This manuscript was published in

Vandecasteele B., De Vos B., Tack F.M.G., 2002. Heavy metal contents in surface soils along the Upper Scheldt river (Belgium) affected by historical upland disposal of dredged materials. The Science of the Total Environment 290 (1-3), 1-14.

The original publication is available at http://www.elsevier.com/

Heavy metal contents in surface soils along the Upper Scheldt river (Belgium) affected by historical upland disposal of dredged materials

Bart Vandecasteele[a,*], Bruno De Vos[a] & Filip M.G. Tack[b]

[a] *Institute for Forestry and Game Management, Ministry of the Flemish Community, Gaverstraat 4, B-9500 Geraardsbergen, Belgium*
[b] *Ghent University, Department of Applied Analytical and Physical Chemistry , Coupure 653, B-9000 Ghent, Belgium*

* Correspondence: B. Vandecasteele. E-mail: Bart.Vandecasteele@lin.vlaanderen.be, Tel. 00 32 54 437136, Fax. 00 32 54 436160

Heavy metal contents in surface soils along the Upper Scheldt river (Belgium) affected by historical upland disposal of dredged materials

Bart Vandecasteele[a,*], Bruno De Vos[a] & Filip M.G. Tack[b]

[a] *Institute for Forestry and Game Management, Ministry of the Flemish Community, Gaverstraat 4, B-9500 Geraardsbergen, Belgium*
[b] *Ghent University, Department of Applied* Analytical *and Physical Chemistry , Coupure 653, B-9000 Ghent, Belgium*
* Correspondence: B. Vandecasteele. E-mail: Bart.Vandecasteele@lin.vlaanderen.be, Tel. 00 32 54 437136, Fax. 00 32 54 436160

Abstract

For several decades, periodical dredging of river sediments has been necessary to allow for shipping traffic on the river Scheldt. Sediments were disposed along the shores in the alluvial plain without concern for the potential presence of contaminants. The aim of this study was to survey the alluvial plains of the Upper Scheldt river in Belgium for the presence of old dredged sediment landfills, and to appraise the heavy metal contamination at these sites. Up to 82% of the areas that were affected by dredged sediment disposal was found to be polluted by at least one of the metals Cd, Cr, Zn or Pb. Concentrations of Cd, Cr and Zn were, in 10% of the cases, higher than 26, 1900 and 2800 mg/kg, respectively. Cu and Ni concentrations were of no environmental concern on any site. Trends in metal concentrations as a function of location and time were explored and discussed. The highest average concentrations of Cd, Cr, Cu, Pb and Zn were found in the most downstream quarter of the Upper Scheldt. Contents of Pb and Cu were significantly lower for sediments disposed after 1965, but no indication for improvement of the sediment quality with time was observed for Cd, Cr and Zn. The pollution levels encountered warrant for caution as most of the soils affected by historical dredged sediment disposal are currently in use for agriculture, nature development or forestry.

Keywords: Heavy metals, dredged sediments, Scheldt

1. Introduction

For several decades, periodical dredging of river sediments has been necessary to allow for shipping traffic on the river Scheldt. Since the nineties dredged sediments have been disposed in confined disposal facilities. Previously, sediments were disposed along the shores in the

alluvial plain without concern for the potential presence of contaminants. An important change in the Scheldt river system was affected by the broadening and deepening of the river to allow for ship traffic up to 2000 tons. These works were executed between 1960 and 1980. The river profile had to be broadened and deepened, leading to a changed sedimentation pattern. The resulting dredged pure sandy soil materials were used for levelling the wet parts of the alluvial plain.

With respect to the river Scheldt, much research has primarily focussed on the river estuary downstream Rupelmonde. Studies about the upstream part in contrast are scarce, although most of the heavy metal input in the estuary is caused by upstream pollution (Regnier & Wollast, 1993). In suspended matter and bottom sediments of the Scheldt Estuary, Verlaan *et al*. (1998) and Verlaan (2000) could distinguish between a fraction of fluvial origin and a fraction of marine origin because of the elevated concentrations of Cr, Pb, Zn, Cu and Cd in the former. Both water quality, suspended matter quality and sediment quality is improving in the Scheldt estuary since two decades (Zwolsman, 1999; Zwolsman *et al*., 1996, Baeyens, 1998). Zwolsman (1999) states that improvement of the water quality eventually will lead to an increased mobilisation of heavy metals in the estuary because of an increased oxygen supply to the sediment.

Important sources of heavy metal pollution in the catchment of the river Scheldt are the transboundary industrial fluxes from Northern France. Transboundary influx of polluted water is screened yearly at the Flemish border (VMM, 1997 ;VMM, 2000a). Approximately 90 % of Cd and Cr in the Flemish surface water are estimated to originate from transboundary pollution (VMM, 2000b). Seuntjens *et al.* (1996) developed an integrated sediment quality assessment tool for the Flemish region. The results of this so-called triad approach revealed that the quality of sediments in the upstream part of the Scheldt in Flanders was very bad based on biological, ecotoxicological and physicochemical criteria (Ministerie van de Vlaamse Gemeenschap, 1995; De Cooman *et al.*, 1998, De Deckere *et al*., 2000).

Research concerning the geochemistry of heavy metals in soils derived from landfilled dredged sediments is a recent topic and shows that adverse effects of heavy metals depend upon the field conditions, of which redox potential and pH are of prime importance. Tack *et al.* (1996) showed that the solubility of Cd, Cu, Pb and Zn as a function of pH increased strongly in an oxidising environment compared to the reduced environment. Metal mobility and availability in selected upland disposal sites in Flanders has been studied in considerable detail (Singh *et al.*, 1996; Singh *et al.,* 1998; Tack *et al.*, 1998; Tack *et al*., 1999). Sequential extraction revealed low residual fractions compared to total contents. This may reflect an important anthropogenic input of metals in these dredged sediment-derived soils (Zhang *et al*., 1990; Vicente-Beckett, 1992). Based on DTPA extraction, Zn, Cd and Cu were estimated to be highly plant-available (Singh *et al.,* 1998). Elevated heavy metal concentrations in the pore water equally raised concerns for enhanced metal availability for plant uptake (Tack *et al.*, 1998). In contrast, long-term leaching and migration of metals to ground water predicted from leaching tests was estimated to be of no environmental concern (Tack *et al*., 1999). Field trials where polluted dredged sediment was applied as a topsoil revealed no migration of metals or organic pollutants 16 months after application (Ruban *et al.*, 1998). However, depending on the geometry of the area, metal transport from the sediment disposal site to surrounding areas by surface runoff may be of concern (Singh *et al.*, 2000).

Several approaches were used to evaluate sediment quality as a function of time. Geochronology was determined by comparison of sediment analyses from different periods (Zwolsman *et al*., 1996), by the use of radionuclide time tracers (Winkels *et al*., 1998; Callender & Van Metre, 1997; Zwolsman *et al*., 1993) or by calculation of sediment deposition rates on sampled points based on time series of monthly surveys (Wiese *et al*.,

1997). An advantage of the use of deposited sediments is that they integrate the fluctuating input of heavy metals over rather long time scales (Regnier & Wollast, 1993).

To estimate the geographical extent and the environmental consequences of historical sediment disposal on land along inland water rivers in Flanders, a detailed survey was initiated in 1997 on behalf of the Flemish Authorities. The primary goal of our study was to survey the alluvial plains of the Upper Scheldt for the presence of old dredged sediment landfills, and to measure and appraise the heavy metal contamination at these sites. Besides, we made an attempt to study the evolution of heavy metal concentrations in these dredged sediment-derived soils as a function of the period of landfilling and the location of the landfill from the data.

2. Materials and methods

Study area

The study area for this research was the Flemish part of the Upper Scheldt (Fig. 1). The catchment area of the river Scheldt covers about 21.600 km² in the north-west of France, the west of Belgium and the south-western part of the Netherlands. The river can be divided in three parts, the Upper Scheldt, the Scheldt river and the Scheldt estuary, which in turn is subdivided in the fluvial, the upper and the lower estuary. The Upper Scheldt is defined as the upstream part of the river which is not submitted to tidal influence. It comprises the section between the source in Saint-Quentin (France) and the city of Ghent. The Scheldt river, between Ghent and Rupelmonde, is the freshwater zone subjected to tidal influence. The estuary is characterised by an increasing salt content towards the North Sea.

