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ASSESSMENT OF TWO THERMALLY TREATED DRILL MUD WASTES FOR LANDFILL CONTAINMENT APPLICATIONS

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ABSTRACT

Offshore oil and gas drilling operations generate significant amounts of drill mud waste, some of which is transported onshore for subsequent thermal treatment (i.e. via thermal remediation). This treatment process results in a mineral waste by-product (referred to as thermally treated drill mud waste, TTDMW, in this paper). Bentonites are originally present in many of the drill mud products and it is hypothesized that TTDMW can be utilized in landfill containment applications (i.e. cover or base liner). The objective of this paper is to examine the feasibility of this application by performing various physical and chemical tests on two TTDMW samples. It is shown that the two TTDMW samples contained relatively small amounts of clay-sized minerals although hydraulic conductivity values are found to be less than 10^{-8} m/s. Organic carbon contents of the samples were approximately 2 percent. Mineralogy characterization of the samples confirmed varying amounts of smectite, however, peak friction angles for a TTDMW sample was greater than 36^{o}. Chemical characterization of the TTDMW samples show potential leaching of barium and small amounts of other heavy metals. Discussion is provided in the paper on suggestions to assist in overcoming regulatory issues associated with utilization of TTDMW in landfill containment applications.

Keywords: Drill mud waste, barrier, landfills, containment, waste management

INTRODUCTION

Recycling of industrial mineral waste by-products for construction purposes represents an environmentally sustainable practice (Sarsby, 2000).. Internationally, one mineral waste that can be generated in large quantities is drill mud waste from offshore oil and gas projects. In 2003, the Protection of the Marine Environment of the North-East Atlantic Commission reported that over 200,000 tonnes of drilling mud waste were generated in the northeast Atlantic offshore region and of this amount; over 100,000 tonnes were brought onshore for treatment or disposal (OSPAR, 2005). Page et al. (2003) examined the feasibility of many different options for reuse of this waste (also referred to as drill cuttings), and four potential options for its recycling were recommended: use in cement manufacture, use in road pavements, bitumen and asphalt, use as fuel, and use in concrete blocks and "ready-mix". The reuse of drilling waste as landfill bottom liners was briefly mentioned by Page et al (2003), mainly in relation to its potential beneficial hydraulic properties. These same authors also state that there is a lack of data on the characterization of the physical and chemical characteristics of drill mud waste.

Thermal desorption (Ntukidem et al., 2002) via indirection thermal recovery processes provides for recycling of the liquid hydrocarbon fraction of this drill mud waste. The mineral waste by-product remaining from the thermal treatment process (referred to as thermally treated drill mud waste, TTDMW, in this paper) may also potentially be recycled (Page et al., 2003). The original composition of a drilling mud will ultimately play some role in the final properties of the TTDMW generated. This composition will vary depending on the drilling conditions and/or environmental regulations for ultimate disposal (NSDEL, 2003), but the basic constituents of drilling muds consist of a base fluid (e.g. water or hydrocarbon), bentonite clay (hydrophilic or hydrophobic), and a multitude of organic and inorganic additives (Bourgoyne et al. 1986). Usually for difficult drilling operations, Oil Based Muds (OBMs) of low toxicity (e.g. synthetic based muds and low toxicity enhanced mineral oil based muds) are preferred in regions with strict environmental regulations due to potential waste management issues after drilling operations are completed. These OBMs often require various forms of organoclays to provide dispersive characteristics for the hydrocarbon based fluid. From a recycling perspective, the bentonite and/or organoclays remaining in TTDMW may be beneficial for landfill containment applications. Bentonite is utilized in many liner and cover applications for municipal solid waste landfills (Lake et al. 2004). Organoclays have been shown in many studies (Smith and Jaffe, 1994; Lo et al., 1997; Xu et al., 1997, Lake and Rowe, 2005) to be efficient at removing volatile organic compounds (VOCs) from solutions or leachates relative to natural clayey soils.

