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Quality of dredged material in the River Seine basin (France). I. Physico-chemical properties

S. Carpentier*a, R. Moillerona, C. Beltranb, D. Hervéc, D. Thévenota

*aCereve, Faculté des Sciences et Technologie, Université Paris XII-Val de Marne, 61, avenue du Général de Gaulle, 94010 Créteil Cedex, France
bCETE Méditerranée, Service Hydraulique, BP 37000, 13791 Aix en Provence Cedex 3, France
cServices de Navigation de la Seine, île de la Loge, 78380 Bougival, France

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Abstract

In rivers, sediments are frequently accumulating persistent chemicals, especially for those that are more contaminated as a consequence of pressure related to environmental pollution and human activity. The Seine river basin (France) is heavily polluted from nearby industrial activities, and the urban expansion of Paris and its suburbs within the Île de France region and the sediments present in the Seine river basin are contaminated. To ensure safe, navigable waters, rivers and waterways must be dredged. In this paper, the quality of the sediment dredged in 1996, 1999 and 2000 is discussed. Physico-chemical characteristics of the sediment itself and of the pore-water are presented. Seine basin sediments show very diverse compositions depending on the sampling site. Nevertheless, a geographic distribution study illustrated that the Paris impact is far from being the only explanation to this diversity, the quality of this sediment is also of great concern. The sediment once dredged is transported via barges to a wet disposal site, where the dredged material is mixed with Seine water in order to be pumped into the receiving site. This sort of dumping might be responsible for the potential release of contaminants to the overlying water from the significantly contaminated sediments. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Dredged material; Sediment quality; Pore-water; River Seine basin

1. Introduction

Dredging rivers is necessary to maintain sufficient sailing depth (Hauge et al., 1998), to reduce the risk of flooding (Hakstege et al., 1998) or to increase outflow during dry periods (Ellery and Mc Carthy, 1998). Dredging is also a crucial operation for some engineering works (Vale et al., 1998). Dredging can also be used as a remedial treatment to improve the environmental quality of the river ecosystem (Besser et al., 1996; Winkels and Stein, 1997; Fioole et al., 1998). Increasing pressure from environmental pollution and human disturbance leads to water quality deterioration. With the influx of polluted water, contaminants are absorbed onto suspended particles and subse-
quently accumulated in the underlying sediment (Hauge et al., 1998; Lau and Chu, 1999).

Dredging methods vary depending on the volume, texture, water content and grain size of the sediments to be removed. Dredging technologies can be roughly divided into three main categories: mechanical, hydraulic and pneumatic dredges (Pollice et al., 1996; Norman et al., 1997). Mechanical dredges are used to remove bottom sediment through direct application of mechanical force. Once dredged, excavated sediment is generally placed into a barge for transportation to the disposal site. Such dredges can be considered as the most cost effective, especially when the dredging site is far from the disposal site, even if leakages of fine-grained material occur when the bucket is lifted from the river. Hydraulic dredges are all equipped with a ‘head’ that cuts or sweeps the material and conveys it to a hydraulic suction pipeline. The pipe either ends on a barge or is directly connected to a disposal facility on the shore. Typically, the amount of re-suspended sediment generated using hydraulic dredges is less than the amount generated by mechanical dredges (Pollice et al., 1996). In fact, hydraulic dredges, such as cutterhead dredges, cause 2–5 mg l\(^{-1}\) TSS background concentrations, whereas mechanical dredges, such as clamshell dredges, cause TSS background concentrations ranging from 10 to 12 mg l\(^{-1}\) (Havis, 1988). Pneumatic dredges use compressed air instead of centrifugal pumps to remove slurry through a pipeline. They can pump material with a relatively high solids content with little generation of turbidity. The method that causes the least environmental impact on the water quality during dredging operations is the hydraulic method. However, mechanical processes allow the dredging of sediments containing high solids concentration, which is an important criteria for the performance of a dredging operation (Hauge et al., 1998).

