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Characterisation and impact study on the environment of sediments dredged in the north of France

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Abstract:
The work presented in this paper is undertaken in the framework of a methodology suggested to improve management of the marine and river sediments dredged in the north of France. The first stage of this methodology consists in determining the main characteristics of the sediments studied in relation to the field of valorisation considered. Thus, in this paper the physical, mineralogical and chemical properties of the two types of studied sediments are presented. Their environmental impacts are explored through leaching tests. From the obtained results, their beneficial use, in the field of road work, is discussed.

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1. Introduction

Dredging consists in moving or removing a material from the aquatic environment. The operation of dredging can be necessary in order to maintain or to improve the conditions of navigation by: the deepening or the widening of the marine channels, removing sludge from the ports, the creation of new waterways and also the removal of the polluted sediments from the aquatic environment.

With the increase of demand of materials in various fields as in: civil engineering work, manufactured goods (bricks), agriculture or rehabilitation of sites (ST. LAWRENCE CENTRE, 1993; ULBRICHT, 2002; LIFE, 2002), the dredged sediments can help to improve the management of standard aggregates and can constitute a new source of materials.

In France, civil engineering field consumes, annually, more than 400 million tons of granular materials with more than half of this quantity in the field of road works (UNPG, 2005; MICHEL, 1997). To ensure a viable valorisation of the dredged sediments in this field, it is important to improve the knowledge of their geotechnical characteristics. Because of the presence of salts, in the marine sediments, of organic matter and pollutants, in the marine and river sediments, the evaluation of their environmental impacts is also of a very great importance.

In this paper, marine sediment and river sediment dredged in the north of France are studied. In the context of a valorisation in the field of road works, the physical, mineralogical and chemical characteristics of the sediments are discussed. The potential impact of these materials on the environment is evaluated through leaching tests.

2. Methodology

The methodology developed to ensure the valorisation of the dredged sediments in the field of roads works is composed of three stages (ZENTAR *and Al*, 2005):

- A stage of characterisation of the raw sediments: this stage includes the identification of the mechanical, physico-chemical and mineralogical characteristics of the raw sediments. The environmental impact of these materials is also evaluated.
- A stage of formulation of a road material: this stage has as a role the improvement of the mechanical performance of the raw sediments to reach the desired performance. This stage includes a stage of characterisation of the added materials and the study of the designed road material.
- A stage of validation: this stage includes the validation of the mechanical characteristics and the study of the impacts on the environment of the designed material.

In the following, the effort will be made on the identification of the physico-chemical and mineralogical characteristics of the raw sediments and on the impact study on the environment of the designed materials in the context of a valorisation in the field of roads construction. The design technique and the mechanical characteristics of the designed materials, described by DUBOIS (2006), will be very briefly discussed in this paper.

3. Presentation of the sites of taking away

The sediments studied in this work are dredged from Dunkirk port for the marine sediment and from the channel of Scarpe for the river sediment (fig. 1).

Dunkirk port, the third French port in term of exchange of goods, ensures an annual traffic of more than 57.11 million tons of goods. Concerning the sedimentary activity, nearly 4 million m^3 of mud every year are dredged (DUBOIS, 2006). The principal proportion of dredged sediments is regarded as not polluted, according to the French legislation (METL & MATE, 2000). In this case, the immersion of the sediment is always possible. In this study, the marine sediment is dredged in the East of the port of Dunkirk. This zone, according to a zoning study carried out in the port (MAC FARLANE, 2004), is regarded as polluted according to the decree of the 14/06/2000.

The Scarpe river extends between Arras and Corbehem in the Pas-de-Calais region. The sampling was carried out in the middle of the channel, near the town of Brebières located at approximately 50 kilometers from the town of Lille (France).