The objective of this study was to examine the suitability of TTDMW material for landfill containment applications (e.g. cover, basal liner). Due to the paucity of information related to physical and chemical properties of TTDMW for landfill containment applications, it is imperative that these properties be defined. Two samples were studied; one originating from the United Kingdom sector of the North Sea and the second from Nova Scotia, Canada (referred to as TTDWW-UK and TTDMW-NS respectively). The original impetus for the study was to examine the nature of the

TTDMW-NS sample to be produced for a proposed indirect thermal recovery treatment process (i.e. the treatment process did not exist at the time of the study). The TTDMW-UK sample served as a field scale "standard" of TTDMW product. To assess the suitability of these two TTDMWs for landfill containment applications, results of various physical, chemical and mineralogical tests are presented that may help to define relevant regulatory and performance issues related to these materials

MATERIALS AND METHODS

Sampling of the TTDMWs

TTDMW-UK was sampled from a larger existing sealed container of dry material in Halifax, Nova Scotia. This material was originally used for offshore drilling activities in the North Sea and derived from a low toxicity OBM (Envirosoil, 2003). This particular TTDMW material was chosen for the research because it is an actual by-product of a full-scale indirect thermal recovery (ITR) process. In addition, a drill mud waste sample from Nova Scotia, Canada was obtained from a local drilling mud waste treatment company. Little technical information is known about the initial characteristics of this drill mud waste except that it was generally considered a low toxicity OBM. Unlike the TTDMW-UK sample, this sample was collected prior to field scale thermal treatment since, at the time of this study, the Nova Scotia drill mud waste thermal treatment facility utilized a different thermal remediation technique than that performed for the TTDMW-UK sample.

To prepare a TTDMW sample from this viscous drill mud waste material, a bench scale thermal desorption process was performed in the laboratory. The sample was heated on a hot plate at 100°C in a fume hood for 16 h to remove the majority of the free liquid product. The material was then placed in a vented oven at 250°C for 1 h. This temperature is representative of typical thermal desorption temperatures used in practice (Ayen and Swanstrom, 1992; Wait and Thomas, 2003). The TTDMW sample generated from this process is hereafter referred to as TTDMW-NS.

Total petroleum hydrocarbon (TPH) concentrations (Atlantic PIRI, 1999) of the two TTDMW samples were as follows: TTDMW-UK, 375 mg/kg (C_{10}-C_{21}) and 253 mg/kg (C_{21}-C_{32}), TTDMW-NS, 550 mg/kg (C_{10}-C_{21}) and 54 mg/kg (C_{21}-C_{32}). It should also be noted that TPH concentrations for the TTDMW-NS sample (and TTDMW-UK) are below the acceptable values for soil on Canadian industrial sites, according to CCME (1999).

Chemical and geotechnical/geo-environmental test procedures

Various chemical and geotechnical/geo-environmental tests were performed on the two TTDMW materials to provide some practical assessment of the potential to recycle TTDMW materials into landfill soil liner systems. Table 1 presents a summary of the tests performed and procedural standards or references. Flexible wall hydraulic conductivity tests were performed on samples compacted with standard energy to moisture contents ranging from 2-4% above the optimum moisture content. Samples were consolidated to an effective stress of 100kPa and subjected to a gradient of 20 across the sample.

Various shear strength tests were also performed on compacted TTDMW-UK samples. Similar tests were not performed on the TTDMW-NS due to lack of available sample. To examine drained strength characteristics, isotropically consolidated undrained (CIU) triaxial tests were performed on saturated samples at effective confining stresses of 50KPa, 100KPa, 200KPa, 300KPa, 400KPa, and 500KPa. Drained direct shear tests at strain rates of less than 0.16 %/min were also performed on submerged compacted samples at initial effective normal stresses of 45KPa, 90KPa, 180KPa, 270KPa, and 360KPa. Direct shear test samples were saturated in a 150mm diameter triaxial cell under an effective confining pressure of 25 kPa prior to sample trimming and testing. Degrees of saturation were greater than 95% for all direct shear samples tested. To evaluate the undrained shear strength characteristics of the TTDMW, unconsolidated undrained (UU) triaxial tests at cell pressures of 100 kPa and 200 kPa were performed. All compacted triaxial samples were prepared utilizing a gyratory compacter to obtain similar densities obtained from standard proctor tests at 2-4% above optimum moisture content.

