cars which can bring down these PAHs emissions drastically. Again, lowest PAHs values were found in the residential and recreational areas C and D.
- **DDT** (range: 1.2 - 450 µg/kg); see Fig. 2F. As for most other components, the inner city sediments showed highest pollution levels, with 13 stations exceeding the class 3_{ns} limits of 20 µg/kg. In the outer city, 18 additional stations belonged to this quality class. There does not seem to be a clear connection with the presence of greenhouses in NW Delft (see Fig. 1). Up to the ban in the late 1970s, DDT had been used extensively as an agricultural pesticide in Europe. The main source of the present DDT load is probably wind-driven, from *e.g.* tropical countries where DDT is still widely being applied.
- **Mineral oil** (range: 0 - 12,000 mg/kg); see Fig. 2G. In contrast to most other observations, the inner city sediments did not show elevated pollution levels. Here the main hotspot was located in the industrial zone B (6 stations with contents ≥ 3000 mg/kg (class 3_{ns}), with a maximum of 12,000 mg/kg). Some other polluted sites were located in small recreational lakes, probably connected to local boating activities.
- **PCBs** (range: 1 - 465 µg/kg); see Fig. 2H. No major sediment pollution was observed for these components; only 8 stations exceeded the class 3_{ns} limits of 200 µg/kg.

POLLUTION AT FOUR SELECTED AREAS IN DELFT

Two apparent hotspots of sediment pollution in Delft were found, *viz.* the inner city and an industrial area near the Rijn-Schie canal. The lowest pollution levels were generally present in the SW residential area, and in the NE recreational site. In the following, a more detailed investigation of sediment pollution in these four areas will be presented (see Fig. 1):

- **Zone A:** Inner city (21 station; 27 observations);
- **Zone B:** Industrial area (12 stations; 14 observations);
- **Zone C:** Residential area (31 stations; 44 observations);
- **Zone D:** Recreational area (25 stations; 29 observations).

Using the Chi square test [8], it was found that, both for zone A and B (inner city; industrial area) most observations showed a non-normal distribution, with a positive skewing towards the higher values. Therefore, it was decided to apply non-parametric statistical testing methods here. These included the use of medians (instead of means) and of Spearman rank correlation calculations (instead of parametric correlations) [7]. In contrast, the residential and recreational areas (zone C and D) showed normal distributions for virtually all the components.

Table 2. Mean and median sediment pollutants values in four areas in Delft. For definitions and units, see Table 1. The areas with the relatively highest pollutants contents are indicated in bold.

		Cr	Ni	Cu	Zn	Cd	Hg	Pb	As	DDT	PCBs	PAHs	OIL
A: Inner city	Median	**45**	**24**	**195**	675	1.6	**1.3**	**325**	**20**	**20**	**76**	**11**	630
B: Industrial	Median	29	20	170	**685**	**1.8**	0.28	120	18	13	**76**	10	**2200**
C: Residential	Mean	32	21	25	140	0.62	0.11	49	12	4.2	5.5	3.4	280
D: Recreational	Mean	23	19	44	185	0.54	0.29	96	9.1	6.8	23	2.2	270

Fig. 2 A-H. Pollution classes for heavy metals and organic micropollutants at 188 sediment stations in Delft (cf. Fig. 1). Indicated are class 2 (“lightly polluted”), class 3 (“polluted”) and class 4 (“highly polluted”) sediments. No corrections were made with respect to “non-standard sediment” composition.