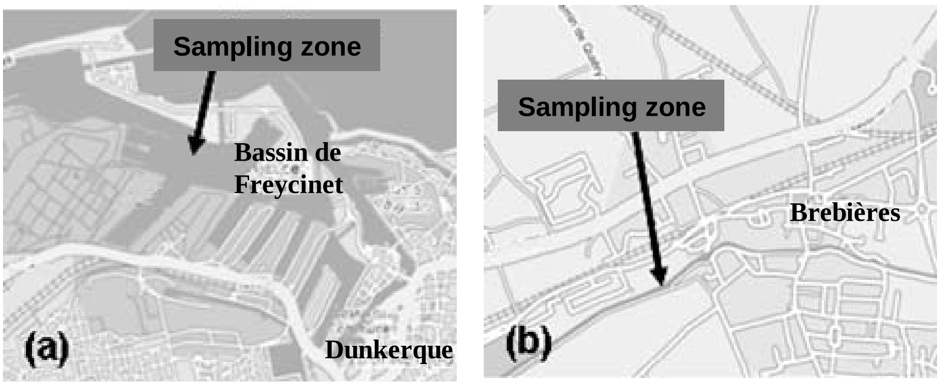

Figure 1. Zones of sampling of the marine sediment (A) and the river sediment (b).

4. Test results and discussion

4.1 <u>Determination of the physical characteristics</u>

In the field of road work, determination of granulometry, quantity and the activity of the clay fraction, organic matters content, and the consistency limits of materials constitute the starting point of any study. The knowledge of these characteristics allows in particular the material classification, in France, according to the earthworks guide (GTR, 1992).

In this study, the granulometric distribution of the sediments was determined by the laser diffraction technique. The obtained results prove that the two types of sediments are comparable. They are composed mainly of fine particles. The proportion of clay

(particles lower than 2 μm) is slightly more important in the river sediment (12%) that in marine sediment (9%), (Table 1).
To characterise the activity of the argillaceous fraction, the method of adsorption of the methylene blue is used (NF P 94-068, 1993). For each type of sediment, the median value of three test results is reported in Table 1. From these results, it appears that the activity of the argillaceous fraction of the river sediment is appreciably more important than the activity of the argillaceous fraction of the marine sediment. This result is partially due to the higher clay content in the river sediment in comparison to the content measured in the marine sediment but also to the type of argillaceous minerals as it will be shown by the x-ray diffraction analyses.
The organic matters (OM) content (MO), for the two types of sediment, was measured by two methods: By the method of loss of mass by ignition at 450 °C (XP P 94-047, 1998) and by the sulfochromic oxidation method (NF ISO 14235,1998). In the second method, the OM content is deduced from the percentage of organic carbon by multiplying this last quantity by 1.72 (MUSTIN, 1987). According to the values presented in Table 1, the test results seem to be comparable for the marine sediment where an OM content of approximately 8% is estimated with the two methods but for the river sediment, the relative difference observed reaches 100%. The over-estimate of the OM content by the method of mass loss by ignition could be induced by the loss of combined water in clays by the heating. This loss of combined water is more marked for argillaceous minerals with a high specific surface (HOLTZAPFFEL, 1985).
The Atterberg limits of the two types of sediment are measured according to standard test NF P 94-051 (1993). The liquid limit (w_L) is measured by using the Casagrande equipment and the plastic limit (w_P) by the technique of the rollers. In term of w_P, the values measured on the two types of sediment are about 30%. For the w_L, the value measured on the marine sediment is almost twice higher than the value measured on the river sediment. According to a complementary study undertaken on the marine sediment, it was shown that this high value of the liquid limit w_L is mainly due to the presence of OM (DUBOIS, 2006). On the basis of the method of determination of OM by sulfochromic oxidation, the OM content of the river sediment is less than half of the content measured in the marine sediment.
In addition to the four basic physical characteristics described and measured above, the density of the solid particles (ρ_s) is measured using a pycnometer with helium of the type Accupyc . A complete cycle of measurements comprises 80 measurements. These measurements are carried out with various pressures. For each type of sediment, the results reported in Table 1 are the median values of 240 measurements taken out of three samples. In terms of results, as envisaged, the measured values are lower than the values known for the mineral particles which constitute the sediments (≈ 2.70 t/m^3). This difference is due mainly to the presence of OM which is lighter than the mineral particles. To check this statement, the density of the solid particles of the sediment, after

ignition at 450 °C during 12 hours, was measured. The result obtained is comparable with the standard values of inorganic materials. Also, the difference observed between the values of ρ_s measured for the samples ignited at 450 °C and the raw sediments are more important for the marine sediment than for the river sediment. This result could support the assumption that the OM content in river sediment is lower than that in the marine sediment.