Mineralogical test procedures

Table 1 provides a summary of mineralogical test methods performed on the two TTDMW samples. Powder pattern x-ray diffraction (XRD) analyses were performed on the minus 0.075mm air-dried fraction of the two TTDMWs to identify the major non-clay minerals present. Clay minerals were identified by performing preferred orientation slides for XRD analysis on the minus 0.002 mm fraction of the soil obtained by centrifugation. Prior to preferred orientation X-ray analysis, various treatments were performed on the air-dried mount samples such as magnesium saturation, ethylene glycol saturation and heating to 550°C. All X-ray analyses were performed with a PW3710 BASED diffractometer, generating copper radiation from a rotating anode source (Department of Earth Sciences, Dalhousie University). The diffractometer was operated at 40 kV and 40 mA with a scan rate of 0.6 deg/min.   Cation exchange capacity analyses (CEC) for TTDMW samples were performed with silver thiourea solution similar to that suggested by Chhabra et al. (1975) while absorbed calcium concentrations were obtained after washing air dried soils with a 5000ppm KCl solution. All cation concentrations were established from atomic absorption measurements performed at the Minerals Engineering Centre at Dalhousie University. X-ray fluorescence (XRF) spectrometry using a Philips PW2510 102 position sample changer (Department of Geology, St-Mary's University) was performed to measure the K_2O composition of the minus 0.075 mm fraction of the samples.

RESULTS AND DISCUSSION

Physical Index testing

Based on the results shown in Table 2, the two TTDMW samples were classified as sandy silt (TTDMW-UK) and silty sand (TTDMW-NS) by the Unified Soil Classification System. To provide an indication of particle texture, high-resolution Scanning Electron Microscope (SEM) was used to examine the TTDMW-UK material, in which an abundant amount of sand and silt sizes were observed (Fig. 1).

Minimal plasticity was evident in the Atterberg Limit tests, especially for TTDMW-NS. The plastic limits of these samples would be considered too low for most compacted clay liner specifications. The specific gravities of the samples (Table 2) were higher than those of typical soils (i.e. 2.6 to 2.9), which was likely due to the presence of barite (specific gravity of 4.2 (Schlumberger, 2005)) used in the original drilling muds. The significant presence of barite in both TTDMW-UK and TTDMW-NS was also confirmed in the x-ray diffraction analysis presented in the following section. The swell index was examined with both de-ionized distilled water and diesel. For both TTDMW-UK and TTDMW-NS, minimal to no swelling was observed with water or diesel on the minus 0.075mm fraction of the material. When bentonite is used as an additive to geosynthetic clay liners or sand-bentonite mixtures, there is often a minimum swell index specified (e.g. min 25 ml of swell per 2 g of bentonite). This swell index is an indication of the hydration capacity of the bentonite to water and is an indirect measurement of the hydraulic conductivity of the bentonite. The TTDMW samples appeared flocculated in water for the swell testing. Even though no significant swelling was observed with diesel for these two materials, an observation of immediate dispersion in the diesel was noted for both TTDWM samples. This hydrophobic nature of the TTDMWs is probably due to the small amounts of residual hydrocarbons remaining in the material after thermal treatment, as well as the organically modified clay present in the original drilling muds.

Mineralogy

XRD random orientation powder patterns for TTDMW-UK and TTDMW-NS showed the primary non-clay minerals were barite and quartz. The high level of barite is not surprising since barite comprises approximately 63% of the minerals and chemicals used in the manufacturing of drilling muds in United States (Crawley et al., 1987). Other major non-clay minerals identified from the powder pattern included calcite, dolomite, and to a lesser extent, feldspars. This large variety of non-clay minerals is not surprising since the drilling process would have encountered a large variety of geological units and hence cuttings from the well development would have been encompassed in the DMW material.