Once dredged, DM can be treated, valorised or disposed. Before any management decision, it is necessary to characterise the DM chemically, physically and mechanically (Boutouil et al., 1997). However, economic and political forces, and management, (i.e. lack of disposal sites) can interfere with the decision following the characterisation of DM. When contamination is too high to allow reuse, DM can either be disposed in controlled storage sites or treated. Disposal sites are of two types (Perrin and Zimmer, 1995):

- terrestrial disposal sites where DM are dumped mechanically or hydraulically towards basins or natural cavities and
- under water disposal sites such as disused quarries, deep zones or some river meanders.

Recently, the number of available disposal sites has rapidly declined due to urbanisation (Austin, 1995; Michelsen, 1998) and because of the large amounts of sediment that have to be dredged (Cuypers et al., 1998). In many cases, dredging for navigation and development projects has been hindered due to the presence of contaminated sediments and a lack of disposal sites (Michelsen, 1998).

2. Material and methods

2.1. Dredging in the Seine basin

The Seine river basin (France) study area includes the rivers Marne, Oise, Yonne and Seine, until Rouen, and covers an area of 68840 km\(^2\). In the Seine basin, mechanical dredging from a boat was chosen to remove, on average, 150 000 m\(^3\) of sediment per year (Fig. 1). Dredging in the Seine basin is mainly related to navigation. Consequently, dredging operations are nearly continuous along the rivers. As a result, hydraulic dredges cannot be used in this case since they need a deposit site within 2.5 km. Obviously, such a configuration does not exist all along the Seine river system. Therefore, mechanical dredging is, in the case of the Seine basin, the most adapted option.

Within the 6 million m\(^3\) DM extracted in France (internal waters only), 70% are dumped into deposit sites or along the riverside, 10% are valorised as embankments and 7% are spread on agricultural land. The rest is either used as an embankment material, dumped into the sea or treated. Since the dredging operations in the Seine basin occurs in urban areas, it is difficult to dump the DM on the riverside, as is currently performed in the north of France for example. Thus, DM is
Fig. 1. Part of the Seine basin concerned in this study and site codes corresponding to the sediment samples collected during 1996, 1999 and 2000 campaigns.
presently dumped at wet disposal sites downstream from Paris. From 1992 to 1998, the site at Triel and from 1999 to 2001, the site at Rouillard (Verneuil sur Seine) have been filled with the DM extracted from the River Oise, downstream from Compiegne, and from the River Seine downstream from Vitry and until Vernon (Fig. 1). It is of great importance to check the quality of the DM before it is dumped, in order to provide its efficient management. This is presented in this paper. As far as micro-pollutants are concerned, heavy metals and organic pollutants such as PAHs, PCB, pesticides etc. will be presented in a further publication.

2.2. Sediment collection

Samples were collected from the bottom of the Seine and Oise rivers (Fig. 1) by core sampler (20–50 cm depth, depending on the sediment structure) in order to assess the sediment quality before dredging. Sixty four samples were collected in May and June 1996 (31 samples), April 99 (18 samples) and May 2000 (15 samples) in strategic areas determined by the Navigation Services of the Seine basin, according to the dredging needs the following year, resulting from the bathymetric data of the river. Once cored, sediments were transferred into fully filled closed glass containers (1 l) and were kept for 48 h at 4 °C before homogenisation and analysis.

3. Methods

The parameters analysed on the sediment are listed in Table 1, together with the parameters measured in the pore-water of the same sediment samples. Pore-water was obtained after centrifugation of a known amount of sediment (3400 rpm at 4 °C, during 20 min) in order to obtain a sufficient volume to carry out analyses. The supernatant was then filtrated through a 0.45-μm pore filter to remove any remaining suspended solids. Most of the parameters were measured following the French AFNOR standard procedures (Table 1). All samples were analysed by laboratories certified by the French Ministry of the Environment, i.e. certified by the COFRAC (comité français d’accreditation). For international quality control, the COFRAC calibration certificates co-operate with other European calibration services (the European co-operation for Accreditation- EA). The harmonisation of the accreditation criteria and procedures are orientated towards the European standards for calibration and testing laboratories, and their