In terms of materials classification, according to the guide of earthworks (GTR, 1992), the marine sediment and the river sediment studied are regarded as moderately organic materials. This is due to the OM content which is higher than 3%. This class of material is indicated as class F. According to the granulometric distribution, the value of the methylene blue and the Atterberg limits, the marine sediment is classified as a material of type A4-F11. This class includes the clay soils which are moderately organic. The river sediment is classified as a material of type A2-F11; this class includes the muddy grounds which are moderately organic (GTR, 1992).

In the field of road work, the use of these materials as embankments materials with low heights can be allowed under specific conditions. For the use in the structure of the roadways, additional tests are necessary for a better evaluation of the mechanical behaviour.

Table 1. Physical characteristics of the studied sediments.

Paramètres	**Sédiment marin**	**Sédiment fluvial**
% < 2µm (Clays)	9%	12%
2µm < % < 63µm (Silts)	72%	80%
63µm < % (Sands)	19%	8%
Methylene blue value (MBV)	3.4	4.2
% MO by ignition	8.6	7.2
% MO by oxidation	7.2	3.1
w_L (%)	89.4	49.5
w_P (%)	35.5	28.0
IP (%)	53.9	21.5
ρ_s (t/m^3)	2.53	2.56
ρ_s (t/m^3) after ignition (450 ^{0}C)	2.69	2.66

4.2 Mineralogical analysis

A good knowledge of the mineralogical composition of materials is essential to anticipate the problems which could be induced during the valorisation of the dredged sediments and can also explain the contradictions observed in the current test results as in the test of loss on the ignition, sulfochromic oxidation and the methylene blue test. In

this study, the crystalline phases in materials are identified by the use of the technique of x-rays diffraction. The equipment used is of the type Siemens D500.
For the studied marine sediment, the three detected principal crystalline phases are: calcite, quartz and halite. For the river sediment, calcite and quartz are the two principal crystalline phases.
To determine the nature of the argillaceous phases, specific analyses were carried out on particles of size lower than 2µm. These analyses consist in characterising the raw sample, the sample after thermal treatment at 450 °C during two hours and material saturated by ethylene glycol vapours. The heat treatment facilitates the detection of Kaolinite and the treatment by ethylene glycol vapours facilitates the detection of the Smectites. From the treatment of the spectra of diffraction obtained, in Table 2, the distribution of the argillaceous phases is reported. From these results, it appears that the argillaceous phases of the marine sediment are composed of Smectites, of Illite and Kaolinite in equal proportions. For the river sediment, the Smectites constitute the main phase. The high proportion of Smectites in the river sediment, due to their important specific surface and the capacity to adsorb water, can explain the value obtained in the test with the methylene blue and partly the contradiction observed between the tests results of mass loss on ignition and the sulfochromic oxidation.

Table 2. Type and proportion of minerals in the studied sediments.

Minerals	***Smectites***	***Illite***	***Kaolinite***	***Chlorite***
Marine Sediment (%)	*30*	*32*	*30*	*8*
River Sediment (%)	*55*	*20*	*15*	*10*

4.3 Environmental aspect

To evaluate the environmental impacts of the raw sediments, in this study leaching tests, with a liquid to solid ratio of 10, are realised according to the specifications defined in European standard (NF EN12457-2, 2002).
The inorganic parameters, such as metals and the anions, are measured in the leachates. The organic parameters, such as polycyclic aromatic hydrocarbons (PAH), mineral oil polychloro biphenyl's (PCBs), the benzene, toluene, the ethyl benzene and the xylene compounds (BTEX) are directly measured on the solid. The thresholds prescribed in the European decision (CEU, 2003) are used to evaluate the potential impact of the materials on the environment. It is important to note that these thresholds were developed to establish criteria for the acceptance of waste in the waste canters. Currently, specific thresholds for the valorisation of the dredged sediments in road work do not exist.
The results of the analyses on the two types of sediments are reported in Table 3. From these results, it appears that contents of chlorides, molybdenum and hydrocarbons, for the marine sediment, and the content of the PAH, for the river sediment, exceed the