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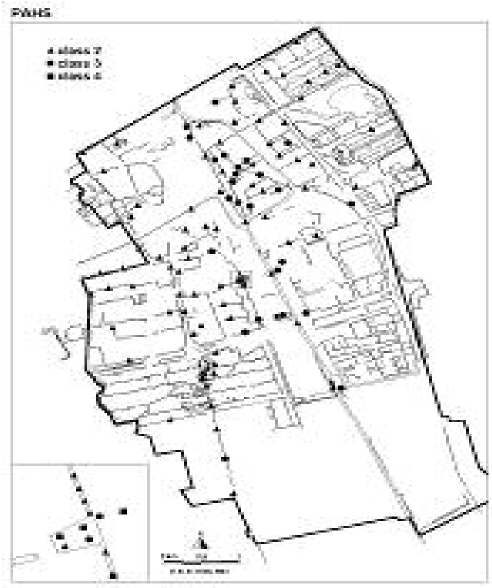
PAHS
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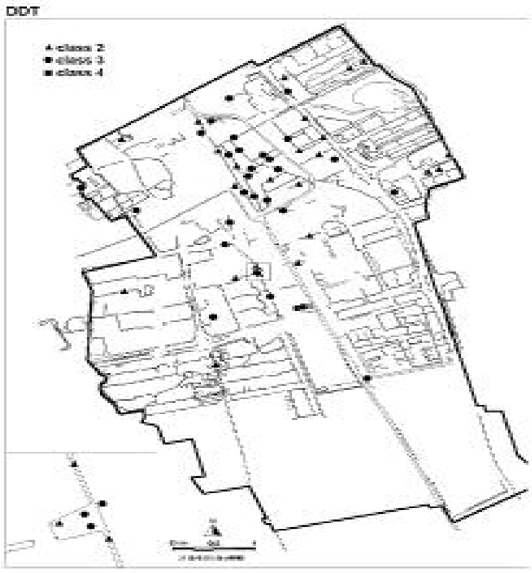
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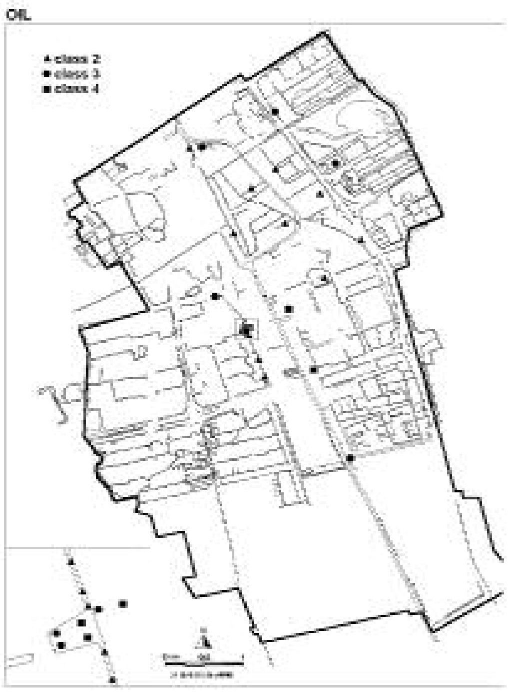
OIL
class 2
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class 4

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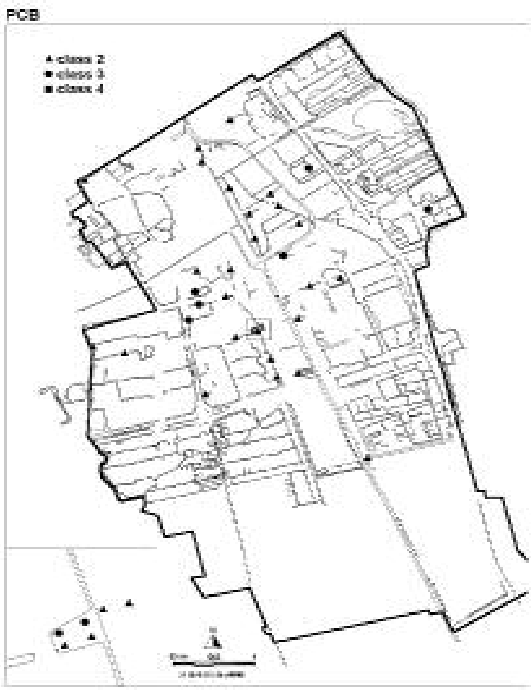
PCB
class 2
class 3
class 4

With respect to the results of the Spearman rank correlation for the inner city sediment data (see Table 3), the following trends can be observed:

- For the heavy metals, the best correlations were found between Hg-Cu and Ni-Cr;
- in general, chromium and copper showed the best correlations with the other heavy metals;
- least significant regressions were observed in case of lead and cadmium;
- Arsenic showed good correlations with all heavy metals except Pb.
- Other significant correlations involved PAHs and PCBs with heavy metals; on the other hand, mineral oil and DDT hardly showed significant regressions with the other components.

