

Soil Washing for Remediation of Non Volatile Hydrocarbon Polluted Soils

G. Belardi*, N. Shehu, L. Passeri

Istituto Trattamento dei Minerali, Via Bolognola, 7-00138, Roma, Italy

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ABSTRACT

This paper illustrates the possibilities of applying what is essentially a physical soil-washing technique for treatment of soil contaminated by semi-volatile hydrocarbons in order to reduce the level of pollution to that permitted by Italian regulations.

The ground around a factory producing lubricating-oil additives consists of silty-sandy soils, which have long been contaminated by heavy hydrocarbons, especially branched alkyl benzenes and naphthalene derivatives. Pollution is particularly marked in some zones (hot-spots) where Total Petroleum Hydrocarbon (TPH) values are as high as 7000ppm. The hydrogeological characterization showed the presence of a gravely soil in a sandy/silty matrix, with big stones making preferential paths to the water circulation. The permeability is 10^{-5} m/s and the water-table occurs at about 3m below ground level. After laboratory and pilot-scale studies on representative samples of in-situ soils with TPH between 2700 and 7000ppm a flow-sheet is proposed for reducing TPH values below the acceptable limits in Italy, namely 700ppm for hydrocarbons with more than 12 carbon atoms and less than 250ppm for those with fewer. Recovery of remediated soil to be returned to the site is around 90%. © 2002 SDU. All rights reserved.

Keywords: Soil-washing; Organic pollutants; Branched alkyl benzenes; Naphthalene derivatives

1. INTRODUCTION

Organic pollutants in soils are listed under three main headings:

- Volatile and non-volatile aromatic and aliphatic hydrocarbons such as oils and benzenes
- Polycyclic aromatic hydrocarbons (PAH)
- Halogenated and non-volatile compounds such as trichlorethylene, perchlorethylene, pesticides and biphenylpolychlorides (PCB).

One of the most frequent organic pollutants in soils is fuel oil which is widely used for energy production and as a commercially-important primary raw material. Organic compounds are also present in various industrial products, for instance, PCBs, which are used for the production of paints, inks, cooling fluids and protective coatings for timber.

The site of concern in this paper is mostly contaminated by branched alkylbenzene and naphthalene derivatives. These compounds were introduced into the soil through tank overflow, container dripping and on-site disposal of unwanted fractions.

The industrial activity, was based on the distillation of various coal fractions including oily fraction, tar and pitch. Some of the TPH contained in these products such as PAH, are considered to be cancer-causing or potentially cancer-causing agents.

The major concerns with contaminated soil at industrial site are potential health and environmental risk. Not only may people be exposed to the contaminants through direct contact to soil particle, but contaminants may also migrate into the groundwater creating an expanding zone of contamination.

* Corresponding author. E-mail: belardi@itm.rm.cnr.it

Three tiers of contaminated soil treatment are in use today, namely traditional treatment technologies, alternative technologies and emerging treatment technologies. Traditional treatment consists of landfilling (Ellis *et al.*, 1986), incineration (Ellis *et al.*, 1986), and solidification/stabilization (Conner, 1994; Brevster *et al.*, 1995; Misra *et al.*, 1995). Alternative technologies include low-temperature thermal treatment (Misra *et al.*, 1995), bioremediation (Groudeva *et al.*, 1993; Misra *et al.*, 1995; Eccles *et al.*, 1996; Groudev *et al.*, 2000; Pèrè, 2000; Porta, 2000) (biosparging, bioslurping, bioventing, biopile, land farming), vapour extraction (Porta, 2000), soil-washing (Scholz and Milanowski, 1983; Sonnen *et al.*, 1989; Hsieh *et al.*, 1989; Valine *et al.*, 1989; Hsieh *et al.*, 1990; ; Masters *et al.*, 1991; Cline *et al.*, 1993; Burson and Elston, 1994; Krstich *et al.*, 1994; Sadler and Krstich, 1994; Cline and Reed, 1995; Griffiths, 1995; Krstich and Gilson, 1995; Tiefel and Schricke, 1995; Abumaizar and Khan, 1996; Semer and Reddy, 1996; Van Benschoten *et al.*, 1997) and chemical extraction (Brevster *et al.*, 1995; Fristad, 1995; Davis and Singh, 1995; Misra *et al.*, 1995; Ganguly *et al.*, 1998; Steele and Pictnel, 1998; Peters, 1999; Djordem *et al.*, 2000). Emerging technologies currently include in-situ vitrification (Porta, 2000), RF processes (Porta, 2000), dechlorination (Porta, 2000), soil ashing (Porta, 2000), electrokinetic (Shapiro *et al.*, 1989; Acar, 1992; Hichs and Tondorf, 1994; Misra *et al.*, 1995; Li and Neretnicks, 1998; Djordem, 2000), permeable reactive barrier (Diependall *et al.*, 1993; Sidkar *et al.*, 1998; Porta, 2000), cyanide oxidation (Porta, 2000), hot air injection (Porta, 2000), hot water extraction (Qi Dai *et al.*, 1998; Porta, 2000), phytoextraction (Misra *et al.*, 1995; Huang *et al.*, 1997; Porta, 2000), flushing (Anderson, 1993; Misra *et al.*, 1995; Peters *et al.*, 1995; Porta, 2000), air sparging (Porta, 2000), and dual-phase extraction (Porta, 2000).

Because of limited extent of the high contamination in depth (about 2-3m), limited surface (about 2500m²) and the scarce biodegradability of the contaminants it was considered to eliminate the pollution through an economical soil treatment technology such as soil washing. In the environmental studies soil-washing is today defined as a physical/chemical separation process to remove or extract contaminants adsorbed or bound to soils.