Several data sources were used to collect information about the (potential) locations and the period of construction of historical dredged sediment landfills and about former dredging activities. These comprised the Soil Map of Belgium at a 1:20.000 scale (Geologisch Instituut, Gent), the archives of the Flemish Government Administration, and information obtained from comparing topographical and vegetation indications on the various editions of the topographical maps at a 1:25.000 scale (Nationaal Geografisch Instituut, Brussel). Based on these data, sites that were levelled up and thus potentially have been filled could be identified.

Recent dredged sediment landfills in the area were sampled in advance to develop an appropriate sampling strategy based on the knowledge of the landfill morphology (Fig. 2). The physical and chemical properties of the potential landfills were investigated by taking sample cores down to 2 m underneath the surface level. Sampling points were restricted to app. 1 point per ha. On dredged sediment landfills the sandy material is found close to the discharge openings while the silt is distributed relatively homogeneously in the horizontal plane over the remaining of the discharge site (van Driel & Nijssen, 1988). Therefore, samples were taken in the silty part of the landfill. Soil samples for analysis were taken from layers with high organic matter content, striking texture differences, anaerobic features or from superficial layers with exceptionally thick Ah/Ap-horizons. Also the underlying original soil layers were sampled.

The soil samples were dried at 40°C until constant weight and grounded to pass a 2 mm sieve using a mechanic grinder (Retsch, Haan, Germany). The co-ordinates of the sampled points were determined using differential GPS (TRIMBLE TDC2 and TSC1, Sunnyvale, CA).

Physical and chemical analysis

Soil pH and electrical conductivity (EC) were measured in a 1:5 soil to water suspension after stirring for 2 h. Organic carbon was determined by the method of Walkley-Black (Bremner & Jenkinson, 1960). $CaCO_3$ content was determined by back-titration with 0.5 M NaOH of the excess of H_2SO_4 added to 1 g air-dried sediment (Nelson, 1982). The grain size distribution of the soil samples was determined using laser diffractometry (Coulter LS200, Miami, FL). The clay fraction is defined as the 0-6 µm fraction. This fraction was found to correlate with the 0-2 µm fraction, determined using the conventional pipette method, except for soil samples with clay contents > 50 % (Fig. 3) (Vandecasteele *et al.*, submitted to European Journal of Soil Science). The maximum clay content measured in the soil samples from the dredged sediment landfills was below this figure (Table 4).

Soil total contents of Cd, Cr, Cu, Ni, Pb, Zn, P and S are aqua regia extractable contents measured with ICP-AES (Varian Liberty Series II, Varian, Palo Alto, CA), Aqua regia destruction was performed using microwave (Milestone 1200 MS Mega) with the following program: 250 W (5 min.), 400 W (5 min.), 600 W (5 min.), 800 W (10 min.), ventilation (10 min.). Quality control of analysis was based on multi-element standards (Merck 11355 ICP standard IV), and external and internal sediment standards. The accuracy of the method was checked by analysis of a reference sediment material (CRM 320: river sediment). Values (in mg/kg DM) obtained were for Cd: 0.53 (certified value: 0.533 ± 0.026) , Cu: 42.9 (certified value: 44.1 ± 1.0), Zn: 124.8 (value for aqua-regia extraction with ICP: 122), Cr: 81.7 (value for aqua-regia extraction with ICP: 79), Ni: 57.8 (value for aqua-regia extraction with ICP: 57), and Pb: 27.7 (value for aqua-regia extraction with ICP: 33). Maximum relative standard deviations on a fourfold analysis of 2 soil samples from different dredged sediment landfills (replicate analysis in 4 different months) were 6.7 % for Cd, 8.0 % for Cr, 3.5 % for Cu, 4.5 % for Ni, 5.2 % for Pb and 11.3 % for Zn.

Evaluation of pollution

Heavy metal content of the dredged sediment landfills was evaluated with reference to the Flemish Decree on Soil Sanitation (VLAREBO, 1996). Background concentration levels for the Flemish soils are defined as these concentrations commonly found in soils that are considered clean, and are a function of soil organic matter and clay content (Tack *et al*., 1997). The pollution criterion and the sanitation standard values for different land use classes were calculated based on risk assessment (Cornelis & Geuzens, 1995). Four classes of land use were distinguished: nature, forestry and agriculture (class 1+2), habitation (class 3), sports and recreation (class 4) and industry (class 5) (Table 1). A dredged sediment landfill was classified as polluted in this survey if in at least one place there was at least one layer where the pollution criterion for forestry, nature and agriculture was exceeded for one of the measured heavy metals.

Identification of dredged sediment landfills

In total, 682 samples from 592 sampling points were available. For many of the sites, their former use as a dredged sediment disposal could not be ascertained by historical records. Dredged sediment disposal sites were therefore identified based on field observations, comparative granulometric analyses and chemical analyses. Criteria were developed, based on

a comparison between reference data from 56 aerobic soil samples of areas positively identified as affected by dredged sediment disposal and 36 samples from undisturbed alluvial soils along the upper Scheldt. Full details about the procedure followed are given in Vandecasteele *et al*. (2000). $CaCO_3$ content was found to be a major distinguishing property between alluvial soils and dredged sediment landfills in the studied area. Dredged sediment landfills were characterised by a $CaCO_3$ content higher than 4 %. Other chemical properties that allowed to conclude that a location probably was affected by dredged sediment disposal were: OM (%) x layer thickness of Ap/Ah horizont (cm) > 180, S (g/kg DM) x layer thickness of Ap/Ah horizont (cm) > 26, EC > 400 µS/cm, C:S ratio < 30, C:P ratio < 10. Using these criteria, 162 sampling points could be identified that likely have affected by dredged sediment disposal.

Dataset

Data from both aerobic and anaerobic soil samples were available (162 sampling points and 211 samples). Only the aerobic dredged sediment-derived soil samples (137 samples from 117 sampling points) were used for studying of the measured concentrations of heavy metals in relation to location along the river Scheldt and time of landfilling.

For each sampled site, the position relative to the river and the time when the landfill was constructed were recorded. Four sections (Fig. 4) were delineated such that the total landfill area was approximately equally distributed over the sections. The section limits were the N60 bridge in Oudenaarde (km 26 from the Flemish border crossing), the Teirlinck old river meander (km 50) and the Ring canal crossing in Zwijnaarde (km 59). The construction periods of the landfills also were categorised into 2 periods, (1) constructed before 1965, and (2) constructed after 1965. The distribution of the samples over periods and river sections is summarised in Table 2.

Statistics

Statistical processing started with an explorative correlation analysis to detect linear relations in the dataset. No interactions between the factor ‘period of landfilling’ and ‘location of the landfill’ was observed by analysis of variance (ANOVA), which implies that both factors can be evaluated separately. First ANOVA for the factor “location of the landfill” was performed (Mathsoft, 1999). Multiple comparison of means was performed according to the Sidak method with 95 % simultaneous confidence intervals (Mathsoft, 1999). This is a very conservative method which also allows for comparison of groups with a different number of elements and in which every pair of factor values is evaluated separately. Differences between the samples from sites landfilled before and after 1965 were compared using the two-sample t-test (Mathsoft, 1999). Sample data were both processed unweighed and with the use of a weight factor to account for the sediment mass represented by a sample as calculated from the area and layer thickness of the sampled site. It was found that both approaches led to similar conclusions. Significance levels based on unweighed data are presented.