The majority of the Delft inner city sediment pollution comes, directly and indirectly, from the river Rhine, and the problems must be connected to water pollution in the Rhine basin itself (Fig. 3). The following main (point and diffuse) pollution sources can be found here [11, 12]:

- **Industrial discharges**. Important contributions in the river Rhine basin have been coming from *e.g.* metal processing industries (Ni, Cr, Cd), leather tanning (Cr) and chemical industries (heavy metals, organics). Industrial inputs have sharply decreased since the 1960s and nowadays only constitute a minor fraction (around 10%) of the total waste loads;
- **Urban sewage treatment plant effluents**, responsible for similar pollutants loadings;
- **Diffuse sources.** These constitute the major contribution (*viz.* 50-80 % [12]) of heavy metal loadings in the Rhine basin; examples are:
    - Corrosion of metal structures, of water distribution networks, of buildings and bridges, etc. (Zn, Cu, Pb);
    - Storm water overflows;
    - Natural and anthropogenic run-off, *e.g.* from agricultural lands;
    - Traffic. Pb has been used extensively as an anti-knocking agent in car petrol. However, its use has gradually been banned since the 1980s. A major source of the PAHs may have been car traffic as well in view of incomplete petrol combustion.

Following the introduction of catalysts in cars this PAHs pollution source has sharply decreased.

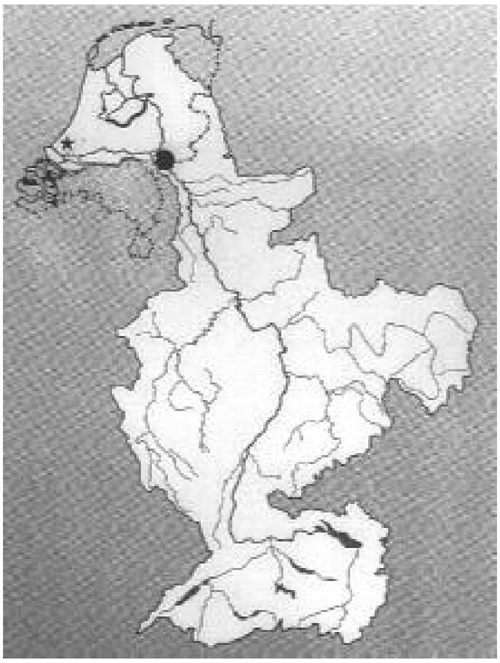

Fig.3. The river Rhine basin with the monitoring station Lobith (●) and Delft (★)

(|------| = 100 km)

It is interesting to compare the inner city sediment data with those of the river Rhine, and in particular with the pollutant contents on river Rhine suspended matter (mg/kg). For this the on-line WATERSTAT data base was used (www.waterstat.nl). The river Rhine monitoring station "Lobith" at the Dutch-German border (see Fig. 3) offered the most comprehensive set of water quality data. Unfortunately, insufficient data were available for river Rhine branches closer to Delft.

The Delft sediment data have all been collected around the year 1995. This year thus represents the age of the top layer of the sediment samples. With an average sludge thickness representing 10 years [1], this would imply that the inner city sediment samples give an average picture of the situation in the year **1990**. To make a reasonable comparison, it was therefore decided to choose the river Rhine median values for the period 1986-1995 (see Table 4).

Table 4. Median contents (mg/kg, except PCBs (in µg/kg)) of pollutants on suspended matter in the river Rhine at Lobith over the years 1986-1995. N.m.= not measured.

Cr	Ni	Cu	Zn	Cd	Hg	Pb	As	DDT	PCBs	PAHs
90	50	100	600	2.5	1.0	125	19	N.m.	90	7.4

* Estimates; these components have only been measured in the latter part of above 10 years period.