Two different commercial approaches have evolved: 1) a primary physical-chemical separation system that treats specific soil fractions, and 2) a primary chemical system that uses various reagents to solubilize contaminants into an aqueous phase that can be treated as a wastewater stream. Both approaches have the same ultimate objective, namely, to return the largest possible volume of soil to the site as "clean" while generating the smallest volume of concentrated residuals.

The physical approach is based on mining and mineral processing principles which involve operations such as dispersion/attrition, particle-size separation, gravity treatment in static and centrifugal systems, electrostatic and magnetic separation, solid-fluid separation and thermal treatment. For environmental/hazardous waste applications, such an approach is very logical since the relevant contaminants exist in specific particle-size fractions in reasonably predictable ways.

Soil-washing can generally be considered feasible for the treatment of soils containing:

- Heavy metals, including lead, chromium, nickel, cadmium, copper, zinc and cyanides;
- "Special" metals, including arsenic and mercury;
- Semi-volatile organics, polynuclear aromatic (PNA) compounds, typically including naphthalene, anthracene, crysene, and the benzo(x) derivatives.

The benefits of soil-washing are substantial, namely:

- The system is exceptionally cost-effective, since it focuses only on the treatment of certain fractions, rather than on the entire waste volume
- The system can treat both organics and inorganics in the same treatment stream
- The soil washing system is a true volume reduction option (volume reduction of greater than 80% on a dry solids basis), and directly supports the recycle and reuse of site materials
- The system complies with current Italian regulations
- Since there is no air emission or wastewater discharge, the system is more readily permitted than traditional remedial alternatives.

Some of the key issues to consider during soil washing are:

- Size of project. On-site technology applications are directly dependent upon volume as an economic parameter. For full-scale soil-washing a volume of more than 5000 tons is required to compete economically with other "conventional" remediation alternatives. For projects where "conventional" alternatives are limited by unusual site conditions or wastes, the minimum volume may be smaller. Smaller volumes can be cost-effectively handled by a pilot plant in some cases
- Particle size/contaminant relationship. The better the natural distribution of coarse and fine-grained materials, the more economical soil washing becomes. Soil washing is not a set, rigid treatment train, but is modified specifically for the actual wastes to be treated. Very substantial volume reduction can be obtained by understanding the particle-size/contaminant relationship and merely by screening and separating wastes for the most appropriate treatment
- Contaminants. Primary candidate contaminants for soil-washing are heavy metals, semi-volatile organics (THP), PNAs, pesticides, PCBs, and low-level radioactive wastes
- The position of the state regulators in selecting on-site approaches

A number of soil washing techniques have been developed and field tested, including the BIOTRAL (Biological Aqueous Treatment System) (EPA/540/R-93/S26), the B.E.S.T. solvent extraction technology (EPA/540/R-93/S26) and the Harman Environmental Services Soil Washing Technique (EPA/540/S-89/013).

Many soil-washing field demonstrations conducted to date have been focused on removing volatile and semi-volatile organic materials from contaminated soils. Soil-washing has achieved 90-99% removal of volatiles and 40-90% removal of semi-volatiles with 85% volume reduction (EPA/S42/R-92/012).

The average cost (including excavation, product management activities, closure activities) for soil washing typically ranges from US\$120 to US\$200/t of soil treated (EPA/S42/R-92/012; EPA-823-B93-S26; EPA/540/R-93/S26; EPA/540/S-89/013).

Cost will vary depending on the volume of soil treated and the fines percentage of the soil.

The purpose of this paper is to propose a methodology of physical soil-washing, which permits the treatment of soils badly polluted with TPH. The method is based on the use of simple, economic schemes which ensure not only highly-efficient pollutants extraction but also very good recovery of remediated soil for return to site.

The study has been conducted in various stages with the aim of:

- checking the efficacy of preliminary dispersion or attrition for the extraction of hydrocarbons present in the soils;
- checking the efficacy of flotation;
- checking the possibilities offered by the application of various gravity separation techniques;
- establishing a final scheme suitable for industrial application.

2. MATERIALS AND METHODS

2.1. Soil preparation

The bulk soil for this study was obtained from selected "hot spots" to ensure that a highly-contaminated sample was collected.

Bulk soil sample up to a depth of 2-3m was collected by a mechanical excavator in a sampling trench. This material was allowed to air dry for 4 days and then dry per-screened to remove the gross oversize fraction (>5mm) staged for recycling after meeting testing requirements.

The coarse fraction of the soil (> 5mm) eventually still out of the law limits can be treated by land farming amended the soil with municipal solid waste compost and fertilized with a solution of ammonium nitrate and sodium phosphate. From studies carried out by Enitecnologie S.p.A. (Fabiani *et al.*, 2001) this treatment led the contaminants in the soil within the Italian law target after 120 days.

The 5mm soil fraction was utilized for experimentation throughout this study. This soil fraction was initially thoroughly homogenized to provide a uniform sample with respect to pollutants concentration. The homogenization process was initiated by manual mixing using a shovel. After homogenization, the soil was passed through a Gilson sample riffler to obtain thoroughly homogenized samples. The samples were then placed in plastic containers to maintain their natural moisture content, and stored at room temperature (about 25°C). Samples used for chemical characterization analysis were held at 4°C.

2.2. Soil analysis

Particle size analysis of the soil samples was performed using sieves of the ISO series and a Sympatec Helos laser granulometer equipped with a He/Ne laser. Total hydrocarbon content was ascertained by means of a DANI 1000 gas chromatograph.