Results

Research in the archives of the Flemish community revealed that in the most upstream part of the study area small quantities of sediment were dredged on specific spots with high sedimentation rates such as sluices, and at the Kortrijk-Bossuit canal outfall. Only once, in 1966, larger quantities section were dredged from a larger section (Table 3). In the downstream part of the river, the situation was similar until 1968, but since 1980, when the calibration works for ship traffic up to 2000 tons were finished, huge amounts of sediment were dredged (Table 3). No files were available about dredging activities between 1968 and 1980. This fact can be explained by the river broadening works being executed in that period. Since 1980, dredging was performed in a more systematic way and larger sections were dredged in each operation.

In total 232 ha of dredged sediment landfills (83 sites) were sampled. Physico-chemical properties of these sites are shown in Table 4. 190 Ha of this area (82 %) was found to be polluted according to the criteria outlined before. Besides, 259 ha (71 sites) levelled up with pure soil material from calibration works were mapped. For another 144 ha (88 sites), the sites were not sampled because they were built-on, they were levelled up with other material (disposal sites) or no clear conclusions could be drawn based on soil chemical properties.

162 Sampled points were situated on dredged sediment disposal sites. On 139 points (86 %) the pollution criterion was exceeded for at least one of the measured heavy metals. The distribution of the measured concentrations of heavy metals over the sanitation standard values (Fig. 5) clearly demonstrates severe pollution with Cr, Cd and Zn. Pb is a minor contaminant, while Cu and Ni are of no environmental concern in the area. In 108 of the 139 sampled points (78 %) with soil pollution, Cr, Cd as well as Zn concentrations are above the sanitation reference values for soil sanitation.

The results of the survey revealed a distinct geographical fragmentation of the polluted dredged sediment landfills. The area of the landfills varied between 0.2 and more than 20 ha and averaged to 2.4 ha. For 73 % of the spots where pollution was found, the polluted sediment was on the surface. For the total area of sites with a superficial polluted sediment layer, the thickness of the polluted layer was between 0-25 cm for 11%, between 25-50 cm for 13%, and between 50-75 cm for 28% of the area. For 35 % of the area designated as contaminated, the thickness of the layer exceeded 1 meter. When the polluted dredged sediment landfill area is plotted against the relative position downstream along the Upper Scheldt (Fig. 6), it is observed that half of the polluted landfills are situated in the most downstream quarter of the study area. Three-quarter of the polluted sites are situated in the downstream half of the area. Three sites show a distinct clustering of landfills (Fig. 6): downstream from the outfall of the canal Kortrijk-Bossuit, at Oudenaarde and the whole downstream part from the Teirlinck old river meander. There is clearly no uniform geographical scatter of the landfills. Both the canal mouth and the Oudenaarde sluice are important sediment traps because of the reduced water velocity.

More than 1/3 of the area of polluted landfills was raised before 1965, while in the next 20 years more than 100 ha was landfilled (Fig. 7). The average area for landfilling was 2.1 ha in the period before 1965, 2.9 ha between 1965 and 1975 and 9.2 ha between 1975 and 1985. After 1985, one single landfill of 13 ha was constructed in the study area.

Correlation analysis is an indicative tool to detect linear trends in the data. An important feature in the data of the aerobic sediment landfills was the strong positive correlation between Cd, Cr and P. There was also a high positive correlation between Zn on one hand and Cd and the clay content on the other hand (Table 5). No significant correlations were

observed between heavy metal concentrations and the soil properties used to identify dredged sediment landfills (see Materials and Methods section), and between the relative position of the sampled points and the soil properties.

Trends in the heavy metal contents of the sediment landfills as a function of location were studied. The river was divided in four sections (Figure 4). ANOVA revealed that the location of the landfill was a significant factor for all heavy metals ($p < 0.001$) in determining metal concentrations in the sediment-derived surface soils (Table 6). From the multiple comparison of means it is concluded that for Cd, Cr and Zn, the major pollutants in the area, the landfills in the 2 downstream sections show significantly higher concentrations than in the other sections. For Pb, the results were similar, except that the difference between section 1 and 3 was not significant.

A similar evaluation was made of the chemical and physical soil parameters. Significant differences were detected for sand content ($p < 0.05$), silt content, S, $CaCO_3$ and EC ($p < 0.001$). Silt and $CaCO_3$ content were significantly lower in the most downstream section, while S en EC were higher (Table 7). The latter parameter gives an indication for tidal influences on the landfilled sediment.

Temporal trends in heavy metal contents of the dredged sediment landfills were assessed by comparison between 2 periods of landfilling, before 1965 and after 1965. From ANOVA it is concluded that measured concentrations of Cd, Cr, Zn and Ni were not significantly different between these periods, while levels of Cu and Pb are significantly lower ($p < 0.001$) in the sites landfilled after 1965 (Table 8). For the chemical and physical soil parameters silt ($p < 0.001$) and organic matter ($p < 0.05$) content decreased significantly, while P and $CaCO_3$ concentrations increased since 1965 ($p < 0.001$).

Discussion

This survey gives important information about the pollution of the terrestrial compartment of the Upper Scheldt river system which is connected to pollution of the water body through dredging activities. Landfilling of dredged sediments caused a pollution with a more permanent character than the sediment pollution itself. When sediments settle and accumulate on locations with reduced current, reduction processes effectively immobilise metals in the sediments (Gambrell, 1994), thus decreasing heavy metal bio-availability. The upper layers of the sediment are subjected to a limited resuspension and may release pollutants to the water column (Tessier *et al.*, 1989; Boughriet *et al.*, 1992). Resuspended matter transported in the Scheldt river will be trapped in the high turbitity zone around Antwerp (Verlaan, 2000; Regnier & Wollast, 1993). This area is dredged yearly and the sediment is disposed in overdepths or on-land at confined disposal sites.

Landfilled calcareous dredged sediments with high OM and clay contents are characterised by a very low leaching potential (Tack *et al.*, 1999) but may exhibit an appreciable bio-availability of the heavy metals (Singh et al., 1998; Tack *et al.*, 1999; Stephens *et al.*, 2001). A significant decline in heavy metal concentrations in these terrestric parts of the river alluvium is therefore not expected to occur within decades, while in contrast further improvement of the aquatic part of the ecosystem can be expected. Beyer & Stafford (1993) calculated that bio-concentration of Cd by earthworms is a risk for higher trophic levels in the foodweb when concentrations in dredged sediment-derived soils exceed 10 mg Cd/kg DM. This concentration is exceeded in the superficial layer of the dredged sediment landfills along the Upper Scheldt in more than half of the sampled disposal sites.

Dredged sediment landfills are mostly concentrated in the downstream section of the Upper Scheldt. These landfills are characterised by the highest heavy metal concentrations in the study area. Nevertheless, heavy metal contamination of the river is mainly due to transboundary influx. This pollution thus is to an important extent transported to the most downstream section of the Upper Scheldt. A combination of factors, higher sedimentation rates and more opportunities for the construction of landfills, gave raise to more landfilling in the downstream section.

Conclusions with respect to changes in sediment quality with time from the current data are valid provided that total concentrations were not significantly altered since disposal. Acidification is a potential process that may lead to a strong mobilisation and leaching of heavy metals in oxidising sediments (Gambrell *et al*, 1991). Dredged sediment-derived soils along the river Scheldt are characterised by appreciable $CaCO_3$-levels (>4 %). Significant leaching of metals thus did not occur and it can be assumed that metal contents in the dredged sediment-derived soils reflects metal contents in the Scheldt river at the time of dredging.

Interpretation of results to detect temporal trends was hampered because of the unbalanced distribution of samples over the selected periods. Only for Cu and Pb a distinct decline in concentrations since 1965 was revealed. There was also a decrease in organic matter and silt content. Both Pb and Cu are known to show a great affinity to organic matter. Data on historical organic matter content of sediments in the Scheldt is lacking, but Wollast (1988) calculated that in 1978 2/3 of the fluvial suspended matter in the Scheldt was related to antropogenic activities. It was characterised by organic matter contents of 40 to 60 %, while the current organic matter content in the suspended matter is 10-15 % (Wollast, 1988). Zwolsman (1999) calculated the average OM content of the fluvial suspended matter in 1987-1988 to be 14 %. Dredged sediments of the river Scheldt are characterised by an initially very high organic matter content compared to normal contents in upland soils in this region. Even in the Scheldt estuary, the organic fraction of fluvial material is considerably higher than in marine material (Verlaan *et al*., 1998). A consequence of landfilling is a decrease in organic matter contents with time which is strongly influenced by landuse (Singh *et al*., 1998; Ruban *et al.*, 1998). Ruban *et al.* (1998) observed a 35 % decline after 28 months in landfilled dredged sediment with an initial high organic matter content (16 %). Despite the breakdown of organic matter which probably already took place since the period of landfilling, it was found in this study that the organic matter content in the dredged sediment landfills constructed before 1965 is higher than in the landfills constructed afterwards (Table 8). This finding is in accordance with the trend of the decreasing organic matter content of the fluvial suspended matter reported by Wollast (1988).

