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# Pollution sources and abatement measures for dredged sediments in the city of Delft (The Netherlands)

## Abstract

*This paper gives an overview of a long-years study on the supply, pollution sources and remediation measures with respect to dredged sediments in the city of Delft (The Netherlands). An inventory at 200 stations showed that especially the inner city canal sediments have strongly been polluted with heavy metals and organic micropollutants. Shipping traffic plays a dominant role in the supply of the sediment; prevention measures should especially be aimed at a reduction of vessel speeds. With respect to sanitation measures of the polluted dredged materials, hydrocyclones were shown to offer promising results for reduction of the waste volumes.*

## Quality classification of dredged sediments in Delft

Like many other municipalities in The Netherlands, the city of Delft has to deal with large problems with polluted dredged materials from its waterways. Around 100,000 $m^3$ of this material will have to be processed in the coming years. Important criterion for the necessary sanitation measures is the sediment quality, as expressed in the Dutch system of quality classes (see Table 1; cf. [1]). Whereas dredged sediments of classes 1-2 may generally be disposed of into the environment without many restrictions, class 3-4 sediments are of unacceptable quality and need (for class 4: with highest urgency) sanitation, *viz.* dredging, disposal in special storage reservoirs and, if possible, sediment cleanup measures [1]. This will increase the processing cost by at least a factor 2.

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| Parameter | 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 | < 0.01 | 0.01 - < 0.02 | 0.02 - < 4.0 | ≥ 4.0 |
| Σ PCB | < 0.1 | 0.1 - < 0.2 | 0.2 - < 1.0 | ≥1.0 |
| Σ PAH | <1 | 1 - < 10 | 10 - < 40 | ≥ 40 |
| Mineral oil | < 1000 | 1000 - < 3000 | 3000 - < 5000 | ≥ 5000 |

**Table 1.**
Definitions of pollution classes (as for the year 1997) with respect to the (heavy) metals chromium, nickel, copper, zinc, cadmium, mercury, lead, arsenic, and organic micropollutants contents (mg/kg) for polluted sediments in The Netherlands; class limits have been normalized with respect to a "standard sediment" with 10% organic matter and 25% lutum (< 2µm) contents. In case of non-standard sediment composition, correction factors were applied, taking into account the stronger, semi-permanent binding capacity of organic-rich, muddy sediments (and similarly for organic-poor, sandy sediments) [cf. 1].
The *overall* sediment quality at a certain station was established using the "worst class" sediment parameter(s).
Σ DDT expresses the sum of all DDT, DDD and DDE congeners.
Σ PCB 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.

Sediment samples down to 20-50 cm depth were taken at 200 stations all over Delft. The samples were analysed by a certified laboratory on grain size distribution, organic matter contents and levels of heavy metals and organic micropollutants (for details, see [2]). The results in Fig. 1 show that for the inner city canals, 95% of the dredged sediments belong to the strongly polluted classes 3 and 4. In the outer city, this percentage lies at around 45%; also the pollution levels here are some 2-10 times lower than in the inner city.