The procedures adopted for preparing samples for analysis were as follows: Representative soil samples were first dried using anhydrous sodium sulphate and extracted with methylene chloride after being subjected to sonic treatment for 15 minutes. The centrifuged supernatant was analysed in a gas chromatograph under the following conditions: ramp at 40°C for 2 minutes, 150°C at 3°C/minute, 150°C for 5 minutes, 280°C at 5°C/minute, 280°C for 5 minutes, detector (FID) at 300°C.

Thermogravimetric analyses were performed with a Stanton 1500 thermobalance. The samples were analysed using the following operating conditions: ramp from room temperature to 700°C at 20°C/minute at atmospheric pressure.

Mineralogical analyses were carried out by means of X-ray diffraction on powders finer than 75µm using a Siemens D500 diffractometer. Tap water was used in all the tests. Static sedimentometers, filter-presses and Buchner funnels were used for solid-liquid separations.

2.3. Rotary-scrub mechanical stirring (dispersion)

Dispersion tests were run using a Denver flotation cell with the air switched off. Representative soil (table 1) samples weighing about 1000g were treated in a pulp containing 40% solids. Dispersion time was held at 10 minutes at an impeller speed of 1200rpm.

Table 1
Particle size distribution and TPH analysis of representative soil sample

Size (mm)	Weight (%)	TPH (ppm)	TPH Distribution (%)
-5 + 2	17.1	447	1.79
-2 + 0.6	31.5	1112	8.22
-0.6 + 0.2	16.4	1117	4.30
-0.2 + 0.063	13.6	1515	4.83
-0.063	21.4	16.113	80.86
Feed	100.00	4264.13	100.00

As can be seen, 80% of the hydrocarbons occur in the minus 63µm fraction which accounts for 21% (w/w) of the sample.

2.4. Attrition scrub apparatus

The attrition scrubbing tests were performed using a WEMCO laboratory attrition scrubber fitted with a high-torque variable speed drive motor and a stainless steel shaft. The attrition apparatus consisted of three bladed impellers and a close-fitting stainless-steel tank with a lid. To optimise the effectiveness of the WEMCO laboratory attrition scrubber, a regulated volume of soil was added to the stainless steel tank. Approximately 1000 grams of moist <5mm soil was used, the appropriate amount of water being added to produce a slurry containing 80% to

85% solids. The speed was set to approximately 1200rpm and attrition was carried out for 10 minutes.

2.5 Wet shaker table

The shaker table experiments were conducted on a Wilfley laboratory concentrating table with a model 13A sand deck. The differential shaking motion was provided by a gear connected to a pulley which was driven by an electric motor.

Water was added to the pre-sized soil (<0.5mm) to form a 20% solids mixture which was fed to the table. Table feed rate, cross-water flow, table-tilt, speed and stroke were adjusted to obtain proper separation based on visual inspection. The "bands" of material flowing off the table were isolated by sample cutters and discharged into containers as tabled samples.

2.6. Flotation cell

Flotation tests were performed in an aspirated KHD Humboldt Wedag AG type MN 935/4 cell equipped with interchangeable agitator-set consisting of the rotor and stator with disintegrator blade made of corrosion resistant material.

Plexiglass flotation containers of 1.2L capacity were employed. Pulp density was adjusted at 35% solids and the impeller speed was fixed at 1200rpm. The amount of air aspirated was kept at 3.9L/min.

2.7. Hydrocyclone

The hydrocyclone used in the laboratory experiments was a Mozley model C124 two inch hydrocyclone in abrasion-resistant polyurethane with field-adjustable cones, bands, and vortex finder such that the "cut-point" interface between coarse and fine-grained materials can be modified to be consistent with treatment requirements. In all the tests the cyclone was equipped with a 11mm vortex finder cap and 6.4mm spigot cap. Operating pressure was 2 atmospheres with feed pulp densities of 15% solids (w/w) which permitted a d50 cut of about 12 μ m.

2.8. Multi-gravity separator (MGS)

A Mozley C900 laboratory MGS separator was adopted. This consists essentially of a slightly tapered open-ended drum 0.6m long and 0.5m in diameter. The drum rotates in a clockwise direction at speeds varying between 150 and 300rpm enabling a centrifugal force equivalent to a gravitational pull of between 6 and 24g to be generated at the drum surface, depending on the nature of the material treated.

The rotational speed of the drum affects the operation of the MGS in two ways. An increase in speed results firstly in an increase in the flow-rate of the slurry in an axial direction towards the tailings end. Secondly, it increases the inertial mass of the mineral particles, increasing their tendency to pin to the drum wall and form a solid layer. A sinusoidal shake with an amplitude varying between 12mm and 25mm in an axial direction, with a frequency between 4 and 6cps, is superimposed upon the motion of the drum. The effect of shaking is to impart an additional shearing action on the particles; this aids the separation process.

The angle between the drum axis and the horizontal, the angle of tilt, may be adjusted between 0 and 10 degrees. The drum consists of a stainless steel cylindrical shell, closed at one end, with a centrifugally cast polyurethane lining. The lining forms a cone, narrowing towards the open, or outer end with a half angle of one degree.

The MGS is fitted with a total of 34 scrapers. Four scraper bars, spaced at 90 degree intervals parallel to the axis of the drum, each hold 8 or 9 scrapers equally spaced along their length. Each scraper is 65mm long and is lightly spring-loaded to just touch the surface of the drum. The edge of each scraper is carefully profiled to match the profile of the drum. All scrapers are set at an angle of 60 degrees with the drum axis and are in such a way arranged that the

“trailing edge” of each overlaps the “leading edge”. During operation the scraper mechanism revolves 2.5% faster than the speed of the drum. Thus the scrapers move slowly across the surface of the drum, completing one revolution of the drum surface for every 40 revolutions of the drum.