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Table 1
List of the physico-chemical parameters and major contamination analysed in the sediment (S) sampled in 1996, 1999 and 2000 and in the pore-water

<table>
<thead>
<tr>
<th>Analysis</th>
<th>Units</th>
<th>Method</th>
<th>Sample</th>
<th>Campaigns</th>
</tr>
</thead>
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<tr>
<td>Humidity</td>
<td>%</td>
<td>NF ISO 11465 X 31-102</td>
<td>S</td>
<td>96, 99, 00</td>
</tr>
<tr>
<td>Organic Matter</td>
<td>%</td>
<td>NFU 44-160</td>
<td>S</td>
<td>96, 99, 00</td>
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<tr>
<td>Total Organic Carbon (TOC)</td>
<td>g kg⁻¹</td>
<td>NF EN 1484 NF T 90-102</td>
<td>S, P</td>
<td>96,99, 00</td>
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<tr>
<td>Fine particles</td>
<td>%</td>
<td>NF X 11-501</td>
<td>S</td>
<td>96, 99, 00</td>
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<tr>
<td>Density</td>
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<td>U 44-175</td>
<td>S</td>
<td>96, 00</td>
</tr>
<tr>
<td>NO₃⁻, NO₂⁻</td>
<td>mg l⁻¹</td>
<td>NF EN ISO 10304-2 T90-046</td>
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<tr>
<td>NH₄⁺</td>
<td>mg l⁻¹</td>
<td>NF T 90-015</td>
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<tr>
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<tr>
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<tr>
<td>P</td>
<td>mg l⁻¹</td>
<td>NF EN ISO 11885</td>
<td>P</td>
<td>00</td>
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</table>

* Loss on ignition at 550 °C. (P): relevant analytical methods.
4. Results and discussion

For economic reasons, the Navigation Services of the Seine basin, in charge with the sampling and analysis, divides the Seine basin sites to be checked into three groups, and all of them are analysed within 3 years of sampling (here, 1996, 1999 and 2000). However, for a given year, the sampling sites are randomly defined, hence temporal evolution will not be taken into account in this paper.

4.1. Sediment quality

French regulations are not clear about the management of dredged materials extracted from rivers. There is presently no available limit that can be used for deciding the fate of DM. This type of regulation is only available for marine and estuarine sediments (French bylaw of the 14/06/2000). Consequently, waste regulations and/or sewage sludge regulations are often used as a reference, as well as the Dutch sediment decree (circular on target values and intervention values for soil remediation, Ministry of Housing, Spatial Planning and Environment, updated 4th February, 2000). The general characteristics of the sediment samples (Table 2) vary significantly from one sample to the other and demonstrate the great variability of the sediment characteristics all along the Seine basin. This makes the management of the DM quite difficult. The main variation is observed on the grain size, which shows that the percentage of fine particles (<50 μm) presents a coefficient of variation of 106% (Table 3). In fact, some areas show very sandy material (V01, V02, V15, V17, V23, S6, S8, S10, S11), whereas others contain a great proportion of silt (V22, V28, O10, O14, S12, S16). This grain size is in fact strongly linked to the organic content of the sediment. Fine sediments (grain size below 50 μm) are usually considered to adsorb organic and metallic pollutants more than coarse fractions (Evans et al., 1990; Warren and Zimmermann, 1994; Perrin and Zimmer, 1995). Such large variability is also observed for organic matter, whether determined with loss on ignition (OM) or the total organic carbon (TOC).

On average, the density of the sediment is a reasonably stable value (average, 1.47; CV, 14%). We can therefore consider this density as a characteristic of the sediment present in the entire Seine basin studied.