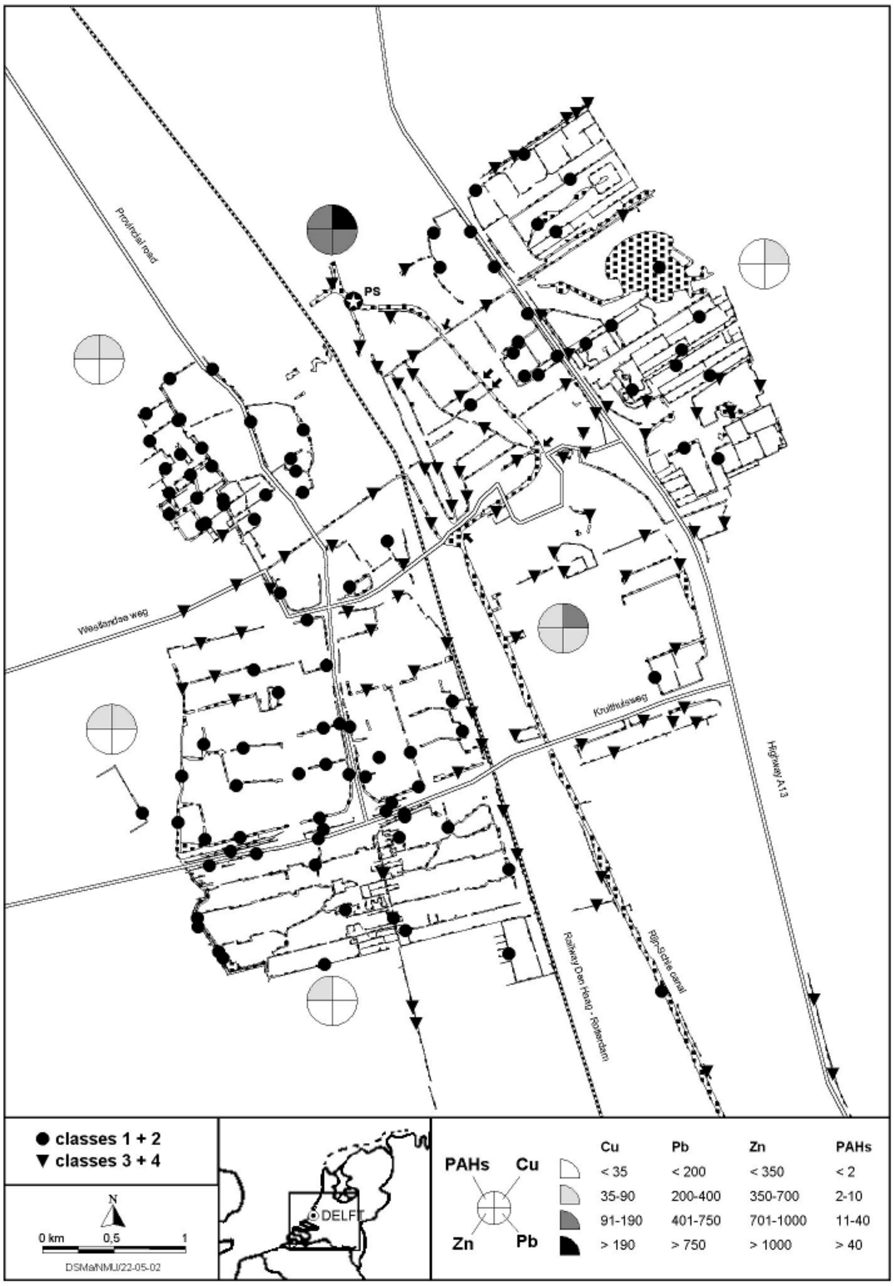

**Figure 1**

The factor most contributing to the sediment pollution in the inner city canals turned out to be the high copper contents, followed by polycyclic aromatic hydrocarbons (PAHs), DDT (including its degradation products), zinc and lead. Studies on the vertical distribution of pollutants in the inner city sediments showed clearly decreasing (*viz.* 20-55%) pollution levels in the upper, most recently deposited sediment materials [2, 3]. In spite of this, 95% of this material still belongs to the quality classes 3-4, a phenomenon that can nearly completely be attributed to the –still too high- copper levels in these top sediments. It can be shown (see later) that the sediment pollution here can mainly be attributed to the import of suspended solids from the main shipping canal "Rijn-Schie canal" (see Fig. 1), with which the Delft inner city canals have open connections. For the outer city dredged materials, Cu and PAHs contents were shown to have similar contributions to the class 3-4 sediment classification.

Pollution in the outer city sediments is evidently of a much lower magnitude than for the inner city. Some "hotspots" of pollution (Kelderman, in prep.) may be observed (see Fig. 2), *viz.*:

- In outer city canals in direct connection with the inner city canal system;
- Sites at or connected to the Rijn-Schie canal;
- Stations located close to roads or highways. Elevated PAHs contents, mostly leading to moderately polluted class 3 sediments, are often observed here. This may probably be attributed to automobile exhaust gases [4, 5]. It is remarkable that the Pb contents in the sediments hardly ever contribute to class 3-4 classification; apparently the replacement of lead by alternative anti-knocking agents in car petrol has resulted into this quality improvement.
- Stretches along the railway Den Haag-Rotterdam. In this case, highly elevated levels of copper (up to > 500 mg/kg) were found. Most important pollution source here will be abrasion of copper particles from the high voltage electricity wires [6].