An examination of the XRD traces for the oriented aggregate mounts resulting from the different treatments performed on the samples showed that both materials behaved similarly. Bentonite is a common ingredient in many drilling fluids and hence the general presence of smectites near 14Å-15Å for the magnesium-saturated samples is expected. Swelling of the 14Å peak upon glycolation (especially for TTDMW-UK) and the increase in intensity of the 10 Å peak after heating the sample to 550°C also suggests the presence of smectite. Kaolinite in both TTDMW samples is suggested by the presence of the 7Å peak, which disappears upon heating the air-dried sample to 550°C. Illite is identified in the samples by its basal reflection at 10Å. Although vermiculite and chlorite may be present in the sample, they are most likely present in trace amounts.

As detailed by Carignan (2005), XRD results were combined with the results of the various geochemical test results to estimate the mineral compositions of the samples. Table 3 shows that the amount of barite is higher in the TTDMW-UK than TTDMW-NS, while the specific gravity was higher for the TTDMW-NS (3.6) than for TTDMW-UK (3.0). This could be because Table 3 results reflect the minus 0.075mm fraction of the material (i.e. 57% of the TTDMW-UK and 23%percent of TTDMW-NS). If the whole soil fraction were considered, smectite contents would be reduced by proportioning to 4%

and 3% for TTDMW-UK and TTDMW-NS respectively. The sand sized fraction most likely contains significant amounts of barite.

Chemical Testing Related To Environmental Regulations

Various Canadian environmental regulations were used to assess the suitability of the materials for landfill containment applications. The results of metal concentrations measured on the samples (Table 4) show relatively high levels of barium, chloride (TTDMW-UK), aluminium and iron. Barium concentrations exceeding the values specified by CCME (2003) were observed for both TTDMW-UK and TTDMW-NS (Antimony detection limits were above those of the guidelines). Thallium was exceeded by the TTDMW-UK sample only.

To examine the leaching potential of various metals from the samples if placed in a landfill environment, the samples were also subjected to the Toxic Characteristic Leachate Procedure, TCLP (US EPA, 1992). When Canadian Soil Quality Guidelines for the protection of environmental and human health for industrial land use (CCME, 2003) (Table 4), along with Nova Scotia Canada regulations for disposal into landfills (NSDEL, 1994) and the more stringent Canadian drinking water guidelines (Health Canada, 2003) are used to evaluate the samples, only TTDMW-NS manganese concentrations exceeded the guidelines for disposal of contaminated solids in landfills. TCLP results for both TTDMW-UK and TTDMW-NS exceeded the drinking water standards for aluminium, iron, lead and manganese. Barium concentrations were exceeded for TTDMW-UK, while cadmium concentration was exceeded by TTDMW-NS. More testing will be needed to assess the degree of variability between TTDMW materials. Others (e.g. Tuncan et al. 1997 and Tuncan et al. 2000) have presented chemical characteristics for drill mud wastes, but the wastes had not undergone thermal desorption for stabilization studies.

Geotechnical Performance Based Testing

The hydraulic conductivities of the two samples were similar (Table 6) and slightly higher than the 1 x 10^{-9} m/s value often specified for municipal solid waste landfill liner systems (NSDEL, 1997). The relatively small amount of clay-sized particles compared to traditional clayey liners is one reason for these high values. As discussed by Rowe et al. (2004), the hydraulic conductivity is only one parameter that can affect contaminant migration through engineered liner systems. The slightly high values obtained above do not preclude the use of TTDMW in liner systems if they are considered as part of a composite liner system or utilized with a geosynthetic clay liner (Lake and Rowe, 2005). For some jurisdictions, an hydraulic conductivity less than 10^{-8} m/s would be considered acceptable for a cover system (NSDEL, 1997).