At least, for Cd, Cr and Zn it can be concluded that concentrations in the dredged sediment were very high in the period before 1965 and that no obvious quality improvement was observed in the period after 1965. Extrapolation of the quality of dredged sediments to that of the river sediment is however very complex. Zwolsman *et al.* (1996) found a distinct improvement in sediment and suspended matter quality in the Scheldt estuary, especially in the most polluted fluvial estuary. In the suspended matter, the authors found decreasing concentrations of Zn since 1972, a peak of Cd around 1980 and a peak of Cr between 1970 and 1975, followed by a drastic decline for both elements. These trends could not be confirmed or denied for the Upper Scheldt since the data only allowed for a reliable comparison between the situation before and after 1965. Results of the spatial trend of heavy metal contamination in landfilled dredged sediment must not be translated straightforward to the quality of the river sediment. Recent landfills were used for disposal of dredged sediments from different locations and larger sections. The relation between sediment quality before and after dredging and landfilling is also not known.

Conclusions

Dredged and landfilled Upper Scheldt sediments were strongly polluted with Cr, Zn, Cd and Pb a long time before 1965 and no clear quality improvement is observed since. Landfilling of heavy metal contaminated dredged sediments caused an important contamination in the terrestrial part of the alluvial system. The ecological restoration of these dredged sediment-derived soils will be a time-consuming process. However, the presence of dredged sediment landfills in the alluvial plain is an indirect consequence of polluted sediments in the river Scheldt which is easily forgotten. Further efforts to reduce inputs in the river bodies remains a priority. While dredging activities and natural processes of sorption/desorption, suspension and flocculation may eventually lead to a gradual improvement in the river sediment quality, pollution of these terrestrial sites with heavy metals has a more permanent character.

Acknowledgements

This project was carried out with financial support from the Waterways and Marine Affairs Administration (AWZ) of the Ministry of the Flemish Community. We are grateful to Carine Buysse, Els Mencke, Anya Derop and Athanaska Verhelst for the accurate soil analyses and to Rik Delameillieure for the help during the field survey. Thanks also to Raf Lauriks and Jürgen Samyn for the critical review and to Paul Quataert for the statistical support.

References

Baeyens W. 1998. Evolution of trace metal concentrations in the Scheldt estuary (1978-1995). A comparison with estuarine and ocean levels. Hydrobiologia 366, 157-167.

Beyer WN, Stafford C. 1993. Survey and evaluation of contaminants in earthworms and in soils derived from dredged material at confined disposal facilities in the great lakes region. Environmental monitoring and assessment 24(2), 151-165.

Boughriet A, Ouddane B, Fischer JC, Wartel M, Leman G. 1992. Variability of dissolved Mn and Zn in the Seine estuary and chemical speciation of these metals in suspended matter. Water Res., 26, 1359-1378.

Bremner JM, Jenkinson DS. 1960. Determination of organic carbon in Soil. 1. Oxidation by dichromate of organic matter in soil and plant materials. Journal of Soil Sciences 11, 394-402.

Cornelis C, Geuzens P. 1995. Voorstel tot normering van bodemverontreiniging door zware metalen en metalloïden. Studie uitgevoerd in opdracht van de OVAM. VITO/MIE/DI/95-03. Afdeling Leefmilieu, april 1995.

De Cooman W, Florus M, Devroede-Vander Linden MP. 1998. Karakterisatie van de bodems van de Vlaamse onbevaarbare waterlopen. AMINAL Afdeling Water, Brussel.

De Deckere E, De Cooman W, Florus M, Devroede-Vander Linden MP. 2000. Karakterisatie van de bodems van de Vlaamse bevaarbare waterlopen. AMINAL Afdeling Water, Brussel.

Gambrell RP, DeLaune RD, Patrick,WH Jr. 1991. Redox processes in soils following oxygen depletion. In: Plant life under oxygen deprivation (eds M.B. Jackson *et al.*), pp. 101-117. SPB Academic Publishing, The Hague, The Netherlands.

Gambrell RP. 1994. Trace and toxic metals in wetlands - a review. Journal of

Environmental Quality, 23, 883-891.
Mathsoft. 1999. S PLUS 2000. Modern Statistics and Advanced Graphics. Guide to Statistics. Data analysis products division. Mathsoft, Seattle.
Ministerie van de Vlaamse Gemeenschap. 1995. Methodologische studie naar de inventarisatie, de ecologische effecten en de saneringsmogelijkheden van de bodems van de Vlaamse waterlopen. Eindrapport – deel evaluatie.
Nelson RE. 1982. Carbonate and gypsum. In: Methods of soil analysis, Part 2. Chemical and microbiological properties, 2nd edn (eds A.L. Page *et al.*), pp. 181-197. ASA, Madison, Wisconsin.
Regnier P, Wollast R. 1993. Distribution of trace metals in suspended matter of the Scheldt. Marine Chemistry 43, 3-19.
Ruban V, Parlanti E, Riffé C, Amblès A, Six P, Jambu P. 1998. Migration of micropollutants in a dredging amended soil in Northern France. Agrochimica 42 (1-2), 59-71.
Satawathananont S, Patrick WHJr, Moore PAJr. 1991. Effect of controlled redox conditions on metal solubility in acid sulfate soils. Plant and Soil, 133, 281-290.
Seuntjens P, De Cooman W, Verheyen R. 1996. Development of a contaminated sediment assessment strategy in Flanders. CATS III, Oostende, 18-20 maart 1996.
Singh SP, Tack FMG, Verloo M. 1996. Extractability and bioavailability of heavy metals in dredged sediment derived surface soils. Chemical Speciation and Bioavailability, 8, 105-110.
Singh SP, Tack FM, Verloo MG. 1998. Heavy metal fractionation and extractability in dredged sediment derived surface soils. Water, Air, and Soil Pollution 102, 313-328.
Singh SP, Tack FMG, Gabriels D, Verloo M. 2000. Heavy metal transport from dredged sediment derived surface soils in a laboratory rainfall simulation experiment. Water, Air and Soil Pollution, 118, 73-86.
Stephens SR, Alloway BJ, Carter JE, Parker A. 2001. Towards the characterisation of heavy metals in dredged canal sediments and an appreciation of 'availability': two examples from the UK. Environ. Pollut. 113[3], 395-401. 2001/8.
Tack FMG, Callewaert OWJJ, Verloo MG. 1996. Metal solubility as a function of pH in a contaminated, dredged sediment affected by oxidation. Environmental Pollution, 91 (2), 199-208.
Tack FMG, Verloo MG, Vanmechelen L, Van Ranst E. 1997. Baseline concentration levels of trace elements as a function of clay and organic carbon contents in soils in Flanders (Belgium). The Science Of The Total Environment, 201, 113-123.
Tack FMG, Singh SP, Verloo MG. 1998. Heavy metal concentrations in consecutive saturation extracts of dredged sediment derived surface soils. Environmental Pollution, 103, 109-115.
Tack FMG, Singh SP, Verloo MG. 1999. Leaching behaviour of Cd, Cu, Pb and Zn in surface soils derived from dredged sediments. Environmental Pollution, 106, 107-114.
Tessier A, Carignan R, Dubreuil B, Rapin, F. 1989. Partitioning of zinc between the water column and the oxic sediments in lakes. Geochim. Cosmochim. Acta, 53, 1511-1522.
Vandecasteele B, De Vos B, Lauriks R, Buysse C, Mergaert K. 2000. Baggergronden in Vlaanderen. Resultaten van de terreininventarisatie naar geografische omvang en verontreiniging. Mededelingen van het Instituut voor Bosbouw en Wildbeheer 2000-1. IBW, Geraardsbergen
Vandecasteele B, De Vos B, Van Meirvenne M. (submitted). Relationship between soil textural fractions determined by the sieve-pipette method and laser diffractometry. Submitted to European Journal of Soil Science.
van Driel W, Nijssen JPJ. Development of Dredged Material Disposal Sites: Implications for Soil, Flora and Food Quality. In: Salomons W, Förstner U (Eds.) Chemistry and biology of solid waste. Dredged material and mine tailings. Berlin: Springer-Verlag, 1988.