Over the whole Seine basin, the sediment contains between 14 and 75% water. In terms of DM management, this type of material cannot always be treated as a typical urban or industrial waste. As far as waste is concerned, the French regulation of the 18 December, 1992, modified by the regulation of the 18 February, 1994, after the French law no. 75-633 about industrial waste, considers a waste as stabilised if its siccity (i.e. dry matter content) is above 35%. This is the case for most samples collected in the Seine basin except 12 from the 64 samples (Table 2).

If we consider previous studies performed in the Seine basin on suspended solids collected from December 1994 to March 1995 (Meybeck et al., 1998), 40–45% of the suspended matter collected by sediment traps were fine particles (<50 μm). However, the sampling method was completely different to the one performed in the present study (core sampling) where the average result found for grain size below 50 μm is 20%. For that reason, we shall presume that sediment trap samples represent the very superficial sediment of the Seine basin. This could explain why the fine particles percentages are much smaller when collected with a core sampler (20%). In a previous study carried out in 1992, Seine sediments downstream from Paris had an average fine particles content reaching 36% (Garban et al., 1995). However, this value was obtained with sediments arising from only three sites on the Seine downstream from Paris, whereas sediments analysed in the present study were sampled in 64 locations, all along the Seine basin including Marne, Oise, Yonne, and Seine upstream and downstream from Paris, hence over a much larger area, presenting a large variability potential (CV = 106%).

When organic matter is assessed, former studies carried out on the Seine river downstream from Paris, show a loss on ignition in sediments collected in 1992 reaching 11% (Garban et al., 1995).
Table 2
General characteristics of the Seine basin sediment collected in 1996, 1999 and 2000, before the dredging operation

<table>
<thead>
<tr>
<th>Site code</th>
<th>Year of collection</th>
<th>TOC (g kg⁻¹)</th>
<th>Density (%)</th>
<th>Humidity (%)</th>
<th>OM (%)</th>
<th>Fine particles (% &lt;50 μm)</th>
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<th>Humidity (%)</th>
<th>OM (%)</th>
<th>Fine particles (% &lt;50 (\mu)m)</th>
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The organic layer of the Paris combined sewer system, which is resuspended and discharged into the Seine river during storms (combined sewer overflows CSO) shows loss on ignition values reaching 60% (Ahyerre, 1999). Total suspended solids of the Seine river downstream from CSO outlets and Paris, present loss on ignition of approximately 20% (Estèbe, 1996). In the present study, the sediments collected in 1996, 1999 and 2000 show an average value of 8%. OM values (loss on ignition) in the Seine basin seem to display average to low contents compared to the different sources of urban organic matter.

In the North of France, Ruban et al. (1998) found high concentrations of total organic carbon (TOC average, 83 g kg\(^{-1}\)) in the sediments of rivers from a very industrialised region of Lille (Ruban et al., 1998), which is twice as much as the average value found in the Seine basin. Stegmann and Krause (1988), found concentrations ranging from 160 to 200 g kg\(^{-1}\) in dredged sediments and sediment sampled directly from the bottom of the Elbe river in Germany. These comparisons tend to indicate that the sediment of the River Seine basin present concentrations a lot lower than those found in heavily polluted areas, such as the north of France or the River Elbe in Germany. The organic matter content being frequently linked to the micropollutant contamination, the Seine river basin DM should not present a great threat as long as their management is concerned. Nevertheless, it is of considerable importance to determine how polluted the same sediments are as far as micropollutants are concerned.

Table 3

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<th>OM (%)</th>
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<th>BOD (mg l(^{-1}))</th>
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<th>NO(_2)^- (mg l(^{-1}))</th>
<th>NH(_4^+) (mg l(^{-1}))</th>
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Concerned. Moreover, checking the properties of the pore-waters of such sediments is of great importance for the management of DM, since this material is bound to undergo important physical and chemical changes during its dredging, transport and dumping, and therefore the pore-water is likely to be released in the overlying or underground water.