In view of the severity of the Delft inner city sediment pollution, most of the research has concentrated on this area. In the following chapters, the quantitative supply, pollution sources and possible remediation measures will further be discussed.

## *Quality classification of dredged sediments in Delft*

For the supply of sediment material into the Delft inner city, the main navigation canal "Rijn-Schie canal", which surrounds the inner city (see Fig.1), is of major importance. This canal directly imports water from the river Rhine into the Delft area; very similar water qualities of the two can therefore be found [7, 8].

At five inlet points, the canals in the Delft inner city are in open connection with the Rijn-Schie canal (see Fig.1.). At the same time, a pumping station takes care of flushing the inner city canals, leading to a permanent flow velocity of *ca.*5 cm/sec. in these canals [9]. Effectively, Rijn-Schie canal water is thus imported into (and later exported out of) the Delft inner city canal system. Together with this, suspended materials will be imported, contributing to the sediment loading. Under completely quiescent conditions in the Rijn-Schie canal (*i.e.* under absence of shipping traffic during at least 2 weeks periods in harsh winters), the suspended solids (SS) contents in the Rijn-Schie canal were found to be very low, *viz.* 3.2 ± 0.8 mg/L[2] [9]. Normally, however, the busy shipping traffic is responsible for **continuously** increased SS contents in the canal, *viz.*12 ± 2 mg/L. An extra factor are the water level variations occurring during ships' passages, effectively leading to a "shock wave" of water with high suspended solids contents (up to 150 mg/L) **into** the Delft inner city canals [9].

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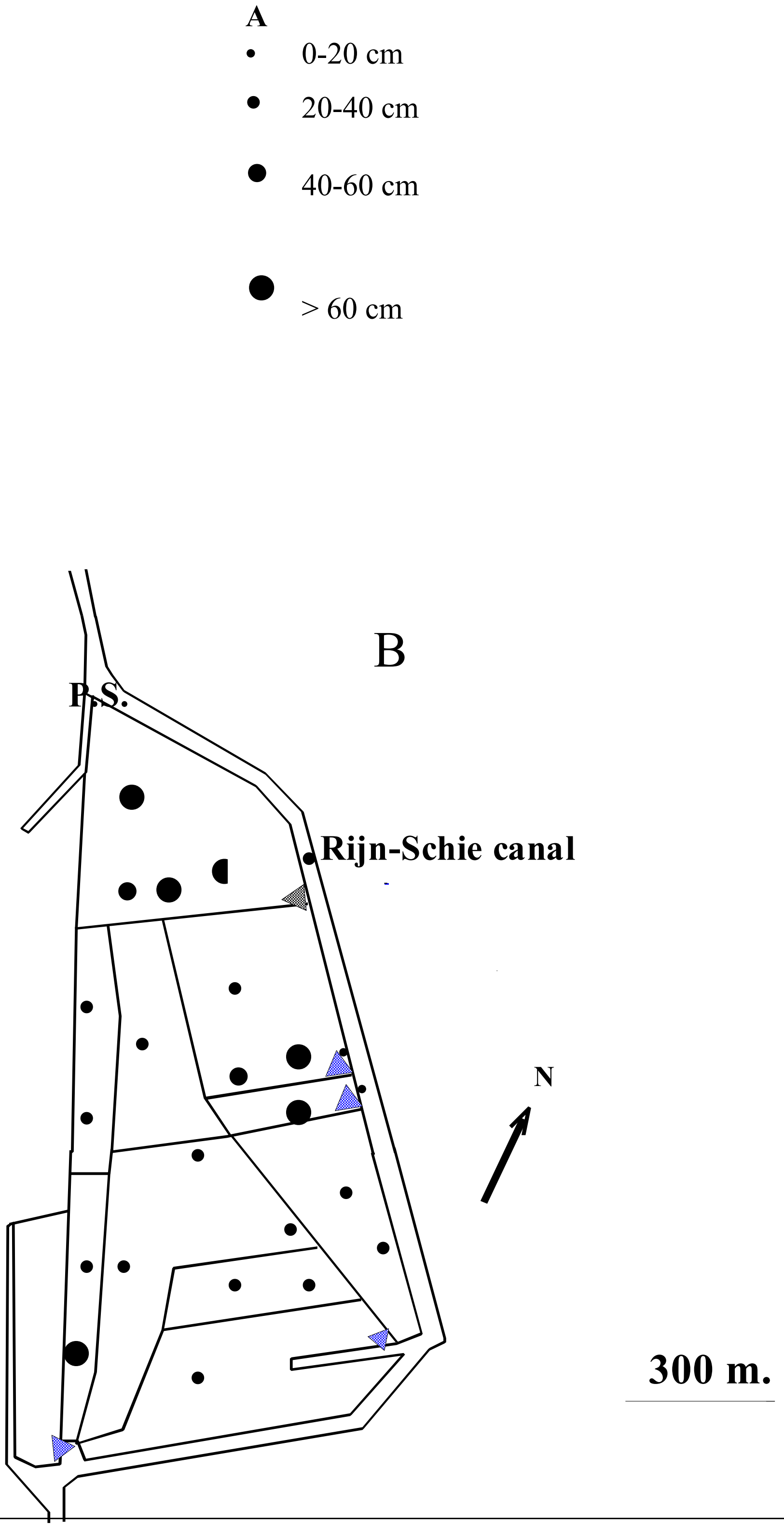
[2] 95% confidence intervals will be used throughout this text