As each scraper moves across the drum surface it ploughs into the layer of heavy mineral particles pinned to the drum surface. Because the scrapers are angled the particles are pushed up the slope towards the open end of the drum. Each scraper moves the heavy mineral particles approximately 35mm. As the “scraped” particles leave the trailing edge of one scraper they are picked up and conveyed a further 35mm by the next scraper. As a result of this action the heavy particles are eventually scraped to the open end of the drum where they are discharged into the concentrate launder. The scrapers are manufactured with the same pour-moulded polyurethane as the drum lining.

Feed slurry is introduced continuously mid-way onto the internal surface of the drum via an accelerator launder in order to reduce turbulence caused by the introduction of the feed. Wash water is introduced via a similar accelerator launder close to the outer end of the drum. Observation indicates that the slurry follows a spiralling pattern on the revolving drum surface. Heavier particles, or particles of higher specific gravity, penetrate the slurry and are pinned to the surface of the drum as a result of the centrifugal forces to form a semi-solid base layer. An intermediate layer forms above this consisting of a relatively dilute suspension of lower specific gravity particles and slime particles. The top layer consists of relatively clear water. The shake provides an additional shearing force on the particles in the flowing film, resulting in improved separation, whilst the specially designed scrapers moving across the drum surface continually re-grade the settled particles, thus minimizing entrainment of gangue.

The high density particles pinned to the surface of the drum are therefore continuously swept up the slope by the scrapers to discharge at the open end as the concentrate. The lower density minerals (or gangue), along with most of the wash water, flow downstream to discharge as tailings via slots at the inner end of the drum. Both products are collected in circumferential launders which discharge below the machine.

Because the centrifugal acceleration imparted to the particles is many times greater than the normal gravitational force acting on a conventional shaking table, separation of the dense and light particles into layers is greatly accelerated.

Wash water is added close to the open end of the drum. The ploughing action of the scrapers turns over the mineral layer allowing entrained lower density gangue particles to be removed by the wash water as the mineral layer leaves the trailing edge of each scraper.

Optimum settings of the operating variables were identified in preliminary tests.

3. RESULTS AND DISCUSSION

3.1. Soil characterization

The soil sample can be classified as sandy-silty with a natural moisture content of about 10%.

The main mineral constituents are anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), quartz (SiO_2), clinoclone ($\text{Mg}_5\text{Al}(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_8$), magnesium-hornblende ($\text{Ca}_2(\text{MgFe})_5(\text{SiAl})_8\text{O}_{22}(\text{OH})$) and muscovite 3T ($(\text{KNa})(\text{AlMgFe})_2\text{Si}_{3.1}\text{Al}_{0.9}$), while there are traces of calcium albite ($(\text{NaCa})(\text{SiAl})_4\text{O}_8$), as is evident from the Figure 1 diffractogram.

The principal organic pollutants are cumene, 2-methyl propyl benzene, 2-methyl butyl benzene, 1,1-dimethyl, 4-methyl indene, diisopropyl benzene, 2-methyl tetrahydrophenanthrene, PM 206, methyl naphthalene and methyl diphenyl, as reported in the Figure 2 gas chromatogram.