Verlaan PAJ. 2000. Marine vs Fluvial Bottom Mud in the Scheldt Estuary. Estuarine, Coastal and Shelf Science 50, 627-638.
Verlaan PAJ, Donze M, Kuik P. 1998. Marine vs Fluvial Suspended Matter in the Scheldt Estuary. Estuarine, Coastal and Shelf Science 46, 873-883.
Vicente-Beckett VA. 1992. Trace metal levels and speciation in sediments of some Philippine natural waters. The Science Of The Total Environment, 125, 345-357.
VLAREBO. 1996. Besluit van de Vlaamse regering houdende vaststelling van het Vlaams reglement betreffende de bodemsanering. Belgisch Staatsblad, 27 maart 1996.
VMM. 1997. Overzicht van de Inkomende Grensoverschrijdende Vuilvrachten in Vlaanderen 1994 en 1995. VMM, Erembodegem, Begium. 218 p.
VMM. 2000a. Overzicht van de Inkomende Grensoverschrijdende Vuilvrachten in Vlaanderen 1997-1998. VMM, Erembodegem, Begium. 164 p.
VMM. 2000b. MIRA-S 2000 Milieu- en natuurrapport Vlaanderen: scenario's. Garant, Leuven-Apeldoorn. 637 p.
Wiese SBO, MacLeod CL, Lester JN. 1997. A Recent History of Metal Accumulation in the Sediments of the Thames Estuary, United Kingdom. Estuaries 20(3), 483-493.
Winkels HJ, Kroonenberg SB, Lychagin MY, Marin G, Rusakov GV, Kasimov NS. 1998. Geochronology of priority pollutants in sedimentation zones of the Volga and Danube delta in comparison with the Rhine delta. Applied Geochemistry 13(5), 581-591.
Wollast R. 1988. The Scheldt Estuary. In W. Salomons et al. (eds)., Pollution of the North Sea: an assessment. Springer-Verlag, Berlin: 183-193.
Zhang JG, Huang WW, Wang Q. 1990. Concentration and partitioning of particulate trace metals in the Changjiang. Water, Air and Soil Pollution, 52, 57-70.
Zwolsman JJG. 1999. Geochemistry of trace metals in the Scheldt estuary. Geologica ultraiectina 177. Universiteit Utrecht,Faculteit aardwetenschappen, Utrecht.
Zwolsman JJG, Berger GW, Van Eck GTM. 1993. Historical input, mobility and retention of major elements and trace metals in salt marshes of the Scheldt estuary. Marine Chemistry 44, 73-94.
Zwolsman JJG, Van Eck GTM, Burger G. 1996. Spatial and temporal distribution of trace metals in sediments of the Scheldt estuary. Estuarine, Coastal and Shelf science 43, 55-79.

Table 1. The pollution criterion and the sanitation standard values of the Flemish Decree on SoilRecovery for 2 extreme soil types (clay content: 2 and 50%, organic matter content: 1 and 20%) as a function of land use

	Cd	**Cr**	**Zn**	**Pb**	**Cu**	**Ni**
background concentration level	0.5 - 2.9	32 - 61	50 - 148	35 - 93	14 - 29	7 - 22
pollution criterion for agriculture and nature	1 - 5.8	89 - 171	385 - 1145	140 - 370	134 - 273	61 - 198
sanitation standard value for agriculture and nature	1.2 - 7.2	111 - 214	481 - 1432	175 - 463	168 - 341	77 - 247
sanitation standard value for habitation	3.6 - 21.8	256 - 495	802 - 2386	614 - 1621	336 - 682	361 - 1162
sanitation standard value for recreation	9 - 54.4	427 - 824	802 - 2386	1315 - 3473	420 - 853	423 - 1360
sanitation standard value for industry	18 - 108.8	683 - 1319	2405 - 7159	2192 - 5788	673 - 1365	538 - 1731

Table 2. Number of aerobic samples from dredged sediment landfills per time period and section

period	section 1	section 2	section 3	section 4	total
< 1965	29	6	16	21	72
> 1965	0	30	20	10	60
unknown	1	0	4	0	5
total	30	36	40	31	137

Sections: 1: from the Flemish border crossing to the N60 Bridge in Oudenaarde; 2: from the N60 Bridge in Oudenaarde to the Teirlinck old river meander; 3: from the Teirlinck old river meander to the Ring canal in Zwijnaarde, 4: from the Ring canal to the center of the city Ghent

Table 3. Amounts of dredged material as a function of time in the upstream and downstream part of thebridge of Kerkhove

dredged amounts (in 10^3 m^3) in the upstream part of Kerkhove bridge
1933 (2.2); 1934 (1.9); 1935 (1.9); 1936 (12.6); 1939 (3.1); 1941 (17.8);
1942 (2.8); 1943 (12.4); 1946 (2.7); 1947 (16.7); 1952 (6.6); 1962 (6.0); 1966 (30.2)
dredged amounts (in 10^3 m^3) in the downstream part of Kerkhove bridge
1944 (4.1); 1945 (63.7); 1946 (36.0); 1957 (22.6); 1962 (29.8); 1963 (52.3);
1968 (23.9); 1980 (571.2); 1982 (306.8); 1983 (22.7); 1987 (157.7)

Table 4. Descriptive data for the landfilled dredged sediment samples

	mean	median	Stdev.	min.	10th perc.	90th perc.	Max.
Cd (mg/ kg DM)	11.9	14.5	10.5	0.5	1.3	26.6	47.2
Cr (mg/ kg DM)	732	1190	724	31	113	1901	2769
Cu (mg/ kg DM)	107	125	65	5	21	208	316
Ni (mg/ kg DM)	31	33	11	7	17	47	70
Pb (mg/ kg DM)	233	220	180	5	34	414	1281
Zn (mg/ kg DM)	1455	1881	984	63	216	2857	4224
%clay	32	38	7	14	23	42	47
%silt	52	47	8	22	41	59	67
%sand	16	14	13	1	3	34	64
P (g/ kg DM)	3.3	4.4	1.8	0.9	1.4	6.2	7.1
S (g/ kg DM)	2.6	2.8	1.9	0.9	1.1	4.9	12.8
N (g/ kg DM)	0.04	0.04	0.01	0.02	0.02	0.05	0.09
%$CaCO_3$	7.4	7.7	2.2	4.0	4.6	10.3	13.7
%OS	8.2	7.9	2.7	3.5	4.8	11.6	16.3
pH-H_2O	7.6	7.5	0.2	7.0	7.3	7.8	8.0
pH-$CaCl_2$	7.3	7.3	0.2	6.9	7.1	7.6	7.7
EC (µS/ cm)	680	759	619	139	190	1836	2290
dry matter (%)	71.1	67.7	12.1	55.4	63.6	78.7	85.2
%LOI	14.8	13.7	4.0	6.1	9.8	20.6	22.7

Table 5. Regression coefficients (R^2) for heavy metals, P and organic matter

elements	Regression coefficients	correlation
Cd - Cr	0,548****	+
Cd - Pb	0,809****	+
Cd - Zn	0,640****	+
Cr - P	0,824****	+
Cu - OM	0,337****	+
Cr - OM	0,072***	+
Cd - OM	0,181****	+
Ni - OM	0,200****	+
Pb - OM	0,265****	+
Zn - OM	0,385****	+