4.2. Pore-water quality

Thirty-one samples of pore-water were also analysed in the sediments collected during the 1996, 1999 and 2000 campaigns (Table 4 and Table 5). As far as the organic content is concerned, the variability observed in the sediment is also found in the pore-water. The average TOC
content of the pore water is 83 mg l\textsuperscript{-1} (CV = 100\%) when the River Seine water, downstream from Paris, showed an average TOC of 7 mg l\textsuperscript{-1} (Seidl et al., 1998). The most striking parameter, which characterises the pore-water, is the ammonium ion. Our results concerning the Seine river basin show relatively high concentrations (61 mg l\textsuperscript{-1}, on average). The ammonium values obtained in a previous study carried out on sediment samples from the Seine basin cored in 1992, gave average values reaching 55 mg l\textsuperscript{-1} (Garban et al., 1995), which fits within the same concentration range as the results obtained in the present study. Indeed, NH\textsubscript{4}\textsuperscript{+} accumulates in anoxic sediment pore-waters due to nitrification of organic nitrogen. The consequence of high concentrations of NH\textsubscript{4}\textsuperscript{+} in pore-waters is a release of NH\textsubscript{4}\textsuperscript{+} when sediments are re-suspended (Stumm and Morgan, 1981). Moreover, it has been found that mortality of C. Fluminea juveniles in river sediments was more closely associated with the presence of unionised ammonia than with the levels of metals, pesticides and PCBs. Similar results were reported for the bivalve Anodonta Imbecillis (Cataldo et al., 2001). As the management of DM in the Seine basin involves re-suspension of the sediment with large quantities of Seine water (9/1 v/v), this method dilutes the NH\textsubscript{4}\textsuperscript{+} contained in the pore-water in the wet disposal site and should cause ammonium contamination of the waters. This ammonium input might cause adverse effects to the overlying waters in the disposal site itself, to the underground waters and to receiving waters if there is a discharge system in the disposal site. This phenomenon has been observed in the case of a disposal site of the River Seine basin used from 1999 to 2001 (Carpentier et al., 2001).

4.3. Geographic distribution and correlation analysis

Geographic distribution with the different results (TOC, density, humidity, fine particles) obtained for the sediment quality before dredging were determined using Mapinfo® Geographic Information System. The Seine basin maps did not reveal any significant effect of the geographic position. Indeed, the fact that some sediments were sampled upstream Paris did not prevent them from being rich in organic matter compared to other samples collected downstream from Paris, as we should expect considering the anthropic effect suggested by this heavily industrialised and urbanised area. Nevertheless, we found (Fig. 2) that sediment organic matter as expressed by loss on ignition, as well as other parameters related to the organic characteristics of the sediment, (i.e. TOC), was more abundant directly downstream from Paris, until the crossing of Seine and Oise rivers, where polluting activities are concentrated. In fact, the impact of Paris conurbation, i.e. the opposition between upstream of and downstream from Paris, leading to the statement that sediments downstream from Paris should be more contaminated
Fig. 2. Organic matter contents (loss on ignition, %) in the sediment collected along the Seine basin during 1996, 1999 and 2000 campaigns.
Table 6
Linear correlation coefficient (r) between parameters analysed in the Seine basin sediments (S) and in their pore-waters (P) in 1996, 1999 and 2000