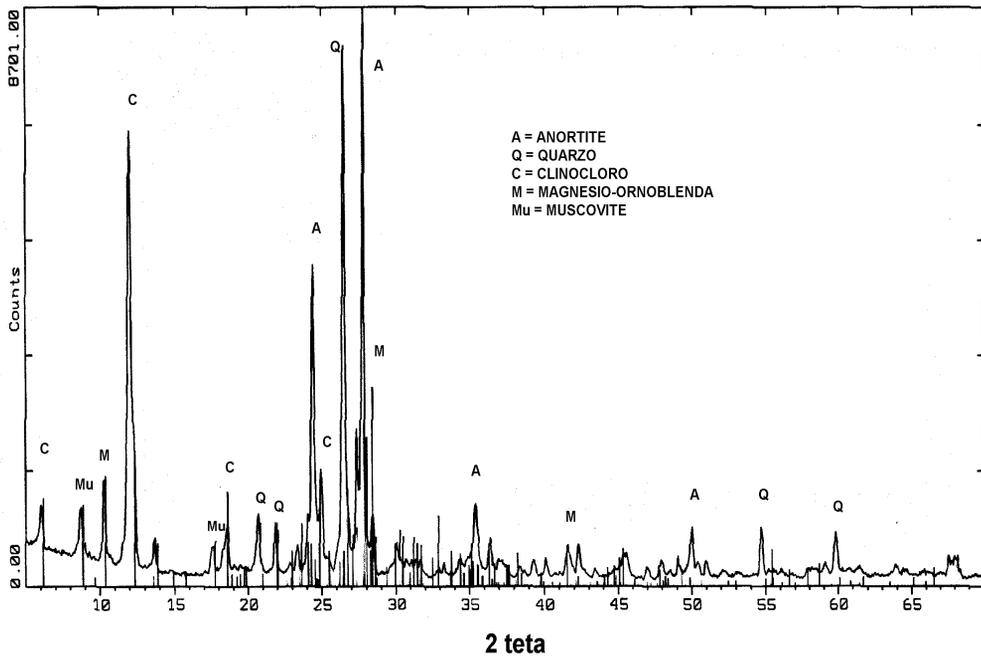


Figure 1. Diffractogram of representative soil sample used in laboratory tests

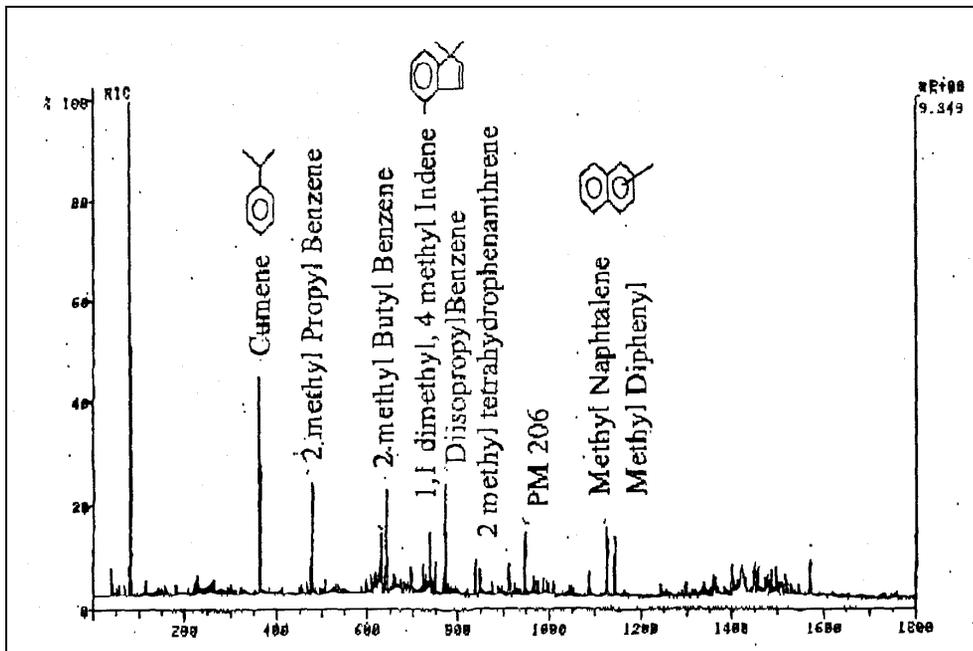


Figure 2. Gas chromatogram of organic pollutants present in representative soil sample

The preponderant presence of non-volatile hydrocarbons is confirmed also by the thermogram of a sample of hydrocarbon having the same composition as that present in the soil and illustrated in Figure 3.

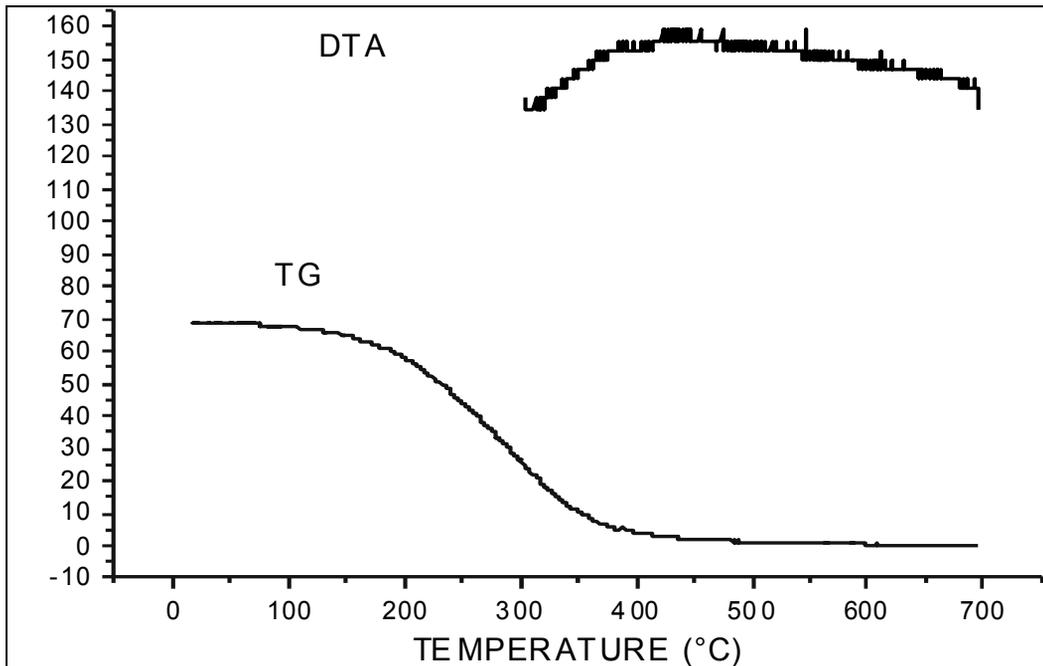


Figure 3. Thermogram of sample representative of organic pollutant present in the soil

The particle size analysis of a representative soil sample is shown in Table 1 together with TPH analysis and distribution in each size fraction.

3.2. Comparative tests

The purpose of these tests was to examine the efficacy of attrition compared with dispersion (mechanical stirring) as regards TPH extraction via flotation using various reagents available on the market. The attrition of particle suspensions for the cleaning of the surface of particles contaminated by adsorbed pollutants can now be considered state-of-art technology (Marini *et al.*, 1997; Neil and Bridgwater, 1994; Schricke, 2000; Schriker and Neesse, 1998; Tiefel *et al.*, 1999; Van Benschoten, 1994). Furthermore, remediation of oil-contaminated soil by froth flotation has many advantages over clean-up processes (Komechi *et al.*, 1998; Niewiadomski *et al.*, 2000). Flotation can be cheap, fast and efficient if optimal conditions are found first to release oil from the soil and then to float it from the process water.

The following surfactants were used in the tests:

S-3 107 Cyanamid (cresylic acid), Aerofroth 77 Cyanamid (mixed C5 to C8 alcohols), sodium oleate, Armac C Akzo Chimie (cocoamine acetate), Aero 825 Cyanamid (anionic petroleum sulphonate), Linaflot (mixture of sodium oleate and aliphatic amines), and pine oil by Cyanamid.

The reagents were prepared in 2% aqueous solution using demineralized water.