**** $p < 0.0001$, *** $p < 0.001$

Table 6. Average concentration of heavy metals (mg/kg dry soil) for the dredged sediment landfills as a function of location (river sections, see Figure 4). Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95 % level of significance)

	section 1	**section 2**	**section 3**	**section 4**
Cd	8.7 a	6.6 a	16.2 b	15.4 b
Cr	439 a	317 a	1015 b	1129 b
Cu	84 a	77 a	107 a	164 b
Ni	28 ab	26 a	36 c	33 bc
Pb	192 ab	121 a	284 bc	337 c
Zn	1098 a	738 a	1834 b	2143 b

Table 7. Average concentration of chemical en physical soil parameters for the dredged sediment landfills as a function of location (river sections, see Figure 4). Means that are not significantly different are denoted with the same letter (Sidak multiple comparison of means at the 95 % level of significance)

	section 1	**section 2**	**section 3**	**section 4**
clay (%)	30.8 a	33.6 a	33.3 a	30.7 a
silt (%)	55.4 b	52.1 b	53.5 b	47.1 a
sand (%)	13.8 ab	14.2 ab	13.2 a	22.2 b
S (g/ kg DM)	2.1 a	2.7 ab	2 a	3.7 b
OM (%)	7.8 a	7.9 a	8.7 a	8.4 a
$CaCO_3$ (%)	7.4 ab	8.6 b	6.9 b	6.7 a
EC (µS/ cm)	402 a	718 a	432 a	1148 b

Table 8. Average concentration of heavy metals and other soil characteristics for the dredged sediment landfills as a function of time. Only the significant differences are shown

	Cu (mg/ kg DM)	Pb (mg/ kg DM)	silt (%)	OM (%)	CaCO3 (%)	P (g/ kg DM)
before 1965	121	294	53.7	8.6	7.0	2.7
after 1965	92	161	49.7	7.6	8.1	4.0

Fig. 1. The Scheldt Estuary and the Upper Scheldt river with the study area indicated.

Fig. 2. Normal structure of a dredged sediment landfill in a horizontal en vertical view with indication of important physical features.

Fig. 3. Scatterplot of the pipette 0-2 μm fraction and the laser diffractometry 0-6 μm fraction. The 1:1 ratio line is also shown.

Fig. 4. The sections distinguished in the study area. Sections: 1: from the Flemish border crossing to the N60 Bridge in Oudenaarde; 2: from the N60 Bridge in Oudenaarde to the Teirlinck old river meander; 3: from the Teirlinck old river meander to the Ring canal in Zwijnaarde, 4: from the Ring canal to the center of the city Ghent. The area displayed on the map is the study area indicated on Fig. 1.

Fig. 5. Classification of the heavy metal concentrations of the samples of the dredged sediment landfills over the sanitation standard values of the Flemish Decree on Soil Recovery. The number of samples in each class is shown on the Y-axis.

Fig. 6. Cumulative distribution of polluted dredged sediment landfills as a function of the relative position along the Upper Scheldt.

Fig. 7. Distribution of the area and the number of polluted dredged sediment landfills over the 4 periods.

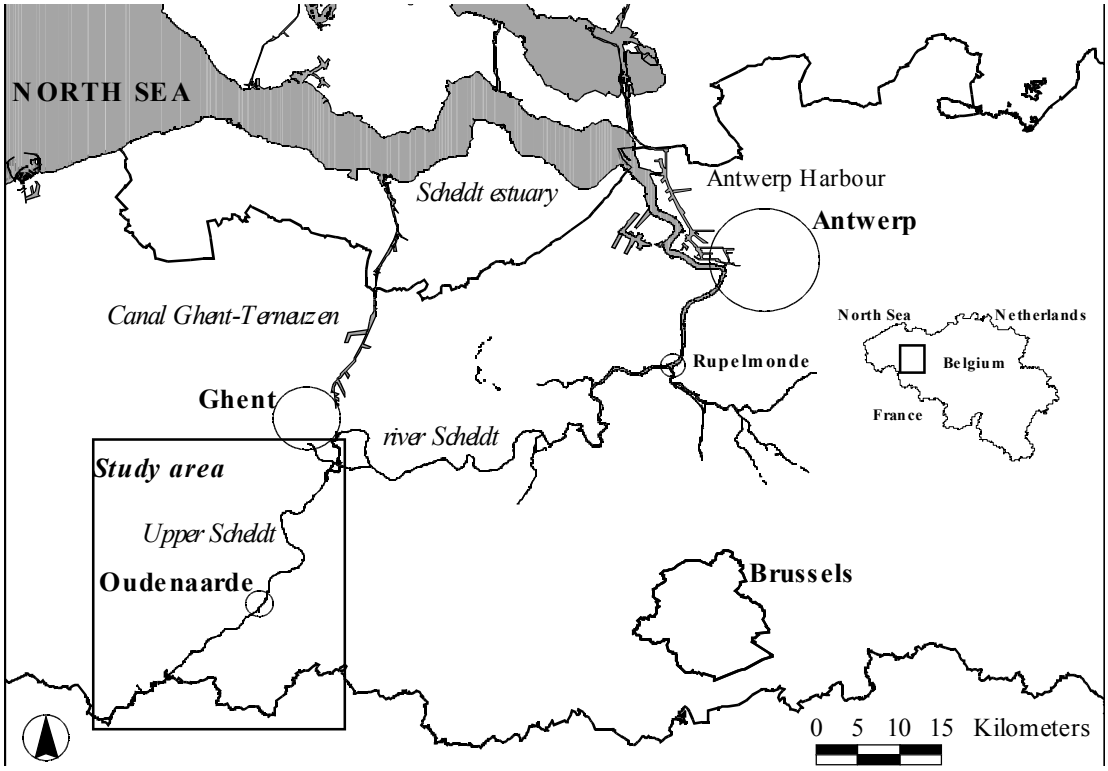
NORTH SEA
Scheldt estuary
Antwerp Harbour
Antwerp
Canal Ghent-Terneuzen
North Sea
Netherlands
Belgium
France
Rupelmonde
Ghent
river Scheldt
Study area
Upper Scheldt
Oudenaarde
Brussels
0 5 10 15 Kilometers

Fig.1.

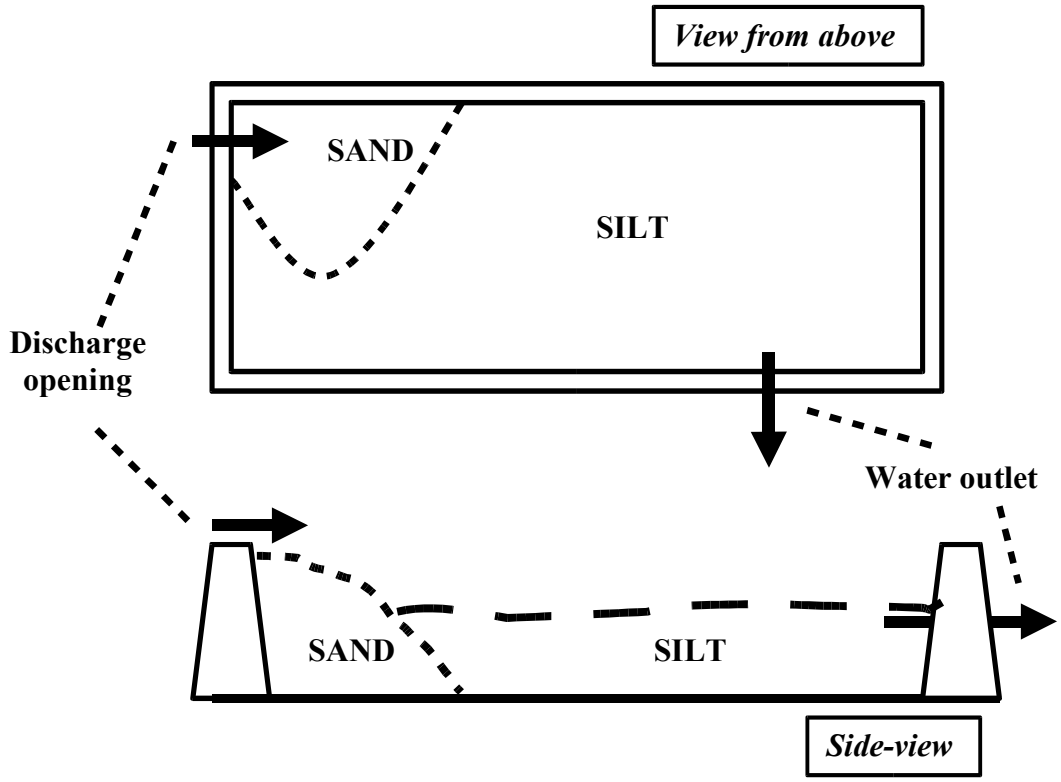
View from above
SAND
SILT
Discharge opening
Water outlet
SAND
SILT
Side-view