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<th>Density (S)</th>
<th>Humidity (S)</th>
<th>OM (S)</th>
<th>Fp. ( ^{a} ) (S)</th>
<th>TOC (P)</th>
<th>COD (P)</th>
<th>BOD(_{5}) (P)</th>
<th>NO(_{3})(^{-}) (P)</th>
<th>NO(_{2}) (P)</th>
<th>NH(_{4})(^{+}) (P)</th>
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<td>-0.43</td>
<td>-0.47</td>
<td>-0.26</td>
<td>-0.38</td>
<td>0.50</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NH(_{4})(^{+}) (P)</td>
<td>0.45</td>
<td>-0.69</td>
<td>0.74</td>
<td>0.49</td>
<td>0.59</td>
<td>0.30</td>
<td>0.44</td>
<td>0.79</td>
<td>-0.45</td>
<td>-0.39</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NTK (P)</td>
<td>0.43</td>
<td>-0.68</td>
<td>0.74</td>
<td>0.49</td>
<td>0.59</td>
<td>0.29</td>
<td>0.43</td>
<td>0.78</td>
<td>-0.46</td>
<td>-0.39</td>
<td>0.99</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>P (P)</td>
<td>-0.03</td>
<td>0.18</td>
<td>0.34</td>
<td>0.15</td>
<td>0.33</td>
<td>-0.21</td>
<td>0.09</td>
<td>0.50</td>
<td>-0.31</td>
<td>-0.07</td>
<td>0.45</td>
<td>0.47</td>
<td>1</td>
</tr>
</tbody>
</table>

\( ^{a} \) Fine particles (% <50 \( \mu \)m).

than those upstream of Paris, was not observed and did not explain the variability of the organic characteristics of the sediment.

In order to assess the possible role of one or several parameters to control other parameters, linear correlation analysis was performed. Correlation analysis with sediment general parameters (Table 6) did not reveal any major trend, apart from very usual ones such as a negative correlation between density and humidity \( r = -0.86 \) or density and TOC \( r = -0.70 \) or a positive correlation between humidity and OM \( r = 0.69 \). Surprisingly, we observed weak correlation between TOC and OM \( r = 0.36 \), or OM assessed by loss on ignition and fine particles content \( r = 0.51 \).

Concerning the sediment pore-waters (Table 6), usual correlations were found between BOD\(_{5}\) and COD \( r = 0.95 \), NH\(_{4}\)\(^{+}\) and TKN \( r = 0.99 \), or between TOC and COD \( r = 0.99 \). This type of correlation should probably be more informative when physico-chemical parameters and micropollutants data are simultaneously taken into account (in a further publication).

The combined linear correlation of sediment parameters and pore-water parameters (Table 6) gave high coefficients for TOC in the sediment and NO\(_{3}\)\(^{-}\) observed in the pore waters \( r = 0.71 \). This correlation could be related to the biological activity that links these parameters (nitrifying bacteria may be more abundant in more organic media).

5. Conclusions

During three campaigns performed in 1996, 1999 and 2000, sediment samples were collected
and analysed before dredging in order to assess their major composition, in the Seine river basin, upstream and downstream from Paris. This quality is important in the case of the Seine basin, as the sediments are dumped into disused quarries. Hence, the composition of these sediments was estimated in order to assess their possible impact after disposal. As far as the particulate matter is concerned, compared to other industrialised river basins, the sediments are not heavily loaded in organic matter. Pore-waters contain high concentrations of ammonium ions, compared to other river basins, that might lead to a management concern, because the dumping of such sediment might result in an increase of ammonium concentration in the receiving waters, i.e. disused quarries, underground water or Seine water. At the disposal sites, where the water is stagnant, high inputs of ammonium ion, as well as phosphorus inputs, could lead to eutrophication concern. Such an impact was in fact observed on the DM disposal site of the River Seine basin, i.e. the Rouillard pond (Carpentier et al., 2001). Therefore, the analysis of ammonium in the sediment pore-waters should be considered as an important parameter to assess the quality of sediments before dredging. However, the main characteristic of the sediment samples is its wide variability from one site to the other. Consequently, the main outcome of this work is that the sediment collection and analysis in each dredged area should be performed before dredging, as often as possible. Indeed, along the Seine basin, the river sediment shows a wide range of properties due to point and non-point sources, but also due to hydrology or navigation, which one is unable to control. Depending upon such parameters, the amount of organic matter and nitrogen of sediment samples, different management procedures for DM could be decided, i.e. dumping in wet disposals, separating fine particles from coarse ones, so the fine ones can be treated, and the coarse ones valorised. Such a procedure might be necessary when national or European regulations appear in relation to the European Framework Directive on water management.

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