The tests were conducted on the basis of increasing reagent additions (from 0 to 1400g/t). Conditioning time was 2 minutes, flotation pH was natural (pH = 6.88-7) and flotation time was 5 minutes.

The results of the most significant tests are summarized in Figures 4 and 5 which also show the remediated soil recovery and the TPH values.

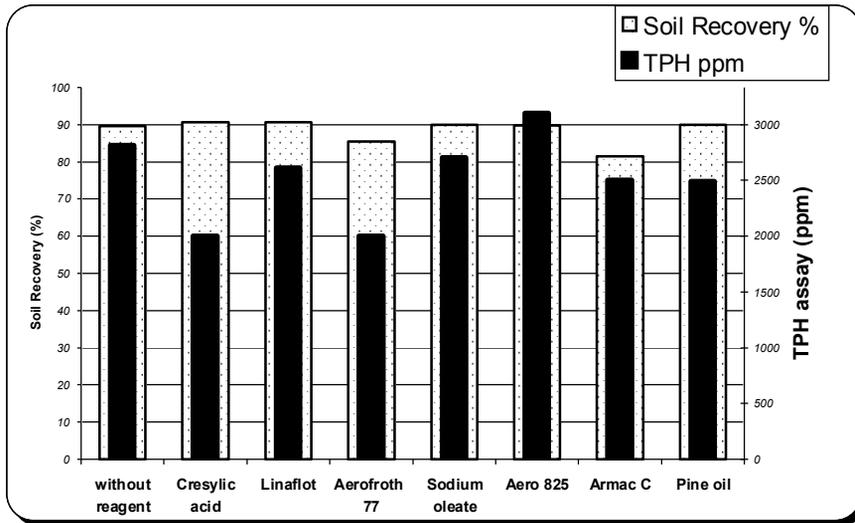


Figure 4. Recovery and TPH values of remediated soil subjected to dispersion and flotation using various reagents

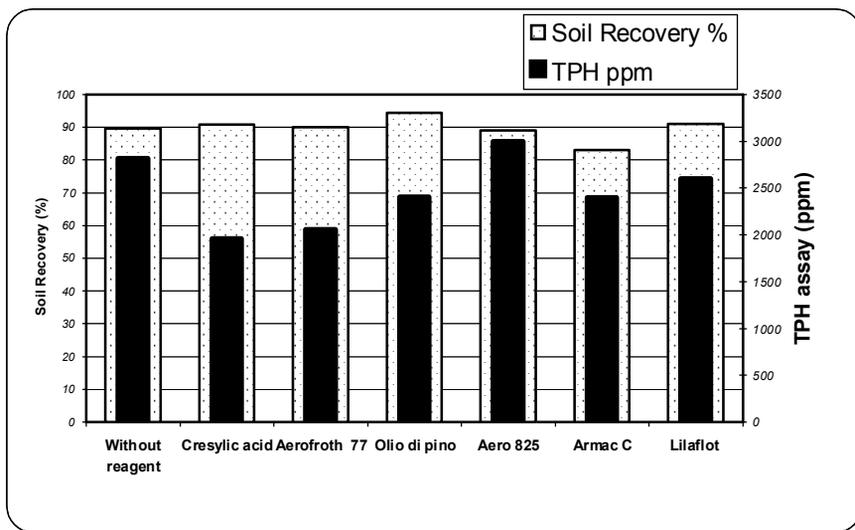


Figure 5. Recovery and TPH values of remediated soil subjected to attrition and flotation using various reagents

With mechanical stirring (dispersion) and flotation the TPH of the treated soil is reduced from 6300ppm to less than 3000ppm, while recovery of hydrocarbons in the float is in the 60 to 70% range. Some authors report (Zhang *et al.*, 1998; Niewiadomski *et al.*, 2000) that the zeta potential of the heavy oil droplets at neutral pH in deionized water without surfactants is negative. With anionic surfactants the zeta potential becomes even more negative as their concentration in solution increases but under the same conditions with cationic reagents, the zeta potential changes from negative to positive. Non-ionic surfactants reduce the negative charge of the oil droplets.

It emerges from the tests that cationic and anionic surfactants give comparable results, but the detergency action of the anionic collector is less efficient. This can be attributed to the fact that excessive adsorption of surfactant hydrophilized oil droplets, and attachment with air bubbles, is lessened. Low oil removal from the surface of sand particles by cationic collector is probably explained by electrostatic interactions of cationic surfactants with negatively charged

silica and feldspat minerals, while it is likely that the slightly better recovery achieved with the use of non-ionic surfactant can be attributed to the coalescence of oil droplets.

TPH extraction efficiency is also influenced by the fact that although the soil is not very clayey it does contain fines which may influence the process because of undifferentiated action of the collector and marked adsorption of oil on these particles.

Attrition greatly improves flotation efficiency. The mineral surface must be made amenable to cleaning, i.e. to abrasion, in the case of attrition. The intensive particle contact, existing at such high solids concentrations, creates a heavy stress on the particle surface which removes the uppermost particle layer and causes adsorption of the contaminants.

As cresylic acid appeared to be the most influential surfactant on oil recovery in the float after attrition, comparative tests were run using 1400g/t of this reagent. Some tests were performed at room temperature adding finely ground South African coal at a rate of 4% (w/w) of the material treated in the dispersion or attrition stages. It has been proven that adsorption of hydrocarbon type contaminants can occur on finely ground coal added to the soil water slurry (Szymocha *et al.*, 1995; Szymocha *et al.*, 2000). This action is attributed to the high affinity of organic compounds towards coal surfaces. In order to improve separation kinetics and efficiency the process is often performed at elevated temperature (85°C) (Szymocha *et al.*, 2000). However, in this study the tests were performed at room temperature (20-25°C).