Fig. 2

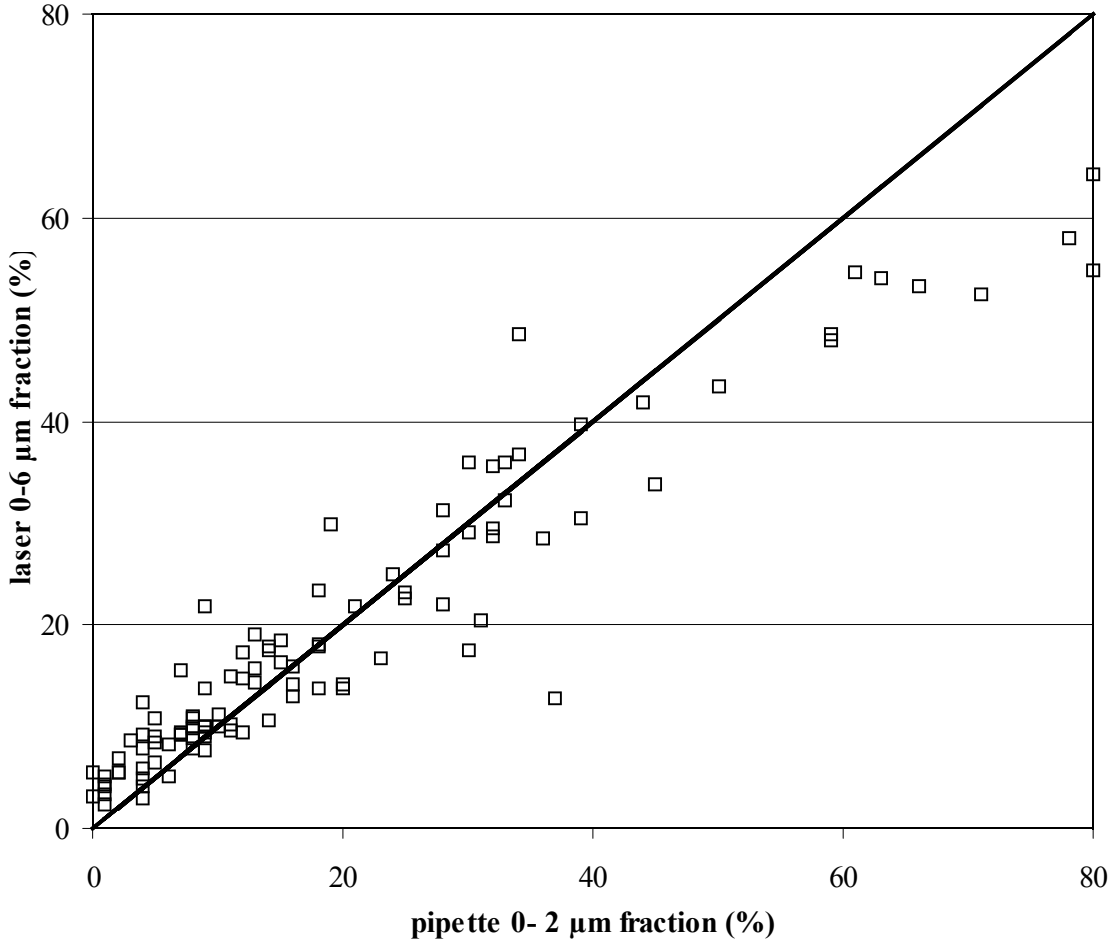
80
60
40
20
0
laser 0-6 µm fraction (%)
0
20
40
60
80
pipette 0- 2 µm fraction (%)

Fig. 3.

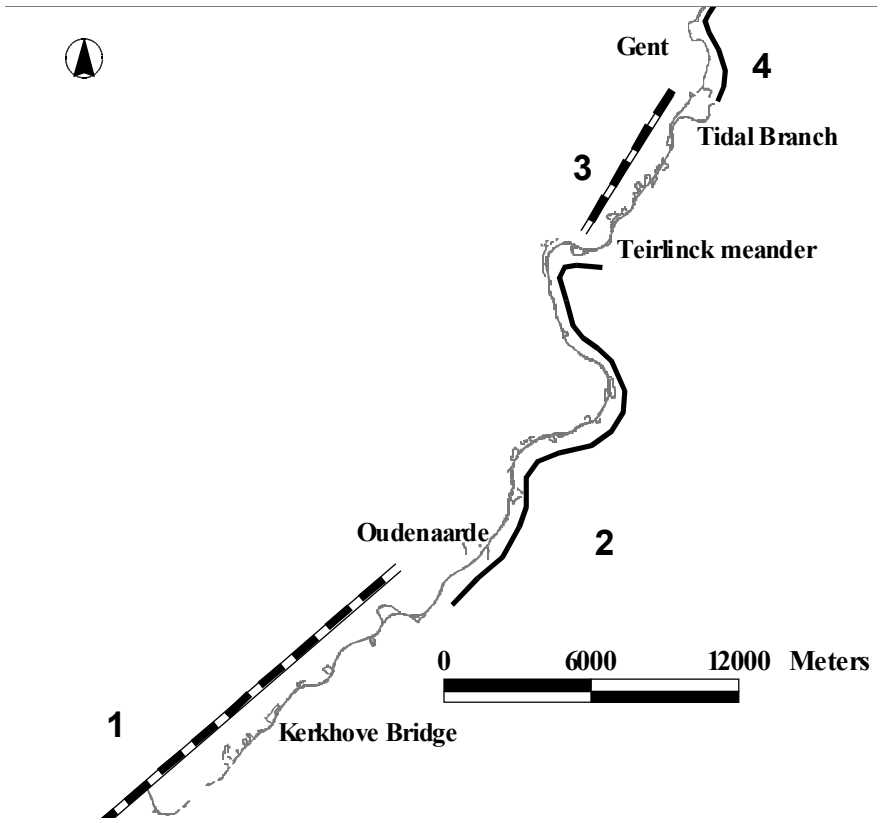
Gent
4
Tidal Branch
3
Teirlinck meander
Oudenaarde
2
0
6000
12000
Meters
1
Kerkhove Bridge

Fig. 4.