**Fig. 2A/B.** Sludge thicknesses along the Delft inner city canals in 1996/97 (▲ = inlet points Rijn-Schie canal; P.S. = pumping station).



Making use of the number of annual ship movements, the amount of imported water and its SS contents, a value of 520 x $10^3$ kg/year was estimated for the annual import of suspended materials into the Delft inner city canals [2]. Compared to this, the SS import under completely quiescent conditions would only be 85 x $10^3$ kg/year.

The external SS import of 520 x $10^3$ kg/year is much higher than the internal sources (*e.g.* via run-off from the streets and through combined sewer overflows); these internal sources were roughly estimated at *ca.* 10 x $10^3$ kg SS/year [10, 11], yielding a total sludge accumulation in the Delft inner city canals of **530 x $10^3$ kg/year**.

**Sludge thickness measurements**

In 1996/97, a research was undertaken to quantify the sludge accumulation in the Delft inner city canals over the period since the last dredging operation, *i.e.* 12 years [12]. Sludge thicknesses were measured with a measuring stick in 5-15 metre intervals. The results of the survey (see Fig. 2) indicate that the sludge thickness in the centre of the inner city is in the range of 20-40 cm. Highest sludge thicknesses, up to 1 metre, can be found in the canals directly connected to the Rijn-Schie canal, and in the canal adjacent to the pumping station P.S. This must be due to, on the one hand, the earlier mentioned import of suspended materials during ships' passages and, on the other, to the accumulation of sludge just before the inlet works of the pumping station. The earlier mentioned "shock waves" during the ships' passages must be responsible for the relatively thin, *viz.* 0-30 cm, sludge layers near the entrance points at the Rijn-Schie canal (see Fig. 2).

With the help of these measured sludge thicknesses and the estimated specific weights and water contents, a total sludge accumulation of **550 x $10^3$ kg/year** was calculated [9]. This value is in excellent agreement with the just reported value of 530 x $10^3$ kg/year, as calculated from the external and internal sludge inputs into the Delft inner city canals.

## *Mass budgets for heavy metals*

Using the annual import of suspended materials from the Rijn-Schie canal and their heavy metal contents, mass budgets for lead, copper and zinc were set up [2]:

HM (ext.) + HM (int.) = HM (acc.) (kg/year), in which:

- HM (ext.) = Heavy metals import via the five inlet points with the Rijn-Schie canal
- HM (int.) = Contributions from heavy metals inside the inner city
- $HM_{acc.}$= Fitting term: accumulated heavy metals in the Delft inner city sediments.

(N.B. as mentioned before it was assumed that no sludge was pumped out via the pumping station: HM (out) = 0).

The results (see Fig. 3) show that only 15-35% of the heavy metals in the Delft inner city canals derive from internal pollution sources, such as run-off from streets and combined sewer overflows. Noteworthy contributions in this respect are corrosion of zinc roofs, use of paints for houseboats and illegal waste discharges [11, 13].