Some 70% of the inner city Cu, Pb and Zn sediment pollution can be ascribed to the Rijn-Schie canal. It was now assumed that this fraction holds for all other components as well. Thus, multiplying the Delft inner city sediment contents (see Table 2) with a factor 0.7 would theoretically yield the suspended matter contents in the river Rhine: $\frac{Delft_{corr.}}{Rhine} = 1.0$; see Fig 4.

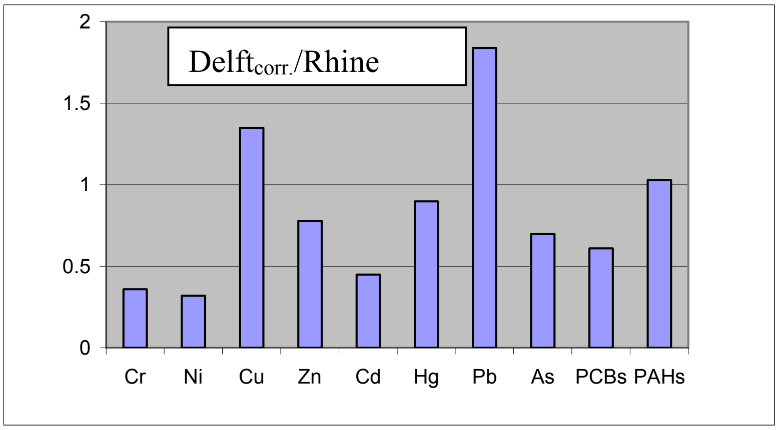

Fig. 4. Ratios for median pollutants contents in the Delft inner city sediment (see Table 2; all values corrected by multiplying by 0.7), and on river Rhine suspended solids (see Table 4).

According to the crude assumptions made here, a difference of ± 0.5 with the theoretical ratio of 1.0 seems acceptable. The following observations can thus be made:

- A good correspondence exists for the parameters Cu, Zn, Hg, As, PCBs and PAHs.
- For Cr, Ni and Cd, we found an **under**estimation in the Delft inner city sediments as compared to river Rhine data.
- For Pb, we found an **over**estimation as compared to river Rhine data.

A number of reasons can explain the above deviations. The relatively low values for Cr, Ni and Cd may be due to a relative “loss” of these components on the way from the river Rhine monitoring station “Lobith” to Delft. This is reasonable, since much sedimentation of suspended matter will have taken place, together with adsorbed heavy metals and organic micropollutants. This may also be the reason why the values for all ratios, except for Pb and Cu, were found to be < 1.0.

The relatively high Pb (and, to some extent Cu) ratios in Fig. 3 seem to indicate an additional Pb input in the Netherlands itself. Although sharply decreased in recent times, atmospheric deposition of lead has, in the 1980s, still been the major diffusive pollution source for the whole river Rhine basin [11], and it may well be that relatively large Pb loadings have derived from traffic in the Netherlands. It is remarkable that, of all heavy metals in the Delft inner city sediment, Pb showed the poorest correlations with all other components (see Table 3).

Industrial area (zone B)

As for the inner city sediments, high sediment pollution levels were often observed here (see Table 2). Remarkably, the largest pollution values were nearly always found in the centre of the area, with a decrease towards the edges (see Fig. 2). In the past, industrial activities connected to metal processing and manufacturing of metal constructions were carried out here without proper pollution control. The relatively high heavy metal contents may be explained by these industrial activities, as may be the exceptionally high "mineral oil" pollution levels.

The abundance of the individual PAHs components follows exactly the same pattern as for the inner city sediments (for details, see [5]). We found the following PAHs ratios: F/PY = 1.6 ± 0.3; P/A = 4.1 ± 0.5. This is a clear indication of urban run-off as the major PAHs pollution source. We could have expected more PAHs of low molecular weights, as found in mineral oil. Over the years, these may have been lost from the sediments, through *e.g.* volatilisation and microbial degradation [14].

Table 5. Results for the Spearman rank correlations between sediment parameters at industrial site B (see Fig. 1; n=14). + = significant ($p<0.01$, i.e. $r \geq 0.66$); ++ = highly significant ($r \geq 0.9$).