The results are summarized in Figure 6 which reveals that coal favours TPH extraction following mechanical stirring but there is no marked difference if it is added in the attrition stage. In any case, attrition alone, followed by flotation ensures soil recovery of 90.83% (w/w) and a TPH value of 1962ppm.

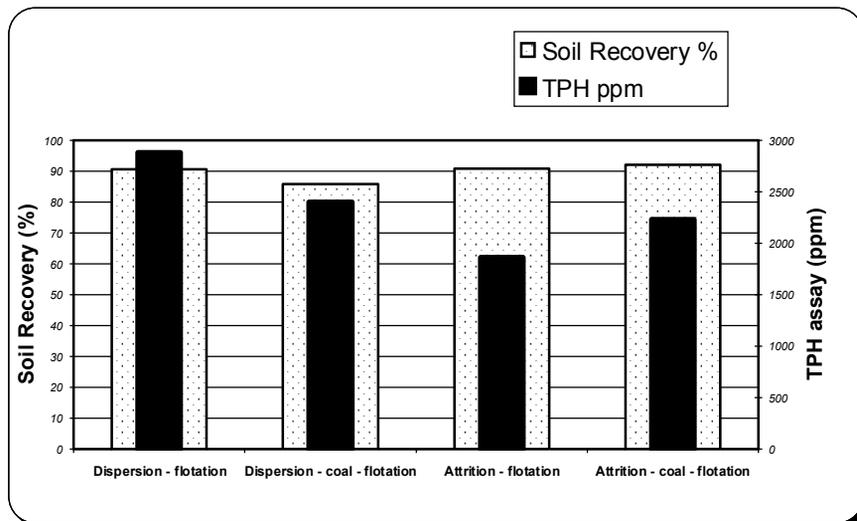


Figure 6. Recovery and TPH values of soil treated in various ways by flotation with 1400g/t cresylic acid

3.3. Gravity separation tests

The need to separate the fine fraction of the soil has been demonstrated by screening at 45µm the tailings remaining after cresylic acid flotation the TPH value in the -0.5 + 0.045mm fraction is 881ppm, of which 800ppm +C₁₂ and 81ppm -C₁₂, with remediated soil recovery of 69.43%

Preliminary tests showed that wet screening at 0.5mm performed after attrition can improve results of wet gravity separation. It also emerged that the -5+0.5mm fraction, which accounts for some 50% of the feed, has an average TPH of 850ppm; this is in line with Italian regulations so the material can be re-used on site. There is also the fact that the -0.5mm fraction is ideal for many wet gravity separation processes.

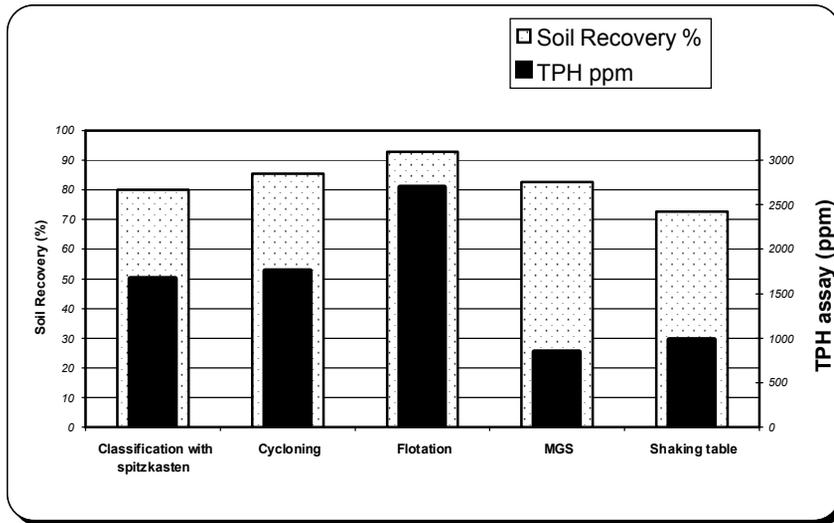


Figure 7. Recovery and TPH values of soil treated by attrition and screening at 0.5 mm then subjected to various gravity and size separations

Another point to underline is that organic pollutants are probably adsorbed on particles finer than 45 μ m, some of which do not float even when large quantities of surfactants are added.

The need to identify the best wet gravity separation process arises from the necessity to achieve maximum recovery of remediated soil. A series of comparative wet gravity separation tests was thus run on the -0.5mm fraction after attrition. The types of equipment used for wet gravity separation were surface sorting (Spitzkasten), cyclones, Multi Gravity Separation (MGS) and shaking tables. A comparative test with cresylic-acid flotation was also performed. The results are illustrated in Figure 7 which reveals that separation involving centrifugation gives good results as regards TPH grade and recovery of remediated soil.

3.4 Final tests

Optimized tests were performed on the basis of the pointers emerging from the work reported above. The most significant process is indicated in Figure 8 which also shows the operating conditions adopted in each stage of the circuit and the results obtained.

It is evident that physical separation ensures extraction of non-volatile hydrocarbons in excess of 92%.

A reusable soil with a pollution level within the limits set by Italian regulations is thus obtained, while the unusable fraction amounts to no more than about 10% (w/w).

On the basis of biodegradation studies performed in lab and pilot scale, a scheme of an integrated treatment of heavy contaminated soils (unusable fraction) has been designed forwarding the slurry to the wastewater treatment plant of the factory where the active sludge microbial population is able to degrade the contaminants. Biodegradation studies of the contaminants in the slurry were carried out by Enitecologie S.p.A. (Fabiani *et al.*, 2001) using a laboratory equipment (Vittadini Mark-2 mini pilot plant) that reproduces an activated sludges wastewater treatment plant. The mini pilot plant was started up by using the activated sludges and the wastewater generated by the industrial plant operating at the site. It was verified that the output parameters (COD, TPH and suspended solids) were below the Italian law levels for treatment wastewaters pointing out that this innovative physical-biological process is considered suitable.