The larger part, *viz.* 65-85%, of the heavy metals in the Delft inner city sediments can be attributed to the import via the Rijn-Schie canal. It is improbable that *local* pollution sources in the Delft area markedly contribute to this pollution load. Around 99% of all point sources in the Delft area have a connection to sewage treatment works; of all effluents of these treatment works, some 90% is discharged on large waters outside the region. On the other hand, the river Rhine is the direct source of intake water in the region, via the Rijn-Schie canal. Results of the statistical techniques Factor and Cluster analysis [14] strongly indicate that the suspended solids in the river Rhine and the Delft inner city canal sediments have one or more common sources [7]. Evidently, abatement measures for the inner city sediment pollution must especially be aimed at tackling the **external** supply of polluted sediments via the Rijn-Schie canal.

The *calculated* heavy metal accumulation data in Fig. 3 are in very good (*viz.* 12-27% difference) with their *measured* values, based on the heavy metal contents in the Delft inner city canal sediments [9]; this can be seen as a clear support for the above mass budget approach.

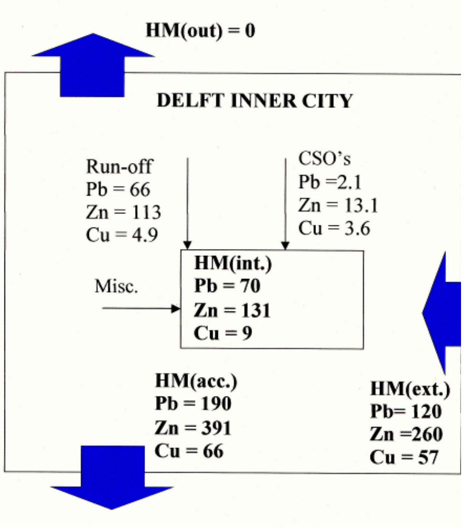


Fig. 3: Mass budgets for the heavy metals Pb, Zn and Cu in the Delft inner city canals, indicating the external (HM(ext.) and internal (HM(int.)) heavy metal loadings onto the system (kg/yr); the latter term comprises the contributions from run-off, combined sewer overflows (CSO's) and miscellaneous terms. HM(out) and HM(acc.), also in kg/yr, represent the heavy metal output (= 0) and accumulation terms, respectively.

## *Abatement measures*

### Shipping traffic on the Rijn-Schie canal

As mentioned before, the intensive shipping traffic on the Rijn-Schie canal must be responsible for the continuously increased suspended solids contents in the imported water. Additional SS imports take place during ships' passages. To assess these effects, a research was carried out on the influence of shipping movements on the suspended solids contents in the water; use was made of a computer model as well as laboratory experiments [15].

The mathematical formulation of the model was based on the shear stress exerted by the ship's propeller on the sediment bed of the canal, giving rise to resuspension of the sediment particles when a certain critical shear stress value would be exceeded [cf. 16]. Also other hydraulic phenomena such as the draw down of the water during a ship's passage were taken into account. In the modelling exercise, uniform conditions were assumed with respect to vessel size and tonnage as well as to water depth and sediment characteristics. The only remaining variable was then the ship's velocity.

The results in Fig. 4 clearly show that under increasing ship's speeds there is a strong increase in the current velocity along the sediment bed. This will result into sediment resuspension and thus into strongly enhanced suspended solids contents in the water. According to the model results, elevated SS contents will especially occur at vessel speeds > 2.5 m/sec. Thus, we can see in Fig. 4 that a vessel speed increase from 2.0 to 4.0 m/sec. will increase the SS contents from *ca.* 20 to >200 mg/L. On the other hand, at vessel speeds < 1.5 m/sec., the SS contents will decrease to values below 5 mg/L.