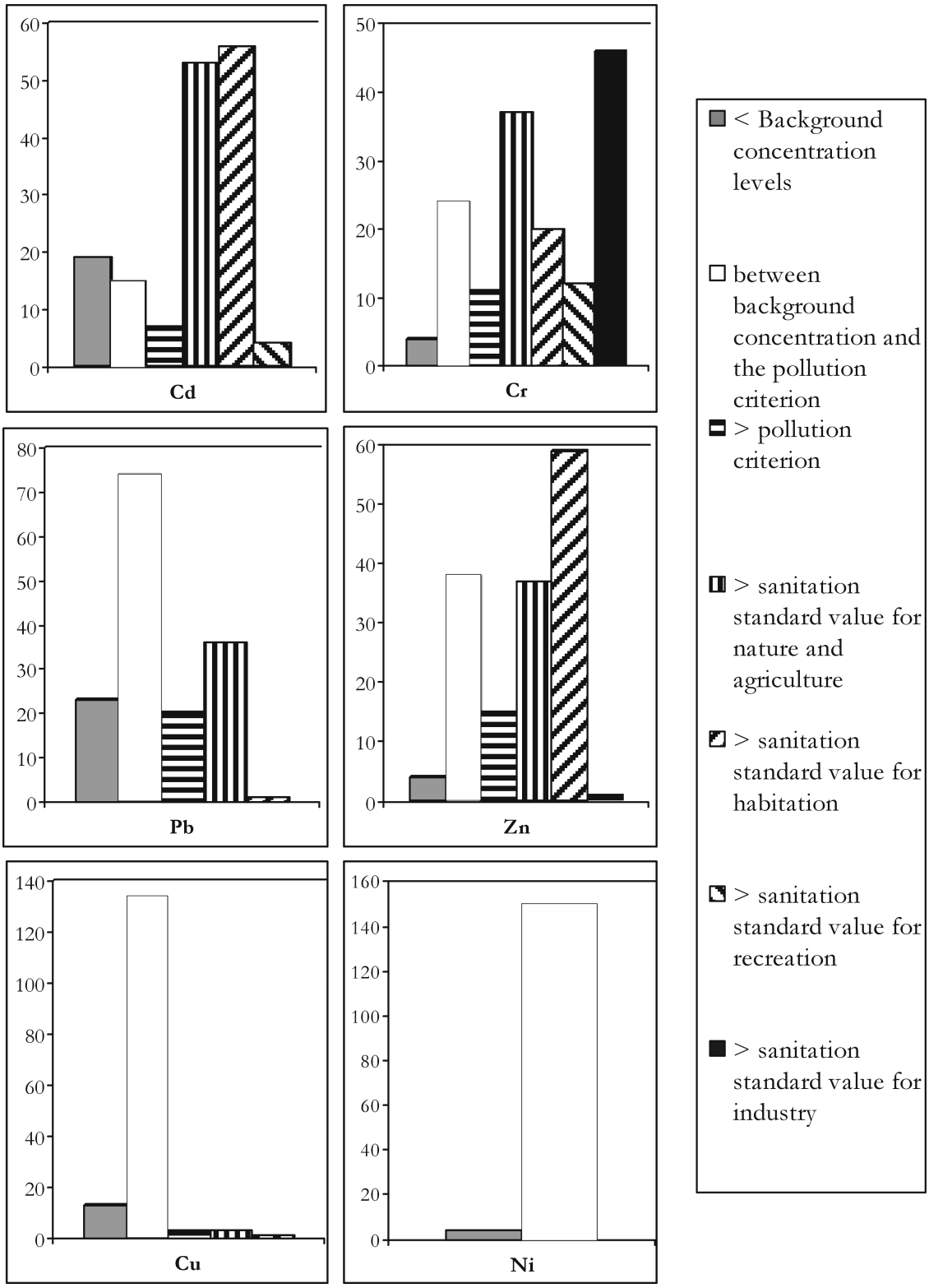
Cd
Cr
Pb
Zn
Cu
Ni
< Background concentration levels
between background concentration and the pollution criterion
> pollution criterion
> sanitation standard value for nature and agriculture
> sanitation standard value for habitation
> sanitation standard value for recreation
> sanitation standard value for industry

Fig. 5.

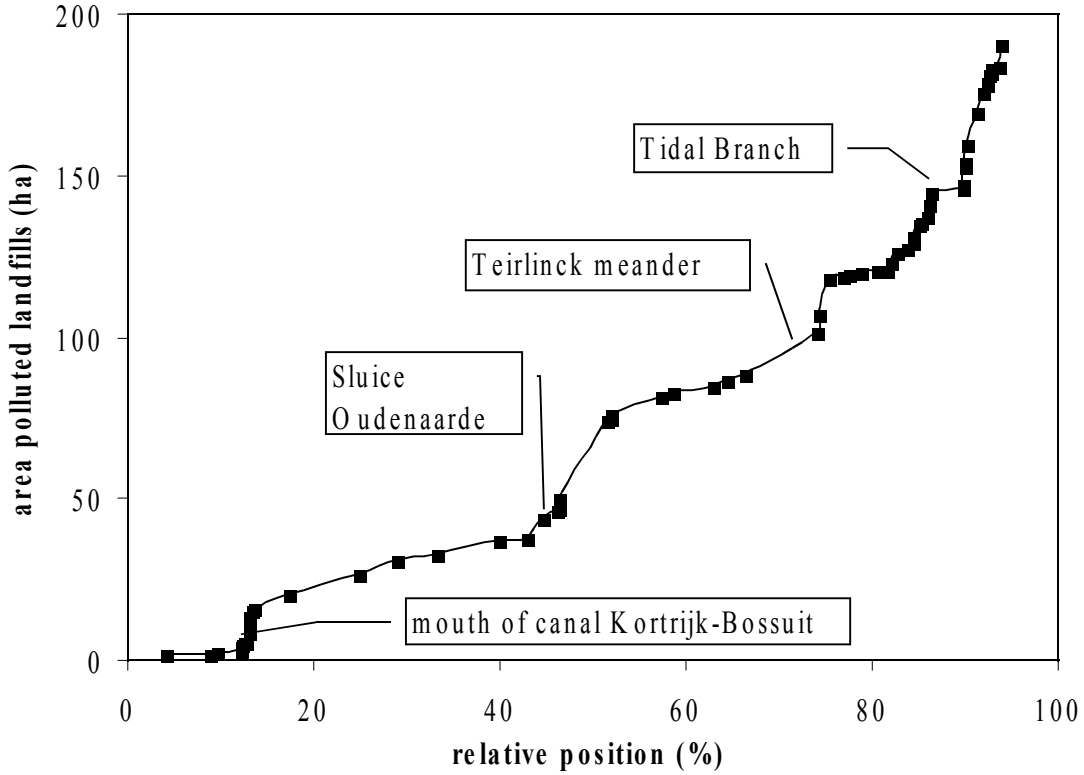
200
150
100
50
0
area polluted landfills (ha)
0
20
40
60
80
100
relative position (%)
Tidal Branch
Teirlinck meander
Sluice
Oudenaarde
mouth of canal Kortrijk-Bossuit

Fig. 6.

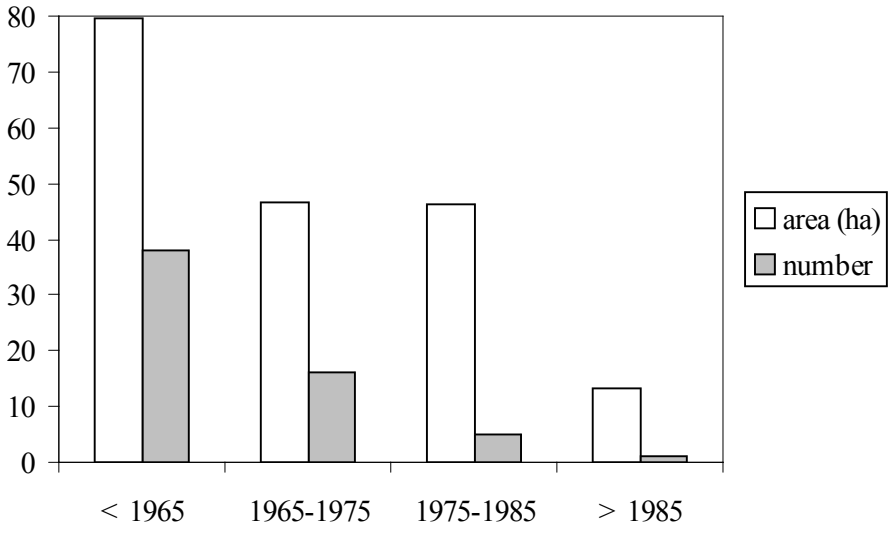
80
70
60
50
40
30
20
10
0
< 1965
1965-1975
1975-1985
> 1985
area (ha)
number

Fig. 7.