Peak friction angles from both CIU triaxial testing and direct shear testing on the TTDMW-UK sample fell within the range of 36^{o} to 39^{o} (Table 6). Undrained shear strengths from CIU and UU triaxial testing varied from 150 kPa to 230 kPa for the various consolidation pressures tested. Smectite minerals present in TTDMW samples can potentially exhibit low friction angles and undrained shear strengths that can make them unattractive on steep to moderately steep slopes of landfills (Mitchell 1993). The grain-size distribution of the TTDMW sample tested, combined with the mineralogical results, suggest these materials will be stable at the typical slope angles for cover or liner applications.

Implications of Results on the Use of TTDMW for Landfill Containment Applications

The low hydraulic conductivities ($<10^{-8}$ m/s) and peak friction angles greater than 36^o (only TTDMW-UK tested) reflect inherent mineralogical characteristics of the samples. Organic carbon contents of approximately 2% suggest the materials have the potential to sorb VOCs, as reported by Lake and Carignan (2006). One of the main disadvantage of these materials involve the potential regulatory issues associated with the risk of leaching metals such as barium and cadmium from a TTDMW liner or cover system in a landfill application. Moreover ,the compacted TTDMW material had only marginal hydraulic characteristics. Even though hydraulic conductivity values lower than 10^{-8} m/s may be suitable to satisfy regulations for cover system applications, the TTDMW material would not meet the standard 10^{-9} m/s hydraulic conductivity value specified for most bottom liner systems (NSDEL, 1997).

Based on these points, it appears that the potential value-added opportunity for use of the TTDMW in landfill liner systems may be its ability to attenuate VOC contaminants; these are often found in municipal solid waste leachates at low concentrations (Rowe et al., 2004) or its potential for use in cover systems. The issues of leaching and hydraulic conductivity limitations could be overcome by utilizing "composite" TTDMW liner systems in combination with geomembranes and/or geosynthetic clay liners. For example, the harmonic mean hydraulic conductivity of a composite TTDMW/GCL liner system would satisfy the common regulatory standard of 1 x 10^{-9} m/s. The leaching of metals, such as barium and cadmium, could be addressed by underlying the TTDMW material with high-density polyethylene (HDPE) geomembrane. Intact HDPE geomembranes have been shown to be excellent diffusion barriers to inorganic compounds, but have also been shown to have relatively high diffusion coefficients to small VOCs (Rowe et al., 2004). The combination of the TTDMW and HDPE geomembrane would constitute a composite liner where the geomembrane could provide proven hydraulic performance and a thick TTDMW liner could provide VOC attenuation (Lake and Carignan, 2006).

SUMMARY AND CONCLUSIONS

The current paper has presented the results of physical and chemical index tests, mineralogical analyses, chemical analyses, and geotechnical hydraulic and strength tests performed on TTDMW-UK and/or TTDMW-NS samples to assess their likely suitability for use in landfill containment systems. Shear strength tests revealed no major stability concerns for moderate slope angles, although there may be some environmental regulatory issues associated with their use. However, several options are offered to overcome these potential obstacles.