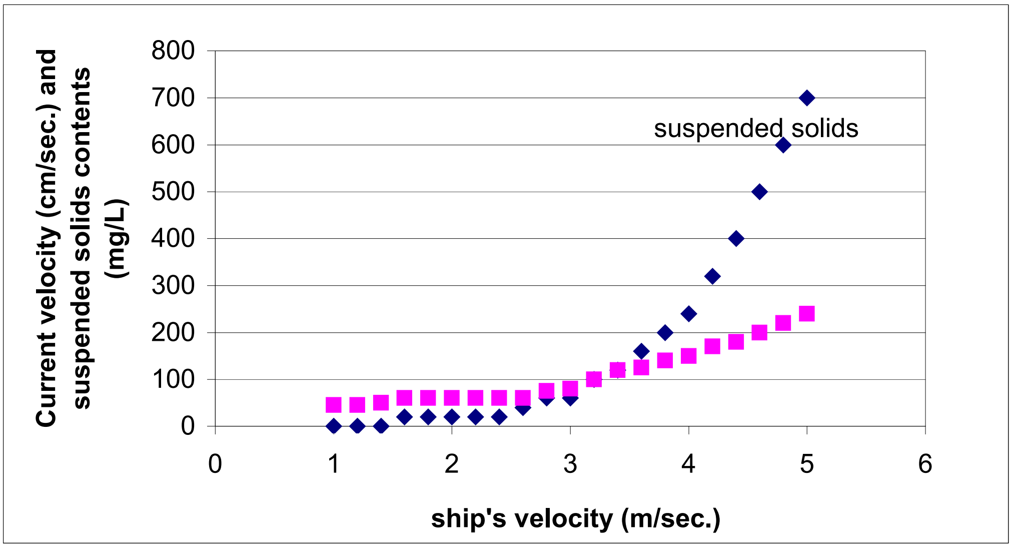


**Fig. 4.** Modelled bottom currents velocities and suspended solids contents in the Rijn-Schie canal as a function of ship's velocities during the passage of ships.

Above modelling results offer a good picture of the real situation. With the estimated average vessel speeds in the Rijn-Schie canal of 3-4 m/sec., the model predicts SS contents of 50-150 mg/L during ships' passages, in very good agreement with the actual measured values in the field [9]. On the other hand, as mentioned before, SS contents of *ca.* 3 mg/L were found during 2-3 weeks periods of complete absence of shipping traffic in the Rijn-Schie canal. Laboratory experiments on the resuspension and settling rates of Rijn-Schie canal sediments with different grain sizes further support above findings [15].

As a management strategy to reduce the import of suspended solids into the Delft inner city canal system, it is therefore recommended to set a maximum speed for ships in the Rijn-Schie canal of 1-1.5 m/sec. Further measures could be constructions for temporary closing off the water inlets, sediment trap devices inside the inlet canals, *etc.* In this way, the import of suspended solids from the Rijn-Schie canal can potentially be reduced with some 85% (*viz.* from 520 to 85 tonnes per year).

### Heavy metal pollution

Reduction of the heavy metal pollution of the dredged sediments in the Delft inner city can especially be achieved through quality improvements of river Rhine water. In the last decades, large progress has been made in this respect [7]. It is remarkable that, in contrast to *e.g.* cadmium and mercury, which mainly derive from point sources of (industrial) pollution, the copper and zinc contents in the river Rhine show a less strong decrease. Thus, the Cd and Hg contents on river Rhine suspended matter decreased with 50-60% over the period 1988-1999, compared to only 20-30% for copper and zinc (with typical present values of 80 and 500 mg/kg, respectively) [cf. 8]. A similar trend has earlier been reported for Delft inner city canal sediments [3, 7]. Probably the latter metals are for a large part coming from *diffuse* sources; management of these sources has been a bottleneck in the ultimate cleanup of surface waters [17, 18]. Thus, high contributions for copper may be expected from the corrosion of water distribution networks as well as from agricultural sources (where Cu is used as a fodder additive). For zinc, corrosion from roofs and other metal structures will play an important role.

Finally, abatement of the internal pollution in the Delft inner city (responsible for 15-35% of the heavy metal pollution of the sediments) can best be achieved by improving water drainage during rainstorms and by sanitation of local pollution sources (*e.g.* houseboats).