	Ni	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**	**As**	**DDT**	**PCBs**	**PAHs**	**Oil**
Cr	+	+	+	+	-	+	-	+	+	-	-
Ni		++	++	+	-	++	+	++	+	-	+
Cu			+	+	+	+	+	-	+	+	++
Zn				++	+	++	+	+	++	+	+
Cd					-	++	+	+	++	+	+
Hg						+	+	-	+	-	+
Pb							+	+	++	+	++
As								-	+	-	+
DDT									+	-	+
PCBs										+	+
PAHs											+

Compared to the inner city, it is apparent that in general even better mutual correlations can be found between the heavy metals, including Pb. This is a clear indication of the presence of one or a few sources of pollution here, probably related to metal processing (plating, pickling, rinsing, *etc.*) [15]. These activities will have given rise to the PCB, mineral oil and PAH pollution as well. The observed significant correlations with DDT are higher than expected and seem to be less in line with "industrial pollution".

Residential and recreational areas (zones C and D)

Relatively lowest values of sediment heavy metals, arsenic and organic micropollutants were observed in the zones C and D (see Fig. 2). The heavy metal and arsenic contents were in the range of the background values for sediments in the Netherlands (see Table 6). Comparable sediment background levels in Europe have been reported elsewhere [15].

Table 6. Mean values of sediment parameters in zone C and D (see Fig. 1), and background values in sediment in The Netherlands [16] (all values in mg/kg)

Component	**Sediment values in residential area C**	**Sediment values in recreational area D**	**Background values in sediment, The Netherlands**
Cr	32	23	100
Ni	21	19	35
Cu	25	44	36
Zn	140	190	140
Cd	0.6	0.5	0.8
Hg	0.1	0.3	0.3
Pb	49	95	85
As	12	9	29

There do not seem to be consistent differences between the residential and recreational areas. It might be expected that Pb and PAHs values would be higher in the residential area (related with local traffic) but this was not observed here. Apparently both the measures on alternative anti-knocking agents in car petrol (for Pb) and the use of catalysts in cars (for PAHs) have been successful. Correlations between heavy metals and arsenic can be seen to be reasonably significant; however correlations were virtually absent in the case of organic micropollutants (see Table 7).

Observed F/PY ratios were 1.3 ± 0.07 and 1.6 ± 0.3, respectively, for the residential and recreational area. Values of 4.0 ± 0.5 and 5.8 ± 1.5 were found for the P/A ratios. All this indicates that urban run-off has been the major PAHs source here.

Table 7. Linear regression results between pollution parameters in the residential and recreational areas C and D (+ = significant ($p<0.01$; i.e. $r>0.48$); ++ = highly significant correlation ($r> 0.8$)). First figure: zone C; second figure: zone D (see Fig. 1).

	Ni	**Cu**	**Zn**	**Cd**	**Hg**	**Pb**	**As**	**DDT**	**PCBs**	**PAHs**	**Oil**
Cr	+	-/-	-/-	-/+	-/-	-/-	+/-	-	-	-	-
Ni		+	-/+	+/-	+	-/+	+/-	-	-	-	-
Cu			+	+/-	+/++	+	-/-	-	-	+/-	-
Zn				++/-	-/+	-/+	+/-	-	-	-	+/-
Cd					-/-	-/-	+/-	-	-	-	+/-
Hg						-/++	-/-	-	-	-	-
Pb							-/-	-	-	-	-
As								-	-	-	-
DDT									-	-	-
PCBs										-	-
PAHs											-

CONCLUSION

Until recently, point and diffuse pollution sources from the river Rhine have had major adverse impacts on the sediment quality in the surface waters of The Netherlands. Sanitation measures in the Rhine Basin over the last decades have led to a gradual improvement of this situation. In Delft, there are legacies of industrial activities that play a role as well. The techniques for disposal and treatment of polluted sediments are all costly and rather ineffective [17]. Therefore, with the drastic sanitation of point sources, abatement of diffuse pollution sources has now become a factor of paramount importance.

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