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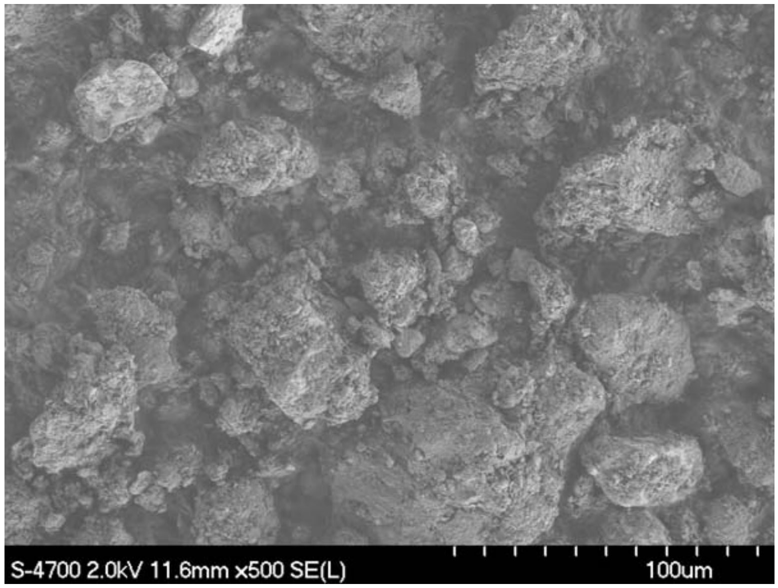
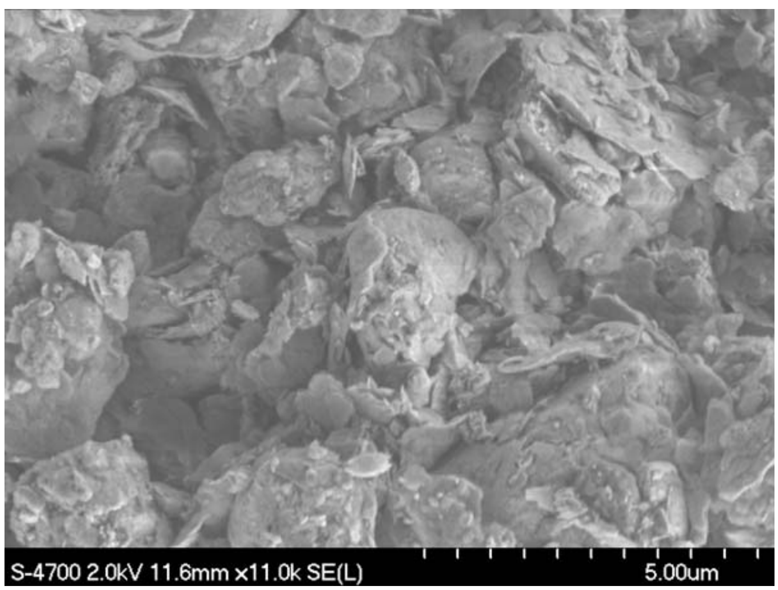
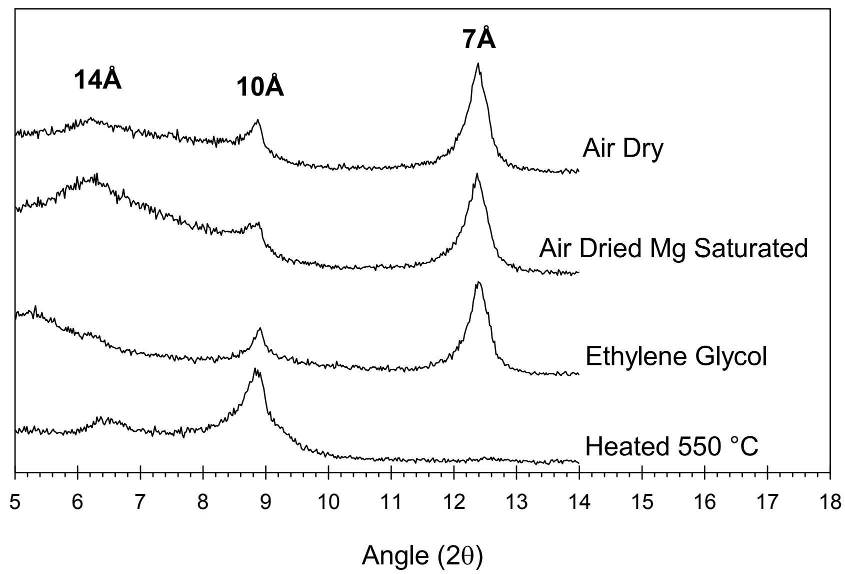


(a)