### Clean-up of the polluted sediments

Generally there is a large tendency for heavy metals and organic micropollutants to be adsorbed onto especially the *fine* fraction of the sediment [13, 16]; this phenomenon can be ascribed to the larger specific surface area of the smaller sediment particles. In Delft sediments a similar pattern was found (see Table II [3, 10]). Separation of the sandy ("clean") and muddy ("polluted") sediment fraction could thus in principle be used as a technique for reducing the amount of waste materials.

| Grain size | Cu (mg/kg) | Cd (mg/kg) | Pb (mg/kg) | Zn (mg/kg) |
|---|---|---|---|---|
| < 0.1 mm | 425 ± 140 | 7.0 ± 1.9 | 650 ± 175 | 1190 ± 270 |
| ≥ 0.1 mm | 95 ± 20 | 3.0 ± 0.6 | 260 ± 55 | 395 ± 95 |

**Table 2.:** Heavy metal contents (averages and 95% confidence intervals at 6 stations) for different grain size fractions of Delft inner city canal sediments.

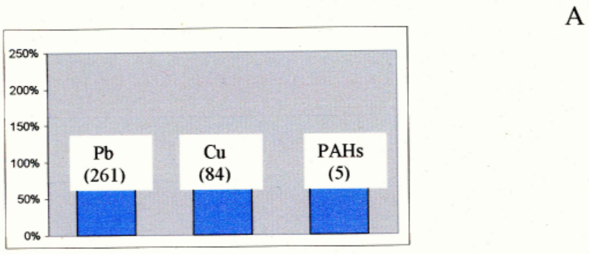
In a large-scale study on possible clean-up techniques of polluted dredged sediments in The Netherlands [19, 20], it was concluded that for sediments polluted with a "cocktail" of heavy metals and organic micropollutants, separation of the fine and crude fraction with the help of hydrocyclones, probably offers the best, cost effective option, provided that the sand (> 0.05 mm) content is > 50% (preferably > 80%).

In a hydrocyclone (see Fig. 5A) separation of the two fractions is achieved after introduction of the diluted sediment slurry on top of the funnel shaped design. Centrifugal forces will be created during the downward transport, leading to a movement of the finer sediment fraction to the centre. Because of the decreasing diameter, this sediment fraction will be pushed upward out of the hydrocyclone ("overflow"), whereas the crude sediment material will be transported down in the underflow. An important process parameter is the $d_{50}$ value of the hydrocyclone, *i.e.* the sediment grain size, which has a 50% probability to go to the underflow (and also 50% to go the overflow). A main disadvantage of hydrocyclones is the relative poor separation capacity between the fine and crude fraction, a capacity that can however be improved by pre-settling of sandy materials in a settling basin, and/or by using a number of hydrocyclones in series [19, 21].

Laboratory experiments were carried out with four Delft inner city canal sediments to test the potential possibilities of hydrocyclones [22]. The set up consisted of three small (typical dimensions 10-40 cm.) pilot plant hydrocyclones in series, with $d_{50}$ values of 60 µm, 16 µm and 10 µm, respectively (see Fig. 5B).

The results indicate that, compared to the original sample, the Pb, Cu and PAHs contents in the 60 µm underflow were reduced with 75-85%, and will probably have yielded a relatively unpolluted class 1-2 sediment (cf. Table 1). Heavy metal contents steadily increased in the subsequent 16 µm underflow, the 10 µm underflow and, finally, the 10 µm overflow. In this last fraction the heavy metal contents were found to be 50-100% higher than in the original, raw sample (N.B. due to the small quantity of materials, PAHs contents could usually not be analysed in the smaller hydrocyclone fractions).

It can be concluded that hydrocyclones offer a promising (pre-) treatment technique for polluted Delft canal sediments. However, this option should, together with others, be evaluated in the light of cost, priorities for sanitation and future developments in the legal framework for polluted sediments in The Netherlands.