limits fixed for inert waste. It is important to note that for the marine sediment, the tests were carried out on a material with a sea water content of 200%.
The study undertaken to determine the mechanical characteristics of the raw sediments for a beneficial use in the field of road work, which is not detailed in this paper, indicated a relatively low bearing capacity (poor values of the immediate bearing capacity value denoted IPI), a high sensitivity to water and poor mechanical characteristics. In order to ensure a viable valorisation, the mechanical performance was increased by modifying the granular distribution of the raw sediments by adding pit sand (SC) and/or dredged sand (SD) in combination with binders.

Table 3. Results of the analysis of the raw sediments.

Elements	Unit	Marine sediment	River sediment
Tests on leachates			
pH		8.3	8.0
Conductivity	mS/cm	7.7	0.2
As	mg/kg	< 0.5	< 0.5
Ba	mg/kg	< 0.2	< 0.2
Cd	mg/kg	< 0.04	< 0.04
Cr	mg/kg	< 0.5	< 0.5
Cu	mg/kg	< 0.5	< 0.5
Hg	mg/kg	< 0.001	< 0.001
Mo	mg/kg	4.19	< 0.5
Ni	mg/kg	< 0.4	< 0.4
Pb	mg/kg	< 0.5	< 0.5
Sb	mg/kg	< 0.06	< 0.06
Se	mg/kg	< 0.1	< 0.1
Zn	mg/kg	0.73	< 0.5
Fluorides	mg/kg	7.1	2.5
Chlorides	mg/kg	27537	30
Sulphates	mg/kg	3718	310
Phenol Index	mg/kg	0.64	< 0.10
TOC	mg/kg	479	78
Test on solids			
TOC	mg/kg	42000	18000
BTEX	mg/kg	< 5	< 5
PCB	mg/kg	0.28	0.055
Hydrocarbones	mg/kg	820	230
PAH	mg/kg	29.1	110

Legend:
- *Inert waste*
- *Non dangerous waste*
- *Dangerous waste*
- *Treatment befor storage*

From preliminary studies undertaken, a quantity of sediments in the mix was constrained by the acceptable OM content in the mix (<3%) and by the quantity of fine particles (< 12%). The content of cement in the mix is equal to 6% by sediment dry weight, quantity typically used in the field. From the preliminary study, four mixes are developed, (Table 4). Two mixes contain marine sediment with or without the pit sand (denoted FM1 and FM2) and two formulations contain river sediment (denoted FF1 and FF2).

In terms of mechanical characteristics, which are not discussed in this paper, the tests carried out on the mixes made it possible to validate the design methodology (DUBOIS, 2006).

With regard to the impact study on the environment, the leaching tests are carried out on monolithic samples (NF X31-211, 2000) and on crushed samples (NF EN12457-2, 2002). The monolithic samples are prepared according to the method for preparing cylindrical samples for mechanical tests (NF P98-230-2, 1993).

From the results of the analyses (Table 5), it appears that the monolithic samples do not indicate particular pollution in the leachates except a moderate quantity of chlorides for the mixes which include the marine sediment.

For the crushed samples, the tests on the mixes containing river sediment (FF1 and FF2) indicate that the materials are inert by comparison with the thresholds established for waste. For the mixes containing the marine sediment (FM1 and FM2), the analyses indicate quantities of molybdenum and chlorides lower than those measured on the raw sediment. However, the content of copper is appreciably higher than the content measured on the raw sediment. This result requires complementary analyses and analyses of the added materials in the mixes. In a general way, according to the chlorides contents measured and of molybdenum, the materials containing the marine sediment exceed the threshold of inert waste and are considered as not dangerous waste.

Table 4. Formulation of materials containing marine sediment and river sediment.