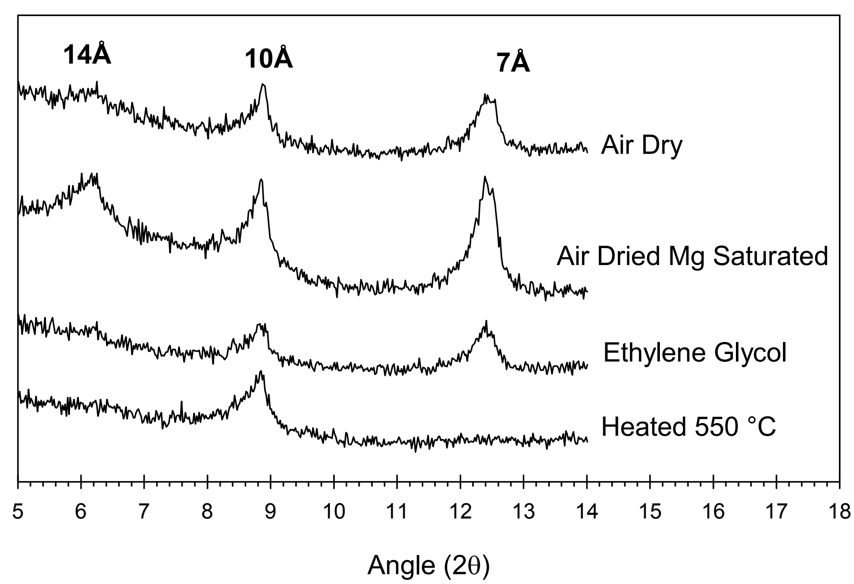


(b)

Figure 1. SEM images, TTDMW-UK



(a)



(b)

Figure 2. Oriented aggregate x-ray diffractograms of (a) TTDMW-UK and (b) TTDMW-NS

Table 1. Tests performed on the TTDMW samples

Test	Method	Size Fraction Tested
Soil metal content	USEPA (1991)	WS
Total Petroleum Hydrocarbons (TPH)	Atlantic PIRI Guidelines (1999)	WS
Toxicity Characteristics Leaching Procedure (TCLP)	USEPA (1992)	WS
Organic carbon	(SSSA, 1996)	WS & <0.075 mm
Specific gravity	ASTM D792	WS
Compaction	ASTM D698	WS
Atterberg limits	ASTM D4318	WS
Grain size	ASTM D422	WS
Swell index	ASTM D5890	<0.075 mm
Hydraulic conductivity	ASTM D5084	WS
Shear Strength	ASTM D3080, D4767, D2850	WS
X-ray diffraction (powder)	USGS, 2001	<0.075 mm
X-ray diffraction (oriented)	USGS, 2001	<0.002 mm
Carbonates	Dreimanis, 1962	<0.075 mm
Cation Exchange Capacity	Chabbra et al. (1975)	<0.075 mm
Glycol retention	SSSA (1986)	<0.075 mm
X-Ray Fluorescence	SSSA (1996)	<0.075 mm

Notes:
[1]WS denotes whole sample
[2]samples compacted at moisture contents ranging from 2 percent to 4 percent above optimum moisture content. Samples subjected to effective confining pressures of 100 kPa to 200 kPa.

Table 2. Geotechnical index and soil chemistry results for the TTDMW samples

Parameter	**TTDMW-UK**	**TTDMW-NS**
Grain size		
Gravel (%)	0	5
Sand (%)	43	72
Silt (%)	47	18
Clay (%)	10	5
Atterberg Limits		
Plastic Limit (%)	26	17
Liquid Limit (%)	34	19
Plasticity Index (%)	8	2
Specific Gravity	3.0	3.6
[1]Maximum Dry Unit Weight (kN/m^3)	17	22
[1]Optimum Water Content (%)	20	13
CEC (meq/100g)	38	17
Specific Surface (m^2/g)	127	178
K_2O (%)	1.2	0.4

Notes: [1]Standard Proctor Energy

Table 3. Semi-quantitative mineralogical analysis of TTDMWs (<0.075 mm)