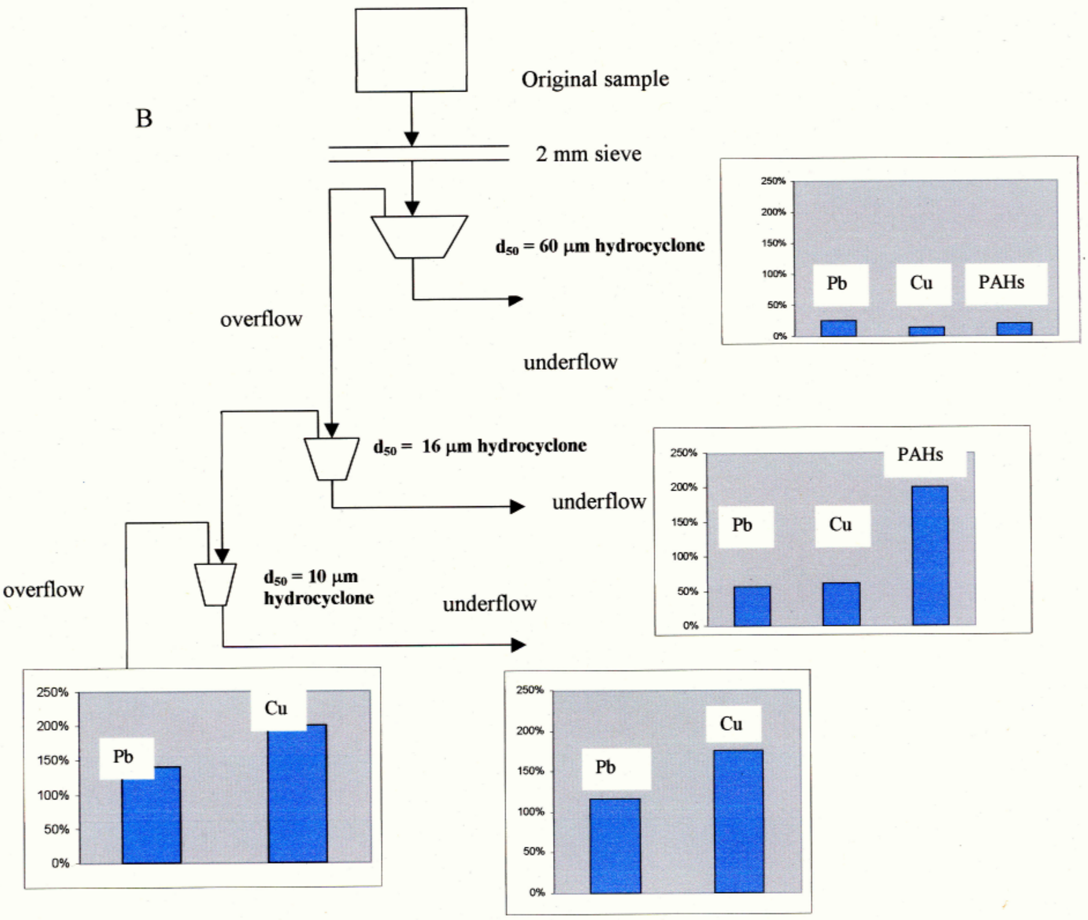


**Fig. 5A.** A hydrocyclone (typical dimensions (L x W x H): 2.5 x 2.5 x 7 metre).
**Fig. 5 B.** Set up of the hydrocyclone experiments for four representative sediments from the Delft inner city canals. The results of the experiments, as averages over the four sediments are also indicated here, *viz.* (clockwise starting from top left: original sample with values expressed as 100% and as mg/kg; 60 µm hydrocyclone underflow; 16 µm underflow; 10 µm under and overflow, respectively.

## *Conclusion*

The main priority for pollution abatement of Delft canal sediments lies in *the inner city canal system*, both from a quantitative and qualitative point of view. It may be expected that the quality of the *outer city sediments* will improve parallel with that of the inner city, since there are often hydrological connections between the two. A clear concern, however, remains the effect of the railroad, through very high copper levels in the nearby waterways.

In this research, the main pollutants in Delft inner city canal sediment turned out to be copper, PAHs, DDT and zinc. For the two heavy metals, the main supply was shown to be diffuse waste discharges into the river Rhine. Continuing efforts will have to be made in the abatement of these non-point sources, after the successful sanitation of point sources during the last decades. The organic micropollutants PAHs and DDT are probably from more local origin. Relatively high contents of DDT (and its degradation products) were found in the Delft sediments, decades after its ban in the 1970's. For PAHs, main pollution sources will be automobile exhaust gases and industrial air emissions. It may be expected that, together with the abatement of industrial emissions, the use of catalysts in cars will drastically bring down the traffic-related PAH pollution loads.

Still, since the Dutch sediment quality classification system is based on the "worst class" parameter, the "chain will be as strong as the weakest link". Therefore, drastic improvements in the dredged sediment quality can only be reached by a balanced, integrated sanitation of all relevant point and non-point sources of pollution.

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