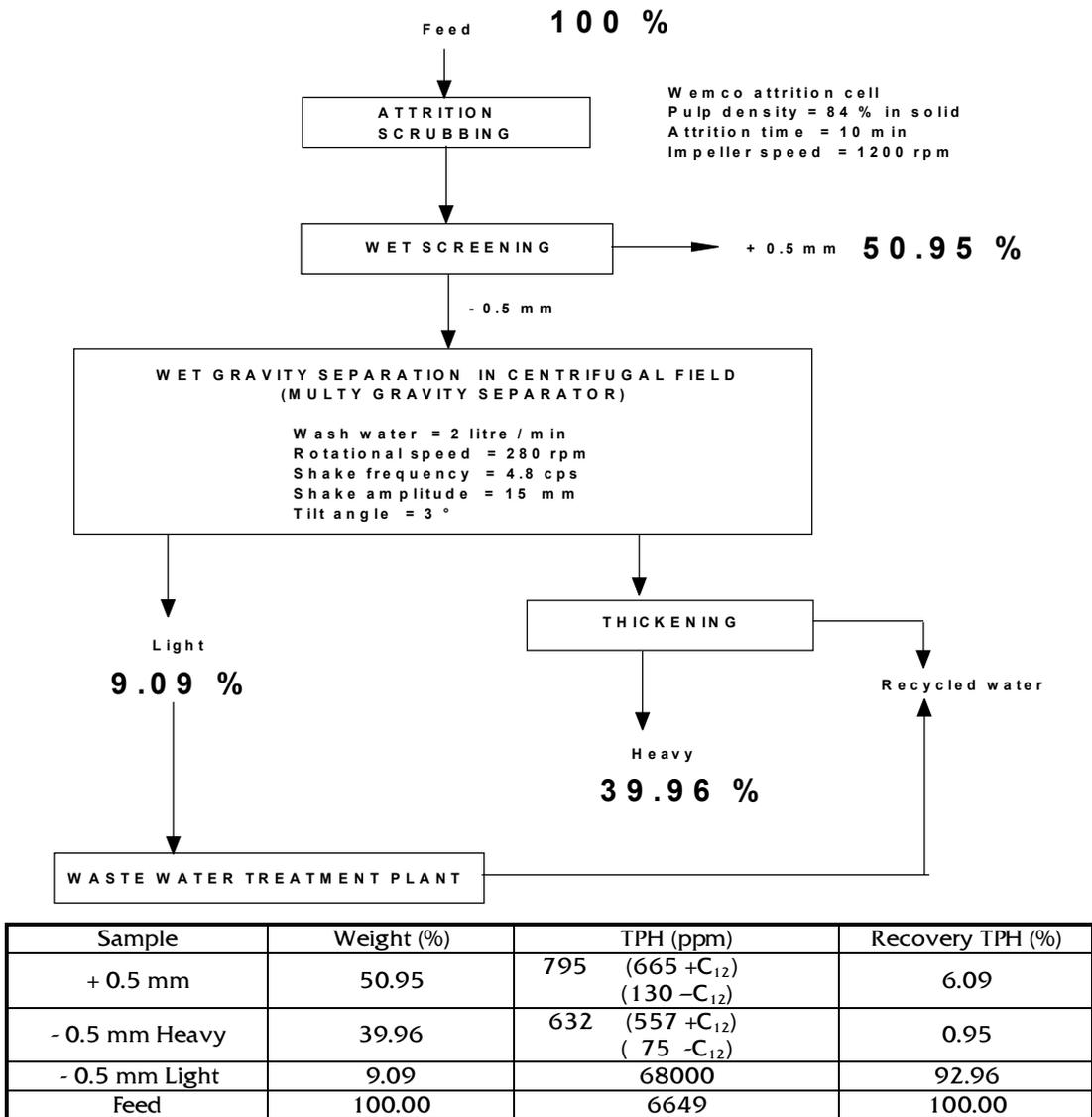


Figure 8. Laboratory scale flowsheet

4. CONCLUSIONS

On the basis of indications provided by laboratory tests a mainly physical treatment process of soil washing has been developed. This is used to clean-up soil from a site polluted by non-volatile hydrocarbons with a high TPH value. The process involves a high-density attrition stage followed by wet screening at 0.5mm and then wet gravity separation of the minus 0.5mm fraction in the centrifugal field.

The gravity separation products consist of a light fraction with particle size finer than 15µm which contains most of the pollutants, and a heavy coarse fraction with TPH values conforming to the limits set by Italian regulations (less than 700ppm hydrocarbons whose structure contains more than 12 C atoms, and less than 250ppm of hydrocarbons with less than 12 C atoms). Total recovery of soil with pollution values within the set limits exceeds 90%, while total extraction of hydrocarbons is higher than 92%. The fraction which must necessarily be rendered inert accounts for 9.09% (w/w) of the feed. The cost for the treatment of about

10.000t of contaminated soil was estimated in 123€/t (140US\$/t) including excavation, pre-screening and staging (7-10.5€/t — 8-12US\$/t), product management activities (3.5-8.8€/t — 4-10US\$/t) and regrading and revegetation (4.4-8.8€/t — 5-10US\$/t).

However in order to consolidate the attained results and to evaluate the cost of the whole intervention, field pilot plant tests of 100kg/h capacity are planned for the second part of the 2002.

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