	FM1	*FM2*	*FF1*	*FF2*
% Marine Sediment	*33*	*33*	*/*	*/*
% River Sediment	*/*	*/*	*23*	*23*
% Dredged Sand	*20*	*61*	*24*	*71*
% Pit Sand	*41*	*/*	*47*	*/*
% Cement	*6*	*6*	*6*	*6*

5. Conclusions

Dredged sediments, according to European classification (CEU, 2001; MATE, 2002), are regarded as waste. To promote their valorisation in road work, it is necessary to better know their physical, mineralogical, chemical and mechanical characteristics but also it is important to evaluate their environmental impacts.

Table 5. Results of analysis of designed materials containing marine and river sediments.

		Tests on leachates							
Test on		**Monolithic samples**				**Crushed samples**			
Parameters	*Unit*	*FM1*	*FM2*	*FF1*	*FF2*	*FM1*	*FM2*	*FF1*	*FF2*
L/S		10	10	10	10	10	10	10	10
pH		11.3	11.3	7.93	8.11	12.1	12.2	11.05	12.08
Conductivity	mS/cm	1.6	1.6	0.7	0.7	4.4	4.4	3.2	3.4
As	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Ba	mg/kg	0.15	0.17	0.2	0.2	0.46	0.47	0.9	1.2
Cd	mg/kg	< 0.04	< 0.04	< 0.04	< 0.04	< 0.04	< 0.04	< 0.04	< 0.04
Cr(VI)	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cr	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cu	mg/kg	1.96	1.98	< 0.5	< 0.5	12.87	12.51	< 0.5	< 0.5
Hg	mg/kg	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Mo	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	2.75	2.70	< 0.5	< 0.5
Ni	mg/kg	< 0.4	< 0.4	< 0.4	< 0.4	< 0.4	< 0.4	0.50	0.46
Pb	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Sb	mg/kg	< 0.06	< 0.06	< 0.06	< 0.06	< 0.06	< 0.06	< 0.06	< 0.06
Se	mg/kg	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Zn	mg/kg	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Fluorides	mg/kg	1.3	1.3	1.4	1.4	5.8	5.7	6.4	6.5
Chlorides	mg/kg	3627	3632	36.3	16.5	7009	8004	122.7	59.3
Sulphates	mg/kg	196	175	38.1	35.5	823	673	122.2	104.0
Phenol Index	mg/kg	0.19	0.23	/	/	0.54	0.63	/	/
TOC	mg/kg	120	131	48	55	530	520	200	211
		Tests on solids							
TOC	mg/kg					12000	10000	4000	4000
PCB	mg/kg					< 0.05	< 0.05	< 0.05	< 0.05
Hydrocarbones	mg/kg					500	450	33	20
PAH	mg/kg					6.5	5.4	6.8	6.3

Inert waste *Non dangerous waste*

Non dangerous waste *Treatment before storage*

In this study, the measured physical characteristics show that the studied sediments are fine materials, made up mainly of silts (passing at 63 µm is higher than 80%), moderately organic and highly plastic. The environmental impact, evaluated through leaching tests, shows a high chloride content, moderate molybdenum contents and

hydrocarbons for the marine sediment. For the river sediment, only the presence of the PAH is reported.
To fulfil the requirements in term of mechanical characteristics for valorisation in road engineering of the dredged sediments, it was necessary to improve the granular distribution of the sediments by adding sand and binders.
On the designed materials, the leaching tests carried out on the monolithic samples did not indicate particular pollution, except moderate chloride content for materials containing marine sediment. For the tests carried out on the crushed samples, one observes moderate amount of copper hydrocarbon and molybdenum for the mixes containing the marine sediment. The mixes containing the river sediment did not indicate particular problems. However, it is important to note in this study that the thresholds used to evaluate the quality of the leachates are established for the acceptance of waste in landfill and that at present specific thresholds for the valorisation of the dredged sediments in road work do not exist.
At this stage of the study, for the mixes qualified in terms of mechanical behaviour and harmlessness on the environment, it seems important to validate their behaviours on a site scale.

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