Mineral	TTDMW-UK (%)	TTDMW-NS (%)
Quartz	17	25
Calcite	8	5
Dolomite	2	2
K-Feldspar	3	1
Na-Feldspar	1	1
Barite	50	45
Illite	9	2
Kaolinite	3	6
Smectite	7	13

Table 4. Soil metal content compared to regulatory values (mg/kg)

Metal	CCME Guidelines Industrial Soils*	TTDMW-UK	TTDMW-NS
Aluminium	-	26,900	4,500
Antimony	40	2	<2
Arsenic	12	10	12
Barium	2,000	**16,900**	**7,100**
Beryllium	8	<5	<5
Boron	-	13	6
Cadmium	22	0.4	0.4
Chromium	87	41	13
Chloride	-	13,500	NT
Cobalt	300	12	4
Copper	91	47	65
Iron	-	25,500	9,200
Lead	600	100	120
Manganese	-	3,525	430
Molybdenum	40	4	2
Nickel	50	35	12
Selenium	3.9	<2	<2
Silver	40	<0.5	<0.5
Strontium	-	525	270
Thallium	1	**2**	0.1
Tin	300	3	NT
Uranium	-	1.3	0.5
Vanadium	130	60	11
Zinc	360	228	100

* CCME (2003)

Note: 1- Results for TTDMW-UK where obtained from Envirosoil (2003). All samples were tested by the private laboratory PSC Analytical Services

2- Bold values indicate excess of CCME (2003) guidelines

3- "NT" denotes Not Tested

4-"<0.1" denotes not detected above the given detection limit

Table 5. Metal leachate extraction analysis (µg/L)

Analyte	Guidelines Leachate*	Guidelines Drinking Water**	TTDMW-UK	TTDMW-NS
Aluminium	500,000	100	**610**	**5100**
Antimony	-	6[1]	<20	<20
Arsenic	5,000	25[1]	<20	<20
Barium	100,000	1,000	**1,300**	660
Beryllium	10,000	-	<50	<50
Boron	500,000	5,000[1]	600	<20
Cadmium	500	5	4	**7.5**
Chromium	5,000	50	<20	<20
Cobalt	5,000	-	<10	24
Copper	100,000	≤ 1,000[2]	81	100
Iron	30,000	≤ 300[2]	**2,000**	**810**
Lead	5,000	10	**143**	**120**
Lithium	250,000	-	61	30
Manganese	5,000	≤ 50[2]	**1,800**	***8,200***
Molybdenum	5,000	-	<20	<20
Nickel	20,000	-	22	39
Selenium	1,000	10	<20	<20
Silver	5,000	-	<5	<5
Strontium	-	-	1,700	2,500
Thallium	-	-	<1	<1
Tin	-	-	<20	<20
Uranium	2,000	20[1]	<1	<1
Vanadium	10,000	-	<20	<20
Zinc	500,000	≤ 5,000[2]	825	1,800

* NSDEL, 1994
** Health Canada (2003)
[1] IMAC (Interim Maximum Acceptable Concentration)
[2] AO (Aesthetic Objective)

Note: 1- Bold values indicate excess of Health Canada (2003) guidelines
2- Italic values indicate excess of NSDEL (1994) guidelines
3- "<0.1" denotes not detected above the given detection limit

Table 6. Geotechnical tests results for the TTDMW samples

Material	Hydraulic Conductivity (m/s)	Peak Friction Angle (º)	Undrained Shear Strength (KPa)
TTDMW-UK	[1]3×10^{-9}	36[3]	150-200[3]
	-	39[4]	180, 230[5]
TTDMW-NS	[2]2×10^{-9}	-	-

[1]Average value of 2 samples, standard deviation of 6×10^{-10} m/s
[2]Average value of 3 samples, standard deviation of 2×10^{-10} m/s
[3] Consolidated Undrained Triaxial Tests
[4] Direct Shear Tests
[5] Unconsolidated, Undrained Triaxial Tests